Simultaneous particle agglomeration and attrition in a high temperature fluidized bed

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Graduate Program in Chemical and Biochemical Engineering
A thesis submitted in partial fulfillment of the requirements for the degree in Master of Engineering Science
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SIMULTANEOUS PARTICLE AGGLOMERATION AND ATTRITION IN A HIGH TEMPERATURE FLUIDIZED BED

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by

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Graduate Program

in

Department of Chemical and Biochemical Engineering

A thesis submitted in partial fulfillment of the requirements for the degree of Master of Engineering Science

The School of Graduate and Postdoctoral Studies
The University of Western Ontario
London, Ontario, Canada

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The thesis by

**Mithun Saha**

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is accepted in partial fulfillment of the requirements for the degree of
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Abstract

Fluid Coking™ is a major refining process to upgrade heavy crude oil or bitumen from oil sands. In Fluid Coking™ bitumen is atomized with steam and sprayed inside a high temperature fluidized bed of coke particles where it thermally cracks into smaller molecules. Fluid Coking™ is a carbon rejection process as solid carbon residues are formed during fragmentation into lighter hydrocarbon vapors. New coke deposits over existing coke particles and, consequently, the particle size increases. In addition, particles agglomeration occurs when the injected liquid does not disperse uniformly on individual hot coke particles but reaches, instead, local concentrations sufficiently large to act as a binder for several solid particles. Attrition nozzles are used to control the particle size to maintain the desired flow properties and reactor operability. In fact, fluidization quality degrades while there are too many particles smaller than 50 \( \mu \text{m} \) or larger than 600 \( \mu \text{m} \). Some of the large agglomerates may interact with the stripper sheds near at the bottom of Fluid Cokers™, resulting in their fouling and can cause premature reactor shut down.

The simultaneous particle agglomeration and attrition observed in Fluid Cokers™ has been simulated at elevated temperature in laboratory scale fluidized unit to study these processes and to develop and apply new technological solutions to reduce the generation of large agglomerates. An experimental technique has been developed to simultaneously perform the spray injection and attrition processes utilizing different nozzle designs and operating conditions, measuring, the initial and the final bed particle size distribution. Various interactions of spray and attrition jets have been tested and a novel liquid dispersion technique has been proposed and studied to reduce the production of large agglomerates.

The results show that, when an attrition jet hits at the base of a spray jet, the formation of large agglomerates is significantly reduced, but a large amount of unwanted fines is then generated. On the other hand, the application of satellite jets at the periphery of spray jets achieved better control of large agglomerates while minimizing fines generation.

Keywords: Fluid Coker™, Fluidized bed, High Temperature, Spray nozzle, Agglomeration, Attrition jet, Satellite jets
Co-Authorship

Chapter 2

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<td>Contributions:</td>
<td>Mithun Saha conducted all experimental work, data analysis and writing. Mehran Soleimani assisted with setting up the fluidized bed, also assisted in some of the experimental work. Cedric Briens and Franco Berruti provided guidance, supervision and revised drafts of the work.</td>
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Table of Contents

CERTIFICATE OF EXAMINATION ................................................................. ii
Abstract .................................................................................................................... iii
Co-Authorship ....................................................................................................... iv
Acknowledgements ............................................................................................... v
Table of Contents ................................................................................................. vi
List of Tables ......................................................................................................... ix
List of Figures ....................................................................................................... x

Chapter 1: Introduction ......................................................................................... 1
  1.1 Present thesis work .................................................................................... 1
  1.2 Canadian oil sands development and Fluid coking™ process .................. 1
  1.3 Particle agglomeration and attrition in fluidized beds- Literature Review .... 5
    1.3.1 Liquid injection and agglomeration ................................................ 5
    1.3.2 Gas jet and particle attrition ............................................................... 8
  1.4 Research objectives ................................................................................... 9
  1.5 References ................................................................................................. 10

Chapter 2: Study of simultaneous particle agglomeration and attrition in a high temperature fluidized bed ......................................................... 13
  2.1 Abstract ..................................................................................................... 13
  2.2 Introduction ............................................................................................... 14
  2.3 Experimental setup and methodology ..................................................... 15
  2.4 Results and discussions: .......................................................................... 21
    2.4.1 Impact of simultaneous spray and attrition jets on macro-agglomerates formation .......................................................... 21
    2.4.2 Impact of simultaneous spray and attrition jets on micro-agglomerates formation and the Sauter-mean diameter of bed particles ....................................................... 22
2.4.3 Impact of simultaneous spray and attrition jets on fines generation........23

2.5 Conclusions............................................................................................................28

2.6 Nomenclature .........................................................................................................28

2.7 References..............................................................................................................29

Chapter 3: Effect of different spray and attrition jet interaction in a high temperature fluidized bed.................................................................30

3.1 Abstract..................................................................................................................30

3.2 Introduction............................................................................................................30

3.3 Experimental setup and methodology....................................................................32

3.4 Results and discussion ...........................................................................................41

3.4.1 Impact of spray and attrition jets interaction on macro-agglomerates formation.................................................................43

3.4.2 Impact of spray and attrition jets interaction on micro-agglomerates formation........................................................................44

3.4.3 Impact of spray and attrition jets interaction on the Sauter-mean diameter of bed particles and on the production of fines...........................45

3.5 Conclusion .............................................................................................................53

3.6 Nomenclature .........................................................................................................53

3.7 References..............................................................................................................54

Chapter 4: Application of satellite jets with spray nozzle in a high temperature fluidized bed..................................................................................................................56

4.1 Abstract..................................................................................................................56

4.2 Introduction............................................................................................................56

4.3 Experimental setup and methodology....................................................................58

4.4 Results and discussion ...........................................................................................63

4.4.1 Impact of peripheral gas jets on macro-agglomerates formation..............64

4.4.2 Impact of peripheral gas jets on micro-agglomerates formation and the Sauter mean diameter of bed particles .................................................64
List of Tables

Table 3.1: Total cyclone catch mass (wt % of bed mass) for only N$_2$ gas injection through attrition and spray nozzles .......................................................................................................................... 41

Table 3.2: Nitrogen gas consumption rate (g/s) with spray and attrition nozzles .......... 42

Table 3.3: Superficial gas velocity in the upper bed region and the freeboard for various bed operating conditions ...................................................................................................................... 42

Table 4.1: Operating conditions with 1.2 mm spray nozzle and 0.13 mm satellite nozzles combination ................................................................................................................................................. 62

Table 4.2: Operating conditions with 2.7 mm spray nozzle and 0.4 mm satellite nozzles combination ................................................................................................................................................. 63
List of Figures

Figure 1. 1: Flow diagram of the Fluid Coking™ Process (House et. al., 2007) ........... 3

Figure 1. 2: Schematic of agglomeration process. (i) Wetting and nucleation, (ii) consolidation and coalescence and (iii) attrition and breakage (Adopted from Iveson et al., 2001) .................................................................................................................................................................................. 7

Figure 2. 1: Schematic of the high temperature fluidized bed with spray and attrition nozzles. (Modified from Feng, 2010) 16

Figure 2. 2: Spray nozzle setup for the fluidized bed ...................................................... 17

Figure 2. 3: Layout of the straight cylindrical spray nozzle (I.D. 2.7 mm and 3.6 mm) .. 17

Figure 2. 4: Typical convergent-divergent nozzle (Throat diameters: 1.7 and 2.4 mm). 18

Figure 2. 5: Attrition gas flow rate with attrition pressure .............................................. 18

Figure 2. 6: Total macro-agglomerate mass as a function of attrition gas flow rate for spray nozzle 2.7 mm ........................................................................................................................................................................... 23

Figure 2. 7: Total macro-agglomerate mass as a function of attrition gas flow rate for spray nozzle 3.6 mm ........................................................................................................................................................................... 24

Figure 2. 8: Samples of bed particles after simultaneous agglomeration and attrition at GLR 1.7% with 2.7 mm spray nozzle and attrition nozzle with 2.4 mm at 0.62 MPa (a) particles size 0.85 - 1.4 mm (b) particles size 1.4 - 2.36 mm ......................................................... 24

Figure 2. 9: Micro-agglomerate mass as a function of attrition gas flow rate for spray nozzle 2.7 mm ........................................................................................................................................................................... 25

Figure 2. 10: Micro-agglomerate mass as a function of attrition gas flow rate for spray nozzle 3.6 mm ......................................................................................................................................................................... 25
Figure 2. 11: Sauter-mean diameter of bed particles (45 µm < \(d_p\) < 0.85 mm) as a function of attrition gas flow rate for spray nozzle 2.7 mm................................. 26

Figure 2. 12: Sauter-mean diameter of bed particles (45 µm < \(d_p\) < 0.85 mm) as a function of attrition gas flow rate for spray nozzle 3.6 mm................................. 26

Figure 2. 13: Fines (\(d_p\) > 45 µm) mass as a function of attrition gas flow rate for spray nozzle 2.7 mm................................................................. 27

Figure 2. 14: Fines (\(d_p\) > 45 µm) mass as a function of attrition gas flow rate for spray nozzle 3.6 mm................................................................. 27

Figure 3. 1: Schematic of the high temperature fluidized bed with spray and attrition nozzle. (Modified from Feng, 2010) 34

Figure 3. 2: TEB spray nozzle (Throat diameter 2.7 mm)......................................................... 35

Figure 3. 3: Schematic of pre-mixer for 2.7 mm TEB nozzle .................................................... 35

Figure 3. 4: Spray nozzle setup for the fluidized bed ................................................................. 35

Figure 3. 5: Attrition nozzle (Throat diameter 1.7 mm)............................................................. 36

Figure 3. 6: Schematic of spray and attrition nozzle positions in fluidized bed................. 37

Figure 3. 7: Macro-agglomerate mass (wt%) as a function of GLR (wt%) for spray only condition ................................................................. 47

Figure 3. 8: Total macro-agglomerate mass reduced (wt %) from spray only condition as a function of attrition angle................................................................. 47

Figure 3. 9: Macro-agglomerate mass reduced from spray only condition for GLR 3.6% as a function of attrition angle ................................................................. 48

Figure 3. 10: Macro-agglomerate mass reduced from spray only condition for GLR 4.6% as a function of attrition angle ................................................................. 48
Figure 3.11: Macro-agglomerate mass reduced from spray only condition for GLR 5.5% as a function of attrition angle

Figure 3.12: Micro-agglomerate mass (wt% of total bed mass) as a function of GLR (wt%) for spray only condition, i.e. in the absence of attrition nozzles

Figure 3.13: Increase in the total mass of micro-agglomerates, relative to the increase observed in the absence of attrition nozzles

Figure 3.14: Total macro-agglomerate mass (wt %) vs. micro-agglomerate mass (wt %)

Figure 3.15: Reduction in the mass of fines and changes in the SMD of the bed particles (45 µm < d_p < 0.85 mm) as a function of GLR for spray only condition

Figure 3.16: SMD of bed particles (45 µm < d_p < 0.85 mm) change ratio as a function of attrition angle

Figure 3.17: Fines (d_p < 45 µm) mass generated as a function of attrition angle

Figure 3.18: Total macro-agglomerate mass (wt %) vs. SMD of bed particles (45 µm < d_p < 0.85 mm)

Figure 4.1: Schematic of the high temperature fluidized bed with spray nozzle. (Modified from Feng, 2010)

Figure 4.2: TEB spray nozzle used for experiments (Throat diameter 1.2 and 2.7 mm)

Figure 4.3: Satellite nozzles (0.13 mm) mounted with 1.2 mm TEB spray nozzle

Figure 4.4: Satellite nozzles (0.4 mm) mounted with 2.7 mm TEB spray nozzle

Figure 4.5: Total macro-agglomerate mass (wt %) as a function of total GLR (wt %) for various spray nozzles
Figure 4. 6: Macro-agglomerate mass reduction with various size cuts for combination of TEB 1.2 mm and Satellite 0.13 mm nozzles from macro-agglomerates mass with TEB 1.2 mm .................................................................................................................................... 66

Figure 4. 7: Macro-agglomerate mass reduction with various size cuts for combination of TEB nozzle 2.7 mm and Satellite 0.4 mm nozzles from macro-agglomerate mass with TEB 2.7 mm .................................................................................................................................... 67

Figure 4. 8: Micro-agglomerate mass (wt %) as a function of total GLR (wt %) for various spray nozzles .................................................................................................................................... 67

Figure 4. 9: Total macro-agglomerate mass (wt %) as a function of micro-agglomerate mass (wt %) for various spray nozzles .................................................................................................................................... 68

Figure 4. 10: SMD of bed particles (45 µm < d_p < 0.85 mm) as a function of total GLR (wt %) for various spray nozzle .................................................................................................................................... 68

Figure 4. 11: Total macro-agglomerate mass (wt%) vs. SMD of bed particles (45 µm < d_p < 0.85 mm) with various spray nozzles .................................................................................................................................... 69

Figure 4. 12: Fines (d_p < 45 µm) mass (wt %) as a function of total GLR (wt%) .................................................................................................................................... 69
Chapter 1: Introduction

1.1 Present thesis work

The research work presented in this thesis deals with experimental studies on particle agglomeration and attrition in a high temperature fluidized bed with gas-liquid injection. Fluidized beds are used for particulate operations in numerous industrial applications due to good solid and gas mixing. Better mixing of solid and gas promote better heat and mass distribution in a fluidized bed, which are critically important characteristic features required by the heavy crude oil upgrading process. The Fluid Coking™ process is used for upgrading heavy hydrocarbons streams in refineries and bitumen extracted from the oil sands. The heavy oil or bitumen are atomized with steam and sprayed inside a high temperature fluidized bed of circulating coke particles. When bitumen droplets come in contact with coke particles, a significant portion of droplets form agglomerates with coke particles. The motivation and objective of this thesis is to achieve a better understanding and control over the agglomerate formation process and, to improve the liquid feed distribution and mixing in Fluid Coking™ processes.

In this chapter, a brief description of Canadian oil sands development and of Fluid Coking™ process will be presented first. A review of recent studies on agglomeration formation and breakage will follow. This chapter concludes with the specific objectives of the research work.

1.2 Canadian oil sands development and Fluid coking™ process

Canada has a vast deposit of heavy oil in the province of Alberta. This heavy oil is a tar like petroleum crude available in the form of a mixture of water, sand, clay and bitumen which is typically referred to as “oil sand”. Most of this petroleum resource is located in the Athabasca region of Northeastern Alberta. The estimated reserve of Alberta oil sands is 1.8 trillion barrels and around 300 billion barrels can be recovered with current
technologies. With this amount of recoverable oil, Canada rank 2\textsuperscript{nd} in terms of available oil resources, just after Saudi Arabia (Government of Alberta, 2009). Oil sands production is different from conventional oil production. The oil deposits within about 100 meters from the surface are extracted by surface mining. For the deeper deposits, the technology applied is known as steam assisted gravity drainage (SAGD), where steam is injected to heat up the bitumen and reduce its viscosity so that it can be collected using horizontal wells and pumped to the surface. Bitumen is a black, thick like heavy crude oil and cannot be processed in conventional refining equipment. It requires a primary upgrading process to convert it into a so-called synthetic crude oil (SCO) before it can be further processed to produce the typical petroleum products of a refinery (waxes, naphtha, gasoil, gasoline, solvents, lubricants, and gaseous products).

Fluid Coking\textsuperscript{TM} is a thermal cracking process and use a non-catalytic chemical reaction to upgrade bitumen to light synthetic crude oil (Gray, 2002). In the thermal cracking process, long chain hydrocarbons break down to smaller chain hydrocarbons at elevated temperatures, and, in the process, reject solid carbon. Fluid Coking\textsuperscript{TM} is a continuous process developed by ExxonMobil Corporation. Currently, Fluid Coking\textsuperscript{TM} process is used by many refineries around the world to convert heavy oil and Fluid Catalytic Cracking (FCC) bottoms to lighter crude oil fractions. Syncrude Canada is the largest producer of synthetic oil from the thermal cracking of bitumen extracted from the oil sands using three of the largest Fluid Cokers\textsuperscript{TM} in the world. Syncrude has produced 107 million barrels or 300,000 barrel per day synthetic crude in the year of 2010. They have capacity to produce over 15 percent of Canadian crude oil demand (Syncrude Canada Ltd., 2011).
Figure 1.1: Flow diagram of the Fluid Coking™ Process (House et. al., 2007)

Figure 1.1 provides a schematic diagram of the Fluid Coking™ process. The Fluid Coker consists of two vessels, a reactor and a regenerator or burner. The reactor has three sections: a scrubber at the top, a reaction section in the middle, and a stripper at the bottom. Coke particles are heated to 600-800 °C in the fluidized bed burner by burning a fraction of the coke with air, and they are transported to upper section of reactor via a hot coke standpipe to bring the required heat to crack the feed bitumen. Feedstock bitumen, heated to around 350 °C and atomized with steam, is sprayed into a bubbling or turbulent fluidized bed of coke particles in the reaction section operating at 510-550 °C (House et al., 2007) using banks of nozzles located at different axial and radial positions. As the liquid droplets come in contact with coke particles, endothermic cracking reactions occur. The products of these reactions are a mixture of gas, light and, heavy oil vapors, and coke. The cracked gases and vapors pass through the scrubber section of reactor and are then directed to condensers where the synthetic crude oil is collected and separated from
the non-condensable gases. The cracking process generates gases and vapors with a higher hydrogen-to-carbon ratio than the original feed by rejecting solid carbon which is deposited over the existing coke particles that are circulating through the system. The coke particles grow in size, flow downwards to the stripper section, where entrapped light hydrocarbons are removed with steam, and are then transported to the burner for the subsequent re-heating cycle. The most desired Fluid Coking™ products are light and heavy gas oils which make up the sweet crude oil. Thus the objective of the Fluid Coking™ process is to maximize gas-oils production while minimizing lighter gases and coke. 

As described above, during the Fluid Coking™ process, the bed particles increase in size due to new coke formation. To maintain a continuous operation and achieve the desired mass flowrates between reactor and burner, the bed particle size needs to be within an optimal size range. If there are too many large particles over 600 µm, slugging, bed defluidization and plugging of the stripper section may occur (McMillan et al., 2007). On the other hand, with excessive fine particles below 50 µm, fluidized bed behavior becomes erratic (Dunlop et al., 1958). Therefore, the bed particle size is controlled by attrition created by jets of high pressure superheated steam from supersonic attrition nozzles located just above the stripper section. The attrition nozzles have a converging-diverging section, to achieve sonic speed at the throat and supersonic speed at the nozzle exit. High speed steam entrains and accelerates slow moving coke particles into the spray jet cavity, and slams them over dense bed particles near the jet tip, where some of them fracture into smaller fragments. These attrition nozzles consume a large percentage of the total steam used in Fluid Cokers™ and improvement of the attrition nozzle efficiency would be highly desirable in terms of considerable savings in steam consumption, in the ability to process more feed in the reactor, and in the consequent reduction in the amount of water which would need to be condensed and separated from the produced synthetic crude oil. It is expected that all these savings would be reflected in a significant enhancement of the economics and environmental friendliness of the Fluid Coking™ process.
In addition to normal particle growth during the coking process, poor feed injection and, consequently, poor liquid-solid contact, account for the formation and growth of larger and wet agglomerates which hinder fluidization quality as well as mass and heat transfer processes, leading to lower yields of desirable liquid products. In addition, these large agglomerates, being wet and rich in hydrocarbons, cause stripper fouling which is typically the main reason for premature reactor shutdowns. To minimize this problem, the reactor operating temperature is kept higher than optimal, causing higher vapor phase cracking of light and heavy gas-oils to lighter non-condensable gases resulting in lower yields of the more valuable condensable fractions. Therefore, a better solution would be to find ways to improve the liquid-solid contact during the interaction between feed jet and solid bed particles, in the reactor section, resulting in a lower amount of large agglomerates. Similarly, improvements in the effectiveness of the attrition nozzles would result in lower steam consumption, higher yields and enhanced reactor operability. These considerations represent the motivation for the research work reported in this thesis.

1.3 Particle agglomeration and attrition in fluidized beds- Literature Review

1.3.1 Liquid injection and agglomeration

The contact between injected liquid and solid particles in a fluidized bed was examined by Leclere et al. (2001) by measuring the vaporization rate of injected liquid feed. They developed a new model to predict the formation of agglomerates depending on the spray droplet size and bed particle size. Gray et al. (2002) indentified the key factors governing wet agglomerates behavior in a fluid coker™ as the Stokes number of the particles, the thickness of the liquid films and the diameter and surface roughness of the particles. They emphasized the importance of an efficient mechanism of spraying the feed on the surface of coke particles creating uniform thin films. The mechanism of agglomerate formation was then studied by Ariyapadi et al. (2003) using an X-ray imaging technique with the injection of non-evaporating radioactive liquid tracer in a fluidized bed; they reported that a significant amount of agglomerates forms at the end of the jet cavity. House et al. (2004) investigated the effect of different injection nozzle configurations on liquid-solid
contact. They reported that the initial contact of liquid droplets with solids can be improved by modifying the way the liquid feed jet interacts with fluidized particles. Bruhns et al. (2004) studied the liquid injection by spraying evaporating liquid in fluidized beds and found that agglomerates form at tip of some spray nozzles. They injected water at a bed temperature of 153°C and the temperature profile in the jet bed interaction region was monitored as a function of liquid evaporation rate. Ariyapadi et al. (2004) developed a model to determine the entrainment of solids and gas into the jet cavity formed by a spray nozzle in a fluidized bed. With injection of non-evaporating liquid using a conventional spray nozzle, McMillan et al. (2007) studied the mixing of liquid and solids. They found that the core of the jet cavity is a liquid rich region, while the annular region is richer of solid particles, and that enhancing the radial mixing along the jet cavity improves the quality of the liquid distribution. In fact, Chan et al. (2004) proposed the implementation of a draft tube mixer positioned coaxially downstream of the feed nozzles, which would enhance the radial mixing of liquid droplets and solid particles. This study derived from earlier findings by Hulet et al. (2003), who investigated the particle entrainment and the stability of the feed jet in a fluidized bed when using a draft tube downstream of the spray nozzle. Portoghese et al. (2007) developed a nozzle performance index measure the liquid-solid mixing characteristics of different liquid injection nozzles. House et al. (2007) reported that higher liquid-to-solid (L/S) ratios in the jet-bed interaction region lead to agglomeration and, consequently, to heat transfer limitations.

Since the contact of liquid feed and solid bed particles in fluidized beds results both in the desirable formation of distributed liquid films over individual mobile particles, and the undesirable formation of small and large particle agglomerates, it is important to better understand the phenomena involved during their formation.

Granulation or agglomeration is the process that binds particles together into larger granules in which the original particles can still be distinguishable. For wet agglomeration, a liquid binder is required. In agglomerates and granules, liquid can exists in three forms producing cohesive forces: - (a) mobile liquid bridges, (b) adsorbed liquid layers on particles, and (c) adhesive or viscous binders (Sherrington and Oliver, 1981).
The binder viscosity has been recognized as an important parameter in controlling the granulation behavior (Ennis, 1990).

Fundamentally there are three rate processes which determine wet granulation behavior (Iveson et al. 2001). Schematic diagrams of these processes are shown in Figure 1.2.

(i) Wetting & Nucleation

(ii) Consolidation & Coalescence

(iii) Attrition & Breakage

Figure 1.2: Schematic of agglomeration process. (i) Wetting and nucleation, (ii) consolidation and coalescence and (iii) attrition and breakage (Adopted from Iveson et al., 2001)

1. Wetting and nucleation: the liquid binder is bought into contact with a dry powder bed, and is distributed throughout the powder bed to form nuclei.
2. Consolidation and growth: collision between two granules, granules and dry powder, granules and bed wall lead to compaction and growth.
3. Attrition and breakage: wet or dry granules break due to impact, wear or compaction.
This thesis work mainly focused on the formation of agglomerates resulting from the simultaneous agglomeration and attrition created by various injection and attrition nozzle configurations and operating conditions.

1.3.2 Gas jet and particle attrition

There have been extensive studies over particle attrition in fluidized beds. Most of the research considered the turbulent jet theory developed by Abromovich (1963), De Michelle (1976) and Xuereb (1991). A high velocity jet within a fluidized bed creates a differential pressure between the formed low density jet cavity and the turbulent shear layers at the jet bed interface, which draws fluidization gas towards the jet. While the flow rate of gas is significantly high, it entrains solid particles inside the jet. Xuereb et al. (1991) and later Felli et al. (2002) observed that particles are entrained mostly near the nozzle tip. Ariyapadi et al. (2003) reported that particles size, density and gas properties will affect the entrainment rate of particles.

The purpose of the application of high velocity gas jets in fluidized beds is particle attrition. There are two modes of particle attrition: abrasion and fragmentation. With abrasion, very fine solid elements (irregularities) are removed from the particle surface, making the particles rounder, while keeping the particle size distribution nearly unchanged. On the contrary, with fragmentation, particles break into smaller pieces and the particle size distribution shifts, often to a bimodal distribution (Werther and Rappenhagen, 1999). Patel et al. (1986) classified the variables causing particle attrition in two categories: particle properties and fluidized bed environment. Particle properties encompass shape, surface roughness and strength. Fluidized bed properties include fluidization velocity, bed temperature and attrition pressure. Lin et al. (1980) investigated elutriation and attrition in fluidized bed and found that the attrition rate is related to the excess fluidization velocity ($U_g - U_{mf}$). They reported that, in the absence of jets, the majority of the attrition process occurs in the bubble wake due to excess fluidization velocity and presented a correlation to estimate the rate of fines generation. Bentham et al. (2004) investigated particle attrition in a single jet region of a fluidized bed and found
that the breakage mechanism works by entrainment of particles from the dense phase region surrounding of gas jet into the dilute jet cavity. The high speed gas accelerates the entrained particles and slams them over the denser bed of particles near the jet tip. They reported a correlation equation for attrition rate as a function of jet velocity.

In a Fluid Coker™, high velocity jet attrition has been achieved using De-Laval type convergent-divergent supersonic nozzles. McMillan et al. (2007) investigated the operating parameters of attrition nozzle on particle attrition and found that larger diameter nozzles operating at high gas flow rate with low density gases show a higher grinding efficiencies, where the grinding efficiency is defined as the ratio of the new particle surface that is created through attrition, to the mass of attrition gas used. Cruz et al. (2010) studied the attrition nozzle design and found that the divergent angle of the nozzle needs to be within the optimum range of 3° to 8°. With higher angles, shock waves occur in the divergent section, whereas, with lower angles, higher friction at the nozzle wall dissipates excessive energy. Li et al. (2011) studied the effects of bed temperature on the particle attrition rate and reported that higher grinding efficacies could be achieved at higher bed temperatures when using a high gas flow rate of low density attrition gas.

1.4 Research objectives

The overall goal of this research work was to understand the simultaneous particle agglomeration and attrition process occurring in a high temperature fluidized bed where liquid feed is injected and distributed over solid particles. The ultimately objective was to reduce the large agglomerate formation in the Fluid Coking™ process by identifying and applying novel concepts utilizing improved liquid and attrition gas injection devices and optimal operating parameters.

The specific objective of the first study reported in the thesis was the determination of the best operating parameters with simultaneous spray and attrition jets in a fluidized bed to minimize production of large agglomerates. Previous studies investigated spray and attrition nozzles separately: in particular, the possible impact of the attrition nozzles on
the break-up of wet agglomerates, before they solidify, was not investigated. Various spray and attrition nozzle sizes and operating parameters were examined. In this study, the spray and attrition nozzles did not interact directly. The generation of large agglomerates, the Sauter-mean diameter of bed particles, and the amount of fines have been measured to quantify the combined performance of the spray and attrition nozzles. To optimize the attrition gas consumption rate, various gas flow rates with various nozzle sizes have been examined. This study required the development of a new experimental system to simulate the agglomeration and attrition processes that occur in a Fluid Coker™.

The objective of the second study of this thesis was the investigation of different types of direct interaction between spray and attrition jets in a hot fluidized bed. An innovative approach was taken to compare the conventional separate (non-interacting) spray and attrition jets injection with attrition jets hitting the spray jet cavity at various locations.

The objective of the third study was to reduce large agglomerate formation by improving the liquid dispersion over the fluidized bed particles. A novel nozzle design used satellite pure gas nozzles positioned at the periphery of the feed spray jet. The satellite gas jets enhanced particle entrainment into the spray cavity and liquid-solid mixing within the spray jet cavity. The effect of this solution on agglomerate formation has been investigated.

1.5 References


Chapter 2: Study of simultaneous particle agglomeration and attrition in a high temperature fluidized bed

2.1 Abstract

In processes such as granulation, liquid is sprayed into a gas solid fluidized bed to bind particles together. In other processes, such as jet milling, gas is injected through supersonic nozzles into a fluidized bed to grind the fluidized particles and reduce their size. In Fluid Cokers™, which are used for heavy oil upgrading, agglomeration and attrition occur simultaneously. Investigation of these simultaneous processes was carried out in a hot fluid bed of coke particles, spraying a sucrose binder solution into the bed, simulating the heavy oil, and attriting the particles with a supersonic nitrogen gas jet. The size distribution of the bed solids was monitored during the process. The effect of the size and operating characteristics of the liquid spray and attrition nozzles on both agglomeration and attrition were analyzed, using the particle size distribution. The objective was to reduce the formation rate of large agglomerates in the bed. The liquid spray nozzle performed better at higher atomization gas to liquid ratios, resulting in a smaller production rate of large agglomerated material. Attrition nozzles that used a higher pressure gas performed better, generating more small particles for the same gas flow rate. Operating the spray and attrition nozzles simultaneously could be beneficial, as the attrition nozzles brake up some of wet agglomerates as they are generated by the spray nozzles.

Key words: Fluid Coking™, Agglomeration, Attrition, Spray nozzle, Fluidized bed, Jet milling
2.2 Introduction

The Fluid Coking™ process was invented and commercialized by ExxonMobil to crack heavy residual oil and bitumen, generating lighter, more valuable hydrocarbons and rejecting carbon as solid coke. In this process, hot bitumen is sprayed using steam atomized nozzles into a fluidized bed of hot coke particles. Droplets of bitumen crack upon contact with the surface of the coke particles at temperatures ranging from 510 to 550 °C (House et al., 2007).

The Fluid Coking™ process is a carbon rejection process, so new coke forms in the bed and deposits over existing coke particles. Coke particles are transferred from the reactor to a burner where a fraction of the accumulated coke is combusted to reheat the coke particle providing the heat for the thermal cracking process. The reheated coke particles are re-circulated to the reactor and come to contact with bitumen feed. Attrition nozzles must be used to prevent the particles from growing beyond the acceptable size range, degrading the reactor operation, as too many particles smaller than 50 µm or larger than 600 µm would deteriorate the fluidization quality (Li et al., 2011). Therefore a series of attrition nozzles, located in the lower part of the Fluid Coker™ reactors, inject pressurized steam inside the bed to break the coke particles before they are stripped from light hydrocarbons and returned to the burner section for reheating. Convergent–divergent nozzles are used to supply steam at supersonic speed. McMillan et al. (2007) has reported that bed particles are entrained into the jet formed by these nozzles, accelerate to high speed and slam on slow moving bed particles near the jet tip, causing particle breakage. One of the important aspects of the convergent-divergent nozzle configuration is the lower consumption of steam compared to regular nozzle (McMillan et al., 2007).

To maximize the yield of valuable liquid products, thermal cracking must occur rapidly. This means that the formation of coke agglomerates must be minimized, since they would limit heat and mass transfer (House et al. 2007). Agglomerates can also result in stripper fouling, leading to plugging and consequently premature reactor shutdown.
Gray et al. (2001) proposed a mechanism of dispersion for steam assisted bitumen injection, where a large number of liquid droplets contact a large number of coke particles, forming temporary agglomerates that are rapidly broken up by shear forces within the bed into individual coke particles covered by thin films of liquid. Chan et al. (1997) invented a more effective spray nozzle which produces smaller droplets of liquid. House et al. (2007) carried out experiments and modeling with gas atomized liquid spray nozzles and showed that smaller agglomerates with a lower liquid content result in a higher liquid yield and better Coker operability.

Previous studies investigated spray and attrition nozzles separately. In particular, the possible impact of the attrition nozzles on the break-up of wet agglomerates, before they solidify, was not investigated. The objective of this study was a better understanding of the commercial Fluid Coker™ operation, under more realistic conditions, by investigating the simultaneous spraying and attrition processes in an agglomerating system operating under simulated conditions and at high temperature.

2.3 Experimental setup and methodology

The simultaneous agglomeration and attrition experiments were conducted using a fluidized column of 1.23 m high, and with a 0.508 m x 0.114 m rectangular cross-section (Figure 2.1). The fluidized bed height was 0.45 m. Bed particles were petroleum coke (with a particle density of 1450 kg/m$^3$ and a Sauter mean diameter of 120 µm). The coke particles were fluidized with nitrogen gas introduced through a sparger distributor and heated to 250° C with an electric heater system immersed in the upper region of the bed, well above the spray and attrition nozzles. Binding liquid (30% sugar solution), with approximately the same viscosity (3 cP) as the bitumen injected in commercial cokers, was injected into the hot bed with a spray nozzle and atomized with nitrogen, at a height of 0.25 m above the gas distributor. Cold nitrogen gas was injected into the hot fluidized bed through an attrition nozzle at a height of 0.13 m above the gas distributor and 0.13 m below the spray nozzle. Two sampling ports, equipped with a screw system to extract the bed material, which were at two different heights (0.13 m and 0.25 m above the gas
distributor), were used to collect bed samples. The bed was equipped with three cyclones: solids collected by the 1st cyclone were recycled to the bed through a dip-leg, while solids collected by the second and the tertiary cyclones were recovered outside the bed.

Two straight tube feed spray nozzles were tested, with different inner diameters (2.7 and 3.6 mm). Figure 2.2 shows how the liquid was fed to the nozzle from a pressurized blow tank, through a hollow cylinder pre-mixer where it was mixed with the atomization gas. Pressure $P_1$ and $P_2$ were set with pressure regulators to achieve the required liquid flow rate and atomization Gas-to-Liquid Ratio (GLR). Figure 2.3 shows the simple, cylindrical geometry of the spray nozzles.

Figure 2. 1: Schematic of the high temperature fluidized bed with spray and attrition nozzles. (Modified from Feng, 2010)
Convergent-divergent nozzles (De-Laval nozzle) were used as attrition nozzles. Attrition nozzles of 2 different sizes (throat diameter: 1.7 and 2.4 mm) were used (Figure 2.4) to get a comprehensive view of the effect of nozzle size on attrition quality for two gas mass flow rates (200 and 320 g/min). In such convergent-divergent nozzles, sonic velocity is reached at the throat and supersonic velocity is attained in the divergent expansion (Smith et al. 2005). Gas mass flow rates 200 and 320 g/min were achieved at 0.52 and 0.62 MPa.
respectively with the 2.4 mm nozzle and at 1.48 and 2.17 MPa respectively with the 1.7 mm nozzle, as showed in Figure 2.5.

![Figure 2.4: Typical convergent-divergent nozzle (Throat diameters: 1.7 and 2.4 mm)](image)

A mass of 22 kg of sieved coke particles (with a Sauter mean diameter of 120 µm and a maximum size of 420 µm, with a minimum fluidization velocity of 0.007 m/s) were first introduced into the bed. The bed particles were then heated with an electric heater to a
target temperature of 250° C at a low fluidization velocity of 0.05 m/s \((U / U_{mf} = 7)\), to maximize heat transfer and minimize heat losses. Once the required bed temperature was reached, the bed was fluidized with a superficial velocity of 0.47 m/s \((U / U_{mf} = 67)\) to ensure that all entrainable particles could be removed from the bed and recovered from the secondary and tertiary cyclones. Therefore, any particle subsequently recovered from these two cyclones resulted from attrition and breakage. For the experiments conducted, the bed was then kept at a superficial velocity of 0.4 m/s \((U / U_{mf} = 57)\) and during the whole process, the superficial velocity was of 0.57 m/s \((U / U_{mf} = 81)\), due to the added contribution of attrition and atomization gas. To achieve simultaneous agglomeration and attrition, the attrition nozzle was kept open until all the injected liquid was dispersed inside the bed. 1200 gm of 30 % sugar solution were sprayed inside the bed over a fixed spray time of 6 minutes at liquid flow rate of 3.33 g/s. As the sugar solution heats up upon contact with hot coke particles, the sugar converts to caramel that coats the particles, while water evaporates: this simulates what occurs in Fluid Cokers\textsuperscript{TM}, when bitumen contacts hot coke particles, vapours are emitted and coke deposits on the particles. In Fluid Cokers\textsuperscript{TM}, the solid coke represents about 20 to 25 wt% of the injected bitumen while with the sugar solution, about 25 wt% of the solution forms caramel (sugar is first deposited and there is a subsequent mass loss as the sugar undergoes caramelization). The liquid was injected at supersonic velocity in the fluidized bed. Including the vaporization of water from the sugar solution, the superficial velocity was 0.6 m/s \((U / U_{mf} = 100)\). With this high superficial velocity, there would be higher entrainment of fine particles from the fluidized bed and due to increased bed turbulence, more bed agglomerates would break down.

High pressure nitrogen gas was simultaneously injected through the attrition nozzle while the liquid was injected through the spray nozzle. After the process, the bed was kept fluidized with the same fluidization velocity, 0.4 m/s, for 15 minutes to cool it down and then samples were collected from the bed through the screw collecting ports and the elutriated fines were recovered from the secondary and tertiary cyclones. The size distribution of these collected samples was analyzed using laser diffraction (HELOS of Sympatec). The bed particle sizes were classified as:
- Fines, with a particle diameter smaller than 45 µm. Such particles would lead to high dust emission in commercial Fluid Cokers™.
- Regular bed particles, with diameter between 45 and 420 µm.
- Micro-agglomerates with diameters below 0.85 mm. Such agglomerates would lead to small, manageable reductions in heat transfer (House et. al., 2007).
- Macro-agglomerates with a diameter larger than 0.85 mm. Such agglomerates would lead to large and problematic reductions in heat transfer (House et. al., 2007).

After each batch process the bed coke particles were unloaded and sieved to recover the macro-agglomerates, which were classified and weighted in four size cuts: 0.85 - 1.4 mm; 1.4 - 2.36 mm; 2.36 - 6.3 mm and above 6.3 mm. The effect of sieving on agglomerates breakage had been evaluated and the impact was found negligible.

The performance of the system was evaluated as follows:

- The quality of the liquid distribution was estimated from the mass of agglomerates that were formed from the original bed particles bound with the sugar from the injected sugar solution. Of particular importance are the macro-agglomerates, with a sieve size larger than 0.85 mm, for which heat and mass transfer processes would be hindered (House et al., 2007).
- The attrition performance was evaluated from:
  - The reduction in agglomerates, as shown from the evaluation of the quality of the liquid distribution.
  - The change in the average particle size of the bed particles was characterized by their Sauter mean diameter, \( d_{psm} \), including the cyclone catches but excluding the macro-agglomerates and the fines.
  - The proportion of fines, i.e. particles smaller than 45 µm. Such particles are unwanted since they lead to dust emissions problems.
2.4 Results and discussions:

The objective of this study was to investigate simultaneous liquid and attrition gas injection in a high temperature fluidized bed and their effect on particle size distribution. When the sugar solution contacts the 250 °C bed particles, its water evaporates and its sugar turns into caramel. As a first approximation, the caramel mass is assumed to be the same as the injected sugar mass (30% of 0.6 kg = 0.18 kg) and the caramel density (1386 kg/m³) is assumed to be the same as the coke particle density (1450 kg/m³).

After each experiment, the final bed mass, \(M_b\) is obtained from the sum of the initial bed mass and caramel mass, i.e. 22.18 kg. It includes both the bed contents and the particles recovered from the secondary and tertiary cyclones.

The bed particles are sorted in size cuts. Because of agglomeration, some cuts will lose volume while others will gain volume.

Knowing the total change in bed mass and the changes in the size distribution of the bed particles, one can estimate the mass of micro-agglomerates, with a diameter smaller than 0.85 mm, that have been produced. Appendix B shows a detailed derivation of the equations that were used and a sample calculation.

Effects with spray and attrition nozzles size and operating parameters on the bed particle size distribution in terms of macro-agglomerate mass, micro-agglomerate mass, Sauter-mean diameter of bed particles (45 µm < \(d_p\) < 0.85 mm) and fines (\(d_p\) < 45 µm) are discussed in the following sections.

2.4.1 Impact of simultaneous spray and attrition jets on macro-agglomerates formation

Figure 2.6 shows that, in the absence of an attrition jet, the macro-agglomerates mass decreased as the atomization gas to liquid ratio (GLR) was increased from 1.7% to 2.6% for a spray nozzle of 2.7 mm. Portoghese et al. (2008) showed the beneficial effect of higher GLR in improving the nozzle performance in a fluidized bed, within certain threshold limits, for various nozzle sizes and liquid flow rates.
With the attrition gas jet, the macro-agglomerate mass was significantly reduced (Figure 2.6). For both attrition nozzles, the production of macro-agglomerates decreased dramatically with increasing attrition gas flow rates. For the same GLR and attrition gas flow rate, the mass of macro-agglomerates was reduced when the smaller attrition nozzle was utilized, as its higher upstream pressure provided more energy to break macro-agglomerates. Zhang et al. (2011) also reported that a higher attrition performance was obtained with an increased attrition pressure along with the use of a smaller size attrition nozzle.

Figure 2.7 shows that similar trend were observed with the 3.6 mm spray nozzle. Figure 2.8 shows an example of the agglomerates that were recovered from the bed after a typical experiment of simultaneous feed injection and particles attrition.

2.4.2 Impact of simultaneous spray and attrition jets on micro-agglomerates formation and the Sauter-mean diameter of bed particles

Figures 2.9 and 2.10 show the mass of micro-agglomerates produced with the spray nozzles of 2.7 mm and 3.6 mm respectively, with simultaneous application of attrition nozzles at various operating conditions. A comparison of Figures 2.9 and 2.10 with Figures 2.6 and 2.7 shows that the increase in the mass of micro-agglomerates with either increasing GLR or the attrition gas flowrate was greater than the corresponding decrease in the mass of macro-agglomerates: large, wet macro-agglomerates were replaced with smaller and drier micro-agglomerates.

The Sauter-mean diameters (SMD) of the bed particles (45 μm < d_p < 0.85 mm) for the experiments with simultaneous liquid and attrition gas injection are illustrated in Figures 2.11 and 2.12. Improving the distribution of liquid droplets over the fluidized bed by raising the GLR produced more micro-agglomerates, and, as shown in the Figures 2.9 and 2.10, the SMD of bed particles increased. With the application of the attrition jet, the macro-agglomerates break down to smaller particles and fines and, as a result, the SMD of bed particles increases. The most significant increase in SMD of the bed particles was observed with the smaller attrition nozzle operated at the higher gas flow rate along with a higher GLR.
2.4.3 Impact of simultaneous spray and attrition jets on fines generation

Figures 2.13 and 2.14 show the mass of fines generated by the attrition performance. In all cases, the best attrition results were obtained with the highest attrition gas flow rate in combination with the smaller attrition nozzle. Attrition was most effective when the feed spray nozzle was operated simultaneously with the 1.7 mm attrition nozzle; with the same attrition gas flow rate, the smaller attrition nozzle expended more energy resulting in a dramatic reduction of large macro-agglomerates and, subsequently, in an increased mass of fines captured by the cyclones. Increasing GLR through the spray nozzles had no impact on attrition performance.

*Figure 2.6: Total macro-agglomerate mass as a function of attrition gas flow rate for spray nozzle 2.7 mm*
Spray nozzle 3.6 mm I.D.

Figure 2.7: Total macro-agglomerate mass as a function of attrition gas flow rate for spray nozzle 3.6 mm

Figure 2.8: Samples of bed particles after simultaneous agglomeration and attrition at GLR 1.7% with 2.7 mm spray nozzle and attrition nozzle with 2.4 mm at 0.62 MPa (a) particles size 0.85 - 1.4 mm (b) particles size 1.4 - 2.36 mm
Figure 2.9: Micro-agglomerate mass as a function of attrition gas flow rate for spray nozzle 2.7 mm

Figure 2.10: Micro-agglomerate mass as a function of attrition gas flow rate for spray nozzle 3.6 mm
Figure 2.11: Sauter-mean diameter of bed particles \((45 \, \mu m < d_p < 0.85 \, mm)\) as a function of attrition gas flow rate for spray nozzle 2.7 mm

Figure 2.12: Sauter-mean diameter of bed particles \((45 \, \mu m < d_p < 0.85 \, mm)\) as a function of attrition gas flow rate for spray nozzle 3.6 mm
Figure 2.13: Fines ($d_p > 45 \text{ \mu m}$) mass as a function of attrition gas flow rate for spray nozzle 2.7 mm

Figure 2.14: Fines ($d_p > 45 \text{ \mu m}$) mass as a function of attrition gas flow rate for spray nozzle 3.6 mm
2.5 Conclusions

A novel method has been developed to investigate the simultaneous particle agglomeration and attrition process occurring in a high temperature fluidized pilot plant simulating the commercial Fluid Coking™ process. The use of higher Gas-to-Liquid ratios (GLR) for spray nozzles allows for better control over the agglomerate size distribution. Applying smaller size of attrition nozzle with similar gas mass flow rate concurrently with feed nozzle, results in a lower amount of macro-agglomerates and a correspondingly higher amount of micro-agglomerates. Attrition nozzle performance increases with higher attrition pressure, but, for the same attrition pressure, there is an optimal nozzle size for an optimal agglomerates breakage.

2.6 Nomenclature

<table>
<thead>
<tr>
<th>Symbol</th>
<th>Description</th>
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<tbody>
<tr>
<td>$U$</td>
<td>Fluidization velocity (m/s)</td>
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<tr>
<td>$U_{mf}$</td>
<td>Minimum fluidization velocity (m/s)</td>
</tr>
<tr>
<td>$GLR$</td>
<td>Gas to Liquid ratio (wt %)</td>
</tr>
<tr>
<td>$SMD$</td>
<td>Sauter mean diameter (µm)</td>
</tr>
<tr>
<td>$M_b$</td>
<td>Mass of bed particle (kg)</td>
</tr>
<tr>
<td>$Q_i$</td>
<td>Volume fraction (%)</td>
</tr>
<tr>
<td>$d_i$</td>
<td>Particle size (µm)</td>
</tr>
<tr>
<td>$N_i$</td>
<td>Number of particles in particle size $d_i$</td>
</tr>
<tr>
<td>$V_i$</td>
<td>Change in volume with particle size $d_i$, (m$^3$)</td>
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Greek Letter

<table>
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<tr>
<th>Symbol</th>
<th>Description</th>
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<tbody>
<tr>
<td>$\rho_p$</td>
<td>Particle density (kg/m$^3$)</td>
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</table>
2.7 References


Chapter 3: Effect of different spray and attrition jet interaction in a high temperature fluidized bed

3.1 Abstract

A high temperature fluidized bed unit was used with simultaneous liquid injection and attrition, using separate nozzles that were located to allow for interactions between the jet cavities that they created in the fluidized bed. The effect of jet interactions on liquid-solid contact and particle attrition was studied. When the attrition nozzle was inclined so that its jet would hit the spray jet near its tip, there was a large reduction in the formation of large liquid-solid agglomerates, but particle attrition was reduced and the formation of fines was increased.

Keywords: Fluid Coker\textsuperscript{TM}, Agglomeration, Attrition, Spray, Jet interaction, Fluidized bed

3.2 Introduction

Fluid Coking\textsuperscript{TM} is used commercially to crack bituminous feed thermally in a fluidized bed of hot coke particles. Bitumen feed is atomized with steam and injected in reactors at around 350°C. The cracking reaction takes place on the surface of coke particles at the temperatures of 510-550°C (House et. al., 2004) and produce gas, liquid oil and solid coke. The gas and liquid oil leave the reactor in gaseous form and the coke deposits over existing bed particles. A challenge in Fluid Coking\textsuperscript{TM} operation is to distribute feed bitumen evenly on fluidized coke particles while avoiding agglomeration. In a Coker, liquids are injected radially inward through spray nozzles with atomization steam, forming a dilute jet cavity with in fluidized bed. Liquid droplets contact with particles both in jet cavity and near the tip of this cavity. During the process, the coke particles become gradually larger due to the deposition of product coke layers and the
agglomeration of smaller particles. Controlling the particle size distribution in a Fluid Coker™ is a major challenge, as it has a large impact on Coker operation. The particle size is controlled with attrition nozzles through which high velocity steam is injected to break the particles. The steam jets entrain bed particles, accelerate them and slam them onto dense bed particles, resulting in attrition and breakage. It is important to keep the bed particle size constant without generating too many fines or consuming too much steam.

Large agglomerates promote slugging and poor fluidization while with too many fines below 50 microns, agglomeration will take place (Dunlop et al., 1958) and dust emissions will increase. Poor heat transfer within wet agglomerates result in wet agglomerates reaching the stripper sheds, at the bottom of the Fluid Coker™, where they can hit the sheds, promoting stripper fouling, which leads to premature shut-down.

In Fluid Coking™, large agglomerates inhibit heat and mass transfer, reducing the yield of valuable product and deteriorating operation. Ariyapadi et al. (2003) used X-ray imaging to show that with the conventional nozzle configuration, agglomerates formed at jet bed interface due to the coalescence of wet particles and droplets near the tip of the spray jet cavity. Bruhns et al. (2005) also observed the formation of wet agglomerates from spray jets in a fluidized bed.

Previous studies have shown that drier agglomerates break up more quickly, eliminating the problems caused by agglomeration (Weber et al., 2006). It is therefore, essential to improve solid-liquid mixing in or near the spray jets to minimize the liquid content of the wet agglomerates. Several studies suggested that solid liquid mixing would be improved by minimizing the liquid flux and maximizing the solids flux at the interface between the dense bed and the spray jet cavity (Iveson et al., 2001). Minimizing the liquid flux is not economical, as it would reduce the unit capacity and it is, thus, important to enhance the solids flux. However, the local liquid to solid ratio varies over the cross section of the jet cavity and with typical spray nozzles, is highest near the jet axis and decreases with the radial distance from the axis (House et al., 2004). McMillan et al. (2005) showed that placing a co-axial tube downstream of the spray nozzle tip enhances solid- liquid mixing
with in jet cavity. This new feed nozzle configuration is known as ESE (Enhanced Solid Entrainment) nozzle is quite effective in reducing the formation of liquid-solid agglomerates (McMillan et al., 2005). However, the application of ESE in Fluid Coking has been limited by concerns about erosion of its tube by high velocity coke particles.

In the Fluid Coking™ process, converging-diverging attrition nozzles are used to maintain the particle size within an optimal range. This configuration of nozzle can provide supersonic velocities and the grinding efficiency of the bed particles can be maximized by optimizing the nozzle geometry, such as the angle of its divergent section (Cruz et al., 2010). Li et al (2011) has studied the effect of bed temperature over attrition performance and reported that higher grinding efficiency can be achieved at higher bed temperatures. The grinding efficiency is defined as:

\[
\text{Grinding efficiency, } \eta = \frac{\text{New particle surface created by attrition}}{\text{Mass of required attrition gas}} = \frac{m^2/s}{kg/s} = \frac{m^2}{kg}
\]

Tuunila and Nystorm (1998) studied with the effect of attrition angle inclination over fluidized bed, using three different downward angles with the horizontal directions 23°, 33° and 43° and reported the highest grinding efficiency with the largest angle: 43°. While Ahlbus (1996) found the similar trend and reported the most effective grinding with 60° inclination.

In current Fluid Cokers™, spray and attrition nozzles are located in different sections and do not interact. The objective of this study was to determine whether interactions between the jet cavities from spray and attrition nozzles could minimize agglomerate formation.

### 3.3 Experimental setup and methodology

Experiments were performed in a hot fluidized bed column, 1.23 m high, with a 0.508 m × 0.114 m rectangular cross-section (Figure 3.1). The fluidized bed height was 0.45 m. Bed particles were petroleum coke with a Sauter mean diameter of 110 µm and a particle density of 1450 kg/m³. The coke particles in the bed were fluidized with nitrogen gas
through a sparger distributor and heated to 250° C with an electric heater system immersed in the upper region of the bed, well above the spray and attrition nozzles. Cold binding liquid (30% sugar solution), with approximately the same viscosity (3 cP) as the bitumen injected in commercial cokers (Gray et al., 2002), was injected inside the hot bed with a spray nozzle using nitrogen for atomization, at a height of 0.25 m above the gas distributor. Cold nitrogen gas was injected in the hot fluidized bed through an attrition nozzle at a height of 0.13 m above the gas distributor and 0.13 m below the spray nozzle. The bed was equipped with three cyclones: solids collected by the 1st cyclone were recycled to the bed through a dip-leg, while solids from the secondary and the tertiary cyclones were recovered outside the bed.
The spray nozzle used in Fluid Coker™ is known as TEB nozzle, which was invented by Terrance E. Base¹. A TEB nozzle with a throat diameter of 2.7 mm was used as spray nozzle. It was scaled down from the commercial scale spray nozzle to use in the hot fluidized unit. Figure 3.2 shows a schematic diagram of the spray nozzle. Figure 3.4 illustrates that the liquid was fed to the nozzle from a pressurized blow tank, through a venture pre-mixer (Figure 3.3) where it was mixed with the atomization gas. Pressure $P_1$ and $P_2$ were set with pressure regulators to achieve the required liquid flow rate and atomization gas flow rate, expressed as GLR (atomization gas to liquid mass ratio).

**Figure 3. 1: Schematic of the high temperature fluidized bed with spray and attrition nozzle. (Modified from Feng, 2010)**
Figure 3. 2: TEB spray nozzle (Throat diameter 2.7 mm)

Figure 3. 3: Schematic of pre-mixer for 2.7 mm TEB nozzle

Figure 3. 4: Spray nozzle setup for the fluidized bed

A convergent-divergent attrition nozzle with a 1.7 mm throat diameter (Figure 3.5) was used with a gas mass flow rate of 320 g/min with an upstream pressure of 2.2 MPa (300
psig). In such a convergent-divergent nozzle, sonic velocity is reached at the throat and supersonic velocity is attained in the divergent channel (Cruz et al., 2010). Two different orientations along with the conventional horizontal attrition nozzle orientation have been tested in this work: 14° and 45° angles with the horizontal plane (Figure 3.6). Preliminary measurements showed that:

- There was no interaction between spray and attrition jets when the attrition nozzle inclination was 0°.
- When the attrition nozzle inclination was 14°, the attrition jet connected with the spray jet cavity near its tip.
- When the attrition nozzle inclination was 45°, the attrition jet connected with the spray jet cavity near its base, just ahead of the spray nozzle tip.

![Attrition nozzle (Throat diameter 1.7 mm)](image)

**Figure 3. 5: Attrition nozzle (Throat diameter 1.7 mm)**
(i) Achieving attrition effect at tip of spray jet (Attrition nozzle positioned at $14^\circ$ angle)

(ii) Achieving attrition effect at spray nozzle tip (Attrition nozzle positioned at $45^\circ$ angle)

This was confirmed by estimating the lengths of both the spray and attrition jet cavities with well established correlations for the types of nozzles used in this study. The length of the gas-liquid jet was calculated with the empirical correlation proposed by Siva (2004).
Where,
\[ \rho_p = \text{Particle density, kg/m}^3 \]
\[ \rho_g = \text{Gas density, kg/m}^3 \]
\[ \rho_L = \text{Liquid density, kg/m}^3 \]
\[ G_L = \text{Superficial mass velocity of the liquid at the nozzle tip, kg/s-m}^2 \]
\[ [S] = \text{mean slip velocity ratio} \]
\[ \varepsilon' = \text{Voidage at nozzle exit} \]
\[ C_g = \text{nozzle geometry parameter} \]

The length of attrition jet, \( L_{gjet} \) was calculated with the empirical correlation proposed by Li (2011)

\[
L_{gjet} = \frac{\alpha}{g^\beta} \left( \frac{1}{(\rho_p - \rho_g)^\beta g} (\rho_g U_g^2)^\beta d_o^{(1-\beta)} \right)
\]

The value of \( \alpha \) and \( \beta \) depends on the particle type and the bed temperature. For bed temperatures ranging 200 to 500° C and coke particles, \( \alpha = 0.38 \) and \( \beta = 0.54 \)

By calculation,
- Gas-Liquid jet length, \( L_{jet} \) = 0.35 m
- Attrition gas jet, \( L_{gjet} \) = 0.35 m
- Horizontal penetration of attrition gas jet for 14°: \( 0.35 \times \cos (14°) = 0.34 \) m
- Horizontal penetration of attrition gas jet for 45°: \( 0.35 \times \cos (45°) = 0.25 \) m
- Vertical penetration of attrition gas jet for 45°: \( 0.35 \times \sin (45°) = 0.25 \) m

A mass of 22 kg of sieved coke particles (\( d_p < 420 \) μm), with a minimum fluidization velocity, \( U_{mf} = 0.007 \) m/s at the bed conditions, were first introduced into the bed. The bed particles were then heated with an electric heater to a target temperature of 250° C at a low fluidization velocity of 0.05 m/s (7 \( \times \) \( U_{mf} \)), to maximize heat transfer and minimize heat losses. Once the required bed temperature was reached, the bed superficial velocity was raised to 0.3 m/s (42 \( \times \) \( U_{mf} \)) and during the actual experiments; the superficial
velocity was in range of 0.48 to 0.49 m/s (68.5 to 70 × \(U_{mf}\)) due to the contributions of the gas streams from the atomization and attrition nozzles.

To achieve simultaneous agglomeration and attrition, gas was started through both attrition and spray nozzles and the liquid flow was immediately initiated through the spray nozzle. It took about 20 s to inject 600 g of 30 % sugar solution into the bed. Both atomization and attrition gas flows were then stopped. As the sugar solution heats up upon contact with hot coke particles, the sugar converts to caramel that coats the particles, while water evaporates: this simulates what occurs in Fluid Cokers\textsuperscript{TM}, when bitumen contacts hot coke particles, vapours are emitted and coke deposits on the particles. In Fluid Cokers\textsuperscript{TM}, the solid coke represents about 20 to 25 wt% of the injected bitumen while with the sugar solution, about 25 wt% of the solution forms caramel (sugar is first deposited and there is a subsequent mass loss as the sugar undergoes caramelization). The liquid was injected at supersonic velocity in the fluidized bed. Including the vaporization of water from the sugar solution, the superficial velocity was about 0.74 to 0.75 m/s (105 to 107 × \(U_{mf}\)). With this high superficial velocity, there would be higher entrainment of fine particles from the fluidized bed and due to increased bed turbulence, more bed agglomerates would break down.

A higher liquid flow rate (30 g/s) had been applied in this study, when compared to the liquid flow rate (3.33 g/s) used in Chapter 2, to achieve a more realistic balance between particle growth and attrition. The superficial gas velocity had been lowered to 0.3 m/s for better simulation of Fluid Coking\textsuperscript{TM} conditions. With this higher liquid flow rate and lower superficial gas velocity, bogging was observed at lower GLRs (1.7 and 2.6%) and experiments were, therefore, conducted with GLRs of 3.6% and higher, to completely eliminate bogging by improving the liquid-solid contact and allowing quicker evaporation. Under bogging conditions, the fluidization quality is highly degraded, leading to unrealistic results.

After the liquid injection, the bed was kept fluidized with the same fluidization velocity of 0.3 m/s for 15 minutes, after which the elutriated fines were recovered from the secondary and tertiary cyclones. The bed particle sizes were classified as:
o Fines, with a particle diameter smaller than 45 µm. Such particles would lead to high dust emission in commercial Fluid Cokers™.

o Regular bed particles, with diameter between 45 and 420 µm.

o Micro-agglomerates with diameters below 0.85 mm. Such agglomerates would lead to small, manageable reductions in heat transfer (House et. al., 2007).

o Macro-agglomerates with a diameter larger than 0.85 mm. Such agglomerates would lead to large and problematic reductions in heat transfer (House et. al., 2007).

After each batch process the bed coke particles were unloaded and sieved to recover the macro-agglomerates, which were classified and weighted in three size cuts: 0.85 - 2 mm; 2 - 4 mm and above 4 mm. The effect of sieving on agglomerates breakage had been evaluated and the impact was found negligible. The particle size distribution of screened samples (d_p < 0.85 mm) was then analyzed using a laser diffraction method (HELOS of Sympatec).

The performance of the system was evaluated as follows:

- The quality of the liquid distribution was estimated from the mass of agglomerates that were formed from the original bed particles bound with the sugar from the injected sugar solution. Of particular importance are the macro-agglomerates, with a sieve size larger than 0.85 mm, for which heat and mass transfer processes will be hindered (House et al., 2007).

- The attrition performance was evaluated from:
  
  o The reduction in agglomerates, as shown from the evaluation of the quality of the liquid distribution.
  
  o The proportion of fines, i.e. particles smaller than 45 µm. Such particles are unwanted since they lead to dust emissions problems.
  
  o The change in the average particle size of the bed particles was characterized with their Sauter mean diameter, d_psm, including the cyclone catches but excluding the macro-agglomerates and the fines.
3.4 Results and discussion

First of all, two baseline experiments were performed with only nitrogen gas injection through attrition and spray nozzles respectively to observe bed behavior in terms of cyclones catch. A bed mass with 22 kg of coke was heated to 250 °C and superficial velocity of 0.3 m/s was attained. Nozzles were operated for 20 s, as for all planned experiments. As shown in Table 3.1, with separate operation of either attrition or spray nozzles, the total mass of secondary and tertiary cyclone catches seems negligible when compared to the total bed mass. For the planned experiments, however, the cyclone catches were always collected, weighed and analyzed for particle size, so that their contribution to the overall size distribution of the combined bed and cyclone catches particles could be included, as it might become significant when there was interaction between spray and attrition jets.

<table>
<thead>
<tr>
<th>Nozzles operating conditions</th>
<th>Total cyclone catch, wt% of total bed mass</th>
</tr>
</thead>
<tbody>
<tr>
<td>Attrition at 2.2 MPa (300 psig) only</td>
<td>0.02</td>
</tr>
<tr>
<td>Spray nozzle with only gas, corresponding to 5.5% GLR</td>
<td>0.03</td>
</tr>
</tbody>
</table>

Table 0.1: Total cyclone catch mass (wt % of bed mass) for only N2 gas injection through attrition and spray nozzles

Table 3.2 shows the nitrogen gas flow rate through the spray nozzle with increasing GLR (Gas to Liquid Ratio), through the attrition nozzle and the total gas consumption rate when the spray and attrition nozzles were operating simultaneously. Table 3.3, presents the superficial gas velocity in the upper bed region and the freeboard, when only fluidization gas introduced through the gas distributor at the base of the bed, when gas was added through the spray and attrition nozzles. The gas pressure upstream of the attrition nozzle, 2.2 MPa (300 psig) is typical for attrition nozzles used in processes such as Fluid Coking™ (Li et al., 2011), while the GLR used for the spray nozzle is higher than
usual for spray nozzles in the Fluid Coking™ process (Portoghese et al., 2008). The higher GLRs were required to avoid bed bogging.

<table>
<thead>
<tr>
<th>Nozzles operating conditions</th>
<th>N₂ gas consumption rate, g/s</th>
</tr>
</thead>
<tbody>
<tr>
<td>3.6% GLR</td>
<td>1.08</td>
</tr>
<tr>
<td>4.6% GLR</td>
<td>1.37</td>
</tr>
<tr>
<td>5.5% GLR</td>
<td>1.66</td>
</tr>
<tr>
<td>2.2 MPa Attrition</td>
<td>10.26</td>
</tr>
<tr>
<td>3.6% GLR and 2.2 MPa attrition</td>
<td>11.34</td>
</tr>
<tr>
<td>4.6% GLR and 2.2 MPa attrition</td>
<td>11.63</td>
</tr>
<tr>
<td>5.5% GLR and 2.2 MPa attrition</td>
<td>11.92</td>
</tr>
</tbody>
</table>

**Table 0.2: Nitrogen gas consumption rate (g/s) with spray and attrition nozzles**

<table>
<thead>
<tr>
<th>Operating conditions</th>
<th>V₉, m/s</th>
</tr>
</thead>
<tbody>
<tr>
<td>Fluidization only</td>
<td>0.30</td>
</tr>
<tr>
<td>With 3.6% GLR and 2.2 MPa attrition</td>
<td>0.48</td>
</tr>
<tr>
<td>With 4.6% GLR and 2.2 MPa attrition</td>
<td>0.49</td>
</tr>
<tr>
<td>With 5.5% GLR and 2.2 MPa attrition</td>
<td>0.49</td>
</tr>
</tbody>
</table>

**Table 0.3: Superficial gas velocity in the upper bed region and the freeboard for various bed operating conditions**

Micro-agglomerates are the agglomerates that are not retained on a sieve with 0.85 mm openings. Since their sieve diameter is of the same order as the diameter of some of the bed particles, they can be identified through sieving. A new method was, therefore developed to calculate their total mass.

When the sugar solution contacts the 250 °C bed particles, its water evaporates and its sugar turns into caramel. As a first approximation, the caramel mass is assumed to be the same as the injected sugar mass (30% of 0.6 kg = 0.18 kg) and the caramel density (1386 kg/m³) is assumed to be the same as the coke particle density (1450 kg/m³).
After each experiment, the final bed mass, $M_b$, is obtained from the sum of the initial bed mass and caramel mass, i.e. 22.18 kg. It includes both the bed contents and the particles recovered from the secondary and tertiary cyclones.

The bed particles are sorted in size cuts. Because of agglomeration, some cuts will lose volume while others will gain volume.

Knowing the total change in bed mass and the changes in the size distribution of the bed particles, one can estimate the mass of micro-agglomerates, with a diameter smaller than 0.85 mm, that have been produced. Appendix B shows a detailed derivation of the equations that were used and a sample calculation.

The objective was to use interactions between spray and attrition jets to reduce the formation of large wet macro-agglomerates ($d_p > 0.85$ mm), which have a detrimental impact on the Fluid Cokers™, while avoiding a major degradation of the attrition performance (House et. al., 2007). All the experiments were repeated once and all the data are reported here.

### 3.4.1 Impact of spray and attrition jets interaction on macro-agglomerates formation

A first set of experiments were conducted with only liquid injection (0.6 kg of 30% sugar solution) with GLRs of 3.6, 4.6 and 5.5%, in the absence of the attrition jet, to analyze the formation of agglomerates with fluidized bed particles. Figure 3.7 shows that all size cuts of macro-agglomerates were reduced with increasing GLR, indicating a better distribution of the liquid on the fluidized particles. This confirms the results obtained by Portoghese et al. (2008). Increasing the GLR improved the liquid-solid contact because of the formation of finer liquid droplets and a higher particle entrainment rate into the spray jet (Hulet et al., 2003 and Briens et al., 2008).

Figure 3.8 illustrates how the use of the attrition jet reduced the total macro agglomerate mass, when compared to the results obtained with the same spray in the absence of the attrition jet. With an attrition nozzle angle of 0°, there was no interaction between spray and attrition jets and the reduction in macro-agglomerate production was significant but
moderate. With an attrition angle of 14°, the attrition jet hit the spray jet cavity near its tip and the reduction in macro-agglomerate production was significant. The largest reduction in macro-agglomerate production was achieved when the attrition nozzle angle was 45° and the attrition jet hit the spray jet cavity near its base (Figures 3.6 and 3.9).

Figures - 3.9, 3.10 and 3.11 show that the production of all macro-agglomerate sizes was reduced when the attrition and spray jets interacted. This result was obtained for all atomization GLRs and, in all cases, the most important reduction in macro-agglomerates, independent of their size, was achieved when the attrition nozzle was 45°, and the attrition jet hit the spray jet cavity near its tip.

3.4.2 Impact of spray and attrition jets interaction on micro-agglomerates formation

Figure 3.12 shows that the mass of micro-agglomerates increased with increasing GLR. The better liquid distribution achieved by increasing the GLR resulted in a reduction of macro-agglomerates (Figure 3.6) and a concurrent increase in the formation of micro-agglomerates (Figure 3.12).

Figure 3.13 shows that the use of an attrition jet resulted in a larger production of micro-agglomerates. With an attrition nozzle angle of 0°, the increase in micro-agglomerate mass was moderate for all GLRs. With an attrition angle of 14°, the attrition jet hit the spray jet cavity near its tip and the increase in micro-agglomerate production was significant. The most important increase in micro-agglomerate production was achieved when the attrition nozzle angle was 45° and the attrition jet hit the spray jet cavity near its base.

Figure 3.14 indicates that there was a reverse correlation between the productions of micro-agglomerates and macro-agglomerates. As the distribution of the liquid binder on the bed particles was improved, the production of macro-agglomerates was reduced and more micro-agglomerates were produced. Interestingly, the increase in the mass of micro-agglomerates was significantly larger than the corresponding reduction in the mass of
macro-agglomerates: as the liquid distribution was improved, the agglomerates that were formed were both smaller and drier.

3.4.3 Impact of spray and attrition jets interaction on the Sauter-mean diameter of bed particles and on the production of fines

Figure 3.15 shows the change in the Sauter mean diameter of the bed particles, exclusive of fines and macro-agglomerates ($45 \mu m < d_p < 0.85 \text{ mm}$) during one simultaneous spray-attrition run. The change was characterized with:

$$\text{SMD of bed particles (45 } \mu \text{m } < d_p < 0.85 \text{ mm) change ratio } = \frac{d_{psm} - d_{psm,fresh}}{d_{psm,fresh}} \times 100$$

Improving the distribution of liquid droplets over the fluidized bed particles by raising the GLR produced more micro-agglomerates, as shown in the previous section (Figure 3.12). Consequently, the Sauter mean diameter of the bed particles ($45 \mu m < d_p < 0.85 \text{ mm}$) increased. An additional, but minor contribution to the increase in particle size was the deposition of a layer of caramel over individual particles. Figure 3.15 also shows that the application of the liquid spray resulted in the loss of some fines. Dunlop et al. (1958) showed that when heavy oil is injected into a bed of coke particles, fines tend to attach to larger particles to form agglomerates and the similar behavior observed in this study shows that the use of sugar solution at moderate temperatures ($250 \ ^\circ \text{C}$) provided a good simulation of injection of heavy oil in Fluid Coking$^{\text{TM}}$. Improving the liquid distribution by increasing the GLR enhanced the reduction in fines, as less binder was wasted in the very wet macro-agglomerates.

Ariyapadi et al. (2003) examined the jet-bed interaction with an X-ray imaging technique and reported about the formation of agglomerates around the jet cavity. Most of the wet agglomerates are formed near the tip of the jet cavity. Since wet agglomerates require less energy to break up than drier and stronger agglomerates, using the attrition jet to disrupt the tips of the spray jet cavity was effective in reducing agglomerate formation. However, disrupting the base of the cavity was most effective: this suggests that, to reduce macro-agglomerate production, it is even more important to disrupt the base of the jet cavity and
increase of the entrainment of dry bed particles into the central core of the jet, resulting in agglomerates that drier and easier to break.

Unfortunately, the interaction of the spray and attrition jets had a detrimental impact on particle attrition. First, Figure 3.16 shows that interactions resulted in larger increases in the Sauter-mean diameter of bed particles, with the largest increase achieved when the attrition jet hit the base of the spray jet cavity. Second, Figure 3.17 indicates that there was a much larger amount of undesirable fines formed when the two jets interacted; here again, the worst results were obtained when the attrition jet hit the base of the spray jet cavity.

Figure 3.18 shows that there is a general relationship between the total macro-agglomerate mass and the Sauter-mean diameter of the bed particles. The Sauter-mean diameter of the bed particles always increased when the macro-agglomerate mass decreased. This clearly shows that, when there is interaction between spray and attrition jets, there is a trade-off between the attrition performance and the improvement of the liquid-solid contact associated with the macro-agglomerate mass reduction.
Spray only condition

Figure 3.7: Macro-agglomerate mass (wt\%) as a function of GLR (wt\%) for spray only condition

Figure 3.8: Total macro-agglomerate mass reduced (wt \%) from spray only condition as a function of attrition angle
Figure 3.9: Macro-agglomerate mass reduced from spray only condition for GLR 3.6% as a function of attrition angle.

Figure 3.10: Macro-agglomerate mass reduced from spray only condition for GLR 4.6% as a function of attrition angle.
Figure 3.11: Macro-agglomerate mass reduced from spray only condition for GLR 5.5% as a function of attrition angle.

Figure 3.12: Micro-agglomerate mass (wt% of total bed mass) as a function of GLR (wt%) for spray only condition, i.e. in the absence of attrition nozzles.
Figure 3.13: Increase in the total mass of micro-agglomerates, relative to the increase observed in the absence of attrition nozzles.

Figure 3.14: Total macro-agglomerate mass (wt %) vs. micro-agglomerate mass (wt %).
Spray only condition

Gas to Liquid Ratio, wt%

Relative reduction in mass of fines, wt%
SMD of (45 micron < \(d_p\) < 0.85 mm) change ratio,

Figure 3.15: Reduction in the mass of fines and changes in the SMD of the bed particles (45 µm < \(d_p\) < 0.85 mm) as a function of GLR for spray only condition

Attrition angle, deg

SMD of bed particles (45 µm < \(d_p\) < 0.85 mm) change ratio, %

GLR 3.6%
GLR 4.6%
GLR 5.5%

Figure 3.16: SMD of bed particles (45 µm < \(d_p\) < 0.85 mm) change ratio as a function of attrition angle
Figure 3. 17: Fines ($d_p < 45 \mu m$) mass generated as a function of attrition angle

Figure 3. 18: Total macro-agglomerate mass (wt %) vs. SMD of bed particles ($45 \mu m < d_p < 0.85 \text{ mm}$)
3.5 Conclusion

Interactions between attrition and spray jets reduced the generation of macro-agglomerates. The most effective interactions were achieved when the attrition jet disrupted the base of spray jet cavity.

There is a trade-off between the attrition performance and the improvement of the liquid-solid contact associated with the reduction in macro-agglomerate production. When the attrition nozzle was located and inclined so that its jet would hit the spray jet near its base, there was a large reduction in the formation of large liquid-solid agglomerates, but the mass of micro-agglomerates was increased, more undesirable fines were generated and the desirable formation, through attrition, of particles larger than 45 µm was reduced.

3.6 Nomenclature

\( U_g \)  Fluidization velocity (m/s)
\( U_{mf} \)  Minimum fluidization velocity (m/s)
\( GLR \)  Gas to Liquid ratio (wt %)
\( SMD \)  Sauter mean diameter (m)
\( L_{jet} \)  Gas-liquid jet length (m)
\( L_{gjet} \)  Attrition gas jet length (m)
\( G_L \)  Superficial mass velocity of liquid at nozzle tip (kg/s-m²)
\( g \)  Gravity (m/s²)
\( d_i \)  Particle diameter (µm)
\( Q_i \)  Volume fraction (%)
\( N_i \)  Number of particles
\( M_b \)  Mass of bed particle (kg)

Greek Letters

\( \rho_p \)  Particle density (kg/m³)
\( \varepsilon \)  Voidage
3.7 References

1US Patent 6003789, “Nozzle for atomizing liquid in two phase flow”


Chapter 4: Application of satellite jets with spray nozzle in a high temperature fluidized bed

4.1 Abstract

A high temperature fluidized bed unit was used to simulate the agglomerate formation in Fluid Cokers™: a sugar solution was injected to simulate heavy oil injection which, in Fluid Cokers™, gives gases and vapors, simulated by the water in the sugar solution, and a solid coke residue, simulated by the caramel deposit formed from the sugar. In Fluid Cokers™, large agglomerates that result from the liquid injection are undesirable since they lead to a reduction in the yield of valuable products and to operating problems. Two different sizes of spray nozzles were used to achieve two different liquid flow rates. Surrounding the spray nozzle with three peripheral sonic gas jets, or “satellite jets” was found to be beneficial. Less large solid-liquid agglomerates were formed and the mass of undesirable bed fines was reduced.

Keywords: Fluid Coker™, Spray, Agglomeration, Periphery gas jet

4.2 Introduction

Fluid coking™ is used commercially to crack bituminous feed thermally in a fluidized bed of hot coke particles. Bitumen feed is atomized with steam and injected in reactors at around 350 °C. The cracking reaction takes place on the surface of coke particles at the temperature of 510-550 °C (House et. al., 2004) and produce gas, liquid oil and solid coke. The gas and liquid oil leave the reactor in gaseous form and the coke deposits over existing bed particles. A challenge in Fluid Coker™ operation is to distribute feed bitumen evenly on fluidized coke particles while avoiding agglomeration. In a Coker, liquids are injected radially inward through spray nozzles with atomization steam,
forming a dilute jet cavity and near the tip of this cavity. In Fluid Coking™, large agglomerates inhibit heat and mass transfer, reducing the yield of valuable product and deteriorating operation. Ariyapadi et al. (2003) used X-ray imaging to show that with the conventional nozzle configuration, agglomerates formed at jet bed interface due to the coalescence of wet particles and droplets near the tip of the spray jet cavity. Bruhns et al. (2005) also observed the formation of wet agglomerates from spray jets in a fluidized bed.

The ideal condition in Fluid Coker™ is to achieve thermal cracking of the injected heavy oil without heat and mass transfer limitations. House et al. (2007) developed a model that showed that the formation of large agglomerates resulted in a drastic reduction in the conversion rate of the heavy oil. Injected liquid then survives for a long time in stable agglomerates and lead to fouling of the stripper baffle, at the bottom of the coker bed, resulting in premature shut-down of the Fluid Coker™. Stripper fouling can be moderated by increasing the bed temperature, but this reduces the production of valuable liquid and increases undesirable SO₃ emissions due to the additional combustion of coke in the burner that reheats the coke particles that are then recirculated to the reactor. Bi et al. (2005) suggested several approach to reduce stripper fouling rate by modifying reactor internal hydrodynamics, changing the feedstock properties and improving spray nozzle performance. Reactor bed hydrodynamics can be changed by varying operating conditions like, fluidization velocity, solid circulation rate or modifying stripper section design. Feed properties can be altered by pre-treatment of injected bitumen. An attractive simple solution would be to change the spray nozzle design to improve its performance (House et al., 2004).

McMillan et al. (2005) and Chan et al. (2006) improved the distribution of the injected liquid on the fluidized solids placing a co-axial draft tube downward of the spray nozzle. McMillan et al. (2005) showed that within the jet cavity formed by the spray nozzle in the fluidized bed, this tube worked by enhancing the mixing of liquid rich regions with peripheral solid-rich regions. A concern that prevents the widespread commercial application of this technology, however, is that tube may foul and plug with coke deposits. House et al. (2007) proposed a new configuration to obtain similar benefits with
a system that would not be subject to fouling. They applied three peripheral gas jets around the spray jet, naming sonic shroud, to entrain more solids into the jet cavity. The study from House et al. (2007), however, used a room temperature system that did not closely model what happens in Fluid Cokers™.

The objective of this study was to explore the potential of peripheral gas jets to reduce formation of large agglomerates, using a more realistic model of heavy oil coking.

4.3 Experimental setup and methodology

Experiments were performed in a hot fluidized bed with column height of 1.23 m, and with a 0.508 m × 0.114 m rectangular cross-section (Figure 4.1). The fluidized bed height was 0.45 m. Bed particles were petroleum coke with a Sauter mean diameter of 110 µm and a particle density of 1450 kg/m³. The coke particles in the bed were fluidized with nitrogen gas through a sparger distributor and heated to 250 °C with an electric heater system immersed in the upper region of the bed, well above the spray and attrition nozzles. Binding liquid (30% sugar solution), with approximately the same viscosity (3 cP) as the bitumen injected in commercial cokers (Gray et al., 2002), was injected inside the hot fluidized bed with a spray nozzle, using nitrogen and atomization at a height of 0.25 m above the gas distributor. The bed was equipped with three cyclones: solids collected by the 1st cyclone were recycled to the bed through a dip-leg, while solids from the secondary and the tertiary cyclones were recovered outside the bed.

The spray nozzle used in Fluid Coker™ is known as TEB nozzle, which was invented by Terrance E. Base. Two TEB spray nozzles with throat diameters of 1.2 and 2.7 mm, respectively, were used to achieve liquid flow rates of 15 and 30 g/s respectively. Figure 4.2 shows a schematic diagram of the spray nozzle.

With the 1.2 mm TEB spray nozzle, satellite nozzles with an inner diameter of 0.13 mm were used and distance from the axis of the satellite nozzles to the spray nozzle axis was 11 mm (Figure 4.3). With the 2.7 mm TEB spray nozzle, satellite nozzles with an inner
diameter of 0.40 mm were used and distance from the axis of the satellite nozzles to the spray nozzle axis was 25.4 mm (Figure 4.4).

Figure 4. 1: Schematic of the high temperature fluidized bed with spray nozzle. (Modified from Feng, 2010)

Figure 4. 2: TEB spray nozzle used for experiments (Throat diameter 1.2 and 2.7 mm)
A mass of 22 kg of sieved coke particles (d_p < 420 µm) were first introduced into the bed. Their minimum fluidization velocity, at operating conditions of 250 °C, was U_{mf} = 0.007 m/s. The bed particles were then heated with an electric heater to a target temperature of 250 °C at a low fluidization velocity of 0.05 m/s (7 × U_{mf}), to maximize heat transfer and
minimize heat losses. Once the required bed temperature was reached, the bed was then kept at a constant superficial velocity of 0.3 m/s ($42 \times U_{mf}$).

600 g of 30% sugar solution were sprayed inside the bed with either a TEB nozzle or a combination of TEB and satellite nozzles. Subsequent to the injection, the bed was kept fluidized with the same fluidization velocity of 0.3 m/s for 15 minutes, after which the elutriated fines were recovered from the secondary and tertiary cyclones. The bed particle sizes were classified as:

- Fines, with a particle diameter smaller than 45 µm. Such particles would lead to high dust emission in commercial Fluid Cokers™.
- Regular bed particles, with diameter between 45 and 420 µm.
- Micro-agglomerates with diameters below 0.85 mm. Such agglomerates would lead to small, manageable reductions in heat transfer (House et al., 2007).
- Macro-agglomerates with a diameter larger than 0.85 mm. Such agglomerates would lead to large and problematic reductions in heat transfer (House et al., 2007).

After each batch process the bed coke particles were unloaded and sieved to recover the macro-agglomerates, which were classified and weighted in three size cuts: 0.85 - 2 mm; 2 - 4 mm and above 4 mm. The particle size distribution of screened samples ($d_p < 0.85$ mm) was then analyzed using laser diffraction method (HELOS of Sympatec).

The performance of the liquid distribution was evaluated from:

- The mass of agglomerates that were formed from the original bed particles bound with the caramel from the injected sugar solution.
  - Of particular importance are the macro-agglomerates, with a sieve size larger than 0.85 mm, for which heat and mass transfer processes will be seriously hindered (House et al., 2007).
  - The mass of micro-agglomerates, with a diameter smaller than 0.85 mm, that were produced, was also obtained.
- The proportion of fines, i.e. particles smaller than 45 µm. Such particles are unwanted since they lead to dust emissions problems.
• The change in the average particle size of the bed particles by Sauter mean diameter, \(d_{\text{psm}}\), including the cyclone catches but excluding the macro-agglomerates and the fines.

Table 4.1 shows the pressure upstream of the spray nozzle for the combination of the 1.2 mm spray nozzle and 0.13 mm satellite gas flows through the satellite nozzles was expressed as total GLR or ratio of the gas mass flow rate to the liquid flowrate through the spray nozzle. With the 1.2 mm spray nozzle, liquid was injected at a flow rate of 15 g/s for 40 s.

Table 4.2 shows similar data of table 4.1 corresponding to the 2.7 mm spray nozzle and 0.4 mm satellite nozzles combination. With the 2.7 mm spray nozzle, liquid was injected at a flow rate of 30 g/s for 20 s.

To get relevant results, it was essential to avoid bogging, the sharp reduction in fluidization quality that is observed when the bed becomes too wet. Due to its lower liquid flow rate, bogging-free operation could be achieved in the fluidized bed with the 1.2 mm spray nozzle and a GLR of 2.5%. With the higher liquid flow rate (30 g/s) of the 2.7 mm spray nozzle, bed bogging was observed at a GLR of 2.5% and results are only shown for the higher GLRs.

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<tr>
<th>Total GLR, wt%</th>
<th>Upstream satellite nozzle pressure, psig</th>
<th>Upstream spray nozzle pressure, psig</th>
<th>Gas flow through each satellite, % of Total</th>
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<td>2.5</td>
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<td>201.5</td>
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Table 0.1: Operating conditions with 1.2 mm spray nozzle and 0.13 mm satellite nozzles combination
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<th>Total GLR, wt%</th>
<th>Upstream Satellite nozzle pressure, psig</th>
<th>Upstream spray nozzle pressure, psig</th>
<th>Gas flow through each satellite, % of Total</th>
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<tr>
<td>3.6%</td>
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<td>12.69</td>
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<tr>
<td>4.6%</td>
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<td>5.5%</td>
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<td>135</td>
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Table 0.2: Operating conditions with 2.7 mm spray nozzle and 0.4 mm satellite nozzles combination

4.4 Results and discussion

All the experiments conducted in this study were fully exploratory, as the operating parameters that would optimize the particle size distribution were unknown. All of the experiments were repeated once and all the data are reported here. The objective was to reduce the generation of large macro-agglomerates ($d_p > 0.85$ mm) by improving the liquid injection into the fluidized bed.

After each experiment, the final bed mass, $M_b$ is obtained from the sum of the initial bed mass and caramel mass, i.e. 22.18 kg. It includes both the bed contents and the particles recovered from the secondary and tertiary cyclones.

When the sugar solution contacts the 250 °C bed particles, its water evaporates and its sugar turns into caramel. As a first approximation, the caramel mass is assumed to be the same as the injected sugar mass (30% of 0.6 kg = 0.18 kg) and the caramel density (1386 kg/m$^3$) is assumed to be the same as the coke particle density (1450 kg/m$^3$).

The bed particles are sorted in size cuts. Because of agglomeration, some cuts will lose volume while others will gain volume. Knowing the total change in bed mass and the changes in the size distribution of the bed particles, one can estimate the mass of micro-
agglomerates, with a diameter smaller than 0.85 mm, that have been produced. Appendix B shows a detailed derivation of the equations that were used and a sample calculation.

4.4.1 Impact of peripheral gas jets on macro-agglomerates formation

Figure 4.5 shows that total using the satellite nozzles greatly reduced the production of undesirable macro-agglomerates, for both spray nozzle sizes. Because the use of atomization gas in costly in commercial units, the operation of the spray nozzle with and without satellite nozzles was performed with the same total GLR, so that, when the satellite nozzles were used, the flow of atomization gas through the spray nozzle was reduced to compensate for the gas used by the satellite nozzles. As shown in Figure 4.5, reducing the GLR through a spray nozzle increased the production of macro-agglomerates and the satellite nozzles, thus, more than compensated for this increase in macro-agglomerates production. The reduction in macro-agglomerates production achieved by the satellite nozzles, for the same total GLR, varied from 15 to 30%. Note that with the larger spray nozzle, the production of macro-agglomerates was higher because the same amount of liquid was injected in half the time, reducing the ability of the fluidized bed to transport the wet solids from the spray region to the rest of the bed.

Figures 4.6 and 4.7 illustrate the relative reduction in the production of macro-agglomerates of various sizes that was achieved with the satellite nozzles, for the same total GLR. The satellite nozzles reduced the production of all sizes of macro-agglomerates.

4.4.2 Impact of peripheral gas jets on micro-agglomerates formation and the Sauter mean diameter of bed particles

Figure 4.8 shows how the production of micro-agglomerates varied with the GLR. Without satellite nozzles, increasing the GLR, which had reduced the macro-agglomerates production (Figure 4.5), increased the generation of micro-agglomerates. The use of satellite nozzles, which had reduced the macro-agglomerates generation (Figure 4.5), also increased the production of micro-agglomerates.
Figure 4.9 shows that, in all cases, breaking up macro-agglomerates resulted in an increased generation of micro-agglomerates. Figure 4.9 also shows that the increase in the mass of micro-agglomerates was much larger than the corresponding reduction in the mass of macro-agglomerates: this indicates that the micro-agglomerates were much drier than the macro-agglomerates that they replaced. Drier and smaller agglomerates are much less of a problem for Fluid Cokers™ (House et. al., 2007) and using satellite nozzle would therefore be highly beneficial.

Figure 4.10 shows that the Sauter mean diameter of the bed particles (45 μm < d_p < 0.85 mm) increased with the GLR with the use of satellite nozzles. This is the result of both the increased generation of micro-agglomerates and the possible increased amount of caramel that forms around individual particles.

Figure 4.11 shows that there was an excellent correlation between the reduction in macro-agglomerates production and the increase in the Sauter mean diameter of the bed particles.

### 4.4.3 Impact of peripheral gas jets on bed fine particles generation

Figure 4.12 shows that, with the smaller, 1.2 mm spray nozzle, nearly all the fines disappeared from the bed. Dunlop et al. (1958) showed that coke fines agglomerate preferentially with larger particles and the injection of liquid, which results in the production of agglomerates, reduce the fines.

Figure 4.12 shows that, with the larger, 2.7 mm spray nozzle, fines did not disappear. Improving the liquid distribution, by either increasing the GLR or using satellite nozzles, reduced the fines. As shown above, when liquid distribution was improved, a small mass of very wet macro-agglomerates was replaced by a much larger mass of drier micro-agglomerates that, therefore, could capture more fines.
Figure 4.5: Total macro-agglomerate mass (wt %) as a function of total GLR (wt %) for various spray nozzles

Figure 4.6: Macro-agglomerate mass reduction with various size cuts for combination of TEB 1.2 mm and Satellite 0.13 mm nozzles from macro-agglomerates mass with TEB 1.2 mm
Figure 4.7: Macro-agglomerate mass reduction with various size cuts for combination of TEB nozzle 2.7 mm and Satellite 0.4 mm nozzles from macro-agglomerate mass with TEB 2.7 mm

Figure 4.8: Micro-agglomerate mass (wt %) as a function of total GLR (wt %) for various spray nozzles
Figure 4. 9: Total macro-agglomerate mass (wt %) as a function of micro-agglomerate mass (wt %) for various spray nozzles.

Figure 4. 10: SMD of bed particles (45 µm < d_p < 0.85 mm) as a function of total GLR (wt %) for various spray nozzle.
**Figure 4.11:** Total macro-agglomerate mass (wt%) vs. SMD of bed particles (45 \( \mu m < d_p < 0.85 \) mm) with various spray nozzles

**Figure 4.12:** Fines (\( d_p < 45 \mu m \)) mass (wt%) as a function of total GLR (wt%)
4.5 Conclusion

Surrounding the spray nozzle with three peripheral sonic gas jets, or “satellite jets” was beneficial as:

- it reduced the production of large macro-agglomerates, which are highly undesirable as they lead to large and problematic reductions in heat transfer.
- it increased the production of smaller, drier micro-agglomerates, which are much less of a problem for Fluid Cokers™.
- it reduced the amount of undesirable fines, which would reduce dust emissions from Fluid Coking plants.

The success of the satellite nozzles is likely due to their ability to enhance the entrainment of bed particles into the liquid-rich regions of the jet cavity that the spray nozzles formed in the fluidized bed. It should be possible to obtain even better results by optimizing the geometry and position of the satellite nozzles.

4.6 Nomenclature

\[ U_g \quad \text{Fluidization velocity (m/s)} \]
\[ U_{mf} \quad \text{Minimum fluidization velocity (m/s)} \]
\[ GLR \quad \text{Gas to Liquid ratio (wt %)} \]
\[ SMD \quad \text{Sauter mean diameter (µm)} \]
\[ d_i \quad \text{Particle diameter (µm)} \]
\[ Q_i \quad \text{Volume fraction (%)} \]
\[ N_{fresh} \quad \text{Number of particles in fresh bed with particle size cut d}_i \]
\[ N_i \quad \text{Number of particles} \]
\[ M_b \quad \text{Mass of bed particle (kg)} \]
\[ \Delta V_i \quad \text{Volume change in particle size cut } d_i \text{ (m}^3\text{)} \]

\textit{Greek Letter}

\[ \rho_p \quad \text{Particle density (kg/m}^3\text{)} \]
4.7 References

US Patent 6003789, “Nozzle for atomizing liquid in two phase flow”


Chapter 5:  Conclusions and Recommendations

5.1 Conclusions

1. A new, high temperature experimental system that simulate the evaporation and coke formation processes that occur in Fluid Coking™ had been developed and utilized in this research project. A sugar solution is sprayed into the fluidized bed of coke particles at 250 °C. The water evaporates and the sugar is transformed into caramel.

2. Under all conditions, increasing the flowrate of atomization gas through the spray nozzle always reduced the production of large, macro-agglomerates, with a diameter larger than 0.85 mm. Such macro-agglomerates are undesirable, since they lead to poor liquid yields and operating problems in Fluid Cokers™. At high atomization gas flowrates, macro-agglomerates are replaced with smaller, drier micro-agglomerates, which would be much preferable.

3. The effect of simultaneous liquid spray and high velocity attrition gas in a fluidized bed of coke particles was investigated, and showed that, even when attrition and spray jets did not interact directly, there was a reduction in the production of macro-agglomerates. Macro-agglomerates were replaced with drier micro-agglomerates. These beneficial effects were enhanced through the direct interaction of the attrition and spray jets, with the best results obtained when the attrition jet hit the base of the spray jet. Unfortunately, the highest reduction in macro-agglomerates formation was associated with the highest generation rate of unwanted fines.

4. Similar beneficial effects on macro-agglomerates reduction were obtained by disrupting the base of the spray jet with small, peripheral gas jets. However, this configuration has also a beneficial effect on the fines, by reducing their generation.
5.2 Recommendations

1. This study considered two extreme cases of direct interaction between the attrition jet and the spray jet, with the attrition jet hitting either the base of the tip of the spray jet cavity. Other types of direct interactions should be investigated.

2. The satellite jets application was found most promising to control the formation of large agglomerates and the generation of fines. To be more realistic, it would be interesting to test the indirect interaction of a spray nozzle with satellite jets simultaneously with a standard attrition jet, as in Chapter 2.
Appendix A

Experimental equipment and procedure:

Experiments were performed in a hot fluidized bed column, 1.23 m high, with a 0.508 m x 0.114 m rectangular cross-section (Figure A.1). The fluidized bed height was 0.45 m. Bed particles were petroleum coke with a Sauter mean diameter of 110 µm and a particle density of 1450 kg/m³. The coke particles in the bed were fluidized with nitrogen gas through a sparger distributor and heated to 250°C with an electric heater system immersed in the upper region of the bed, well above the spray and attrition nozzles. Cold binding liquid (30% sugar solution), with approximately the same viscosity (3 cP) as the
bitumen injected in the commercial Coker, was injected inside the hot bed with a spray nozzle using nitrogen for atomization, at a height of 0.24 m above the gas distributor. Cold nitrogen gas was injected in the hot fluidized bed through an attrition nozzle at a height of 0.13 m above the gas distributor and 0.11 m below the spray nozzle. The bed was equipped with three cyclones: solids collected by the 1st cyclone were recycled to the bed through a dip-leg, while solids from the secondary cyclone (Figure A.2) were recovered outside the bed and solids from tertiary cyclone was negligible.

![Figure A.2. Schematic diagram of secondary cyclone](image)

A mass of 22 kg of sieved coke particles (d<sub>p</sub> < 420 µm), with a minimum fluidization velocity, U<sub>mf</sub> = 0.007 m/s at the bed conditions, were first introduced into the bed. The bed particles were then heated with an electric heater at a low fluidization velocity of 0.05 m/s (U<sub>mf</sub>) for 1.5 to 2 hours to achieve target temperature of 250 °C. Once the required bed temperature was reached, the bed superficial velocity was raised to 0.3 m/s.
(42 \times U_{mf}) and was kept fluidized for 15 minutes to achieve uniform bed temperature. Spray and attrition nozzles were then operated with designed experimental conditions.

After the simultaneous spray and attrition nozzle operation, the bed was kept fluidized with the same fluidization velocity of 0.3 m/s for 15 minutes, to cool down the unit. After which the bed particles were recovered using outlet valve and during unloading the bed was fluidized at minimum fluidization velocity of 0.007 m/s. Elutriated fines were then recovered from the secondary cyclone. The bed particle sizes were classified as:

- Fines, with a particle diameter smaller than 45 \mu m. Such particles would lead to high dust emission in commercial Fluid Cokers™.
- Regular bed particles, with diameter between 45 and 420 \mu m.
- Micro-agglomerates with diameters below 0.85 mm. Such agglomerates would lead to small, manageable reductions in heat transfer (House et. al., 2007).
- Macro-agglomerates with a diameter larger than 0.85 mm. Such agglomerates would lead to large and problematic reductions in heat transfer (House et. al., 2007).

After each batch process the bed coke particles were sieved to recover the macro-agglomerates, which were classified and weighted in three size cuts: 0.85 - 2 mm; 2 - 4 mm and above 4 mm. The effect of sieving on agglomerates breakage had been evaluated and the impact was found negligible. The particle size distribution of screened samples (d_p < 0.85 mm) was then analyzed using a laser diffraction method (HELOS of Sympatec).
Appendix B

Micro-agglomerate mass (wt %) calculation:

Sample calculation for liquid injection at 30 g/s with 3.6% GLR and attrition at 2.2 MPa with 0° angle orientation (Chapter 3)

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<tr>
<th>Particle size, µm</th>
<th>Fresh coke Cumulative dist., Q%</th>
<th>Cumulative dist., µm</th>
<th>2nd Cyclone</th>
<th>3.6% GLR and 0° attrition angle Cumulative dist., µm</th>
<th>Bed particle g</th>
<th>Total particle g</th>
<th>Cumulative dist., Q%</th>
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Table B1: The cumulative distribution for total bed particles with fresh bed and after experiment
When the sugar solution contacts the 250 °C bed particles, its water evaporates and its sugar turns into caramel. As a first approximation, the caramel mass is assumed to be the same as the injected sugar mass (30% of 0.6 kg = 0.18 kg) and the caramel density (1386 kg/m$^3$) is assumed to be the same as the coke particle density (1450 kg/m$^3$).

After each experiment, the final bed mass, $M_b$, is obtained from the sum of the initial bed mass, 22.00 kg and caramel mass, e.g. 0.18 kg, for a total of 22.18 kg. It includes both the bed contents and the particles recovered from the secondary and tertiary cyclones.

The bed particles are sorted in size cuts. If $Q_i$ is the volume fraction of the bed particles (including the cyclone catches) in size cut $i$, the number of particles in this size cut is given by:

$$N_i = Q_i \times \frac{M_b}{\rho_p} \times \frac{1}{\pi \frac{d_i^3}{6}}$$

where,

$M_b$ = Mass of bed particles, kg
$Q_i$ = Volume fraction in particle size cut $d_i$
$\rho_p$ = Particle density, kg/m$^3$

So, the change in number of particles from that of the fresh bed, $N_{\text{fresh}}$, in each particle size cut $d_i$, can be calculated from the difference in number of particles between fresh and final bed, $(N_i - N_{i,\text{fresh}})$ and the volume change in each size cut given by:

$$\Delta V_i = (N_i - N_{i,\text{fresh}}) \times \frac{\pi}{6} d_i^3$$

A positive value for $\Delta V_i$, i.e. a gain in volume of the particles in this cut, indicates fines generation for $d_{pi} < 45$ µm or agglomerate formation $d_{pi} > 45$ µm.

A negative value $\Delta V_i$, means that particles have disappeared from this cut, either because of their breakage into smaller particles, or as was more likely in our experiments, due to agglomeration.
Table B2: Change in volume with each size cut of particles

Table B2 shows that the volume of generated fines, which can be obtained by summing up the new volume of particles for particle sizes $d_p < 45 \, \mu m$, was 0.0003 m$^3$ in this example, or 0.435 kg, assuming a particle density of 1450 kg/m$^3$.

The total volume of macro-agglomerates, which are larger than 0.85 mm, is obtained separately by sieving. Since they are no particles in the fresh solids with a diameter larger
than 0.85 mm, all the particles larger than 0.85 mm in the final solids are the result of agglomeration.

Determining the volume of micro-agglomerates, with a diameter smaller than 0.85 mm, is more complex. Table B2 shows, that in this case, cuts below 180 µm lost volume, while cuts above 180 µm gained volume. This shows that, during the experiments, some particles smaller than 180 µm disappeared and formed agglomerates larger than 180 µm.

The total volume of micro-agglomerates can, therefore, be obtained from the gain in volume of all the cuts smaller than 0.85 mm and larger than the diameter above which all the volume changes were positive, 180 µm in this example. This gives 0.00426 m³ in this case. Assuming a density of 1450 kg/m³, this gives a mass of about 6.18 kg, or 27.84% of the bed mass. Note that this method slightly underestimates the mass of micro-agglomerates, since several particles of 50 µm, for example, could have agglomerated into agglomerates smaller than 180 µm.

**Nomenclature:**

\[ d_i \] Particle diameter (µm)

\[ Q_i \] Volume fraction (%)

\[ N_{fresh} \] Number of particles in fresh bed with particle size cut \( d_i \)

\[ N_i \] Number of particles

\[ M_b \] Mass of bed particle, including secondary and tertiary cyclone catches (kg)

\[ \Delta V_i \] Volume change in particle size cut \( d_i \) (m³)

**Greek Letter**

\[ \rho_p \] Particle density (kg/m³)
Curriculum Vitae

Name: Mithun Saha

Post-secondary Education and Degrees:
Bangladesh University of Engineering and Technology, Dhaka, Bangladesh
2003 - 2008 B. Sc. in Mechanical Engineering.

Honours and Awards:
Western Graduate Scholarship
2010 - 2012

Related Work Experience:
Research Assistant
Institute for Chemicals and Fuels from Alternative Resources
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2010 - 2012