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Residence Time Distribution Measurements in a Downer Reactor & Gas-Solids Separator

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Abstract

An integrated gas-solids separator and stripper was developed, tested, and optimized for a circulating fluidized bed (CFB) downer at the Institute for Chemicals and Fuels from Alternative Resources (ICFAR) at Western University. The downer, designed for pyrolytic co-processing of heavy oil and biomass to valuable liquid fuels and chemicals, capitalizes on the plug flow behavior of gravity-assisted downward gas-solids flow, which has been proven in previous work. However, the effect of the reactor exit on unit performance has not been studied. Therefore, a comprehensive assessment of the gas-solids separator performance in a 0.07 m diameter, 1.34 m tall cold model downer was done in this thesis.

A novel, fast and cost effective pressure-response-based gas RTD measurement technique was developed that was able to screen potential separator designs. Several other conventional experimental methods, including solids RTD measurement using phosphorescent tracer and stripping efficiency measurement using CO₂ tracer, were used to further assess the separator performance. The separator performance was found to be strongly dependent on the separator cone diameter. A novel, objective Separator Performance Index (SPI) was developed to assess separator performance in a comprehensive manner. Separator performance was shown to increase dramatically with the separator cone size.

A 6.3 cm diameter, 60° internal angle cone-shaped separator performed best among several tested separator designs in terms of maximum solids collection efficiency (> 99.9%), good control of mean residence time (~ 0.5 s), and least gas backmixing. The cone-shaped separator performance was maximized with fully turbulent superficial gas velocities greater than 0.85 m/s, high solids-to-gas mass loading ratios greater than 10 kg/kg, and with stripping gas injected 30 cm below the gas outlet in the range of 6 % to 15 % by volume of the downer gas flowrate, which are preferred conditions for process intensification. Separator performance was shown to decrease with particle size. However, enhanced heat and mass transfer result with smaller particles. Therefore, the
use of stripping gas is essential to minimize gas backmixing in the separator when using small particles to achieve favorable pyrolysis reaction kinetics.

Keywords

multiphase flow, fluidization, fast fluidization, circulating fluidized bed, CFB, downer, pyrolysis, biomass, heavy oil, cyclone, separator, separation, gas-solids separation, residence time distribution, RTD, tracer, helium tracer, phosphorescent tracer, pressure response, deconvolution, axial dispersion, mixing, contact time
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Chapter 1

1 Introduction

Part A – Background

1.1 The Circulating Fluidized Bed Downer

At the Institute for Chemicals and Fuels from Alternative Resources (ICFAR) at Western University, numerous biomass conversion processes have been recently developed and investigated (e.g. Berruti et al., 2007; Berruti et al., 2009; Xu et al., 2009; Cascarosa et al., 2011; Latifi et al., 2014) to respond to growing demand for renewable chemicals and fuels (e.g. International Energy Agency, 2013; Peter & Lehmann, 2008). Biomass, including low-value industrial organic residues, has been identified in most countries as an abundant, renewable feedstock for fuel and chemical products (e.g. Offermann et al., 2011; Yamamoto et al., 2001). Hence, ICFAR’s biomass conversion processes have been developed for fundamental research at the laboratory and pilot scales on the path to commercial development. These processes have used mainly existing fluidized bed reactor technology for pyrolysis, torrefaction, and gasification reactions. Novel mechanical fluidization technology has also been developed as demonstrated by Briens et al. (2010), Cascarosa et al. (2011), Lance et al. (2011) and Latifi et al. (2014). The main products of interest have been bio-oil, biochar, combustible gases, and other specific chemicals from various types of agricultural, forestry, and industrial organic residues.

Among these fluidized bed-based processes, a circulating fluidized bed (CFB) downer reactor was developed for the simultaneous pyrolysis of heavy crude oil and biomass. This process has a historical basis in the work of Bergougno, Briens, and colleagues at Western University on the topics of ultra-rapid pyrolysis of heavy crude oil and fast gas-solids separation in a CFB downer during the 1980s and 1990s (e.g. Vogiatzis et al., 1989; Gauthier, 1990; Herbert et al., 1998). The more recent work at ICFAR (McMillan et al., 2006) has retained important lessons from the former, especially concerning mixing of feedstock with catalyst or heat-bearing solids and carrier gas upstream of the downer inlet, and in rapid separation of products from spent catalyst or solids. Mixing
and separation are especially important when the reaction is driven by heat addition and removal as opposed to catalytic conversion, as used in the fluid catalytic cracking (FCC) process.

In the pyrolysis reaction, the feedstock (heavy crude oil, solid biomass, or both) undergoes rapid (< 1 s) thermal decomposition at high temperature (roughly 500 °C to 600 °C), in the absence of oxygen, to produce gas, liquid, and solid products. Significant heat is required to bring the heat-carrying solids to the proper temperature. Therefore, it is important to recover heat from the solids through recirculation and by burning non-condensable gases produced by pyrolysis to provide further heat for the reaction. The purpose of the process using a downer reactor was to maximize the liquid yield to produce bio-oil from biomass and as an initial step in upgrading heavy crude oil to intermediate products. A downer reactor configuration was chosen over other reactor types for several demonstrated advantages (Bayle, 1996; Cheng et al., 2008; Cheng et al., 2013; Gauthier et al., 1992):

- to capitalize on the gravity-assisted, co-current gas and solids flow, thereby reducing the potential for reverse, upward flow and backmixing;
- to approach near to plug flow fluid mechanics and reactor performance;
- to achieve very short, well-controlled contact time between the gas phase (i.e. reacting feedstock) and solids phase (i.e. heat-carrier sand and / or catalyst).

In all applications, these proven advantages were aimed to maximize the yield of a narrow range of desired products. The co-current downward flow conditions of the downer all but eliminate the troublesome backmixing of the core-annulus flow regime observed in CFB riser reactors (Berruti & Kalogerakis, 1989; Brereton & Grace, 1993; Grace et al., 1999; Zhang et al., 2001). The main reason preventing extensive industrial use of the downer configuration is that most industrial riser installations in refineries were developed before the first reports of a downer, coupled with very little further capital expenditures on CFB reactor columns. In riser units, particle clustering and gravity work against the desired upward flow, leading to cascading downward streams of gas and solids along the wall. Zhang et al. (2001) demonstrated clearly in a direct comparison between a 10 cm diameter riser and a 10 cm diameter downer that, although a
core-annulus structure existed in the riser and the downer, the radial profile of solids concentration was much more uniform in the downer.

CFB downer reactors typically have four main sections, as illustrated in Figure 1.1:

1. **Inlet mixer:**
   Feedstock, carrier / fluidizing medium, fresh solids and / or recirculated solids enter the reactor and are mixed to rapidly initiate feedstock reaction.

2. **Downer:**
   The reaction continues as all phases are transported along the height of the vessel.

3. **Phase separator / quench:**
   Gas and solids phases are rapidly separated to terminate the reaction. Product quench may be integrated in the separator or located immediately downstream to further reduce secondary product reaction and to condense liquid products.

4. **Riser:**
   Catalyst and / or heat-bearing particles are regenerated, reheated, and recirculated to the inlet mixer.

![Figure 1.1 – Conceptual layout of a CFB downer reactor](image)

Unfortunately, the flow structure at the reactor inlet and in the gas-solids separator is not ideal and leads to gas and solids backmixing. Brust & Wirth (2004) studied the hydrodynamics of the gas-solids flow at the downer inlet, where it was found that high superficial gas velocities and low inlet solids velocities were essential to achieve plug flow-like behavior in the downer. Conversely, low superficial gas velocities and high
inlet solids velocities were found to cause severe backmixing in the downer. Johnston et al. (1999) also found that the gas and solids distributor designs had a very significant impact on the downer hydrodynamics.

In a similar fashion, the flow structure in the gas-solids separator at the downer exit, regardless of the separator design, is complex and almost certainly backmixed. This is due to sharp turns in the gas phase required for effective phase separation and the high likelihood of stagnant recirculation zone formation. No previous experimental studies have investigated the effect of the separator on the flow structure at the downer exit or the relative impact on the overall residence time distribution and reaction kinetics. However, many authors have stressed the importance of rapid, efficient phase separation with minimal backmixing for optimum downer performance. Therefore, there is a need to study the hydrodynamics and backmixing of candidate gas-solids separation devices.

### 1.2 The Cone-Shaped Gas-Solids Separator

To meet the challenging needs of biomass pyrolysis and heavy crude oil upgrading in a downer reactor, Huard (2009) developed a novel gas-solids separator for ICFAR’s CFB downer process. The separator was designed to achieve fast, efficient separation of the reacting gas phase from the heat-bearing solids phase and to minimize gas backmixing at the downer exit. The separator featured a cone-shaped particle deflector mounted above a gas outlet pipe, as shown in Figure 1.2. During operation, the incoming gas-solids suspension was diverted toward the downer wall by the particle deflector. Solids accumulated along the wall and were separated inertially and centrifugally as the gas turned sharply to exit the downer via the gas outlet pipe. The solids fell by gravity into a collection tank.
Extensive cold model testing showed that the gas-solids separation efficiency was 99.04% to 99.99% for a wide range of operating conditions representative of biomass and heavy oil pyrolysis in the downer. This range of measured efficiency was deemed sufficiently high for pyrolysis in the downer. However, the work by Huard (2009) was limited only to investigation of gas-solids separation phenomena. The extent of backmixing of both the gas and solids phases in the separator was unknown. Hence, the hydrodynamics of the gas-solids flow in the separator were deemed crucial to study in the present work to elucidate the effects of the separation process on the flow at the downer exit.

1.3 Motivations for Study of the Residence Time Distribution and Separator Performance in CFB Reactors

Measurement of the residence time distribution (RTD) is essential to understand hydrodynamics, flow structures, and backmixing in a CFB. Only Gauthier (1992) has previously measured the RTD in the cyclone of a downer reactor. Moreover, no studies have measured the RTD of both the gas and solids phases in the same downer. Given the
potentially crucial (though unknown) influence of the gas-solids separator on the product yield and composition, measurement of the gas and solids phase RTDs in ICFAR’s downer gas-solids separator were deemed necessary and were performed for this thesis. However, in order to implement appropriate and successful RTD measurement methods, an understanding of previous CFB RTD studies in the literature was required and performed in the current chapter.

Reaction conversion in riser and downer reactors is of primary interest in the design and operation of CFBs (Kunii & Levenspiel, 1997). A maximum yield of very specific products is desired; this yield depends on the conversion rate and the unit throughput. Since the reaction kinetics and conversion in CFB units are time-dependent phenomena, they are both therefore dependent on the residence time distribution and gas-solids contact time. This is especially important for catalytic cracking or fast pyrolysis reactors, in which the desired product is an intermediate product: in both types of reactor, it is important to keep the vapor residence time within a narrow band, as longer residence times lead to undesired non-condensable gases. Hence, knowledge of the flow and mixing behavior is of critical importance in CFB design and operation. The flow patterns and mixing / contact experienced by each phase determine the amount of time spent in different zones of the reactor by each phase, which in turn impact on the conversion and product yield.

Many researchers have studied the residence time distribution of one or both phases to gain insight into the mixing behavior and hydrodynamics in CFB risers and downers. In the simplest sense, the RTD is a description of all possible time values that elements of a substance could spend in a reactor or vessel. There is one condition on this definition, which is that representative particles of the substance cannot re-enter the vessel after crossing any of its boundaries. This condition ensures that only unique, continuous transits through the vessel are counted in the RTD measurement, which gives the true RTD. However, due to diffusion, turbulence effects, and other complicating flow conditions, the true RTD is an asymptotic limit that can never be fully realized in practice. Very close approximations to the true RTD can be obtained in tracer
experiments with careful treatment of the boundary conditions and of the injection and detection of tracer (Levenspiel et al., 1970; Harris et al., 2002a).

Response function RTDs describe mathematically how the concentration of one phase evolves with time between two particular locations in a reactor. The RTD effectively “smears” and shifts the inlet concentration versus time curve to give a new concentration curve at the outlet, which has a larger average time and wider distribution. The inlet and outlet concentration curves are linked to each other via convolution of the inlet concentration curve with the RTD. The RTD captures all of the particular flow structures and the state of mixing existing between the measurement boundaries. With proper treatment of inlet and outlet boundary conditions and tracer injection and detection, the response function RTD approximates the true RTD (Levenspiel, 1999). True RTDs and their approximations have the property that they can almost always be modeled as a probability density function, which can be analyzed using statistical moments (Harris et al., 2003a).

Figure 1.3 shows an illustrated fictitious experimental response function RTD curve for the downer section of a CFB. The illustration also shows fictitious tracer concentration versus time curves for the downer inlet and outlet. The RTD curve has two peaks, with one prominent primary peak followed by a smaller secondary peak. The secondary peak is an indication that significant backmixing may exist somewhere in the unit, or else the flow through the reactor may be split between parallel non-contacting streams. It is interesting to note that the RTD is blind to the spatial tracer concentration distribution within the unit. However, one can gain spatial detail and identify problematic zones by performing tracer experiments at different locations in the unit, at the risk of increasing inaccuracy and experimental error due to improper boundary selection and tracer injection and detection methods.
1.4 Chapter Objectives

The main goal of this work was to allow for maximum liquid product yield and selectivity of the pyrolysis process in the downer by characterizing and optimizing the performance of the cone-shaped gas-solids separator. In order to achieve this main goal, the following objectives were set out for the present chapter:

- Understand previous work in CFB reactors on hydrodynamics, mixing, RTDs, and separator performance characteristics
- Apply the findings of the literature search to:
  - Understand how CFB reactor / separator performance has previously been assessed
  - Set out realistic performance objectives for the gas-solids separator
o Develop reliable, accurate techniques to measure the RTD of both the gas and solids phases in the gas-solids separator

The present chapter presents background and a review of the literature on the topics of CFB column exit and separator performance characteristics used in the literature, experimental measurement and analysis of residence time distributions and hydrodynamics in CFB reactors, and application of stripping gas to improve CFB separator / regenerator performance.

Part B – Literature Review

1.5 CFB Exit / Separator / Stripper Performance

1.5.1 Introduction & Importance

The effects of the column exit in CFB risers have been studied extensively in several works, with a comprehensive review provided by Chan et al. (2010). Experimental results have shown that the exit configuration has a huge impact on hydrodynamics and mixing, with abrupt exits having much stronger gas and solids recirculation than smooth exits (Pugsley et al., 1997). Gas and solids recirculation at the riser exit has also been shown to impact on the overall unit hydrodynamics. However, as noted earlier, no previous studies have investigated the effect of the downer exit on the local and overall unit hydrodynamics and performance. Furthermore, since ICFAR’s gas-solids separator is essentially an integrated downer exit and fast separator, there is a need first to understand CFB column exit effects and second to review separator performance characteristics in the literature. Finally, since stripping gas is meant to be used to reduce gas backmixing in ICFAR’s gas-solids separator, and since stripping is normally performed on the solids before they flow to the regenerator of fluid catalytic crackers and fluid cokers, it is also important to understand how stripping performance has been assessed in the literature. Therefore, a broad definition of “separator performance” must necessarily be used in the context of ICFAR’s integrated gas-solids separator.
1.5.2 Performance Characteristics

The column exit effects in CFB risers in the literature have typically focused on four main topics:

i) Retrograde length of influence relative to column height (Harris, Davidson, & Thorpe, 2003b)

ii) Axial and radial mixing of both the gas and solids phases (concentration distributions, recirculation) (Zhou, Grace, Lim, & Brereton, 1995)

iii) Solids reflux and recirculation (Gupta & Berruti, 2000)

iv) Solids RTD (Harris, Davidson, & Thorpe, 2003b)

In general, as noted above, abrupt exits were found to have stronger particle refluxing than smooth exits, effectively making them behave as gas-solids separators. Gupta and Berruti (2000) indicated that particle characteristics were also important with regard to exit effects, with Geldart Group A particles leading to less severe exit effects than Geldart Group B particles. However, in spite of the difference between abrupt and smooth riser exits, the impact of exit geometry on the solids RTD was shown to be distinguishable, though limited, by both Rhodes et al. (1991) and by Harris et al. (2003b). Since refluxing has not been shown to be a prominent phenomenon in CFB downer exits, only the solids RTD is expected to be relevant with regard to ICFAR’s gas-solids separator performance among the topics listed above.

Primary gas-solids separator performance has been assessed in cyclones and special separator designs in both CFB riser and downer units. An extensive review of gas-solids separator performance characteristics is provided by Huard (2009). Separators of all types have been assessed mainly according to:

i) Solids collection efficiency

ii) Pressure drop

iii) Gas underflow

iv) Gas RTD

The solids collection efficiency is always desired to be maximized, but usually comes at the expense of other performance metrics, namely pressure drop. Gas underflow refers to the fraction of the total gas stream that is entrained with the collected solids stream into
the diplegs (in FCC units). Underflow is typically desired to be minimized since product vapors can be degraded into undesirable permanent gases through excessive residence time in the separator and diplegs. Gauthier (1991) and Gauthier et al. (2005) both studied gas underflow in CFB gas-solids separators of two different designs and found that the addition of separator sealing gas (i.e. stripping gas) greatly aided to reduce excessive gas residence time and reaction.

1.5.3 Effect of Particle Size on Separator Performance

The effect of particle size on fluid-solids and fluid-fluid separator performance is likely the single most important consideration affecting the separator design. In most studies on dust cyclones, hydrocyclones, and demisters of various designs, the grade efficiency curve is typically used to characterize a separator’s ability to remove particles of different sizes (e.g. Vaughan, 1988; Hoffmann et al., 1992; Yang et al., 2010). The grade efficiency curve is a plot of the particle collection efficiency plotted for specific ranges of particle size, as shown in Figure 1.4. Most authors have used experimental grade efficiency curves either to calibrate analytical collection efficiency models (e.g. Maynard, 2000) or to determine whether the collection efficiency for a specific separator design and range of particle size will be sufficient for a given application. In general, experimental grade efficiency curves are S-shaped, where particle collection efficiency increases with particle size. This is due to two main effects: the terminal velocity of a particle in either the gravity field or a centrifugal field increases with particle size, assisting in particle collection. Agglomeration and clustering of particles may further complicate the situation and result in non-monotonic behavior of collection efficiency with changing particle size.
Particle size and particle size distribution (PSD) have also been shown to have a significant effect on the hydrodynamics, mixing, and performance in conventional fluidized bed reactors (Grace & Sun, 1991). Among Geldart Group A PSDs with the same average size, wider PSDs have been shown to result in an expanded dense phase, lower effective viscosity, smaller bubble size, better gas-solids contacting, and higher conversion in tests spanning the bubbling to fast fluidization regimes. However, Zhu et al. (1995) identified a need for research on the effect of particle size in CFB downers. Wang et al. (2005) tested the effect of three different coal particle size distributions (<280 μm, 280 μm to 450 μm, and 450 μm to 600 μm) and 250 μm silica sand on the pyrolytic conversion of coal to gas and liquid products in a CFB downer. The authors found that the liquid yield, and specifically of desired aliphatics and methylphenols, decreased with increasing particle size. This was attributed to reduced gas-solids heat transfer and increased secondary reactions with the large PSD. However, to the author’s knowledge, no other studies exist in the literature investigating the effect of PSD on downer hydrodynamics and performance.

1.5.4 Motivations
Given that several different performance characteristics have previously been used to assess different parts of CFB units surrounding the exit and gas-solids separator, it is important to recognize how ICFAR’s gas-solids separator can be developed to integrate several of these components into one effective separator. Therefore, a comprehensive
evaluation of all relevant factors under realistic conditions must be considered. The following performance characteristics and other effects are deemed relevant to investigate further both in the literature and in ICFAR’s cone-shaped gas-solids separator:

i) Solids collection efficiency (performed by Huard, 2009)
ii) Gas RTD
iii) Solids RTD
iv) Pressure drop (reviewed by Huard, 2009)
v) Gas underflow / solids stripping
vi)Particle size

The remainder of this chapter is devoted to a literature review of CFB gas and solids RTD measurement and modelling, of previous stripping gas studies, and of specific objectives for this thesis.

1.6 Gas Phase CFB RTDs in the Literature

1.6.1 Introduction & Importance

Historically, most residence time distribution and phase mixing studies have been performed in riser reactors rather than downers. This is due to more extensive industrial application of riser reactors, most of which were developed and constructed before the downer had been conceived and investigated in academic settings. Some of the findings in riser reactors are applicable to downer reactors since the operating conditions are similar in both reactor types. Gas RTD measurement in CFBs is useful to:

- Gain insight into gas flow patterns in the unit
- Estimate gas axial and radial mixing variables, such as dispersion coefficients
- Predict reaction kinetics when the gas is reacting
- Optimize the reactor design

Much controversy has abounded in the literature regarding the actual flow structure in risers. Some authors asserted that gas backmixing was negligible and assumed plug flow behavior. Others reported plug flow behavior in the core but significant backmixing in the annulus. Li & Weinstein (1989) measured gas backmixing across the full spectrum of fluidization regimes and found that backmixing was strongly dependent on operating
conditions, and especially on the gas superficial velocity, in the fast fluidization regime. Vandewalle et al. (2002), on the other hand, claimed that the main cause of gas backmixing in CFB risers was by adsorption of the gas phase on active catalyst particles. Since several differing opinions exist, the current section reviews the work done to obtain the gas phase RTD in CFB risers and downers. Much less work exists on this topic in the literature than for solids phase RTD studies.

1.6.2 Experimental Methods

All RTD studies in CFB reactors can be categorized according to:

- the type, injection method, and injection location of tracer used to measure the RTD
- the type and location of tracer detector
- the type of model used to fit the measured RTD.

These categories will be employed to summarize and discuss the various experimental methods used in gas and solids RTD studies. Table 1 summarizes relevant test conditions for all of the gas phase RTD studies discussed here.

A number of different gas tracers have been used in gas phase RTD tests in CFB units. Helium was used by Yang et al. (1983) and Bader et al. (1988) to study the gas RTD. Gauthier et al. (2005) also used helium tracer to study the gas phase RTD in an FCC riser using a novel riser termination device. Helium is inert and has unique thermal properties making it relatively easy to detect with heat transfer-detection methods. Highly cost prohibitive and scarce radioactive argon was used by Patience & Chaouki (1993), while non-radioactive argon was used by Brust & Wirth (2004) and Dry & White (1989). In Kagawa et al. (1991), the authors used ozone decomposition, while propane was used in Vandewalle et al. (2002) and Smolders & Baeyens (2000a). Namkung & Kim (1998) found that the axial gas dispersion coefficient (a parameter used to assess the closeness to plug flow, which is discussed later) determined using CO$_2$ as tracer was overestimated in all regions of the riser and by as much as 62%. The authors explained that CO$_2$ was adsorbed on the solid phase particles and was extensively back-mixed. The implication
for gas RTD studies is that adsorbing tracers are not representative and cannot be used if the main carrier gas is non-adsorbent.

Two modes of tracer injection are possible: pulse and step. In pulse injection mode, the intention is to rapidly inject a small amount of tracer so as to approximate a Dirac delta function. Gas pulse injections were performed in studies by Dry & White (1989), Patience & Chaouki (1993), Vandewalle et al. (2002), and Brust & Wirth (2004). Only Patience & Chaouki specified the method by which a tracer pulse injection was performed, which in their case was done manually using a syringe. Patience & Chaouki injected a very small amount of tracer (9 mg), which ensured that the riser hydrodynamics were not disturbed by the injection. In step injection mode, the intention is to rapidly switch the flow from the carrier fluid to tracer or vice versa so as to approximate a perfect step function. Step injections were performed by Namkung & Kim and by Brust & Wirth. Namkung & Kim injected tracer at equal velocity to the superficial gas velocity in the riser via an upward-pointing tube whose radial position could be adjusted. In both pulse and step injection modes, Levenspiel & Turner (1970) demonstrated that very significant errors in the measured RTD were present when the velocity profile at the injection point was not uniform and, under this same condition, tracer was injected uniformly across the cross-section at equal flowrates.

As for tracer detection methods, unlike in some solids phase RTD studies, the response time of the detectors was not a problem. Mass spectrometry (MS) was used in Dry & White (1989) and in Brust & Wirth (2004) to detect argon gas. Gas chromatography (GC) was used by Smolders & Baeyens (2000a) and Vandewalle et al. (2002). Patience & Chaouki (1993) used a NaI scintillator counter. Detectors sensing differences in heat transfer were typically used when helium was used as tracer. Heat flux probes, such as thermal conductivity detectors and hot wire anemometers, are relatively simple to implement and are replaced at low cost. However, MS, GC, and heat flux detection methods all require sampling lines which, depending on their length, may allow for significant tracer diffusion and dispersion, thereby skewing the measurement. Sampling line lengths were not specified in any of the studies discussed here, and therefore the accuracy of the measurements should be regarded with some skepticism. The presence of
solids makes gas tracer sampling even more difficult since solids inevitably accumulate and cake on the sampling line tip. Heat flux probes also have the disadvantage of relatively low sensitivity. Radioactive tracer detectors are the least obtrusive but most cost prohibitive among the detectors discussed here. Levenspiel & Turner (1970) noted that RTD measurement errors also existed when the velocity profile at the detection location was non-uniform combined with disproportionate tracer sampling.
Table 1.1 - Test conditions for various gas phase CFB RTD studies

<table>
<thead>
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</tr>
</thead>
<tbody>
<tr>
<td>Brust &amp; Wirth (2004)</td>
<td>Downer</td>
<td>8.6</td>
<td>0.15</td>
<td>0 – 7</td>
<td>25 – 120</td>
<td>Ambient</td>
<td>FCC</td>
<td>--</td>
<td>85</td>
<td>Argon</td>
<td>Pulse &amp; Step</td>
<td>Mass spectrometer</td>
</tr>
<tr>
<td>Dry &amp; White (1989)</td>
<td>Riser</td>
<td>7.2</td>
<td>0.09</td>
<td>2.0 – 8.0</td>
<td>36 - 227</td>
<td>Ambient</td>
<td>FCC</td>
<td>1370</td>
<td>71</td>
<td>Argon</td>
<td>Pulse</td>
<td>Mass spectrometer</td>
</tr>
<tr>
<td>Patience &amp; Chaouki (1993)</td>
<td>Riser</td>
<td>5</td>
<td>0.083</td>
<td>4 - 8</td>
<td>20 - 140</td>
<td>Ambient</td>
<td>Sand</td>
<td>2630</td>
<td>277</td>
<td>Radioactive argon</td>
<td>Pulse</td>
<td>NaI scintillation</td>
</tr>
<tr>
<td>Smolders &amp; Baeyens (2000a)</td>
<td>Riser</td>
<td>6.5</td>
<td>0.1</td>
<td>3.5 – 4.5</td>
<td>0 - 38</td>
<td>Ambient</td>
<td>FCC Sand</td>
<td>1700 2600</td>
<td>70 90</td>
<td>Propane</td>
<td>--</td>
<td>--</td>
</tr>
<tr>
<td>Vandewalle et al. (2002)</td>
<td>Riser</td>
<td>6.5</td>
<td>0.1</td>
<td>4</td>
<td>6 - 25</td>
<td>Ambient</td>
<td>FCC Sand</td>
<td>1000 1500</td>
<td>70 120</td>
<td>Propane</td>
<td>Pulse</td>
<td>Gas chromatograph</td>
</tr>
<tr>
<td>Namkung &amp; Kim (1998)</td>
<td>Riser</td>
<td>5.3</td>
<td>0.1</td>
<td>1.5 – 4.5</td>
<td>14 – 62</td>
<td>Ambient</td>
<td>FCC</td>
<td>1720</td>
<td>65</td>
<td>Helium &amp; CO₂</td>
<td>Step</td>
<td>Gas chromatograph</td>
</tr>
</tbody>
</table>
1.6.3 Experimental Results

Most gas phase RTD studies in risers demonstrated significant gas backmixing. This was indicated by the presence of short peak heights and long tails on the measured RTD curves, as shown in Figure 1.5. Increasing mean residence times with decreasing gas velocity and with increasing solids loading were observed in all studies, which is the same effect observed in many solids RTD studies. A bimodal RTD was observed by Vandewalle et al. at $U_g = 4$ m/s, which indicated strong recirculation. Dry & White and Smolders & Baeyens observed that the peak spread in the RTD signal decreased with increasing gas velocity, which indicated decreasing backmixing, while the peak spread increased with the solids circulation rate. However, Smolders & Baeyens concluded that the gas phase behaved in plug flow – in conflict with other studies reviewed here.

Brust & Wirth demonstrated that a downer reactor could be operated near the plug flow regime, but this behavior was highly contingent on the gas-solids mixing at the entrance to the downer. The authors introduced secondary gas in the gas-solids mixer to enhance inlet mixing. The use of secondary gas was observed to increase gas backmixing at the downer inlet. High superficial gas velocities in the downer coupled with low secondary gas velocities were essential to reduce the gas-solids backmixing at the downer inlet. The method of gas-solids distribution and flow at the downer inlet were also found in Johnston et al. (1999) to have a strong impact on the developed flow in the downer.

Figure 1.5 – Experimental RTDs in a riser reactor: (a) modified from Patience & Chaouki (1993); (b) modified from Dry & White (1989)
Figure 1.6 below compares typical gas RTDs measured by Brust & Wirth in the respective downer and riser sections of their apparatus under identical operating conditions. The figure demonstrates a much narrower, taller peak in the downer as compared to the riser, indicating significantly less backmixing in the downer.

Figure 1.6 – Experimental gas phase RTDs: downer versus riser (modified from Brust & Wirth, 2004)

1.6.4 Motivations

Given the above discussion, wherein no previous work has been done on the gas RTD in a CFB downer gas-solids separator, it is imperative to determine the effect of the separator on the overall gas backmixing in the unit. Average nominal residence times in unique separator designs have been calculated but have not been measured (e.g. Gartside & Woebcke, 1981). No estimates of the extent of backmixing or peak spread have been performed nor measured. Since the average residence time in the separator can be statistically significant relative to the overall residence time for the rest of the unit, it is reasonable to assume that the impact on the reaction kinetics might also be significant. Therefore, the need exists to measure and compare the RTD in ICFAR’s separator for several designs and optimized in order to minimize gas backmixing. Furthermore, a
simpler and better gas RTD measurement method should be developed to avoid the cost prohibitive nature of radioactive tracers and the experimental dispersion errors induced by gas sampling as in mass spectrometry.

1.7 Solids Phase CFB RTDs in the Literature

1.7.1 Introduction & Importance

Measurement of the solids RTD in CFB risers and downers is very useful for several purposes. In non-catalytic gas / solid reactions, such as biomass pyrolysis, the main applications of the solids RTD are to ensure adequate gas / solids contact times and to assess axial and radial heat and mass transfer (Huang, Qian, Zhang, & Wei, 2006). In other situations, the solids RTD is used to control solids mixing and residence time, to characterize reactor hotspots, and to identify unreacted material (Lackermeier & Werther, 2002).

Many different experimental techniques have been employed to directly measure the solids phase RTD, all of which involve the use of a solid tracer material that is assumed to accurately represent the flow patterns and characteristics (e.g. density, size, shape) of the actual solids phase. These various experimental methods and results are reviewed in the following section.

1.7.2 Experimental Methods

In short, the experimental procedure for measuring the solids phase RTD in a CFB riser / downer involves:

i. Pulsed or step injection of a small amount of tracer particles, or introduction of a single tracer particle into the surrounding flow,

ii. Measurement of tracer concentration with time as the tracer flows past the detector(s).

Figure 1.7 shows a typical arrangement for a tracer injection / detection system in a CFB riser. Depending on the locations and methods of tracer injection and detection, the RTD may be measured between any two locations.
Many different tracers and tracer injection techniques have been used to measure the solids phase RTD in CFB reactors. These various methods are grouped by tracer particle property: radioisotope, ferromagnetic, salt, chemical, luminescent (i.e. fluorescent and phosphorescent), and other miscellaneous types. An excellent summary of previous experimental investigations into the solids RTD in CFB risers is provided in Harris et al. (2003a) and in Gao et al. (2012), including information about: tracer type, riser geometry (e.g. height $H$ and diameter $D$), and operating conditions (e.g. superficial gas velocity, solids flux, reactor temperature, particle properties). Huang et al. (2006) also provide a summary of previous solids phase RTD studies. Their critical reviews are omitted here for conciseness.

Solids phase RTD studies in downers were reported in Roques et al. (1993), Wei et al. (1994), and Huang et al. (2006), in which all used phosphorescent tracer particles. The use of phosphorescent tracer particles seems to be the most reasonable compromise between RTD accuracy and ease of implementation. Phosphorescent particles used in RTD studies were activated using strong visible light flashes to create very close
approximations to true pulse injections. However, preferential activation of particles near the wall versus particles near the column centerline may be a problem if the flash brightness is too low or when operating at high solids mass flux. Excessive tracer may also be activated if the light flash is not collimated. Radioactive tracers can give accurate results but require long down times between experiments as the tracer is recovered and the reactor is decontaminated from residual tracer. Use of salt tracer is easy to implement but results in large errors in the injection and detection / sampling stages. Organic chemical tracer is moderately difficult to implement and has problems with re-adsorption on the solid particles.

The response time of solids tracer detection and sampling is one of the greatest limiting factors of RTD measurement. For example, in Bader et al. (1988), Rhodes et al. (1991), and Zheng et al. (1992), the temporal resolution of salt sampling was on the order of 0.5 s, which is far too long for accurate RTD measurement according to Harris et al. (2003a). Scintillation counters used with radioactive tracers (e.g. Ambler et al., 1990, Patience et al., 1990) have extremely short response times and are generally quite accurate, but they may erroneously detect radiation too early or too late if the emitted radiation is not collimated into the detector. Nearby particles at rest (e.g. solids collected in a cyclone dipleg) can also be detected erroneously if proper shielding is not used. Gas chromatography (GC) is often used to detect organic tracer desorbed from solid particles. GC also has adequate response times but may give erroneous RTD results if the organic tracer re-adsorbs onto the solid particles and is not captured by the detector. Photomultiplier tubes and photocells are typically used with phosphorescent and fluorescent tracer particles. They have good temporal resolution but their sensitivity may be reduced at heavy solids loadings by non-phosphorescent particle shielding (Yan et al., 2009). Like radiation detectors, these sensors may also detect tracer too early or too late if the emitted light is not collimated into the detector.

Harris et al. (2002a) listed the following numerous advantages of the phosphorescent tracer technique:

- instantaneous, non-intrusive activation of tracer by a light pulse,
- simple, non-intrusive online detection of the tracer by a light detector,
• the tracer is identical to the rest of the bed material (when not blended with the actual bed material used in the reaction),
• no extra particles or gas are added to disturb the flow,
• no tracer accumulation in the bed since the bed particles deactivate, and
• low cost compared with radioactive tracer studies.

However, the method also has a few minor disadvantages:
• excessive or improper activation of tracer if the light flash is not plane collimated,
• early or late detection of activated tracer if the detected light is not collimated at the detector, and
• preferential activation and/or detection of particles near the wall versus particles near the column centerline if the flash brightness is too low or when operating at high solids mass flux.

Detection of tracer outside of the measurement plane would tend to skew the measured RTD to be narrower and earlier than the true RTD due to detector bias toward the slightly brighter solids upstream of the detector. Overall, the phosphorescent technique seems to be the most reasonable compromise between RTD accuracy, ease of implementation, and cost after taking precautions for proper tracer activation and detection.

1.7.3 Experimental Results

Experimentally determined solids phase RTDs in CFB risers/downers are used to:
• estimate the gas-solids contact time;
• quantify the extent of backmixing in the reactor;
• identify problematic flow regions;
• apply the findings to tune hydrodynamics and reaction kinetics models.

Solids backmixing can have a strong negative impact on conversion and is typically to be avoided. Experimental RTDs have also given evidence for the existence of a core-annulus flow structure in risers (e.g. Rhodes et al., 1991; Harris et al., 2003a).

The effect of superficial gas velocity and solids circulation rate on the measured RTD in risers/downers has been studied extensively. On the whole, the results were fairly consistent. Most authors concluded that increasing the superficial gas velocity led to a
decrease in the mean residence time, decreased axial dispersion, and a tendency toward plug flow in both the gas and solids phases (e.g. Ambler et al., 1990; Rhodes et al., 1991; Smolders & Baeyens, 2000b; Harris et al., 2003a; Yan et al., 2009). Harris et al. (2003a) demonstrated that increasing gas velocity decreased the RTD signal’s variance (i.e. a measure of peak spread) and breakthrough time (i.e. time required for tracer to be initially detected).

The solids loading has a less clear influence on the solids RTD. In general, increasing solids flux has been found to cause increased mean residence time, increased axial dispersion, shrinking peak height, increased tail length, and in some cases the appearance of a second peak in the RTD (e.g. Harris et al., 2003a; Ambler et al., 1990; Smolders & Baeyens, 2000b). However, conflicting results were reported in Rhodes et al. (1991) and Yan et al. They found that axial dispersion actually decreased slightly with increasing solids flux. They acknowledged that this relationship was quite weak and claimed that the effect of the superficial gas velocity was much more influential on mixing.

The riser diameter has also been shown to have an effect on the solids RTD. Conflicting conclusions are given in Rhodes et al., where it was found that increasing the riser diameter decreased backmixing, and Pugsley et al. (1997), who found an opposite trend. An interesting result in downer reactors was obtained by Huang et al. (2006), where it was found that there was essentially no significant scale-up effect on backmixing when increasing the reactor diameter. They also claimed that the radial solids mixing was more intense in the larger diameter reactor, which they surmised to be advantageous for downer scale-up.

Figure 1.8 compares the solids axial Péclet number versus superficial gas velocity in a similar sized riser versus downer as reported by Harris et al. (2003) and Wei et al. (1994), respectively. The dimensionless Péclet number, \( \text{Pe} = \frac{LU_p}{D_{ax}} \), where \( L \) is the column length, \( U_p \) is the bulk particle velocity, \( D_{ax} \) is the axial dispersion coefficient, is the ratio of convective transport to diffusion-like or dispersion-like transport. Higher Péclet numbers indicate less backmixing approaching plug flow behavior. The results showed that the axial Péclet number was roughly five times greater in the downer versus the riser.
The solids flux was similar between the two studies. However, it must be noted that there were significant differences between the column inlet and outlet configurations in the two studies; therefore, the differences in the reported Péclet numbers should not be considered absolute or authoritative. Furthermore, Wei and Zhu (1996) showed that dispersed particles in risers have axial Péclet similar to the solids in a downer, indicating near plug flow behavior for the dispersed particles.

Figure 1.8 – Comparison of typical solids axial Péclet number observed in a CFB downer (from Wei et al., 1994) versus a CFB riser (from Harris et al., 2003a)

1.7.4 Motivations

Since axial dispersion in downers has been shown to be very low compared to risers, the corresponding relative impact of the gas-solids separator on the overall solids RTD may be significant, but is unknown. As shown, the solids RTD has not previously been measured in the separator of a CFB downer. Moreover, the impact of solids RTD on the separator performance has also not been investigated. Therefore, there is a need to measure the solids RTD in ICFAR’s cone-shaped gas-solids separator and to determine the impact of the RTD on separator performance. Furthermore, there is a need to integrate
both the gas and solids RTD results into a realistic flow model, which has not been done in a downer gas-solids separator.

1.8 RTD Modelling Approaches

The majority of RTD modelling has been applied to CFB risers due to their more frequent use and because of their relatively more complex flow patterns as compared to CFB downers. Most authors have found that plug flow and modified plug flow with axial dispersion models adequately described both the gas and solids RTDs in downers (Wei et al., 1994; Bayle, 1996; Huang et al., 2006). Much more complex models have been developed to describe the core-annulus structure and recirculation observed in risers (Ambler et al., 1990; Harris et al., 2002b). Furthermore, very similar models have been applied to both the gas and solids phases. These modelling approaches are reviewed briefly here.

In a plug flow reactor model, as used by Bayle (1996) to describe the gas and solids RTDs in a CFB downer, axial dispersion is assumed to be negligible. The RTD in this case, \( E(t) \), is represented by a Dirac delta function, \( \delta(t) \), shifted by the mean residence time (\( \tau \)) relative to the vessel entrance time (\( t \)):

\[
E(t) = \delta(t - \tau).
\]

Since axial dispersion is assumed to be negligible in this model, the RTD peak spread is zero. This condition is a theoretical limit that can never be truly achieved in practice (Levenspiel, 1999). However, the model was shown to represent the gas phase flow well in the work by Bayle.

Axial dispersion models (ADMs) similar to the following expression were applied in numerous studies to model the RTD in CFB risers (e.g. Brust & Wirth, 2004; Dry & White, 1989; Wei et al., 1994; Huang et al., 2006):

\[
\Delta \sigma^2 = 2\Delta t^2 \frac{D_{ax}}{U_g L} \left(1 - \frac{D_{ax}}{U_g L}\right) \left(1 - e^{-U_g L/P_{ax}}\right),
\]

(1.2)
where $\sigma^2$ is the signal variance, $t$ is time, $D_{ax}$ is the axial dispersion coefficient, $U_g$ is the gas superficial velocity, and $L$ is the characteristic length of the vessel. Writing equation (1) in terms of the Péclet number one obtains

$$\Delta \sigma^2 = 2\Delta t^2 \text{Pe}^{-1} \left(1 - \text{Pe}^{-1}\frac{1}{1 - e^{-\text{Pe}}}ight),$$

in which the closed-closed Danckwerts boundary conditions are usually assumed implicitly:

At $z = 0$: $U_g C\bigg|_{z=0^-} = \left(U_g C - D_{ax} \frac{dC}{dz}\right)_{z=0^+}$, and

$$\text{At } z = L: \left.\frac{dC}{dz}\right|_{z=L^-} = 0,$$

where $z$ is the distance along the vessel, $C$ is the concentration of tracer, $z = 0^-$ and $z = 0^+$ refer to the points immediately upstream and downstream of the inlet boundary, respectively, and $z = L^-$ refers to the point immediately upstream of the outlet boundary.

In this model the peak spread is proportional to the axial dispersion coefficient, which is typically used as an adjustable parameter to fit the experimental data. Peak spread is also linearly proportional to the vessel length. The ADM has been popular in the literature since it has only two parameters and is very simple to fit to experimental data. However, the appropriate use of this model depends on the multiphase flow phenomena actually occurring in the unit, and the correct treatment of the boundary conditions at the entrance and exit to the reactor zone being modeled (Levenspiel & Fitzgerald, 1983; Briens et al., 1995). Closed boundary conditions signify plug flow upstream and downstream of the reactor such that the reactor can be considered to be a closed vessel. Open boundary conditions exist where there is no significant transition in flow regime at the entrance and exit of the reactor. Levenspiel (1999) stated that the ADM can be used for either type of boundary condition only for small amounts of dispersion (i.e. near plug flow). For larger extents of dispersion, one must ensure that closed boundary conditions exist to get a good approximation to the true RTD.

Another popular class of models has been the core-annulus tracer balance approach for CFB risers. This model class assumes the existence of:

- a dense, fast fluidized region at the bottom of the riser,
dilute, rapid bulk flow in the core of the riser, and
• a thin, annular, dense, slow flow region along the wall.

These models were characterized by several mass and momentum exchange coefficients between the different flow regions, which attempted to describe the actual flow structures in the unit. The equations for this type of model are rather lengthy and tedious, and are thus not shown here for conciseness. The authors in Kagawa et al. (1991), Patience & Chaouki (1993), Kruse & Werther (1995), and Vandewalle et al. (2002) developed core-annulus gas tracer balance models in risers. Ambler et al. (1990) and Harris et al. (2002b) developed core-annulus solids tracer balance models in risers. The models were used with varying degrees of success. In any case, one of the main advantages of the downer is that the problematic core-annulus structure is virtually eliminated as shown by Zhang et al. (2001).

As previously mentioned, the time-dependent tracer concentration at an exit boundary is equivalent to convolution of the time-dependent tracer concentration at the entrance with the true RTD. Therefore, numerical deconvolution may be used to directly obtain the response function RTD in systems where either the tracer concentration is measured at entrance and exit, or only at the exit so long as the tracer injection is well characterized. A typical deconvolution procedure was described in Brust & Wirth (2004). Deconvolution directly gives the RTD without the need to fit any model parameters. However, the process is extremely sensitive to noise in the input signals and must therefore be used with heavy data smoothing and filtering.

The last modelling technique to be discussed here is the use of compartment models. In this method, each reactor zone, with its particular RTD, is assumed to be made up of plug flow and mixed flow compartments with recycle flow (Levenspiel, 1999). The method of compartments was applied by Huard et al. (2011) to model the RTD of single phase gas flow in a downer. The work by Huard et al. is provided in Appendix E. The advantages of compartment models are that the reactor entrance / exit boundary conditions do not need to be considered and the models can be superimposed to any level of complexity. Its
disadvantages are less accurate agreement with experimental data and use of several fitting parameters.

1.9 Use of Stripping Gas

Stripping gas has traditionally been used in the fluid catalytic cracking (FCC) process in the spent catalyst bed downstream of the gas-solids separator to improve recovery of entrained hydrocarbon vapor prior to catalyst regeneration (Avidan et al., 1990; McKeen and Pugsley, 2003; Gao et al., 2008). It has also been used and studied extensively in the fluid coking process to strip residual liquid hydrocarbons in addition to hydrocarbon vapor recovery (Bi et al., 2005; Cui et al., 2006). The main issues of academic interest in both applications are to measure and model hydrodynamics, mass and heat transfer, and stripping efficiency in order to optimize the stripper design and operating conditions. In both FCC and fluid coker strippers, the particle bed is operated in either of the bubbling or fast fluidized regimes.

Gao et al. used a thermal conductivity detector (TCD) in a gas chromatograph to measure the concentration of \( \text{O}_2 \) tracer (and thus the stripping efficiency) at several heights along the stripper in a cold model FCC riser. The stripper was operated without any stripper internals and with downward sloped baffles. The authors found that stripping efficiency increased with decreasing height above the gas distributor and with increasing superficial gas velocity. The V-shaped baffle configuration also resulted in better stripping efficiency than the stripper with no internals. Cui et al. measured the stripping efficiency in the standpipe of a cold model fluid coker using helium tracer detected by a TCD. The authors acknowledged that the non-adsorbing helium did not accurately represent adsorbing hydrocarbon vapors, but deemed the accuracy of the method sufficient for ranking different stripper configurations. Stripping efficiencies greater than 99.7 % were reported with a highly developed and complex stripper design. Hulet et al. (2008) used a \( \text{CO}_2 \) tracer technique to measure the entrainment of gas into a horizontal gas jet used to transport solids between two segregated fluidized beds. The solids were non-porous and there was negligible adsorption of the \( \text{CO}_2 \) on the particles. Tracer was injected upstream of the fluidization gas windbox and detected by a \( \text{CO}_2 \) probe and transmitter in the exhaust line of one of the fluidized beds. The authors successfully related the \( \text{CO}_2 \)
concentration measured at the probe to the flowrate of entrained gas in the gas jet. Among the techniques described here, the CO\textsubscript{2} tracer method was the most cost effective method and provided good accuracy.

### 1.10 Specific Objectives

Based on the preceding discussion, the following specific objectives are proposed for this thesis, with reference to where they are discussed in detail:

- Describe the implementation of conventional experimental methods (Chapter 2) for measurement of:
  - Separator pressure drop
  - Local gas velocity
  - Solids collection efficiency
  - Solids RTD
  - Solids stripping efficiency

- Develop a simple, novel gas RTD measurement method (Chapter 3) that:
  - Eliminates dispersion in sampling lines
  - Avoids the difficulties and cost of working with radioactive tracer
  - Has a fast response, is cost effective, and can be applied to other reactor types and situations

- Measure and assess gas-solids separator performance and compare different separator designs (Chapter 4 and 5) based on:
  - Solids collection efficiency
  - Solids stripping efficiency
  - Gas and solids RTD shape and peak spread

- Develop novel separator performance metrics that combine the performance characteristics listed above (Chapter 4)

- Understand the hydrodynamics of the gas-solids separation process and describe their relation to the gas and solids RTDs and to the separator performance (Chapters 3 and 4)

- Identify problematic flow structures (i.e. backmixing) (Chapters 3, 4, and 5)
• Identify optimum operating conditions for fast, efficient gas-solids separation with minimum backmixing (Chapters 3 and 4)
• Remedy problematic flow structures through optimization of the separator geometry, operating conditions, and by addition of solids stripping gas (Chapters 3 to 6)
• Demonstrate the effect of particle size on the gas-solids separator performance (Chapters 3, 4, and 5)
• Demonstrate the effect of stripping gas on separator performance (Chapter 6)

1.11 Notation

\[ D_{ax} \quad \text{Axial dispersion coefficient [m}^2\text{/s]} \]
\[ E(t) \quad \text{Residence time pulse distribution [--]} \]
\[ L \quad \text{Characteristic vessel length [m]} \]
\[ \text{Pe} \quad \text{Péclet number} \]
\[ t \quad \text{Time [s]} \]
\[ U_g \quad \text{Superficial gas velocity [m/s]} \]
\[ z \quad \text{Axial distance along reactor vessel [m]} \]
\[ \delta(t) \quad \text{Dirac delta function [--]} \]
\[ \sigma^2 \quad \text{RTD variance [s}^2\text{]} \]
Chapter 2

2 Experimental Equipment & Conventional Measurement Methods

Several conventional experimental methods used frequently in academic research and in industry were used in the following chapters of this thesis to complement the novel experimental gas RTD measurement method described in Chapter 3. These conventional methods were used to assess the separator performance in a comprehensive manner. Conventional methods were used for measurement of the separator pressure drop, separation zone volume, local gas velocity, local solids concentration, solids collection efficiency, solids RTD, and solids stripping efficiency in ICFAR’s cold model downer and gas-solids separator. In addition to conventional methods, the main experimental equipment used in this thesis is described in detail in the present chapter to provide one consolidated description of the repeatedly used equipment and conditions.

2.1 ICFAR’s Cold Model Downer & Gas-Solids Separator

In recent years, the Institute for Chemicals and Fuels from Alternative Resources (ICFAR) has been developing and testing reactor technology for the conversion of biomass and heavy oil feedstocks to useful bio-oil, bio-char, syngas, and other valuable chemical products via pyrolysis. Among the various biomass conversion processes developed at ICFAR, a downer reactor was designed for the pyrolysis of biomass and heavy oil feedstocks to maximize the liquid yield. The downer configuration was selected over other reactor types for careful control of thermal cracking reactions and gas-solids contact times.

To help achieve maximum liquid yield and careful control of cracking and contact time, a novel gas-solids separator was developed and tested (Huard, 2009; Huard et al., 2010b) in a full-scale cold model downer. The gas-solids separator was designed to integrate aspects of the circulating fluidized bed (CFB) reaction column exit, of primary gas-solids separation, and of product vapor recovery using stripping gas into one effective
separation-plus-stripping device. In order to assess the characteristics and performance of the integrated gas-solids separator in a comprehensive manner, and to optimize the design of the separator, the cold model downer was modified and supplemented with new instrumentation.

A 6.9 cm diameter ($D$), 134 cm tall ($L$), transparent acrylic cold model downer apparatus used in Huard (2009) and Huard et al. (2011) was modified for the work described in this thesis. Figure 2.1 shows a process and instrumentation diagram for the cold model downer and associated equipment. Figure 2.2 illustrates to scale the geometry of the downer apparatus and gas-solids separator with some of the internals and instrumentation used for the majority of the studies in this thesis. The vertical position of the separator could be adjusted such that the cone rim was a maximum of 10.5 cm above the gas outlet pipe to a minimum of 1.8 cm below the gas outlet, as illustrated in Figure 2.3. The downer was not equipped with a recirculation loop and was therefore operated in a once-through mode. The gas and solids outlets were located 99 cm and 134 cm below the downer inlet, respectively. The gas outlet diameter was 0.95 cm. Solids exiting the downer were collected in a cylindrical tank of diameter 20 cm and height 22 cm. The main changes to the system from Huard (2009) were positive pressure air delivery from a compressor (versus vacuum pressure delivery by an axial fan blower installed in the downer exhaust line) and far greater process instrumentation and control including converging-diverging nozzle gas mass flowrate controllers, flowmeters, pressure transducers, and data acquisition.
Figure 2.1 – Process and instrumentation diagram for the cold model downer
Figure 2.2 – Illustration of (a) the cold model downer, (b) gas-solids separator, and (c) top view of sheds and tracer sparger
Compressed air at room temperature was supplied to the apparatus for fluidization. The mass flowrate of air was controlled using a bank of three converging-diverging nozzles of various sizes (two at 0.20 cm diameter, one at 0.31 cm diameter) upstream of the downer. The air mass flowrate could be controlled up to 10 g/s, which resulted in a maximum superficial gas velocity of 2 m/s. A 750 W Omega AHP-7561 inline electrical heater was installed just downstream of the converging-diverging nozzles to heat the incoming air stream for local solids concentration measurements (whose procedure is explained in further detail below). Silica sand (particle density = 2650 kg/m$^3$, Sauter mean diameter = 180 μm, full particle size distribution shown in Figure 2.4) was delivered to the downer by pressurized gravity flow from a feed tank mounted directly above the downer up to a maximum flowrate of 100 g/s. This allowed operation of the cold model downer for roughly three minutes at the highest solids flowrate. The sand particle size distribution was measured using a Sympatec GmbH HELOS H2316 particle size analyzer. The gas and solids were mixed in a Y-shaped pipe fitting immediately upstream of the downer inlet. Solids escaping from the gas-solids separator were captured in a 1 μm mesh filter bag.

Figure 2.3 – Illustration of separation length: a) negative separation length with cone rim below height of gas outlet, b) zero separation length with cone rim at same height as gas outlet, c) positive separation length with cone rim above height of gas outlet
As shown in Figure 2.2, sheds were used to segregate the downer from the gas-solids separation zone, and were located 14 cm above the gas outlet. The sheds had a criss-cross pattern, consisting of two rows of three sheds in each row, as shown in Figure 2.2. The purpose of the sheds was to create gas jets entering the separation zone, thereby inducing strong mixing with the tracer injected immediately downstream, and to prevent gas recirculation back into the downer. In this way the sheds created an approximation to a true closed boundary condition essential to accurate RTD measurement (Levenspiel, 1999). Assuming that the gas mixing condition entering the separator was representative of most downers, an axial dispersion coefficient ($D_{ax}$) of 0.2 m$^2$/s can be assumed (Brust & Wirth, 2004). Over the range of superficial gas velocities resulting in fully turbulent gas flow in the downer (from a minimum of roughly $U_g = 0.8$ m/s), the dispersion number ($D_{ax}/U_gL_d$) at the sheds had a minimum value of around 0.1, where $L_d$ was the length of the downer. A dispersion number of 0.1 is characteristic of “intermediate” dispersion (Levenspiel, 1999), and is reasonable for gas flow. However, the assumption of $D_{ax} = 0.2$ m$^2$/s was quite conservative and so it is reasonable to assume that dispersion at the sheds was actually quite low.

### 2.2 Separator Pressure Drop Measurement

As will be shown in Chapter 3, the gas RTD was measured using a novel pressure-response method by correlating the transient separator pressure drop to the composition

![Figure 2.4 – Particle size distribution of sand used in the majority of experiments](image)
of a gas mixture of downer air plus helium tracer during a step injection. Differential pressure transducers (OMEGA® PX142-001DV & PX142-005DV) were used to measure the pressure drop across the gas tracer orifice plate flow meter and the gas-solids separator. The PX142-001DV transducer, located at P₄ as shown in Figure 2.5, was used to measure the orifice plate flow meter pressure drop and had a full scale range of 7 kPa. The PX142-005DV transducer, with pressure taps at P₁ and P₂ as shown in Figure 2.5, was used to measure the separator pressure drop and had a full scale range of 35 kPa. The separator pressure drop was measured between the tracer sparger and roughly 40 cm along the length of the gas outlet pipe directly upstream of the solids filter bag, as shown in Figure 2.5. An Omega PX181-100G5V gage pressure transducer (full scale range of 690 kPa), located at P₃ as shown in Figure 2.5, was used to monitor the absolute pressure in the separation zone. All pressure transducers had response times of 1 ms. Voltage signals produced by the pressure transducers and all other electronic equipment were sampled at 500 Hz by two National Instruments USB-6008 data acquisition cards.

Figure 2.5 – Locations of pressure transducers in the separator
2.3 Gas Tracer Injection

As shown in Figure 2.1, helium tracer was supplied from a compressed gas cylinder for the gas RTD measurement. The flow of helium tracer and tracer substitute air were controlled individually using converging-diverging nozzles (0.32 cm diameter) and McMaster-Carr (catalog no. 5489T411) solenoid valves. The flowrate of tracer and tracer substitute gas was measured by an orifice plate flowmeter installed in a 6 mm ID tracer injection line. Tracer was injected over the downer cross-section through a 5 cm diameter circular ring sparger with 18 downward-pointing 0.08 cm diameter holes spaced equally around the ring as shown in Figure 2.2. The sparger hole diameter was chosen to give a sparger pressure drop much greater than the separator pressure drop, which was assumed to give good distribution of tracer over downer cross-section.

2.4 Separation Zone Control Volume Measurement

The combined volume of the separator, solids tank (at various fill levels), and part of the gas outlet pipe was measured to provide a check of reasonableness of the measured gas RTD mean response time. This was done by injecting a small sample of air with measured initial pressure (~ 300 kPa) and known volume (1.05 L) into the sealed separation zone (occupied by air at ~ 100 kPa) and measuring the change in pressure in both the sealed separator and the sample volume vessel. The volume of the separation zone was then calculated by Avogadro’s Law,

$$V = \frac{\Delta P_{samp}}{\Delta P_{sep}} V_{samp},$$  \hspace{1cm} (2.1)

where $V$ is the separation zone volume,

$\Delta P_{samp}$ is the change in pressure of the sample volume,

$\Delta P_{sep}$ is the change in pressure of the separation zone, and

$V_{samp}$ is the sample volume.

The apparatus used for the control volume measurement experiments is shown in Figure 2.6. The sample volume was equipped with an Omega PX181-100G5V gage pressure transducer (full scale range of 690 kPa). The sample volume was sealed on one end by a manually-operated ball valve and by a normally-closed solenoid valve between the volume and the separation zone. The separation zone was sealed by the same solenoid
valve, a blind flange installed 25 cm above the separation zone, and a gate valve in the gas exhaust line. An on-off switch was used to control the sample volume injection into the separation zone. All pressure signals were connected to the data acquisition system described earlier.

2.5 Local Gas Velocity & Solids Concentration Measurement

2.5.1 Equipment

Local gas velocity measurements were made in the region between the gas outlet and the solids collection tank to characterize the gas hydrodynamics and to gain insight into the gas-solids separation and solids stripping phenomena. A 10 kΩ resistance, 0.12 cm bead diameter Murata NTC thermistor was used as a heat flux sensor to measure the local gas velocity and local solids loading in the region between the gas outlet and the solids collection tank. The thermistor, shown in Figure 2.7, was supplied with a constant voltage of 12 VDC, and had a measured response time of 0.14 s. Although the thermistor was very simple to implement and calibrate, its main disadvantage was that it could not distinguish the direction of gas flow. An OMEGA® K-type thermocouple was used in combination with the thermistor to measure the local temperature of the gas-solids mixture during local solids concentration experiments. The thermocouple was always
located 5 cm below the thermistor so as not to disturb the flow passing over the thermistor. The vertical position of both the thermocouple and thermistor could be adjusted between 8.5 cm to 26.4 cm below the gas outlet pipe, while both instruments were able to span the diameter of the downer, as shown in Figure 2.2.

![Figure 2.7 – Photograph of Murata NTC thermistor (from www.digikey.ca)](image)

### 2.5.1.1 Local Gas Velocity Method

Before measuring the local gas velocity using the thermistor, a calibration of thermistor resistance versus superficial gas velocity was performed at three temperatures ($T_\infty = 19.1 \, ^\circ\text{C}$, $T_\infty = 22.2 \, ^\circ\text{C}$, and $T_\infty = 25.0 \, ^\circ\text{C}$) spanning the full range of temperature observed in the actual runs. The calibration was performed with the thermistor located 90 cm downstream of the downer inlet at the pipe centerline. For fully turbulent flow, over the range of Reynolds number $2300 < \text{Re} < 7000$, the entrance length ($L_e$) for fully developed flow was estimated to be between 110 cm and 130 cm from the following expression (White, 2003):

$$L_e \approx 4.4D \text{Re}^{1/6}.$$  \hspace{1cm} (2.2)

Although the thermistor was located near the end of the predicted entrance region, the flow was assumed to be nearly fully developed. Furthermore, the purpose of the experiments was to estimate the volume occupied by non-stagnant gas below the gas outlet. As such, very accurate velocity measurements were not required.

A non-linear regression of thermistor resistance versus gas velocity was obtained by calibration, which allowed for measurement of the local gas velocity in the region below the gas outlet. The complete velocity measurement calibration procedure and results are
described in Appendix A. To measure the local gas velocity, the thermistor voltage \( V \) and resistance \( R \) were measured along with the local temperature using the thermocouple during steady state gas-only operation over a period of 30 s. These measurements were used to interpolate in the thermistor resistance versus gas velocity and temperature calibration regressions. The measurement time was sufficient to measure the average local gas velocity and associated fluctuations occurring over spans of several seconds.

2.5.1.2 Local Solids Concentration Method

Local solids concentrations were measured by comparing the difference in heat transfer from the thermistor to the flow media between gas-only and gas-plus-solids operations. During each local solids loading experiment, the downer air was pre-heated to \( \sim 50 ^\circ \text{C} \) while the solids were kept at room temperature in order to “amplify” the relative heat transfer between gas-only and gas-plus-solids operation. Similar techniques were used successfully by McMillan et al. (2006) and by Fushimi et al. (2012) to determine extents of liquid / solid and solid / solid mixing, respectively, in CFB downer units. In this thesis, heat transferred from the hot air to the cooler particles through contact and mixing in the downer. When the gas / solids mixture reached the thermistor located below the gas outlet, higher solids concentrations were indicated by greater changes in the measured thermistor power and temperature.

During each run, the initial, steady thermistor power \( Q_1 \) was first measured during gas-only operation. In gas-only operation, the thermistor was initially at some temperature \( T_{s1} \) close to the temperature of the heated air \( T_1 \sim 50 ^\circ \text{C} \). Room temperature solids \( T_2 \sim 20 ^\circ \text{C} \) were then injected along with the hot air. The thermistor power \( Q_2 \) during gas / solids injection was again measured, with the thermistor temperature changing to \( T_{s2} \) proportionally with the local solids concentration. Since the temperature of the gas and solids entering the unit was not controlled, the temperature of each phase entering the downer was not constant between runs. Therefore, it was necessary to normalize the change in thermistor power to the change in thermistor temperature during the run. The ratio of change in thermistor power to change in thermistor temperature due to the local concentration of solids was given by:
\[
\frac{\Delta Q}{\Delta T_s} = \frac{(\text{Gas/solids mixture thermistor power}) - (\text{Gas only thermistor power})}{(\text{Gas/solids mixture thermistor temperature}) - (\text{Gas only thermistor temperature})}
\]

\[
\frac{\Delta Q}{\Delta T_s} = \frac{Q_2 - Q_1}{T_{s2} - T_{s1}} = \frac{V_2^2/R_2 - V_1^2/R_1}{f(R_2) - f(R_1)},
\]

where \( \Delta Q \) is the change in thermistor power, and \( \Delta T_s \) is the change in thermistor temperature.

The thermistor surface temperature \( (T_s) \) was determined by interpolation from a thermistor resistance versus temperature calibration described in Appendix A. Larger values of \( \Delta Q/\Delta T_s \) indicated higher solids concentrations since total heat transfer increased with increasing solids concentration. Local solids concentration radial profiles were measured by adjusting the thermistor radial position in the range \(-1 < r/R_D < 1\) as shown in Figure 2.2(b).

### 2.6 Phosphorescent Pigment Particle Size Distributions

In order to determine the effect of particle size on the gas RTD, solids RTD, solids collection efficiency, and stripping gas efficiency, two particle size distributions (PSDs) were prepared by grinding fresh phosphorescent pigment used in the solids RTD experiments described below in Section 2.8. In this way, sand normally used in the downer was represented by the phosphorescent pigment ground to approximately the same average size. The pigment type was GTA LLC PLO-7A alkaline earth aluminate with particle density \( = 3600 \text{ kg/m}^3 \), which was significantly different from the density of the sand \( (2500 \text{ kg/m}^3) \). The HELOS analyzer used to measure the PSD of the sand required wet samples and thus could not be used with the phosphorescent pigment, which partially dissolved in water. Therefore, the full PSD of the pigment was obtained by sieve analysis. The fresh pigment had a mass mean size of around 800 μm. One half of the fresh pigment mass was ground to a large average size whose mass median size was \( \bar{d}_p = 620 \mu\text{m} \) (approximate Sauter mean diameter \( \bar{d}_{PSM} = 550 \mu\text{m} \)). The other half of the fresh pigment was ground to a small average size representative of the sand used in normal
downer operation. The small size pigment was ground to a mass mean size of $d_p = 220 \mu m$ (approximate Sauter mean diameter $d_{PSM} = 160 \mu m$).

Although the pigment particle density was significantly different than that of the silica sand, the pigment particle characteristics were deemed adequately similar to assume similar behavior between the two particle types. The calculated terminal velocity in still ambient air for the Sauter mean diameter of sand and of the small pigment PSD was 1.26 m/s and 1.36 m/s, respectively, giving a difference of 7%.

In order to determine the effect of particle size on the collection efficiency of fine particles smaller than 100 \(\mu m\), both the small and large PSDs were seeded with small glass beads ($d_{PSM} = 60 \mu m$) in the amount of 15\% of the total batch mass. The PSD of the glass beads is also shown in Figure 2.8.

![Figure 2.8 – Experimental pigment particle size distributions](image-url)
2.7 Solids Collection Efficiency

2.7.1 Overall Collection Efficiency

Huard (2009) described the method by which the overall solids collection efficiency was measured in this thesis. Since the downer was operated in a once-through mode, the total mass of a solids batch was measured prior to each solids collection efficiency experiment using either a Shimadzu UW2200H balance with a capacity of 2.200 kg and precision of 0.01g for small batches up to 2 kg or a 30 kg capacity balance with 0.1 g precision for larger batches up to 10 kg. The solids were loaded into the solids feeding tank while the downer was operating with gas only. A manually operated ball valve was used to inject solids from the tank while the time of injection was measured using a hand held timer. The filter bag mass was measured prior to and after each run on the Shimadzu balance to determine the mass of escaped solids.

2.7.2 Grade Efficiency Measurement

To determine the collection efficiency of specific particle size ranges (i.e. grade efficiency), the size distributions of the solids loaded into the feeder tank and of the “escaped” solids captured in the 1 μm mesh size bag filter were measured and compared. The bag filter was manually shaken gently to dislodge any particles trapped in the filter cloth until the mass of the bag filter was constant. Although this method was effective at retrieving greater than 90 % of the mass of the “escaped” solids, a small amount of particles were permanently trapped in the filter cloth. The HELOS particle size analyzer was used to measure the PSD of representative samples of ~ 60 μm glass beads added to the bulk sand or pigment batches. Sieve analysis was used to measure the PSD of representative samples of the phosphorescent pigment. Representative samples of the total injected solids batch and of the escaped solids were obtained using a Humboldt riffle-type splitter. Knowing the masses and PSDs of the injected and captured “escaped” particles, the collection efficiency of each particle size range was calculated.
2.8 Solids RTD Measurement

The solids RTD was measured using the phosphorescent pigment tracer technique used in several previous studies (e.g. Roques et al., 1993, Wei et al., 1994, Huang et al., 2006). This method was deemed to provide the most reasonable compromise between RTD accuracy, ease of implementation, and cost based on the literature review discussed in Chapter 1.

2.8.1 Solids RTD Equipment

To measure the solids RTD, the sheds and tracer gas sparger were removed to prevent particles from rebounding upward as they entered the gas-solids separator. The small phosphorescent pigment PSD described in Section 2.6 was used as tracer to substitute for the sand normally used in the cold model downer. The PSDs of the tracer pigment and sand are compared in Figure 2.9. The difference in the Sauter mean diameter between the pigment and sand was roughly 12%, though as noted above, the difference in terminal velocity between the two particle types was only 7%.

![Figure 2.9 – Comparison of the particle size distributions between silica sand and phosphorescent tracer pigment](image)

Two camera flash heads were mounted opposite each other across the downer diameter and were used to activate tracer entering the downer. The flash heads were mounted
external to the transparent downer wall. As shown in Figure 2.10, the flash heads were mounted 7 cm below the downer inlet. The light emitted by the flash heads was collimated into a roughly rectangular beam 2 cm high by 10 cm wide as illustrated in Figure 2.10. The resulting activation area in the downer was nearly a cylindrical slice of average thickness slightly larger than 2 cm.

Tracer was detected using Futurlec GL105 10 kΩ resistance, light-active photocells. The photocell response time was 60 ms. In order to track the bulk trajectory of tracer particles at numerous locations along the downer, sets of photocells were mounted at five locations.
along the height of the downer, as shown in Figure 2.11. At each vertical position, four photocells, all connected in series in a ring, were mounted around the downer circumference at 90° angles to provide tracer detection over the entire cross-section. With all photocells in a ring connected in series, an analog average of the emitted light from activated tracer was measured by the photocell ring. All photocells connected in a ring were also connected in series with an external resistor used to measure the combined resistance of the photocell ring. Each photocell ring circuit was powered by 12 VDC. Voltage signals from each photocell ring and their associated external resistors were sampled at 5 ms intervals. Two photocell rings were mounted upstream of the gas-solids separator, at \( z = 69 \) cm and 87 cm below the downer inlet, respectively. Three photocell rings were mounted between the gas and solids outlets at \( z = 105 \) cm, 118 cm, and 133 cm from the downer inlet.

![Figure 2.11 – Illustration of downer showing vertical position and circumferential arrangement of photocell tracer detectors (downer not shown to scale)](image-url)
2.8.2 Experimental Conditions & Procedure

Solids RTD experiments were performed over the ranges of superficial gas velocities $U_g = 0.75 \text{ m/s}$ to $U_g = 1.4 \text{ m/s}$ and solids loading ratios $\frac{m_s}{m_g} = 1.8$ to $\frac{m_s}{m_g} = 46$. The cone separation length was kept constant at $L_s/D = 0$ (see Figure 2.3) since it is shown later not to have a significant impact on the gas RTD in Chapter 3 nor on the solids RTD in Chapter 5. During a solids RTD experiment, steady gas and solids flow were first achieved prior to tracer activation. Tracer pulses were then activated repeatedly in roughly five second intervals to give several (typically four or five) replicates per run. The typical residence time of solids in the unit was less than one second, and it will be shown in Chapter 5 that the tracer phosphorescence decayed to less than 1 % of its initial activity within one second. Thus, a five second replicate interval was more than sufficient to safely assume that any activated tracer from the previous replicate had either left the system or was no longer significantly active to affect the measurement.

2.9 Solids Stripping Experiments

To demonstrate the effect of an integrated separator / stripper device on separator performance, the cold model downer was augmented with additional equipment and instrumentation to perform stripping gas experiments using carbon dioxide as tracer. Figure 2.12 shows the schematic diagram of the apparatus used for the stripping gas experiments. CO$_2$ tracer was used to measure the stripping efficiency, which is defined in detail later in Chapter 6, and is essentially a comparison of the concentration of CO$_2$ at the downer inlet where tracer was injected to the solids outlet, where the presence of CO$_2$ indicated poor stripping.

CO$_2$ tracer was delivered to the downer from a pressurized cylinder, whose flowrate was controlled using a calibrated converging-diverging nozzle and a normally closed solenoid valve. CO$_2$ tracer was injected over the downer cross-section using a 5 cm diameter circular sparger ring with 18 upward-pointing 0.08 cm diameter holes spaced equally around the ring. The CO$_2$ sparger ring was located 9 cm downstream of the downer inlet, as shown in Figure 2.13. An inexpensive CO$_2$ detector (CO2Meter, Inc. K-30 10,000 ppm) was installed in a 6 mm diameter gas sampling line located 3 cm upstream of the
solids collection tank. The sample line tip was located at the downer radial centerline, as indicated in Figure 2.13. The CO₂ detector response time was 20 s, and the CO₂ concentration was sampled at 0.1 s intervals. Although the CO₂ detector was inexpensive and very simple to implement with no detector calibration required, its very slow response time prevented its additional use for gas RTD experiments. The full scale response of the detector was 1 % CO₂ by volume; therefore, the injected CO₂ volume flowrate was always kept to less than 0.5 % of the total downer air plus stripping air flowrates. The resolution of the CO₂ detector was about 0.1 % of the full scale response, i.e. 10 ppm.

Figure 2.12 – Schematic of stripping gas experimental apparatus and instrumentation
During stripping experiments, stripping air was injected over the downer cross-section using a 5 cm diameter circular sparger ring with 18 horizontal-pointing 0.08 cm diameter holes spaced equally around the ring. The vertical position of the stripping gas sparger could be adjusted between 8 cm to 30 cm below the gas outlet, as shown in Figure 2.13. The helium tracer sparger and gas velocity thermistor and thermocouple were used in this study with the same configurations described in Sections 2.3 and 2.5.

Solids collection efficiency, gas RTD, and solids RTD experiments were performed to determine the overall impact of stripping gas on separator performance. Silica sand of the
same type and size described in Section 2.1 was used for the solids collection efficiency and gas RTD experiments with stripping gas. The small pigment PSD was used for the solids RTD experiments. The superficial gas velocity was varied between 0.6 m/s to 1.2 m/s for all stripping experiments. During stripping experiments with solids, the solids mass flowrate was varied between \( \dot{m}_s = 0.007 \) kg/s and \( \dot{m}_s = 0.118 \) kg/s, which corresponded to solids-to-gas loading ratios of \( \dot{m}_s / \dot{m}_g = 1.1 \) to 31.

### 2.10 Notation

- **D**: Vessel diameter [m]
- **\( L_e \)**: Velocity entrance length [m]
- **\( P_{samp} \)**: Pressure of sample volume [Pa]
- **\( P_{sep} \)**: Pressure of separation zone [Pa]
- **\( Q \)**: Thermistor power [W]
- **\( Q^* \)**: Temperature-specific change in thermistor power [W/K]
- **\( R \)**: Thermistor resistance [\( \Omega \)]
- **Re**: Reynolds number [--]
- **\( T_s \)**: Thermistor surface temperature [K]
- **\( V \)**: Thermistor voltage [V]
- **\( \mathcal{V} \)**: Separation zone volume [m\(^3\)]
- **\( \mathcal{V}_{samp} \)**: Sample volume [m\(^3\)]
Chapter 3

3 Gas RTD Measurements in a CFB Downer Gas-Solids Separator

A unique, simple, fast, and cost-effective pressure response-based technique was developed to measure the gas residence time distribution (RTD) for the gas-solids separator of ICFAR’s CFB downer reactor. Heat flux sensing thermistors were also used to measure the local gas velocity and local solids concentration in the separator in order to provide additional information on the gas backmixing and gas-solids separation phenomena.

3.1 Introduction

Several gas phase RTD studies have previously been performed in circulating fluidized bed (CFB) riser reactors (e.g. Dry & White, 1989; Patience & Chaouki, 1993; Vandewalle et al., 2002). Most authors reported significant gas backmixing in the riser, decreased backmixing with increasing gas velocity, and increased backmixing with increasing solids flux. Only Gauthier (1991) and Brust & Wirth (2004) measured gas backmixing in a downer reactor and found that backmixing was reduced at high gas velocities. Brust & Wirth further demonstrated that the gas phase in the downer was much closer to ideal plug flow than in the riser in their CFB unit under identical conditions.

Previous RTD studies in CFB reactors have often neglected the impact of the reactor outlet and gas-solids separator on gas backmixing in the unit. However, Vandewalle et al. (2003) showed that the extent of gas backmixing in a riser depended mainly on whether gas adsorption on the solids phase was possible or not possible. When non-adsorbing

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1 A version of this chapter has been presented orally at The 14th International Conference on Fluidization and published in the conference proceedings as follows:

inert sand was used, gas recirculation in the riser was minimized and the gas behaved in essentially plug flow. However, when gas adsorption was possible on “active” catalyst, significant gas refluxing accompanied the usual solids refluxing at the riser exit. When compared to the more traditional CFB risers, CFB downers benefit from a nearly plug flow behavior of both gas and particles, and are attractive for processes such as heavy oil or biomass pyrolysis, where backmixing would result in the overcracking of valuable products. It is, therefore, essential that the gas-solids separator, at the downer exit, does not introduce major backmixing of gas or particles. Since the effect of downer exit and gas-solids separation on overall unit hydrodynamics has not been investigated, there is both a need and opportunity to investigate the gas RTD in ICFAR’s downer gas-solids separator.

Huard et al. (2011) demonstrated a hot wire anemometer measurement technique to detect helium tracer in a gas-only downer. This hot wire method could not be adapted successfully for a multiphase system with solids. Despite measures taken to minimize the effect of gas sampling lines used to protect the hot wires, significant gas backmixing and tracer velocity fluctuations occurred in the sampling lines, which prevented accurate measurement of the downer and separator pulse response. Therefore, a simple, robust pressure based technique has been developed to measure the separator response to a helium tracer downstep. A related pressure based technique was developed concurrently by Morales (2013) to monitor liquid vaporization in a laboratory scale experimental model of the fluid coking process. However, no similar technique has been applied in CFB units in the literature.

There were two distinctive advantages to use of the pressure-response based gas RTD method:

1. The measurements were extremely simple and inexpensive to implement since only pressure transducers and flowmeters were required for instrumentation. This is compared especially to the complexity and cost of radioactive gas tracer, which otherwise gives the most accurate gas RTD measurements.

2. The pressure measurements were global and represented the overall system response over the entire inlet and outlet boundaries, thereby circumventing issues
related to tracer sampling. This is compared especially to sampling using thermal conductivity, gas chromatography, and mass spectrometry tracer detection, in which significant tracer dispersion is possible and likely in tracer sampling lines. These other sample-based detection techniques are the most commonly used in the literature.

The primary disadvantage of the pressure response method was that significant calibration was required to accurately determine the concentration of helium tracer in the bulk downer air and solids mixture.

Given the needs for a better gas RTD measurement technique and for investigation of the effects of the downer exit on gas backmixing and hydrodynamics, the pressure response based method was developed and applied in the present chapter. This method was used to measure the gas RTD and backmixing in the downer reactor and around the gas-solids separator with and without solids. The main objectives of the present chapter were to describe and demonstrate:

- The novel pressure-response-based gas RTD measurement technique,
- Local gas velocity measurement using a thermistor,
- Estimation of local solids concentration using a thermistor,
- RTD modelling using a “reverse deconvolution” algorithm, and
- Experimental results using all techniques listed above for one selected gas-solids separator design.

### 3.2 Background

#### 3.2.1 Gas RTD Modelling

Previous RTD studies in CFB downer units have typically used a form of the axial dispersion model, e.g. Brust & Wirth (2004), to describe both the gas and solids RTDs in the downer. However, the impact of the gas-solids separator on the overall downer RTD has not been investigated. In preliminary experiments, the two-parameter axial dispersion model did not provide a good fit of tracer pulse response data measured in the gas-solids separator. Instead, combinations of several two-parameter, asymmetrical peak
Exponential distributions $E_i(t)$ were found to fit the experimental data more closely, and were given by:

$$E_i(t) = 4a_i \exp\left[-2a_i(t-t_{0i})\right]\left[1-\exp\left[-2a_i(t-t_{0i})\right]\right], \quad (3.1)$$

where $a_i$ is the peak amplitude,

$t$ is time, and

$t_{0i}$ is the initial peak time, i.e. the time at which the signal starts increasing above its baseline of 0.

Note that this form assumes that the area enclosed by $E_i(t)$, $A_{Ei}$, is unity, as for a true RTD (Levenspiel, 1999).

Experimental RTD data can be fitted with increasing accuracy by summing several weighted expressions $E_i(t)$ of the form given in Equation (3.1):

$$E(t) = \sum_{i=1}^{N} \alpha_i E_i(t), \quad (3.2)$$

where $\alpha_i$ is the weight factor for the peak function $E_i(t)$. To preserve the condition $A_E = 1$, all $\alpha_i$ must sum to unity, since $A_{Ei} = 1$ for all $E_i(t)$. The weight factor $\alpha_N$ for the last ($N^{th}$) term $E_N(t)$ in Equation (3.2) can be written in terms of the other weight factors:

$$\alpha_N = 1 - \sum_{j=1}^{N-1} \alpha_j. \quad (3.3)$$

In this thesis, combinations of up to three exponential distributions ($N \leq 3$) were found to fit the experimental data with sufficient accuracy.

For use in downstep response experiments, it can be shown that the normalized discrete cumulative distribution $F(t_j)$ at time $t_j$ is related to the discrete probability distribution $E(t_j)$ by:

$$F(t_j) = F(t_{j-1}) - \frac{E(t_j)}{\sum_{j=1}^{\infty} E(t_j)} ; \quad 0 \leq j \leq \infty, \quad F(t_0) = 1 \quad (3.4)$$

Helium tracer concentrations were measured at two locations in the separator to give an inlet tracer concentration signal, $X(t)$, and an outlet tracer concentration signal, $Y(t)$. The
outlet signal was related to the inlet signal through discrete numerical convolution with the RTD, $E(t)$:

$$Y(t) = (X * E)(t),$$  \hspace{1cm} (3.5)$$

where $*$ indicates the convolution operation. The discrete numerical convolution $(X * E)(t)$ is defined as:

$$(X * E)[t] = \sum_{\theta=0}^{\infty} X[\theta]E[t-\theta] = \sum_{\theta=0}^{\infty} X[t-\theta]E[\theta],$$  \hspace{1cm} (3.6)$$

where $\theta$ was a dummy variable required to reflect and shift one of the independent variables. Qualitatively, the convolution of $X(t)$ with $E(t)$ effectively “smears” and elongates $X(t)$, transforming it into the outlet concentration signal, $Y(t)$. In this way $E(t)$ describes the overall state of mixing between the two measurement points.

In each experiment, the inlet and outlet tracer concentrations, $X(t)$ and $Y(t)$, were measured, from which $E(t)$ was solved numerically by a “reverse deconvolution” algorithm. Deconvolution, as described by Brust & Wirth (2004), was not used in this study because of its tendency to be unstable and very sensitive to input signal noise. Instead, by reverse deconvolution, the residual error between the fitting function, $Y_{fit}(t)$, and the experimental signal, $Y_{exp}(t)$, was minimized by adjusting the parameters for $E(t)$ in Equations (3.1) and (3.2) using an iterative solver. The disadvantages of reverse deconvolution are that the assumption of a certain expression form for $E(t)$ may not accurately represent all of the observed experimental RTDs, as well as longer computational time. Figure 3.1 outlines the basic steps of the reverse deconvolution algorithm.

Figure 3.1 – Flow chart describing the RTD reverse deconvolution algorithm
In this chapter, as in most RTD studies, the mean residence time (i.e. response time) and standard deviation of the pulse response function, $E(t)$, were used to quantify the state of mixing in the vessel. The mean response time ($\tau$) was given by:

$$
\tau = \frac{\int_0^\infty t \cdot E(t) dt}{\int_0^\infty E(t) dt}.
$$

(3.7)

The standard deviation of the RTD was given by:

$$
\sigma = \sqrt{\frac{\int_0^\infty (t - \tau)^2 E(t) dt}{\int_0^\infty E(t) dt}}.
$$

(3.8)

Since the control volume of the vessel was not fixed and varied with the operating conditions, as will be discussed in detail in Sections 3.4.1.5 and 3.4.1.6, different mean residence times were possible at the same gas flow rate. Therefore, the coefficient of variation (COV) was used instead of the standard deviation as a measure of the spread of the RTD. In this way the spread of RTDs with significantly different mean residence times could be compared fairly. The coefficient of variation was given by:

$$
\text{COV} = \frac{\sigma}{\tau}.
$$

(3.9)

### 3.3 Experimental

#### 3.3.1 Cold Model Downer Apparatus

The 6.9 cm diameter ($D$), 134 cm tall ($L$), transparent acrylic cold model downer apparatus described in Chapter 2.1 was used in the present chapter. The sole gas-solids separator design used in this chapter consisted of a 6.3 cm base diameter, $60^\circ$ total internal angle hollow cone separator mounted above the gas outlet pipe, as shown in Figure 3.2. The vertical position of the separator could be adjusted such that the cone rim was a maximum of 10.5 cm above the gas outlet pipe to a minimum of 1.8 cm below the gas outlet. This vertical position was defined as the separation length ($L_s/D$).
3.3.2 Experimental Conditions & Procedure

3.3.2.1 Gas RTD Experiments

During each downstep experiment, helium tracer was injected steadily through the tracer sparger then cut off sharply in a downstep into the bulk air flow in the downer. Helium was chosen as tracer due to its significant density difference with air at room temperature. This difference resulted in a different separator pressure drop when an equivalent portion of the bulk air was replaced completely by helium, which was easy to detect. In the current chapter, tracer was detected by measuring the corresponding change in the
pressure drop of the bulk flow across the gas-solids separator, i.e. between locations P\textsubscript{1} and P\textsubscript{2} as shown in Figure 2.5 due to the change in gas composition. The gage pressure was also measured for verification at location P\textsubscript{3}, as shown in Figure 2.5.

The steady state concentration of helium during tracer injection was 5 vol.% of the combined total gas flowrate, which was assumed to have a small, insignificant effect on the downer hydrodynamics. To verify this assumption, the gas RTDs for steady state helium concentrations of 3 vol.%, 5 vol.%, and 7 vol.% at $U_g = 0.80 \text{ m/s}$ were compared. The mean residence time and standard deviation of the pulse responses varied less than 10% between runs, which was deemed adequate. Gas RTD experiments were performed over the range of superficial gas velocities $U_g = 0.15 \text{ m/s}$ to $U_g = 1.2 \text{ m/s}$, which covered the full range of laminar to fully turbulent flow regimes. The solids loading was varied from zero to $\dot{m}_s/\dot{m}_g = 15 \text{ kg/kg}$. The separation length was also varied over $-0.2 \leq L_s/D \leq 1.5$.

3.3.2.2 Gas RTD Numerical Procedure

An iterative solver was developed in Visual Basic to solve automatically for the transient air and tracer mole balances, transient tracer injection, and gas RTD fitting parameters by the reverse deconvolution algorithm. For each gas RTD experiment, the routine printed and graphed the results in a Microsoft Excel spreadsheet to provide visual evidence of a reasonable solution. The code for the iterative solver is provided in Appendix G.

3.4 Results

3.4.1 Gas Mixture Composition Modelling

3.4.1.1 Introduction

A brief introduction to the various components of the gas mixture composition modelling is required, as the model is complex and follows several steps. The solution steps followed in the gas mixture composition modeling and other associated tasks are shown as part of the overall reverse deconvolution algorithm in Figure 3.3. At steady state conditions, the molar flowrates of the downer air and helium tracer at the separator inlet and outlet were measured constants. However, during a helium tracer downstep injection,
the transient molar flowrates of air, \( n_{\text{air},o}(t) \), and helium, \( n_{\text{he},o}(t) \) at the gas outlet were unknown, and therefore represented two unknown variables. These two variables were solved in two equations, namely the tracer mole balance and the separator pressure drop empirical model. The development of these two equations is discussed in detail later in this section.

As shown in Figure 3.3, the primary measured inputs in the algorithm were the tracer input signal, \( X(t) \), the separator pressure drop (\( \Delta P_{\text{sep}} \)), and the control volume (\( V \)):

- The tracer input signal in this case was the injected helium tracer molar flowrate, \( \dot{n}_{\text{he},i}(t) \), which was fitted by the tracer injection model, as will be shown in Section 3.4.3.
- The separator pressure drop was modeled as a linear sum of the contributions of the flowrates through the separator exhaust of air, helium, and solid particles (Section 3.4.1.3). Separate calibration experiments were performed to determine the empirical model parameters.
- The control volume was difficult to determine a-priori since it depended on variables such as the level of solids in the separator tank. Separate experiments were conducted to determine the control volume (Section 3.4.1.5).

The separator pressure drop and the control volume were used to calculate the experimental tracer output signal, \( Y_{\text{exp}}(t) \), through iterative solution of the tracer mole balance and implicitly formulated separator pressure drop empirical model (Figure 3.3). The tracer output signal was taken as the helium tracer molar flowrate at the gas outlet, \( \dot{n}_{\text{he},o}(t) \). Convolution of the injected tracer molar flowrate, \( \dot{n}_{\text{he},i}(t) \), with the RTD, \( E(t) \), calculated to obtain the fitted outlet tracer molar flowrate, \( Y_{\text{fit}}(t) \). This study used an equation with adjustable parameters for the RTD, \( E(t) \), as shown in Equation (3.1). These adjustable parameters (\( a_i, a_i \) and \( t_{0i} \) in Figure 3.3) were optimized iteratively in the reverse deconvolution algorithm to fit \( Y_{\text{exp}}(t) \), which was equal to \( \dot{n}_{\text{he},o}(t) \).
Figure 3.3 – Gas RTD reverse deconvolution algorithm showing gas mixture composition model and other associated steps (highlighted in grey)
3.4.1.2 Tracer Mole Balance

During helium tracer downstep injections, a constant molar flowrate of helium injected at the entrance to the separation zone was suddenly stopped to measure the transient response of the separator pressure drop to the step injection. Figure 3.4 illustrates the separation zone and control volume used for the pressure drop analysis. A tracer mole flowrate balance written for a control volume consisting of the separation zone, the solids collection tank, and the gas outlet pipe with one inlet at the height of the tracer sparger and one outlet in the gas outlet pipe was given by

\[ \dot{n}_{\text{tot}} = \dot{n}_{\text{He,o}} + \dot{n}_{\text{air,o}} = \dot{n}_{\text{He,i}} + \dot{n}_{\text{air,i}} - \dot{n}_{\text{accum}} = \dot{n}_m - \frac{dP_{\text{sep}}}{dt} \frac{\mathcal{V}}{RT_\infty}, \]  

(3.10)

where \( \dot{n}_{\text{tot}} = \dot{n}_{\text{He,o}} + \dot{n}_{\text{air,o}} \)

was the total molar flowrate at the gas outlet [mol/s],

\( \dot{n}_m = \dot{n}_{\text{He,i}} + \dot{n}_{\text{air,i}} \)

was the total molar flowrate at the separation zone inlet [mol/s],

\( \dot{n}_{\text{accum}} = \frac{dP_{\text{sep}}}{dt} \frac{\mathcal{V}}{RT_\infty} \)

was the total molar flowrate accumulation in the separation zone due to the transient separator pressure change [mol/s],

\( P_{\text{sep}} \) was the absolute pressure in the separation zone [Pa],

\( t \) was the time [s],

\( \mathcal{V} \) was the control volume [m³],

\( R \) was the ideal gas constant [J/kg/mol], and

\( T_\infty \) was the ambient temperature [K].

Since the pressure drop across the separation zone was measured, it was preferable to express Equation (3.10) in terms of the separator pressure drop (\( \Delta P_{\text{sep}} \)). \( P_{\text{sep}} \) was interchangeable with \( \Delta P_{\text{sep}} \) since the two were measured simultaneously and were confirmed to change at the same rate with respect to time. Therefore,
\[
\dot{n}_{\text{tot}} = \dot{n}_{\text{in}} - \frac{d(\Delta P_{\text{sep}})}{dt} \frac{V}{RT_{\infty}}.
\]  

(3.11)

\[
\Delta P_{\text{sep}}(t) = \Delta P_{\text{air}} + \Delta P_{\text{He}} + \Delta P_{s},
\]  

(3.12)

where \( \Delta P_{\text{air}} \) was the contribution to the total separator pressure drop from air, \( \Delta P_{\text{He}} \) was the contribution to the total separator pressure drop from helium tracer, and

3.4.1.3 Separator Pressure Drop Regression Model

To determine the transient concentration of tracer in the gas outlet, and thereby obtain the gas RTD, an extensive calibration was performed for the steady-state separator pressure drop versus steady air, helium, and solids flowrates. A non-linear regression with one adjustable parameter was then developed to fit the calibration data. The regression assumed that the separator pressure drop could be treated as a linear sum of contributions from each of the components in the gas / solids mixture in the following manner:

Figure 3.4 – Illustration (to scale) of the maximum possible control volume and inlet and outlet gas flows used in the separator pressure drop modelling
\( \Delta P_s \) was the contribution to the total separator pressure drop from solids, which encompassed gas-particle, particle-particle, and particle-wall friction losses.

Using this approach, validated below, the separator pressure drop regression was built starting from the separator pressure drop for single phase air-only flow, and was expanded with increasing complexity by adding contributions from each new component in the mixture.

Figure 3.5(a) shows the experimental single phase air-only steady separator pressure drop (\( \Delta P_{air} \)) and regression fit. The data were fitted very well by an expression of the form

\[
\Delta P_{air} = C_1 \frac{n_{air}^2 M_{air}}{P_{sep}},
\]

(3.13)

where \( C_1 \) was an empirical coefficient determined from the calibration, \( n_{air} \) was the mole flowrate of air, and \( M_{air} \) was the molecular weight of air.

The empirical coefficient (\( C_i \)) was used to tune Equation (3.16) to the calibration data, and was found to be specific to the design of the separator. It is shown in the subsequent chapter that \( C_i \) had a specific value for each tested candidate separator design.
Figure 3.5 – Experimental separator pressure drop and regression model fit for: (a) single phase air, (b) single phase air plus helium, and (c) dual phase air plus solids
Figure 3.5(b) shows the experimental contribution from helium tracer to the total separator pressure drop in air plus helium flow. The concentration of helium in the calibration tests was varied between 0 vol.% and 8 vol.%, which spanned the range of helium concentrations used in the gas RTD tests. The calibration data were fitted well by an expression of the form

$$\Delta P_{He} = C_2 \left( \dot{n}_{air} M \right)^{\gamma_2} \dot{n}_{He}^{\gamma_3},$$

where $C_2$, $\gamma_2$, and $\gamma_3$ were empirical fitting parameters determined from the calibration, $\dot{n}_{He}$ was the mole flowrate of helium, and $M$ was the gas mixture average molecular weight.

Separate variables were used for the flowrates of helium and air, $\dot{n}_{He}$ and $\dot{n}_{air}$, respectively, instead of the total gas flowrate combined with gas mixture properties, for the sole reason that the transient flowrates of each component did not change at the same rate with respect to time through the downstep experiment. Therefore, each component had to be treated as separate dependent variables. A single-equation separator pressure drop model using only mixture properties was originally developed which accurately predicted the separator pressure drop but was unsuccessful at predicting complex transient flow phenomena in the separator. The gas mixture average molecular weight ($M$) in Equation (3.14) was calculated from

$$M = xM_{He} + (1 - x)M_{air},$$

where $M_{He}$ was the molecular weight of helium, and $x$ was the mole fraction of helium.

The empirical fitting parameters $C_2$, $\gamma_2$, and $\gamma_3$ were found to be nearly constant under all conditions and for all separator types, as shown in the following chapter. The experimental data were fitted best with empirical exponents $\gamma_2 = 1.36$ and $\gamma_3 = 0.75$. The helium contribution to the separator pressure drop was roughly one order of magnitude less than the contribution from the carrier air since the helium concentration was kept less than 10 % of the total gas flow. The results of Figure 3.4(b) demonstrate that the assumption that the separator pressure drop could be treated as a linear sum of contributions from each of the components was valid for air/helium mixtures.
Figure 3.5(c) shows the experimental contribution from solids to the total separator pressure drop in air plus solids flow. The solids loading was varied between 0 and 16 kg/kg, which spanned the range of solids loading used in the gas RTD tests. The calibration data were fitted well by an expression of the form

$$\Delta P_s = (C_4 \dot{n}_{air} M + C_5) \dot{m}_s^{\gamma_s},$$

where $C_4$, $C_5$, and $\gamma_s$ were empirical fitting parameters determined from the calibration, and

$\dot{m}_s$ was the mass flowrate of solids.

The solids contribution to the total separator pressure drop increased linearly with the air mole flowrate, and was roughly one order of magnitude less than the contribution from the bulk air flow.

Summing Equations (3.13), (3.14), and (3.16), the total steady-state separator pressure drop was given by

$$\Delta P_{sep} = C_4 \dot{n}_{air}^2 \frac{M}{P_{sep}} + C_5 \dot{n}_{air} M \dot{n}_{He}^{\gamma_s} \dot{n}_{air}^{\gamma_s} + (C_4 \dot{n}_{air} M + C_5) \dot{m}_s. \quad (3.17)$$

Each term in the total separator pressure drop in Equation (3.20) was a function of the air mole flowrate. Furthermore, $M_{air}$ in Equation (3.16) was changed to $M$ to reflect the fact that the gas was composed of two species during all gas RTD experiments. Since the experimental transient air mole flowrate ($\dot{n}_{air}(t)$) was unknown aside from its steady-state values, Equation (3.17) was rearranged to solve for $\dot{n}_{air}(t)$ implicitly by iteration.

To account for transient effects not encompassed by the steady-state separator pressure drop regression, $\dot{n}_{air}$ was split into two terms: a constant steady-state air mole flowrate ($\dot{n}_{air,ss}$) and a transient air mole flowrate ($\dot{n}_{air,tr}$). One adjustable fitting parameter ($C_{t,tr}$) was introduced in Equation (3.17) to achieve proper agreement with the experimental transient separator pressure drop during gas RTD runs, and the final separator pressure drop model was given by

$$\Delta P_{sep} = \left(C_1 \dot{n}_{air,ss}^2 + C_{1,ss} \dot{n}_{air,ss}^2 \right) \frac{M}{P_{sep}} + C_5 \dot{n}_{air} M \dot{n}_{He}^{\gamma_s} \dot{n}_{air}^{\gamma_s} + (C_4 \dot{n}_{air} M + C_5) \dot{m}_s. \quad (3.18)$$
Since \( \dot{n}_{air,ss} \) was constant, no new variables were introduced when splitting \( \dot{n}_{air} \) into two terms. Furthermore, since the total separator pressure drop was dominated by the contribution by the downer air, it was not helpful to split the helium flowrate, \( \dot{n}_{He} \), into steady-state and transient components.

### 3.4.1.4 Mixture Composition Model

In each gas RTD experiment, the tracer mole balance and separator pressure drop, given by Equations (3.11) and (3.18), were solved simultaneously for the unknown transient gas flowrates \( \dot{n}_{air}(t) \) and \( \dot{n}_{He}(t) \). These equations, taken together, were defined as the gas mixture composition model. At each time step, the mixture composition at the separator outlet was assumed to be in a new pseudo-steady-state condition, which allowed for the use of Equation (3.18). The mixture composition model was subject to the boundary conditions:

\[
\begin{align*}
\dot{n}_{He}(t = 0) &= \dot{n}_{He,1} = \text{constant} \\
\dot{n}_{He}(t = \infty) &= 0 \\
\dot{n}_{air}(t = 0) &= \dot{n}_{air}(t = \infty) = \dot{n}_{air,ss} = \dot{n}_{air,1} = \text{constant}.
\end{align*}
\]

Figure 3.6 shows a typical mixture composition model solution for a sample downstep RTD experiment without solids. Figure 3.7 shows a typical mixture composition model solution for a sample downstep RTD experiment with solids. The separator pressure drop predicted by the mixture composition model fit the experimental data very well in all runs. The experimental separator pressure drop shown in Figure 3.6(a) exhibited the following features:

1. A sharp drop starting at \( t = 0 \) that corresponded to the abrupt termination of helium tracer flow, then
2. A dip below the final steady-state value for a short period of time, and
3. A recovery to the final steady-state value.

This behavior was observed in several instances and was successfully predicted by the mixture composition model. However, in most cases, as shown by example in Figure 3.7, the separator pressure drop simply decreased steadily without a dip below the final steady-state value.
The mixture composition model was able to account for all observed flow events, including the complex situation when the total gas mixture molar flowrate reached steady-state before all of the helium tracer was swept from the control volume after the downstep (as shown in Figure 3.6) due to very poor mixing. The example mixture composition model solution shown in Figure 3.6 predicted the following events during the gas RTD run:

1. A rush of air from the downer into the separation zone (i.e. control volume) in the first moments after the downstep occurred. This was due to a sudden decrease in pressure in the separation zone relative to the downer when the downstep occurred. Although the air flowrate temporarily surged, the total gas mixture flowrate at the outlet always decreased toward the final steady-state value. Finally, while the rush of air occurred, the outlet tracer flowrate decreased since the flow of tracer at the separator inlet had been terminated. The initial rush of air from the downer likely introduced an error into the measured RTD, which demonstrated one limitation of the pressure response method. It may be possible to eliminate this error by conducting downstep experiments with different tracer to downer gas flowrate ratios, and extrapolating the results to a downstep size of zero.

2. The total gas mixture flowrate at the gas outlet continued to decrease and reached steady state. Meanwhile, the air flowrate at the gas outlet also decreased and reached its steady-state value, but did not necessarily settle there.

3. In some instances, such as shown in Figure 3.6, tracer was accumulated in stagnant regions such as the solids collection tank due to bypassing and poor mixing. In this situation, after the total gas mixture flowrate at the gas outlet reached steady state, the air flowrate at the gas outlet actually dipped below its final steady-state value while any remaining tracer was eventually flushed from the control volume. This always occurred well after the end of the tracer downstep and after the rush of air into the control volume had passed.
Figure 3.6 – Mixture composition model sample results ($U_g = 0.86$ m/s, 60° cone, helium off injection method, no solids): (a) separator pressure drop; (b) helium tracer inlet and outlet molar flowrates; (c) air and total outlet molar flowrates

Figure 3.7(a) shows the separator pressure drop during a typical gas RTD run with solids. In this example the superficial gas velocity was $U_g = 1.0$ m/s, with $m_s/m_g = 10$ and $L_S/D = 0$. The figure shows steady separator pressure drops for air only, air and solids, and air, solids, and helium tracer flows. The separator pressure drop was dominated by the contribution from air only, while the respective contributions from solids and helium tracer were quite small. Following the tracer downstep, the separator pressure drop
decreased gradually over a period of roughly 4 s before reaching a final steady-state value, as shown in Figure 3.7(b). Unlike the more complicated example shown in Figure 3.6, the mixture composition model predicted that there was no change in the air molar flowrate at the gas outlet during the tracer downstep as shown in Figure 3.7(d). Instead, all of the change observed in the total gas molar flowrate at the gas outlet was due solely to the change in the tracer molar flowrate at the outlet. The outlet tracer molar flowrate was observed to decrease very sharply just after the equally sharp downstep, followed by a more gradual decrease in the outlet tracer flowrate, as shown in Figure 3.7(c). The more gradual decrease in outlet tracer flowrate occurred as tracer in the separation zone and solids tank were eventually flushed from the reactor. Although very low outlet tracer flowrates could not be seen due to the scale used in Figure 3.7(c), the model predicted that the outlet tracer molar flowrate finally decreased to less than 1% of the maximum outlet tracer flowrate roughly 2.7 s after the downstep occurred.

Figure 3.7 – Mixture composition model sample results with solids ($U_g = 1.0$ m/s, 60° cone, helium off injection method, $\dot{m}_s/\dot{m}_g = 10$)
3.4.1.5 Separation Zone Control Volume Measurement

The maximum theoretical control volume ($V$) was composed of the separator, the solids collection tank (at various fill levels), and part of the gas outlet pipe, as indicated by the shaded region in Figure 3.4 above. However, the exact size of the actual control volume was not known and could only be estimated during any particular gas RTD experiment for several reasons:

- the solids collection tank was partially full,
- the gas occupying the solids collection tank may or may not have been partially or fully stagnant, thereby acting as an effective “dead” volume, and
- the control volume may have fluctuated with respect to time due to flow pattern resonance, oscillations, or other transient effects.

The maximum theoretical control volume was measured to provide an empirical upper limit on the value $V$ to be used in the tracer mole balance Equation (3.10). The separation zone volume was measured by injecting a small sample of gas with known pressure and volume into the sealed separator and solids tank, and measuring the change in the sealed separator pressure, as described in Chapter 2.4. Figure 3.8 shows the change in pressure for both the injected sample volume and separation zone during a typical volume measurement. Volume measurements were performed with $\pm 6\%$ experimental uncertainty. An uncertainty analysis for the volume measurements is provided in Appendix B.
Figure 3.8 – Separator volume experiment sample pressure signals

Figure 3.9 shows the measured separation zone volume as a function of the solids collection tank fill level. As expected, the measured volume $V$ decreased with increasing solids tank fill level. A linear regression fit the experimental data well. Therefore, since the tank fill level was easily measured, the maximum possible separation zone volume could be estimated from the linear regression.

Figure 3.9 – Effect of voidage and tank fill level in the collected solids tank on separation zone volume
3.4.1.6 Importance of Volume in the Gas Mixture Composition Model

Since the effective separator control volume was not known in any gas RTD experiment, $V$ was used as an adjustable parameter in the total gas mole balance in Equation (3.10) to minimize the residual error between the experimental and fitted tracer molar outlet flowrates. The experimental tracer mole flowrate was determined from the iterative solution of the separator pressure drop model in Equation (3.18). Typical solutions of Equation (3.10) showed that the molar accumulation term was dominant in the gas mole balance. This was indicated by the presence of a false tracer pulse when the assumed value of $V$ was much larger than the “actual” effective gas volume (i.e. non-stagnant, active volume), yet still less than the maximum theoretical value. The actual effective separator volume was taken as the value that resulted in the minimum sum of squared errors between the fitted and measured tracer molar flowrate in the gas outlet. Examples of false tracer pulses due to overestimates in the control volume ($V$) are shown for one sample gas RTD run in Figure 3.10(a). The actual effective separator volume in this example ($V = 2.5$ L) resulted in the minimum sum of squared errors for the fitted tracer molar flowrate. The tracer molar outlet flowrate was fitted very poorly when $V > 4$ L in the example shown in Figure 3.10. Overall, the optimized effective control volume was always less than 4 L for all gas RTD experiments, which represented partial penetration into the solids collection tank.
The fact that the “actual” effective gas volume was any value other than the full volume of the separator plus solids collection tank is counterintuitive. It would seem reasonable to expect that the control volume for the separation zone would also include the solids tank. However, it is important to distinguish the effective volume from the control volume. There are several reasons why the effective volume was much smaller than the volume of the separator plus solids collection tank. The most important reason, which
will be shown by the local gas velocity measurements in Section 3.4.7, is due to the gas velocity distribution below the gas outlet. The gas velocity decreased rapidly with increasing distance below the gas outlet, which meant that there was very strong bypassing of gas from the gap between the cone rim and downer wall directly to the gas outlet. Therefore, the solids tank effectively acted as a dead volume. Another reason for the small value for the effective volume was that very low flowrates of tracer (< 5 % of the steady tracer injection flowrate before the downstep) could not be resolved accurately using the mixture composition model. Therefore, very low concentrations of tracer caught in the mostly stagnant solids tank and eventually exiting the reactor were likely truncated from the RTD.

Figure 3.10(b) shows the effect of separator volume on the gas RTD ($E(t)$). When the actual volume ($V = 2.5$ L) was used in the gas mole balance equation, $E(t)$ was composed of a sharp primary peak at $t = 0$ followed by a long secondary tail from roughly $0.5 \, s < t < 2.3 \, s$. When the separator volume was increased above $V > 4$ L, the secondary peak feature was lost and the primary peak was shifted to increasingly later times. This result further suggested that the effective volume was much smaller than the total separation zone volume since a smaller effective volume value in the mixture composition model resulted in the ability to capture more of the RTD features long after the downstep.

### 3.4.2 Tracer Injection Method

An ideal tracer downstep injection fully and instantaneously replaces the tracer with the bulk material before or after the step injection (Levenspiel, 1999). In the current study, a comparative test was done between:

1. Injection of helium at a rate of 5 mol % of the total gas flow, followed by a quick switch using a three-way solenoid valve to 5 mol % substitute air (helium $\rightarrow$ air), and
2. Injection of helium at a rate of 5 vol % of the total gas flow, followed by rapid termination of helium flow (helium $\rightarrow$ off).

This test was done to determine the suitability of the helium $\rightarrow$ off method, which was simpler to operate and less disruptive to the overall flow hydrodynamics than the helium $\rightarrow$ air method. Figure 3.11(a) compares sample separator pressure drop signals for both
injection modes. The change in $\Delta P_{sep}$ during a downstep was roughly the same for both injection types but with opposite sign. Further, since the tracer molar flowrate at the gas outlet, $\dot{n}_{He,o}$, was directly proportional to the separator pressure drop, $d(\Delta P_{sep})/dt$, as shown in Equation (3.10), and therefore directly related to the gas RTD, $d(\Delta P_{sep})/dt$ was compared for both injection modes since it was a rough approximation for the gas RTD. Figure 3.11(b) shows smoothed $d(\Delta P_{sep})/dt$ signals for both injection modes. The mean time ($\tau$) and standard deviation ($\sigma$) of $d(\Delta P_{sep})/dt$ for both injection modes were found to be less than 5% different. Therefore, the simpler helium $\rightarrow$ off method was deemed acceptable for all further gas RTD experiments.

![Figure 3.11](image)

Figure 3.11 – Comparison of tracer injection methods ($U_g = 0.99$ m/s, no solids): (a) raw separator pressure drop, (b) smoothed separator pressure drop derivative
3.4.3 Tracer Injection Model

Due to the volume of accumulated tracer in the injection line between the solenoid valve and the sparger, which was 30 cm long, the step injection into the separator volume was not perfectly abrupt. The downstep was always performed in less than 0.5 s, but was usually injected in 0.1 s to 0.2 s, similar to the injection shown in Figure 3.12(b). Typical average residence times in the separator were on the order of 0.5 s. Consequently, the actual downstep time was significant. Therefore, a Tracer Injection Model was developed to account for the imperfect downstep injection. The Tracer Injection Model ($\hat{n}_{He,in}(t)$) was then convolved with the gas RTD ($E(t)$), to correctly model the tracer outlet molar flowrate ($\hat{n}_{He,out}(t)$). The Tracer Injection Model was based on the measured pressure drop across the tracer sparger ($\Delta P_{spg}$), which was treated as an orifice-type restriction. A derivation for the model is provided in Appendix C. The measured tracer inlet mole flowrate was fitted well by the Tracer Injection Model, which was given by

$$\hat{n}_{he,i}(t) = \hat{n}_0 \sqrt{\frac{\Delta P_{spg}(t) + \Delta P_{sep}(t) + P_{atm}}{\Delta P_{spg,0} + \Delta P_{sep,0} + P_{atm}}} \Delta P_{spg,0}, \quad (3.22)$$

where $\hat{n}_0$ was the measured, constant injected tracer flowrate at $t = 0$,

$\Delta P_{spg,0}$ was the measured, constant sparger pressure drop at $t = 0$, and

$\Delta P_{sep,0}$ was the measured, constant separator pressure drop at $t = 0$. 
Figure 3.12 – Helium tracer injection model sample results: a) sparger pressure drop; b) inlet molar flowrate
3.4.4 Effect of Gas Flowrate on Gas RTD and Active Gas Volume without Solids

3.4.4.1 Shape of Gas RTD

Figure 3.13 shows the general effect of gas flowrate on the gas RTD in the separator when no solids were present. As the superficial gas Reynolds number ($Re_g$) increased, the height of the primary peak at $t = 0$ grew, while the secondary pulse became taller and narrower with increasing gas Reynolds number ($Re_g$). A strong primary peak was observed because of the combined effects of bypassing due to the gas velocity distribution plus a rush of air at the time of the downstep. A secondary peak was observed due to the late emergence of a small amount of tracer from the solids outlet region. The results indicated that backmixing below the gas outlet decreased with increasing gas flowrate. As the gas flowrate increased, more complete mixing and deeper gas penetration toward the solids outlet occurred due to increased turbulence. The implication of deeper gas penetration toward the solids outlet was a smaller stagnant gas volume in the solids collection tank.
3.4.4.2 Active Gas Volume

With regard to the gas RTD, the “active” (i.e. non-stagnant) gas volume was used as a separator performance indicator instead of the mean residence time to quantify the depth of gas penetration below the gas outlet and into the solids tank. The active gas volume concept is illustrated in Figure 3.14. Since significant gas bypassing was observed in the separator, the active gas volume was calculated from the observed separator mean residence time, which was always much smaller experimentally than what would be expected from the nominal quotient of total separation zone volume and gas volume flowrate ($\tau = \frac{V}{Q}$). However, knowledge of the gas penetration depth was more useful than the mean residence time since it could be used to modify the separator design, to optimize the location of stripping gas injection, and to help explain the gas-solids separation phenomena.
Figure 3.14 – Idealized illustration of active gas volume concept

Figure 3.15 shows the general effect of gas flowrate on the active gas volume when no solids were present. The active gas volume ($V_g$) shown in Figure 3.15 was calculated using the experimental mean residence time ($\tau$) values in the following expression:

$$V_g = \frac{\tau \dot{m}_g}{\rho_g},$$

(3.23)

where $\dot{m}_g$ was the total gas mass flowrate, and $\rho_g$ was the average gas mixture density.

As the gas Reynolds number ($Re_g$) increased, the active gas volume increased in a non-linear fashion, indicating deeper gas penetration below the gas outlet. According to the trend line shown in Figure 3.15, the active gas volume reached the solids collection tank at $Re_g \approx 3300$. For $Re_g \geq 3300$, the active gas fully penetrated into the solids tank and occupied part of its volume. Although gas flow in the solids tank likely had an undesirable impact on the gas / solids separation process due to interaction with the particle bed, the reduction in stagnant gas volume was much more beneficial to avoid gas recirculation and backmixing, as discussed in the following section.
The results in Figure 3.15 also indicated that there was no apparent impact of superficial gas flow regime on the observed trend for the cone separator geometry presented in this work. However, the superficial flow regime did have an impact for other separator types, as discussed in the following chapter. In general, gas penetration below the gas outlet increased exponentially with increasing gas flowrate and downward momentum in the cone gap, as will be shown later in this chapter.

### 3.4.4.3 Gas Backmixing

Figure 3.16 shows the effect of gas flowrate on the coefficient of variation (COV) of the pulse response gas RTD. This method provided a means of comparing peak spread between experiments with large differences in gas flowrate and other operating conditions. The results in Figure 3.16 showed that RTD peak spread decreased in a power
law fashion with increasing gas flowrate. This trend indicated that mixing below the gas outlet became more intense and provided further evidence that the stagnant gas volume reduced in size with increasing gas flowrate. However, future experiments should be performed at $\Re_g > 10^4$ to confirm the continuing decreasing trend in COV with increasing gas flowrate.

Several important conclusions and recommendations are drawn from the results in Figure 3.15 and Figure 3.16:

1. The active gas volume increased rapidly with increasing gas flowrate, which was undesirable since gas penetration into the solids collection tank likely disrupts the collected particle bed. Also, at sufficiently high gas flowrates, the mean residence time actually increased with the gas flowrate due to the rapidly increasing active gas volume. This was undesirable since the gas-solids separation process should be completed as rapidly as possible for good pyrolysis reaction control.
2. Turbulence and mixing were enhanced with the increasing active gas volume, which reduced gas backmixing, as shown in Figure 3.17.

3. Without changing any aspects of the apparatus as tested, there should be a range of gas flowrates to achieve an acceptable trade-off between gas backmixing and depth of gas penetration into the solids tank. This range would be dictated by the allowable mean residence time and solids content in the product stream for the gas-solids separation process.

4. Increasing active gas volume (and eventual increase in mean residence time) with gas flowrate would have a general undesirable impact on reaction kinetics. However, there is a trade-off between minimized active gas volume and gas backmixing, since backmixing decreases with increasing gas flowrate.

5. To avoid gas penetration into the solids tank, the length of the downer between the gas outlet and the solids tank should be varied to determine its effect on gas backmixing, active gas volume, and solids collection efficiency.

Figure 3.17 – Relationship between RTD peak spread (COV) and active gas volume
3.4.5 Effect of Solids Loading on Gas RTD

Figure 3.18 shows several sample gas RTDs at various solids loading values, which demonstrated the general effect of solids loading on the gas RTD. The pulse response was observed to widen with increasing solids loading and the mean residence time became longer. This indicated that gas was entrained deep into the solids outlet and the solids collection tank when solids were introduced. The result suggested that stripping gas introduced below the gas outlet would be beneficial in the pyrolysis reaction when the solids loading ratio is high to prevent product vapor entrainment and degradation in the solids collection tank. In other words, if the process were to be intensified by increasing the solids loading, there would be a greater need for stripping gas.

![Sample gas RTDs at various solids loading ratios](image)

Figure 3.18 – Sample gas RTDs at various solids loading ratios ($Re_g = 5700; U_g = 1.1$ m/s)

Shown in Figure 3.19 below is the active gas volume plotted versus the solids loading at various superficial gas velocities. The most obvious feature was the significant scatter in the data. The scatter may be explained by either the experimental method not being accurate or reliable with solids present, or, if the method was indeed reliable, that the gas flow pattern was made unstable with solids. Given that the scatter observed in the gas-only active gas volume and RTD peak spread trends was much smaller, the method can probably be expected to be reliable. It should also be noted that the average mean response time for each data set increased slightly with $Re_g$, which was also observed in
gas-only runs for $Re_g > 4500$. This provided further evidence that the gas RTD results with solids were reasonable.

For the range of tested $Re_g$ shown in Figure 3.19 ($Re_g = 3400$ to $Re_g = 5700$), the gas-only runs demonstrated that the active gas certainly reached the solids tank. The addition of solids seemed to make gas mixing in the separator and solids tank more complex and unstable. In general, there was a weak upward trend in the mean response time with increasing solids loading at $U_g = 0.68$ m/s, and no effect of solids loading for $U_g \geq 0.86$ m/s. This result meant that the active gas volume increased with the solids loading. This would seem to make sense as the tracer would likely have been more easily entrained and carried into the solids tank with increasing solids loading. The following lessons are learned from the results in Figure 3.19:

1. When solids are introduced, tracer mixing in the solids tank becomes very complex as the tracer is entrained downward with the solids.
2. The active gas must be prevented from entering and mixing into the solids tank, which makes the use of stripping gas imperative.

Figure 3.20 shows the effect of solids loading on the gas RTD peak spread. As with the mean residence time trends, the scatter observed in the COV plots was quite large, which was a likely indication of gas flow pattern instability. Peak spread was observed to
decrease with increasing solids loading at the lowest superficial gas velocity, while there was no statistically significant change at the highest two superficial gas velocities. Since the active gas volume was shown to increase with solids loading, the corresponding decrease in peak spread suggested that gas backmixing was slightly reduced as solids loading increased, possibly since the stagnant volume of gas in the solids tank also decreased.

Figure 3.20 – Impact of solids loading ratio on pulse response coefficient of variation

\[(L_S/D = 0)\]

### 3.4.6 Effect of Separation Length on Gas RTD without Solids

Figure 3.21 shows the effect of separation length \((L_S/D)\) on the gas RTD at \(U_g = 0.68\) m/s (Re\(_g\) = 3400) and without solids. One might expect that the mean response time and active gas volume would decrease with increasing separation length (i.e. as the cone moves up, away from the gas outlet), since there is more space available between the cone and the gas outlet for the gas to exit the separator. However, the trends and confidence interval band widths indicated that there was no statistically significant change in the RTD with separation length. The impact of separation length may have been very small compared to the effect of the size of the cone of the tested separator, where a high gas velocity annular jet was formed in the narrow gap. The effect of cone size on the gas flow pattern and gas RTD is discussed in Chapter 5.
3.4.7 Preliminary Comparison of Experimental and Numerical Gas RTDs

In the first part of a two-part study by Yu et al. (2014), the cold flow operation of ICFAR’s downer gas-solids separator design was simulated numerically using the commercial computational fluid dynamics (CFD) code ANSYS Fluent. The work by Yu et al. (2014) is provided for reference in Appendix E, which gives all relevant details of the theoretical framework on which the CFD model was based. The goal of the first part of the study was to improve upon the preliminary CFD simulations by Huard (2009) and to validate the simulated multiphase hydrodynamics and solids collection efficiency of the gas-solids separator by comparing with experimental results. The solids collection efficiency predicted by CFD by Yu et al. (2014) was in good agreement with the experimental results. Yu et al. (2015) then implemented heat transfer and reaction kinetics equations in the second part of the same CFD study. The gas RTD was simulated for the entire downer and gas-solids separator, as shown in Yu et al. (2015), while the gas RTD for the gas-solids separator only was performed by X. Yu at Aston University in the United Kingdom. The CFD separator RTD was shared for comparison with experimental results in this thesis.
In short, the multiphase gas-solids hydrodynamics in the downer and separator were simulated using an Eulerian-Eulerian method, wherein both phases were treated as continuous interpenetrating media. This method is contrasted with the Eulerian-Lagrangian method, wherein the solids phase is treated as a discrete dispersed phase, which is more physically meaningful than in the Eulerian-Eulerian method. However, the Eulerian-Lagrangian approach is limited to very small solids volume fractions and neglects particle-particle interactions (Cortés & Gil, 2007). For this reason, and for the ability to simulate the multiphase flow at high solids concentrations, the Eulerian-Eulerian approach was deemed more appropriate and was thus selected for the CFD simulations.

To simulate the gas RTD in the separator, a post-processing massless tracer particle method was used. The massless tracer method, described in detail by Yu et al. (2015), Mellin et al. (2014), and Aubin et al. (2009), involved the release of a large number (~500) of massless particles distributed uniformly over the downer cross section at the same height where the experimental tracer sparger was located (14 cm above the gas outlet pipe). The simulated tracer particles released at the same height as the actual tracer sparger followed gas streamlines that were calculated from the previously solved gas velocity field in the actual simulation.

The simulated gas velocity field and trajectories of a sample of all tracer particles in the downer and gas-solids separator are shown in Figure 3.22, which were performed by Yu et al. (2015). In the cold model scenario, ~ 90 % of the tracer particles successfully reached the gas outlet, while the remaining ~ 10 % of tracer was retained and recirculated in the downer for the entire period of simulated process time (~ 8 s). It is interesting to note that the reverse gas flow pattern observed below the gas outlet (Figure 3.22(a)) was very similar to the original simulations performed by Huard (2009). Furthermore, the predicted tracer particle path lines showed a very sharp reversal just below the cone rim, which indicated that the majority of the gas did not penetrate far below the gas outlet, as demonstrated by the experimental gas RTD and local gas velocity experiments described in Section 3.4.8.
Figure 3.22 – Simulated (a) gas velocity field and (b) massless tracer particle path lines and residence time predicted by CFD (modified from Yu et al., 2015)

Figure 3.23 compares the experimental and CFD predicted gas RTDs at $U_g = 0.67$ m/s and for solids loading values in the range of 1.0 wt/wt to 20 wt/wt. The most striking feature observed in the experimental RTDs at all solids loading values was the sharp initial peak immediately after the downstep, which was not observed in the simulated RTDs. The sharp initial peak in the experimental results indicated very strong gas bypassing, with a significant fraction of tracer exiting the separator almost instantaneously after the downstep. The main reason explaining the presence of the sharp initial peak in the experiments but not in the CFD results was the tracer injection method. In the experiments, a strong rush of air from the downer was observed in the first instants after the downstep due to the sudden decrease in pressure in the separator. This pulse of air may have caused the sharp initial tracer bypass immediately after the downstep.
However, in the CFD simulation, the injection of tracer was “perfect” since the hydrodynamics were not disturbed by massless tracer particles. Therefore, it may be useful to modify the CFD simulations in future to approximate the tracer downstep injection used in the experiments, since a massless tracer method cannot be implemented experimentally.

In general, the CFD-predicted RTDs were much more uniform and narrower than the experimental RTDs. One main peak centred roughly at $t \approx 0.2$ s was observed in the CFD RTDs, as well as small secondary and tertiary peaks between 0.5 s to 1 s after the downstep. Similar, though much smaller, peaks occurring between zero to 1 s after the downstep were also observed in the experimental RTDs. Additional peaks occurring more than 1 s after the downstep were observed in the experimental RTDs, but not in the simulated RTDs. The main reason explaining the wider distribution of peaks in the experimental RTDs was that roughly 10 – 20 % of the tracer particles CFD RTDs were manually removed from the RTD calculation since they were observed to recirculate for a very long time in the separation zone or were backmixed into the downer. Therefore, if these particles were included in the CFD RTDs, additional peaks occurring later than 1 s after the downstep would also have been observed.
Local gas velocity measurements were performed in the region between the gas outlet and the solids collection tank to better understand the gas flow pattern and gas-solids separation process. The measurements were also used to determine the relationship between the local gas velocity distribution, the gas RTD, and the active gas volume, and whether a distinct boundary existed between the active and stagnant gas volumes.
3.4.8.1 Effect of Gas Flowrate on Gas Velocity Profiles

Figure 3.24 shows experimental normalized local gas velocity profiles versus height below the gas outlet for several values of $\text{Re}_g$ spanning the full range of laminar to turbulent superficial gas flow regimes. The gas velocity probe was located at $r/R_D = 0.71$, with the cone positioned at $L_S/D = 0$. All gas velocity profiles were fitted reasonably well with an exponential decay expression of the form:

$$U^* = \frac{U(r, z)}{U_g} = a_{U_z} \exp\left(-\frac{z}{b_{U_z}}\right),$$  \hspace{1cm} (3.24)

where $U$ was the local gas velocity,
$r$ was the radial position,
$z$ was the vertical position,
$a_{U_z}$ was the gas velocity fitting exponential intercept, and
$b_{U_z}$ was the gas velocity fitting exponential slope.

Figure 3.24 – Effect of sensor height below gas outlet ($z$) on normalized gas velocity ($U^*$) for various superficial Reynolds Numbers ($\text{Re}_g$) ($r / R_D = 0.71; L_S / D = 0$)
The purpose of curve fitting the local gas velocity profiles was to determine whether a distinct gas penetration length existed below the gas outlet. A direct consequence of fitting with an exponential decay function was that there was no distinct gas penetration length. Instead, gas flowing through the cone gap at high velocity penetrated along the downer wall and reversed direction *gradually* over a long vertical distance. The reversing flow then turned upward in the central core toward the gas outlet.

The exponential decay of gas velocity with height below the gas outlet also provided very important insight into the gas-solids separation process. If it is assumed that the gas flow pattern is not significantly altered with the introduction of solids, which is approximately true for low solids concentrations, then the most effective separation occurs just downstream of, and very near to, the cone rim. At this location the centrifugal force acting on a particle is strong since the gas velocity is highest, and a large fraction of this high velocity gas stream reverses direction to exit the separator. Moving downward, away from the cone and gas outlet, the centrifugal force decreases with the gas velocity, which should lead to less efficient gas-solids separation. However, as shown in Section 3.4.8.2, the vertical gas velocity profiles in the central core were equal in magnitude to the annulus, and thus most particles resisting the weaker centrifugal force could not be suspended by the low gas velocity far below the gas outlet.

The fitted exponential intercepts \( a_{Uz} \) for the data shown in Figure 3.24 were found to be statistically equal at all tested values of Re\(_g\) using an F-statistic test at a 5% significance level. The consequence of this result was that the gas velocity profile curves converged to the same normalized gas velocity at \( z = 0 \) when extrapolating from the curve fits. This result was a convenient check that the data trends were reasonable since the ratio of the gas velocity in the cone gap at \( z = 0 \) to the superficial gas velocity should be a constant value and should depend on the cone geometry only.

Figure 3.25 shows the fitted exponential slope \( b_{Uz} \) as a function of Re\(_g\). The fitted exponential slopes \( b_{Uz} \) for the data shown in Figure 3.24 were fitted very well by a decreasing power law expression of the form:
\[ b_{Uz} = a_{bRe} \text{Re}_g^{-b_{bRe}}, \quad (3.25) \]

where \( a_{bRe} \) and \( b_{bRe} \) were curve fitting parameters.

The exponential slope \( (b_{Uz}) \) decreased with increasing \( \text{Re}_g \). This indicated that \( b_{Uz} \) was a measure of how downward momentum of gas along the wall was preserved as a function of the momentum of the superficial gas. Although the results showed that the superficial gas flow regime did not have an impact on \( b_{Uz} \) for the tested separator geometry, it is shown in Chapter 4 that the superficial flow regime had a very important effect for other separator geometries.

Figure 3.25 – Effect of superficial Reynolds number (\( \text{Re}_g \)) on fitting parameter \( (b_{Uz}) \)

### 3.4.8.2 Effect of Radial Position

Figure 3.26 shows the normalized local gas velocity profiles (\( U^* \)) versus height below the gas outlet at two radial positions \( (r / R_D = 0 \) and \( r / R_D = 0.71 \)). At all tested superficial gas velocities, which spanned the full range of laminar to turbulent superficial gas flow regimes, there was no statistically significant difference between the velocity profiles at the two radial positions. All data at both radial positions were fitted reasonably well by the exponential decay function of Equation (3.24). For each tested value of \( \text{Re}_g \), all best-fit parameters for \( U^* \) were statistically equal between the two radial positions according to an F-statistic test at a 5 % significance level. Similar matching fits were achieved for
each pair of data sets at all tested values of $Re_g$. Therefore, the effect of sensor radial position was deemed not to have a significant impact on $U^*$ measurements over the tested range of $z$. It should be noted that reported values of $U^*$ were scalar variables since the thermistor was not able to distinguish the flow direction.

![Graphs showing normalized local gas velocity](image)

Figure 3.26 – Comparison of local gas velocity measurements at $r/R_D = 0$ (filled symbols) and $r/R_D = 0.71$ (open symbols with dot) ($L_S/D = 0$)

**3.4.8.3 Relationship between Gas RTD & Gas Flow Pattern**

Figure 3.27 shows the experimental active gas volume ($V_g$) plotted against the local gas velocity fitting exponential slope ($b_{U_z}$). The active gas volume was inversely proportional to $b_{U_z}$. Larger $b_{U_z}$ meant that the gas velocity decayed more quickly as one moved further
down from the gas outlet. The results in Figure 3.27 indicated that the active gas volume increased with the downward gas momentum. Since the downward gas momentum was proportional to the gas velocity in the cone gap, the active gas volume was thus directly related to the cone geometry. This result is discussed in further detail for several separator configurations in the following chapter.

The data plotted in Figure 3.27 were fitted well by an exponential decay expression of the form:

\[
V_x = a_{Vb} \exp\left(\frac{b_{Vb}}{b_{Uz}}\right), \tag{3.26}
\]

where \(a_{Vb}\) and \(b_{Vb}\) were empirical fitting parameters.

Given this kind of relationship, one may perform either a gas RTD measurement or a set of local gas velocity measurements to estimate the result of the other measurement. In such a scheme, if one type of measurement were to be performed, the values \(a_{Vb}\) and \(b_{Vb}\) would require calibration by doing a small number of measurements of the other type.

![Figure 3.27 – Relationship between active gas volume and downward gas momentum](image)

Figure 3.27 shows the gas RTD coefficient of variation (COV) plotted against the local gas velocity fitting exponential slope \(b_{Uz}\). COV increased linearly with \(b_{Uz}\), which indicated that the gas RTD peak spread and backmixing increased with decreasing
downward gas momentum. As the gas flowrate increased, gas penetrated further below the gas outlet and occupied a larger active volume. This led to more intense mixing and, thus, smaller gas RTD peak spread. Since downward gas momentum \( (b_{Uz}) \) decayed in a power law fashion with \( \text{Re}_g \), as shown in Figure 3.25, the gas RTD peak spread also decreased in similar fashion with \( \text{Re}_g \).

Figure 3.28 – Relationship between gas RTD spread and downward gas momentum

3.4.8.4 Effect of Separation Length

Figure 3.29 shows the effect of separation length on the measured local gas velocity profiles. The gas velocity trends shifted downward on the vertical axis with increasing separation length. As the cone was moved upward and further away from the gas outlet (i.e. increasing \( L_S/D \)), the space between the cone and the gas outlet increased, allowing for more of the gas to “turn” toward the outlet, with less gas flowing below the gas outlet. The slope of the curve fit at \( L_S/D = 0.5 \) was substantially smaller than for the other separation lengths, which indicated that downward momentum was best preserved at \( L_S/D = 0.5 \). The gas velocity fitting exponential slope \( (b_{Uz}) \) was found to be statistically equal for \( L_S/D = 0 \) and \( L_S/D = 1.0 \) using an F-statistic test at a 5% significance level. This result agreed with the observation made at \( L_S/D = 0 \) that there was no statistically significant effect of separation length on active gas volume, since active gas volume and \( b_{Uz} \) were shown to be linked. However, the difference in slope observed at \( L_S/D = 0.5 \)
showed that separation length likely had some effect on downward gas momentum and active gas volume. Further investigation of this relationship is required.

Figure 3.29 – Effect of separation length on normalized local gas velocity profiles

\( \text{Re}_g = 5500; \ r/R_D = 0; \text{no solids} \)

3.4.8.5 Effect of Solids Tank Fill Level

Figure 3.30 shows the normalized local gas velocity \( (U^*) \) plotted against the solids collection tank fill fraction. The superficial gas flow was fully turbulent \( (\text{Re}_g = 5500) \) and \( U^* \) was measured at \( z = 26.4 \text{ cm} \) below the gas outlet. The results showed that the solids tank fill level had no impact on the measured local gas velocity measured just upstream of the solids tank inlet. At \( \text{Re}_g = 5500 \), the active gas volume was previously shown to penetrate into the solids tank. Therefore, as the solids tank reached capacity, the collected solids bed did not have a significant impact on the gas flow pattern in the separator.
3.4.9 Local Solids Concentration Measurements

Local solids concentration measurements were performed using the same type of thermistor used for local gas velocity measurements to demonstrate the usefulness of the probe. A preliminary understanding of the solids concentration distribution in the separator region below the gas outlet was made possible by the measurements. This information was used to gain further insight into the gas/solids separation process.

3.4.9.1 Effect of Superficial Solids Loading

Local solids concentration profiles were measured in the separator region below the gas outlet using the same type of thermistor used to measure local gas velocities in 3.4.7. A temperature method similar to those described by McMillan et al. (2006) and by Fushimi et al. (2012) was used in this study. The downer air was pre-heated to ~ 50 °C before injection in the unit, and the solids were injected at room temperature. Local solids concentrations were determined by measuring the heat transfer between the thermistor and the gas/solids mixture passing over the probe.
Figure 3.31 shows the temperature-specific thermistor power ($Q^*$) radial profiles at three values of the superficial solids loading ratio. The measurements were performed for $L_s/D = 0$, $z = 8.5$ cm, and at $U_g = 0.79$ m/s ($Re_g = 5600$), which ensured fully turbulent superficial gas flow. Larger values of $Q^*$ indicated higher solids concentrations since heat transfer increased with increasing solids concentration. One could reasonably expect that the solids concentration would be highest at the walls and minimum at the centerline. However, as shown in Figure 3.31, $Q^*$ was maximum at $r/R_D = 1$ and decreased across the pipe diameter to a minimum at $r/R_D = -1$. The trends were similar at all tested superficial solids loading ratios.

The apparent minimum solids concentration at $r/R_D = -1$ can be explained by the location of the radial measurement profile. As shown in Figure 3.31, the radial measurement profile was in line with the gas outlet as viewed from above. Therefore, some solids that would otherwise have reached the thermistor between $-1 < r/R_D < 0$ were deflected away from the measurement profile line by the gas outlet. Furthermore, this effect was amplified going from $r/R_D = 0$ to $r/R_D = -1$ since the gas outlet pipe was oriented downward at a $45^\circ$ angle with decreasing $r/R_D$, meaning that the gas outlet was closer to the measurement point at $r/R_D = -1$. A measurement profile unobstructed by the gas outlet would have been preferable; however, the available measurement profile was limited by the location of existing ports on the downer unit.
Figure 3.31 – Effect of solids loading ratio on gas/solids mixture heat transfer radial profiles ($U_g = 0.79$ m/s, $Re_g = 5600$, $z = 8.5$ cm, $L_s/D = 0$)

A relationship was developed between the measured thermistor power and the superficial solids loading ratio in order to estimate the local solids loading ratio based on thermistor power measurements. Figure 3.32 shows the superficial solids loading ratio plotted against the area-weighted measured thermistor power for the same conditions described above. As mentioned above, the gas/solids mixture heat transfer increased with the solids loading. The data in Figure 3.32 were fitted very well by a power law relationship. It should be noted that the curve fit applied only to the specific operating conditions and separator geometry of the measurements shown in the figure. In other words, unique curves of the type shown in Figure 3.32 would be generated for different vertical positions, separator geometry, and superficial gas velocities. The curve fit shown in the figure was then used to estimate the local solids concentration along a measurement profile for a given set of operating conditions.
Figure 3.32 – Relationship between superficial solids loading ratio and area-weighted specific thermistor power ($U_g = 0.79 \text{ m/s}$, $Re_g = 5600$, $z = 8.5 \text{ cm}$, $L_s/D = 0$)

Figure 3.33 shows normalized local solids loading radial profiles for various superficial solids loading ratios using the curve fit shown in Figure 3.32. The local solids loading measurements were normalized to the superficial solids loading. As shown, the data were overlaid for all tested superficial solids loading ratios at $z = 8.5 \text{ cm}$. This indicated that the radial profile was independent of superficial solids loading ratio at $z = 8.5 \text{ cm}$.
3.4.9.2 Solids Concentration Distribution with Height

Figure 3.33 – Normalized local solids loading ratio radial profiles for various superficial solids loading ratios ($U_g = 0.79$ m/s, $Re_g = 5600$, $z = 8.5$ cm, $L_s/D = 0$)

**Figure 3.34** shows specific thermistor power radial profiles at three different heights below the gas outlet. The tests were performed with $U_g = 0.79$ m/s ($Re_g = 5600$), with a solids loading ratio of 6.4, and at $L_s/D = 0$. In general, the thermistor power at the downer centerline ($r/R_D = 0$) increased relative to the walls with increasing distance below the gas outlet. This indicated that particles were migrating increasingly from the walls to the central core as the distance below the gas outlet increased. Increased solids concentration in the central core far below the gas outlet was also observed by Yu et al. (2014) in numerical simulations of the same setup. The work by Yu et al. is provided for reference in Appendix E. Yu et al. predicted that solids migrated back toward the centreline with increasing $z$ for $\dot{m}_{sol}/\dot{m}_g > 10$ at $\dot{m}_g = 4$ g/s ($U_g \sim 0.7$ m/s, $Re_g \sim 4000$). Although the migration of solids toward the centerline was undesirable, the gas velocity was low far below the gas outlet, and thus the impact on gas/solids separation was thought to be small.
Figure 3.34 – Effect of thermistor height on gas / solids mixture heat transfer ($U_g = 0.79$ m/s, $Re_g = 5600$, $\dot{m}_{sol} / \dot{m}_g = 6.4$, $L_S/D = 0$)

3.5 Conclusions

In the first part of the present chapter, a simple, novel pressure-based measurement technique was developed and described to measure the gas RTD in a CFB downer gas-solids separator. The pressure response method was fast and avoided issues related to tracer sampling. The gas RTD was measured for a 60° internal angle, 6.3 cm rim diameter cone separator under various operating conditions. The separator pressure drop was related to the transient flowrates of helium tracer and air, and hence to the gas RTD, by a pressure drop regression and gas mixture composition model. The model was able to predict complex gas flows during the helium tracer downstep experiments.

The gas RTD experiments demonstrated that the active volume of non-stagnant gas increased exponentially with the gas flowrate. At high gas flowrates, the active gas penetrated far below the gas outlet into the solids collection tank. The experiments also demonstrated that gas RTD peak spread and gas backmixing decreased in a power law fashion with increasing gas flowrate. Gas entrainment and backmixing in the solids tank...
was significant for gas flow with solids. A slight decrease in gas backmixing at high solids loading was observed.

Local gas velocities were measured in the region between the gas outlet and the solids tank using a heat flux sensing thermistor. As the distance below the gas outlet increased, the local gas velocity decreased exponentially. Hence, there was no distinct gas penetration length below the gas outlet. In any case, a strong relationship was discovered between the gas velocity distribution below the gas outlet and the gas RTD. The active gas volume was shown to increase exponentially with the downward gas momentum developed in the gap surrounding the separator cone. Peak spread and backmixing were found to decrease linearly with increasing gas momentum in the cone gap.

The heat flux sensing thermistor was also used to measure the local solids concentration in the separator. The solids concentration radial profile was asymmetric about the downer centerline partly due to shielding from the gas outlet tube, which was in line with the measurement profile. Solids were also observed to migrate back to the centerline far below the gas outlet, where the gas velocity was low and where particles were difficult to entrain into the exiting gas stream.

3.6 Recommendations

Future studies based on the work presented in this chapter should be done in a high pressure metallic downer. This would allow for gas RTD measurements identical to those described in this paper to be performed at higher superficial Reynolds numbers to confirm the trends shown here. The tests in this work were limited to \( \text{Re}_g < 7000 \) for the rated pressure of the system. It is also recommended to test the effect of the gas outlet pipe diameter on the RTD, since the separator pressure drop was dominated by the kinetic pressure drop at the gas outlet. The effects of pressure and bulk gas density on the gas RTD could also be investigated by introducing various levels of backpressure in the gas exhaust line.

Another crucial modification to the system that should be studied is to increase the length of the downer between the gas outlet and the solids collection tank. For the separator
geometry tested in this chapter, the active gas was found to penetrate into the solids tank at relatively low superficial gas velocities (~ 0.6 m/s). To allow for complete gas-solids separation to occur in the downer, the separator region below the gas outlet should be extended coupled with an integrated solids stripper in the separator. This may also further reduce gas backmixing.
### 3.7 Notation

<table>
<thead>
<tr>
<th>Symbol</th>
<th>Description</th>
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</thead>
<tbody>
<tr>
<td>$a$</td>
<td>RTD peak amplitude [--]</td>
</tr>
<tr>
<td>$a_{bRe}$</td>
<td>Curve fitting parameter [--]</td>
</tr>
<tr>
<td>$a_{Uz}$</td>
<td>Gas velocity fitting exponential intercept [--]</td>
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<tr>
<td>$a_{4b}$</td>
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</tr>
<tr>
<td>$A_E$</td>
<td>Area under the RTD peak function [--]</td>
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<td>$b_{bRe}$</td>
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<td>RTD step function [--]</td>
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<td>Total molar flowrate at the gas outlet [mol/s]</td>
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<td>$\Delta P_{He}$</td>
<td>Contribution to the total separator pressure drop from helium [Pa]</td>
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<td>$\Delta P_{s}$</td>
<td>Contribution to the total separator pressure drop from solids [Pa]</td>
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<td>Superficial gas velocity [m/s]</td>
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<td>Active gas volume [m$^3$]</td>
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<td>$x$</td>
<td>Mole fraction of helium [mol/mol]</td>
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<td>Inlet tracer concentration [(mol/s)/(mol/s)]</td>
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<td>Outlet tracer concentration [(mol/s)/(mol/s)]</td>
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<td>Experimental outlet tracer concentration [(mol/s)/(mol/s)]</td>
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<tr>
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</tr>
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<td>Empirical fitting exponent [--]</td>
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</table>
\( \gamma_3 \)  \hspace{1cm} \text{Empirical fitting exponent [-]} \\
\( \gamma_4 \)  \hspace{1cm} \text{Empirical fitting exponent [-]} \\
\( \theta \)  \hspace{1cm} \text{Dummy time variable [s]} \\
\( \rho_g \)  \hspace{1cm} \text{Gas density [kg/m}^3\text{]} \\
\( \sigma \)  \hspace{1cm} \text{RTD standard deviation [s]} \\
\( \tau \)  \hspace{1cm} \text{RTD mean residence time [s]}
Chapter 4

4  Gas-Solids Separator Design Comparison\textsuperscript{2}

In this chapter, the performance of several separator designs were compared and ranked objectively using several experimental and analytical techniques described in Chapters 2 and 3. The best separator design and operating conditions were identified according to the criteria of least gas backmixing and highest gas-solids separation efficiency, which were captured by a novel, objective separator performance index. The effect of particle diameter on separator performance was also investigated. A 6.3 cm rim diameter cone separator was shown to be the best design among several tested separator geometries.

4.1 Introduction

Several authors have previously acknowledged the crucial importance of both the downer inlet and outlet configurations on the reactor performance, especially for severe, rapid cracking applications, where the mean residence time in the downer is ideally a fraction of a second (Gauthier et al., 1992; Johnston et al., 1999; Cheng et al., 2008). Therefore, traditional cyclones with mean residence times on the order of 1 – 2 s are not acceptable. Though solids collection efficiency and gas backmixing are the two main performance characteristics of any gas-solids separator in a CFB environment, an integrated downer exit and gas-solids separator design is essential to meet stringent reaction performance constraints.

As previously reported by Cheng et al.(2008) and by Huard et al. (2010), several unique rapid gas-solid separators have been developed specifically for the demanding reaction requirements in CFB downers. These separators typically had mean gas residence times

\textsuperscript{2} A version of this chapter has been presented orally at The 14\textsuperscript{th} International Conference on Fluidization and published in the conference proceedings as follows:

on the orders of 0.05 – 1 s (e.g. Gartside & Woebcke, 1981; Gauthier, 1991), which were faster than traditional cyclones, but at the expense of solids collection efficiency. In some cases the solids losses were up to 100 times greater in special gas-solids separator designs (e.g. Gartside & Woebcke’s half-turn separator design) than in traditional cyclones under identical operating conditions. To the author’s knowledge, only Gauthier (1991) specifically measured the gas RTD in the gas-solids separator of a downer reactor. The RTD results were used to optimize the design of a modified uniflow cyclone, wherein the gas exited in the same direction as the solids instead of reversing direction and exiting at the top of the unit as in a traditional cyclone.

In the present chapter, several candidate gas-solids separator designs were evaluated using experimental and analytical techniques developed in Chapters 2 and 3, and through development of new performance metrics. The objectives of this chapter were to:

- Identify the best separator design, geometry, and operating conditions based on the criteria of:
  - minimal gas backmixing
  - maximum solids collection efficiency
- Develop a new, comprehensive separator performance metric that accounts for the performance criteria listed above
- Explain the difference in performance between different separator designs

4.2 Experimental

4.2.1 Cold Model Downer Apparatus

The 6.9 cm diameter \(D\), 134 cm tall \(L\), transparent acrylic cold model downer apparatus described in Chapter 2.1 was used in the present chapter to test several new separator designs. Figure 4.1 shows all tested separator designs, which included: 60° internal angle cones with three different rim diameters, a bell-shape, no separator (tube outlet only), an “ideal” sparger outlet, and a tube-in-tube outlet. Three different cone diameters were used to determine the effect of cone diameter and cone gap velocity on the gas and solids RTDs and gas-solids separation efficiency. The “ideal” sparger outlet was designed to provide representative cross-sectional sampling of the gas stream
without consideration for the gas-solids separation efficiency. A tube outlet without any particle deflector was used as a type of baseline case. The tube-in-tube outlet was designed to force the active gas stream to always penetrate to the solids tank (i.e. to occupy roughly the same volume) regardless of the superficial gas velocity. The effect of cone angle on separator performance was not investigated in this thesis since gas-solids separation efficiency was found to be reduced in cone angles greater than 60° (Huard, 2009). A steep cone angle was necessary to prevent particles from rebounding upward from the cone surface and back into the downer.

![Figure 4.1 – Tested separator geometries (drawn to scale): a) small 60° cone, b) medium 60° cone, c) large 60° cone, d) bell shape, e) tube outlet, f) “ideal” sparger outlet, g) tube-in-tube outlet](image)

### 4.2.2 Experimental Conditions & Procedure

Local gas velocity and gas RTD measurements were made using the methods described in Chapters 2 and 3, respectively. In the present chapter, gas RTD experiments were performed over the range of superficial gas velocities $U_g = 0.19$ m/s to $U_g = 1.6$ m/s ($Re_g = 900$ to $Re_g = 8500$). No solids were used in the gas RTD or local gas velocity
measurements since the solids loading was shown in the previous chapter to have only a minor impact on the gas RTD and actually introduced significant scatter in the active gas volume and peak spread. The steady state concentration of helium during tracer injection was 5 vol% as used in Chapter 3. Solids collection efficiency experiments were performed using the method described in Chapter 2.7 over the range of superficial gas velocity $U_g = 0.8$ m/s to $U_g = 1.5$ m/s and over the range of solids loading $\dot{m}_s/\dot{m}_g = 1.1$ to $\dot{m}_s/\dot{m}_g = 31$. The cone separation length was kept constant at $L_s/D = 0$ in the present study since it was shown not to have a significant impact on the gas RTD in Chapter 3. Since solids loading and separation length were both removed as independent variables, the current chapter focused mainly on fundamental differences in the gas flow patterns between separator designs.

4.3 Results

4.3.1 Solids Collection Efficiency Performance

4.3.1.1 Overall

The most important function of the gas-solids separator is efficient removal of solids from the gas stream. As such, the solids losses for each separator design were measured and used as an initial screening tool to determine if any designs could be immediately rejected from consideration for use in the pyrolysis reaction. Figure 4.2 shows the overall solids losses averaged over all operating conditions for all tested separators except for the tube-in-tube outlet. As expected, the completely unshielded sparger and tube outlets had the greatest solids losses of 20.0 % and 8.1 %, respectively. Since previous gas-solids separator designs in downers tested in the literature had overall solids losses less than 10 %, and usually less than 2 % to 3 % (see Huard et al., 2010), both the sparger and tube outlets were deemed unsuitable for use in the downer. However, these “separator” designs were also tested as baseline cases in terms of gas RTD performance and gas velocity distribution. The cone-shaped and bell-shaped separators had the least solids losses since they all shielded the gas outlet from oncoming particles. The bell-shaped separator, small cone, and medium cone had average solids losses of 1.1 % to 1.7 %, while the large cone had the lowest solids losses at 0.07 %. All shielding-type separators
were deemed potentially suitable for use in the downer pyrolysis process, with the large cone having a strong advantage in solids collection efficiency.

Figure 4.2 – Comparison of overall average solids losses for several tested separator designs

4.3.1.2 Effect of Particle Size on Solids Collection Efficiency

4.3.1.2.1 General Observations

A more detailed solids collection efficiency study was performed in the large 60° cone to demonstrate how solids losses in the most efficient separator was affected by particle size, while also varying several other operating conditions. The effects of particle size, superficial gas velocity, and solids loading were assessed by measurement of grade efficiency curves. Two phosphorescent pigment particle size distributions (PSDs) with average particle sizes $\bar{d}_p = 220 \, \mu m$ and $\bar{d}_p = 620 \, \mu m$, and an experimental procedure described in Chapter 2.7.2, were used to determine the grade efficiency and effect of particle size.

Figure 4.3 shows the total solids losses plotted against the solids loading for the two tested average particle sizes. The superficial gas velocity was 1.2 m/s for the small PSD, and was varied at three levels in the large PSD ($U_g = \{0.61, 0.96, 1.2\} \, m/s$). For both
particle size groups, solids losses generally decreased with increasing solids loading, which was likely due to clustering in the cone gap and below the gas outlet. Overall, solids losses for the large particles were lower than for the small particles for $\frac{m_s}{m_g} > 4$. However, the difference in losses between the two particle sizes was very small since both particle sizes were, on the whole, quite large and thus easily collected. There was no significant impact of the gas velocity on the solids losses for the large 620 µm particles.

Figure 4.3 – Effect of solids loading, gas velocity, and particle size on total solids losses

(large 60° cone, $L_s/D = 0$, solids tank fill level < 30 %)

Figure 4.4 shows grade efficiency curves for six different test conditions corresponding to the small and large PSDs, for two superficial gas velocity levels ($U_g = 0.60$ m/s and $U_g = 1.2$ m/s), and for solids loading values in the range of 4.0 kg/kg to 24 kg/kg. In general, for both tested PSDs, the collection efficiency curves shown in Figure 4.4 decreased with particle size down to a threshold particle size, at which point the trend reversed direction. For the small 220 µm PSD seeded with 60 µm glass beads, and under all tested conditions, the collection efficiency decreased with particle size down to $d_p \sim 15$ µm, at which point the efficiency increased with decreasing particle size. For the large 620 µm PSD, a similar reversal in the grade efficiency trend was observed at $d_p \sim 150$ µm. The collection efficiency then increased with decreasing particle size down to $d_p \sim 60$ µm.

![Graph showing the effect of solids loading, gas velocity, and particle size on total solids losses.](image-url)
Particles less than 60 µm in size were collected with roughly equal efficiency in the large PSD.

Figure 4.4 – Effect of particle size, gas velocity, and solids loading on separator grade efficiency (large 60° cone, \(L_s/D = 0\))

The observed trend reversals at very fine particle sizes may have been due to particles escaping past the filter bag, thereby being considered as “collected,” or by a measurement error in the particle size analyzer. However, as the filter bag mesh size was 1 µm, and the smallest particles in the glass bead PSD were on the order of 20 µm, it was unlikely that particles were lost beyond the bag filter. Furthermore, there was no particular reason to
suspect a measurement error in the particle size analyzer, whose reported measurement range was 0.1 μm to 8.75 mm. The results shown in Figure 4.4 for the large PSD demonstrated that, for all tested conditions, the collection efficiency decreased with particle size down to \(d_p \sim 150\) μm. Since the minimum efficiency in the large PSD occurred at a relatively large particle size, the experimental errors suggested above (i.e. particles escaping past filter bag and particle size analysis errors) were very unlikely. Another possible explanation for the dip in collection efficiency for \(d_p \sim 150\) μm was that the large PSD underwent significant particle attrition and generated particles of size \(d_p \sim 150\) μm which escaped from the separator. However, this explanation was ruled out by measurement of the PSD of the injected particles before and after each experiment. There was no observed change in the PSD to the injected particles throughout the experiments, which confirmed that there was insignificant attrition of particles in the entire batch mass.

A more likely explanation for increasing collection efficiency with decreasing particle size below the threshold value was preferential agglomeration of fines and large particles by various mechanisms, including particle-particle collisions, electrostatic effects, and clustering along the walls. Preferential agglomeration and decreased entrainment of fine particles above the transport disengaging height has been demonstrated clearly by Bénoni et al. (1994) for polyethylene and sand particles in gas-solid fluidized beds. This phenomenon was also likely at play in the separator studied in this thesis. Particle-particle collisions would likely have been more frequent in the small PSD since the total number of particles in the system was much greater than in the large PSD at the same solids mass flowrate. This may partially explain why the threshold value for the small PSD was much lower than for the large PSD. However, in concept, fine particles agglomerating with larger particles would have had a much higher terminal velocity on average in the large PSD than the small PSD, making them more difficult to entrain in the exiting gas stream. This explanation was in good agreement with the observed higher collection efficiencies of particles smaller than 50 μm in the large PSD over all tested operating conditions. A general conclusion to be drawn from the results in Figure 4.4 was that the mean particle size had a very significant impact on the collection of fines and on the grade efficiency curves.
4.3.1.2.2 Effect of Solids Loading

For the 220 µm PSD, the results in Figure 4.4 showed that for $d_p > 70 \mu m$, the collection efficiency trend was slightly higher at the larger solids loading level at equal gas velocity. This result agreed with previous experiments showing that the collection efficiency generally increases with solids loading. However, for $d_p < 70 \mu m$, the efficiency trend was lower at the larger solids loading, which indicated that fines were lost increasingly at higher solids loading. There remains a need to further explore and explain this phenomenon. The overall collection efficiency was slightly higher at the larger solids loading (97.9 % at $\dot{m}_s/\dot{m}_g = 24$ versus 97.3 % at $\dot{m}_s/\dot{m}_g = 6.6$) because particles larger than 70 µm represented most of the mass of the PSD, which were collected more efficiently at the higher solids loading. For the 620 µm PSD, the effect of solids loading was clearer, where the observed efficiency trend was greater at all particle sizes at higher solids loading while comparing at equal gas velocity.

4.3.1.2.3 Effect of Gas Velocity

Gas velocity had opposite effects on the collection efficiency between the small and large PSDs. For the 220 µm PSD, particles smaller than 70 µm were collected up to 15 % less efficiently at $U_g = 1.2$ m/s than at $U_g = 0.60$ m/s at similar solids loading. However, for $d_p > 70 \mu m$, the collection efficiency increased with gas velocity. The lower collection efficiency of fines at higher gas velocity was likely due to two effects: first, the lower terminal velocity of the fines, making them easier to entrain, and second, deeper penetration of gas into the solids tank. For the large PSD, the observed collection efficiency trend at $U_g = 1.2$ m/s was roughly equal to or greater than the trend at $U_g = 0.60$ m/s at all particle sizes. This indicated that the terminal velocity of particles and clusters in the large PSD was sufficiently high to make entrainment in the gas stream very difficult.
4.3.2 Gas RTD Comparison

4.3.2.1 Separator Gas RTDs

Separator gas RTDs were calculated from the separator pressure drop during downstep experiments using the gas mixture composition model, tracer injection model, assumed RTD function forms, and reverse deconvolution method described in Chapter 3. Figure 4.5 shows the separator gas RTD for all tested separator designs at $U_g = 0.9$ m/s with no solids and separation length $L_S/D = 0$. All of the tested designs showed a strong sharp peak at $t = 0$ followed by a small secondary peak. Secondary peaks were undesirable and indicated the very late emergence of recirculating or trapped tracer, as demonstrated in Chapter 3. All designs also varied in the size and position of the secondary peak. The difference in responses between different separator types was typically very distinct. At $U_g = 0.9$ m/s, gas RTDs for the large 60° cone and the tube outlet showed the least spread, which indicated minimal backmixing relative to the other separator designs. The bell-shaped separator always showed significant secondary peaks long after the downstep under all conditions, and was therefore not studied further in this work.
Figure 4.5 – Comparison of separator gas RTDs to a tracer downstep for all tested separator types at identical conditions ($U_g = 0.9 \text{ m/s, } \text{Re}_g \sim 4500, \frac{L_s}{D} = 0$, no solids)

4.3.2.2 Effect of Gas Flowrate and Superficial Flow Regime

4.3.2.2.1 Mean Residence Time

The tube-in-tube outlet, shown in Figure 4.1(g), was initially introduced by Huard et al. (2013) to explain the observed opposite mean residence time trends between the shielded (cone) separators and the unshielded (tube and sparger) outlets. A demonstration using the tube-in-tube outlet was performed to show that when the RTD control volume was held approximately constant by forcing the gas to occupy the entire volume between the gas and solids outlets, the mean residence time trend should decrease with increasing gas flowrate, as expected by the nominal calculation $\tau = \frac{V}{Q}$. The nominal calculation assumed a somewhat arbitrary, constant control volume bounded by the sheds and gas and solids outlets. The demonstration was successful and showed that when the control
volume was partially fixed, the tube-in-tube mean residence time trend decreased with increasing gas flowrate up to $Re_g < 5500$, as shown in Figure 4.6. For $Re_g > 5500$, the mean residence time increased with the gas flowrate as the active gas expanded into the solids collection tank. A very similar trend was observed for the large cone, which indicated that the gas flowing downward through the cone gap and along the downer wall penetrated far below the gas outlet, and the active gas volume was always occupying almost the entire volume between the gas and solids outlets. Meanwhile, the mean residence time trends in the unshielded outlets increased with the gas flowrate for $Re_g > 2300$.

Figure 4.6 – Effect of gas flowrate on mean residence time for different separator types (no solids)
4.3.2.2.2 Active Gas Volume

In Chapter 3, the active gas volume was shown to increase and the peak spread to decrease with increasing gas flowrate for the large 60° cone. Turbulence and mixing below the gas outlet were thought to be enhanced with increasing gas flowrate due to the increased downward gas velocity in the cone gap, which also resulted in larger active gas volumes. Larger active gas volumes were generally preferred as they corresponded with smaller stagnant gas volumes in the solids collection tank. However, the benefits of a larger active gas volume were reduced at very high gas flowrates since the mean residence time actually increased with gas flowrate, which would be undesirable for the pyrolysis reaction. Figure 4.7 shows the effect of gas flowrate on the active gas volume for all separator designs except for the bell-shaped separator and tube-in-tube outlet. The active gas volume was plotted against the superficial Reynolds number in order to simultaneously account for the effect of the superficial gas flow regime as well as gas flowrate. In general, the active gas volume increased with the gas flowrate for all separators except for the small and medium 60° cones. The data for the small and medium cones showed huge scatter and very weak downward trends with increasing gas flowrate. These cone sizes produced very unstable gas flow patterns and were deemed unsuitable for use in the pyrolysis reaction. In spite of this conclusion, the small and medium cones were further studied along with the tube outlet to help explain the effect of cone diameter on the separator performance.

The sparger outlet and tube outlet trends were very similar, with the sparger trend shifted upward on the vertical y-axis relative to the tube outlet trend. For both separator types, the active gas volume increased in a rapid linear trend with gas flowrate for laminar superficial flow. There was then a sudden drop in the active gas volume as the gas transitioned out of the fully laminar regime and mixing became more intense. A change from laminar to laminar / turbulent transition flow occurred at $Re_g = 2200$ and $Re_g = 1900$ for the sparger and tube outlets, respectively. The difference in transition Reynolds number was likely related either to the different gas velocities at the outlet holes or the difference in outlet plane inclination angle between the two separators. For both
Separators, following the shift out of fully laminar superficial flow, the active gas volume again increased linearly with gas flowrate.

As shown in Chapter 3, and again in Figure 4.7, the active gas volume trend for the large 60° cone increased exponentially with increasing gas flowrate. The extrapolated curve fit lay neatly between the fitted curves for the sparger and tube outlets for \( \text{Re}_g > 6000 \), which suggested possible convergence in the active gas volume for these separator types. There was no transition behavior at low \( \text{Re}_g \) for the large cone because of the high, turbulent gas velocity developed in the cone gap, even at low gas flowrates. The importance of this phenomenon is discussed in detail in Section 4.3.4.

Figure 4.7 – Effect of gas flowrate on active gas volume for several separator designs

\( (L_s/D = 0 \text{ for all cone-type separators, no solids}) \)
4.3.2.2.3 Peak Spread

Figure 4.8 shows the effect of gas flowrate (Re_g) on the gas RTD coefficient of variation (COV) for all tested separators. A solid trend line is shown for the large 60° cone to indicate that the data were fitted by a regression, while for all other separators, trend lines are dashed to indicate that they are meant only as visual aids. In general, COV decreased with increasing Re_g for the sparger and tube outlets and for the large 60° cone, which shall be termed Group 1 separators. However, COV was observed to increase with gas flowrate for the small and medium cones and for the tube-in-tube outlet, which shall be termed Group 2. The main difference between Group 1 and Group 2, which is demonstrated in detail in Section 4.3.4, to explain the observed trends was the difference in downward gas velocity at the wall relative to the upward gas velocity in the central core. This relative difference in velocity was much higher in Group 1 versus Group 2 separators, which helped induce greater turbulence and mixing near the gas outlet in Group 1 separators.
As observed for the active volume trends in Figure 4.7, the scatter in COV in Figure 4.8 was very significant for the small and medium cones, which further proved that these cone sizes were unstable and not suitable for a rapid pyrolysis reaction. Significant scatter and a similar upward trend with increasing gas flowrate were also observed for the tube-in-tube outlet. The tube outlet showed rapidly increasing COV with increasing gas flowrate for laminar superficial flow. At $Re_g \approx 2600$, a transition occurred in the COV trend, which decreased in power law fashion with increasing gas flowrate.

Finally, for the large cone, COV decreased with increasing gas flowrate across the entire range of tested $Re_g$. Peak spread was also roughly two to three times less for the large cone than for all other tested separators. This result was attributed to the high gas velocity
induced in the cone gap for the large cone, which helped to induce very intense mixing below the gas outlet.

4.3.2.3 Effect of Particle Size

4.3.2.3.1 Active Gas Volume

Figure 4.9 shows the active gas volume plotted against the solids loading ratio for the small and large PSDs at three superficial gas velocity levels, $U_g = \{0.7, 0.9, 1.1\}$ m/s. Opposite trends were observed between the small and large PSDs. For the small PSD, the active gas volume generally increased with solids loading at all tested gas velocities, with very deep penetration into the solids tank. Huge scatter was observed in the active gas volume at $U_g = 0.7$ m/s ($Re_g = 3400$), which indicated an unstable gas flow pattern with solids present. The scatter in the active gas volume was greatly reduced at $U_g = 0.9$ m/s ($Re_g = 4600$) and $U_g = 1.1$ m/s ($Re_g = 5700$), which indicated more stable gas flow at higher gas velocities.

For the large PSD, the active gas volume generally decreased with increasing solids loading and with decreasing superficial gas velocity. Gas penetrated into the solids tank at low solids loading, but far less so than with the 220 µm particles. The scatter in the active gas volume trends was also generally smaller than observed in the small PSD. The active gas volume generally increased with superficial gas velocity and downward gas momentum, which was in agreement with the results shown in Chapter 3. The results indicated much more efficient gas / particle disengagement and less gas entrainment into the solids tank with the large PSD and with increasing solids loading.
Figure 4.9 – Effect of particle size distribution, gas flowrate, and solids flowrate on active gas volume (large 60° cone, $L_s/D = 0$)

4.3.2.3.2 Gas RTD Peak Spread

Figure 4.10 shows the effect of PSD, solids flowrate, and gas flowrate on the gas RTD coefficient of variation (peak spread). Due to the scatter in the plots, there was no statistically significant effect of any of the independent variables on the gas peak spread. Since it was shown that gas entrainment into the solids tank was reduced with the large
PSD, the results in Figure 4.10 indicated that there was neither advantage nor penalty on peak spread for operating with the large PSD.

Figure 4.10 – Effect of particle size distribution, gas flowrate, and solids flowrate on active gas volume (large 60° cone, \( L_g/D = 0 \))
4.3.3 Separator Performance

For rapid pyrolysis in a CFB downer the performance of the gas-solids separator was assessed on three fundamental criteria:

1. Maximum gas-solids separation efficiency
2. Minimum deviation from the theoretical mean residence time assuming a constant control volume bounded by the sheds and gas and solids outlets
3. Minimum gas backmixing, i.e. minimum gas RTD peak spread.

Minimum deviation from the theoretical mean residence time indicated minimum stagnant volumes, which was a desirable outcome to prevent gas recirculation, and in the case of the biomass or heavy oil pyrolysis, minimized excessive reaction in the separator. Figure 4.11 compares both the overall average fraction of escaped (uncollected) solids, the root mean square of deviations from the theoretical mean residence time trend, and the average gas RTD peak spread over all tested conditions for the sparger and tube outlets and for all cone sizes. The large 60° cone was superior to all other tested separators on all three performance metrics, and was very clearly the best design in terms of higher solids collection efficiency and lower peak spread. Roughly 100 times fewer solids were lost using the large cone compared to the sparger and tube outlets, wherein the gas outlet hole(s) were completely unshielded to oncoming particles. However, the performance rank of all other separator designs was not clear according to Figure 4.11.
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In order to objectively compare all separators on all three metrics (solids mass lost, deviation from nominal mean residence time, and gas RTD COV), a combined performance metric was developed to rank all separators. For the purpose of this study, each factor was given an equal weight. Depending on actual process economics, each factor may be weighted differently. The Separator Performance Index (SPI) was defined as

\[
SPI = \frac{1}{(\text{Avg. Solids Losses}) \times (\text{Avg. Dev. from Theo. Mean Res. Time}) \times (\text{Avg. Peak Spread})}
\]

\[
SPI = \frac{1}{(1-\eta) \left[ \sqrt{(\tau - \tau_{\text{nom}})^2} \right] (\text{COV})}, \tag{4.1}
\]

where \(\eta\) is the particle collection efficiency, and

\(\tau_{\text{nom}}\) is the nominal theoretical residence time.

The particle collection efficiency was defined by Huard (2009) as

Figure 4.11 – Comparison of separator performance by overall average solids mass lost, deviation from theoretical mean residence time, and RTD COV
\[ \eta = 1 - \frac{\text{Mass Solids Lost}}{\text{Total Mass Solids}}. \quad (4.2) \]

With SPI defined in this way, the desirable outcomes of fewer solids losses, controlled active gas volume, and smaller peak spread resulted in larger SPI. Therefore, larger SPI meant better separator performance.

Shown in Figure 4.12 is the SPI plotted against the unobstructed flow area \((A_f)\) for all cone sizes and for the tube and sparger outlets. With separator performance defined as shown in Equation (4.2), the SPI gave a very clear ranking of all separators, which was (from best to worst):

1. large 60° cone, with 81% of the cross-section area obstructed by the cone,
2. medium 60° cone (40% of the cross-section area obstructed),
3. small 60° cone (13% of the cross-section obstructed),
4. tube outlet (0% of the cross-section obstructed), and
5. sparger outlet (0% of the cross-section obstructed).

The unobstructed flow area was defined as the downer cross-sectional area available to the gas-solids mixture entering the separator in the plane of the gas outlet pipe. An illustration of the unobstructed flow area for the medium 60° cone is shown in Figure 4.13. In the case of the sparger outlet, the unobstructed flow area was larger than the downer cross-section area because the sparger tube was inclined 45° to the horizontal.

The SPI values shown in Figure 4.12 were calculated using the average solids collection efficiency, the root mean of summed squared deviations from the nominal residence time, and the average COV over all tests for each separator design. The observed trend showed that separator performance was inversely proportional to the unobstructed flow area, which in turn was a function of the cone rim diameter and of the angle of inclination of the outlet plane. The data were fitted reasonably well by a power law expression. The relationship between cone rim diameter, local gas velocity, and separator performance is discussed in detail in Section 4.3.4. In the case of the tube and sparger outlets, since no cone existed in these configurations, the unobstructed flow area was very large, and
consequently the particle separation efficiency was extremely poor, which greatly diminished the separator performance.

Figure 4.12 – Comparison of separator designs using Separator Performance Index (SPI) 
\(L_\theta/D = 0\) for all cone-type separators

![Comparison of separator designs using SPI](image1)

Figure 4.13 – Illustration of unobstructed flow area: (a) elevation view of separation zone, (b) top view of section AA’
4.3.4 Relationship between Separator Design, Gas Flow Pattern, and Separator Performance

4.3.4.1 Local Gas Velocity Measurements

Local gas velocity measurements were made in several separator designs using the thermistor-based technique described in Chapter 2.5.1.1. The purpose of these measurements was to better understand differences in the gas flow pattern for each separator design, which could then be used to explain the observed separator performance behavior. Figure 4.14 shows vertical normalized gas velocity profiles for all cone sizes and for the tube outlet at \( r/R_D = 0.71 \), with \( L_S/D = 0 \) and \( \text{Re}_g = 5500 \). The gas velocity decreased with increasing distance below the gas outlet for all separator designs. In general, the gas velocity profiles shifted downward on the vertical y-axis \((U^*)\) with increasing unobstructed flow area, which reflected the fact that higher gas velocities were induced in smaller cone gap areas. For \( z > 0.20 \text{ m} \), cone size ceased to have an impact on the gas velocity near the solids outlet.

![Figure 4.14 – Comparison of vertical normalized gas velocity profiles for three different cone separator sizes and tube outlet separator (Re\(_g\) = 5500; L\(_S\)/D = 0; r/R = 0.71; no solids)](image-url)
4.3.4.2 Vertical Gas Velocity Profile Fitting

All data sets shown in Figure 4.14 and for all other tested values of Re\(_g\) were fitted reasonably well by Equation (3.24):

\[
U^* = a_{Uz} \exp \left( - \frac{z}{b_{Uz}} \right).
\]

Both fitting parameters \(a_{Uz}\) and \(b_{Uz}\) were found to be statistically equal for the small and medium cones based on an F-statistic test with a 5% significance level.

Figure 4.15 shows the effect of gas flowrate and superficial flow regime (Re\(_g\)) on the fitting parameters \(a_{Uz}\) and \(b_{Uz}\). For the tube outlet and for both the small and medium cones, the trends showed that the fitting exponential intercept \(a_{Uz}\) increased almost identically with Re\(_g\). The meaning of this result was that for these separator types, the gas velocity at \(z = 0\) (i.e. at the height of the gas outlet) predicted by the extrapolated trend line was equal. However, for the large cone, \(a_{Uz}\) was constant with respect to Re\(_g\), and the predicted extrapolated value of \(a_{Uz}\) was always significantly higher than for all other separator types across all tested Re\(_g\).
Meanwhile, for all separator types, a power-law-type decay was observed in the fitting exponential slope $b_U$ with increasing $Re_g$. There was also a transition peak in $b_U$ at $Re_g \sim 2000$ for all separator types except for the largest cone. The meaning of this transition peak was that downward gas momentum was preserved most poorly at $Re_g \sim 2000$. When downward momentum is not preserved, the active gas volume would be expected to be very small. This explanation was actually in very good agreement with the observed active gas volume trend for the tube outlet shown in Figure 4.7, wherein the minimum active gas volume was at $Re_g \sim 2000$. 

Figure 4.15 – Effect of superficial Reynolds number ($Re_g$) on: (a) fitted exponential intercept ($a_U$); (b) fitted exponential slope ($b_U$)
As noted, a transition peak in $bU_z$ was not observed for the large cone. The unique behavior for the large cone was due to the high gas velocity formed in the gap between the cone rim and the downer wall that guaranteed turbulent flow in the gap for all tested $Re_g$. The Reynolds number in the gap ($Re_{gap}$) was estimated from the following expression:

$$Re_{gap} = \frac{4m_{air}}{\pi \mu (D - D_{cone})},$$

(4.3)

where $D_{cone}$ was the cone rim diameter.

For the tube outlet, since $D_{cone} = 0$, $Re_{gap}$ in (4.4) simply reduced to the superficial Reynolds number ($Re_g$).

Figure 4.16 shows the calculated values of $Re_{gap}$ plotted against $Re_g$ to demonstrate roughly when the gas stream transitioned to fully turbulent flow in the cone gap for all cone sizes and for the tube outlet. Over the entire tested range of $Re_g$, $Re_{gap}$ was fully turbulent ($Re_{gap} > 4000$) for the large cone. For all other separator types, $Re_{gap}$ transitioned through all flow regimes over the tested range of $Re_g$.

![Figure 4.16 – Calculated Reynolds number ($Re_{gap}$) in the gap between the separator cone rim and the downer wall for all tested values of superficial Reynolds number ($Re_g$)](image-url)
4.3.4.4 Counter-Flow Reynolds Number

All results discussed up to this point indicated the importance of downward gas momentum generated in the cone gap on the gas flow pattern, active gas volume, mixing, and the gas RTD. Greater downward momentum was surmised to induce stronger turbulence and more intense mixing below the gas outlet. Based on evidence from numerical simulations of the gas flow in the gas-solids separator performed by Huard (2009) and Yu et al. (2014), a simplified, idealized concept of the gas flow pattern below the gas outlet was developed. This idealized gas flow pattern is illustrated and compared to the gas flow pattern predicted by CFD by Yu et al. (2014) in Figure 4.17. In this flow pattern model, gas flowing downward through the annular cone gap, termed “gap flow,” continued along the downer wall and remained fairly distinct from the upward flowing gas in the core, termed “core flow.” Gas continually “leaked” from the gap flow to merge with the core flow and exit the separator via the gas outlet. The diameter of the core was assumed to be roughly equal to the size of the cone in the model. The CFD results of Yu et al. (2014) generally confirm the proposed idealized flow pattern, as shown in Figure 4.17.

Figure 4.17 – Idealized illustration and numerical simulation of the gas counter-flow pattern near the gas outlet: (a) idealized illustration, (b) – (d) CFD results
In order to account for the difference in momentum between the downward annular gap flow and the upward core flow, the “counter-flow” Reynolds number \( (Re_{cf}) \) was introduced. The counter-flow Reynolds number was defined as:

\[
Re_{cf} = Re_g + \left| Re_{gap} - Re_{core} \right|
\]

\[
Re_{cf} = \frac{4\dot{m}_{air}}{\pi \mu D} + \frac{4\dot{m}_{air}}{\pi \mu (D - D_{cone})} - \frac{4\dot{m}_{air}}{\pi \mu D_{cone}}
\]

\[
Re_{cf} = \frac{4\dot{m}_{air}}{\pi \mu D} \left( 1 + D \right) \left( \frac{1}{D - D_{cone}} - \frac{1}{D_{cone}} \right)
\]

The difference in magnitude in momentum between the downward and upward streams, \( \left| Re_{gap} - Re_{core} \right| \), was thus added to \( Re_g \) to give the counter-flow Reynolds number \( (Re_{cf}) \).

With \( Re_{cf} \) defined in this way, counter-flow below the gas outlet was assumed to enhance turbulence and mixing.

The trends observed in Figure 4.15 for \( a_{Uz} \) and \( b_{Uz} \) were best interpreted by plotting these fitting parameters versus \( Re_{cf} \), as shown in Figure 4.18. The counter-flow Reynolds number \( (Re_{cf}) \) allowed for clearer interpretation of \( a_{Uz} \) and \( b_{Uz} \) trends for selected separator designs. \( Re_{cf} \) was taken equal to \( Re_g \) for the tube outlet separator. As shown in Figure 4.18(a), transitions in \( a_{Uz} \) occurred at \( Re_{cf} \sim 2000 \) and \( Re_{cf} \sim 7000 \) to 10,000. If experiments were to be performed using the large cone at very low \( Re_g < 400 \), the \( a_{uz} \) trend might be expected to decrease sharply and converge with the trend for all other separators. However, these experiments were not performed since downer operation at such low gas flowrates was outside the area of practical interest for biomass pyrolysis.
Figure 4.18 – Effect of the counter-flow Reynolds number ($Re_{cf}$) on: a) fitted exponential intercept ($a_{Uz}$); b) fitted exponential slope ($b_{Uz}$)

Figure 4.18(b) shows the effect of counter-flow Reynolds number on the fitted exponential slope ($b_{Uz}$). One transition peak in $b_{Uz}$ was observed for the tube outlet and small and medium cones at $Re_{cf} \sim 2000$. This transition peak occurred roughly where the gas flow in the unobstructed flow area transitioned from fully laminar flow. No such transition was observed in the large cone since the smallest tested value of $Re_{cf}$ was roughly 7000. However, the existence of a transition peak for the large cone at $Re_{cf} = 2000$ was assumed for the sake of subsequent analysis. All data shown in Figure 4.18(b)
for $\text{Re}_{cf} > 2000$, and for all separator types, were fitted well by a modified form of Equation (3.25) using $\text{Re}_{cf}$:

$$b_{Uz} = a_1 \text{Re}^{-b_2 \text{Re}}.$$  

(4.5)

### 4.3.4.5 Counter-Flow Turbulence Index

In order to help explain why different maximum values of $b_{Uz}$ were observed for each separator design as shown in Figure 4.18(b), the “counter-flow turbulence index” ($n_{cf}$) was introduced. Referring to the right-hand side of Equation (4.5), $|n_{cf}|$ was defined as the term multiplied with $\text{Re}_{g}$:

$$|n_{cf}| = 1 + D \left( \frac{1}{D - D_{cone}} \right) - \frac{1}{D_{cone}};$$

$$\text{Re}_{g} = |n_{cf}| \text{Re}_{s}.$$  

(4.6)

The counter-flow turbulence index accounted for how turbulence and mixing were enhanced near the gas outlet relative to the superficial gas flow due to the size of the unobstructed flow area (i.e. cone size). Figure 4.19 shows the large variation of $n_{cf}$ with normalized separator cone diameter ($D_{cone}/D$). As shown, $|n_{cf}|$ approached infinity as $D_{cone}$ approached either 0 or $D$. The turbulence index $|n_{cf}|$ also approached 1 as $D_{cone}$ approached $D/2$. Hence, turbulence near the gas outlet was minimally enhanced relative to the superficial gas flow for $D_{cone} \sim D/2$, and was maximally enhanced for $D_{cone} \sim D$ and $D_{cone} \sim 0$. 


Figure 4.19 – Turbulence index for selected separator designs

Based on the results shown in Figure 4.19, the “best” possible separator design would apparently be the largest possible cone diameter in order to maximize turbulence and mixing while still being able to shield the gas outlet from particles. However, in practice, a trade-off exists with regard to the actual pyrolysis reaction, since the potential to plug the cone gap with tar and solids (sand, char, coke, and unreacted feedstock), and thereby trigger a costly reactor upset, increases with the cone diameter. Furthermore, as the gap velocity increases, the active gas volume also increases. In the limit as $D_{cone}$ approaches $D$, the active gas volume might in theory diverge to an extremely large value, resulting in an unacceptably long mean residence time.

Figure 4.20 shows the relationship between the transition peak value of $b_{Uz}$ and the counter-flow turbulence index. The following procedure was used to determine the value of $|n_{cf}|$ for the tube outlet:

1. The maximum transition peak values of $b_{Uz}$ were estimated graphically from Figure 4.18(b)
2. The maximum transition peak values ($b_{Uz,max}$) for all separator designs including the tube outlet were then fitted with an exponential expression of the form:
\[ b_{Uz, \text{max}} = a_{bn} \exp(b_{bn} |n_{cf}|), \quad (4.7) \]

where \( a_{bn} \) and \( b_{bn} \) were fitting parameters.

3. The absolute turbulence index \( |n_{cf}| \) for the tube outlet was then calculated by interpolation in Equation (4.8), assuming that the tube outlet behaved like a cone-type separator.

4. The effective \( D_{\text{cone}} \) for the tube outlet was calculated from the predicted value of \( n_{cf} \).

Figure 4.20 – Effect of absolute counter-flow turbulence index (\( |n_{cf}| \)) on transition peak value of fitted exponential slope (\( b_{Uz, \text{max}} \))

The maximum fitted exponential slope \( (b_{Uz, \text{max}}) \) was strongly dependent on the turbulence index, which in turn was a function of the cone diameter. This meant that the local gas velocity and mixing below the gas outlet were dominated by the cone diameter. Another interesting result to note, as shown in Figure 4.19, was that the point indicating the predicted turbulence index and effective \( D_{\text{cone}} \) for the tube outlet lay close to the theoretical turbulence index curve. This result indicated that the assumption of cone-type behavior in the tube outlet was reasonable. The effective cone diameter for the tube outlet
was $D_{cone} = 1.1$ cm, which was exactly equal to the diameter of the gas outlet tube. This result suggested that the outlet diameter also had an important effect on the gas flow pattern and the turbulence index should be modified in future work to reflect this fact.

### 4.3.4.6 Effect of Turbulence Index on Separator Performance

To this point, the absolute turbulence index $ln_{cfd}$ was used to interpret the local gas velocity measurements. However, the more crucial separator performance index was actually related to the real value of $n_{cf}$. Figure 4.21 shows the separator performance index (SPI) plotted against $n_{cf}$ for the tube outlet and cone separators. In this formulation of $n_{cf}$, the turbulence indices for the small cone and tube outlet were both negative, where negative values indicated simply that $D_{cone} < D/2$. By formulating $n_{cf}$ in this manner, SPI increased exponentially with the turbulence index. This was a very convenient result which demonstrated that the cone diameter should be maximized (within reason to prevent reactor maintenance issues) to achieve optimum gas-solids separator performance. Some potential reactor maintenance issues in the actual pyrolysis reaction include plugging by condensed viscous liquids and tars and by excessive solids flow piling up on the cone and in the cone gap. The large 60° cone diameter of 6.3 cm represents a rough upper limit on the cone size (or 90% of the downer diameter). Furthermore, in the cold model downer tested, the solids mass flowrate should be kept below 0.1 kg/s to prevent separator plugging. Finally, since the experiments in this chapter were performed at room temperature, it cannot be guaranteed that separator performance would be the same at high temperature in the pyrolysis reaction.
4.4 Conclusions

In the present chapter, using experimental and analytical tools described in Chapters 2 and 3, the large 60° cone separator was shown to perform significantly better than all other tested separator designs in terms of maximum solids collection efficiency, least deviation from the average theoretical mean residence time, and least gas backmixing. For all designs, the objective Separator Performance Index metric improved dramatically with increasing cone size. As the cone size increased, the unobstructed flow area in the plane of the gas outlet decreased, leading to higher cone gap velocities which enhanced turbulence and mixing. The gas flow pattern and local gas velocity profiles below the gas outlet were also shown to be dominated by cone size.

Two particle size distributions with average sizes of 220 μm and 620 μm were used in the downer to test the effect of particle size on the particle collection grade efficiency and the gas RTD. Since both average particle sizes were quite large, the solids losses were quite low – on the order of 0.1%. The collection efficiency of all fine particles smaller than 70 μm was significantly higher in the large PSD, which was surmised to be due to
agglomeration of fines with large particles with high terminal velocities. However, the collection efficiency of particles in the size range of 100 μm to 200 μm was actually superior in the small PSD. The gas RTD was found to be much more stable and the active gas volume smaller with the large PSD, which indicated smoother gas-solids disengagement in the large PSD. Particle size was found to have no impact on gas RTD peak spread.

### 4.5 Notation

- $A_f$: Unobstructed flow area [m$^2$]
- $a_{bRe}$: Curve fitting parameter [--]
- $a_{Uz}$: Gas velocity fitting exponential intercept [--]
- $b_{bRe}$: Curve fitting parameter [--]
- $b_{Uz}$: Gas velocity fitting exponential slope [--]
- $C_1$: Empirical separator pressure drop coefficient [--]
- COV: RTD coefficient of variation [--]
- $D$: Downer diameter [m]
- $D_{cone}$: Separator cone diameter [m]
- $d_p$: Average particle diameter [μm]
- $E(t)$: RTD peak function [--]
- $L$: Downer height [m]
- $L_{g/D}$: Separation length [--]
- $M$: Average molecular weight of gas mixture [kg/mol]
- $m_{s,lost}$: Mass of solids lost [kg]
- $m_{s,total}$: Mass of solids injected [kg]
- $\dot{m}_s/\dot{m}_g$: Solids loading ratio [(kg/s)/(kg/s)]
- $\dot{n}_{air}$: Air molar flowrate [mol/s]
- $n_{cf}$: Counter-flow turbulence index [--]
- $\dot{n}_{He}$: Helium molar flowrate [mol/s]
- $P_{sep}$: Absolute pressure in the separation zone [Pa]
- $\Delta P_{air}$: Contribution to the total separator pressure drop from air [Pa]
\( \Delta P_{sep} \) Pressure drop across the separation zone [Pa]
\( r/R_D \) Normalized radial position [--]
\( Q \) Gas volume flowrate \([\text{m}^3/\text{s}]\)
\( \text{Re}_{cf} \) Counter-flow Reynolds number [--]
\( \text{Re}_{core} \) Core flow Reynolds number [--]
\( \text{Re}_g \) Superficial gas Reynolds number [--]
\( \text{Re}_{gap} \) Cone gap Reynolds number [--]
\( \text{SPI} \) Separator Performance Index \([\text{s}^{-1}]\)
\( t \) Time [s]
\( U_g \) Superficial gas velocity [m]
\( U^* \) Normalized local gas velocity [m/s]
\( \mathcal{V} \) Control volume \([\text{m}^3]\)
\( \mathcal{V}_g \) Active gas volume \([\text{m}^3]\)
\( z \) Distance below gas outlet [m]
\( \eta \) Solids collection efficiency [--]
\( \mu \) Gas dynamic viscosity \([\text{Pa}\cdot\text{s}]\)
\( \rho \) Gas density \([\text{kg}/\text{m}^3]\)
\( \sigma \) RTD standard deviation [s]
\( \tau \) RTD mean residence time [s]
\( \tau_{nom} \) RTD nominal theoretical mean residence time [s]
Chapter 5

5 Solids RTD Measurements in a CFB Downer Gas-Solids Separator

In the present chapter, the solids RTD was measured in the downer and gas-solids separator using phosphorescent tracer activated by light flashes and detected using photocells as described in Chapter 2.8. With photocells mounted at several vertical positions, RTD peak spread was found to increase linearly with height along the downer. The solids mean residence time was found to decrease with increasing gas and solids flowrates. A one-dimensional (1D) particle trajectory model was found to fit the measured solids mean residence times reasonably well under high solids loading conditions. Finally, the measured solids mean residence time in the separator gave a good estimate of the gas/solids contact time in the separator.

5.1 Introduction

Many works in the literature have studied the impact of CFB riser termination on gas and solids hydrodynamics, backmixing, and RTD (e.g. Kunii & Levenspiel, 1995; Pugsley et al., 1997; Cheng et al., 1998; Gupta & Berruti, 2000; Reddy & Nag, 2001; Lackermeier & Werther, 2002; De Wilde et al., 2003; Harris et al., 2003a; Harris et al., 2003b; Yan et al., 2003; Mabrouk et al., 2008; Daga & Kumar, 2009; Chan et al., 2010; Wu et al., 2010; Van engelandt et al., 2011; Lopes et al., 2012; Zhang et al., 2013). The most striking feature of gas/solids flow at the top of the riser motivating this abundance of research is the recirculation and recycling of solids in the exit region of the riser. This phenomenon can be very detrimental when plug flow behavior is desired, as in hydrocarbon cracking and fluid pyrolysis, but can also be beneficial for combustion, where longer solids residence times are desired (Lackermeier & Werther, 2002). In general, as noted in Chapter 1.5.2, the main performance characteristics used to assess the performance of the riser exit have been the solids refluxing ratio, axial and radial mixing of both the gas and solids phases (concentration distributions, recirculation), solids refluxing and recirculation, and the solids RTD.
The above cited works in risers were mainly concerned with the local distribution of solids as well as axial and radial pressure profiles as proxies of gas and solids flow patterns. However, very few studies have investigated the effect of the exit configuration on the performance of the downer reactor, which was likely due to the fact that refluxing has not been found to be a major issue. Given that there is a lack of information regarding the downer exit, but also recognizing that solids recirculation is not a concern, the solids RTD would likely give the most relevant information with regard to the solids flow pattern at the downer exit.

Measurement of the solids RTD in CFB risers and downers is very useful for several purposes. In non-catalytic gas / solid reactions, such as biomass pyrolysis, the main applications of the solids RTD are to ensure adequate gas / solids contact times and to assess axial and radial heat and mass transfer (Huang, Qian, Zhang, & Wei, 2006). In other situations, the solids RTD is used to control solids mixing and residence time, to characterize reactor hotspots, and to identify uncombusted material (Lackermeier & Werther, 2002).

The phosphorescent tracer technique has been used in several solids RTD studies in both CFB risers and downers, as cited and described in Chapter 1. The main advantages of the technique are that tracer activation and detection are both fast and non-intrusive, and it is cost effective for small pilot scale units. However, the phosphorescent tracer dominates the cost of the method, which becomes prohibitively expensive for very large industrial systems. Preferential activation and detection of particles along the walls versus particles in the core can be a problem in some systems. Experimental errors may also be large if the tracer activation and detection sources are not collimated combined with significant particle dispersion. Overall, the phosphorescent technique seems to be the most reasonable compromise between RTD accuracy and ease of implementation after taking precautions for proper tracer activation and detection.

In the present chapter, phosphorescent particles were used with an original arrangement of photocells to track bulk solids transits at short distance intervals along the height of the downer and gas-solids separator. Several photocells at each vertical test location were
connected in series in a ring around the downer circumference to measure an analog cross-sectional average signal. Tracer detector rings were placed at five different heights to measure the RTD between many different points along the downer. To the author’s knowledge, only Kirbaş (2004) measured the solids RTD between more than two locations. Since the overall unit RTD is essentially accumulated in increments between detectors, the main advantage of the approach used here is the detection of distinct zones where problematic mixing or flow may exist. One disadvantage of this approach is that RTD accuracy decreases at detectors mounted between the reactor inlet and outlet, where special boundary condition treatment is not applied.

In the present chapter, the solids RTD was measured for several changing operating conditions and in several different areas of the unit to determine the behavior of the solids phase in relation to the gas phase and to gain insight into the gas-solids separation phenomena. The objectives of the present work were to:

- Quantify the range of the mean residence time and solids RTD peak spread while changing the gas and solids flowrates and separator cone size
- Identify the best operating conditions by considering both the gas and solids RTDs
- Estimate the gas-solids contact time
- Determine the behavior of the solids phase in relation to the gas phase
- Gain insight into the gas-solids separation phenomena
- Identify the possible existence of distinct reactor zones

5.2 Background

In order to predict the transit of solid tracer through the downer and gas-solids separator, a 1D model of the solids trajectory was developed. Considering a spherical particle in free fall in a 1D gas stream with velocity \( U_g \) as illustrated in Figure 5.1, the force balance acting on the particle in the vertical direction is given by:

\[
m_p \frac{d^2 z}{dt^2} = F_g - F_k - F_d,
\]

where \( m_p \) is the particle mass,
$z$ is the direction of gas and particle motion,

\[ \frac{d^2z}{dt^2} \] is the particle acceleration in the $z$ direction,

$F_g$ is the gravity force acting on the particle,

$F_b$ is the buoyancy force acting on the particle, and

$F_d$ is the drag force acting on the particle.

Figure 5.1 – Illustration of force balance on a particle in free fall in moving air

For a particle of characteristic size ($d_p$) with density ($\rho_p$) and volume ($V_p$) falling at velocity ($U_p$), and with gas density ($\rho_g$), the solution for the particle acceleration is given by:

\[
\rho_p V_p \frac{d^2z}{dt^2} = g \rho_p V_p - g \rho_g V_p - \frac{1}{2} \rho_g C_D A_p U_{\text{slip}}^2
\]

\[
\rho_p \left( \frac{\pi}{6} d_p^3 \right) \frac{d^2z}{dt^2} = g \left( \rho_p - \rho_g \right) \left( \frac{\pi}{6} d_p^3 \right) - \frac{1}{2} \rho_g C_D \left( \frac{\pi}{4} d_p^2 \right) (U_p - U_g)^2
\]

\[
\frac{d^2z}{dt^2} = \frac{g \left( \rho_p - \rho_g \right)}{\rho_p} - \frac{3}{4} \frac{\rho_g}{\rho_p d_p} C_D (U_p - U_g)^2,
\]

where $g$ is the gravitational acceleration, and

$C_D$ is the particle drag coefficient.

The following assumptions were made in developing the model described above:

- 1D particle transit (i.e. negligible axial / radial dispersion),
- Monosize particles of two different sizes \( d_p = 220 \mu m \) and \( d_p = 620 \mu m \) representing the small and large particle size distributions described in Chapter 2.6,
- The gas velocity was equal to the superficial gas velocity everywhere in the downer, from the downer inlet to the gas outlet,
- The gas velocity was set to zero below the gas outlet,
- One-way coupling from the gas phase to the solids phase, and
- Negligible particle-particle and particle-wall interactions.

The assumptions of negligible particle-particle and especially of particle-wall interactions were dubious; however, the goal in this work was simply to identify zones in the downer where the solids flow deviated from ideal 1D flow. Furthermore, the model did not account for changes in gas velocity due to the cone gap or gas flow pattern below the gas outlet. Note that the usual drag force expression written in terms of the freestream velocity \( U_g \), as given in White (2003), was modified to account for the slip velocity \( U_{slip} \) between the gas and ideal particle. The initial particle velocity at \( z = 0 \) (i.e. at the flash unit) was predicted using the same 1D model based on the known piping geometry and gas flowrate upstream of the downer inlet.

The drag coefficient \( C_D \) was calculated using explicit best-fit interpolation formulae described by Briens (1991) for spherical particles and using the explicit correlation proposed by Haider & Levenspiel (1989) for non-spherical particles. Since the sphericity of the tracer particles was not measured or known, the sensitivity of the predicted particle trajectory on sphericity was also investigated. Briens’ interpolation formulae were based on the following expression:

\[
C_D = a \text{Re}_p^{-b},
\]  

(5.3)

where \( \text{Re}_p = \rho_g(U_p - U_g)d_p/\mu_g \) was the particle Reynolds number, and

- \( a \) and \( b \) were interpolation fitting parameters for specific ranges \( \text{Re}_p \), which are given in Table 5.1.

The explicit correlation of Haider & Levenspiel was given by:
\[ C_D = \frac{24}{\text{Re}_p} \left[ 1 + \exp\left(2.3288 - 6.4581\phi + 2.4486\phi^2\right)\text{Re}_p^{0.0964+0.5565\phi} \right] \]
\[ + \frac{\text{Re}_p \exp\left(4.905 - 13.8944\phi + 18.4222\phi^2 - 10.2599\phi^3\right)}{\text{Re}_p + \exp\left[1.4681 + 12.2584\phi - 20.7322\phi^2 + 15.8855\phi^3\right]} \], \hspace{1cm} (5.4)

where \( \phi \) was the particle sphericity.

Table 5.1 – Spherical particle drag coefficient interpolation fitting parameters (from Briens, 1991)

<table>
<thead>
<tr>
<th>Interval No.</th>
<th>( \text{Re}_p ) Range</th>
<th>a</th>
<th>b</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>( 0 &lt; \text{Re}_p &lt; 0.15 )</td>
<td>24.54</td>
<td>0.996</td>
</tr>
<tr>
<td>2</td>
<td>( 0.15 &lt; \text{Re}_p &lt; 0.84 )</td>
<td>26.68</td>
<td>0.9537</td>
</tr>
<tr>
<td>3</td>
<td>( 0.84 &lt; \text{Re}_p &lt; 2.8 )</td>
<td>27.09</td>
<td>0.8732</td>
</tr>
<tr>
<td>4</td>
<td>( 2.8 &lt; \text{Re}_p &lt; 8 )</td>
<td>24.32</td>
<td>0.7677</td>
</tr>
<tr>
<td>5</td>
<td>( 8 &lt; \text{Re}_p &lt; 19.8 )</td>
<td>19.58</td>
<td>0.662</td>
</tr>
<tr>
<td>6</td>
<td>( 19.8 &lt; \text{Re}_p &lt; 64.5 )</td>
<td>15.9</td>
<td>0.5911</td>
</tr>
<tr>
<td>7</td>
<td>( 64.5 &lt; \text{Re}_p &lt; 252 )</td>
<td>10.61</td>
<td>0.4939</td>
</tr>
<tr>
<td>8</td>
<td>( 252 &lt; \text{Re}_p &lt; 585 )</td>
<td>4.035</td>
<td>0.3193</td>
</tr>
<tr>
<td>9</td>
<td>( 585 &lt; \text{Re}_p &lt; 1490 )</td>
<td>1.869</td>
<td>0.1988</td>
</tr>
<tr>
<td>10</td>
<td>( 1490 &lt; \text{Re}_p &lt; 1740 )</td>
<td>0.9573</td>
<td>0.1046</td>
</tr>
<tr>
<td>11</td>
<td>( 1740 &lt; \text{Re}_p &lt; 4330 )</td>
<td>0.9912</td>
<td>0.1107</td>
</tr>
<tr>
<td>12</td>
<td>( 4330 &lt; \text{Re}_p &lt; 8760 )</td>
<td>0.2952</td>
<td>-0.0341</td>
</tr>
<tr>
<td>13</td>
<td>( 8760 &lt; \text{Re}_p &lt; 12000 )</td>
<td>0.09314</td>
<td>-0.162</td>
</tr>
</tbody>
</table>

The particle acceleration in (5.2) can be integrated discretely once to give the particle velocity and twice to give the particle position as functions of time, thereby giving the 1D particle trajectory in the \( z \) direction:

\[
\frac{dz}{dt}_{t+\Delta t} = \frac{dz}{dt}_{t} + \frac{d^2z}{dt^2} \Delta t, \quad (5.5)
\]

\[
z_{t+\Delta t} = z_t + \frac{1}{2} \left( \frac{dz}{dt}_{t} + \frac{dz}{dt}_{t+\Delta t} \right) \Delta t. \quad (5.6)
\]

5.3 Experimental

5.3.1 Cold Model Downer Apparatus

The cold model downer apparatus described in Chapter 2.1 was also used in the present study. The sheds and tracer sparger were removed to prevent particles from rebounding.
upward as they entered the gas-solids separator. The same three cone diameters used in
the previous Chapters ($D_{cone} = \{ 2.5 \text{ cm}, 4.4 \text{ cm}, 6.3 \text{ cm} \}$) were again used here.

5.3.2 Solids RTD Equipment, Conditions, & Method
The solids RTD measurement equipment, operating conditions, and method used in this
chapter are described in Chapter 2.8. All three cone separators tested in Chapter 4 were
tested in this chapter, with a particular focus on the large cone separator, whose
performance was demonstrated to be much greater than all other tested separator designs.

5.4 Results
5.4.1 Tracer Phosphorescence Decay
In preliminary experiments, the tracer particle phosphorescence was observed to decay
completely within 60 s after being flashed with light, with 99 % of the full decay
occurring within 1 s after activation. Therefore, the transient decay of tracer phosphorescence was measured and modeled to account for phosphorescence decay in
the solids RTD model. In order to distinguish the photocell’s apparent response to
phosphorescence decay from its response to a light flash, the photocell response was
measured and modeled in both situations and deconvolved to obtain the true
phosphorescence decay response.

First, the photocell baseline response to a light flash in the absence of tracer was
measured. Next, a sample of phosphorescent tracer was suspended in a water / gelatin
solution and was then activated using a light flash as shown in Figure 5.2. The tracer /
water / gelatin suspension represented the approximate uniform distribution of particles
flowing in the downer. The photocell responses to both flash-only ($X_p(t)$) and flash
activated tracer decay ($Y_p(t)$) were measured as shown in Figure 5.3. A function form was
assumed for the true phosphorescence decay identity ($D(t)$), which was convolved with
$X_p(t)$ to fit $Y_p(t)$. The reverse deconvolution algorithm described in Chapter 3 was then
used to solve for the fitting parameters of $D(t)$. The function form that best represented
the raw true decay ($d(t)$) was given by:

$$d(t) = \frac{1}{1 + kt}, \quad (5.7)$$
where $k$ was the empirical phosphorescence decay constant.

The true decay identity ($D(t)$) was then obtained by normalizing $d(t)$ to the area under the curve of $d(t)$:

$$D(t) = \frac{d(t)}{\sum_{\Delta t} d(t) \Delta t}.$$  

(5.8)

Figure 5.2 – Phosphorescent tracer suspended in water / gelatin solution at several stages of decay following light flash activation

$t = 0.5 \text{ s} \quad t = 7.5 \text{ s} \quad t = 15 \text{ s}$

Figure 5.3 – Comparison of photocell flash-only and flash activated tracer decay responses
As shown in Figure 5.3, \( Y_p(t) \) was fitted well by the convolution \( (X_p*D)(t) \) for \( t < 3 \) s after the flash activation of tracer. In all solids RTD experiments, all tracer exited the reactor in less than 1.5 s, and thus the form assumed for the raw true decay function defined in (4.4) was sufficient. The empirical phosphorescence decay constant was found to be \( k \sim 500 \text{ s}^{-1} \).

### 5.4.2 Sample Photocell Pulse Response

Figure 5.4 shows the raw photocell response at all detector locations to a sample tracer pulse in the downer. The sample experiment was performed with \( U_g = 0.75 \text{ m/s} \) and \( \dot{m}_s/\dot{m}_g = 27 \). A strong, sharp peak was observed at \( t = 0 \) at all photocells, which corresponded to the tracer activation flash. The ability to detect the flash at all locations allowed for its use as a waveguide. The strength of the flash-induced peak at \( t = 0 \), indicated by the strength of the photocell voltage, decreased with distance from the flash heads along the downer. Activated tracer was initially detected at the first photocell ring at \( z = 69 \) cm at roughly \( t = 80 \) ms. The final detected tracer particles passed by the last photocell at \( z = 133 \) cm at roughly \( t = 950 \) ms. In general, one main peak was observed at each photocell height. As the activated tracer flowed downward, the photocell response peak became broader and spread out due to dispersion and mixing of tracer. The peak height also decreased with increasing length along the downer due to the phosphorescence decay and dispersion of the tracer.
5.4.3 Solids RTD Modelling

The reverse deconvolution algorithm used successfully in the gas RTD studies was applied to model the solids RTD. However, one additional convolution calculation was required to account for the phosphorescence decay. Therefore, the fitted photocell response at height $z$ was given by:

$$Y_{fi}(t) = [X_p * D] * E(t), \quad \text{(5.9)}$$

where $E(t)$ was the solids RTD function between the flash and the photocell at $z$. $E(t)$ was described by a sum of two exponential peak functions as given in Equation (3.1). In most cases, however, the reverse deconvolution algorithm converged to a solution requiring only one peak to fit the photocell response ($Y(t)$). Only the fitting parameters for $E(t)$ were optimized since the photocell flash response $X_p(t)$ was measured and $D(t)$ was known from the phosphorescence decay test. Five total adjustable parameters were used to model $E(t)$ (peak 1: amplitude, time shift, weight fraction; peak 2: amplitude and time shift).
Figure 5.5 compares the photocell response for a flash-only event (\(X_p(t)\)) to the experimental and fitted photocell responses (\(Y_p(t)\) and \(Y_{p,fit}(t)\), respectively) during the sample RTD experiment described in 5.4.2 at \(z = 69\) cm. The experimental photocell response \((Y(t))\) was fitted well by the double convolution \([(X_p*D)*E](t)\). The optimized solids RTD function \(E(t)\) is also shown in the inset plot in Figure 5.5, which was a single peak with a tail initiated at \(t \sim 100\) ms. Nearly all activated tracer particles completed their transit past the photocell at \(z = 69\) cm by \(t < 500\) ms. A tiny amount of tracer fines formed a thin film on the downer wall in view of the photocells in most experiments, which led to a very small, negligible non-zero photocell response for several seconds after the tracer pulse.

Figure 5.5 – Sample photocell response modelling for a typical solids RTD experiment

\((U_g = 0.75\) m/s, \(\dot{m}_s/\dot{m}_g = 27, d_{P,avg} = 220\) µm, large 60° cone, \(z = 69\) cm)
5.4.4 Experimental Variability of Solids RTD Experiments

For each solids RTD experiment, several (typically four or five) replicates were performed to increase confidence in the measurements. Plots of tracer mean residence time and standard deviation at three different vertical positions in a sample RTD experiment are shown in Figure 5.6. 95% confidence intervals are shown to indicate the typical level of repeatability of the mean residence time and standard deviation, which were used to characterize the solids RTD. As shown in Figure 5.6(a), the size of the mean time confidence interval at each vertical position was typically 10 – 20% of the full scale value. However, the size of the standard deviation confidence interval decreased with distance along the downer, as shown in Figure 5.6(b), which indicated that mixing became more uniform and complete moving along the downer. In practice, an average of all replicate photocell voltage signals at each vertical position was taken in order to greatly reduce the total number of convolution calculations.

5.4.5 Effect of Particle Sphericity on 1D Trajectory Model

Figure 5.6(a) shows the particle trajectory predicted by the 1D particle trajectory model (Equations (5.2) & (5.6)) for various values of the particle sphericity ($\phi$) for $U_g = 0.75$ m/s. The particle sphericity was varied over the range $\phi = 0.67$ (strongly non-spherical) to $\phi = 1$ (perfectly spherical), where $\phi = 0.67$ represented the limit of applicability of the drag coefficient correlation proposed by Haider & Levenspiel. Over the range $z = 69$ cm to $z = 135$ cm, i.e. the range of $z$ where photocells were installed, the predicted mean residence time varied no more than 8% over the range of tested particle sphericities, which was less than the variability of the experimental mean residence times. Therefore, since the tracer sphericity was not known or measured in this study, but the shape of the particles could be deemed “irregular,” an approximate mid-range value of $\phi = 0.8$ was assumed for the 1D model.
5.4.6 Effect of Solids Loading on Solids RTD in Overall Unit

Using the double convolution expression in (4.8) and the reverse deconvolution algorithm to optimize the solids RTD function, the RTD mean time and standard deviation were then calculated for all solids RTD experiments. Figure 5.7 shows the mean residence time at all detector locations for $U_g = 0.75$ m/s and for three solids loading values, $\dot{m}_s/\dot{m}_g =$
3.3, $\dot{m}_s / \dot{m}_g = 10$, and $\dot{m}_s / \dot{m}_g = 33$. Individual data points shown in the figure represented the average over several replicates by fitting one single RTD function to the average of all replicate photocell voltage signals. The solids mean residence time increased with height below the tracer flash unit (which was 7 cm below the downer inlet). The trends shifted downward along the vertical y-axis with increasing solids loading, approaching the predicted ideal 1D particle transit at the highest solids loading. However, this result did not mean necessarily that particles at high solids loadings actually approached ideal 1D behavior. It is possible that particle clustering increased with solids loading, thereby increasing the terminal velocity of heavier clusters, leading to shorter mean residence times. Another possible explanation for the observed behavior is decreased dispersion / backmixing with increasing solids loading.

Figure 5.7 – Effect of vertical position and solids loading on experimental mean solids residence time ($U_g = 0.75$ m/s, $d_{p,avg} = 220$ µm, large 60° cone, $L_s/D = 0$)

Figure 5.8 shows the solids RTD standard deviation ($\sigma$) versus height for the same conditions just described for the mean residence time. The standard deviation, which was a measure of RTD peak spread, increased in a nearly linear trend with height along the
downer at all tested solids loading values due to radial / axial dispersion and backmixing. The slopes of all linear regressions used to fit the data were statistically equal for all solids loading values based on an F-statistic test at a 5% level of significance. The assumption of a linear fit meant that the RTD spread increased at the same rate at every position along the height of the downer, with no local variations due to internals or gas flow pattern changes. The scatter in the data demonstrated that an assumption of a linear fit was reasonable but slightly inaccurate. The only difference between the trends at different solids loading values was the linear intercept, which meant that the injected solids had already undergone varying degrees of dispersion / backmixing by the time they reached the flash unit at \( z = 7 \) cm.

![Figure 5.8 – Effect of vertical position and solids loading on solids RTD standard deviation (\( U_g = 0.75 \) m/s, \( d_{P,avg} = 220 \) µm, large 60° cone, \( L_s/D = 0 \))](figure)

5.4.7 Effect of Gas Flowrate on Solids RTD in Overall Unit

Figure 5.9 shows the mean residence time versus height along the downer for \( \dot{m}_s/\dot{m}_g \sim 16 \) kg/kg and at three superficial gas velocities, \( U_g = \{0.75, 1.1, 1.4\} \) m/s. The data at all tested gas velocities at all vertical positions were very similar. As such, the gas velocity appeared to have almost no impact on the total unit mean residence time as measured
from the flash heads to the solids outlet. However, this result was not true with regard to the separation zone, as shown in Figure 5.10. All data in Figure 5.9 were best fitted by the 1D particle trajectory model for $U_g = 0.75 \text{ m/s}$. This result provided an indication that either axial or radial dispersion, or both, increased with gas flowrate, which resulted in solids flow as if it were being transported by an ideal 1D gas stream whose velocity was $U_g = 0.75 \text{ m/s}$. However, by observing only Figure 5.9, it was not clear what form of dispersion was enhanced with increasing gas velocity.

![Figure 5.9 – Effect of vertical position and gas flowrate on solids mean residence time](image)

Figure 5.9 – Effect of vertical position and gas flowrate on solids mean residence time

\[
\left( \frac{\dot{m}_s}{\dot{m}_g} \sim 15 \right) \quad d_{p,\text{avg}} = 220 \mu\text{m}, \text{large } 60^\circ \text{ cone, } L_S/D = 0
\]

As shown in both Figure 5.7 and Figure 5.9, the mean residence time from $z = 0$ to $z = 69 \text{ cm}$ was typically shorter than the predicted 1D trajectory curve at a given superficial gas velocity. The most likely explanation was that a high velocity gas jet at the downer inlet was formed due to the inlet piping, which accelerated the particles to a high initial velocity entering the downer.
5.4.8 Effect of Gas and Solids Flowrates on Separator Solids RTD

Figure 5.10 shows the mean residence time in the large 60° cone gas-solids separator as functions of both the gas and solids flowrates. The solids mean residence time in the separator decreased with increasing solids loading at all gas velocities. This could be explained by increased particle clustering with increasing solids loading. Particle clustering in the cone gap would be expected to increase as the probability of particle-particle collisions increases with increasing solids loading. The mean residence time trends generally decreased with increasing gas velocity, though the trends for $U_g = 1.1$ m/s and $U_g = 1.4$ m/s were very similar. In theory, the downward drag force on a particle should increase with downward gas velocity. Therefore, it is reasonable to expect the particle residence time to decrease with increasing gas velocity, which is in agreement with the observed results.

![Figure 5.10](image)

Figure 5.10 – Effect of both gas and solids flowrate on solids mean residence time in the separation zone (large 60° cone, $d_{P,avg} = 220 \, \mu m$, $L_S/D = 0$)

Figure 5.11 shows the effect of gas flowrate on the solids RTD standard deviation. Peak spread was observed to increase in a roughly linear fashion with height along the downer at all tested gas velocities. The slopes of the lines of best fit were statistically equal for all tested gas velocities according to an F-statistic test with a 5 % level of significance. This
indicated that dispersion and backmixing were independent of gas velocity for $70 \text{ cm} < z < 135 \text{ cm}$. The observed shift between trends along the vertical $y$-axis was likely due to different mixing behaviors at the downer inlet, as previously observed in Figure 5.8. The extent of initial dispersion / backmixing as the solids reached the flash unit seemed to depend on both the gas and solids flowrates.

Figure 5.11 – Effect of vertical position and gas flowrate on solids RTD standard deviation ($\hat{m}_s/\hat{m}_g \sim 15$, $d_{p,avg} = 220 \mu\text{m}$, large $60^\circ$ cone, $L_s/D = 0$)
5.4.9 Effect of Particle Size on Solids RTD

5.4.9.1 Solids RTD in Downer

Figure 5.12 shows the measured solids mean response time at several positions along the downer for both the small (220 µm) and large (620 µm) PSDs, at three superficial gas velocity levels $U_g = \{0.75, 1.1, 1.4\}$ m/s, and at three different solids loading levels at each gas velocity. For both particle size groups, the mean response time trends shifted downward along the vertical y-axis with increasing solids loading. More frequent particle clustering with increasing solids loading, leading to higher average particle terminal velocities, was likely responsible for the decrease in residence time at high solids loadings. This explanation is in agreement with previous studies on particle clustering in CFB downers, where the cluster slip velocity has been shown to increase with particle cluster size (Nova et al., 2004; Lanza et al., 2012), and where particle cluster frequency has been shown to increase with solids mass flux (Manyele et al., 2010).

As noted in Section 5.4.7, the gas flowrate appeared to have very little impact on the total unit mean residence time for the 220 µm particles, which was further confirmed across the full range of solids loadings in Figure 5.12.

The 1D particle trajectory model was applied to establish an ideal baseline for comparison to the measured particle residence times in both PSDs. For the small PSD, the mean residence time trends deviated significantly from ideal 1D particle trajectories under all conditions, which indicated significant solids dispersion and backmixing. The strong dispersion and backmixing indicated that the smaller PSD particles were very sensitive to gas mixing and turbulence. Deviation from the 1D model increased with gas velocity in the small PSD. Both the experimental and model trends showed that particles decelerated along the full length of the downer, which indicated that the average initial velocity of the particles entering the downer was greater than the average terminal velocity. Overall, mean residence times for the small PSD were significantly longer than for the large PSD due to a lower average terminal velocity. This result may have a profound impact on the gas-solids contact time and heat transfer, and is left for future study.
Figure 5.12 – Effect of solids loading, superficial gas velocity, and particle size on solids mean response time at several points in the downer (large 60° cone, \(L_s/D = 0\))

For the large PSD, the experimental mean residence time trends shown in Figure 5.12 followed the 1D particle trajectory model for \(z < 100\) cm, indicating little dispersion and backmixing over this region, thereby indicating low sensitivity to gas velocity fluctuations. However, for \(z > 100\) cm, i.e. through the separator, particles slowed down due to impact and redirection on the separator cone surface and possibly due to radial redirection from the downer wall. The 1D particle trajectory model predicted that a
“perfect” representative particle would accelerate over the entire length of the downer toward its eventual terminal velocity well below the unit. The experimental data for \( z < 100 \) cm followed this predicted trend, which indicated that the average entry velocity of particles at the downer inlet was lower than the average terminal velocity.

Figure 5.13 shows the downer solids RTD standard deviation at several positions along the downer for the same conditions described above for the solids mean residence time. For both PSDs, the standard deviation increased with vertical position from the downer inlet as backmixing and dispersion accumulated in the direction of flow. However, the solids RTD peak spread in the small PSD was roughly two to three times greater than the peak spread observed in the large PSD at all conditions.

For the small PSD, the standard deviation trends decreased with increasing solids loading and with increasing gas velocity. The experimental data were best fitted with linear trend lines indicating a constant rate of increase in peak spread and backmixing with increasing vertical position. Given that the mean residence time trends deviated increasingly from ideal 1D axial trajectories with increasing gas velocity, while the peak spread decreased with increasing gas velocity, it is reasonable to surmise that radial mixing was strongly enhanced at higher gas velocities with little impact on the apparent axial backmixing. This result is very encouraging for biomass pyrolysis since intense radial gas-solids mixing and heat transfer are required for proper feedstock conversion, but with as little axial backmixing as possible.

For the large PSD, the standard deviation trends generally decreased with increasing gas velocity. However, unlike with the small PSD, there was no significant impact of solids loading on RTD standard deviation. This result indicated that, regardless of the frequency of particle clustering, individual particles in the large PSD were sufficiently massive so that the drag force was significantly weaker than the gravity force. The experimental data for the large PSD were best fitted by power law trend lines, which indicated that the rate of increase of peak spread increased with vertical position, especially for \( z > 100 \) cm in the separator. Therefore, significant, undesirable axial backmixing occurred in the separator in the large PSD.
5.4.9.2 Solids RTD in Separator

Figure 5.14 shows the solids mean residence time and RTD coefficient of variation (COV) in the gas-solids separator for the small and large PSDs. The gas velocity was tested at three levels ($U_g = \{0.75, 1.1, 1.4\} \text{ m/s}$) and the solids loading varied in the range...
2 kg/kg ≤ \( \dot{m}_s / \dot{m}_g \) ≤ 46 kg/kg. At all tested values of \( U_g \), the following phenomena were observed:

- the separator mean residence time decreased with increasing solids loading as the frequency and / or size of particle clusters increased,
- the separator mean residence time decreased with increasing gas velocity as the downward drag force increased, and
- the separator mean residence time was almost always shorter for the large PSD than for the small PSD over all tested solids loading values due to the higher terminal velocity of the larger particles.

At \( U_g = 0.75 \) m/s, the mean residence time trends for both particle sizes appeared to converge. There are several possible explanations for this observed behavior, including:

- the frequency of particle clustering increased at a faster rate with increasing solids loading in the small PSD compared to the large PSD,
- particle backmixing decreased with increasing solids loading in the small PSD, and
- knowing that particles in the large PSD entered the separator at a higher velocity than in the small PSD, the average particle cluster size in the small PSD approached or exceeded the average size in the large PSD.

Opposite trends in the separator RTD COV with varying solids loading were observed between the two PSDs. The separator RTD peak spread decreased with increasing solids loading in the small PSD, yet increased with solids loading in the large PSD. This phenomenon remains in need of further study and explanation. The COV trends in the small PSD shifted downward on the vertical y-axis with increasing gas velocity, which indicated that backmixing decreased with increasing downward drag force. For the large PSD, the COV trends remained roughly equal with increasing gas velocity, which indicated that the drag force was insignificant relative to the gravity force for large particles.
5.4.10 Effect of Cone Size on Solids RTD

Figure 5.15 and Figure 5.16 show the effect of cone size on the solids mean residence time and RTD standard deviation, respectively, for the entire unit at $U_g = 1.1$ m/s and $\dot{m}_s/\dot{m}_g \sim 9$. There was very little observable difference in either the mean residence time
or the standard deviation in the separator ($z > 78$ cm) between any of the tested cone sizes.

![Graph](image)

Figure 5.15 – Effect of vertical position and separator cone size on solids mean residence time ($U_g = 1.1$ m/s; $\dot{m}_s / \dot{m}_g \sim 9$, $d_{P,avg} = 220$ µm, $L_0/D = 0$)

Referring to Figure 5.15, at $z = 69$ cm, the mean residence time was in line with a 1D transit curve predicted for $U_g = 1.9$ m/s, which was significantly larger than the actual superficial gas velocity $U_g = 1.1$ m/s. The high velocity of particles arriving at the photocell at $z = 69$ cm was likely due to the formation of a gas jet at the downer inlet and concentration of particles close to the downer centerline due to the inlet piping configuration. For $z > 92$ cm, which corresponded to the zone below the gas outlet, all data were in line with the predicted 1D transit curve for $U_g = 0.87$ m/s, which was significantly less than the actual $U_g = 1.1$ m/s. This indicated that significant solids
backmixing occurred between \( z = 69 \text{ cm} \) and the solids tank, likely due to particle-particle and particle-wall interactions and due to the complex gas flow pattern around the gas-solids separator. The most likely explanation for the apparent increase in backmixing and reduction in average downward solids velocity for \( z > 69 \text{ cm} \) is that the average particle velocity at the wall is low relative to the centerline in downers, and this difference increases with distance from the downer inlet, as shown clearly by Zhang et al. (2001). Particles slow down at the wall due mainly to the no-slip condition but also due to complex particle-particle interactions.

Referring to Figure 5.16, cone size was observed to have minimal impact on the solids RTD peak spread in the separator. In all cases, the peak spread increased linearly with height along the downer, and the standard deviation trends for all cone sizes were fitted by one unique linear regression. Similarly, referring to Figure 5.17, the mean separator residence time trend lines for all cone sizes were fitted by one identical trend line, whose adjustable parameters were equal according to an F-statistic test with a 5% level of significance. These results indicated that cone size had no impact on either the mean residence time or the extent of solids backmixing. Therefore, cone size had no observable impact on the solids RTD.

![Figure 5.16](image)

**Figure 5.16 – Effect of vertical position and cone size on solids RTD standard deviation**

\( (U_g = 1.1 \text{ m/s}; \: \bar{m}_s / \bar{m}_g \sim 9, \: d_{P, \text{avg}} = 220 \mu \text{m}, \: L_s / D = 0) \)
5.4.11 Gas-Solids Contact Time

Gas-solids contact time is an important parameter for predicting the pyrolysis reaction kinetics. One of the main purposes of measuring the solids RTD in thermochemical conversion processes is to estimate the gas / solids contact time. In this type of reaction, it is desirable to minimize the contact time in the separator. Although the “separator residence time” has a different meaning for the gas and solids phases since both phases exit the separator via different outlets, the RTD data from both phases can be used to estimate the contact time.

In Chapter 4, an idealized gas flow pattern model was proposed wherein the gas travels down the wall with the solids, gradually disengages from the solids and reverses direction, flows upward in the core, and then exits the separator via the gas outlet. This proposed gas flow pattern model was found to be in good agreement with the measured gas velocity distributions for all tested cone sizes. Assuming that the idealized gas flow pattern model was mostly accurate, a simple approximation to the time spent by the gas
in the separator before reversing direction is roughly half of the total residence time. In Chapter 3, it was shown that there was no significant difference in magnitude in the measured gas velocity along the wall versus the downer centerline. Therefore, the gas did probably spend around half of the total separator residence time flowing downward in contact with solids along the wall.

Figure 5.18 shows the full mean residence time for both the gas and solids phases in the large cone gas-solids separator as a function of the superficial gas velocity and for \( \dot{m}_s / \dot{m}_g \sim 15 \). A trend line is also shown indicating 50% of the full gas residence time in the separator. This 50% trend agreed very well with the measured solids mean residence time. Therefore, the solids mean residence gave a reasonable estimate of the gas-solids contact time in the separator for the 220 µm particles. However, for thermochemical conversion in the downer, there are additional considerations aside from the gas-solids contact time. The product gas may still react after disengaging with the solids since it must still reverse direction to exit the separator and to be quenched. This additional reaction time must be considered in the design and operation of the downer.
Figure 5.18 – Comparison of separator mean residence times for both gas and solids phases (\( \dot{m}_s / \dot{m}_g \sim 15 \), \( d_{P,avg} = 220 \mu m \), \( L_s/D = 0 \), large 60° cone)

5.5 Conclusions

In the present chapter, the solids RTD was measured using a phosphorescent tracer technique. Particle trajectories were tracked through the downer and separator using photocells mounted at five different positions along the downer height. A 1D particle trajectory model was developed to predict the mean residence time and was useful to identify zones in the downer and separator that deviated from ideal 1D particle flow. The effect of cone size on both the mean residence time and RTD standard deviation was found to be negligible. Therefore, since the large 60° cone performed better than all other separator designs on all other metrics, the large cone was deemed to be the most suitable separator for the downer pyrolysis process. Finally, the solids mean residence time in the separator gave a good estimate of the gas / solids contact time in the separator.

In general, the solids RTD behavior was strongly dependent on the particle size. For both tested particle sizes (220 \( \mu m \) and 620 \( \mu m \)), the mean residence time was observed to decrease with increasing solids loading, which was likely due to increased particle
clustering. For the small particle size group representative of the sand used in the pyrolysis process, the following conclusions are drawn:

- Gas velocity was found to have minimal impact on the particle mean residence time across the entire unit
- Particle mean residence time in the separator decreased with increasing gas velocity
- For all operating conditions, the rate of solids RTD peak spread was observed to increase linearly with height along the downer for $z > 69$ cm
- Small particles were much more dispersed and mixed than large particles, indicating the smaller particles’ strong sensitivity to gas mixing and turbulence

Large particles spent roughly half the time in the downer as spent by small particles under otherwise identical conditions. This result will likely have an important impact on the gas-solids contact, mixing, and heat transfer for either biomass and/or heavy oil pyrolysis in the downer. It is also an unfortunate result since larger biomass particles usually take longer to react in pyrolysis reactors. Solids RTD peak spread increased with distance along the downer for both particle sizes, though the solids RTD was much narrower in the large PSD. Radial mixing was surmised to be superior in the small PSD, which is desirable for the endothermic pyrolysis reaction. Therefore, smaller particle sizes are recommended for the downer pyrolysis process.

5.6 Notation

- $a$: Drag coefficient interpolation fitting parameter [--]
- $b$: Drag coefficient interpolation fitting parameter [--]
- $b_{bRe}$: Curve fitting parameter [--]
- $C_D$: Particle drag coefficient [--]
- $D$: Downer diameter [m]
- $d(t)$: Raw true phosphorescence decay function [--]
- $D(t)$: Phosphorescence decay identity function [--]
- $d_p$: Particle size [μm]
- $\bar{d_p}$: Average particle diameter [μm]
List of symbols:

- $E(t)$: RTD peak function [-]
- $F_b$: Buoyancy force [N]
- $F_d$: Drag force [N]
- $F_g$: Gravity force [N]
- $g$: Gravitational acceleration [m/s$^2$]
- $k$: Empirical phosphorescence decay constant [s$^{-1}$]
- $L$: Downer height [m]
- $m_p$: Particle mass [kg]
- $m_s/m_g$: Solids loading ratio [(kg/s)/(kg/s)]
- $Re_g$: Superficial gas Reynolds number [--]
- $Re_p$: Particle Reynolds number [--]
- $t$: Time [s]
- $U_g$: Superficial gas velocity [m]
- $U_p$: Particle velocity [m]
- $U_{slip}$: Particle slip velocity [m]
- $V_p$: Particle volume [$m^3$]
- $V_p$: Photocell voltage [V]
- $X_p(t)$: Photocell response to flash [V]
- $Y_{fit}(t)$: Fitted photocell response [V]
- $Y_p(t)$: Photocell response to activated tracer [V]
- $z$: Distance from flash unit [m]
- $\rho_g$: Gas density [kg/m$^3$]
- $\rho_p$: Particle density [kg/m$^3$]
- $\sigma$: RTD standard deviation [s]
- $\tau$: RTD mean residence time [s]
- $\phi$: Particle sphericity [--]
Chapter 6

6 Control of the Gas RTD Using Stripping Gas

In this chapter, stripping gas was introduced into the large 60° gas-solids separator between the gas outlet and solids collection tank in order to control the gas RTD in the separator. A CO\textsubscript{2} tracer technique was developed to measure the efficiency of the stripping process while varying several other operating parameters. The stripping efficiency and gas flow pattern stability were found to increase with the amount of stripping gas up to a maximum of 15 % by volume of the superficial gas flowrate. However, solids losses also increased with stripping gas flowrate. Stripping gas in amounts greater than 4 % by volume were effective at preventing the active gas volume from reaching the solids tank.

6.1 Introduction

As discussed in Chapter 1.9, stripping gas has mainly been used in the fluid catalytic cracking (FCC) process to recover entrained hydrocarbon vapor in the spent catalyst bed (Avidan et al., 1990, McKeen & Pugsley, 2003, Gao et al., 2008). It has also been applied in the fluid coking process to strip residual liquid hydrocarbons as well as for hydrocarbon vapor recovery (Bi et al., 2005, Cui et al., 2006). In both applications, the main issues of interest are to measure and model the multiphase hydrodynamics, mass and heat transfer, and stripping efficiency in order to optimize the stripper design and operating conditions. In the FCC process, stripping efficiencies greater than 80 % are normal (Gao et al.), while in the fluid coking process, stripping efficiencies greater than 99 % have been achieved (Cui et al.).

In the present chapter, stripping air was injected below the gas outlet, immediately upstream of the solids outlet, to achieve very simple stripping of the separated solids stream. In the actual pyrolysis process, steam would be used as the stripping gas. The stripping air used in the present chapter was also used to prevent the active gas volume from reaching the solids collection tank (i.e. to prevent gas underflow). Based on the results of the literature review performed in Chapter 1.9, the CO\textsubscript{2} tracer technique was
deemed to be the best method to assess the stripping efficiency due to the linear response of the sensor and with no need for sensor calibration. Therefore, a CO$_2$ tracer technique was used to measure the stripping efficiency. Internals were not tested in this chapter since the main purpose was to establish baseline stripping performance using the simplest possible configuration and to demonstrate the CO$_2$ tracer technique.

The main objectives of the present chapter were to:

- Demonstrate a simple, accurate CO$_2$ tracer technique for measuring the stripping efficiency
- Use tools and techniques developed in previous chapters to assess gas and solids backmixing when stripping gas is present
- Demonstrate the effectiveness of an integrated gas-solids separator / stripper
- Optimize the stripping gas conditions for:
  - Minimum gas backmixing
  - Most stable gas flow pattern
  - Minimum solids losses

6.2 Experimental

A detailed description of the stripping gas equipment and method used in the current chapter is provided in Chapter 2.9.

6.3 Results

6.3.1 CO$_2$ Tracer Technique for Stripping Efficiency Measurement

During each stripping efficiency experiment, stripping air was injected steadily at a measured flowrate. An upstep injection of CO$_2$ was then performed near the downer inlet while the transient CO$_2$ concentration was measured near the solids outlet. The measured CO$_2$ concentration at the solids outlet typically reached a final steady state value within 120 s after starting the steady CO$_2$ injection. Figure 6.1 shows transient CO$_2$ concentration curves during sample experiments for various stripping gas flowrates in the range of 0 to 23 % by volume of the downer air flowrate. The CO$_2$ molar concentration at
the gas outlet \((x_{CO2,go})\) was calculated on the basis of the combined total gas flowrate, and was given by

\[
x_{CO2,go} = \frac{\dot{n}_{CO2}}{\dot{n}_{CO2} + \dot{n}_{air} + \dot{n}_{str}},
\]

(6.1)

where \(\dot{n}_{CO2}\) was the CO\(_2\) molar flowrate,

\(\dot{n}_{air}\) was the downer air molar flowrate, and

\(\dot{n}_{str}\) was the stripping air molar flowrate.

As shown in Figure 6.1, the CO\(_2\) concentration at the solids outlet decreased with increasing stripping gas percentage \((\dot{n}_{str}/\dot{n}_{g} * 100\%)\). This indicated that the stripping gas was working to prevent the active downer air from reaching the solids outlet. With no stripping gas present, the transient CO\(_2\) concentration response was “underdamped” and oscillated significantly around the final steady state value. This result indicated an unstable gas flow pattern. As the stripping gas concentration was increased above zero, the measured transient CO\(_2\) concentration response behaved like an overdamped system and was very stable around the final steady state value, which indicated a stable gas flow pattern. The CO\(_2\) concentration at the solids outlet decreased with increasing stripping gas flowrate, which indicated that the stripping efficiency increased with the stripping gas flowrate.
Figure 6.1 – Sample experimental CO\(_2\) molar percentage signals at the solids outlet at various stripping gas percentage values \((U_g = 0.95 \text{ m/s}, z_{str} = 30 \text{ cm}, \text{ large } 60^\circ \text{ cone, } L_S/D = 0, \text{ no solids})\)

In each stripping efficiency experiment, once the CO\(_2\) concentration transient response settled to a final value, the steady state CO\(_2\) concentration was used to calculate the stripping efficiency. The stripping efficiency \((\eta_{str})\) was given by

\[
\eta_{str} = 1 - \frac{x_{CO_2,so}}{x_{CO_2,go}},
\]

where \(x_{CO_2,so}\) was the mole fraction of CO\(_2\) measured at the solids outlet, and \(x_{CO_2,go}\) was the mole fraction of CO\(_2\) calculated on the basis of the total gas flowrate in the gas outlet.

6.3.2 Stripping Efficiency Reproducibility

Figure 6.2 shows the effect of CO\(_2\) detector location on the measured stripping efficiency at \(U_g = 0.6 \text{ m/s}\) and with the stripping gas sparger located at \(z_{str} = 20 \text{ cm}\) below the gas outlet. In an initial set of experiments, the CO\(_2\) sampling line tip was located 35 cm below the gas outlet (a few centimeters into the solids tank) at the downer centerline. However, for practical reasons, CO\(_2\) detection was easier to perform from the downer wall at a port located 32 cm below the gas outlet, just upstream of the solids tank. Both sets of data shown in Figure 6.2 were fitted by a power law expression whose adjustable parameters
were equal according to an F-statistic test with a 5% level of significance. The results showed that the stripping efficiency reproducibility was acceptable between the two CO$_2$ detection locations. Therefore, all further stripping efficiency experiments were performed from the more convenient downer wall location 32 cm below the gas outlet.

Figure 6.2 – Effect of CO$_2$ detector line tip location on stripping efficiency

\[ \frac{\dot{n}_{str}}{\dot{n}_g} \times 100\% \]%

Figure 6.3 shows the reproducibility of a set stripping efficiency experiments performed at \( U_g = 1.2 \) m/s with the stripper gas sparger located at \( z_{str} = 30 \) cm. The reproducibility of the stripping efficiency with 0% stripping gas was poor since the gas flow pattern was unstable in this condition. A power law curve was fitted to all data and 95% confidence intervals calculated for the curve fit to give an estimate of the experimental uncertainty. When the stripping efficiency replicate data at 0% stripping gas were included in the curve fit, the average size of the 95% confidence interval was 5 stripping efficiency percentage points. However, when the replicates at 0% stripping gas were excluded from the curve fit, the reproducibility improved significantly, with the size of the confidence interval shrinking to 2 stripping efficiency percentage points.
6.3.3 Stripping Efficiency Experiments

Stripping gas experiments were performed with CO₂ tracer upstep injections to determine the effect of stripping gas percentage, stripping gas sparger position, downer gas flowrate, solids flowrate, and particle size on the stripping gas efficiency. Figure 6.4 shows the stripping efficiency plotted against the stripping gas percentage at three superficial gas velocities \( U_g = \{0.6, 0.95, 1.2\} \text{ m/s} \) and at three different stripping gas sparger locations \( z_{str} = \{8, 20, 30\} \text{ cm} \). Figure 6.5 shows the effect of solids loading on stripping efficiency. The effect of each test parameter on the stripping efficiency is discussed individually below.

6.3.3.1 Effect of Stripping Gas Sparger Height

As shown in Figure 6.4, the effect of stripping gas sparger height on stripping efficiency was much more significant than the stripping gas percentage or superficial gas velocity. The stripping efficiency improved dramatically from roughly 45 % to greater than 90 % as the stripping gas sparger moved downward from \( z_{str} = 8 \text{ cm} \) to \( z_{str} = 30 \text{ cm} \). With the

Figure 6.3 – Reproducibility of stripping efficiency experiments

\( (U_g = 1.2 \text{ m/s}, z_{str} = 30 \text{ cm}, \text{large } 60^\circ \text{ cone, } L_S/D = 0, \text{ no solids}) \)
stripping sparger located near the gas outlet at \( z_{str} = 8 \text{ cm} \), the stripping gas was very ineffective and strongly disturbed the overall gas flow pattern, resulting in very low stripping efficiencies on the order of 45 %. When the stripping sparger was located 5 cm above the solids outlet at \( z_{str} = 30 \text{ cm} \) (i.e. 5 cm above the solids outlet), the stripping gas was effective at preventing the downer gas from reaching the solids outlet and did not negatively affect the gas flow pattern, resulting in very high stripping efficiencies above 90 %. At \( z_{str} = 20 \text{ cm} \), intermediate stripping efficiencies around 80 % were observed. At each sparger height, similar stripping efficiency trends were observed between data sets with varying superficial gas velocity and stripping gas percentage.

### 6.3.3.2 Effect of Stripping Gas Percentage

As shown in Figure 6.4, in general, stripping efficiency was observed to increase with stripping gas percentage up to an asymptotic value, above which stripping gas flowrate no longer had an impact on efficiency. As the upward momentum of the stripping gas increased with the stripping gas flowrate, the active downer gas penetrated less toward the solids outlet, which resulted in increased stripping efficiency. For all data sets across all tested conditions, the stripping efficiency tended to approach an asymptotic value as the stripping gas percentage increased beyond 15 %. This result suggested a general rule that no more than 15 % stripping gas was required to achieve maximum stripping efficiency for any given set of operating conditions.

With the stripping sparger located at \( z_{str} = 8 \text{ cm} \), and with no stripping gas injected, the average measured stripping efficiency was 68 %. However, when the stripping gas flowrate increased from zero to 5 %, the stripping efficiency decreased sharply to a minimum average value of 40 %. The stripping efficiency then increased with stripping gas flowrate to an average asymptotic value of 48 % for \( \frac{\dot{n}_{str}}{\dot{n}_{g}} \geq 15 \% \). The decrease in efficiency relative to the zero stripping gas condition for all non-zero stripping gas values was likely due to a strong disruption to the gas flow pattern, which actually forced, rather than prevented, the downer gas toward the solids outlet. In any case, since most of the active downer gas flow turned toward the gas outlet above \( z = 8 \text{ cm} \), the stripping gas was still weakly effective at preventing active gas penetration to the solids outlet. Therefore,
the non-monotonic stripping efficiency trend at $z_{str} = 8$ cm appeared to be due to a trade-off between the harmful sparger location being too near the gas outlet and the general positive impact of increased stripping gas efficiency.

When the stripping sparger was shifted downward and away from the gas outlet beyond $z_{str} \geq 20$ cm, the stripping efficiency increased monotonically with stripping gas flowrate at all tested superficial gas velocities. For $z_{str} = 20$ cm, the stripping efficiency quickly reached an average asymptotic value of 82 % for $\dot{n}_{str}/\dot{n}_g > 5 \%$. With $z_{str} = 30$ cm, the stripping efficiency increased monotonically with stripping gas flowrate.

### 6.3.3.3 Effect of Superficial Gas Velocity

In general, as shown in Figure 6.4, stripping efficiency was observed to decrease with increasing superficial gas velocity. This was due to deeper active gas penetration toward the solids outlet with increasing downer gas flowrate, as previously demonstrated in Chapters 3 and 4. However, at $z_{str} = 30$ cm, a slight increase in stripping efficiency with increasing gas velocity was observed. This was likely due to near complete gas reversal upstream of $z_{str} = 30$ cm, and hence minimal disruption to the gas flow pattern at this sparger position. All data sets at $z_{str} = 30$ cm appeared to converge to the same stripping efficiency of 91 %. However, future work should confirm whether an asymptotic stripping efficiency does indeed exist for $z_{str} = 30$ cm.
6.3.3.4 Effect of Solids Loading

In Chapter 3, it was shown that the downer gas was entrained increasingly into the solids tank with increasing solids loading. Hence, one might expect the stripping efficiency to decrease with solids loading since more gas is reaching the solids tank. However, as shown in Figure 6.5, the stripping efficiency apparently increased with solids loading at all tested stripping gas flowrates. A possible explanation was that the filter tip on the CO₂ sampling line became caked with solids sooner in the run as the solids loading increased.
If the CO\textsubscript{2} sampling line filter were eventually plugged, gas sampling would fail and the CO\textsubscript{2} concentration measurement would be inaccurate (and likely too low since the CO\textsubscript{2} was injected in an upstep). However, the sampling line filter was not checked for caking nor was the gas flowrate in the CO\textsubscript{2} sampling line measured. Thus there was no evidence for the proposed explanation for the observed trends, which leaves the proposed explanation solely as speculation.

Figure 6.5 – Effect of solids loading and stripping gas percentage on stripping efficiency

\(\text{\(U_g = 0.60\, \text{m/s}, z_{str} = 30\, \text{cm}, \text{large 60° cone}, L_{S/D} = 0\)}\)

### 6.3.4 Effect of Stripping Gas on Solids Losses

Since the stripping gas flowed opposite the direction of the falling solids, some solids were expected to be entrained with the stripping gas, leading to a decrease in solids collection efficiency. Gas/solids separation efficiency experiments were thus performed to determine the magnitude of the impact of stripping gas on the solids losses. Figure 6.6 shows 9 total plots of solids collection efficiency experimental results. The collection efficiency was plotted against the solids tank fill level at three different superficial gas velocity levels \(U_g = \{0.8, 1.2, 1.5\}\, \text{m/s}\), at three different solids loading levels \(\dot{m}_s/\dot{m}_s = \{4, 9, 13\}\), and at three stripping gas percentage levels \(\dot{n}_{str}/\dot{n}_g = \{0, 5, 10\}\%\). The observed range of solids losses for all tests was 0.1\% to 0.6\%. 
Figure 6.6 – Effect of gas flowrate, solids flowrate, stripping gas percentage, and solids tank fill level percentage on solids losses (large 60° cone, $L_S/D = 0$)
As shown in Figure 6.6, solids losses always increased with solids tank fill level. With the solids tank level increasing after every run, particles splashing on the collected bed caused previously captured solids to be ejected from the bed. These ejected particles were more likely to be entrained back into the downer as the tank level rose. In general, solids losses also increased with gas flowrate and stripping gas flowrate. As the gas flowrate increased, the active gas reached deeper into the solids tank, thereby increasing the amount of entrained solids. The addition of stripping gas exacerbated this problem by reducing the downward drag force on the particles, thereby making particles more easily entrained in the upward flowing gas stream in the core. However, solids losses were always less than 0.6 %, which was deemed sufficient for the pyrolysis process.

At $U_g = 0.80 \text{ m/s}$, increased solids loading led to a moderate increase in solids losses. However, for all other tested gas velocities, solids losses were reduced with increasing solids loading. One possible explanation for the general decrease in solids losses with increasing solids loading was the higher frequency of particle-particle collisions and clustering. Particle clusters are easier to separate from the gas stream since their terminal velocity is higher. Overall, minimum solids losses of less than 0.2 % were observed for $U_g = 0.80 \text{ m/s}$ and $\dot{m}_s/\dot{m}_g = 4$, as shown in the top left plot of Figure 6.6. However, since gas backmixing was shown in previous chapters to be reduced at higher gas velocities, the next best set of conditions leading to low solids losses were considered. At $U_g = 1.5 \text{ m/s}$ and $\dot{m}_s/\dot{m}_g = 13$, the solids losses were always less than 0.3 % as shown in the bottom right plot in Figure 6.6. Higher gas velocity and higher solids loading lend themselves to process intensification and so these conditions were deemed the best for the pyrolysis reaction.

6.3.5 Effect of Stripping on the Gas RTD

An important consideration for the use of stripping gas was its impact on the gas RTD. Therefore, gas RTD measurements were performed using the pressure response technique described in Chapter 3. Figure 6.7 shows the effect of stripping gas flowrate on the active gas volume for $U_g = 1.2 \text{ m/s}$, with the stripping sparger located at its optimum position of $z_{str} = 30 \text{ cm}$. No solids were used in the gas RTD experiments in this chapter since the
stripping efficiency accuracy was shown to be in question with solids present. The active gas volume data and its scatter decreased with increasing stripping gas percentage, reaching a constant value above 6 % stripping gas. For \( \dot{n}_{str}/\dot{n}_g \leq 4 \% \), the scatter in the active gas volume was very large, which provided further indication of unstable gas flow. Furthermore, at low stripping gas flowrates, the active gas volume reached the solids tank. As the stripping gas flowrate increased to \( \dot{n}_{str}/\dot{n}_g \geq 6 \% \), the scatter in the data was very small and the active gas was prevented from reaching the solids tank. Therefore, the optimum range of stripping gas flowrates was 6 % to 15 %, recalling also that the stripping efficiency reached a plateau at 15 % stripping gas.

![Figure 6.7 – Effect of stripping gas percentage on active gas volume (\( U_g = 1.2 \) m/s, large 60° cone, \( L_g/D = 0 \), \( z_{str} = 30 \) cm, no solids)](image)

Figure 6.7 – Effect of stripping gas percentage on active gas volume (\( U_g = 1.2 \) m/s, large 60° cone, \( L_g/D = 0 \), \( z_{str} = 30 \) cm, no solids)

Figure 6.8 shows the effect of stripping gas flowrate on the gas RTD coefficient of variation (COV), i.e. the gas backmixing. Over the range \( 0 \leq \dot{n}_{str}/\dot{n}_g \leq 10 \% \), the stripping gas flowrate appeared not to have any impact on the gas RTD COV, which meant that there was no impact on RTD peak spread. In spite of the fact that the stripping gas did not reduce the extent of gas backmixing, several other benefits were obtained by the use of stripping gas as shown throughout this chapter.
Figure 6.8 – Effect of stripping gas percentage on gas RTD coefficient of variation ($U_g = 1.2$ m/s, large 60° cone, $L_s/D = 0$, $z_{str} = 30$ cm, no solids)

6.3.6 Effect of Stripping Gas on Gas Velocity Distribution

In order to demonstrate how the optimum stripping gas conditions interacted with the gas flow pattern below the gas outlet, local gas velocity measurements were performed using the thermistor technique described in previous chapters while injecting stripping gas. Figure 6.9 shows the effect of stripping gas flowrate on the normalized local gas velocity measured at $z = \{9, 14, 19, 26\}$ cm below the gas outlet for $U_g = 0.59$ m/s and with the stripping sparger located at $z_{str} = 30$ cm. The trends at $z = 9$ cm and $z = 26$ cm represented two extremes, with all other trends showing intermediate behavior between the two extremes. At $z = 26$ cm, a linear increase in $U^*$ with stripping gas was observed, which was due solely to the increase in stripping gas because so little downer air penetrated so far below the gas outlet. However, at $z = 9$ cm, the bulk of the gas was mainly downer air, and $U^*$ decreased with increasing stripping gas flowrate as the stripping gas displaced and slowed the downward momentum of the downer air flow. At $z = 14$ cm and $z = 19$ cm, the $U^*$ trends exhibited features of both extremes. At $z = 14$ cm, $U^*$ decreased with increasing stripping gas flowrate for $n_{str}/n_g \leq 4\%$. However, as the stripping gas flowrate increased, the downward gas momentum was sufficiently inhibited, and $U^*$ was
observed to increase with stripping gas flowrate for $\dot{n}_{str}/\dot{n}_g > 4\%$ due solely to the increase in stripping gas flowrate. The results shown in Figure 6.9 support the earlier conclusion that stripping gas should be injected as far below the gas outlet as possible to minimize the disruption to the gas flow pattern.

![Figure 6.9](image)

Figure 6.9 – Effect of stripping gas on normalized local gas velocity at various velocity sensor heights ($U_g = 0.59\, \text{m/s}, L_S/D = 0, z_{str} = 30\, \text{cm}, r/R_D = 0$)

### 6.3.7 Effect of Stripping Gas on Solids RTD

Solids RTD experiments were performed with stripping gas using the phosphorescent tracer technique described in Chapter 2.8. Figure 6.10 shows the solids mean residence time in the separator plotted against the solids loading for stripping gas flowrates of 0 and 10\%, with $U_g = 1.1\, \text{m/s}$ and $U_g = 1.4\, \text{m/s}$. At both superficial gas velocities, the solids mean residence time decreased with increasing solids loading. However, the stripping gas had no measurable impact on the measured solids mean residence time in the separator. At each tested superficial gas velocity, each data set for different stripping gas values were fitted by identical trend lines. The fitting parameters for the two curve fits at each gas velocity were equal according to an F-statistic test with a 5\% level of significance.
Since the gas residence time was shown to decrease with increasing stripping gas flowrate, the gas-solids contact time in the separator likely also decreased with increasing stripping gas percentage. Therefore, the use of stripping gas was a simple and effective method to control and stabilize the gas RTD and gas-solids contact with only a minor increase to the solids losses and no impact to the solids RTD.

Figure 6.10 – Effect of solids loading, gas flowrate, and stripping gas percentage on measured solids mean residence time in separator (large 60° cone, $L_S/D = 0, z_{str} = 30 \text{ cm}$)

6.4 Conclusions

In the present chapter, stripping gas was introduced below the gas outlet to minimize downer gas penetration near the solids outlet and to stabilize the gas flow pattern. The stripping gas performance was assessed on the basis of the stripping efficiency as well as its effect on the solids losses, gas RTD, gas flow pattern, and solids RTD. The stripping efficiency and overall gas flow pattern stability both increased with stripping gas flowrate up to about 15 % by volume of the downer gas flowrate. Above this threshold a plateau in stripping efficiency was reached. The stripping gas also successfully prevented the active gas from reaching the solids tank for stripping gas percentages greater than 6 %.
Therefore, the optimum range of stripping gas flow rates was 6 % to 15 % by volume of the downer gas flow rate. By maximizing the stripping efficiency, stabilizing the gas flow pattern, and preventing the active gas volume from reaching the solids tank, the gas RTD was greatly improved using stripping gas. Furthermore, from the standpoint of the pyrolysis reaction, even if the stripping gas had not reduced the active gas volume, it would still have a beneficial effect by reducing the amount of hydrocarbon vapors reaching the lowest part of the downer. This would help to minimize the excessive production of undesirable non-condensable gases by hydrocarbon vapor overcracking.

Solids losses generally increased with both the stripping gas and downer gas flowrates, and generally decreased with increasing solids loading and solids tank fill level. In all cases, the solids losses never exceeded 0.6 %, which was deemed adequate for the pyrolysis process. In order to minimize gas backmixing and to maximize the benefits of stripping gas, the superficial gas velocity should be operated above 1.5 m/s and the solids loading greater than 10 kg/kg. The stripping gas was found to have no impact on the solids RTD. In general, the use of stripping gas was a simple and effective method to control and stabilize the gas RTD and gas-solids contact with only a minor increase to the solids losses and no impact to the solids RTD.

### 6.5 Notation

<table>
<thead>
<tr>
<th>Symbol</th>
<th>Description</th>
<th>Unit</th>
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</thead>
<tbody>
<tr>
<td>COV</td>
<td>RTD coefficient of variation [-]</td>
<td></td>
</tr>
<tr>
<td>D</td>
<td>Downer diameter [m]</td>
<td></td>
</tr>
<tr>
<td>(d_p)</td>
<td>Average particle diameter [(\mu m)]</td>
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</tr>
<tr>
<td>(E(t))</td>
<td>RTD peak function [-]</td>
<td></td>
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<tr>
<td>L</td>
<td>Downer height [m]</td>
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<tr>
<td>(\dot{m}_s/\dot{m}_g)</td>
<td>Solids loading ratio [(kg/s)/(kg/s)]</td>
<td></td>
</tr>
<tr>
<td>(\dot{n}_s)</td>
<td>Air molar flowrate [mol/s]</td>
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</tr>
<tr>
<td>(\dot{n}_{CO2})</td>
<td>CO(_2) molar flowrate [mol/s]</td>
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<tr>
<td>(\dot{n}_{str})</td>
<td>Stripping air molar flowrate [mol/s]</td>
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</tr>
<tr>
<td>(\dot{n}_{str}/\dot{n}_g \times 100%)</td>
<td>Stripping gas percentage [%]</td>
<td></td>
</tr>
<tr>
<td>Symbol</td>
<td>Description</td>
<td>Units</td>
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<tr>
<td>----------</td>
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</tr>
<tr>
<td>$\text{Re}_g$</td>
<td>Superficial gas Reynolds number</td>
<td>[--]</td>
</tr>
<tr>
<td>$t$</td>
<td>Time</td>
<td>[s]</td>
</tr>
<tr>
<td>$U_g$</td>
<td>Superficial gas velocity</td>
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<tr>
<td>$U^*$</td>
<td>Normalized local gas velocity</td>
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<td>CO$_2$ molar concentration at gas outlet</td>
<td>(mol/s)/(mol/s)</td>
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<tr>
<td>$x_{\text{CO}_2,so}$</td>
<td>CO$_2$ molar concentration at solids outlet</td>
<td>(mol/s)/(mol/s)</td>
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<tr>
<td>$z$</td>
<td>Distance below gas outlet</td>
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</tr>
<tr>
<td>$z_{str}$</td>
<td>Stripping gas sparger distance below gas outlet</td>
<td>[m]</td>
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<tr>
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<td>Solids collection efficiency</td>
<td>[--]</td>
</tr>
<tr>
<td>$\eta_{str}$</td>
<td>Stripping efficiency</td>
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</tr>
<tr>
<td>$\sigma$</td>
<td>RTD standard deviation</td>
<td>[s]</td>
</tr>
<tr>
<td>$\tau$</td>
<td>RTD mean residence time</td>
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Chapter 7

7 Conclusions & Recommendations

This chapter presents the main conclusions drawn from the work reported in this thesis and provides recommendations for future work.

7.1 Conclusions

The work reported in this thesis was based on and greatly extended the author’s Master’s thesis work and related publications on gas-solids separation in ICFAR’s novel gas-solids separator (Huard, 2009). Several, though not all, of the recommendations by Huard were implemented and investigated in this thesis, with particular focus placed on gas RTD measurements and backmixing in the separator. A literature review was performed to identify the most relevant metrics and methods by which to comprehensively assess the performance of ICFAR’s integrated gas-solids separator / stripper. Based on the review, a need was identified to develop a better gas RTD measurement technique that was less costly and less complex than radioactive tracer and avoided errors associated with tracer sampling. Using a novel pressure-response-based gas RTD measurement technique that satisfied the need for a better RTD method, and several other conventional experimental and analytical methods, the separator / stripper design and performance was assessed and optimized on the basis of: solids collection efficiency, gas and solids backmixing via gas and solids RTD measurement, and stripping efficiency. The effects of several main process design and operating parameters on all separator performance metrics were also investigated, including: separator geometry, bulk downer gas flowrate, stripping gas flowrate, downer gas flow regime, solids flowrate, and particle size. Local gas velocity measurements in the separator were also performed at several stages of the work to explain and confirm proposed gas flow patterns and gas-solids separation phenomena.

The following list summarizes the main findings in this thesis:

1. The transient separator pressure drop during helium tracer downstep injections was successfully correlated to the transient flowrates of helium tracer, air, and solids, and hence to the gas RTD, by a pressure drop regression and gas mixture
composition model. The model was able to predict complex gas flows during the helium tracer downstep experiments.

2. Gas RTD experiments demonstrated that the active volume of non-stagnant gas increased with the gas flowrate for most separator designs. At high gas flowrates, the active gas penetrated far below the gas outlet into the solids collection tank. Gas entrainment into the solids tank increased on the whole in the presence of solids versus without solids, but the active gas volume generally decreased with increasing solids flowrate. For the large 60° cone, gas RTD peak spread and gas backmixing decreased in a power law fashion with increasing gas flowrate.

3. Local gas velocity measurements below the gas outlet showed that there was no distinct gas penetration length below the gas outlet. The active gas volume was shown to increase exponentially with the gas velocity in the gap surrounding the separator cone.

4. The large 60° cone performed better than all other tested separator designs in terms of maximum solids collection efficiency (> 99.9% for 180 µm Sauter mean diameter sand), least deviation from the theoretical mean residence time trend, and smallest gas RTD peak spread (i.e. least backmixing). Two main separator design classes were tested, which included shielded and unshielded gas outlets. Among the shielded gas outlet separators, three different cone deflector diameters were tested.

5. The separator performance, quantified objectively by the Separator Performance Index (SPI) developed in this thesis, was found to be strongly dependent on the separator cone diameter. Separator performance increased with cone size. As the cone size increased, the gas outlet was better shielded from oncoming particles, and a higher gas velocity was induced in the gap between the cone rim and the downer wall. Greater gap velocities resulted in deeper gas penetration below the gas outlet, which promoted turbulence and mixing and helped to prevent stagnant gas zones.

6. Solids RTD experiments showed that the cone diameter had no impact on the solids mean residence time or backmixing in the separator. As such, the large 60° remained the best separator design on all performance metrics. Furthermore, for
an average particle size of 220 µm, the gas-solids contact time in the large cone separator was approximated closely by the solids mean residence time in the separator. At high superficial gas velocities, the gas-solids contact time in the separator was on the order of 0.5 s.

7. The mean residence time and active gas volume in the large 60° cone separator were greatly reduced and the separator gas flow pattern stability increased with the injection of stripping gas just upstream of the solids outlet. The optimum stripping gas flowrate range was 6 % to 15 % by volume of the downer gas flowrate to achieve significant active volume reduction and high stripping efficiencies greater than 90 %. A minor (though acceptable) penalty on the solids collection efficiency was incurred by using stripping gas.

8. Particle size had a very significant impact on all aspects of separator performance. The solids collection efficiency of fine particles smaller than 45 µm was 5 % to 20 % higher in a large 620 µm-average PSD compared to a small 220 µm-average PSD. The gas RTD was found to be much more stable and the active gas volume smaller with a large 620 µm-average PSD compared to a small 220 µm-average PSD. However, based on the results of the solids RTD experiments, radial mixing was surmised to be far superior with the small PSD, which is advantageous for heat and mass transfer in the endothermic pyrolysis reaction. Therefore, the use of stripping gas is crucial to minimize the negative effects of the small PSD on the gas RTD, thereby allowing for use of the small PSD in the downer.

7.2 Recommendations for Optimum Separator Performance & Future Work

In order to maximize solids collection efficiency and to minimize excessive vapor reaction time and backmixing in the separation zone, the large 6.3 cm rim diameter, 60° cone separator should be used in the downer pyrolysis process. Optimum separator performance can be achieved under all of the following conditions implemented together:

a. Cone separation length $L_s/D = 0$ (i.e. cone rim in line with the gas outlet)

b. By operating the downer in the fully turbulent flow regime ($U_g > 0.85$ m/s for the 7 cm diameter downer tested), which lends itself to process intensification
c. High solids-to-gas mass loading ratios greater than 10 kg/kg, which lends itself to process intensification

d. Stripping gas flowrates in the range of 6 % to 15 % by volume of the downer gas flowrate with the stripping gas sparger located 30 cm below the gas outlet

e. A narrow particle size distribution with an average size around 200 µm to enhance radial mixing in the downer but to avoid excessive solids losses in the separator.

The following recommendations are proposed for further potential increases in performance to the large 60° cone separator:

- The distance between the gas and solids outlets should be varied, but preferably extended. By extending the distance between the gas and solids outlets coupled with the use of stripping gas, the active gas volume may be prevented from reaching the solids collection tank, along with minimizing gas backmixing.

- The gas outlet diameter should be varied and optimized to determine its effect on separator pressure drop, gas velocity distribution, gas flow pattern, and gas RTD. As tested, the separator pressure drop in the 0.95 cm diameter gas outlet was dominated by the kinetic pressure drop at the entrance to the gas outlet. A change to the kinetic pressure drop would be expected to result in changes to the gas velocity distribution, gas flow pattern, and consequently the gas mixing and RTD.

- Gas RTD experiments should be performed at $\text{Re}_g \sim 10^4$ with and without stripping gas to confirm the increasing active gas volume and decreasing gas RTD peak spread trends with increasing gas flowrate. In addition to a potential increase in separator performance brought on by enhanced turbulence and mixing, further process intensification may be possible in the downer, thereby increasing unit throughput.

- Gas RTD experiments should be performed in a metallic downer, which would allow for the effects of pressure, temperature, and gas density to be investigated.
References


Appendix A – Gas Velocity & Solids Concentration Probe Calibration

A 10 kΩ, 0.12 cm bead diameter Murata NTC thermistor was used as a heat flux sensor to measure the local gas velocity and local solids loading in the gas-solids separator. The thermistor had a measured response time of 0.14 s. An OMEGA® K-type thermocouple was used in combination with the thermistor to measure the local temperature of the gas-solids mixture during local solids loading experiments.

To measure the local gas velocity, the thermistor voltage \( V_s \) and resistance \( R_s \) were measured along with the gas freestream temperature \( T_\infty \) using a thermocouple. These measurements were used to interpolate in thermistor resistance versus gas velocity and temperature calibration regressions. Figure A.0.1 shows the calibration data used to develop regression equations between the superficial gas velocity \( U_g \), \( R_s \), and \( T_\infty \). The calibration data were fitted well by an expression of the form:

\[
U_g = b_{UR} \left[ 1 - \exp \left( \frac{R_i - R_s}{d_{UR}} \right) \right]^2, \quad (\text{A.0.1})
\]

Where: 
\( b_{UR} \) was a fitting constant,
\( d_{UR} \) was a temperature calibration parameter, and 
\( R_i \) was a reference thermistor resistance accounting for the gas temperature.

The reference thermistor resistance \( R_i \) was given by the expression:

\[
R_i = c_{UR} + d_{UR} \ln \left( 1 - \sqrt{2}/2 \right), \quad (\text{A.0.2})
\]

where \( c_{UR} \) was a temperature calibration parameter.
Values for the temperature calibration parameters \( c_{UR} \) and \( d_{UR} \) were obtained at each tested freestream temperature shown in Figure A.0.1 by fitting Equations (A.1) and (A.2) to the calibration data. Both \( c_{UR} \) and \( d_{UR} \) were fitted reasonably well by linear curve fits. The linear calibration equations are shown in Figure A.0.2.
Local solids concentrations were measured by comparing the difference in heat transfer from the thermistor to the flow media between gas-only and gas plus solids operations. During each solids loading experiment, the thermistor power \(Q\) was first measured during gas-only operation, then again during solids injection. Since the gas and solids entered the unit at different temperatures, the thermistor power during solids injection was reported relative to the gas-only condition. Furthermore, since the gas and solids did not enter the unit at the same temperature between runs, the change in thermistor power was made specific to the difference in thermistor temperature between gas-only and gas plus solids conditions,

\[
\frac{\Delta Q}{\Delta T_s} = \frac{(\text{Gas/solids mixture thermistor power}) - (\text{Gas only thermistor power})}{(\text{Gas/solids mixture thermistor temperature}) - (\text{Gas only thermistor temperature})}
\]

\[
\frac{\Delta Q}{\Delta T_s} = \frac{Q_2 - Q_1}{T_{i2} - T_{i1}} = \frac{V_2^2/R_2 - V_1^2/R_1}{f(R_2) - f(R_1)},
\]

where \(\Delta Q\) is the change in thermistor power, and
\(\Delta T_s\) is the change in thermistor temperature.

\(V_{i1}\) and \(R_{i1}\) are the thermistor voltage and resistance for gas-only flow, and
\( V_{s2} \) and \( R_{s2} \) are the thermistor voltage and resistance for gas plus solids flow.

The thermistor surface temperature was determined by interpolation from a thermistor resistance versus temperature calibration. The calibration was performed by immersing the thermistor in a stirred beaker full of cold municipal tap water. The water beaker was placed on a small hot plate to slowly heat the water. Water temperature was measured at 30 second intervals from a digital readout using a thermocouple immersed at the same location in the beaker as the thermistor. The thermistor resistance was measured at one (1) second intervals as the water beaker temperature increased slowly from 12 °C to 48 °C over a time period of 35 minutes, which resulted in a heating rate of roughly 1 °C/minute. The response time (time to reach 63% of the steady state value) of the thermocouple was roughly 10 seconds. Therefore, at each temperature reading every 30 seconds, which corresponded to three time constants, the thermocouple read roughly 95% of the full change in the measurement period, which was deemed adequate.

Figure A.0.3 shows the thermistor resistance versus ambient temperature calibration data. The data were fitted very well by an expression of the form:

\[
R_s = a_{RT} e^{b_{RT} T_s}, \tag{A.0.3}
\]

where \( a_{RT} \) and \( b_{RT} \) were empirical calibration constants.

![Thermistor resistance versus ambient temperature calibration](image-url)
Appendix B – Volume Measurement Uncertainty Analysis

The combined volume of the separator, solids tank (at various fill levels), and part of the gas outlet pipe was measured by injecting a small sample of air with measured initial pressure (~ 300 kPa) and known volume (1.05 L) into the sealed separation zone (occupied by air at ~ 100 kPa) and measuring the change in pressure in both the sealed separator and the sample volume vessel. The volume of the separation zone was then calculated by Avogadro’s Law,

\[ V = \frac{\Delta P_{\text{samp}}}{\Delta P_{\text{sep}}} V_{\text{samp}} \]  \hspace{1cm} (B.0.1)

where \( V \) is the separation zone volume,
\( \Delta P_{\text{samp}} \) is the change in pressure of the sample volume,
\( \Delta P_{\text{sep}} \) is the change in pressure of the separation zone, and
\( V_{\text{samp}} \) is the sample volume.

The uncertainty in \( V \) was estimated by calculating the differential using the quotient rule:

\[ \delta V = \left[ \left( \delta \Delta P_{\text{samp}} \right) V_{\text{samp}} + \Delta P_{\text{samp}} \left( \frac{\delta V_{\text{samp}}}{V_{\text{samp}}} \right) \right] \Delta P_{\text{sep}} + \Delta P_{\text{samp}} V_{\text{samp}} \left( \frac{\delta \Delta P_{\text{sep}}}{\Delta P_{\text{sep}}} \right). \] \hspace{1cm} (B.0.2)

where \( \delta \) indicates the uncertainty in the measured quantity.

The sample volume was measured by filling the sample vessel with water and emptying into the contents into a graduated cylinder. The uncertainty in the sample volume was estimated to be one graduation of the cylinder, which was 50 mL.

The uncertainty in \( \Delta P_{\text{samp}} \) and \( \Delta P_{\text{sep}} \) were estimated by calculating the confidence intervals in the measured steady state pressure signals before and after the sample volume was injected into the separator vessel. For pressure measurements, the 95% confidence interval is given by:
\[ \delta \Delta P = 1.96 \left( \frac{\sigma_{p1}}{\sqrt{n_{p1}}} + \frac{\sigma_{p2}}{\sqrt{n_{p2}}} \right), \]  

(B.0.3)

where \( \sigma \) is the standard deviation of the pressure signal,

\( n \) is the number of pressure measurements, and

subscripts 1 and 2 refer to before and after sample injection, respectively.

For the example volume measurement shown in Figure B.0.1, the uncertainties in the pressure measurements were \( \delta \Delta P_{\text{samp}} = 375 \) Pa and \( \delta \Delta P_{\text{sep}} = 149 \) Pa. Therefore, the uncertainty in the separator volume was

\[ \delta V = \left[ 375 \times 1.05 \times 10^{-3} + 2.441 \times 10^3 \times 5 \times 10^{-5} \right] \times 2.115 \times 10^3 + 2.441 \times 10^3 \times 1.05 \times 10^{-3} \times 149 \]

\[ \delta V = 0.681 \text{L} \]

Since the separator volume was 11.9 L in the example, the uncertainty in the measurement was \( \pm 0.681 \) L, or 5.7 % of the total value. The uncertainty in all volume measurements was less than 6 %.

Figure B.0.1 – Example pressure signals during separator volume measurement
Appendix C – Derivation of Transient Tracer Flowrate during Downstep Experiment

During a gas RTD downstep experiment, a steady flow of tracer was abruptly cut off using a fast-acting solenoid valve. However, due to accumulation of tracer in the injection line between the solenoid valve and the tracer sparger inside the downer, the actual injection of tracer was not perfectly abrupt and was significant relative to the mean gas residence time in the separator. Therefore, the transient injected tracer mole flowrate was modeled as a function of the tracer sparger pressure drop in order to convolve with the gas RTD function to get the modeled outlet tracer flowrate.

Figure C.0.1 illustrates the tracer injection setup used in gas RTD experiments. The pressure of the tracer gas, composed solely of helium, was measured just downstream of the solenoid valve as well as in the downer just downstream of the tracer sparger. The pressure drop across the tracer sparger ($\Delta P_{spg}$) was treated as fluid flow across an orifice, whose pressure drop was given by

$$\Delta P_{spg} \propto \rho U_{spg}^2,$$  \hspace{1cm} (C.1)

where $\rho$ is the gas density, and $U_{spg}$ is the gas velocity at the sparger holes.

The gas velocity at the sparger holes was proportional to the tracer mass flow rate ($\dot{m}_{he,i}$) and the gas density by

$$U_{spg} \propto \frac{\dot{m}_{he,i}}{\rho}.$$  \hspace{1cm} (C.2)

Therefore,

$$\Delta P_{spg} \propto \frac{\dot{m}_{he,i}^2}{\rho}.$$  \hspace{1cm} (C.3)

Assuming that the tracer behaves as an ideal gas,

$$\rho = \frac{PM}{RT},$$  \hspace{1cm} (C.4)

where $P$ is the gas pressure at the sparger holes, $M$ is the gas molecular weight,
\( R \) is the universal gas constant, and 
\( T \) is the gas temperature.

Substituting (C.3) into (C.4),
\[
\Delta P_{spg} \propto \frac{\dot{m}_{he,i}^2 RT}{P M} = \frac{\dot{n}_{he,i}^2 RT}{P},
\]  
(C.5)
where \( \dot{n}_{he,i} \) was the tracer mole flowrate.

The pressure drop in the tracer injection line can be neglected relative to the tracer sparger pressure drop. Therefore, the pressure \( (P_1) \) was approximately equal to the pressure at the sparger holes \( (P_{spg}) \),
\[
P_{spg} \Delta P_{spg} \propto \dot{n}_{he,i}^2 RT.
\]  
(C.6)
Since \( P_{spg} = \Delta P_{spg} + \Delta P_{sep} + P_{atm} \), and introducing a constant of proportionality \( (K) \) accounting for the gas constant \( (R) \) and the approximately constant temperature,
\[
(\Delta P_{spg} + \Delta P_{sep} + P_{atm}) \Delta P_{spg} = K \dot{n}_{he,i}^2 = K \left( \frac{dn_{he,i}}{dt} \right)^2.
\]  
(C.7)
Rearranging (C.7),
\[
\frac{dn_{he,i}}{dt} = \frac{1}{\sqrt{K}} \sqrt{\Delta P_{spg}(t) + \Delta P_{sep}(t) + P_{atm}} \Delta P_{spg}(t).
\]  
(C.8)
Therefore, the transient injected tracer mole flowrate \( (\dot{n}_{he,i}) \) was simply a function of the known, measured pressures \( \Delta P_{sep} \) and \( \Delta P_{spg} \).
Just prior to the downstep at $t = 0$, the injected tracer mole flowrate was steady (i.e. constant) and known; therefore,

$$K = \left( \frac{P_{spg} + \Delta P_{sep} + P_{atm}}{\dot{n}_{he,i}} \right) \Delta P_{spg} \bigg|_{t=0}.$$  \hfill (C.9)

Next, assuming again that the tracer behaves as an ideal gas,

$$n_{he,i} = \frac{P_{spg} \nu_{spg}}{RT},$$ \hfill (C.10)

where $\nu_{spg}$ was the volume of the tracer injection line downstream of the solenoid valve combined with the volume of the tracer sparger.

Differentiating with respect to time to get the mole flowrate,

$$\frac{dn_{he,i}}{dt} = \frac{1}{RT} \left( \frac{dP_{spg}}{dt} \nu_{spg} + \frac{d\nu_{spg}}{dt} \right).$$ \hfill (C.11)

Since the tracer injection line and sparger volumes were constant,

$$\frac{dn_{he,i}}{dt} = \frac{\nu_{spg}}{RT} \frac{dP_{spg}}{dt}.$$ \hfill (C.12)

Substituting (C.10) into (C.7) and rearranging,

$$\left( P_{spg} + \Delta P_{sep} + P_{atm} \right) \Delta P_{spg} = K \left( \frac{\nu_{spg}}{RT} \right)^2 \left( \frac{d(\Delta P_{spg})}{dt} \right)^2 \frac{RT}{\nu_{spg}} \sqrt{\frac{\left( \Delta P_{spg} + \Delta P_{sep} + P_{atm} \right) \Delta P_{spg}}{K}}.$$ \hfill (C.13)

Substituting (C.9) into (C.13) and rearranging,

$$\frac{d(\Delta P_{spg})}{dt} = \frac{\dot{n}_0 RT}{\nu_{spg}} \sqrt{\frac{\left( \Delta P_{spg} + \Delta P_{sep} + P_{atm} \right) \Delta P_{spg}}{(\Delta P_{spg,0} + \Delta P_{sep,0} + P_{atm}) \Delta P_{spg,0}}},$$ \hfill (C.14)

where $\dot{n}_0$ was a shorthand for the constant injected tracer flowrate at $t = 0$,

$\Delta P_{spg,0}$ was a shorthand for the constant sparger pressure drop at $t = 0$, and

$\Delta P_{sep,0}$ was a shorthand for the constant separator pressure drop at $t = 0$. 
Finally, substituting (C.12) into (C.14),

\[
\dot{n}_{he,i}(t) = \frac{d\dot{n}_{he,i}}{dt} = \dot{n}_0 \left[ \frac{\Delta P_{spg}(t) + \Delta P_{sep}(t) + P_{am}}{(\Delta P_{spg,0} + \Delta P_{sep,0} + P_{am})\Delta P_{spg,0}} \right].
\] (C.15)

Returning to (C.13), by integrating numerically over the sparger pressure drop from onset to completion of the downstep, the total time required for all tracer to be injected \((t_{\text{step}})\) was obtained. The solution to (C.14) was given by

\[
t_{\text{step}} = \sqrt{K} \int_0^{\Delta P_0} \frac{1}{\Delta P_{spg} + \Delta P_{sep} + P_{am}} d(\Delta P_{spg}).
\] (C.16)

A specified step size of \(\Delta(\Delta P_{spg})\) equal to 0.1% of the maximum sparger pressure drop occurring at \(t = 0\) was sufficient to give step size independent results. Finally, the tracer mole flowrate \((\dot{n}_{he,i})\) was then calculated using Equation (C.12).
Appendix D – Qualitative Comparison of Separator Pressure Drop & Gas RTD of Several Separator Designs

This appendix describes the development of a pressure drop regression for several different gas-solids separator designs. Sample transient separator pressure drop signals for various separator designs are compared to qualitatively assess backmixing in the various separator designs.

A.1 Separator Pressure Drop Regression

The separator pressure drop regression described in Chapter 3.4.1 was used to model the separator pressure drop of the tested separator designs illustrated in Figure 4.1. As shown in Figure D.0.1, the air-only pressure drop for three selected separator designs was fitted well by the air-only contribution to the separator pressure drop model expression given by Equation (2.20):

\[
\Delta P_{sep} = C_1 \frac{n_{air}^2 M}{P_{sep}}. \tag{0.1}
\]

The designs for which data is shown in Figure D.0.1 were chosen to represent the maximum range in the separator pressure drop, and thereby demonstrate the applicability of the regression to all tested separator designs. Based on the results in Figure D.0.1, the fitting parameter \( C_1 \) was found to be statistically equivalent for the tube outlet and the large 60° cone based on an F-statistic test with a 5% significance level. Therefore, it was concluded that the additional kinetic pressure drop across the cone gap was statistically insignificant relative to the kinetic pressure drop at the gas outlet tube. Thus, the presence of a cone had no relevant impact on the separator pressure drop.
2.2 Qualitative Comparison of Pressure Response and Tracer Flowrate in Tested Separator Designs

Figure D.0.2 shows experimental separator pressure drop signals during tracer downstep experiments for all tested separators except for the tube-in-tube outlet at identical conditions. The superficial gas velocity was $U_g = 0.9 \text{ m/s}$ (Re$_g \sim 4500$), while the separation length was $L_S/D = 0$, and no solids were present. A qualitative description of the state of mixing in the separator can be made from the slope of the $\Delta P_{sep}$ curve since the tracer outlet molar flowrate is proportional to $d(\Delta P_{sep})/dt$. Longer times required for the separator pressure drop to settle to steady state corresponded roughly to longer mean residence times and higher active gas volumes. Large changes in $d(\Delta P_{sep})/dt$ over the course of the run corresponded to fluctuations in the tracer molar flowrate, which indicated a large spread (i.e. backmixing) in the RTD. Based on the sample signals shown in Figure D.0.2, the RTDs for all of the separator types could be expected to be similar except for the 60° small cone, where $d(\Delta P_{sep})/dt$ took noticeably more time to settle to steady state than for the other separator designs.
Figure D.0.2 – Comparison of experimental separator pressure drop during tracer downstep for all tested separator types at identical conditions ($U_g = 0.9$ m/s, $Re_g \approx 4500$, $L_g/D = 0$, no solids)

Figure D.0.3 shows the tracer outlet molar flowrate predicted by the gas mixture composition model during the same downstep experiments at the same identical conditions described above. Qualitative differences between separator designs were more readily apparent when looking at transient tracer outlet flowrate signals. At $U_g = 0.9$ m/s, the small and medium 60° cones and bell-shaped separator showed long secondary tracer peaks leaving the system over the period of 0.5 s to 2 s after the downstep, which indicated significant backmixing. The tracer outlet flowrate signals for the tube outlet, sparger outlet, and large 60° cone were very similar to each other. Small secondary peaks were observed indicating little backmixing. The secondary peaks were narrowest in the large 60° cone and tube outlet. Therefore, the tube outlet and large 60° cone were expected to have the least gas backmixing.
Figure D.0.3 – Comparison of predicted tracer outlet molar flowrate during downstep for all tested separator types at identical conditions ($U_g = 0.9$ m/s, $Re_g \sim 4500$, $L_g/D = 0$, no solids)
Appendix E – Fast Pyrolysis Process Intensification: Study of the Gas Phase Residence Time Distribution and Backmixing in a Downer Reactor

E.1 Introduction

The Institute for Chemicals and Fuels from Alternative Resources (ICFAR) is developing and testing reactor technology for the conversion of biomass and heavy oil feedstocks to useful bio-oil, bio-char, syngas, and other valuable chemical products via pyrolysis. Among the various biomass conversion processes developed at ICFAR, a downer reactor was designed and manufactured for the pyrolysis of biomass and heavy oil feedstocks to maximize the liquid yield. The downer configuration was selected over other reactor types for careful control of thermal cracking reactions and gas-solid contact times. To achieve these goals, a novel gas-solids separator was developed and tested (Huard, 2009; Huard et al., 2010) specifically for the downer to quickly and efficiently separate heat-bearing sand particles from product vapors. The purpose of this study is to ensure that the separator does not introduce excessive gas backmixing, which would degrade the downer performance.

Several gas phase RTD studies have previously been performed in circulating fluidized bed (CFB) riser reactors (Dry & White, 1989; Gauthier, 1991; Smolders & Baeyens, 2000; Gauthier et al., 2005). Most authors reported significant gas backmixing in the riser and increased backmixing with increasing solids flux. However, only Brust and Wirth (2004) measured gas backmixing in a downer reactor and found that backmixing was reduced at high gas velocities. Hence, in the present study, the gas residence time

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3 A version of this chapter has been presented orally at The 14th International Conference on Fluidization and published in the conference proceedings as follows:

distribution (RTD) and backmixing in the downer reactor and around the gas-solids separator were investigated in the absence of solids. A simple hot wire anemometry technique was developed to measure the concentration of helium tracer in the model downer and around the gas-solids separator. Some initial results of the study are presented to demonstrate the applicability of hot wires in measuring the gas phase RTD.

E.2 Background

Knowledge of the RTD in a reactor may be obtained by measuring the concentration of a tracer in the reactor as time proceeds. Thin filament hot wire anemometers were used in this study to detect the presence of helium tracer in air. These sensors detect differences in the heat transfer characteristics of gas flow over the thin electrified resistive film or filament. Heat generated by the resistive element must be dissipated by the flowing gas, which results in decreased detector resistance or voltage. For fluid flow past a resistive element, the heat balance for the hot wire is given by

\[ \frac{V_w^2}{R_w} = h_w A_{ws} (T_w - T_g), \]  

(1)

where \( V_w \) is the voltage applied to the detector, \( R_w \) is the resistance of the detector, \( h_w \) is the heat transfer coefficient between the detector and the surrounding fluid, \( A_{ws} \) is the heat transfer surface area of the detector, \( T_w \) is the temperature at the surface of the detector, and \( T_g \) is the gas temperature.

Variation in gas composition as the gas flows over the hot wire is detected in the constant-voltage mode as a change in the probe resistance \( R_w \). For most hot wire filaments, the resistance is linearly related to the probe temperature in the form

\[ R_w = m \Delta T + b, \]  

(2)

where \( \Delta T = T_w - T_g \) and \( m \) and \( b \) are empirical coefficients of resistivity specific to the filament material. Substituting Equation (2) in (1) and solving for \( \Delta T \) gives

\[ \Delta T = \frac{-b + \sqrt{b^2 - \frac{4mV_w^2}{R_w A_{ws}}}}{2m}. \]  

(3)
Changes to $\Delta T$ in Equation (3) are due only to changes in $h_w$ and possibly to $V_w$ if not operated in constant-voltage mode. The hot wire may be treated as a smooth cylinder, so that $h_w$ can be determined by the Churchill-Bernstein correlation for external cross-flow over a cylinder (Churchill & Bernstein, 1977). For known gas properties, extreme values of $R_w$ may be calculated for pure carrier gas flow and for pure tracer. Measured values of $R_w$ between these extreme values represent mixtures of carrier and tracer, and the concentration of tracer $C_{He}$ may be inferred by interpolating in a theoretical curve of $R_w$ for different gas mixture properties.

The time-dependent input and output tracer concentration signals $c_{in}(t)$ and $c_{out}(t)$ are related to the RTD function $e(t)$ by the convolution integral:

$$c_{out} = e * c_{in} = \int_{-\infty}^{\infty} e(t_1 - t) c_{in}(t_1) dt_1 = \int_{-\infty}^{\infty} e(t_1) c_{in}(t - t_1) dt_1, \quad (4)$$

where $t$ refers to the time domain and $t_1$ is a shift of one of the functions relative to the other in the time domain. In the present study, $c_{in}$ and $c_{out}$ were measured and $e(t)$ was unknown; hence, deconvolution was used to obtain the RTD function between any two given concentration signals. A fast Fourier transform algorithm was used to convert measured concentration signals to the frequency domain, where deconvolution is simplest. After obtaining the RTD in the frequency domain, the inverse Fourier transform was used to convert the RTD $e(t)$ back to the time domain.
E.3 Materials & Method

A model downer similar to the one used by Huard et al. (2010) and Huard (2009) constructed of transparent acrylic was used to perform cold flow gas phase RTD experiments as shown in Figure E.0.1. The downer had an internal diameter of 6.99 cm and a total height of 133.5 cm. The cone deflector was attached to an adjustable rod to change the distance $L_S$ between the gas outlet and the cone rim from -1.40 cm to 6.99 cm. Values of $L_S < 0$ indicated that the cone rim was below the gas outlet.

[Sketch of Experimental Model Downer]

Compressed air was used as the carrier gas in the downer. Three calibrated sharp-edged orifice nozzles with diameters of 0.20 cm, 0.22 cm, and 0.31 cm were used to control the mass flowrate of air in the downer. The range of superficial gas velocities $U_g$ in the downer was 0.30 m/s to 1.27 m/s for the present study. A tracer injection line was installed upstream of the downer as depicted in Figure E.0.2. As shown in Figure E.0.2, air was permitted to flow into the downer inlet either through the tracer gas injection line or the air bypass line.

Before injection, solenoid valves on the injection line were opened and the bypass line was closed. Helium was then permitted to flow through the open injection line and into
the downer for about one minute to flush the injection line. Both solenoid valves were then closed to trap tracer in the line. Next, the air bypass line was opened with a slight restriction in the line for strong preferential flow to the injection line during injection. Data logging was then initiated on the data acquisition system. Finally, the tracer injection was performed by opening sequentially the solenoid valves at the inlet and outlet of the injection line, respectively. The inlet valve was first opened to equilibrate the pressure between the flowing air and the trapped helium, while the outlet valve was opened about two seconds later to flush the injection line with air and inject the helium pulse into the downer.

![Figure E.0.2 – Schematic Diagram of Experimental Apparatus](image)

Three hot wire anemometers (Probes 1 to 3) were used to measure the concentration of helium, whose approximate locations in the downer are shown in Figure E.0.2. Probe 1 was installed 5.1 cm below the downer inlet to measure the incoming helium concentration signal. Probe 2 was located 16.5 cm above the gas outlet just upstream of the gas-solids separator, and Probe 3 was located 47.0 cm downstream of the entrance to
the gas outlet. These detection points allowed measurement of the RTD through the entire
downer and across the gas-solids separator. A 6.0 VDC regulated power supply was used
to power each probe circuit, which were operated in a mode similar to constant-voltage
mode. This allowed for a very simple electronic setup but slightly sacrificed probe
sensitivity compared to other standard operation modes. Each detector was connected in a
Wheatstone bridge circuit to increase the sensitivity of the detectors.

E.4 Results & Discussion

E.4.1 Effect of Gas Velocity

Gas phase RTD measurements were performed in the absence of solids to gain insight
into potential gas backmixing in the reactor and around the gas-solids separator. The gas
velocity $U_g$ and separation length $L_S/D$ were adjusted to understand their respective
effects on the RTD in the downer. The effect of gas velocity was investigated for the
range of gas velocities $U_g = 0.30$ m/s to 1.27 m/s. This range corresponded to average
residence times of 3.9 s to 0.92 s between the downer inlet and the gas outlet, and to
residence times of 5.0 s to 1.2 s when also including the reactor volume below the gas
outlet. Hence, the gas outlet was an open boundary through which the flow could
penetrate.

Figures E.3(a) and E.3(b) show typical normalized helium concentration $C_{He}$ curves
measured at Probes 1, 2, and 3 and the corresponding RTD curves for $U_g = 1.05$ m/s and
$L_S/D = 0$. Concentration measurements were acquired at 500 Hz. Huard (2009) showed
that maximum particle removal efficiency in the separator occurred at $L_S/D = 0$ for the
operating conditions expected in the actual pyrolysis process in the downer reactor.
Hence, this condition was of particular focus in the present study.
The RTDs shown in Figure E.3(b), obtained by deconvolution of the concentration signals, indicated some spreading and potential bypassing of the initial helium pulse in the downer upstream of the gas-solids separator. This was indicated by a bimodal RTD curve $e_{1-2}$ (i.e. RTD between Probes 1 and 2) in Figure E.3(b). A small secondary peak was also observed in curve $e_{2-3}$ as shown in Figure E.3(b), which indicated some backmixing around the gas-solids separator. Although future introduction of solids would likely increase backmixing in the reactor, these initial results confirmed the possibility to operate the downer close to the plug flow regime.

The severity of backmixing was gauged by analyzing the average residence time and variance of the measured RTDs. Figure E.4 shows the actual average residence time and variance of the measured RTD curves for the range of tested gas velocities. As shown in Figure E.4(a), the measured residence times in the downer before the separator were reasonably close to the nominal values calculated by dividing the reactor volume by the volume flowrate, which confirmed near plug flow and use of the deconvolution technique. Based on the RTD variance curves shown in Figure E.4(b), the gas-solids separator was the major contributor to RTD spreading for all tested gas velocities. This result indicated very little RTD spreading and backmixing in the downer before the separator.

Another important indicator of reactor performance was the effective gas penetration length $L_p$. This indicator represented how far the gas “effectively” flowed past the gas
outlet based on the measured residence time compared to the nominal residence time between the inlet and the gas outlet. In other words, measured residence times longer than the nominal values gave $L_P > 0$, indicating gas flow below the outlet. Measured residence times shorter than the nominal values gave $L_P < 0$, indicating either no gas flow past the outlet or bypassing. The effect of gas velocity on $L_P$ is shown in Figure E.4(c). The results show that gas did not penetrate past the gas outlet for velocities $U_g \leq 0.79$ m/s, which corresponded to short durations in the gas-solids separator. Long penetration lengths past the outlet were observed for gas velocities $U_g \geq 1.05$ m/s. Penetration past the gas outlet did not necessarily correspond to backmixing, but the relative duration in the separator was much longer for $L_P > 0$. This result also indicated that future use of stripping gas below the gas outlet would likely decrease the penetration length and separation time.

Figure E.0.4 – Effect of Gas Velocity on Average Residence Time, Variance, and Effective Penetration Length
E.4.2 Effect of Separation Length

The effect of separation length on the residence time, variance, and penetration length in the downer was investigated over the range of $L_s/D = -0.2$ to $L_s/D = 1.0$. The gas velocity was set at $U_g = 1.27$ m/s to represent the actual pyrolysis process operating conditions. As shown in Figure E.5(a), a modest increase in the residence time with decreasing separation length was observed for $0 \leq L_s/D \leq 1.0$. Considering also the gas-particle separation efficiency, which increased with decreasing separation length to a maximum at $L_s/D = 0$ for similar operating conditions (Huard, 2009), the present results suggest that there may be a trade-off between separation efficiency, residence time, and backmixing for optimum process performance in the downer. However, for $L_s/D = -0.2$, an enormous increase in the residence time, variance, and penetration length was observed. This indicated very significant backmixing below the gas outlet and gas flow reaching the solids outlet. Hence, this operating condition must be avoided to achieve plug flow in the reactor and to avoid pyrolysis product vapor degradation.
E.5 Conclusion

A simple hot wire anemometry technique was developed and used successfully to measure the gas phase RTD in a downer reactor. The measured RTD curves show near plug flow behavior in the downer for most tested operating conditions. Some gas backmixing was observed around the gas-solids separator at high gas velocity and for very short separation lengths. The most severe backmixing occurred for $L_S/D = -0.2$. The best operating conditions for near plug flow in the reactor were $U_g = 1.27$ m/s and for separation lengths $L_S/D \geq 0$.

E.6 Notation

$A_{ws}$  Hot wire heat transfer surface area [m$^2$]

$b$  Empirical coefficient of electrical resistivity [$\Omega$]

$C_{He}$  Normalized concentration of helium [-]
\( c_{in}(t) \) Input tracer concentration [kg/m\(^3\)]
\( c_{out}(t) \) Output tracer concentration [kg/m\(^3\)]
\( D \) Downer internal diameter [cm]
\( D_w \) Hot wire diameter [mm]
\( E(s) \) Residence time distribution function in the frequency domain [-]
\( e(t) \) Residence time distribution function in the time domain [-]
\( h_w \) Heat transfer coefficient between the hot wire and the surrounding fluid [W/m\(^2\)K]
\( L_S \) Vertical distance between gas outlet and cone deflector rim [cm]
\( m \) Empirical coefficient of electrical resistivity [Ω/K]
\( R_w \) Hot wire probe resistance [Ω]
\( s \) Frequency domain [s\(^{-1}\)]
\( T_w \) Hot wire surface temperature [K]
\( T_g \) Ambient gas temperature [K]
\( t \) Time [s]
\( U_g \) Superficial gas velocity [m/s]
\( V_w \) Voltage applied to hot wire [V]
\( \sigma^2 \) Variance [s\(^2\)]
\( \tau_{avg} \) Average residence time [s]

E.7 References


Appendix F – A CFD study of biomass pyrolysis in a downer reactor equipped with a novel gas-solid separator - I: hydrodynamic performance

F.1 Introduction

The escalating global concern over the exhaustion of non-renewable energy sources lead to the recent development of a range of novel technologies for the use of renewable energy resources, such as biomass, solar and wind. Among these resources and technologies, biomass pyrolysis has emerged as a very promising renewable alternative for bio-oil production. In a large commercial scale, this could be carried out in a dual fluidized bed (DFB) system with various optional arrangements. The schematics in Figure F.0.1 demonstrate examples of these arrangements. In this study, we are interested in the downer-riser type of a dual fluidized bed, shown in Figure F.0.1(b), where the biomass pyrolysis takes place in the downer side of the reactor, while the riser side is used for combustion, thus providing the heat required for the pyrolysis through the circulating inert heat carrier solid (such as sand). This arrangement has the following specific advantages for bio-oil production through fast pyrolysis:

i) The downer pyrolysis reactor can be operated with very low carrier gas (e.g. nitrogen) flow rates, which is desirable in some cases to reduce up-stream pre-heating and downstream processing.

ii) Reducing the gas and solid back-mixing (Mrgain et al., 2000; Zhang et al., 2001; Huang et al., 2006) thus, limiting the spread of the gas/solid residence time distribution, i.e. near to plug flow.

iii) Relatively low cost, simple operation/control and high energy efficiency.

iv) The char combustion in the second reactor will guarantee sustainable operation and better control of the pyrolysis temperature in the first reactor.

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4 A version of this chapter has been published in Fuel Processing Technology:

However, in order to achieve high conversion efficiency (more than 70% bio-oil yield) in a downer reactor there remains two main technical challenges:

i) Control of the pyrolysis gas residence time within the hot zone of the reactor (ideally 1-2 seconds). Longer residence time of the pyrolysis gas at high temperature initiates a range of undesirable side reactions, which could adversely affect the quality of the product bio-oil (Bridgwater & Peacocke, 2000; Hoekstra et al., 2012)

ii) Control of the downstream contact between the pyrolysis gas and bio-char. The bio-char, formed during pyrolysis, acts as a vapor cracking catalyst, therefore should be separated as soon as the pyrolysis vapor is released (Jahirul et al., 2012)
Char, as well as other entrained fine particles, can primarily be separated from the pyrolysis gas by using conventional cyclones (reverse and co-current flow types). However, this carries the risk of increasing the contact time between the gas and char inside the cyclone. In addition, the cyclone inlet is commonly placed external to the reactor or away from the pyrolysis zone, thus, causing extra contact time between the solid and gas. The extensive review conducted by Huard et al. (2010) and Cheng et al. (2008) on downer reactors and rapid gas-solid separation techniques revealed that there are limited attempts on implementing new design methods for rapid gas-solid separation in these reactors.

Recent research at the Institute for Chemicals and Fuels from Alternative Resources (ICFAR) has led to the development of a novel gas-solid separation device for a downer pyrolysis reactor. The device features a cone-shaped solid deflector positioned above a gas outlet pipe, both positioned concentrically in the downer pipe (see Figure F.0.2). This was designed to achieve primary solid-gas separation and gas removal within the same device (Huard, 2009). The separator allows for better control of the pyrolysis vapor residence time, therefore, reducing the severity of vapor over-cracking compared to other fast separation methods. Experimental work by Huard et al. (2010) has shown that this separator can achieve very high solid-gas separator efficiency above 99.99% when using spherical silica sand particles of 200 µm diameter.

Computational Fluid Dynamics (CFD) modeling is one of the powerful tools to analyse gas-solid flow behavior, including that involves intense heat transfer and chemical reactions. The co-authors from the ICFAR have previously used an Eulerian-Lagrangian modeling approach to investigate the effect of the particle elasticity on the separator efficiency in the same novel separator investigated in this study (Huard, 2009). While this approach revealed important details of the particle-wall collision and its effects on the separator efficiency and mechanism, the simulation domain was limited to the separator zone only and the total solid volume fraction was limited to a maximum of 4×10⁻⁵. The Eulerian-Eulerian (also referred to as two-fluid) is another modeling approach that has the advantage of being robust and realistic in computational time, especially when considering a large number of particles or large simulation domain. Unlike the Eulerian-
Lagrangian approach, which treats each single particle as a dispersed phase in the continuum fluid flow, the Eulerian-Eulerian approach treats both of the fluid and solid phases as an interpenetrating continuum. Studies on the Eulerian-Eulerian simulation of solid-gas hydrodynamics in a downer reactor have been previously reported by Ropelato et al. (2005), Kim et al. (2011) and Samruamphianskun et al. (2012). This modeling approach was found to be especially useful in predicting the effects of inlet design and flow conditions on the solid distribution and dispersion behavior. This CFD modeling approach has also been used by different researchers to study the phenomena of solid-gas separation in cyclones (Winfield et al., 2013; Kepa, 2010; Chu et al., 2011).

In this study, the main objectives are:

i) to develop a valid Eulerian-Eulerian (multi-fluid) CFD model capably of predicting the detailed hydrodynamic behaviour in a downer reactor equipped with a novel gas-solid separation device;

ii) to use the developed model in investigating the effect of the operating conditions and various separator design parameters on the overall hydrodynamics, with particular focus on the separator efficiency;

iii) to provide a platform for the development of a predictive model of the pyrolysis reactions and yield in the downer reactor equipped with the novel gas-solid separator.

The investigation was carried out theoretically and experimentally in a cold flow reactor model equipped with the ICFAR novel gas-solid separator and gas removal mechanism, as described in details in Section 0. The theoretical transient model was solved in three-dimensional coordinates using the Eulerian-Eulerian (two-fluid) approach, employing constitutive relations from the kinetic theory of granular flow (KTGF) (Jenkins & Savage, 1983). In the second part of this study, the developed hydrodynamic model will be extended to include heat transfer and reaction kinetics to demonstrate the advantages of the ICFAR separator in improving the performance and product quality in a biomass downer pyrolysis reactor.
F.2 Experiments and Procedure

The experimental work described here was carried out by the co-investigators at the ICFAR in Canada. The equipment consisted of a cold flow gas-solid flow downer of 133.5 cm height and 7.0 cm diameter, equipped with the ICFAR novel gas-solid separator as shown in Figure F.0.2 and Figure F.0.3. This separator included a gas removal pipe and a cone deflector, where the bottom of the deflector and tip of the pipe were located 98.6 cm below the downer inlet. A solid collection tank of 20.4 cm diameter and 21.8 cm height was placed at the bottom of the downer column around 34.9 cm below the cone deflector. Compressed air at room temperature was supplied to the downer from a bank of calibrated sonic orifice nozzles. The Sauter mean diameter of the particle mixture used was 188 µm and its skeletal density was 2650 kg/m³. The particle size distribution of the mixture is shown in Figure F.0.4.

Figure F.0.2 – Schematic diagram of the experimental apparatus
Table 0.1 – Dimensions of the downer reactor and the cone separator

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<th>Value</th>
<th>Parameters</th>
<th>Value</th>
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<td>$D_{go}$ [cm]</td>
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</tr>
<tr>
<td>$\alpha$ [degree]</td>
<td>60, 90, 120</td>
<td></td>
<td></td>
</tr>
</tbody>
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Figure F.0.3 – Illustration of experimental (a) cold model downer setup and (b) gas-solid separation zone
The solid particles were delivered to the downer column from an air pressurized tank mounted above the downer main air inlet. The total mass flowrate of air in the downer was $\dot{m}_g = 0.0039 \text{ kg/s}$, which corresponded to a superficial gas velocity of $U_g = 0.73 \text{ m/s}$. The solids mass flowrate was adjusted by changing the feed tank air pressure and this was varied between $\dot{m}_s = 0.017 \text{ kg/s}$ and $0.083 \text{ kg/s}$, which corresponded to solids-to-gas loading ratios of $\dot{m}_s / \dot{m}_g = 4.3$ to 21. The gas-solids mixture flowed co-currently in the downer before entering the gas-solids separation zone. Three different cone deflectors with various internal angles of 60°, 90° and 120° were used. The downward falling particles were collected in the tank at the bottom of the unit, while the gas stream, along with any entrained particles, exited the system through the gas outlet pipe mounted in the centre of the downer cross section and below the cone deflector. A bag filter connected at the end of the gas exhaust line was used to collect the particles entrained in the exiting gas stream.
At the start of each experiment, the total mass of solids fed into the system, $m_{in}$, was measured. The mass of the entrained solids collected in the bag filter, $m_{collected}$, was then measured at the end of each experiment. Thus, the experimental percentage total solids separation efficiency, $\eta$, was calculated from the following expression:

$$\eta = \left(1 - \frac{m_{collected}}{m_{in}}\right) \times 100\%$$

(9.1)

The mass flowrate of solids was determined by measuring the total mass of solid collected in the filter bag and tank against the recorded time.

**F.3 Hydrodynamic Model**

The overall reactor hydrodynamics and gas-solid separation was investigated using the Eulerian-Eulerian (multi-fluid) model approach based on the Kinetic Theory of Granular Flow (KTGF). The developed model was solved using the CFD software ANSYS FLUENT (Ver. 14). In order to mimic the wide size distribution of the solid mixture used in the experiment, the simulation was carried out using a solid mixture of three different particle sizes, as detailed in section 3.3. The main model equations for non-reacting isothermal gas-solid flow are given by:

**Continuity equations:**

$$\frac{\partial (\alpha_g \rho_g)}{\partial t} + \nabla (\alpha_g \rho_g \bar{u}_g) = 0$$

(9.2a)

$$\frac{\partial (\alpha_s \rho_s)}{\partial t} + \nabla (\alpha_s \rho_s \bar{u}_s) = 0$$

(9.2b)

$$\sum_{i=1}^{3} \alpha_{s_i} + \alpha_g = 1$$

(9.2c)

**Momentum equations:**
\[
\frac{\partial (a_g \rho_g \vec{u}_g)}{\partial t} + \nabla (a_g \rho_g \vec{u}_g \cdot \vec{u}_g) = -a_g \nabla P + \nabla \vec{t}_g - \sum_{i=1}^{3} \beta_{gs_i} (\vec{u}_g - \vec{u}_{s_i}) + a_g \rho_g g \quad (9.3a)
\]

\[
\frac{\partial (\alpha_s \rho_s \vec{u}_{s_i})}{\partial t} + \nabla (\alpha_s \rho_s \vec{u}_{s_i} \cdot \vec{u}_{s_i}) = -\alpha_s \nabla P - \nabla P_{s_i} + \nabla \vec{t}_{s_i} + \beta_{gs_i} (\vec{u}_g - \vec{u}_{s_i}) + \sum_{j=1,j \neq i}^{3} \beta_{s_i s_j} (\vec{u}_{s_j} - \vec{u}_{s_i}) + \alpha_s \rho_s g \quad (9.3b)
\]

where:

\[
\vec{t}_k = \left( \lambda_k - \frac{2}{3} \mu_k \right) (\nabla \cdot \vec{u}_k) \vec{l} + 2 \mu_k \vec{S}_k \quad (9.4a)
\]

\[
\vec{S}_k = \frac{1}{2} \left( \nabla \vec{u}_k + (\nabla \vec{u}_k) \tau \right) \quad (9.4b)
\]

\( k \) represents solid or gas phase.

To obtain the granular temperature, the FLUENT code was optionally set to use a partial differential equation (Pseudo Energy Equation) as follows (Ding & Gidaspow, 1990):

\[
\frac{3}{2} \left[ \frac{\partial (\alpha_s \rho_s \theta_{s_i})}{\partial t} + \nabla (\alpha_s \rho_s \theta_{s_i} \vec{u}_{s_i}) \right] = \left( -P_{s_i} \vec{l} + \vec{t}_{s_i} \right) \cdot \nabla \vec{u}_{s_i} + \nabla (\kappa_T \nabla \theta_{s_i}) - \gamma_T + \sum_{k=1}^{3} \phi_{k s_i} \quad (9.5)
\]

The various closure and constitutive relations used in the model are given in Table 0.2. In order to take into consideration the solid-solid frictional stresses at the dense regions of the reactor, the friction equation proposed by Syamlal (1987), as given in Equation T1-5, was used. Due to the highly turbulence of the flow near the deflector zone the standard
K-epsilon turbulence and energy dissipation equations proposed by Launder and Spalding (1972) were also incorporated in the model and these are given as follows:

Turbulence momentum equations:

\[
\frac{\partial (\alpha_g \rho_g k_g)}{\partial t} + \nabla (\alpha_g \rho_g \bar{u}_g k_g) = \alpha_g G_{k,g} + \nabla \left( \alpha_g \frac{\mu_l g}{\sigma_k} k_g \right) - \alpha_g \rho_g \varepsilon_g + \alpha_g \rho_g \Pi_{k,g} \tag{9.6a}
\]

Turbulent kinetic energy dissipation equation:

\[
\frac{\partial (\alpha_g \rho_g \varepsilon_g)}{\partial t} + \nabla (\alpha_g \rho_g \bar{u}_g \varepsilon_g) = \nabla \left( \alpha_g \frac{\mu_l g}{\sigma_\varepsilon} \varepsilon_g \right) + \alpha_g \frac{\varepsilon_g}{k_g} (C_1 \varepsilon G_{k,g} - C_2 \rho_g \varepsilon_g) + \alpha_g \rho_g \Pi_{\varepsilon,g}
\]

(9.6b)

where:

\[
G_{k,g} = \mu_{l,g} \left( \nabla \bar{u}_g + \left( \nabla \bar{u}_g \right)^T \right) \cdot \nabla \bar{u}_g
\]

\[
C_\mu = 0.09, C_{1\varepsilon} = 1.44, C_{2\varepsilon} = 1.92, \sigma_k = 1, \sigma_\varepsilon = 1.3
\]
Table 0.2 – Constitutive relations for the gas–solid flow

\[ P_s = \rho_s \bar{\rho}_s \left[ 1 + \sum_{j=1}^{n} \left( \frac{d_s + d_j}{4d_j^2} \right) \rho_s \alpha_s \Theta_s \Theta_j \right] \quad (T1 - 1) \]

**Solid shear viscosity**
\[ \mu_s = \mu_{s,\text{ref}} + \mu_{s,\text{kin}} + \mu_{s,\text{fr}} \quad (T1 - 2) \]

**Collisional viscosity** [21]
\[ \mu_{s,\text{coll}} = \frac{1}{2} \alpha_s \rho_s d_s g_0 s_{\Theta_s} (e_{s,s} + 1) \left( \frac{\rho_s}{\Theta_s} \right)^{1/2} \quad (T1 - 3) \]

**Kinetic viscosity** [22]
\[ \mu_s = \frac{\alpha_s \rho_s d_s g_0 s_{\Theta_s} (e_{s,s} + 1) \left( \frac{\rho_s}{\Theta_s} \right)^{1/2}}{4s_{\Theta_s} (3s_{\Theta_s} - 1)} \quad (T1 - 4) \]

**Friction viscosity** [23]
\[ \mu_{s,\text{fr}} = \frac{\alpha_s \rho_s d_s g_0 s_{\Theta_s} (e_{s,s} + 1) \left( \frac{\rho_s}{\Theta_s} \right)^{1/2}}{4s_{\Theta_s} (3s_{\Theta_s} - 1)} \quad (T1 - 5) \]

**Bulk viscosity** [24]
\[ \lambda_s = \frac{4}{3} \alpha_s \rho_s d_s g_0 s_{\Theta_s} (e_{s,s} + 1) \left( \frac{\rho_s}{\Theta_s} \right)^{1/2} \quad (T1 - 6) \]

**Radial distribution function**
\[ g_{0,s,s} = \left( 1 - \frac{2e_{s,s}}{\sigma_{s,s}^2} \right)^{1/2} + \frac{1}{2} \sigma_{s,s} \sum_{j=1}^{n} \alpha_s \rho_j d_j \frac{\alpha_s}{\sigma_{s,s}^2} \frac{\alpha_j}{\sigma_{s,j}^2} \quad (T1 - 7a) \]
\[ g_{0,s,j} = \frac{d_j \sigma_{s,j}^2 - d_s \sigma_{s,s}^2 \rho_j}{d_s \sigma_{s,s}^2} \quad (T1 - 7b) \]
\[ \sigma_{s,j} = \frac{1}{2} \sum_{j=1}^{n} \alpha_j \rho_j d_j \quad (T1 - 7c) \]

**Gas viscosity**
\[ \nu_g = -H_g + H_G, \quad \mu_g = \frac{C_p \alpha_s \rho_s}{e_g} \quad (T1 - 8) \]

**Gas-solids drag coefficient** [25]
\[ \beta_{g,s} = \frac{30 \alpha_s}{\alpha_g \beta_g} C_D \left( \frac{\rho_s}{\rho_g} \right) \left( |u_s - \bar{u}_s| \right) \quad (T1 - 9) \]

\[ \nu_{r,s} = 0.5 \left( A - \text{0.06Re}_s \sqrt{(0.06Re}_s)^2 + 0.12Re_g (28 - A) + A^2 \right) \quad (T1 - 10) \]

\[ A = \alpha_s^{1.14}, \quad B = 0.8 \alpha_s^{0.28} \left( \alpha_s \leq 0.85 \right) \]
\[ B = 0.8 \alpha_s^{0.25} \left( \alpha_s > 0.85 \right) \quad (T1 - 11) \]

\[ C_D = \left( 0.63 + \frac{18}{\sqrt{\text{Re}_s}} \right)^2 \quad (T1 - 12) \]

\[ \text{Re}_s = \frac{\rho_s u_s |u_s| \bar{u}_s}{\mu_s} \quad (T1 - 13) \]

**Solid–solid drag coefficient** [19]
\[ \beta_{s,j} = \left( \frac{1}{2} \sigma_{s,s}^2 \sigma_{s,j}^2 \right) \frac{\alpha_s \rho_s \rho_j \sigma_{s,s}^2 \sigma_{s,j}^2 |u_s - \bar{u}_s|}{2\sigma_{s,s}^2 \sigma_{s,j}^2} \quad (T1 - 14) \]

**Diffusion coefficient of granular energy** [21]
\[ \nu_{\Theta_s} = \frac{150 \nu_{s} \alpha_s \rho_s}{384 (e_{s,s} + 1)^2} \left[ 1 + 9 \alpha_s g_0 s_{\Theta_s} (e_{s,s} + 1) \right] \quad (T1 - 15) \]
\[ + 2 \alpha_s 
\[ \nu_{\Theta_s} = \frac{12}{d_s \rho_s \alpha_s \beta_g \Theta_s^{2/3}} \quad (T1 - 16) \]

**Collisional energy dissipation** [24]
\[ \gamma_{\Theta_s} = \frac{12}{d_s \rho_s \alpha_s \beta_g \Theta_s^{2/3}} \quad (T1 - 17) \]

\[ \gamma_{\Theta_s} = \frac{12}{d_s \rho_s \alpha_s \beta_g \Theta_s^{2/3}} \quad (T1 - 18) \]
F.3.1 Computational Domain and Meshing

Figure F.0.5 shows the computational domain and the meshing used in solving the model. This was generated using a finite volume method with hybrid cells of structured and unstructured grids, giving a total of 30,785 cells. In order to capture the steep hydrodynamic variations around the walls of the separation device (the conical deflector and the gas exit pipe), the grid size was refined by setting the minimum and maximum grid size at 0.3 and 1.0 cm respectively. In the rest of the simulation domain the minimum and maximum grid size was set at 1.0 and 5.0 cm respectively. The impact of the grid size on the solution accuracy was initially tested by setting three different meshing schemes and the grid size used in this study was found to give acceptable grid independent solution.

F.3.2 Computation Procedure

The model equations were solved using the finite volume approach. First-order discretization schemes were used for the solution of the convection terms in all governing
equations. The relative error between any two successive iterations was specified by using a convergence criterion of $10^{-3}$ for each scaled residual component. The phase-coupled SIMPLE (PC-SIMPLE) algorithm (Vasquez & Ivanov, 2000), which is an extension of the SIMPLE algorithm to multiphase flows, was applied for the pressure-velocity coupling. The linearized equations for governing equations were solved using a block algebraic multigrid method. In order to ensure easy convergence of the various partial differential equations (PDE) in the model, the Courant–Friedrichs–Lewy (CFL) condition for three-dimensional PDE is followed:

$$C = \frac{u_x \Delta t}{\Delta x} + \frac{u_y \Delta t}{\Delta y} + \frac{u_z \Delta t}{\Delta z} \leq C_{\text{max}}$$  \hspace{1cm} (9.7)$$

where $C_{\text{max}}$ is specified by the CFL condition to fall within the range of ~1-5 (Courant et al., 1928). In this study, a time step of 0.005 seconds was found to satisfy this condition.

### F.3.3 Boundary and Simulation Conditions

The particle-wall restitution coefficient and the specularity coefficient are two important parameters in determining the dynamics of particles at the wall region. The following wall boundary conditions were employed in the model (Johnson & Jackson, 1987):

$$u_{s_i,w} = - \frac{6 \mu_{s_i} \alpha_{s,\text{max}}}{\sqrt{3} \theta_{s_i} \pi \rho_s \alpha_{s_i} g_{0,s_i,s_i}} \frac{\delta u_{s_i,w}}{\delta n}$$  \hspace{1cm} (9.8)$$

$$\theta_{s_i} = \frac{k_{s_i} \theta_{s_i}}{\gamma_w} \frac{\delta \theta_{s_i,w}}{\delta n} + \frac{\sqrt{3} \pi \rho_s \alpha_{s_i} u_{s_i,\text{tip}}^2 g_{0,s_i,s_i} \theta_{s_i,3}}{6 \alpha_{s,\text{max}} \gamma_w}$$  \hspace{1cm} (9.9a)$$
\[
\gamma_w = \frac{\sqrt{3} \pi (1 - e_{s_{l,w}}^2) \rho_s \alpha_{s_l} g_{0,s_l} \theta_{s_l}^3}{4 \alpha_{s,\text{max}}} \tag{9.9b}
\]

where \(\varphi\) is the specularity coefficient and \(e_{s_{l,w}}\) is the particle–wall restitution coefficient.

In order to reasonably match the particle size distribution used in the experiments, the simulations were carried out assuming the solid mixture to consist of three different particle sizes. The fraction of each particle size group was estimated from the experimental size distribution given in Figure F.0.4. The simulation particle sizes and percentages are given in Table 0.3. For the gas phase, the velocity at the wall was assumed zero (no slip condition). Table 0.4 summarizes the various operating conditions considered in the simulations. Some of these conditions were carefully selected to allow for the comparison of the model predictions with the corresponding experimental data.

**Table 0.3 – Particle size distribution employed in the CFD model**

<table>
<thead>
<tr>
<th>Particle size (µm)</th>
<th>Solid phase I</th>
<th>Solid phase II</th>
<th>Solid phase III</th>
</tr>
</thead>
<tbody>
<tr>
<td>Volume fraction (%)</td>
<td>(X_{10} = 124)</td>
<td>(X_{50} = 206)</td>
<td>(X_{90} = 328)</td>
</tr>
</tbody>
</table>

**Table 0.4 – Gas and solid phase boundary/operating conditions used in the CFD model**

<table>
<thead>
<tr>
<th>Boundary/operating condition</th>
<th>Experiment</th>
<th>Model</th>
</tr>
</thead>
<tbody>
<tr>
<td>Gas mass flow rate, (\dot{m}_g) [kg/s]</td>
<td>0.0039, 0.0239, 0.0439</td>
<td>0.0039(^a), 0.0239, 0.0439</td>
</tr>
<tr>
<td>Gas density, (\rho_g) [kg/m(^3)]</td>
<td>1.2</td>
<td>1.2(^a)</td>
</tr>
</tbody>
</table>
| Gas dynamic viscosity [kg/(m·s)] | \(1.8 \times 10^{-5}\) | \(1.8 \times 10^{-5}\)
| Gas outlet pressure, \(P_{sg}\) [Pa] | 0 (ambient) | 0\(^a\) (ambient) |
| Particle density, \(\rho_p\) [kg/m\(^3\)] | 2650 | 2650\(^a\) |
| Solid mass flow rate, \(\dot{m}_{s_{l}}\) [kg/s] | 0.017, 0.083 | 0.004\(^a\), 0.02, 0.04, 0.08 |
| Mean particle size, \(\bar{d}_s\) [µm] | 188 | Mixture\(^a\) (124, 206, 328) |
| Angle of cone deflector [degree] | 60, 90, 120 | 60\(^a\), 90, 120 |
| Separation length, \(L_s\) [m] | 0 | 0\(^a\), 0.035, 0.07 |

\(^a\) Default simulation conditions, unless otherwise specified.
F.4 Results and Discussion

F.4.1 Mechanism of Gas-Solid Separation

It is postulated that the drag and gravity forces, the last two terms in the left hand side of Eq. 3b, are the main forces dominating the hydrodynamic behavior of the gas-solid phases within the separator zone. Figure F.0.6 gives an overall description of the flow structure with close zoom-in at the cone deflector region. The gas velocity in the gap between the deflector and the wall is very high due to the considerable pressure drop, similar to gas expansion through a throttling device. In the region under the cone and below the gas exit pipe there is an upward gas drag force due to the high reverse gas phase velocity. However, the extremely dilute solid concentration in this zone means very little solids are being entrained. It is therefore desirable to minimize the upward gas drag force in this region in order to achieve high separator efficiency. On top of the cone deflector, the solid phase is diverted radially towards the wall and then accelerates through the gap between the deflector and the downer wall, pushed by a strong gas drag force. The influence of the gravity force in this region is also significant due to the high solid concentration. Accordingly, it is believed that any particles entrained through the exit pipe are falling under the influence of two different drag mechanisms:

i) Reverse gas flow (upward) under the cone deflector due to the abrupt gas pressure drop at the tip of the gas exit pipe. This makes the tip of the exit pipe act as a vacuum to the surrounding solids.

ii) Radial gas flow from the walls towards the core in the region just below the cone deflector. This results in the solids being first dragged towards the core, and then further dragged/sucked by the gas leaving through the exit pipe.
Figure F.0.6 – Example of the gas–solid flow structure in the overall downer column and around the separation device. (a) Gas phase velocity vectors, with the color code restricted to a maximum of 5m/s to allow visualization. (b) Solid phase volume fraction in a section, with the color code restricted to a maximum of $4 \times 10^{-4}$ to allow visualization.

This is to some extent similar to the solid-gas separation mechanism in a cyclone, where in both cases the reverse gas flow in the core is responsible of solid entrainment. However, in the cyclone, the particles move radially towards the walls under the influence of centrifugal forces, while in the cone deflector, the particles are deflected radially by the cone wall to fall under the strong downward gas drag force in the “throttling” gap, as described earlier. It is worth noting that the modeling results reported by the co-authors from the ICFAR suggested that the solid rebound upon hitting the walls, investigated through changing the wall-particle restitution coefficient, may have a dominant role in the mechanism of gas-solid separation in the cone deflector. This hypothesis will be discussed in some details in the following sections.
Figure F.0.7 shows the predicted solid concentration and the gas velocity profiles at the level of 3.9 cm below the tip of the gas exit pipe (see Figure F.0.6(b) for the sampling line level). These profiles reveals a very interesting hydrodynamic behavior where the solid concentration profile is shown to take the shape of a dense-wall and dilute-core, while the gas velocity takes the shape of an upward parabolic flow profile at the core and a downward flow at the walls. It appears that, due to the existence of the cone deflector, the overall flow pattern below this device has been completely changed from the classic gas-solid down flow pattern, commonly observed in downer reactors, to a more complex flow similar to that existing in a turbulent solid-gas flow riser.

![Figure F.0.7](image)

Figure F.0.7 – Example of the radial profiles of the (a) axial gas velocity and (b) solid volume fraction. The data was taken at the sample level of 3.9 cm below the gas exit tip

### F.4.2 Gas Disengagement Height (GDH)

Figure F.0.8 shows that there are four distinct flow zones each with characteristic flow behavior. These are mainly arising from the changes induced by the cone deflector and these can be described as flows:

**Zone I:** This is where fully developed flow and uniform distribution of the solid and gas phases take place, typical to that observed in a conventional downer reactor.

**Zone II:** This the where both of the solid and gas phases are first hitting the inclined plane to create a dense moving solid layer at the cone walls before being pushing by a
strong gas drag force through the gap between deflector and the downer walls. Under the cone, the lowest solid concentration in the whole system exists and the gas is removed through the exit pipe driven by the rapid pressure drop at the exit pipe tip.

**Zone III:** This is where the disengagement of gas from the gas-solid flow mixture takes place. The overall flow hydrodynamics in this region is very complex due to the effect of sharp changes in pressure, which consequently leads to reverse gas flow towards the top and radial solid movement from the dense walls towards the dilute core.

**Zone IV:** This is where the solid phase is mainly concentrated at the walls. The radial flow diminishes and the particles fall under the strong influence of the gravity force before entering the solid collection tank at the bottom of the downer system.
Figure F.0.8 – Sectional view of the solid and gas axial velocity vectors demonstrating the various characteristic flow zones in the downer reactor and around the separation device. To allow visualization of the low ranges, the magnitude of the velocity vector was restricted to a maximum of 5m/s. The color code bar indicates the range of gas/solid velocity.

As described earlier, the main objective of the cone separator is to allow for fast and efficient separation of the gas from the downward gas-solid flow stream. In a biomass pyrolysis downer reactor, this separation should ideally take place immediately at the
level of the gas outlet pipe tip and with zero solid entrainment. The first reason is to prevent undesirable secondary gas reactions by removing the gas from the reactor hot zone, and second, to prevent catalytic char cracking by limiting the contacts between the solid and gas. However, in reality, the gas separation from the solid-gas stream takes place a little further down beyond the level of the tip of the gas removal pipe. It is therefore particularly interesting to quantify the height of Zone III, in which the gas separation takes place. This is defined here as the gas disengagement height (GDH), analogous to the definition of the transport disengaging height (TDH) in gas-solid fluidized beds. The method used in this study to estimate the GDH is demonstrated in Figure F.0.9. The GDH is defined at the intercept of the lines tangential to the low pressure gradient curve and the steep changing pressure gradient curve, or alternatively, the GDH can be estimated from plotting the axial gas velocity against height as shown in Figure F.0.9(b). The pressure gradient method is similar to the method used by Geldart et al. (2010) in determining the transport disengaging height (TDH).

![Figure F.0.9](image)

**Figure F.0.9** – Illustration of the method used to determine the GDH demonstrated in typical results obtained at the default model operating conditions (a) pressure gradient method (b) axial gas velocity method.

Figure F.0.10 shows the result of a sensitivity analysis of the GDH to a range of operating conditions. Please note that the y-axis in Figure F.0.10 represents the summation of the GDH and the separation distance $L_s$, where the separation distance is defined as the
distance from the cone rim to the tip of the gas exit pipe. The GDH range under the various operation conditions considered in the simulation was found to fall between 2.5 cm and 6 cm. It is clear that the GDH is most sensitive to the solid loading and the solid flow rate. The increase in the separation distance from 0 cm to 7 cm and the solid flow rate from 0.004 kg/s to 0.08 kg/s caused a corresponding increase in GDH of around 30% for both cases. Clearly, the cone angle and gas flow rate appear to cause negligible effect in this regard. It should be noted that, while it is desirable to decrease the GDH as discussed earlier, this does not necessarily mean improving the separation efficiency, as will be demonstrated in the next section.

Figure F.0.10 – The effect of operation conditions on the gas disengagement height. The default simulation conditions (Table 0.4) were used in all simulations, unless otherwise specified.

F.4.3 Separator Efficiency

The theoretical separator efficiency was obtained by dividing the predicted solid flow rate at the gas exit pipe (entrained solids) by the inlet solid mass flow rate, such that,
The separator efficiency was analysed with respect to various operating conditions. This also included a sensitivity analysis of the separation efficiency towards varying the wall-particle interactions mechanism, through changing the particle-wall restitution coefficient and specularity coefficient. Both parameters appear in the solid boundary condition of Eq. 8-9. The first coefficient is a measure of the degree of energy loss when the particles hit the walls, hence determining the rebound velocity, and the second coefficient defines the angle of rebound. It is therefore possible to determine the effect of the wall surface material and particle properties on the separation efficiency through changing these two parameters in the model.

In this study, 100% separator efficiency was obtained when operating with: large particle size of \(d_p = 328 \mu\text{m}\), separation length \(L_s = 0\ \text{cm}\), cone angle \(\theta = 60^\circ\), gas mass flow rate \(m_g = 0.0039\ \text{kg/s}\) and high solid flow rate \(m_s = 0.08\ \text{kg/s}\). This was found to dramatically decrease when decreasing the particle size. This result is in good agreement with the experimental study by Huard et al. (2010) where it was shown that the separator efficiency, when using FCC catalyst of \(d_p = 43 \mu\text{m}\) and glass beads of \(d_p = 63 \mu\text{m}\), is much lower than that achieved with sand of \(d_p = 200 \mu\text{m}\). Figure F.0.11 shows the values of the predicted separator efficiency obtained within the range of operating conditions considered in this study. The detailed results and discussion on the effect of each of these parameters on the overall hydrodynamics and separator efficiency are given in the next sections. According to this data, it is concluded that the sensitivity of the separator efficiency towards the operating conditions can be ranked in order of decreasing impact on the separation efficiency as follows; (1) separation length (2) cone angle (3) gas flow rate (4) solid flow rate (5) particle physical properties (expressed in terms of the restitution and specularity coefficients). Note that, the impact of particle size on the separation efficiency comes on top of all the above parameters.
Figure F.0.11 – Overall sensitivity analysis of the effect of the operating conditions on the separator efficiency. The default simulation conditions (Table 0.4) were used in all simulations, unless otherwise specified.

F.4.4 Effect of the Separation Length ($L_s$)

The effect of separation distance on the separator efficiency was studied using three different separation lengths of 0 cm, 3.5 cm and 7 cm, which corresponds to the normalized separation length ($L_s/D$) of 0, 0.5 and 1 respectively. All the other parameters were set at the default values.

Figure F.0.12 shows the separator efficiency as a function of the normalized separation length. The overall trend indicates a negative impact on the separator efficiency. The maximum mean efficiency was 99.986% and this dropped to 99.633% at $L_s/D=1$. The greater separation efficiency achieved with the particle size of 206 µm compared to the size group of 324 µm can be explained by the fact that the concentration of this particle group (60 wt%) was greater than the latter one (20 wt%). Therefore, the more frequent particle-particle interaction within the same group can neutralized part of radial velocity which may cause entrainment of particle. The same phenomenon was observed in Figure F.0.15. In terms of sensitivity, the effect of the separation length on the separation efficiency is the highest compared to the other parameters investigated, as shown earlier in Figure F.0.11. It was also demonstrated earlier that the separation distance has also a
relatively high effect on the GDH. The velocity vectors shown in Figure F.0.13 indicate that the increase in $L_s$ resulted in the creation of two vortices in the space between the cone deflector and the tip of the gas exit pipe. This can be attributed to the strong radial gas flow in this region, resulting from the considerable pressure drop at the exit pipe.

Figure F.0.12 – Effect of the separation length on the separator efficiency. The simulation was carried out using the default operating conditions
Figure F.0.13 – Gas velocity vectors at various normalized separation lengths. To allow visualization of the low ranges, the magnitude of the velocity vector was restricted to a maximum of 5 m/s. The color code bar indicates the range of gas velocity.

Figure F.0.14 shows the changes in the solid concentration and velocity profiles with changing the separation length at the sample level of 3.9 cm below the bottom of the cone deflector. It is clear that the solid concentration increases with increasing $L_{s}/D$, while the vertical upward gas velocity at the core decreases. This suggests that the upward gas drag force may have limited influence on the separation efficiency. It is the increased radial gas velocity (radial drag), the subsequent formation of vortices and the increased solids concentration at the core that collectively play the dominant role in decreasing the separator efficiency as the separation length increases.
Figure F.0.14 – Effect of the separation length on the (a) radial profiles of axial gas velocity and (b) solid volume fraction (for the selected particle size $d_s = 124\ \mu m$). The data was taken at the sample level of 3.9 cm below the gas exit pipe tip.

F.4.5 Effect of the Cone Deflector Angle ($\theta$)

The effect of the cone deflector angle on the separator efficiency was studied using various angles $\theta = 60^\circ$, $90^\circ$ and $120^\circ$. All other operating conditions were set at the default values. Figure F.0.15 shows that the separator efficiency decreases with increasing the cone angle. The maximum mean separator efficiency (taking into account the three particle sizes) was 99.986%, this dropped to 99.869 % efficiency when the angle is increased to $120^\circ$. This trend is in satisfactory agreement with the experimental data of Huard et al. (2010); however, the experiments showed less pronounced changes compared to the predictions, and this may be attributed to the differences between the particle size distribution in the experiment and the assumed size mixture in the model.
Figure F.0.15 – Effect of the cone deflector angle on the separator efficiency. The experiment data was obtained from Hurad et al. (2010). The simulation was carried out using the default operating conditions.

In Figure F.0.16 the magnitude and direction of the gas velocity vectors suggest that as the cone angle increases there is a greater chance the particles rebound more in the reverse direction from the cone inner surface and normal to the gas exit. This would slow down the particles and make them easier to be entrained, thus having a negative impact on the separator efficiency. In Figure F.0.17 there is clear increase in the solid concentration on top of the cone’s upper surface due to flattering of the cone external surface as shown in Figure F.0.17; however this is not expected to have contributed to the change in the separation mechanism or efficiency.
Figure F.0.16 – Gas velocity vectors at various cone deflector angles. To allow visualization of the low ranges, the magnitude of the velocity vector was restricted to a maximum of 5 m/s. The color code bar indicates the range of gas velocity.

Figure F.0.17 – Solid volume fraction for the particle size of ds=324 µm at two different deflector angles. To allow visualization of the low ranges, the solid volume fraction was restricted to $5 \times 10^{-4}$. The color code bar indicates the range of solid volume fraction.
F.4.6 Effect of the Gas Mass Flow Rate

The effect of inlet gas mass flow rate on the gas separator efficiency was investigated at three different flow rates of 0.0039 kg/s, 0.0239 kg/s and 0.0439 kg/s and a fixed solid flow rate of 0.004 kg/s. This corresponds to inlet gas velocities of 0.73 m/s, 4.5 m/s and 8.2 m/s, respectively. All other operating conditions were set to the default values. Figure F.0.18 shows that the effect of the gas mass flow rate on the separator efficiency is negligible. This is in good agreement with the experimental observation reported by Huard et al. (2010).

![Figure F.0.18](image)

Figure F.0.18 – Effect of the gas mass flowrate on the separator efficiency. The simulation was carried out at the default operating conditions.

To gain further understanding on the effect of gas flow rate on the overall hydrodynamics, Figure F.0.19 shows the gas velocity vectors as function of the gas mass flow rate. It is clear that there is a significant change in the magnitude of the gas velocity but little change in the flow pattern. There is also evidence of a significant change in the solid concentration around the cone deflector as shown in Figure F.0.20. Despite this, such a dramatic change caused no effect on the separator efficiency due to counterbalance of forces, which are described as follows:
i) At a high gas velocity, there is considerable increase in the pressure drop between the gas exit pipe and its surroundings, hence high upward gas velocity (drag force), as shown in Figure F.0.21(a). However, this is counterbalanced by the considerable reduction in the solid concentration in the wall and the core region below the exit pipe, as shown in Figure F.0.21(b).

ii) At a low gas velocity, there is high solid concentration at the wall (i.e. high gravity force), as shown in Figure F.0.21(b). This is associated with low pressure drop between the wall and the tip of the exit pipe. Hence, there is reduction in the solid migration from the wall to the core (i.e. low radial gas drag force) or solid carry over by the reversing gas (i.e. low upward gas drag force)

![Figure F.0.19 – Gas velocity vectors at various gas flow rates. The color code bars indicate the range of gas velocity](image)
Figure F.0.20 – Solid volume fraction at various gas mass flow rates for the particle size $d_s = 206 \, \mu m$. To allow visualization of the low ranges, the magnitude of the solid volume fraction is restricted here to a maximum of $2 \times 10^{-4}$. The color code bar indicates the range of the solid volume fraction.

Figure F.0.21 – Effect of the gas flowrate on the (a) radial profiles of axial gas velocity and (b) solid volume fraction (for the selected particle size $d_s = 206 \, \mu m$). The data was taken at the sample level of 3.9 cm below the gas exit pipe tip.
According to the above analysis, it is concluded that the gas velocity has little effect on the separator efficiency, at least within the operating conditions considered here. In biomass pyrolysis, however, the gas velocity has a critical effect on the product quality due to its effect on the gas and solid residence time. The residence time can be quantified through the average gas velocity, particularly within the GDH region, as discussed in Section 4.2. The interrelation between the gas velocity, gas/solid residence times and the GDH in a downer pyrolysis reactor is a complex one and requires careful optimization in order to achieve the best product quality.

F.4.7 Effect of Solid Mass Flow Rate

The effect of solid mass flow rate on the separator efficiency was investigated using four different flow rates of \( m_s = 0.004 \) kg/s, 0.02 kg/s, 0.04 kg/s and 0.08 kg/s at a fixed gas mass flow rate of \( m_g = 0.0039 \) kg/s. This corresponded to solid to gas flow ratios (solid loading) of \( m_s/m_g = 1, 5, 10 \) and 20 respectively. All the other operating conditions were set to the default values. The experimental and predicted results, shown in Figure F.0.22, suggest that the separator efficiency improves as the solid loading increases within the range of \( \dot{m}_s/\dot{m}_g < 10 \), beyond which the efficiency appears to be independent of solid loading. This trend is less pronounced in the predicted data, which show very limited changes. Quantitatively, there is an over-prediction of separator efficiency when compared with the experiment data; particularly at low solid loading.
Figure F.0.22 – Effect of the solid loading on the separator efficiency. The simulation was carried out at the default operating conditions.

Figure F.0.23 shows the velocity vectors as a function of the solid loading. In the wall region below the cone deflector, there is a clear change in the magnitude and direction of the gas velocity vector, particularly in the right hand side below the gas exit pipe. This implies an increased downward gas drag force, which positively adds to the solid gravity force. It is therefore concluded that as the solid loading increases the amount of solid entrained by the reversing gas at the central region below the cone deflector reduces. The solid concentration in the wall region massively increases while the core region remains relatively constant which can be seen in Figure F.0.24. The solid concentration and velocity profiles at the sample level, shown in Figure F.0.25, indicate considerable hydrodynamic changes below the cone deflector as the solid loading increased. The gas velocity, however, shows exactly the opposite behavior with the axial velocity in the core region more than doubled when increasing the solid flow rate from 0.004 to 0.08 kg/s, while the velocity near the walls is slightly increased. Because the increase in the axial upward gas velocity in the centre takes place in a region that is at extremely low in solid concentration, the separator efficiency remains almost independent of the increase in
solid loading. Accordingly, it is recommended to operate this downer reactor at a high solid flow rate for the following three main advantages:

i) Increased upward gas velocity towards the gas outlet pipe within the GDH region, therefore reducing the gas residence time in the reactor.

ii) Improved separator efficiency, as evident from the experimental and predicted results.

iii) Increasing the reactor processing capacity for biomass pyrolysis.

Figure F.0.23 – Gas velocity vectors at various solid flowrates. To allow visualization of the low ranges, the magnitude of the velocity vector was restricted to a maximum of 5m/s. The color code bar indicates the range of gas velocity
Figure F.0.24 – Solid volume fraction at various solid flow rates for the selected particle size $d_s = 206 \, \mu m$. The color code bars indicate the range of solid volume concentration.

Figure F.0.25 – Effect of the solid mass flow rate on (a) radial profiles of axial gas velocity and (b) solid volume fraction (for the selected particle size $d_s = 206 \, \mu m$). The data was taken at the sample level of 3.9 cm below the gas exit pipe tip.
F.4.8 Effect of the Particle Restitution and Specularity Coefficients

It is understood that the particle size plays a major role on the separator efficiency such that the larger the particles size the higher the separator efficiency. Another important parameter of interest here is the degree of particle momentum loss or rebound upon hitting the solid surfaces, which is defined in the model through the restitution coefficient and specularity coefficient. The effective particle-wall restitution coefficient ($e_{sw}$) was determined experimentally by the co-authors from the ICFAR by measuring the rebound velocities of silica sand when hitting various types of solid surfaces; giving restitution coefficients ranging from 0.73 (Plexiglas surface) to 0.48 (paper surface). In this study, the same range of particle restitution coefficient was implemented in the model to investigate the effect of this parameter on the separator efficiency. The effect of the specularity coefficient ($\varphi$) was investigated by using values of $\varphi=0$, 0.1 and 1.0, thus covering the two extreme ends of particle-wall interaction; free slip condition at $\varphi=0$ and no slip condition at $\varphi=1$. Reported studies (e.g. Jin et al., 2010) have shown that the restitution coefficients have an effect on the solid velocity, gas velocity and solid concentration. It is also understood that as the restitution coefficient increases there is a corresponding increase in the wall shear stress. The specularity coefficient, on the other hand, has been reported to have a pronounced effect on the solid concentration, as increasing this parameter results in reducing the solid concentration at the wall.

Figure F.0.26 shows a comparison between the predicted and measured separator efficiency as a function of the particle restitution coefficient. It is clear that the predicted separator efficiency is a very weak function of this parameter. This is in good agreement with some of the reported literature (e.g. Jin et al., 2010) which suggest that the particle restitution coefficient (in the range 0.6~0.99) has limited effects on the solid velocity, gas velocity and the solid concentration in circulating fluidized bed reactors. The experimental data shows a slight decrease in the efficiency as the restitution coefficient decreases; however, this is still within a very limited range.
Figure F.0.26 – Effect of the particle–wall restitution coefficient ($e_{s,w}$) on the separator efficiency. The simulation was carried out at the default operating conditions.

Figure F.0.27 shows the changes in the predicted separator efficiency with changing the value of the secularity coefficient. While there is clear drop in the efficiency as the secularity coefficient increases, this is still within a very limited range. This change is believed to be a result of the increase in the wall shear stress (no-slip condition), which in turn results in hindering the downward flow of the dense wall layer and hence giving rise to particle migration from the wall to the core followed by entrainment by the reversing gas towards the exit pipe. This phenomenon is demonstrated by the changes in the solid velocity and concentration profiles shown in Figure F.0.28. The secularity coefficient appears to have a significant effect on the gas velocity and solid concentration at the wall regions, which is in good agreement with the observation reported by Jin et al (2010), and in spite of this there is a negligible effect on the separator efficiency.
Figure F.0.27 – Effect of specularity coefficient on the separator efficiency. The simulation was carried out at the default operating conditions.

Figure F.0.28 – Effect of the specularity coefficient on the (a) radial profiles of axial gas velocity and (b) solid volume fraction (for the selected particle size $d_s = 206 \, \mu m$). The data was taken at the sample level of 3.9 cm below the tip of the gas exit pipe.
F.5 Conclusions

The hydrodynamics in a downer pyrolysis reactor equipped with a novel gas-solid separator have been investigated theoretically using an Eulerian-Eulerian (two-fluid) CFD model. The novel separator, which consists of a cone deflector and a gas outlet pipe, was designed by the co-authors from the ICFAR (Canada). The model predictions were compared with experimental measurements of separator efficiency. This study revealed interesting hydrodynamic features around the cone deflector, where due to the restriction of the flow passage and solid deflection towards the walls, the region below the deflector in the downer reactor was completely transformed to behave like a riser, characterized by distinct upward gas flow at the core and dense falling solid layer at the walls. These distinct hydrodynamic features allowed for high efficiency of gas-solid separation up to 100%. A new method for estimating the gas disengagement height (GDH) was developed to help in estimating the gas residence time in this novel reactor. This study also included detailed sensitivity analysis of the separator efficiency towards the various operating conditions, including the effect of particle restitution and secularity coefficients. In the second part of this study, the present hydrodynamic model will be extended to include reaction kinetics and heat transfer to simulate the reactor thermochemical performance during the pyrolysis of biomass.
F.6 Notation

\( a \)  
Gap between conical deflector and reactor wall (m)

\( A \)  
Model parameter (-)

\( B \)  
Model parameter (-)

\( C \)  
Courant number (-)

\( C_D \)  
Drag coefficient (-)

\( C_{\mu}, C_{1\varepsilon}, C_{2\varepsilon} \)  
Constants (-)

\( C_{fr,s_i s_j} \)  
Friction coefficient between solid phase \( i \) and phase \( j \) (-)

\( d_{s_i} \)  
Particle diameter of solid phase \( i \) (m)

\( D_c \)  
Reactor diameter (m)

\( D_{go} \)  
Diameter of gas outlet pipe (m)

\( e_{s_i s_j} \)  
Particle-particle restitution coefficient (-)

\( e_{s_i w} \)  
Particle-wall restitution coefficient (-)

\( g \)  
Gravity (m s\(^{-2}\))

\( g_0 \)  
Radial distribution function (-)

\( G_{k,g} \)  
Production of turbulent kinetic energy (kg m\(^{-1}\) s\(^{-2}\))

\( \overline{I} \)  
Unit vector (-)

\( I_{2D} \)  
Second invariant of the deviatoric stress tensor (s\(^2\))

\( L_1, L_2 \)  
Reactor dimension (m)

\( L_s \)  
Separation length (m)

\( k_g \)  
Turbulence kinetic energy (m\(^2\) s\(^{-2}\))

\( m_{collected}, m_{in} \)  
Mass of collected and fed solid particles respectively (kg)
\(m_g, m_s\)  Mass flow rate of gas and solid respectively (kg s\(^{-1}\))

\(P\)  Pressure (pa)

\(\bar{S}\)  Strain rate (s\(^{-1}\))

\(Re_{si}\)  Reynolds number of solid phase \(i\) (-)

\(t\)  Time (s)

\(\bar{u}_g, \bar{u}_{si}\)  Gas and solid velocity vector (m s\(^{-1}\))

\(u_{si,w}\)  Particle velocity at wall (m s\(^{-1}\))

\(v_{r,si}\)  Terminal velocity correlation (-)

\(X_{10}, X_{50}, X_{90}\)  Particle size at accumulative volume fraction at 10%, 50%, 90%

**Greek symbols**

\(\alpha\)  Angle of conical deflector (Degree)

\(\alpha_g, \alpha_{si}\)  Volume fraction of gas and solid phase \(i\) respectively (-)

\(\beta\)  Momentum exchange coefficient (kg m\(^{-3}\) s\(^{-1}\))

\(\gamma_{\theta_{si}}\)  Collisional energy dissipation (kg m\(^{-1}\) s\(^{-3}\))

\(\varepsilon_g\)  Turbulent dissipation rate (m\(^2\) s\(^{-3}\))

\(\eta\)  Separation efficiency (-)

\(\Theta_{si}\)  Granular temperature of solid phase \(i\) (m\(^2\) s\(^{-2}\))

\(\kappa_{\theta_{si}}\)  Diffusion coefficient of granular energy (kg m\(^{-1}\) s\(^{-1}\))

\(\lambda_{si}\)  Particle bulk viscosity (kg m\(^{-1}\) s\(^{-1}\))

\(\mu_{l,g}, \mu_{t,g}\)  Viscosity of gas phase due to laminar, turbulent flow (kg m\(^{-1}\) s\(^{-2}\))

\(\mu_{si,col}\)  Viscosity of solid phase \(i\) due to collision (kg m\(^{-1}\) s\(^{-1}\))

\(\mu_{si,kin}\)  Viscosity of solid phase \(i\) due to kinetics (kg m\(^{-1}\) s\(^{-1}\))

\(\mu_{si,fr}\)  Viscosity of solid phase \(i\) due to friction (kg m\(^{-1}\) s\(^{-1}\))

\(\Pi_{k,g}\)  Influence of solid phases on gas phase (m\(^2\) s\(^{-3}\))

\(\Pi_{e,g}\)  Influence of solid phases on gas phase (m\(^2\) s\(^{-4}\))
\( \rho_s, \rho_g \)  
Solid and gas densities respectively (kg m\(^{-3}\))

\( \overline{\tau} \)  
Shear stress tensor (kg m\(^{-1}\) s\(^{-2}\))

\( \sigma_k, \sigma_\varepsilon \)  
Constants (-)

\( \phi \)  
Angle of Internal friction (Degree)

\( \phi_{kSi} \)  
Energy exchange between phase k and solid phase i (kg m\(^{-1}\) s\(^{-1}\))

\( \varphi \)  
Specularity coefficient (-)

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### F.7 References


Appendix G – Gas Composition Model Iterative Solver

This appendix provides code developed in Visual Basic to automatically obtain the gas residence time distributions from a batch of experimental pressure drop signals by solving a gas mixture composition model iteratively for each experiment. The code is initiated from Microsoft Excel, and the code outputs and prints equations and results to a Microsoft Excel spreadsheet. The welcome screen to the solver is shown in Figure G.0.1.

![Welcome screen to gas composition model iterative solver](image)

```vbnet
Option Base 1
Sub routine()

'Declaring variables
Dim headers, i, ii, t1, t2, expl As Integer
Dim solverdir, filetype, spath, sfil, sfil1 As String
Dim newbook As Workbook
Dim v As Range
Dim pavg, vavg, vres, t_step, C_1t As Single
Dim dp_s(), vres1() As Single
Dim DPs_chart, Fno_chart, Fnhei_chart, Fnheo_chart, x_chart, F_chart, E_chart As ChartObject

'Declaring constants
solverdir = Workbooks("GAS COMPOSITION MODEL ROUTINE.xlsm").Worksheets("Sheet1").Range("B4")
filetype = Workbooks("GAS COMPOSITION MODEL ROUTINE.xlsm").Worksheets("Sheet1").Range("B5")
```

headers = Workbooks("GAS COMPOSITION MODEL ROUTINE.xlsm").Worksheets("Sheet1").Range("B6")
t1 = Workbooks("GAS COMPOSITION MODEL ROUTINE.xlsm").Worksheets("Sheet1").Range("B7")
t2 = Workbooks("GAS COMPOSITION MODEL ROUTINE.xlsm").Worksheets("Sheet1").Range("B8")
dt = Workbooks("GAS COMPOSITION MODEL ROUTINE.xlsm").Worksheets("Sheet1").Range("B9")
dt1 = Workbooks("GAS COMPOSITION MODEL ROUTINE.xlsm").Worksheets("Sheet1").Range("B10")
exp1 = Workbooks("GAS COMPOSITION MODEL ROUTINE.xlsm").Worksheets("Sheet1").Range("B11")
a_dps = Workbooks("GAS COMPOSITION MODEL ROUTINE.xlsm").Worksheets("Sheet1").Range("B14")
b_dps = Workbooks("GAS COMPOSITION MODEL ROUTINE.xlsm").Worksheets("Sheet1").Range("B15")
a_dphe = Workbooks("GAS COMPOSITION MODEL ROUTINE.xlsm").Worksheets("Sheet1").Range("B16")
b_dphe = Workbooks("GAS COMPOSITION MODEL ROUTINE.xlsm").Worksheets("Sheet1").Range("B17")
c_1 = Workbooks("GAS COMPOSITION MODEL ROUTINE.xlsm").Worksheets("Sheet1").Range("B18")
gamma1 = Workbooks("GAS COMPOSITION MODEL ROUTINE.xlsm").Worksheets("Sheet1").Range("B19")
c_3 = Workbooks("GAS COMPOSITION MODEL ROUTINE.xlsm").Worksheets("Sheet1").Range("B20")
gamma2 = Workbooks("GAS COMPOSITION MODEL ROUTINE.xlsm").Worksheets("Sheet1").Range("B21")
gamma3 = Workbooks("GAS COMPOSITION MODEL ROUTINE.xlsm").Worksheets("Sheet1").Range("B22")
c_He = Workbooks("GAS COMPOSITION MODEL ROUTINE.xlsm").Worksheets("Sheet1").Range("B23")
c_2 = Workbooks("GAS COMPOSITION MODEL ROUTINE.xlsm").Worksheets("Sheet1").Range("B24")
c_5 = Workbooks("GAS COMPOSITION MODEL ROUTINE.xlsm").Worksheets("Sheet1").Range("B25")
c_6 = Workbooks("GAS COMPOSITION MODEL ROUTINE.xlsm").Worksheets("Sheet1").Range("B26")
gamma4 = Workbooks("GAS COMPOSITION MODEL ROUTINE.xlsm").Worksheets("Sheet1").Range("B27")
a_dpsphe = Workbooks("GAS COMPOSITION MODEL ROUTINE.xlsm").Worksheets("Sheet1").Range("B28")
b_dpsphe = Workbooks("GAS COMPOSITION MODEL ROUTINE.xlsm").Worksheets("Sheet1").Range("B29")
vol_sparg = Workbooks("GAS COMPOSITION MODEL ROUTINE.xlsm").Worksheets("Sheet1").Range("B30")
gamma_sp = Workbooks("GAS COMPOSITION MODEL ROUTINE.xlsm").Worksheets("Sheet1").Range("B31")
c_ltr = Workbooks("GAS COMPOSITION MODEL ROUTINE.xlsm").Worksheets("Sheet1").Range("B32")
vol = Workbooks("GAS COMPOSITION MODEL ROUTINE.xlsm").Worksheets("Sheet1").Range("B33")

'Specifying path to experimental files to be analyzed
spath = "C:\Signal_analysis\data_files"
ChDir spath
sfil = Dir("*.*" & filetype)
'Do While loop performs operations on each data file in the specified experimental file path

i = 0
Do While sfil <> ""
    i = i + 1
    'Open next data file
    If Len(sfil) - Len(filetype) - 1 > 31 Then
        sfil1 = Mid(sfil, 1, 31)
    Else
        sfil1 = Mid(sfil, 1, Len(sfil) - Len(filetype) - 1)
    End If
    Set v = Workbooks.Open(sfil).Worksheets(sfil1).Range("B" & headers + 1 & ":D" & headers + 10000)

    'Create a new workbook to output results
    Application.DisplayAlerts = False
    Set newbook = Workbooks.Add
    With newbook
        .Title = sfil1
        .SaveAs filename:=sfil1 & ".xlsm", FileFormat:=52
    End With
    Application.DisplayAlerts = True
    '''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''''
\[ dp_s(ii) = a_dps \times v(ii, 1) + b_dps \]
\[ \text{Workbooks(sfil1 & ".xlsm").Worksheets("Sheet1").Range("C" \\ ii + 1)} = dp_s(ii) \]
\[ \text{Workbooks(sfil1 & ".xlsm".Worksheets("Sheet1").Range("D" \\ ii + 1).Formula} = "\text{Sheet2!$B$1*Sheet1!B" & ii + 1 & "+Sheet2!$B$2"} \]

Next ii

'1-02 Goal seek tracer orifice plate pressure drop to zero during the second steady state period
\[ \text{Workbooks(sfil1 & ".xlsm".Worksheets("Sheet2").Range("A3") = "DP_he2"} \]
\[ \text{Workbooks(sfil1 & ".xlsm".Worksheets("Sheet2").Range("B3").Formula} = "AVERAGE(Sheet1!$D$" & t2 / dt + 2 & "$:Sheet1!$D$" & mm + 1 & _ ")" \]
\[ \text{Workbooks(sfil1 & ".xlsm".Worksheets("Sheet2").Range("B3").GoalSeek _ goal:=0, _ } \\
\text{changingcell:=Workbooks(sfil1 & ".xlsm".Worksheets("Sheet2").Range("B2")} \]

'1-03 Calculate time of downstep
\[ vavg = 0 \]
\[ vres = 0 \]
For ii = 1 To t1 / dt
\[ vavg = vavg + v(ii, 3) \]
Next ii
\[ vavg = vavg / (t1 / dt) \]
ReDim vres1(t1 / dt)
For ii = 1 To t1 / dt
\[ vres1(ii) = \text{Abs}(v(ii, 3) - vavg) \]
Next ii
\[ \text{vresmax = WorksheetFunction.Max(vres1)} \]
For ii = t1 / dt To mm
\[ \text{If v(ii, 3) < vavg - 1.5 * vresmax Or v(ii, 3) > vavg + 1.5 * vresmax Then} \]
\[ t\_step = ii \times dt \]
Exit For
End If
Next ii
\[ \text{Workbooks(sfil1 & ".xlsm".Worksheets("Sheet2").Range("A4") = "t\_step"} \]
\[ \text{Workbooks(sfil1 & ".xlsm".Worksheets("Sheet2").Range("B4") = t\_step} \]

'1-04 Calculate transient tracer inlet molar flowrate through sparger (based on calibration performed 11/05/2013)
\[ \text{Workbooks(sfil1 & ".xlsm".Worksheets("Sheet2").Range("E21") = "a\_DPsparg"} \]
\[ \text{Workbooks(sfil1 & ".xlsm".Worksheets("Sheet2").Range("F21") = a\_dpsphe} \]
\[ \text{Workbooks(sfil1 & ".xlsm".Worksheets("Sheet2").Range("E12") = "DP\_hesparg1"} \]
\[ \text{Workbooks(sfil1 & ".xlsm".Worksheets("Sheet2").Range("F12").Formula} = "=Sheet2!$F$21*AVERAGE(Sheet1!D2:D" & t1 / dt + 2 & "")" \]
Workbooks(sf1 & ".xlsm").Worksheets("Sheet2").Range("E13") = "DP_s1"
Workbooks(sf1 & ".xlsm").Worksheets("Sheet2").Range("F13").Formula = 
"=AVERAGE(Sheet1!C2:C" & t1 / dt + 1 & ")"
Workbooks(sf1 & ".xlsm").Worksheets("Sheet2").Range("E14") = "F_nhei1"
Workbooks(sf1 & ".xlsm").Worksheets("Sheet2").Range("F14").Formula = 
"=C_He * SQRT(2*AVERAGE(Sheet1!D2:D" & t1 / dt + 1 & ")+Sheet2!$F$12+Sheet2!$F$13+101500)/0.004008/8.314/293*AVERAGE(Sheet1!
D2:D" & 
Workbooks(sf1 & ".xlsm").Worksheets("Sheet2").Range("E18") = "V_sparg"
Workbooks(sf1 & ".xlsm").Worksheets("Sheet2").Range("F18") = vol_sparg
Workbooks(sf1 & ".xlsm").Worksheets("Sheet2").Range("E15") = "C_dpsparg"
Workbooks(sf1 & ".xlsm").Worksheets("Sheet2").Range("F15").Formula = 
"=(Sheet2!$F$12*(Sheet2!$F$12+Sheet2!$F$13+101500)^" & _
"(Sheet2!$F$20-1)/Sheet2!$F$14^Sheet2!$F$20)^((Sheet2!$F$20-1)/Sheet2!$F$20)*Sheet2!$F$18/8.314/293"
Workbooks(sf1 & ".xlsm").Worksheets("Sheet2").Range("E16") = "dDP"
Workbooks(sf1 & ".xlsm").Worksheets("Sheet2").Range("F16").Formula = 
"=-(Sheet1!$F$12*(1001-" & ii + 1 & ")/1000+1"
Workbooks(sf1 & ".xlsm").Worksheets("Sheet3").Range("A1") = "DP_spargi"
Workbooks(sf1 & ".xlsm").Worksheets("Sheet3").Range("B1") = "F_i(DP)"
Workbooks(sf1 & ".xlsm").Worksheets("Sheet3").Range("C1") = "t_i"
Workbooks(sf1 & ".xlsm").Worksheets("Sheet3").Range("D1") = "F_nhei"
Workbooks(sf1 & ".xlsm").Worksheets("Sheet3").Range("E1") = "F_nhei*dt_i"
For ii = 1 To 1000
Workbooks(sf1 & ".xlsm").Worksheets("Sheet3").Range("A" & 
For ii = 1 To 999
Workbooks(sf1 & ".xlsm").Worksheets("Sheet3").Range("D" & 
Next ii
For ii = 1 To 999
Workbooks(sf1 & ".xlsm").Worksheets("Sheet3").Range("B" & 
Next ii
"& _
'1-05 Smooth separator pressure drop signal with normal probability distribution functions

'1-05-01 Print headers
Workbooks(sfil1 & ".xlsm").Worksheets("Sheet2").Range("A5") = "DP_s SMOOTHING PARAMETERS"
Workbooks(sfil1 & ".xlsm").Worksheets("Sheet2").Range("A6") = "Peak->"
Workbooks(sfil1 & ".xlsm").Worksheets("Sheet2").Range("A7") = "Mean"
Workbooks(sfil1 & ".xlsm").Worksheets("Sheet2").Range("A8") = "SD"
Workbooks(sfil1 & ".xlsm").Worksheets("Sheet2").Range("A9") = "c"
Workbooks(sfil1 & ".xlsm").Worksheets("Sheet2").Range("A10") = "alpha"
Workbooks(sfil1 & ".xlsm").Worksheets("Sheet2").Range("B6") = "1"
Workbooks(sfil1 & ".xlsm").Worksheets("Sheet2").Range("C6") = "2"
Workbooks(sfil1 & ".xlsm").Worksheets("Sheet2").Range("D6") = "3"
Workbooks(sfil1 & ".xlsm").Worksheets("Sheet2").Range("A11") = "SUM(DP_s sm pk)"
Workbooks(sfil1 & ".xlsm").Worksheets("Sheet2").Range("A12") = "DP_s1"
Workbooks(sfil1 & ".xlsm").Worksheets("Sheet2").Range("A13") = "DP_s2"
Workbooks(sfil1 & ".xlsm").Worksheets("Sheet2").Range("A14") = "S_dpssm"
Workbooks(sfil1 & ".xlsm").Worksheets("Sheet2").Range("E7") = "S_dpdt2"

'1-05-02 Initialize pressure drop smoothing parameters
Workbooks(sfil1 & ".xlsm").Worksheets("Sheet2").Range("B7").Formula = t_step
Workbooks(sfil1 & "xlsm").Worksheets("Sheet2").Range("C7").Formula = t_step + 0.1
Workbooks(sfil1 & "xlsm").Worksheets("Sheet2").Range("D7").Formula = t_step + 0.2
Workbooks(sfil1 & "xlsm").Worksheets("Sheet2").Range("B8") = 0.1
Workbooks(sfil1 & "xlsm").Worksheets("Sheet2").Range("C8") = 0.2
Workbooks(sfil1 & "xlsm").Worksheets("Sheet2").Range("D8") = 0.5

Workbooks(sfil1 & "xlsm").Worksheets("Sheet2").Range("B10") = 0.5
Workbooks(sfil1 & "xlsm").Worksheets("Sheet2").Range("C10") = -0.1
Workbooks(sfil1 & "xlsm").Worksheets("Sheet2").Range("D10") = -0.1

'1-05-03 Calculates sampled time signal
Workbooks(sfil1 & "xlsm").Worksheets("Sheet1").Range("Z1") = "Time tr"
For ii = 1 To mm * dt / dt1
    Workbooks(sfil1 & "xlsm").Worksheets("Sheet1").Range("Z" & ii + 1).Formula = _
    "=INDEX(A2:A" & mm + 1 & "," & dt1 / dt & _
    "*(" & ii & "+1/2))"
    Next ii
    Application.Calculate

'1-05-04 Calculates sampled pressure drop signal
For ii = 1 To mm * dt / dt1
    Workbooks(sfil1 & "xlsm").Worksheets("Sheet1").Range("E" & ii + 1).Formula = _
    "=INDEX(Sheet1!C2:Sheet1!C" & mm + 1 & "," & dt1 / dt * (ii - 1 / 2) & ")"
    Next ii

'1-05-05 Calculates steady state average pressure drops
Workbooks(sfil1 & "xlsm").Worksheets("Sheet2").Range("B12").Formula = _
    "=AVERAGE(Sheet1!E2:Sheet1!E" & t1 / dt1 + 1 & ")"
Workbooks(sfil1 & "xlsm").Worksheets("Sheet2").Range("B13").Formula = 
    "=AVERAGE(Sheet1!E" & t2 / dt1 + 2 & ":Sheet1!E" & _
    mm * dt / dt1 + 1 & ")"

'1-05-06 Calculates smoothed pressure drop signal
For ii = 1 To mm * dt / dt1
    Workbooks(sfil1 & "xlsm").Worksheets("Sheet1").Range("F" & ii + 1).Formula = _
    "=IF(Sheet1!Z" & ii + 1 & ",Sheet2!B12,
    "<Sheet2!$B$4,Sheet2!$B$12,IF(Sheet1!Z" & ii + 1 & ">") & t2 & 
    ",Sheet2!$B$13," & _
    "(Sheet2!$B$12-
    Sheet2!$B$13)*(Sheet2!$B$10*NORMDIST(Sheet1!Z" & ii + 1 & 
    ",Sheet2!$B$7,Sheet2!$B$8,FALSE)+" _
    & "Sheet2!$C$10*NORMDIST(Sheet1!Z" & ii + 1 & 
    ",Sheet2!$C$7,Sheet2!$C$8,FALSE)+Sheet2!$D$10*" _
'1-05-07 Calculate derivative of separator pressure drop signal and its absolute value
For ii = 2 To mm * dt / dt1 - 1
    Workbooks(sfil1 & ".xlsm").Worksheets("Sheet1").Range("$H$" & ii + 1).Formula = 
        "=(Sheet1!$F$" & ii + 2 & "+-Sheet1!$F$" & ii & ")/2/" & dt1
Next ii

'1-05-08 Calculates pressure drop smoothing objective optimization function
Workbooks(sfil1 & ".xlsm").Worksheets("Sheet2").Range("B14").Formula = 
    "=SUMXMY2(Sheet1!$E$2:Sheet1!$E" & t2 / dt1 + 1 & "Sheet1!$F$2:Sheet1!$F$" & t2 / dt1 + 1 & ")"

'1-05-09 Runs Solver to optimize pressure drop smoothing
Workbooks(sfil1 & ".xlsm").Worksheets("Sheet2").Activate
SolverReset
SolverOptions maxtime:=600, iterations:=1000,
    scaling:=True, convergence:=0.0000000001
    solveradd cellref:="$B$8:$D$8", relation:=3,
    formulatext:="1e-6"
SolverOk SetCell:="$B$14", MaxMinVal:=2, ValueOf:="0",
    ByChange:= _"$B$7:$D$8,$B$10:$D$10"
SolverSolve userfinish:=True

'1-06 Calculate absolute pressure in separator
Workbooks(sfil1 & ".xlsm").Worksheets("Sheet1").Range("J1") = "P_s"
For ii = 1 To mm * dt / dt1
    Workbooks(sfil1 & ".xlsm").Worksheets("Sheet1").Range("$J$" & ii + 1).Formula = 
        "=Sheet1!$F$" & ii + 1 & "+101500"
Next ii

'1-07 CALCULATE SEPARATOR PRESSURE DROP CONSTANTS, NOMINAL DOWNER AIR MOLAR FLOWRATE, AND DOWNER VOLUME

'1-07-01 Prints headers and calculates nominal flowrates
Workbooks(sfil1 & ".xlsm").Worksheets("Sheet2").Range("A15") = "C_2"
Workbooks(sfil1 & ".xlsm").Worksheets("Sheet2").Range("B15") = c_2
Workbooks(sfil1 & ".xlsm").Worksheets("Sheet2").Range("A16") = "C_4"
Workbooks(sfil1 & ".xlsm").Worksheets("Sheet2").Range("B16") = "C_4"
Workbooks(sfil1 & ".xlsm").Worksheets("Sheet2").Range("B16") = "C_4"
Workbooks(sfil1 & ".xlsm").Worksheets("Sheet2").Range("A16") = "C_4"
Workbooks(sfil1 & ".xlsm").Worksheets("Sheet2").Range("E1") = "C_lss"
'1-07-02 Loads solids mass flowrate (if applicable)
F_ms = Workbooks("GAS COMPOSITION MODEL ROUTINE.xlsm").Worksheets("Sheet1").Range("K" & i + 4)

'1-07-03 Calculates air inlet molar flowrate
If exp1 = 1 Or exp1 = 4 Then
  Workbooks(sfil & ".xlsm").Worksheets("Sheet2").Range("B17").Formula = _
  "=((Sheet2!$B$13-(" & c_5 & "*Sheet2!$B$17*0.02897+" & c_6 & ")*" & F_ms & "] & gamma4 & ")*((Sheet2!$B$13+101500)*" & _
  "/8.314/293]^" & gamma1 - 1 & 
  "/Sheet2!$F$1/0.02897)^(1/" & gamma1 & ")"
Else
  Workbooks(sfil & ".xlsm").Worksheets("Sheet2").Range("B17").Formula = _
"=((Sheet2!$B$12-
Sheet2!$B$16*(Sheet2!$B$17*Sheet2!$F$2)^ gamma3 & gamma2 & "-(" & c_5 & 
"*Sheet2!$B$17*Sheet2!$F$2+" & c_6 & ")*" & F_ms & 
"^" & gamma4 & 
")*((Sheet2!$B$12+101500)/8.314/293)^ gamma1 - 1 & "/Sheet2!$F$1/Sheet2!$F$2)^(1/" & gamma1 & ")"
End If
With Application
 .Calculation = xlCalculationAutomatic
 .Iteration = True
 .MaxIterations = 20
 .MaxChange = 0.0000000001
End With
With Application
 .Calculation = xlCalculationManual
 .Iteration = False
End With

'1-08 CALCULATE ITERATIVE COMPONENTS

'1-08-01 Prints column headers
 Workbooks(sfil1 & ".xlsm").Worksheets("Sheet1").Range("K1") = "DP_she"
 Workbooks(sfil1 & ".xlsm").Worksheets("Sheet1").Range("L1") = "DP_sa"
 Workbooks(sfil1 & ".xlsm").Worksheets("Sheet1").Range("AA1") = "DP_ss"
 Workbooks(sfil1 & ".xlsm").Worksheets("Sheet1").Range("M1") = "DP_sahes"
 Workbooks(sfil1 & ".xlsm").Worksheets("Sheet1").Range("N1") = "F_nao"
 Workbooks(sfil1 & ".xlsm").Worksheets("Sheet1").Range("O1") = "F_nhei theo"
 Workbooks(sfil1 & ".xlsm").Worksheets("Sheet1").Range("P1") = "F_no"
 Workbooks(sfil1 & ".xlsm").Worksheets("Sheet1").Range("Q1") = "F_nheo exp"
 Workbooks(sfil1 & ".xlsm").Worksheets("Sheet1").Range("R1") = "x"
 Workbooks(sfil1 & ".xlsm").Worksheets("Sheet1").Range("S1") = "M"
 Workbooks(sfil1 & ".xlsm").Worksheets("Sheet1").Range("T1") = "DP_s fit"
 Workbooks(sfil1 & ".xlsm").Worksheets("Sheet1").Range("U1") = "F(t) model"
 Workbooks(sfil1 & ".xlsm").Worksheets("Sheet1").Range("AF1") = "F_naotr"
 Workbooks(sfil1 & ".xlsm").Worksheets("Sheet1").Range("AG1") = "F_naoss"

'1-08-02 Resets time of downstep and associated signal
times to sampled signal downstep time, then recalculates workbook
'This step ensures that the later convolution calculation
will result in a smooth, continuous signal
 t_step1 = t_step
 n_step = WorksheetFunction.MRound(t_step, dt1) / dt1 + 2
t_step = Workbooks(sfil1 & ".xlsm").Worksheets("Sheet1").Range("Z" & n_step)

Workbooks(sfil1 & ".xlsm").Worksheets("Sheet2").Range("B4") = t_step

If Workbooks(sfil1 & ".xlsm").Worksheets("Sheet2").Range("B8") = _
t_step1 Then Workbooks(sfil1 & ".xlsm").Worksheets("Sheet2").Range("B8") = t_step

If Workbooks(sfil1 & ".xlsm").Worksheets("Sheet2").Range("C8") = _
t_step1 Then Workbooks(sfil1 & ".xlsm").Worksheets("Sheet2").Range("C8") = t_step

With Application
  .Calculation = xlCalculationAutomatic
  .Iteration = True
  .MaxIterations = 20
  .MaxChange = 0.0000000001
End With

With Application
  .Calculation = xlCalculationManual
  .Iteration = False
End With

'1-08-03 Codes for iterative signals
For ii = 1 To mm * dt / dt1

  '1-08-03-01 Separator pressure drop contribution from tracer
  Workbooks(sfil1 & ".xlsm").Worksheets("Sheet1").Range("K" & ii + 1).Formula = 
  "+=Sheet2!$B$16*(Sheet1!N" & ii + 1 & 
  "*Sheet1!S" & ii + 1 & ")^" & gamma3 & "+Sheet1!Q" & ii + 1 & ")^" & gamma2

  '1-08-03-02 Separator pressure drop contribution from downer air
  Workbooks(sfil1 & ".xlsm").Worksheets("Sheet1").Range("L" & ii + 1).Formula = 
  "+=IF(Sheet1!Z" & ii + 1 & "<Sheet2!$B$4," & _
  "Sheet2!$F$1*Sheet1!N" & ii + 1 & "^" & gammal & 
  "*0.02897*(8.314*293/Sheet1!J" & ii + 1 & ")^" & gammal - 1 & _
  "+(Sheet2!$F$1*Sheet1!AG" & ii + 1 & "^" & gammal & 
  "*0.02897+Sheet2!$F$6*Sheet1!AF" & ii + 1 & "^" & gammal & _
  "*Sheet1!S" & ii + 1 & ")^" & gammal - 1 & ")" & (8.314*293/Sheet1!J" & ii
  + 1 & ")^" & gammal - 1 & "

  '1-08-03-03 Separator pressure drop contribution from solids
  Workbooks(sfil1 & ".xlsm").Worksheets("Sheet1").Range("AA" & ii + 1).Formula = 
  "+=" & c_5 & "N" & ii + 1 & "S" & ii + 1 & "^" & c_6 & 
  "F_ms" & "^" & gammal & F_ms & "^" & gammal

  '1-08-03-04 Separator pressure drop
  Workbooks(sfil1 & ".xlsm").Worksheets("Sheet1").Range("M" & ii + 1).Formula = 
  "+Sheet1!K" & ii + 1 & "+Sheet1!L" & ii + 1 & _ 

"}
'1-08-03-05 Air outlet molar flowrate
alpha1 = Workbooks(sfil1 & ".xlsm").Worksheets("Sheet2").Range("B10")
If alpha1 <= 1 Then
'Air outlet molar flowrate lower bounded - cannot be less than F_nai
Workbooks(sfil1 & ".xlsm").Worksheets("Sheet1").Range("N" & ii + 1).Formula = _
"=IF(Sheet1!Z" & ii + 1 & 
& ii + 1 & ">P" & ii + 1 & ",P" _
& ii + 1 & ",AF" & ii + 1 & "+AG" & ii + 1 & ")")"
Else
'No lower bound on air outlet molar flowrate
Workbooks(sfil1 & ".xlsm").Worksheets("Sheet1").Range("N" & ii + 1).Formula = _
"=IF(Sheet1!Z" & ii + 1 & 
& ii + 1 & ">P" & ii + 1 & ",P" & ii + 1 & ",AF" & ii + 1 & "+AG" & ii + 1 & ")")"
End If

'1-08-03-06 Fitted tracer inlet molar flowrate
If exp1 = 1 Or exp1 = 4 Then
Workbooks(sfil1 & ".xlsm").Worksheets("Sheet1").Range("O" & ii + 1).Formula = _
"=IF(Sheet1!Z" & ii + 1 & 
& dt & ",Sheet2!$F$14,IF(Sheet1!Z" & ii + 1 & ">Sheet2!$B$4+Sheet2!$F$17,0,INDEX(Sheet3!$D$2:$D$1000,MATCH(Sheet1!Z" & ii + 1 & ",Sheet3!$C$2:$C$1000))")"
Else
Workbooks(sfil1 & ".xlsm").Worksheets("Sheet1").Range("O" & ii + 1).Formula = _
"=IF(Sheet1!Z" & ii + 1 & 
& dt & ",Sheet2!$F$14,IF(Sheet1!Z" & ii + 1 & ")")"
End If

'1-08-03-07 Total outlet molar flowrate
Workbooks(sfil1 & ".xlsm").Worksheets("Sheet1").Range("P" & ii + 1).Formula = _
"=IF(Sheet2!$B$17+Sheet1!O" & ii + 1 & "- Sheet2!$B$18/8.314/293*Sheet1!H" & ii + 1 & "- Sheet2!$B$17+Sheet1!O" & ii + 1 & ")")

'1-08-03-08 Tracer outlet molar flowrate
'Calculates tracer outlet flowrate F_nheo - without condition that F_nheo >= F_nhei at all times
Workbooks(sfil1 & ".xlsm") .Worksheets("Sheet1") .Range ("Q" & ii + 1).Formula = _
"=IF(Sheet1!P" & ii + 1 & "+Sheet1!N" & ii + 1 & ">0,0,Sheet1!P" & ii + 1 & "\/Sheet1!P" & ii + 1 & "")"

'1-08-03-09 Tracer outlet mole fraction
'x=F_nheo/F_no
Workbooks(sfil1 & ".xlsm") .Worksheets("Sheet1") .Range ("R" & ii + 1).Formula = _
"=IF(Sheet1!Q" & ii + 1 & "+ Sheet1!Q" & ii + 1 & ">0,0,Sheet1!Q" & ii + 1 & ")")

'1-08-03-10 Gas mixture molecular weight
Workbooks(sfil1 & ".xlsm") .Worksheets("Sheet1") .Range ("S" & ii + 1).Formula = ":=Sheet1!R" & ii + 1 & _
"*0.004008+(1-Sheet1!R" & ii + 1 & ")*0.02897"

'1-08-03-11 Fitted separator pressure drop
'If exp1 = 1 Or exp1 = 4 Then
Workbooks(sfil1 & ".xlsm") .Worksheets("Sheet1") .Range ("T" & ii + 1).Formula = _
"=IF(Sheet1!Z" & ii + 1 & "<Sheet2!$F$3,Sheet2!$F$1*Sheet2!$B$17^gamma1 & *0.02897*(8.314*293/Sheet1!J" & ii + 1 & ")^gamma1 - 1 & "+Sheet2!$B$16*(Sheet1!N" & ii + 1 & _
"*0.02897)^gamma1 & gamma3 & "+Sheet1!Q" & ii + 1 & ")"

Else
Workbooks(sfil1 & ".xlsm") .Worksheets("Sheet1") .Range ("T" & ii + 1).Formula = _
"=Sheet2!$F$1*Sheet1!AG" & ii + 1 & "^gamma1 & gamma3 & "+Sheet1!Q" & ii + 1 & ")"

End If
End If

'1-08-03-11 Air outlet transient and steady-state molar flowrate initialization
Workbooks(sfil1 & ".xlsm”).Worksheets("Sheet1").Range("AF" & ii + 1) = 0
Workbooks(sfil1 & ".xlsm”).Worksheets("Sheet1").Range("AG" & ii + 1).Formula = "=Sheet2!$B$17"

Next ii

'1-08-04 Worksheet iterations
With Application
  .Calculation = xlCalculationAutomatic
  .Iteration = True
  .MaxIterations = 20
  .MaxChange = 0.0000000001
End With
With Application
  .Calculation = xlCalculationManual
  .Iteration = False
End With

'1-08-05 Recalculates air transient molar flowrate
For ii = 1 To mm * dt / dt1
  '1-08-05-01 Air outlet transient molar flowrate
  Workbooks(sfil1 & ".xlsm”).Worksheets("Sheet1").Range("AF" & ii + 1).Formula = 
    "=IF(Sheet1!Z" & ii + 1 & 
  "<Sheet2!B$4,0,IF(Sheet1!S" & ii + 1 & 
  "+" & c_5 & "*N" & ii + 1 & 
  "+" & c_6 & ")*" & F_ms & "^" & gamma4
End If

  '1-08-05-02 Air outlet steady-state molar flowrate
  Workbooks(sfil1 & ".xlsm”).Worksheets("Sheet1").Range("AG" & ii + 1).Formula = 
    "=IF(Sheet1!Z" & ii + 1 & 
  "<Sheet2!B$4,Sheet2!B$17,IF(Sheet1!S" & ii + 1 & 
  "+" & c_5 & "*N" & ii + 1 & 
  "+" & c_6 & ")*" & F_ms & "^" & gamma4
End If

'1-08-05-03 Air outlet steady-state molar flowrate
With Application
  .Calculation = xlCalculationAutomatic
  .Iteration = False
  .MaxChange = 0.0000000001
End With
With Application
  .Calculation = xlCalculationManual
  .Iteration = True
End With
'1-08-06 Worksheet iterations
With Application
    .Calculation = xlCalculationAutomatic
    .Iteration = True
    .MaxIterations = 20
    .MaxChange = 0.0000000001
End With
With Application
    .Calculation = xlCalculationManual
    .Iteration = False
End With

'1-09 ADJUST C1_ss TO GET CORRECT SUPERFICIAL GAS MOLAR FLOWRATE

'1-09-01 Loads nominal gas molar flowrate
F_ng = Workbooks("GAS COMPOSITION MODEL ROUTINE.xlsm").Worksheets("Sheet1").Range("M" & i + 4)
Workbooks(sfil1 & ".xlsm").Worksheets("Sheet2").Range("E23") = "F_nai nom"
Workbooks(sfil1 & ".xlsm").Worksheets("Sheet2").Range("F23") = F_ng

'1-09-02 Runs Solver routine to adjust superficial gas molar flowrate
Workbooks(sfil1 & ".xlsm").Worksheets("Sheet2").Range("F24").Formula = 
"=SUMXMY2(B17,F23)"
Workbooks(sfil1 & ".xlsm").Worksheets("Sheet2").Activate
SolverReset
SolverOptions maxtime:=300, iterations:=50, precision:=1E-20,
convergence:=1E-20
solveradd cellref:="$F$1", relation:=1, formulatext:="4e9"
solveradd cellref:="$F$1", relation:=3, formulatext:="1e7"
SolverOk SetCell:="$F$24", MaxMinVal:=2, ValueOf:=0,
ByChange:="$F$1"
With Application
    .Calculation = xlCalculationAutomatic
    .Iteration = True
    .MaxIterations = 20
    .MaxChange = 0.0000000001
End With
SolverSolve userfinish:=True
With Application
    .Calculation = xlCalculationManual
    .Iteration = False
End With

'1-10 ADJUST C_2 AND a_dpshe TO FIT SEPARATOR PRESSURE DROP (AND END OF DOWNSTEP IF TRACER INJECTION TYPE IS HELIUM --> OFF)
'1-10-01 Prints headers and calculates optimization equations
Workbooks(sfil1 & ".xlsm").Worksheets("Sheet2").Range("F21") = a_dpsphe
Workbooks(sfil1 & ".xlsm").Worksheets("Sheet2").Range("A22") = "S_DPsc2"
Workbooks(sfil1 & ".xlsm").Worksheets("Sheet2").Range("B22").Formula = 
"=SUMXMY2(Sheet1!F2:Sheet1!F" & mm * dt / dt1 + 1 _ 
& ",Sheet1!T2:Sheet1!T" & mm * dt / dt1 + 1 & ")"
Workbooks(sfil1 & ".xlsm").Worksheets("Sheet2").Range("E4") = "S_F2c2"
Workbooks(sfil1 & ".xlsm").Worksheets("Sheet2").Range("F4").Formula = 
"=SUMPRODUCT(Sheet1!U" & t2 / dt1 + 1 _ 
& ":Sheet1!U" & mm * dt / dt1 + 1 & ",Sheet1!U" & t2 / dt1 
+ 1 & ":Sheet1!U" & mm * dt / dt1 + 1 & ")"
Workbooks(sfil1 & ".xlsm").Worksheets("Sheet2").Range("E5") = "S_DPsF2c2"
Workbooks(sfil1 & ".xlsm").Worksheets("Sheet2").Range("F5").Formula = 
"=Sheet2!B22+Sheet2!F4^(ABS(1-Sheet2!F4))"
Workbooks(sfil1 & ".xlsm").Worksheets("Sheet2").Activate
SolverReset
SolverOptions maxtime:=300, iterations:=50, convergence:=100

'1-10-02 Sets Solver constraints to optimize c_2
solveradd cellref:="$B$15", relation:=3, formulatext:="1e4"
solveradd cellref:="$B$15", relation:=1, formulatext:="4e6"
SolverOk SetCell:="$B$22", MaxMinVal:=2, ValueOf:="0",
ByChange:="$B$15"

'1-10-03 Worksheet iterations
With Application
  .Calculation = xlCalculationAutomatic
  .Iteration = True
  .MaxIterations = 20
  .MaxChange = 0.0000000001
End With

'1-10-04 Recalculate iterative signals if there is an error
If WorksheetFunction.IsError(Workbooks(sfil1 & ".xlsm").Worksheets("Sheet2").Range("B22")) = True Then
  With Application
    .Calculation = xlCalculationManual
    .Iteration = True
  End With
End If
Workbooks(sfil1 & ".xlsm").Worksheets("Sheet1").Activate
Workbooks(sfil1 & ".xlsm").Worksheets("Sheet1").Range("J2:U2").Select
Selection.AutoFill Destination:=Range("J2:U1001")
Selection.AutoFill Destination:=Range("AF2:AG2")
Application.Calculate
Workbooks(sfil1 & ".xlsm").Worksheets("Sheet2").Activate
End If

'1-10-05 Runs Solver to optimize c_2
SolverSolve userfinish:=True
With Application
  .Calculation = xlCalculationManual
  .Iteration = False
End With

'1-11 OPTIMIZE TRACER MOLE BALANCE

'1-11-01 Codes and calculates tracer mole balance optimization function
Workbooks(sfil1 & ".xlsm").Worksheets("Sheet2").Range("E10") = "n_hei+n_hevol"
Workbooks(sfil1 & ".xlsm").Worksheets("Sheet2").Range("F10").Formula = 
"=SUM(INDEX(Sheet1!O2:O" & mm * dt / dt1 + 1 & _
  ",Sheet2!B4/" & dt1 & ":INDEX(Sheet1!O2:O" & mm * dt / dt1
  + 1 & "," & mm * dt / dt1 & "))*" & dt1 & _
  "+Sheet2!B12*Sheet2!B18/8.314/293*Sheet2!B19"
Workbooks(sfil1 & ".xlsm").Worksheets("Sheet2").Range("E11") = "n_heo"
Workbooks(sfil1 & ".xlsm").Worksheets("Sheet2").Range("F11").Formula = 
"=SUM(INDEX(Sheet1!Q2:Q" & mm * dt / dt1 + 1 & _
  ",Sheet2!B4/" & dt1 & ":INDEX(Sheet1!Q2:Q" & mm * dt / dt1
  + 1 & "," & mm * dt / dt1 & "))*" & dt1
Workbooks(sfil1 & ".xlsm").Worksheets("Sheet2").Range("E9") = "S%_nhei"
Workbooks(sfil1 & ".xlsm").Worksheets("Sheet2").Range("F9").Formula = "=(F10-
  F11)/F10*100"
With Application
  .Calculation = xlCalculationAutomatic
  .Iteration = True
  .MaxIterations = 20
  .MaxChange = 0.0000000001
End With
With Application
  .Calculation = xlCalculationManual
  .Iteration = False
End With

'1-11-02 Restarts tracer mole balance optimization if solution has not converged in a previous attempt
go13 = 0
13
If go13 > 0 Then
  Workbooks(sfil1 & ".xlsm").Worksheets("Sheet2").Range("F6")
= C_1tr
  With Application
    .Calculation = xlCalculationAutomatic
    .Iteration = True
    .MaxIterations = 20
    .MaxChange = 0.0000000001
  End With
With Application
  .Calculation = xlCalculationManual
  .Iteration = False
End With

End If

'1-11-03 Adjusts tracer mole balance optimization function if net balance cannot be achieved within 1%
If WorksheetFunction.IsError(Workbooks(sfil1 & ".xlsm").Worksheets("Sheet2").Range("B22")) = True Then
  Workbooks(sfil1 & ".xlsm").Worksheets("Sheet1").Activate
  Workbooks(sfil1 & ".xlsm").Worksheets("Sheet1").Range("J2:U2").Select
  Selection.AutoFill Destination:=Range("J2:U1001")
  Workbooks(sfil1 & ".xlsm").Worksheets("Sheet1").Range("AF2:AG2").Select
  Selection.AutoFill Destination:=Range("AF2:AG1001")
  Application.Calculate
  Workbooks(sfil1 & ".xlsm").Worksheets("Sheet2").Activate
End If
S1 = Workbooks(sfil1 & ".xlsm").Worksheets("Sheet2").Range("F9")
If S1 > 1 Then
  Workbooks(sfil1 & ".xlsm").Worksheets("Sheet2").Range("F9").Select
  Selection.Copy
  Range("F8").Select
  Workbooks(sfil1 & ".xlsm").Worksheets("Sheet2").Range("G8").Formula = "=(F9-F8)/F8*100"
  Application.Calculate
End If

'1-11-04 Optimizes tracer mole balance manually by stepping through values of C_ltr
With Application
  .Calculation = xlCalculationAutomatic
  .Iteration = True
  .MaxIterations = 20
  .MaxChange = 0.0000000001
End With
If S1 < 0.99 Then
  C_ltr1 = Workbooks(sfil1 & ".xlsm").Worksheets("Sheet2").Range("F1")
  Workbooks(sfil1 & ".xlsm").Worksheets("Sheet2").Range("F6")
  = C_ltr1
  S2 = Workbooks(sfil1 & ".xlsm").Worksheets("Sheet2").Range("F9")
  C_ltr2 = C_ltr
  Workbooks(sfil1 & ".xlsm").Worksheets("Sheet2").Range("F6")
  = C_ltr2
  C_ltr3 = C_ltr2 - C_ltr1
  frac = 0.5
  ii = 0
Do While S1 > 1 Or S1 < 0.99
C_ltr3 = C_ltr2 - C_ltr1
If S1 < 0.99 Then
    C_ltr2 = C_ltr2 - frac * C_ltr3
    Workbooks(sfil1 & ".xlsm").Worksheets("Sheet2").Range("F6") = C_ltr2
If Workbooks(sfil1 & ".xlsm").Worksheets("Sheet2").Range("F9") > 1 Then
    C_ltr1 = C_ltr2
    C_ltr2 = C_ltr2 + frac * C_ltr3
    C_ltr3 = C_ltr2 - C_ltr1
End If
ElseIf S1 > 1 Then
    C_ltr2 = C_ltr2 + frac * C_ltr3
    Workbooks(sfil1 & ".xlsm").Worksheets("Sheet2").Range("F6") = C_ltr2
    If Workbooks(sfil1 & ".xlsm").Worksheets("Sheet2").Range("F9") > 1 Then
        C_ltr1 = C_ltr2
        C_ltr2 = C_ltr2 + frac * C_ltr3
        C_ltr3 = C_ltr2 - C_ltr1
    End If
Else: Exit Do
End If
End If
S1 = Workbooks(sfil1 & ".xlsm").Worksheets("Sheet2").Range("F9")
i = i + 1
Workbooks(sfil1 & ".xlsm").Worksheets("Sheet2").Range("G6") = ii
Loop
Else
    C_ltr1 = Workbooks(sfil1 & ".xlsm").Worksheets("Sheet2").Range("F1")
    Workbooks(sfil1 & ".xlsm").Worksheets("Sheet2").Range("F6") = C_ltr1
    S2 = Workbooks(sfil1 & ".xlsm").Worksheets("Sheet2").Range("G8")
    C_ltr2 = C_ltr
    Workbooks(sfil1 & ".xlsm").Worksheets("Sheet2").Range("F6") = C_ltr1
    C_ltr3 = C_ltr2 - C_ltr1
    frac = 0.1
    i = 0
    Do While S1 > 1 Or S1 < 0.99
    C_ltr3 = C_ltr2 - C_ltr1
    If S1 < 0.99 Then
        C_ltr3 = C_ltr2 - C_ltr1
        C_ltr2 = C_ltr2 - frac * C_ltr3
        Workbooks(sfil1 & ".xlsm").Worksheets("Sheet2").Range("F6") = C_ltr2
    If Workbooks(sfil1 & ".xlsm").Worksheets("Sheet2").Range("G8") > 1 Then
        C_ltr1 = C_ltr2
        C_ltr2 = C_ltr2 + frac * C_ltr3
        C_ltr3 = C_ltr2 - C_ltr1
    End If
ElseIf S1 > 1 Then


\[ C_{\text{l}tr3} = C_{\text{l}tr2} - C_{\text{l}tr1} \]
\[ C_{\text{l}tr2} = C_{\text{l}tr2} + \text{frac} \times C_{\text{l}tr3} \]

```vba
Workbooks(sfil1 & ".xlsm") .Worksheets("Sheet2") .Range("F6") = C_{\text{l}tr2}
If Workbooks(sfil1 & ".xlsm") .Worksheets("Sheet2") .Range("G8") > 1 Then
    C_{\text{l}tr1} = C_{\text{l}tr2}
    C_{\text{l}tr2} = C_{\text{l}tr2} + \text{frac} \times C_{\text{l}tr3}
    C_{\text{l}tr3} = C_{\text{l}tr2} - C_{\text{l}tr1}
End If
Else: Exit Do
End If
S1 = Workbooks(sfil1 & ".xlsm") .Worksheets("Sheet2") .Range("G8")
ii = ii + 1
Workbooks(sfil1 & ".xlsm") .Worksheets("Sheet2") .Range("G6") = ii
Loop
End If
With Application
  .Calculation = xlCalculationManual
  .Iteration = False
End With
```

'1-12 CALCULATE RTD

'1-12-01 Initialize RTD fitting parameters and optimization functions
Workbooks(sfil1 & ".xlsm") .Worksheets("Sheet2") .Range("A24") = "E(t) PARAMETERS"
Workbooks(sfil1 & ".xlsm") .Worksheets("Sheet2") .Range("A25") = "Peak->"
Workbooks(sfil1 & ".xlsm") .Worksheets("Sheet2") .Range("A26") = "a"
Workbooks(sfil1 & ".xlsm") .Worksheets("Sheet2") .Range("A27") = "b"
Workbooks(sfil1 & ".xlsm") .Worksheets("Sheet2") .Range("A28") = "c"
Workbooks(sfil1 & ".xlsm") .Worksheets("Sheet2") .Range("A29") = "alpha"
Workbooks(sfil1 & ".xlsm") .Worksheets("Sheet2") .Range("B25") = "1"
Workbooks(sfil1 & ".xlsm") .Worksheets("Sheet2") .Range("C25") = "2"
Workbooks(sfil1 & ".xlsm") .Worksheets("Sheet2") .Range("B26") = 50
a2 = Workbooks(sfil1 & ".xlsm") .Worksheets("Sheet2") .Range("B7")
b2 = Workbooks(sfil1 & ".xlsm") .Worksheets("Sheet2") .Range("B8")
a3 = Workbooks(sfil1 & ".xlsm") .Worksheets("Sheet2") .Range("C7")
b3 = Workbooks(sfil1 & ".xlsm") .Worksheets("Sheet2") .Range("C8")
Workbooks(sfil1 & ".xlsm") .Worksheets("Sheet2") .Range("C26") = a2
'1-12-02 codes pulse E(t) and downstep F(t) signals in spreadsheet

Workbooks(sfil1 & ".xlsm").Worksheets("Sheet1").Range("V1") = "E(t)"
Workbooks(sfil1 & ".xlsm").Worksheets("Sheet1").Range("W1") = "F(t) conv"
Workbooks(sfil1 & ".xlsm").Worksheets("Sheet1").Range("X1") = "F_nhei norm"

For ii = 1 To mm * dt / dt1
  'Dirac plus 2 pulse peaks
  Workbooks(sfil1 & ".xlsm").Worksheets("Sheet2").Range("B26") = 1 / dt1
  Workbooks(sfil1 & ".xlsm").Worksheets("Sheet1").Range("V" & ii + 1).Formula = 
    "=Sheet2!$B$29*if(and(or(" & dt1 / dt * (ii - 1 / 2) & "=Sheet2!$B$27/" & dt & "," & dt1 / dt * (ii - 1 / 2) & "," & dt & ") & ">Sheet2!$B$27/" & dt & ")) & dt1 / dt * (ii - 3 / 2) & "<Sheet2!$B$27/" & dt & ")) & 
    "+Sheet2!$C$29*PERSONAL.xlsb!pulsepeak(Sheet2!$C$26,Sheet2!$C$27,Sheet2 !$C$28,"_ & "INDEX(Sheet1!$A$2:Sheet1!$A$" & mm + 1 & "," & mm + 1 & ")") & 
    "+Sheet2!$D$29*PERSONAL.xlsb!pulsepeak(Sheet2!$D$26,Sheet2!$D$27,Sheet2 !$D$28,"_ & "INDEX(Sheet1!$A$2:Sheet1!$A$" & mm + 1 & "," & mm + 1 & ")") & 
  'Downstep F(t)
  Workbooks(sfil1 & ".xlsm").Worksheets("Sheet1").Range("AH" & ii + 1) = dt1
  Workbooks(sfil1 & ".xlsm").Worksheets("Sheet1").Range("W" & ii + 1).Formula = 
    "=1-SUMPRODUCT(Sheet1!V$2:V" & ii + 1 & ",Sheet1!AH$2:AH" & ii + 1 & ")/Sheet2!$B$30"
  Workbooks(sfil1 & ".xlsm").Worksheets("Sheet1").Range("X" & ii + 1).Formula = 
    "=(Sheet1!O" & ii + 1 & ")-AVERAGE(Sheet1!O" & t2 / dt1 + 2 & ":O" & mm * dt / dt1 + 1 & ")/(AVERAGE(Sheet1!O2:O" & t1 / dt1 + 1 & ")-AVERAGE(Sheet1!O" & t2 / dt1 + 2 & ":O" & mm * dt / dt1 + 1 & "))"

Next ii

'1-12-03 codes convolution F_nheo(t) = F_nhei(t) * E(t) in spreadsheet
Workbooks(sfill & ".xlsm").Worksheets("Sheet1").Range("Y1") = "CONV(E*F_nhei)"
Workbooks(sfill & ".xlsm").Worksheets("Sheet1").Activate
For ii = 1 To n_step - 1
    Workbooks(sfill & ".xlsm").Worksheets("Sheet1").Range("Y" & ii + 1).Formula = ":=Sheet1!Q" & ii + 1
Next ii
Workbooks(sfill & ".xlsm").Worksheets("Sheet1").Range("Y" & n_step & ":Y" & mm * dt / dt1 + 1).Select
    Selection.FormulaArray = ":=PERSONAL.xlsb!convolution(Sheet1!V" & n_step & ":O" & mm * dt / dt1 + 1 & "," & dt1 & "," & dt1 & ",0)"
Application.Calculate

'1-12-04 Calculates mean residence time from sampled E(t) and time signals
Workbooks(sfill & ".xlsm").Worksheets("Sheet2").Range("A33") = "tau"
Workbooks(sfill & ".xlsm").Worksheets("Sheet2").Range("B33").FormulaArray = ":=SUMPRODUCT(Sheet1!$Z2:Sheet1!$Z" & mm * dt / dt1 + 1 & ",ABS(Sheet1!$V2:Sheet1!$V" & mm * dt / dt1 + 1 & ")/SUM(ABS(Sheet1!$V2:Sheet1!$V" & mm * dt / dt1 + 1 & "))-Sheet2!$B$4"

'1-12-05 Calculates nominal residence time
Workbooks(sfill & ".xlsm").Worksheets("Sheet2").Range("C33").Formula = ":=(101500+Sheet2!$B$13)*Sheet2!$B$18/Sheet2!$B$21/8.314/293"

'1-12-06 Codes RTD objective optimization function S in spreadsheet
'Residual between experimental and convolved tracer outlet molar flowrate only
Workbooks(sfill & ".xlsm").Worksheets("Sheet2").Range("A31") = "S_tot"
Workbooks(sfill & ".xlsm").Worksheets("Sheet2").Range("B31").Formula = ":=SUMXMY2(Sheet1!$Q$2:Sheet1!$Q$" & mm * dt / dt1 + 1 & ",Sheet1!$Y$2:Sheet1!$Y$" & mm * dt / dt1 + 1 & ")"  
't_tail = t_step + WorksheetFunction.Max(0.1, Workbooks(sfill & ".xlsm").Worksheets("Sheet2").Range("F17"))
Workbooks(sfill & ".xlsm").Worksheets("Sheet2").Range("B31").Formula = ":=SUMXMY2(Sheet1!$Q$2:Sheet1!$Q$" & mm * dt / dt1 + 1 & "Sheet1!$Y$2:Sheet1!$Y$" & mm * dt / dt1 + 1 & ")"
With Application
    .Calculation = xlCalculationAutomatic
    .Iteration = True
    .MaxIterations = 20
    .MaxChange = 0.0000000001
End With
With Application
  .Calculation = xlCalculationManual
  .Iteration = False
End With

'1-12-08 Codes for new module to be added to experiment spreadsheet with code for running a Solver routine
Set VBProj = Application.Workbooks(sfil1 & ".xlsm").VBProject
VBProj.References.AddFromFile solverdir & "\SOLVER.XLAM"
Set VBProj = Application.Workbooks("GAS COMPOSITION MODEL
ROUTINE.xlsm").VBProject
Application.ExecuteExcel4Macro
"PERSONAL.xlsb!AddModuleToProject("" & sfil1 & ","" & solverdir & ","")"

'1-12-09 Runs RTD Solver routine from module programmed into experimental spreadsheet
Workbooks(sfil1 & ".xlsm").Worksheets("Sheet2").Activate

'1-13 CALCULATE PULSE RESPONSE STATISTICAL MOMENTS AND PRINT RESULTS TO MASTER SHEET
Workbooks(sfil1 & ".xlsm").Worksheets("Sheet1").Range("AD1") =
"E(t) full"
For ii = 1 To mm
  'Dirac impulse plus 2 pulse peaks
  Workbooks(sfil1 & ".xlsm").Worksheets("Sheet1").Range("AD" & ii + 1).Formula =
  "=Sheet2!$B$29*if(and(or(sheet1!a" & ii + 1 & 
  ">=Sheet2!$B$27,sheet1!a" & ii + 1 & ">=Sheet2!$B$27)," _
  "& "sheet1!a" & ii + 1 & ")" _
  "& "+Sheet2!$C$29*PERSONAL.xlsb!pulsepeak(Sheet2!$C$26,Sheet2!$C$27,Sheet2
!$C$28,sheet1!a" & ii + 1 & ")" _
  "& "+Sheet2!$D$29*PERSONAL.xlsb!pulsepeak(Sheet2!$D$26,Sheet2!$D$27,Sheet2
!$D$28,sheet1!a" & ii + 1 & ")"
  Next ii
Workbooks(sfil1 & ".xlsm").Worksheets("Sheet2").Range("AD1") =
"tau"
Workbooks(sfil1 & ".xlsm").Worksheets("Sheet2").Range("B33").FormulaArray =
"=SUMPRODUCT(Sheet1!A2:Sheet1!A" & mm + 1 & 
  "& ",ABS(Sheet1!AD2:Sheet1!AD" & mm + 1 & 
  ")/SUM(ABS(Sheet1!AD2:Sheet1!AD" & mm + 1 & 
  ")-Sheet2!$B$4"
Workbooks(sfil1 & ".xlsm").Worksheets("Sheet1").Range("AE1") =
"res_tau"
For ii = 1 To mm
  Workbooks(sfil1 & ".xlsm").Worksheets("Sheet1").Range("AE" & ii + 1).Formula =
  "=(Sheet1!A" & ii + 1 & ")^2"
  Next ii
Workbooks(sfil1 & ".xlsm").Worksheets("Sheet2").Range("AD1") =
"SD"
Workbooks(sfil1 & ".xlsm").Worksheets("Sheet2").Range("B34").FormulaArray =
"=SQRT(SUMPRODUCT(Sheet1!AE2:Sheet1!AE" & mm + 1 & 
  "& ",ABS(Sheet1!AD2:Sheet1!AD" & mm + 1 & 
  ")/SUM(ABS(Sheet1!AD2:Sheet1!AD" & mm + 1 & 
  ")"))"
Workbooks("GAS COMPOSITION MODEL ROUTINE.xlsm").Worksheets("Sheet2").Range("A" & i) = sfill
Workbooks("GAS COMPOSITION MODEL ROUTINE.xlsm").Worksheets("Sheet2").Range("B" & i) = _
    Workbooks(sfill & ".xlsm").Worksheets("Sheet2").Range("B33")
Workbooks("GAS COMPOSITION MODEL ROUTINE.xlsm").Worksheets("Sheet2").Range("C" & i) = _
    Workbooks(sfill & ".xlsm").Worksheets("Sheet2").Range("B34")
Workbooks("GAS COMPOSITION MODEL ROUTINE.xlsm").Worksheets("Sheet2").Range("D" & i) = _
    Workbooks(sfill & ".xlsm").Worksheets("Sheet2").Range("F1")
Workbooks("GAS COMPOSITION MODEL ROUTINE.xlsm").Worksheets("Sheet2").Range("E" & i) = _
    Workbooks(sfill & ".xlsm").Worksheets("Sheet2").Range("F6")
Workbooks("GAS COMPOSITION MODEL ROUTINE.xlsm").Worksheets("Sheet2").Range("F" & i) = _
    Workbooks(sfill & ".xlsm").Worksheets("Sheet2").Range("B13")
Workbooks("GAS COMPOSITION MODEL ROUTINE.xlsm").Worksheets("Sheet2").Range("G" & i) = _
    Workbooks(sfill & ".xlsm").Worksheets("Sheet2").Range("F9")

'1-14 PRINT CHARTS

    '1-14-01 Separator pressure drop vs time chart
    Set DPS_chart = Workbooks(sfill & ".xlsm").Worksheets("Sheet1").ChartObjects.Add(Left:=1632, Width:=384, Top:=0, Height:=225)
    With DPS_chart
        .Chart.ChartType = xlXYScatterSmoothNoMarkers
        .Chart.HasLegend = True
        .Chart.HasTitle = True
        .Chart.ChartTitle.Text = "Separator Pressure Drop"
    End With
        .Name = Workbooks(sfill & ".xlsm").Worksheets("Sheet1").Range("E1")
        .Values = Workbooks(sfill & ".xlsm").Worksheets("Sheet1").Range("E2:E" & mm * dt / dt1)
        .XValues = Workbooks(sfill & ".xlsm").Worksheets("Sheet1").Range("Z2:Z" & mm * dt / dt1)
    End With
        .Name = Workbooks(sfill & ".xlsm").Worksheets("Sheet1").Range("G1")
        .Values = Workbooks(sfill & ".xlsm").Worksheets("Sheet1").Range("G2:G" & mm * dt / dt1)
        .XValues = Workbooks(sfill & ".xlsm").Worksheets("Sheet1").Range("Z2:Z" & mm * dt / dt1)
    End With
        .Name = Workbooks(sfill & ".xlsm").Worksheets("Sheet1").Range("T1")
        .Values = Workbooks(sfill & ".xlsm").Worksheets("Sheet1").Range("T2:T" & mm * dt / dt1)
        .XValues = Workbooks(sfill & ".xlsm").Worksheets("Sheet1").Range("Z2:Z" & mm * dt / dt1)
    End With
With DPs_chart.Chart.Axes(xlCategory)
    .MinimumScale = WorksheetFunction.MRound(t_step, 1) - 1
    .MaximumScale = WorksheetFunction.MRound(t_step, 1) + 3
End With

'1-14-02 Air and total outlet molar flowrates charts
Set Fno_chart = Workbooks(sfil1 & ".xlsm").Worksheets("Sheet1").ChartObjects.Add(Left:=1632, Width:=384, Top:=225, Height:=225)
With Fno_chart
    .Chart.ChartType = xlXYScatterSmoothNoMarkers
    .Chart.HasLegend = True
    .Chart.HasTitle = True
    .Chart.ChartTitle.Text = "Outlet Molar Flowrates"
End With
    .Name = Workbooks(sfil1 & ".xlsm").Worksheets("Sheet1").Range("P1")
    .Values = Workbooks(sfil1 & ".xlsm").Worksheets("Sheet1").Range("P2:P" & mm * dt / dt1)
    .XValues = Workbooks(sfil1 & ".xlsm").Worksheets("Sheet1").Range("Z2:Z" & mm * dt / dt1)
End With
    .Name = Workbooks(sfil1 & ".xlsm").Worksheets("Sheet1").Range("N1")
    .Values = Workbooks(sfil1 & ".xlsm").Worksheets("Sheet1").Range("N2:N" & mm * dt / dt1)
    .XValues = Workbooks(sfil1 & ".xlsm").Worksheets("Sheet1").Range("Z2:Z" & mm * dt / dt1)
End With
With Fno_chart.Chart.Axes(xlCategory)
    .MinimumScale = WorksheetFunction.MRound(t_step, 1) - 0.5
    .MaximumScale = WorksheetFunction.MRound(t_step, 1) + 3
End With

'1-14-03 Tracer inlet molar flowrate chart
Set Fnhei_chart = Workbooks(sfil1 & ".xlsm").Worksheets("Sheet1").ChartObjects.Add(Left:=1632, Width:=384, Top:=450, Height:=225)
With Fnhei_chart
    .Chart.ChartType = xlXYScatterSmoothNoMarkers
    .Chart.HasLegend = True
    .Chart.HasTitle = True
    .Chart.ChartTitle.Text = "Tracer Molar Flowrate"
End With
    .Name = Workbooks(sfil1 & ".xlsm").Worksheets("Sheet1").Range("AB1")
    .Values = Workbooks(sfil1 & ".xlsm").Worksheets("Sheet1").Range("AB2:AB" & mm * dt / dt1)
    .XValues = Workbooks(sfil1 & ".xlsm").Worksheets("Sheet1").Range("Z2:Z" & mm * dt / dt1)
End With
    .Name = Workbooks(sfil1 & ".xlsm").Worksheets("Sheet1").Range("O1")
.Values = Workbooks(sfil1 & ".xlsm" ).Worksheets("Sheet1" ).Range("Q2:O" & mm * dt / dt1)
.XValues = Workbooks(sfil1 & ".xlsm" ).Worksheets("Sheet1" ).Range("Z2:Z" & mm * dt / dt1)
End With
With Fnheo_chart.Chart.Axes(xlCategory)
 .MinimumScale = WorksheetFunction.MRound(t_step, dt1) - 0.1
 .MaximumScale = 9
End With

'1-14-04 Tracer outlet flowrate chart
Set Fnheo_chart = Workbooks(sfil1 & ".xlsm" ).Worksheets("Sheet1" ).ChartObjects.Add(Left:=1632, Width:=384,
Top:=675, Height:=225)
With Fnheo_chart
 .Chart.ChartType = xlXYScatterSmoothNoMarkers
 .Chart.HasLegend = True
 .Chart.HasTitle = True
 .Chart.ChartTitle.Text = "Tracer Molar Flowrate"
End With
 .Name = Workbooks(sfil1 & ".xlsm" ).Worksheets("Sheet1" ).Range("Q1")
 .Values = Workbooks(sfil1 & ".xlsm" ).Worksheets("Sheet1" ).Range("Q2:Q" & mm * dt / dt1)
 .XValues = Workbooks(sfil1 & ".xlsm" ).Worksheets("Sheet1" ).Range("Z2:Z" & mm * dt / dt1)
End With
 .Name = Workbooks(sfil1 & ".xlsm" ).Worksheets("Sheet1" ).Range("Y1")
 .Values = Workbooks(sfil1 & ".xlsm" ).Worksheets("Sheet1" ).Range("Y2:Y" & mm * dt / dt1)
 .XValues = Workbooks(sfil1 & ".xlsm" ).Worksheets("Sheet1" ).Range("Z2:Z" & mm * dt / dt1)
End With
With Fnheo_chart.Chart.Axes(xlCategory)
 .MinimumScale = WorksheetFunction.MRound(t_step, 1) - 0.5
 .MaximumScale = WorksheetFunction.MRound(t_step, 1) + 3
End With

'1-14-05 Tracer mole fraction chart
Set x_chart = Workbooks(sfil1 & ".xlsm" ).Worksheets("Sheet1" ).ChartObjects.Add(Left:=1632, Width:=384,
Top:=900, Height:=225)
With x_chart
 .Chart.ChartType = xlXYScatterSmoothNoMarkers
 .Chart.HasLegend = True
 .Chart.HasTitle = True
 .Chart.ChartTitle.Text = "Tracer Mole Fraction"
End With
With x_chart.Chart.SeriesCollection.NewSeries
 .Name = Workbooks(sfil1 & ".xlsm" ).Worksheets("Sheet1" ).Range("R1")
 .Values = Workbooks(sfil1 & ".xlsm" ).Worksheets("Sheet1" ).Range("R2:R" & mm * dt / dt1)
.XValues = Workbooks(sfil1 & 
".xlsm").Worksheets("Sheet1").Range("Z2:Z" & mm * dt / dt1)
End With
With x_chart.Chart.Axes(xlCategory)
    .MinimumScale = WorksheetFunction.MRound(t_step, 1) - 0.5
    .MaximumScale = WorksheetFunction.MRound(t_step, 1) + 3
End With

'1-14-06 Downstep response chart
Set F_chart = Workbooks(sfil1 & 
".xlsm").Worksheets("Sheet1").ChartObjects.Add(Left:=1632, Width:=384, 
Top:=1125, Height:=225)
With F_chart
    .Chart.ChartType = xlXYScatterSmoothNoMarkers
    .Chart.HasLegend = True
    .Chart.HasTitle = True
    .Chart.ChartTitle.Text = "Step Response"
End With
    .Name = Workbooks(sfil1 & 
".xlsm").Worksheets("Sheet1").Range("U1")
    .Values = Workbooks(sfil1 & 
".xlsm").Worksheets("Sheet1").Range("U2:U" & mm * dt / dt1)
    .XValues = Workbooks(sfil1 & 
".xlsm").Worksheets("Sheet1").Range("Z2:Z" & mm * dt / dt1)
End With
with F_chart.Chart.SeriesCollection.NewSeries
    .Name = Workbooks(sfil1 & 
".xlsm").Worksheets("Sheet1").Range("W1")
    .Values = Workbooks(sfil1 & 
".xlsm").Worksheets("Sheet1").Range("W2:W" & mm * dt / dt1)
    .XValues = Workbooks(sfil1 & 
".xlsm").Worksheets("Sheet1").Range("Z2:Z" & mm * dt / dt1)
End With
With F_chart.Chart.Axes(xlCategory)
    .MinimumScale = WorksheetFunction.MRound(t_step, 1) - 0.5
    .MaximumScale = WorksheetFunction.MRound(t_step, 1) + 3
End With

'1-14-07 Pulse response chart
Set E_chart = Workbooks(sfil1 & 
".xlsm").Worksheets("Sheet1").ChartObjects.Add(Left:=1632, Width:=384, 
Top:=1350, Height:=225)
With E_chart
    .Chart.ChartType = xlXYScatterSmoothNoMarkers
    .Chart.HasLegend = True
    .Chart.HasTitle = True
    .Chart.ChartTitle.Text = "Pulse Response"
End With
With E_chart.Chart.SeriesCollection.NewSeries
    .Name = Workbooks(sfil1 & 
".xlsm").Worksheets("Sheet1").Range("V1")
    .Values = Workbooks(sfil1 & 
".xlsm").Worksheets("Sheet1").Range("V2:V" & mm * dt / dt1)
    .XValues = Workbooks(sfil1 & 
".xlsm").Worksheets("Sheet1").Range("Z2:Z" & mm * dt / dt1)
End With
With E_chart.Chart.Axes(xlCategory)
  .MinimumScale = WorksheetFunction.MRound(t_step, 1) - 0.5
  .MaximumScale = WorksheetFunction.MRound(t_step, 1) + 3
End With

'1-15 SAVE AND CLOSE THE OPEN WORKBOOK
Workbooks(sfil).Close savechanges:=True
Workbooks(sfill & ".xlsm").Close savechanges:=True

'ON TO THE NEXT WORKBOOK
sfil = Dir
Loop

End Sub
Curriculum Vitae

Name: Martin Huard

Post-secondary Education and Degrees:

University of Saskatchewan
Saskatoon, Saskatchewan, Canada
2003-2008 B.E.

The University of Western Ontario
London, Ontario, Canada
2008-2009 M.E.Sc.

The University of Western Ontario
London, Ontario, Canada
2010-2015 Ph.D.

Honours and Awards:

Mitacs Accelerate Plus
Doctoral Fellowship
2010-2011

Natural Science and Engineering Research Council Alexander Graham Bell Canada Graduate Scholarship (CGS)
2011-2014

Western University Graduate Thesis Research Award
2013

Related Work Experience:

Graduate Research Assistant
The University of Western Ontario
2008-2015

Teaching Assistant
The University of Western Ontario
2010

Research Engineer
Agri-Therm Inc.
2010-2011

Research Engineer
Alberta Innovates – Technology Futures
2014-
Publications:


