The Application Of Thick-source Alpha-particle Spectrometry To The Detection Of Disequilibrium In The Radioactive Decay Series Of Uranium-238 In Rocks And Minerals

Patrick George Killeen
THE APPLICATION OF THICK-SOURCE ALPHA PARTICLE SPECTROMETRY TO THE DETECTION OF DISEQUILIBRIUM IN THE RADIOACTIVE DECAY SERIES OF URANIUM-238 IN ROCKS AND MINERALS

by

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ABSTRACT

The natural radiation from rocks and minerals is primarily from the U-238 and Th-232 radioactive decay series. This is comprised of alpha particles of 23 different energies, along with beta and gamma radiation.

Although the analysis of rocks for uranium content by gamma-ray spectrometry is a rapid and efficient method it depends on the crucial assumption that the U-238 decay series is in radioactive equilibrium. A method of detecting the presence of disequilibrium in the U-238 decay series has been developed. A silicon semiconductor detector and a 1024 channel pulse height analyzer were used to obtain over 250 alpha particle energy spectra of uraniferous rocks and minerals. "Infinitely thick" sources were used to avoid the problems involved in preparation of "infinitely thin" sources. The problem of generating theoretical energy distributions for infinitely thick sources was solved. Complete theoretical spectra were computed for comparison with experimentally obtained alpha particle energy spectra of rocks and minerals. The presence and extent of disequilibrium in the U-238 decay series in a rock is determined from the residual spectrum obtained by subtracting a fitted theoretical spectrum from the measured spectrum of a rock.
ACKNOWLEDGEMENTS

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Special thanks is given to the following for financial assistance: the Geological Survey of Canada for financing field work and collection of the Blind River area uranium ore samples; the National Research Council which supported an assistantship of which the writer was a recipient; and the Ontario Government for financial aid in the form of Ontario Graduate Fellowships.
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CHAPTER 1

INTRODUCTION

1.1 Introduction

Fortuitously the sun was not shining on Wednesday, February 26th and Thursday, February 27th, 1896, and the photographic plates which Henri Becquerel had intended to use were left in the darkness of a drawer which also contained a specimen of Uranium sulphate. This led to the discovery of the phenomenon to which Madame Curie, in 1898, gave the name 'radioactivity'. Radioactivity can be defined as "spontaneous emission of particles from the atomic nucleus of a chemical element, changing its atomic weight or number, and thus yielding a different element", (Stein, 1954). Nuclear physics has been a direct development from the discovery of radioactivity.

The three types of radiation from radioactive substances, of interest here, are alpha particles, beta particles, and gamma rays. An alpha particle is a doubly ionized Helium atom, and consequently has an atomic mass of 4 and a double positive charge. Beta particles are essentially the same as electrons, the only difference being their place
of origin, i.e., beta particles are emitted from the atomic nucleus of a radionuclide, while an electron is an extra-nuclear particle originating from an atomic orbit. Gamma rays are photons emitted after a radioactive decay by either alpha or beta emission, when the nucleus of the daughter element is in an excited state. The gamma radiation represents the energy emitted as the daughter nucleus goes to the ground state.

Since alpha particles and gamma rays are emitted with discrete energies characteristic of the decaying radionuclide a measurement of the energies of these radiations can yield information concerning the identity and amount of radionuclide. This is the basis of alpha-particle-, and gamma-ray-spectrometry. Beta particles on the other hand are not emitted with discrete energies, but have in general a continuous distribution of energies with a maximum energy which is unique to the radionuclide. General descriptions of radioactivity are available in Lapp and Andrews, (1963), Rutherford, et al. (1951), Kaplan, (1964), and Arya, (1968). More detailed discussions are given by Evans, (1955), and by Siegbahn, (1968).

1.2 Natural Radioactivity

There are several naturally occurring radionuclides, the most familiar being potassium-40 and those that comprise the decay series of uranium and thorium. Wahl and Bonner, (1951) list additional naturally occurring radioisotopes
of rubidium, samarium, lutetium, and rhenium, Kohman and Saito, (1954) add radioisotopes of lanthanum, bismuth, tungsten, and neodymium to the list, and Adams, (1962) adds isotopes of indium, tellurium, and iodine. In addition to these there are cosmic ray induced radioisotopes such as tritium and carbon-14.

The naturally occurring radioisotopes which are the concern of this study, are those of the Uranium-238, Thorium-232, and Uranium-235 decay series. The three decay series are shown in Figure 1. The radioelements in these decay series along with Potassium-40 are the major sources of natural radioactivity (i.e. radioactivity in rocks). This is because of their specific activity and abundances compared to all other natural radioisotopes (Adams, 1962).

In order to determine the distribution of the radioactive elements in the earth, representative sampling of the earth with analysis of these major sources of radioactivity must be accomplished. Radiometric methods of analysis have been developed to increase the speed and accuracy of these determinations. A knowledge of the distribution of uranium and thorium is of considerable economic significance. Such knowledge is also valuable in the computation of the radiogenic contribution to terrestrial heat flow, in understanding geologic processes involving geochemical differentiation, and in interpreting the history of the earth and its place in relation to the evolution of the universe.
Classification of natural radioactivity (after Rosholt, 1957)

Figure 1.
1.3 Gamma Ray Spectrometry

The most commonly used method of analysis for uranium and thorium in geologic materials is that of gamma-ray spectrometry. The method involves the counting of gamma rays of energies usually chosen as 1.76 Mev and 2.62 Mev for the uranium and thorium analyses respectively. These gamma radiations are actually produced by daughter products bismuth-214 in the U-238 decay series and thallium-208 in the Th-232 decay series. The gamma ray count rate is then related to the amount of parent, assuming there is a relation between the amount of daughter and parent. This assumption is more precisely defined by stating that the radioactive decay series is assumed to be in a state of secular equilibrium.

1.4 Radioactive Equilibrium

A radioactive decay series such as that for U-238 is said to be in a state of secular equilibrium when the number of atoms of each daughter in the series being produced is equal to the number of atoms of that daughter being lost by radioactive decay. To express this mathematically a short review of the laws of radioactive decay must be given first (e.g. Arya, 1968).

The rate of loss by decay is proportional to the amount of radioactive element present, i.e.
$$\frac{dN_1}{dt} = -\lambda_1 N_1 \quad [1.1]$$

where \( N_1 \) = the amount of element \( N_1 \)

\( \lambda_1 \) = the decay constant for the element \( N_1 \)

then the rate of decay of the daughter of \( N_1 \) is given by the difference between its loss rate and its production rate from its parent.

e.

$$\frac{dN_2}{dt} = \lambda_1 N_1 - \lambda_2 N_2 \quad [1.2]$$

if at time \( t = 0 \), only parent \( N_1 \) is present in amount \( N_{10} \), then integrating equation \([1.1]\) we get:

\[ N_1 = N_{10} e^{-\lambda_1 t} \quad [1.3] \]

This can be substituted into equation \([1.2]\) giving:

$$\frac{dN_2}{dt} = \lambda_1 N_{10} e^{-\lambda_1 t} - \lambda_2 N_2 \quad [1.4]$$

Collecting terms in \( N_2 \), and multiplying by \( e^{\lambda_2 t} \) we get an equation which integrates to become:

$$N_2 e^{\lambda_2 t} = \frac{\lambda_1}{\lambda_2 - \lambda_1} N_{10} e^{(\lambda_2 - \lambda_1) t} + C \quad [1.5]$$
Using the initial conditions that at $t=0, N_2=0$ we find $C$ and arrive at:

$$
N_2 = \frac{\lambda_1}{\lambda_2 - \lambda_1} N_{10} (e^{-\lambda_1 t} - e^{-\lambda_2 t}) \quad [1.6]
$$

In the case of a parent with a half life much greater than its daughter i.e. $\lambda_1(\ll \lambda_2$ equation $[1.6]$ reduces to:

$$
N_2 = (\frac{\lambda_1}{\lambda_2}) N_{10} (1-e^{-\lambda_2 t}) \quad [1.7]
$$

After a long period of time i.e. $t$ is much greater than the half life of the daughter $(t \gg (1/\lambda_2))$ equation $[1.7]$ becomes

$$
N_2 = (\frac{\lambda_1}{\lambda_2}) N_{10} \quad [1.8]
$$

Thus the amount of daughter is constant. Its rate of production from parent is equal to its rate of loss by its own decay. This is then a state of secular equilibrium.

For a radioactive decay series, secular equilibrium then implies that

$$
\lambda_1 N_1 = \lambda_2 N_2 = \lambda_3 N_3 = \ldots \ldots \lambda_n N_n \quad [1.9]
$$

The solution for the number of atoms of the nth member of the series was derived by Bateman (1910), assuming initially that only the parent existed. Then:
\[ N_n(t) = c_1 e^{-\frac{\lambda_1 t}{2}} + c_2 e^{-\frac{\lambda_2 t}{2}} + c_3 e^{-\frac{\lambda_3 t}{2}} + \ldots \]
\[ c_n e^{-\frac{\lambda_n t}{2}} \]

where

\[ c_1 = \frac{\lambda_1 \lambda_2 \cdots \lambda_{n-1}}{(\lambda_2 - \lambda_1)(\lambda_3 - \lambda_1) \cdots (\lambda_n - \lambda_1)} N_{10} \]

\[ c_2 = \frac{\lambda_1 \lambda_2 \cdots \lambda_{n-1}}{(\lambda_1 - \lambda_2)(\lambda_3 - \lambda_2) \cdots (\lambda_n - \lambda_2)} N_{10} \]

\[ \vdots \]

\[ c_n = \frac{\lambda_1 \lambda_2 \cdots \lambda_{n-1}}{(\lambda_1 - \lambda_n)(\lambda_2 - \lambda_n) \cdots (\lambda_{n-1} - \lambda_n)} N_{10} \]

1.5 Radioactive Disequilibrium

The question then becomes whether the assumption of secular equilibrium, required for analysis by gamma-ray spectrometric techniques, is valid for the geologic material being analyzed for its uranium content.

If one or more of the daughter products is being lost by any process other than radioactive decay, then equation [1.9] is not satisfied. Since each daughter product is a different chemical element, they will
behave differently in the same environment. For example, Radon-222 in the U-238 decay series is a gas, and it could possibly be lost to the decay system during its 3.8 day half life. The chemical solubility of radium and uranium and thorium isotopes also differ, and preferential leaching of isotopes is another possibility.

1.6 Objectives of this Research

The problem of radioactive equilibrium in the U-238 decay series has been studied primarily by elaborate and tedious radiochemical analysis techniques involving combinations of mass spectrometric isotope ratio determinations, and more recently, thin source alpha particle spectrometric analysis of some of the radiochemically separated isotopes. Another promising method possible with the development of high resolution lithium drifted germanium detectors involves counting gamma rays of low energies but this involves the interference of the Compton spectrum at low energies, and a very low efficiency gamma-ray detector.

Since most techniques involve elaborate time consuming sample preparation, with the consequent possibility of introducing disequilibrium, one of the present objectives was to develop a technique of detecting the presence of disequilibrium which minimized sample preparation time. The characteristic energies of the
alpha particles were to be utilized in alpha spectrometry with a large silicon semi-conductor detector, which provides very good energy resolution. Ideally, the method should not only detect the presence of disequilibrium in a rock sample, but also identify the radioelement which is the cause of the disequilibrium.
CHAPTER 2

ALPHA PARTICLE SPECTROMETRY

2.1 Theoretical Alpha Particle Spectra

A study of the energy spectra of alpha particles emitted by radioisotopes of the U-238 decay series along with the U-235 series and the Th-232 series which are associated with it, requires such knowledge as the peak energies, peak intensities or activities, order of decay and half lives of all isotopes in the series. Most of this data is available in 'Table of Isotopes' compiled by Lederer, Hollander, and Perlman (1968). Table 1 lists the pertinent data for the U-238 series, and the Th-232 series for radioactive equilibrium conditions. The energies are given in Mev, and intensity is in relative percentages where the number of decays of parent element represents 100%. The range in air is measured in centimeters and is taken from Nogami and Hurley (1948). The range in air is not given for the U-235 series shown in Table 2 since the U-235 component in the alpha spectrum is negligible compared to the U-238 component, and the data is not required here. Since most rocks contain thorium along
### U-238 Decay Series in Equilibrium

<table>
<thead>
<tr>
<th>ENERGY</th>
<th>PERCENT</th>
<th>DECAY ORDER</th>
<th>ISOTOPE</th>
<th>HALF LIFE</th>
<th>RANGE IN AIR</th>
</tr>
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<tbody>
<tr>
<td>4.20</td>
<td>77.0</td>
<td>1.0</td>
<td>U238</td>
<td>4.51E09Y</td>
<td>2.65</td>
</tr>
<tr>
<td>4.15</td>
<td>23.0</td>
<td>1.0</td>
<td>U238</td>
<td>4.51E09Y</td>
<td>2.65</td>
</tr>
<tr>
<td>4.80</td>
<td>72.0</td>
<td>2.0</td>
<td>U234</td>
<td>2.47E05Y</td>
<td>3.21</td>
</tr>
<tr>
<td>4.75</td>
<td>28.0</td>
<td>2.0</td>
<td>U234</td>
<td>2.47E05Y</td>
<td>3.21</td>
</tr>
<tr>
<td>4.70</td>
<td>76.0</td>
<td>3.0</td>
<td>Th230</td>
<td>8.00E04Y</td>
<td>3.09</td>
</tr>
<tr>
<td>4.65</td>
<td>24.0</td>
<td>3.0</td>
<td>Th230</td>
<td>8.00E04Y</td>
<td>3.09</td>
</tr>
<tr>
<td>4.85</td>
<td>94.6</td>
<td>4.0</td>
<td>Ra226</td>
<td>1602 Y</td>
<td>3.26</td>
</tr>
<tr>
<td>4.60</td>
<td>5.4</td>
<td>4.0</td>
<td>Ra226</td>
<td>1602 Y</td>
<td>3.00</td>
</tr>
<tr>
<td>5.50</td>
<td>100.0</td>
<td>5.0</td>
<td>Rn222</td>
<td>3.83 Day</td>
<td>4.05</td>
</tr>
<tr>
<td>6.00</td>
<td>100.0</td>
<td>6.0</td>
<td>Po218</td>
<td>3.05 Min</td>
<td>4.66</td>
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<tr>
<td>7.70</td>
<td>100.0</td>
<td>7.0</td>
<td>Po214</td>
<td>1.6E-04S</td>
<td>6.91</td>
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<td>5.30</td>
<td>100.0</td>
<td>8.0</td>
<td>Po210</td>
<td>138.4D</td>
<td>3.84</td>
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### Th-232 Decay Series in Equilibrium

<table>
<thead>
<tr>
<th>ENERGY</th>
<th>PERCENT</th>
<th>DECAY ORDER</th>
<th>ISOTOPE</th>
<th>HALF LIFE</th>
<th>RANGE IN AIR</th>
</tr>
</thead>
<tbody>
<tr>
<td>4.01</td>
<td>77.0</td>
<td>1.0</td>
<td>Th232</td>
<td>1.41E10Y</td>
<td>2.10</td>
</tr>
<tr>
<td>3.95</td>
<td>23.0</td>
<td>1.0</td>
<td>Th232</td>
<td>1.41E10Y</td>
<td>2.10</td>
</tr>
<tr>
<td>5.42</td>
<td>71.0</td>
<td>2.0</td>
<td>Th228</td>
<td>1.91 Y</td>
<td>3.95</td>
</tr>
<tr>
<td>5.34</td>
<td>28.0</td>
<td>2.0</td>
<td>Th228</td>
<td>1.91 Y</td>
<td>3.95</td>
</tr>
<tr>
<td>5.68</td>
<td>94.5</td>
<td>3.0</td>
<td>Ra224</td>
<td>3.64 D</td>
<td>4.28</td>
</tr>
<tr>
<td>5.45</td>
<td>5.5</td>
<td>3.0</td>
<td>Ra224</td>
<td>3.64 D</td>
<td>4.00</td>
</tr>
<tr>
<td>6.28</td>
<td>100.0</td>
<td>4.0</td>
<td>Rn220</td>
<td>55.0 Sec</td>
<td>5.00</td>
</tr>
<tr>
<td>6.78</td>
<td>100.0</td>
<td>5.0</td>
<td>Po216</td>
<td>0.15 Sec</td>
<td>5.64</td>
</tr>
<tr>
<td>6.09</td>
<td>9.8</td>
<td>6.0</td>
<td>Bi212</td>
<td>60.6 Min</td>
<td>4.73</td>
</tr>
<tr>
<td>6.04</td>
<td>24.9</td>
<td>6.0</td>
<td>Bi212</td>
<td>60.6 Min</td>
<td>4.73</td>
</tr>
<tr>
<td>8.78</td>
<td>64.0</td>
<td>7.0</td>
<td>Po212</td>
<td>3.0E-07S</td>
<td>8.57</td>
</tr>
</tbody>
</table>

**TABLE 1. Statistics for Alpha Particle Decay of U-238 and Th-232**
### U-235 Decay Series in Equilibrium

#### U-235 Complex

<table>
<thead>
<tr>
<th>ENERGY</th>
<th>PERCENT</th>
<th>ORDER</th>
<th>ISOTOPE</th>
<th>HALF LIFE</th>
</tr>
</thead>
<tbody>
<tr>
<td>4.60</td>
<td>4.6</td>
<td>1.0</td>
<td>U235</td>
<td>7.1E08 Y</td>
</tr>
<tr>
<td>4.56</td>
<td>3.7</td>
<td>1.0</td>
<td>U235</td>
<td>7.1E08 Y</td>
</tr>
<tr>
<td>4.42</td>
<td>4.0</td>
<td>1.0</td>
<td>U235</td>
<td>7.1E08 Y</td>
</tr>
<tr>
<td>4.40</td>
<td>57.0</td>
<td>1.0</td>
<td>U235</td>
<td>7.1E08 Y</td>
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<tr>
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<td>1.0</td>
<td>U235</td>
<td>7.1E08 Y</td>
</tr>
<tr>
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<td>3.0</td>
<td>1.0</td>
<td>U235</td>
<td>7.1E08 Y</td>
</tr>
<tr>
<td>4.22</td>
<td>5.7</td>
<td>1.0</td>
<td>U235</td>
<td>7.1E08 Y</td>
</tr>
</tbody>
</table>

#### PA-231 Complex

<table>
<thead>
<tr>
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<th>ORDER</th>
<th>ISOTOPE</th>
<th>HALF LIFE</th>
</tr>
</thead>
<tbody>
<tr>
<td>5.05</td>
<td>10.0</td>
<td>2.0</td>
<td>PA231</td>
<td>3.25E04 Y</td>
</tr>
<tr>
<td>5.02</td>
<td>23.0</td>
<td>2.0</td>
<td>PA231</td>
<td>3.25E04 Y</td>
</tr>
<tr>
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<td>24.0</td>
<td>2.0</td>
<td>PA231</td>
<td>3.25E04 Y</td>
</tr>
<tr>
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<td>22.0</td>
<td>2.0</td>
<td>PA231</td>
<td>3.25E04 Y</td>
</tr>
<tr>
<td>4.73</td>
<td>11.0</td>
<td>2.0</td>
<td>PA231</td>
<td>3.25E04 Y</td>
</tr>
</tbody>
</table>

#### TH-227 Complex

<table>
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<th>ORDER</th>
<th>ISOTOPE</th>
<th>HALF LIFE</th>
</tr>
</thead>
<tbody>
<tr>
<td>6.04</td>
<td>24.0</td>
<td>3.0</td>
<td>TH227</td>
<td>18.2 D</td>
</tr>
<tr>
<td>5.98</td>
<td>23.0</td>
<td>3.0</td>
<td>TH227</td>
<td>18.2 D</td>
</tr>
<tr>
<td>5.96</td>
<td>3.0</td>
<td>3.0</td>
<td>TH227</td>
<td>18.2 D</td>
</tr>
<tr>
<td>5.76</td>
<td>20.0</td>
<td>3.0</td>
<td>TH227</td>
<td>18.2 D</td>
</tr>
<tr>
<td>5.71</td>
<td>4.9</td>
<td>3.0</td>
<td>TH227</td>
<td>18.2 D</td>
</tr>
<tr>
<td>5.70</td>
<td>8.2</td>
<td>3.0</td>
<td>TH227</td>
<td>18.2 D</td>
</tr>
<tr>
<td>5.69</td>
<td>3.6</td>
<td>3.0</td>
<td>TH227</td>
<td>18.2 D</td>
</tr>
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</table>

#### RA-223 Complex

<table>
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<th>ISOTOPE</th>
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</tr>
</thead>
<tbody>
<tr>
<td>5.75</td>
<td>9.1</td>
<td>4.0</td>
<td>RA223</td>
<td>11.43 D</td>
</tr>
<tr>
<td>5.71</td>
<td>53.7</td>
<td>4.0</td>
<td>RA223</td>
<td>11.43 D</td>
</tr>
<tr>
<td>5.61</td>
<td>26.0</td>
<td>4.0</td>
<td>RA223</td>
<td>11.43 D</td>
</tr>
<tr>
<td>5.54</td>
<td>9.1</td>
<td>4.0</td>
<td>RA223</td>
<td>11.43 D</td>
</tr>
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</table>

#### RN-219 Complex

<table>
<thead>
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<th>PERCENT</th>
<th>ORDER</th>
<th>ISOTOPE</th>
<th>HALF LIFE</th>
</tr>
</thead>
<tbody>
<tr>
<td>6.82</td>
<td>81.0</td>
<td>5.0</td>
<td>RN219</td>
<td>4.0 S</td>
</tr>
<tr>
<td>6.55</td>
<td>11.5</td>
<td>5.0</td>
<td>RN219</td>
<td>4.0 S</td>
</tr>
<tr>
<td>6.42</td>
<td>7.5</td>
<td>5.0</td>
<td>RN219</td>
<td>4.0 S</td>
</tr>
</tbody>
</table>

#### PO-215

<table>
<thead>
<tr>
<th>ENERGY</th>
<th>PERCENT</th>
<th>ORDER</th>
<th>ISOTOPE</th>
<th>HALF LIFE</th>
</tr>
</thead>
<tbody>
<tr>
<td>7.38</td>
<td>99.9</td>
<td>6.0</td>
<td>PO215</td>
<td>1.78E-03 S</td>
</tr>
</tbody>
</table>

#### BI-211

<table>
<thead>
<tr>
<th>ENERGY</th>
<th>PERCENT</th>
<th>ORDER</th>
<th>ISOTOPE</th>
<th>HALF LIFE</th>
</tr>
</thead>
<tbody>
<tr>
<td>6.62</td>
<td>84.1</td>
<td>7.0</td>
<td>BI211</td>
<td>2.15 M</td>
</tr>
<tr>
<td>6.28</td>
<td>15.9</td>
<td>7.0</td>
<td>BI211</td>
<td>2.15 M</td>
</tr>
</tbody>
</table>

**Table 2. Statistics for Alpha Particle Decay of U-235**
with uranium, the thorium series data are important. The Th/U ratio in most rocks in the lithosphere is about 4.0 (Adams, 1962).

The relative activities of the series are also important. The specific activities given by Hyde et al. (1964) are:

1 mg of pure natural Uranium with isotopes in proper ratios emit 1501 alphas per minute.

<table>
<thead>
<tr>
<th>Isotope</th>
<th>Activity (dpm per milligram)</th>
</tr>
</thead>
<tbody>
<tr>
<td>U-238</td>
<td>733.6</td>
</tr>
<tr>
<td>U-234</td>
<td>733.6</td>
</tr>
<tr>
<td>U-235</td>
<td>33.7</td>
</tr>
</tbody>
</table>

also $U-235/U-238 = 0.711$ weight percent (Hyde et al. 1964) or an atom ratio of $U-238/U-235 = 137.8$ (Hyde et al. 1964). The specific activity for Th-232 is 246 dis/min/mg. (Hyde et al. 1964). Thus the Th-232 series is roughly one third as active as the U-238 series, but in an average rock the amount of thorium is roughly 4 times that of uranium so the contributions are comparable.

The theoretical alpha particle energy spectra for equilibrium conditions are plotted as relative intensities in Figures 2, 3, and 4 for the U-238, Th-232, and U-235 series respectively. For each figure the amount of parent is assumed to be 100%. A theoretical spectrum combining both the U-238 and Th-232 series is shown in Figure 5. This spectrum represents that expected for a sample with a Th/U ratio = 3.5. Here the parent U-238 is taken as 100% and all isotopes of both series are relative to the U-238.
ALPHA PARTICLE ENERGY SPECTRUM
U-238 DECAY SERIES IN EQUILIBRIUM.

PEAK MEV ISOTOPE
1  4.13  U238
2  4.18  U238
3  4.59  RA226
4  4.62  TH230
5  4.68  TH230
6  4.72  U234
7  4.77  U234
8  4.78  RA226
9  5.30  P0210
10 5.48  RN222
11 6.00  P0218
12 7.68  P0214

ENERGY (MEV)

RELATIVE INTENSITY (%) 0 20 40 60 80 100

Figure 2
ALPHA PARTICLE ENERGY SPECTRUM
TH-232 DECAY SERIES IN EQUILIBRIUM.

PEAK MEV ISOTOPE
1 3.95 TH232
2 4.01 TH232
3 5.34 TH228
4 5.42 TH228
5 5.45 RA224
6 5.68 RA224
7 6.04 BI212
8 6.09 BI212
9 6.28 RN220
10 6.78 PO216
11 8.78 PO212

Figure 3
ALPHA PARTICLE ENERGY SPECTRUM
U-235 DECAY SERIES IN EQUILIBRIUM.

<table>
<thead>
<tr>
<th>PEAK MEV</th>
<th>ISOTOPE</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>4.22</td>
</tr>
<tr>
<td>2</td>
<td>4.32</td>
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<tr>
<td>3</td>
<td>4.37</td>
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<td>4</td>
<td>4.40</td>
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<td>5</td>
<td>4.42</td>
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<tr>
<td>6</td>
<td>4.56</td>
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<tr>
<td>7</td>
<td>4.60</td>
</tr>
<tr>
<td>8</td>
<td>4.73</td>
</tr>
<tr>
<td>9</td>
<td>4.94</td>
</tr>
<tr>
<td>10</td>
<td>5.01</td>
</tr>
<tr>
<td>11</td>
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<td>13</td>
<td>5.54</td>
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</tr>
<tr>
<td>28</td>
<td>6.82</td>
</tr>
<tr>
<td>29</td>
<td>7.38</td>
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</tbody>
</table>

Figure 4
THORIUM DECAY SERIES

PEAK MEV ISOTOPE
1  3.95  TH232
2  4.01  TH232
12 5.34  TH228
13 5.42  TH228
14 5.45  RA224
16 5.68  RA224
18 6.04  BI212
19 6.09  BI212
20 6.28  RN220
21 6.78  PO216
23 8.78  PO212

ALPHA PARTICLE ENERGY SPECTRUM

THORIUM/URANIUM RATIO=3.5

URANIUM DECAY SERIES

PEAK MEV ISOTOPE
3  4.13  U238
4  4.18  U238
5  4.59  RA226
6  4.62  TH230
7  4.68  TH230
8  4.72  U234
9  4.77  U234
10 4.78  RA226
11 5.30  PO210
15 5.48  RN222
17 6.00  PO218
22 7.68  PO214

Figure 5
The highest peaks in Figure 5 (#20 and 21) are from the Th-232 series as expected since the ratio of activities of U-238/Th-232 is 733.6/246.0 and the Th/U ratio is 3.5, slightly larger than the activity ratio. A situation more representative of a uranium ore sample with a ratio of Th/U = 1.0 is shown in Figure 6. Most uranium ore contains a certain amount of thorium, but the average ratio for rocks mentioned earlier does not hold for the anomalous conditions of an ore body.

Armed with the knowledge of the theoretical alpha particle energy spectra, the actual measurement of the spectra can now be considered.

2.2 The Range of Alpha Particles

The interaction of alpha particles with matter causes a loss in energy which is related to the amount of material through which the alpha particle travels. For any given material, and a given initial energy for the alpha particle there is a particular range or mean free path. The range-energy relationship for alpha particles originally formulated by Geiger in 1910 as a range-velocity relation is described by Glasstone (1967) as:

\[ R = 3.09 E^{3/2} \]  

where \( R \) = range in air in centimeters, and \( E \) = energy
Figure 6
in Mev. The following paragraph is a quote from Glasstone (1967) p.252 "Actually, the Geiger formula and its equivalent equations are approximate, at best, and in any event they are applicable only for alpha particles with ranges from 3 to 7 cm. of air. (This range is equivalent to 4.0 Mev to 8.0 Mev energies - author) At lower ranges R is approximately proportional to $v^{3/2}$ and $E^{3/4}$, and at higher ranges to $v^4$ and $E^2$ "(where $v =$ velocity).

Since the alpha particles which are emitted from the natural decay series are in the range of 4.0 to 8.8 Mev, the best relation is that of equation (2.1). This relation is for range in air. The Bragg-Kleeman relation (Lapp and Andrews, 1963) is an approximate expression for range of alpha particles in solids:

$$R_s = \frac{3.2 \times 10^{-4} \, RA^{1/2}}{\rho} \quad [2.2]$$

where $R_s =$ range in the solid of density $\rho$, and of mass number $A$. $R$ is the range in air. Discussion of the accuracy of this equation has been given by Nogami and Hurley (1948), Beharrel (1949), Yagoda (1949), Whaling (1958), and Cherry (1963), and seems to range from claims of 5% accuracy to 25%.

For a compound or mixture of elements the effective atomic weight is obtained from the square of the sum of the square roots of the individual elements. The summation is a weighted sum using the percentages of the elements.
present for the weighting factor. (Lapp and Andrews, 1963)

2.3 Thin Source Alpha Particle Spectrometry

The shape of the energy distribution of a monoenergetic alpha particle being emitted from a source will be dependent upon the amount of material through which it passes. From equation (2.1) it can be seen that there is a given range for the given alpha particle energy. If the thickness of the source layer through which it travels is greater than the range of the alpha particle, it will not be emitted from the source at all, but it will be totally absorbed. For source thickness less than the range of alpha particles, it will be emitted but with an energy less than its initial energy. In a homogeneous distribution of alpha emitters in the source, the alphas originating at the surface are unaffected and are able to reach the detector with no energy loss. As a result, a continuous distribution of alpha energies from zero up to the initial energy of emission, are recorded. This complex 'tail' or energy distribution will have a shape that is dependent on the source thickness. To simplify the determination of the alpha particle energy, and amount of radionuclease in the source, an 'infinitely' thin source would be desirable. This would approach the theoretical 'spike' shaped energy spectra shown in Figures 2 to 6. Unfortunately, for comparison and quanti-
tative analysis, the thickness of the 'infinitely' thin source must be reproducible and uniform. Methods such as vacuum sublimation from a hot filament, electrospraying, and electrodeposition are a few of the source preparation techniques. Koralev and Kocharov (1958) prepare sources of 50 micrograms per cm$^2$ by this latter method. Hill (1961) crushes the source to maximum particle size of one micron and sprays the sample onto a cellulose sheet for entry in an ionization chamber. Mayneord and Hill (1959) use this technique in application to biological material. Baranov and Zelenkov (1959) describe the use of scintillation spectrometers, pulsed ionization chambers and magnetic alpha spectrometers in thin source alpha particle studies. Deal and Chanda (1969) describe a method involving the extraction of the radioactive isotopes by a chelate which is selective in its extraction properties for heavy elements. The source is evaporated from a solution onto a disk. Facchini et al (1956) use a thin source technique for alpha spectrometry with an ionization chamber to analyze uranium and thorium minerals.

The source preparation in all thin source alpha spectrometry is rather elaborate, and the loss of short lived daughters in preparation could be a problem in studying the state of radioactive equilibrium of the sample. In addition the count rate from thin sources is usually very low due to the very small amount of sample involved.
2.4 Thick Source Alpha Particle Spectrometry

Although thick sources have the advantage of a minimum of source preparation, they have not been widely used because of the effect of the energy distribution at lower energies than the initial peak energy of the alpha particle. Before the development of high resolution instrumentation, thick sources were utilized in counting the total alpha activity of the decay series, with no discrimination between the various particle energies. Some interesting work with thick sources was done by Evans (1934) and by Finney and Evans (1935) who determined the thorium content of geological samples by studying absorption of alphas and assuming radioactive equilibrium. Keevil and Grasham (1943) computed the number of alphas emitted from thin and thick sources for given geometries, which for analysis requires a knowledge of the amount of one decay series to compute the amount of the other. One of the earliest attempts to obtain an alpha particle assay of uranium and thorium radioactive ores is described by Peirson (1951). The method involved an ionization chamber for a detector and a pulse height discrimination circuit which could be varied. This represented an improvement over earlier absorption measurement techniques. Kulp et al (1952) describes two types of scintillation counters representing improvements on previous alpha counting techniques for both thin and thick sources.
Chudacek (1958) derives an expression for the energy spectrum of alpha particles emitted from sources of varying thicknesses. Since this is particularly relevant to the present study, the equation and technique were investigated thoroughly in relation to the energy spectra obtained from radioactive ore samples. It was found that the distribution given by Chudacek (1958) is more applicable to results from a spectrometer of extremely poor resolution. The energy distribution is representative of the envelope of all alpha particles emitted by the source, and does not yield information on the shape of the individual distributions from each isotope as they appear in an experimental spectrum obtained from a high resolution spectrometer such as the one used in this study.

An expression for the differential energy alpha spectrum from an infinitely thick source was derived by Graeffe and Nurmia (1961). This was also investigated in detail by the author, and although the resulting spectrum computed for an entire decay series was better than that described above, it did not fit the experimental spectra within satisfactory limits. This is possibly a result of the uncertainty of the stopping cross sections which Graeffe and Nurmia (1961) themselves admit.

Abrosimov and Kocharov (1962) discuss the shape of the energy distribution of alpha particles for varying
source thickness. Application of their theory to the particular geometry and detecting system used for this study, led to the development of energy distributions for the entire U-238 and Th-232 decay series which closely matched the experimental results. The application of the equations developed by Abrosimov and Kocarov (1962) to the present study is discussed in detail in a later chapter.
CHAPTER 3

SEMICONDUCTOR PARTICLE DETECTORS

3.1 Introduction

Since the semiconductor particle detector has only come into use relatively recently, a more detailed discussion than usual is warranted. This section deals with a review of the development history of the semiconductor detector, its principle of operation, factors affecting its performance, and finally the selection of a detector for the present application.

3.2 Historical Summary

The earliest work on the use of single crystals for particle detection began in about 1945. It was only in 1949 that sufficiently pure germanium and silicon became available for experimentation on particle detection. By 1962 considerable progress had been made and intensive development of particle detectors had begun in laboratories in many countries (Miller et al, 1962). The International Atomic Energy Agency in 1962 published a bibliography on semiconductor particle detectors comprising abstracts for 612 papers on the subject up to May of that year.
By 1966 the emphasis on semiconductor particle detectors had begun to swing from their development and improvement to their application in nuclear spectroscopy. Hollander and Perlman (1966) discussed this new revolution in nuclear radiation counting with the object of introducing the semiconductor detectors to nuclear spectroscopists. They did this by comparing results obtained by the new technique to results by the old techniques.

Tavendale (1967) reviewed the state of detector technology and performance up to February 1967. The need for still further improvements was pointed out, in particular in relation to the efficiency of these solid state detectors. Coche (1968) states that ".....research on detectors having a higher efficiency than lithium drifted germanium and which are able to operate at room temperature.......is not yet very far advanced."

The main feature which has spurred interest in the development of semiconductor detectors is their extremely high resolution. A review of the physics of semiconductor particle detectors will clarify this and other advantages that these detectors have.

3.3 The Physics of Semiconductor Particle Detectors

Semiconductors, intrinsic and extrinsic, and the band theory of solids is well covered by such authors as Kittel (1968), and by Dekker (1959). In particular
the physics of silicon surface barrier detectors, such as that used in this project, will be discussed.

A silicon p-n junction radiation detector is shown in Figure 7, after Miller (1968). The detector consists of a thin conducting gold layer front surface, a thin n-type region, and a thick p-type region. When a bias voltage is applied, the electrons and holes of the n-type and p-type semiconductors respectively, move away from the junction, leaving a depleted region. When an incident particle passes through the depleted region it produces electron-hole pairs, losing 3.6 electron volts for each pair produced in silicon and 2.9 e.v. for germanium. If the depleted region is sufficiently thick to entirely stop the incident particle, then the number of electron-hole pairs will be representative of the energy of the incident particle except for that portion lost in passing through the extremely thin gold surface and n-type layers. If the bias voltage is sufficiently high these electrons and holes will be swept out of the depleted region before they can recombine. This then represents the charge pulse which is fed to a charge-sensitive preamplifier for entry into the pulse height analysis system.

The high resolution possible with semiconductor detectors results from the fact that only a small energy (3.6 e.v.) is required to form an electron-hole pair,
Silicon p-n junction radiation detector (after Miller, 1968)

Figure 7
compared to about 30 e.v. to form an ion pair in a
gaseous ionization chamber, and about 350 e.v. for a
photoelectron in a scintillation detector (Miller, 1968).
Another advantage of the semiconductor detector is that
its sensitive region (the depletion region) can be varied
to accomodate particles of higher energies, simply by
increasing the bias voltage and moving the electrons
and holes farther from the junction. A full discussion
of the advantages of semiconductor detectors can be
found in Taylor (1963), Dearnaley and Northrop (1963),
and in Bertolini and Coche (1968).

3.4 Selection of a Detector for this Study

The choice of a detector was dependent upon
numerous factors such as (a) the expected count rates
for the samples to be studied, (b) the resolution required
to separate the peaks of interest in this application,
(c) the financial aspect, namely the cost of the detector
to be chosen.

Ideally a detector with a very large area would
be chosen to give maximum count rate since the activity
of most rock samples was not expected to be high. Un-
fortunately choosing maximum area is incompatible with
also choosing highest resolution since the detector noise
is proportional to area (Ortec Inc., 1970).

Selection of the required depletion depth was
determined from curves of alpha particle energy against range in microns of silicon. Since the highest energy alpha particle in this study has an energy of 8.8 Mev, a depletion depth of 60 microns would be sufficient. The required bias voltage to produce this depletion depth was determined from a nomogram supplied by Ortec Inc. A bias of 80 volts would produce a depletion depth of 100 microns, thus guaranteeing the complete absorption of the alpha particle in the detector.

The specifications for the silicon surface barrier detector chosen for this study are given in the next chapter along with descriptions of associated electronic instrumentation.
CHAPTER 4

INSTRUMENTATION

4.1 Introduction

A silicon semiconductor detector was used in conjunction with a multichannel pulse height analyzer to obtain the alpha particle spectra. Pulses from the detector passed through a low noise preamplifier and amplifier before entering the pulse height analyzer. A separate bias supply produced the required depletion region in the detector. The detector and sample were enclosed in a vacuum chamber. A thermocouple vacuum gauge was used for vacuum measurements. A combination mechanical pump and diffusion pump provided the vacuum for the chamber. A block diagram of the instrumentation is shown in Figure 8, and a photo of the experimental setup is given in Figure 9.

4.2 The Detector

An n-type silicon surface barrier detector (Ortec Model BA-070-950-100) with an active area of 950 mm$^2$ was mounted in the top of the vacuum chamber, sensitive side downward. The detector had a minimum
Figure 9. Experimental Arrangement

<table>
<thead>
<tr>
<th>Symbol</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>F</td>
<td>A to D converter</td>
</tr>
<tr>
<td>PTP</td>
<td>paper tape punch</td>
</tr>
<tr>
<td>T</td>
<td>thermocouple vacuum gauge</td>
</tr>
<tr>
<td>C</td>
<td>vacuum chamber</td>
</tr>
<tr>
<td>M</td>
<td>memory unit</td>
</tr>
<tr>
<td>PA</td>
<td>preamplifier</td>
</tr>
<tr>
<td>X-Y</td>
<td>XY recorder</td>
</tr>
<tr>
<td>PLSR</td>
<td>pulser</td>
</tr>
<tr>
<td>R</td>
<td>readout unit</td>
</tr>
<tr>
<td>AMP</td>
<td>amplifier</td>
</tr>
<tr>
<td>BIAS</td>
<td>detector bias</td>
</tr>
<tr>
<td>CHT</td>
<td>oscilloscope</td>
</tr>
</tbody>
</table>
Figure 3. Experimental arrangement

- P- to converter
- T- paper tape punch
- H- thermocouple vacuum gauge
- C- vacuum chamber
- M- memory unit
- R- readout unit
- PA- preamplifier
- AMP- amplifier
- XY- XY recorder
- BIAS- detector bias
- PUL- pulser
- OSCI- oscilloscope
sensitive depth of 100 microns when operated with a bias of 80 volts.

The manufacturer's (Ortec Inc.) alpha resolution measurement is "the full width half maximum (FWHM) of a 5.5 MeV thin Am-241 alpha source spectrum line, measured with detector and source in vacuum at the stated bias voltage, and includes the noise contribution of an Ortec amplifier system". The manufacturer's guaranteed alpha resolution was 70 keV,FWMH, but by actual measurement was 54.4 keV,FWMH. Noise width was guaranteed at 65 keV, FWHM, and by actual measurement was 40 keV,FWHM. The gold front surface plating was rated at 81 micrograms/cm² representing about 10 keV energy loss for alpha particles. Reverse current is 0.95 microamps at a bias of 80 volts.

The gold plated front surface of the detector is guaranteed to produce less than 20 keV energy loss from a 5.5 MeV alpha particle. This is a manufacturer's upper limit. Fabri et al (1967) conclude that a gold layer of 50 to 100 microgram/cm² contributes about 10-14 keV to the FWHM of a 6 MeV alpha particle.

4.3 Detector Bias Supply

An Ortec Model 428 Detector Bias Supply, capable of providing bias of either polarity up to 1000 volts for two detectors was used. Noise and ripple specifications rate noise and ripple at less than 0.0002%. Reverse
current monitor jacks are provided and reverse current was observed while applying bias to the detector in order to be able to detect any possible catastrophic breakdown or abnormal increase in reverse detector current. A positive bias of 180 volts was applied, 100 volts across a protective loading resistor and 80 volts being across the detector. In order to protect the detector, bias was increased slowly allowing reverse current to decrease before increasing bias again. The detector bias connection was made through the preamplifier allowing a single cable connection to the detector assembly for both signal and bias.

Reverse current is due to leakage conductance of the reverse biased detector. This was rated at 0.95 microamps for the rated bias voltage. A Fluke electronic galvanometer was connected to monitor outlets on the bias supply in order to monitor reverse current.

4.4. Preamplifier and Amplifier

An Ortec Model 109A preamplifier was chosen because of its low noise characteristics achieved through a field effect transistor input stage. The preamplifier is located physically close to the detector (about 10 inches). This minimizes the noise caused by the detector-preamplifier cable. The charge sensitivity is 150 mV/Mev (Si) in the X 10 gain position and 15 mV/Mev (Si) in the X 1 gain
position. Test pulses can be inserted at a test pulse connection for calibration purposes. The preamplifier inverts the negative output signal of the n-type Si detector, and feeds it via a cable driver through a cable connected to the remotely located main amplifier. An Ortec Model 435A active filter amplifier was chosen as a high performance instrument with high resolution and wide gain range. The active filter elements provide semi-gaussian pulse shaping resulting in superior resolving time capabilities without increasing the noise of the system. Noise is 10 micro volts at maximum gain of 1800.

Both amplifier and preamplifier have maximum counting rates which are much higher than any encountered in this project.

4.5 Power Supply

A combination Ortec Model 401/402 system modular bin and power supply furnishes standard regulated D.C. power supply voltages to the bias supply, preamplifier, amplifier, and pulse generator. The following are the manufacturer's specifications:
Regulation: less than 0.05% for line voltage variations of ± 10% or 100% changes in rated load. Ripple and noise: less than 3 mV peak to peak. Stability: 0.05%/ 8 hours after warmup.
4.6 Pulse Generator

A modular pulse generator (Ortec Model 480) provides simulated alpha particle detections which can be fed into the test pulse connectors on the preamplifier. A calibration control enables the unit to be adjusted to read directly in terms of alpha particle energy in Mev. Pulse frequency is that of the A.C. line. The Pulse generator can be calibrated using a single radioactive source. It then becomes equivalent to multiple monoenergetic sources for calibration of the pulse height analyzer.

4.7 Pulse Height Analyzer

The pulse height analyzer consists of three units, all of which are Nuclear Data equipment. These are the ND 181 P analog to digital converter, the ND 181 M 1024 Channel memory unit, and the ND 181 R readout control unit. Readout accessories are described in section 4.8.

4.7.1 Analog to Digital Converter (F unit)

This unit contains a linear amplifier, a Wilkinson-type analog to digital converter (Chase, 1961), various discriminator circuits, live time clock circuitry, logic circuitry, power supply and driver units for front panel address and count indicator lamps. The internal linear amplifier was not used in this project as the 435A
amplifier was considered a more advanced lower noise design. During pulse height analysis, the F unit generates a number of channel advance pulses that is proportional to the amplitude of the pulse. The M unit counts these pulses using the result to index a storage channel in the memory. There a count is either added or subtracted from the indexed channel as selected by the operator.

4.7.2 The Memory Unit (M unit)

The M unit is basically a digital computer containing a channel scaler, a memory register, a 1024 word magnetic core memory, memory drive circuits, a digital to analog converter, and various logic circuits. The unit totalizes (max. 1 million per channel) and stores data supplied by the F unit or some other external source such as the paper tape reader. It also performs digital to analog conversion of stored data for readout to external devices such as C.R.T. or XY recorder.

4.7.3 The Readout Control Unit (R unit)

This unit provides a selection of digital and analog readouts to external devices in addition to data reduction and integration (summation) capabilities. The R unit provides logarithmic readout with decade lines for either a CRT or XY recorder in addition to linear display on the same devices. Also included are circuitry
to operate a Tally paper tape perforator for output data and a paper tape reader for input data. Typewriter read-out is also available but not used in this system.

Data reduction capability allows the operator to multiply data contained in the second half of the memory by a constant with the result being added to or subtracted from data in the first half. Thus, for example, a 100 hour background stored in the second half could be multiplied by 0.25 and subtracted from a 25 hour spectrum stored in the first half. Since the entire 1024 channel was utilized for each spectrum in this project, data reduction was done later by computer. Data integration is also possible i.e. the sum of the counts in any number of selected adjacent channels can be obtained. The sum can be multiplied by a constant as mentioned above. Maximum summation time is 1.0 second.

4.8 Data Readout Accessories

4.8.1 The C.R.T. Display

A Nuclear Data ND 410 Display Monitor provides visual analog representation of the stored data on a 5" diameter C.R.T.. Logarithmic and linear displays are available. The stored data may be displayed in halves or quarters if desired or overlapping of halves or quarters for comparison of spectra. Every sixteenth channel displayed is an intensified dot to facilitate counting
channels on the visual display. Exact channel location of peaks is possible from the screen without going to digital readout on the F unit's address lights panel.

4.8.2  The XY Recorder

The Houston Omnipgraphic model HR 97 XY recorder provides a more permanent display for filing or graphical measurements than the C.R.T.. Paper is 11" x 17" and is held flat by vacuum. Log and linear plots are available. Decade lines are provided with the log plot. The XY recorder normally operates as a point plotter, but can be made to operate as a line plotter, joining the points.

4.8.3  The Paper Tape Unit

A Tally model 420 paper tape perforator combined with a model 424 paper tape reader provides digital output for input to a computer for data analysis. The tape perforator operates at 60 characters per second and punches the complete 1024 channel of data in less than two minutes. The data is presented in a 4 line B.C.D. (binary-coded-decimal) format. The paper tape is punched in an eight level IBM computer compatible form. Levels 1-4 present the data. Level 5 is for space and odd parity check. Carriage return is represented by the second, third, fourth, fifth, and seventh levels; zero by the sixth level; and end of message by the eighth level.
Thus each channel is represented by a six digit B.C.D. number, followed by a space, and every eighth number is followed by a carriage return. At the end of the tape, after all the data is read out, an end of message is punched. The paper tape is taken to the computing center and is read by a PDP-10 computer's optical paper tape reader. Since the data format is not exactly compatible with the computer, a special program was written to read and convert it for use. (See Appendix D) It is then stored on magnetic tape and is ready for handling by the data analysis programs.

The paper tape reader allows the operator to input to the memory data previously stored on paper tape, for purposes of data reduction, integration or re-display by C.R.T. and XY recorder. For example, by reading in 1024 channels in subtract mode, a background may be subtracted from a 1024 channel spectrum already in storage. Paper tape is read at a rate of 60 characters per second.

4.9 The Vacuum System

In order to prevent interactions with air molecules and consequent loss of energy of alpha particles in travelling from the rock source to the detector, both source and detector must be enclosed in a vacuum. A combination mechanical pump-diffusion pump assembly (Welch Scientific #1392) was chosen to provide sufficiently
high vacuum to eliminate alpha particle energy loss, to work outside the critical pressure region between 1 micron and 50 microns of Hg where the problem of breakdown on insulators, etc. is most acute, and to provide the required vacuum in a short time. A block diagram of the vacuum system is shown in Figure 10, and a photo of the experimental arrangement is shown in Figure 11.

Ultimate pressure is 0.001 millitorr (microns Hg.). The mechanical pump is used as a 'backing' or 'roughing' pump to reduce the pressure to a level at which the diffusion pump can operate i.e. at least 0.1 torr. The diffusion pump operates with electrically heated Octoil, and is water cooled at a rate of 0.04 gal./minute. The system was designed to have a minimum of piping between the pumps and the chamber to maintain maximum pumping rates. Valves are installed in appropriate positions to allow the vacuum chamber to be isolated for sample changing without shutting down the vacuum system. A sample is placed on the sample holder in the Ortec Model 805 stainless steel chamber, the vacuum release valve A is closed, and valves C and D are closed to isolate the diffusion pump. Valve B is opened and the chamber and pipes are then pumped down to .1 torr or less. Then valve D is opened and the mechanical pump continues to provide 'backing' for the diffusion pump. Finally B is closed
THE VACUUM SYSTEM

Figure 10
Figure 11. Apparatus for Vacuum System

C - vacuum chamber
D - semiconductor detector
H - sample holder
PA - preamplifier
T - thermocouple gauge
I - ionization gauge
Figure 11. Apparatus for Vacuum System

T - vacuum chamber
H - semiconductor detector
M - sample holder
PA - preamplifier
I - thermocouple gauge
I - ionization gauge
and C is opened so that the system is in its normal operating configuration. The system design is based on descriptions of vacuum systems in Guthrie (1963). A thermocouple vacuum gauge located between B and C measures the vacuum in the chamber down to $10^{-3}$ torr. Below this an ionization gauge must be used. A connection designed for insertion of the ionization gauge is located at E in Figure 10. This is usually kept plugged, and is not used except if the vacuum system appears to be changing its characteristics such as increasing pump-down time. In the early setting up of the vacuum system, the ionization gauge was used to insure the system was tight and free of leaks. The system achieves a vacuum of better than $10^{-3}$ torr in about 2 minutes after a sample has been changed; when this pressure is reached the bias voltage can be applied to the detector located at the top of the chamber and the counting can be started.
CHAPTER 5
RADIOACTIVE EQUILIBRIUM AND ALPHA SPECTROMETRY

5.1 Introduction

Rosholt (1958) subdivided the U-238 decay series into five distinct groups for equilibrium studies. These consist of the uranium group, Th-230 alone, Ra-226 alone, the Rn-222 group, and the Pb-210 group. (See Figure 1, Chapter 1) The uranium group contains U-238 and its immediate daughters up to and including U-234. The Rn-222 group contains Rn-222 and its immediate daughters up to and including Po-214. The Pb-210 group contains the remaining daughters in the series. When the relative abundances of each group is known, the extent of disequilibrium is completely defined. Each group can be assumed to be in equilibrium as a unit, but disequilibrium may exist between groups.

The usual method of studying equilibrium involves radiochemical analysis, thin-source alpha spectrometry, gamma-ray spectrometry, or a combination of several of these techniques. A review of previous investigations is outlined below.
5.2 Previous Investigations

The most pertinent investigations on uranium analyses and radioactive disequilibrium relating to the present study are summarized below.


Adams et al (1958) determined the thorium and uranium contents in sedimentary rocks by a combination of gamma-ray spectral analysis, total alpha activity counting, and fluorometric uranium determinations. Bunker and Bush (1966) described a combination of graphical and mathematical interpretation techniques applied
to gamma-ray spectrometry to analyze for uranium, thorium, and radium. Stern and Stieff (1959) discussed some analyses of Ra-226 and uranium in Colorado Plateau secondary uranium ores.

Mero (1960) described a method of determination of the presence of equilibrium by the use of gamma-ray spectrometry, involving comparison of gamma count rates at 0.190 Mev and 0.240 Mev. Coquema et al (1963) described a combination radiochemical analysis and alpha, beta, and gamma counting technique used to study radioactive disequilibrium and its relation to geochronology. Richardson (1964) studied drill cores of Conway granite of New Hampshire with a radiochemical separation, electrodeposition, and thin source alpha spectrometric analysis. The top 15 feet were deficient in U-234, another example of isotopic fractionation of uranium in nature. Somayajulu et al (1966) reported on the disequilibrium observed in the U-238 series in basalt samples from Hawaii, Japan, and Iwo Jima. Their technique was a combination of radiochemical separation and mass spectrometry.

A large proportion of the investigations on radioactive equilibrium in rocks have been carried out by Rosholt of the United States Geological Survey. Rosholt (1954,1957) described quantitative radiochemical methods for determining the sources of natural radioactivity.
Rosholt (1958) discussed the natural migration of uranium and its decay products, and outlined the five groups mentioned in the introduction above. In all of Rosholt's papers, the method used is radiochemical separation of the isotope to be studied, followed by analysis by thin source alpha spectrometry. Rosholt (1959) discussed the geochemical processes affecting ore deposits and their relation to radioactive disequilibrium. His studies indicated six basic types of disequilibrium can occur in present-day radioactive deposits, although since he is primarily concerned with sedimentary-type deposits, the possibility exists of finding new types of disequilibrium in igneous deposits.

Rosholt (1960) described uranium migration in sandstone-type ore deposits. In another paper Rosholt and Dooley (1960) described the radiochemical analysis technique which they improved since the early work of Rosholt (1954). Rosholt (1961a), Robinson and Rosholt (1961) and Rosholt (1961b) elaborated on uranium migration in sandstones as studied above, relating the amount of disequilibrium to the apparent date of migration. Rosholt et al (1963) described isotopic fractionation of uranium in sandstone. The surprising result of U-234 being leached preferentially to U-238 is considered due in part to changes in oxidation states. Pierce and Rosholt (1961) discussed isotopic disequilibrium in uranium-bearing
asphaltite nodules in dolomite. Dooley et al (1964) reported on radioactive disequilibrium in roll features of the uranium deposits of Shirley Basin, Wyoming, using Rosholt's technique.

Rosholt et al (1964) reported fractionation of uranium isotopes and daughter products in weathered granite and uranium-bearing sandstone in the Wind River Basin of Wyoming. Ratios of U-235/U-234 and of U-234/U-238 were determined with a mass spectrometer. Rosholt, Butler, Garner, and Shields (1965) discussed U-234/U-238 ratios in Wyoming and in Colorado uranium ores. Deficiencies of U-234 of 40 to 60% in the ores were reported. The date of migration of the U-234 in the above mentioned areas was discussed by Rosholt, Tatsumoto, and Dooley (1965). Rosholt and Ferreira (1965) discussed U-234 deficiencies of 7 to 29% in samples from the Wentz mine in Wyoming, in relation to the water table.

5.3 Possible Locations of Disequilibrium in Alpha Spectra

Since the most exhaustive studies of equilibrium in rocks have been done by Rosholt and his co-workers, the classification of disequilibrium patterns outlined by Rosholt (1959) will most likely contain any cases to be determined in the present study. The first three types have U-238 in excess of the equilibrium amount which would be required to support the Ra-226 present.

Type 1. Daughter product deficiency U > Pa-231 >
Th-230 $\rightarrow$ Ra-226.

Type 2. Time related daughter product deficiency. This is a special case of type 1 wherein the amount of daughters present all require the same amount of time to be produced from the parent uranium. That is, it is a stage in time before equilibrium has been established since the initiation of decay of the U-238.

Type 3. Th-230 deficiency. The last three types have Ra-226 in excess of the amount that would be present in equilibrium with the U-238 present.

Type 4. Daughter product excess, represented by a low uranium content. Portions of the decay series may have daughters in equilibrium with each other.

Type 5. Ra-226 excess.

Type 6. Only Ra-226 and its daughter products are present. Often Ra-228, a daughter of Th-232 is also present.

5.4 Discussion of Possible Causes of Disequilibrium

The following discussion is a summary of the postulations made by Rosholt (1959) to explain some of the causes of the 6 types of disequilibrium.

Type 1 and 2 ores are formed when the uranium migrates to its present location at a time less than that required by its daughter products to reach approximate equilibrium i.e. less than 300,000 years ago. Alternatively,
there could be preferentially greater leaching of daughter products than of uranium. This second possibility occurs in carnottite and other deposits containing uranium fixative agents such as vanadium or phosphate. Type 1 ores are more likely formed in this manner.

Type 3 disequilibrium is believed due to deposition of uranium and other daughter products rather recently, without Th-230.

Type 4 is the result of leaching of uranium.

Type 5 disequilibrium occurs in pyritic ores and is the result of differential leaching. Sulphates formed in the rock retains the radium, whereas the sulphuric acid leaches and removes the uranium.

Type 6 disequilibrium is associated with oil-and-gas-field brines, and radioactive hot spring deposits. They are the result of precipitation of radium salts from large volumes of water.

5.5 Limitations of the Present Study

5.5.1 Energy Resolution of the System

An inherent limitation of the alpha spectrometric method is the inability to distinguish between radioactive isotopes having alphas of energies closer together than are resolvable by the spectrometer. The resolution of the silicon surface barrier detector used in this study is 55 KeV at about 5 Mev. This means that for example
the 4.77 Mev U-234 peak cannot be resolved from the 4.78 Mev Ra-226 peak, and the 4.68 Mev Th-230 peak cannot be distinguished from the 4.72 Mev U-234 peak. When a rock contains thorium and its decay series, the possibility of more combinations of unresolvable peaks results. The solution is a detecting system of higher resolution, but this necessitates going to a smaller area detector, resulting in a decreased count rate, and increased counting times.

In the case of a group of say 3 peaks, the presence of disequilibrium can still be detected, but which of the isotopes is the cause may be indeterminate. However, a study of other isotopes in the series may indicate which isotope in the group is the most likely to be the cause of the disequilibrium.

5.5.2 Low Intensity Alpha Peaks

When a particular alpha decay results in alpha particles of either of two different energies, usually alphas of one energy occur much more often than the other. For example Ra-226 emits an alpha of 4.78 Mev 94% of the time and a 4.59 Mev alpha 6% of the time. The 4.59 Mev alpha peak will be of such low intensity that it may be undetectable, but any disequilibrium due to Ra-226 will show up in the high intensity peak. The entire U-235 decay series is of low intensity relative to the U-238
series and consequently is undetectable without very long counting times.

5.5.3 Low Concentrations of Uranium in the Rock

A lower limit of detectability is also set by the uranium content of the rock. Since the volume of rock is extremely small (say 64 cm$^2$ area and less than 100 microns thick) the amount of uranium contributing alpha particles to the spectrum is extremely small. In practice it has been determined that a uranium content of less than 1% U$_3$O$_8$ presents difficulties and a lower limit of about 0.1% U$_3$O$_8$ is set by the length of counting time possible without significant shift in the electronic parameters of the instrumentation.

5.5.4 Counting Times and System Stability

As mentioned above, after a certain amount of time the inherent drift in the measuring instruments prevents a good spectrum from being formed. The main problem is a slight shift in gain with time which causes a loss in resolution and broadening of the spectral peaks. A time of 72 hours is the maximum possible counting time to still retain a relatively good spectrum. This is an upper limit achieved when temperature and humidity were relatively stable, and was not possible to obtain in the summer when cool nights and hot days affected the instrument, even
though it was in an air conditioned room.

5.6 Problems in the Analysis of Experimental Alpha Spectra

5.6.1 Smoothing the Statistical Scatter

In order to smooth the statistical scatter which is present in any spectrum, an appropriate smoothing function had to be chosen which would effectively filter out the scatter, yet retain the peaks present in the spectrum. The smoothing was necessary for comparison and display purposes, and was not a part of the equilibrium analysis itself. A running least squares polynomial smoothing function was chosen. A comparison of different smooths for an infinitely thick source spectrum computed using 5, 7, and 9 points with 2nd and 5th degree polynomials is shown in Figure 12. The smoothing function essentially fits a polynomial to the chosen number of points by the least squares method, computes the central value, and moves on one point further, repeating the least squares fit, and computing the next central value. This of course requires an odd number of points. The method is described by Savitzky and Golay (1964) for application to similar spectral data to that obtained in this study. A similar technique has recently been described by Wood and Hockens (1970). The 9 point smooth with a 2nd degree polynomial was chosen for comparison of alpha spectra. The smoothing program is given in Appendix B.
VARIATION IN SMOOTHING FOR TAPE 252 KATANGA 14857

Figure 12
5.6.2 Compensation for Gain and Threshold Shifts During Counting

During the counting time of a single spectrum, the gain of the system could be considered constant, while over the time of several spectra, an appreciable gain shift could occur. For comparison purposes, and for purposes of fitting theoretical spectra to experimental spectra, the shift had to be compensated by mathematically re-computing the spectrum for a different gain. For example, consider the drastic case of a spectrum which was made with a gain setting such that the energy width per channel was 11 Kev. To compare this with a spectrum with an energy per channel value of 10 Kev would be impossible. In the first case 6.66 Mev would fall in channel 600, while the second case 6.66 Mev would fall in channel 666. Usually there was no threshold shift (shift of the zero energy channel). A computer program by Schonfeld (1966) originally developed for gamma-ray spectra was modified to perform the required shifts in the alpha spectra. The modified version is given in Appendix C. An example of the use of the shifting program is shown in Figure 13. Five values of gain shift are used from 0.9800 to 1.0200, these being typical of the amount of shifting required in the experimental data obtained in this study. In Figure 13 the gain shift of 1.0000 represents no gain shift, i.e. the original spectrum. The shift of 1.01302 was the best
VARIATION IN GAIN SHIFT
FOR
50% U3O8 SYNTHETIC ROCK

ALL SPECTRA ARE NORMALIZED TO BOTTOM SPECTRUM
3 POINT SMOOTH WITH A POLYNOMIAL OF DEGREE 2

Figure 13
value of gain shift, and the dashed line through 7.68 Mev passes through the half slope of the 7.68 Mev peak.

5.6.3 Background Subtraction and Contamination

In some alpha decays a relatively large recoil energy is imparted to the isotope emitting the alpha particle. For this reason some radioactive contamination of the detector is possible, particularly from highly radioactive samples which cause a large number of recoil atoms to strike the detector.

Dooley et al (1964) reported a small background contamination of their detector by Ra-224, and Bi-212 from the Th-232 series and Ra-223 from the U-235 series which tend to accumulate on the detector.

The Ra-224 has energies 5.68(95%), 5.45(5%), & T_{1/2}=3.64 days.

Bi-212 has energies 6.09(10%), 6.04(25%), & T_{1/2}=60.5 mins.

Ra-223 has energies 5.54-5.75 low intensities T_{1/2}=11.2 days.

The relatively short half lives of the isotopes involved prevented any significant buildup on the detector.

Similar contamination of the detector was observed in this study, but this did not present a problem if time was allowed for decay of the contaminants between analyses. Even this was unnecessary except after counting extremely radioactive samples.

A special problem, however, arose when the detector used in this study was contaminated inadvertently
by recoil atoms of Po-210 from a polonium source used for energy calibration of the system. Due to inexperience the danger of contamination was not realized, and when the calibration source first arrived from the manufacturer, it was used for several days before a background count was taken. The presence of a large spike of 5.30 Mev was observed in the background—effectively an internal calibration source. The half life of Po-210 is 138.4 days, and it therefore became a relatively permanent feature of the background. Since infinitely thick sources were eventually used, this did not present much of a problem. The Po-210 peak showed as a narrow spike riding on the infinitely thick source spectrum. For rocks of moderate to high activity such as pitchblende samples, the background remained a negligible factor. For rocks of low activity this internal calibration source proved to be very useful in monitoring the gain, as small gain shifts were recorded as movement or broadening of the 5.30 Mev Po-210 peak. This was particularly important for spectra obtained over long counting times. These were in fact the only spectra in which the peak showed. It may be stated, however, that ideally the energy of the internal calibration source should lie outside the region of interest rather than in the middle of it.

The normal background due to the Po-210 contamination is shown on the top of Figure 14 as tape 185.
Examples of background obtained from various source contaminations.

Figure 14.
This shows the large spike of 5.30 Mev Po-210, and a small peak believed due to the 5.54 Mev and 5.61 Mev peaks of Ra-223 from the U-235 series. The peak is only 10 counts high with a counting time of 10 hours. The Po-210 peak which is truncated extends to slightly less than 600 counts, a count rate of under one count per minute in the highest count channel.

The background shown in the center of Figure 14 (tape 203) was taken after counting a weathered oxidized pitchblende sample (number 2824). This shows four new peaks introduced into the background, in addition to the 5.30 Mev Po-210, and the 5.54 + 5.61 Mev Ra-223. The Ra-223 has increased to a height of 30 counts in 10 hours of background counting. The 11.2 day half life means it will decay relatively slowly, and is usually present in the background in some small amount, as Dooley et al (1964) reported. A peak 30 counts high in 10 hours is insignificant in this study.

The four new peaks are located at approximately 6.0 Mev, 6.3 Mev, 6.6 Mev, and 7.7 Mev respectively. These can be explained as follows:

6.0 Mev This can be either the 6.00 Mev peak of Po-218 in the U-238 series with a half life of 3.05 minutes, or the 6.04 and 6.09 Mev peaks of Bi-212 from the Th-232 series. Since the previously counted rock showed no thorium contribution to
the spectrum, the latter possibility is discounted. Since Po-218 has a short half life, nearly all the counts under the 6.00 Mev peak were obtained in the first hour of counting time of the background.

6.3 Mev This is either the 6.28 Bi-211 peak from U-235 with half life of 2.16 minutes, or the 6.28 Mev Rn-220 peak from the Th-232 series. Again, since there is no indication of thorium in the previous spectrum, the latter possibility is eliminated.

6.6 Mev The only possibility is that of the 6.62 Mev Bi-211 peak from the U-235 series. This further substantiates the conclusions on the previous peak discussed.

7.7 Mev The only possibility is the 7.68 Mev Po-214 peak from the U-238 decay series. This is a daughter of the Po-218 peak discussed above. Although the Po-214 has a half life of 160 milliseconds, it is supported by beta emitters located between it and the Po-218. These have half lives of 26.8 minutes and 19.7 minutes.

The bottom of Figure 14 shows a 10 hour background (tape 222) which was taken after counting a spectrum of synthetic rock of 50% epoxy and silica sand and 50% \( \text{U}_3\text{O}_8 \) powder. This represents a more solid non-weathered rock, and the small peaks of less than 10 counts in 10
hours at 6.0 Mev (Po-218) and 7.68 Mev (Po-214) are the only background contamination produced.

As a final note on Figure 14, the variation in the statistical scatter from top to bottom in tape 185, 203, and 222 are the result of reducing counts to equivalent times of 600 minutes from original counting times of 72 hours, 10 hours, and 23 hours respectively.

An extreme example of the contamination possible from counting a thorium sample is shown in Figure 15. From top to bottom is shown four backgrounds taken consecutively, for 24 hours each. Tape 144 at the top was started immediately after counting the alpha spectrum of a sample of thorite from Bancroft, Ontario. The sample was weathered and porous in appearance. The two new peaks in the background are due to the 6.04 and 6.09 Mev peaks of Bi-212 in the Th-232 series ($T_{1/2}=60.5$ minutes) and its immediate daughter 8.78 Mev Po-212 ($T_{1/2}=0.30$ milli-seconds). It should be stressed that this represents an extreme contamination not normally observed after counting a thorium-rich sample. In general it can be stated that the only time a significant contamination occurs after counting either a uranium-rich or thorium-rich rock, is when the rock was porous and weathered. Even then, the contamination is very short lived. The only really significant background is the 5.30 Mev Po-210 peak, and it is not even necessary to subtract this from the spectrum.
Figure 15

Background decay shown by four successive counts.
as the method of analysis chosen is not affected by this background. It will only show up in the residual spectrum after the state of equilibrium has been determined for the rock sample.
CHAPTER 6
PRELIMINARY EXPERIMENTAL WORK

6.1 Introduction

Preliminary investigations were concerned with thin sources and the objective of developing a simple method of source preparation. Areas under the experimental energy distribution peaks were to be compared to determine if they were present in equilibrium proportions. Results indicated there was no simple source preparation technique for thin sources. The preliminary work was most useful in yielding an explanation for some of the peaking effects observed in a few of the infinitely thick source spectra obtained later when a final procedure was adopted.

6.2 Thin Sources
6.2.1 Collimating the Alpha Particles

Since one of the objectives of this research was to maintain simplicity of sample preparation to reduce overall time of an equilibrium analysis, a method of producing the same effect as an infinitely thin source through the use of collimation of the alpha particles
was attempted. The method consisted of producing a collimator by drilling a large number of very fine holes in a sheet of lucite, through which the alpha particles must pass to reach the detector. This would effectively permit only those alphas leaving the source within a certain measurable angle from the normal to the source to pass through the collimator. All others emitted at shallower angles would strike the walls of the collimating holes and be absorbed. The method depends on the theory that, of all the angles of emission from the surface, the vertical angle would contain the largest proportion of particles being emitted with the true initial energy of the alphas. (Ghiorso et al 1949).

This method would involve practically no sample preparation other than to cut the sample to a size to fit inside the vacuum chamber, and beneath the collimator. The method was tried on several samples of crushed uranium oxide (35%) and thorium oxide (34%), which were obtained from the Department of Energy, Mines and Resources in Ottawa. The resulting alpha spectra are shown in Figures 16 and 17. The large narrow peaks observed, although at first quite promising, were determined to be difficult to reproduce, appeared to be shifted in energy from the expected energies by an unknown non-linear amount, and finally took an inordinately long counting time to produce.

The results, however, are useful in explaining
Figure 17

TAPE NO. 23.
34% THO₂, COLLIMATED.
TIME (MIN.) 1000.0
some of the effects observed when counting infinitely thick sources of very porous samples, as is discussed in a later chapter.

6.2.2 Source Preparation

6.2.2.1 Powders

Since artificial methods of producing a thin source were not satisfactory, the relatively simple source preparation of crushing and powdering the sample to 5 or 10 micron size was attempted. The powdered sample (34% ThO₂) was spread thinly on the adhesive surface of a piece of masking tape and counted. The resulting spectrum is shown in Figure 18. This thin source was much too thick and in fact had the spectrum of the infinitely thick source finally adopted. Further attempts at preparation of extremely thin layers of the powders of the samples of 35% U₃O₈, 34% ThO₂ and both powders together are shown in Figures 19, 20, and 21 respectively. The results improved considerably but the energy distribution tails of the isotopes still interfered with each other. The spectra were also unfortunately un reproducible due to a lack of control over the source thickness. A study of the distribution as shown in Figure 20 indicates that the sources were probably approaching infinitely thick for the low energy alpha particles, while approaching infinitely thin for the highest energy alpha particles.
TAPE NO. 24.
35% U308 THIN POWDER.
TIME (MIN.) 1220.0

Figure 19
TAPE NO. 25
34% THO2 THIN POWDER
TIME (MIN.) 1425.0

ENERGY (MEV) 5.0
3.0
1.0
NO. OF COUNTS
TAPE NO. 26.
34% TH02 + 35% U308 POWDER
TIME (MIN.) 2880.0

Figure 21
A comparison of highest and lowest energy peaks shows the rate of fall off of the energy distribution of the 8.78 Mev alpha particle is more rapid than the 4.0 Mev alpha peak energy distribution.

So in fact the infinitely thin sources were "semi-ininitely thick", ie. they were thick for low energy alphas, and relatively thin for the high energy particles.

6.2.2.2 Thin Sections

An alpha particle energy spectrum of a thin section of a sample of pitchblende was obtained, and found to be infinitely thick. Further polishing of the thin section until it was barely visible resulted in no noticeable improvement. The resulting spectrum is shown in Figure 22, an example of an infinitely thick source spectrum.

6.2.2.3 Acetate Peels

Several other techniques were attempted, one of these being the acetate peel method. A cut surface of the rock is moistened with acetone, and a sheet of plastic is placed flush against the surface of the wet rock. The acetone partially dissolves the plastic which then adheres to the rock. When it has dried the plastic sheet is peeled from the rock, and in doing so it removes a very
TAPE NO. 21.
PITCHBLEND THIN SECTION
TIME (MIN.) 1080.0

Figure 22
thin layer of rock from the surface. This method also produced infinitely thick sources, and in addition would not be suitable for every rock since the grains of rock most easily removed from the rock would be those with the lowest strength or hardness, and thus in effect would not be a representative sample.

6.3 Increasing Concentration of Heavy Minerals

While conducting the preliminary work on thin sources, it was realized that rocks with low concentrations of uranium and thorium would take a very long counting time. Several methods of concentrating the uranium and thorium minerals while not disturbing the radioactive equilibrium were attempted.

6.3.1 Heavy Liquid Separation

The crushed sample was placed in a separating funnel which contained tetrabromoethene, a heavy liquid with specific gravity 3.42. The heavy minerals settled to the bottom and the lighter minerals floated to the top. This was rather slow and tedious since only a very small amount could be separated at a time. The heavy minerals were a very small percentage of the rock sample, and had a tendency to be 'rafted' to the surface with the huge bulk of the lighter minerals. Another difficulty was that the unwanted heavy minerals such as pyrite also
separated out. Tests had to be run to determine whether the heavy liquid had any effect on the radioactive equilibrium of the sample. Samples were counted before and after heavy liquid separation with no noticeable effect on the relative peak heights of infinitely thick sources.

6.3.2 The Super Panner

An attempt was made to utilize the 'super panner', an automated panning system similar to that used by gold prospectors panning for gold. By judicious adjustment of the flow of water, the angle of the pan, and the rate of movement, mineral separation could be accomplished. This was determined to be unsatisfactory and time consuming.

6.3.3 Autoradiography and Hand Picking 'Hot Spots'

Several autoradiographs were made of Elliot Lake conglomeratic uranium ore, in order to determine the location of the most radioactive areas on the rock surface. These were then chipped out with the use of a vibrator with a tungsten carbide tip. Although the method may be satisfactory to locate a large crystal of a radioactive mineral in a relatively barren ground mass, it was not satisfactory for such disseminated uranium mineralization as is present in the Elliot Lake conglomeratic ore.

Figure 23 shows an autoradiograph of Elliot Lake uranium
Figure 23. Autoradiographs of Elliot Lake Conglomeratic Uranium Ore (Positive), actual size.
ore. Since the photo is a positive print, the location of the radioactivity is indicated by light areas.

6.4 Discussion

The difficulty in obtaining reproducible spectra for the sources, along with the sample preparation time involved in producing a thin source led to the adoption of infinitely thick sources for the equilibrium studies. A theoretical infinitely thick source alpha particle energy spectrum was developed which closely matched the experimental spectra, eliminating any need to depend on comparisons to rocks believed to be in radioactive equilibrium.
CHAPTER 7

THE TECHNIQUE DEVELOPED FOR THE PRESENT EQUILIBRIUM STUDY

7.1 Introduction

The formula for the energy distribution of alpha particles from an infinitely thick source developed by Abrosimov and Kocharov (1962) discussed in Chapter 2 forms the basis for the development of an alpha particle energy spectrum for a complete radioactive decay series. The original equation depends upon a knowledge of the number of alpha particles emitted by the entire source per unit time \( N_0 \), the thickness of the source \( h \), and the constant of proportionality \( A \) from the range-energy equation:

\[
R = AE^N
\]  

[7.1]

where \( R \) = the range of the alpha particle and \( N \) is a constant equal to approximately 1.5 depending upon the effective atomic number of the source material.

Note: in equations [7.2] to [7.14], \( n \) is used for the constant \( N \) to distinguish it from alpha counts, \( N \).

The general formula for computing the number of alpha counts \( dN \) in an energy range \( dE \) of the energy
distribution is given by: (Abrosimov and Kocharov, 1962)

\[
\frac{dN}{dE} = \frac{N_0 A_n}{4\hbar} E^{n-1} \quad [7.2]
\]

7.2 The Theoretical Alpha Spectrum of an Entire Decay Series

Extending the above relation for the alpha particle energy distribution of a single radioisotope to include several isotopes of various initial energies and intensities is accomplished as follows:

The total count under a particular alpha particle energy distribution is the sum of all the counts of all energies below the maximum energy E_max, i.e. the initial energy of the alpha particle:

\[
\int_0^{E_{\text{max}}} dN = \int_0^{E_{\text{max}}} \frac{N_0 A_n}{4\hbar} E^{n-1} dE \quad [7.3]
\]

\[
N_{\text{total}} = \left[ \frac{N_0 A_n}{4\hbar} \right] \frac{E_{\text{max}}^n}{n} \quad [7.4]
\]

For a given peak we can arbitrarily determine its peak count rate but all others in the decay series will then have count rates relative to that chosen for the first peak, i.e. the relative total counts under the peak energy distributions will be determined by their relative in-
tensities, and ranges in the rock.

Consider peak 1 (which can be any peak)

\[
\frac{dN}{dE_{\text{max1}}} = \left[ \frac{N_0 A_n}{4h} \right]_1 E_{\text{max1}}^{n-1} \tag{7.5}
\]

Integrating eqn. [7.5] in the same way as [7.3] we get:

\[
N_{\text{tot1}} = \left[ \frac{N_0 A_n}{4h} \right]_1 \frac{E_{\text{max1}}^n}{n} \tag{7.6}
\]

Solving for the constant in eqn. [7.5]:

\[
\left[ \frac{N_0 A_n}{4h} \right]_1 = \frac{\frac{dN}{dE_{\text{max1}}}}{E_{\text{max1}}^{n-1}} \tag{7.7}
\]

Since \( \frac{dN}{dE_{\text{max1}}} \) is the count which we can arbitrarily choose in the largest energy increment (or channel in the case of a pulse height analyzer) containing the initial energy of the alpha particle (\( E_{\text{max1}} \)), we can solve for the constant on the left side of eqn. [7.7].

Then the entire distribution can be computed from eqn. [7.2]:

\[
\frac{dN}{dE_i} = \left[ \frac{N_0 A_n}{4h} \right]_1 E_i^{n-1} \tag{7.8}
\]
Now for the second peak we know that as in eqn. [7.6],

$$N_{\text{tot}2} = \left[ \frac{N_0 \text{An}}{4h} \right]_2 \frac{E_{\text{max}2}^n}{n} \quad [7.9]$$

Solving for the new constant in eqn. [7.9],

$$\left[ \frac{N_0 \text{An}}{4h} \right]_2 = \frac{N_{\text{tot}2}}{\left[ \frac{E_{\text{max}2}}{n} \right]^n} \quad [7.10]$$

The ratio of the total count under the two energy distributions is now dependent on the relative ranges, percentages, and activity of each isotope:

$$\frac{N_{\text{tot}2}}{N_{\text{tot}1}} = \frac{\text{Range of } 2}{\text{Range of } 1} \times \frac{\% \text{ of } 2}{\% \text{ of } 1} \times \frac{\text{Activity of } 2}{\text{Activity of } 1}$$

$$= R_{21} \quad [7.11]$$

Therefore substituting for $N_{\text{tot}2}$ from eqn. [7.11] into eqn. [7.10]

$$\left[ \frac{N_0 \text{An}}{4h} \right]_2 = \frac{N_{\text{tot}2}}{\left[ \frac{E_{\text{max}2}}{n} \right]^n} = \frac{R_{21} \left[ N_{\text{tot}1} \right]_n}{\left[ \frac{E_{\text{max}2}}{n} \right]^n}$$
and substituting for $N_{\text{tot1}}$ from eqn. [7.6] we get:

$$
= R_{21} \left[ \frac{N_0 A_n}{4\hbar} \right]_1 \left[ \frac{E_{\text{max1}}}{n} \right]_n \cdot \frac{n}{E_{\text{max2}}} 
$$

$$
= R_{21} \left[ \frac{N_0 A_n}{4\hbar} \right]_1 \left[ \frac{E_{\text{max1}}}{E_{\text{max2}}} \right]^n
$$

[7.12]

Substituting this value in eqn. [7.2], the energy distribution for the second peak is then:

$$
\frac{dN}{dE_1} = \left[ \frac{N_0 A_n}{4\hbar} \right]_2 E_1^{n-1}
$$

$$
= R_{21} \left[ \frac{N_0 A_n}{4\hbar} \right]_1 \left[ \frac{E_{\text{max1}}}{E_{\text{max2}}} \right]^n E_1^{n-1}
$$

[7.13]

Similarly for a third peak:

$$
\frac{dN}{dE_1} = R_{31} \left[ \frac{N_0 A_n}{4\hbar} \right]_1 \left[ \frac{E_{\text{max1}}}{E_{\text{max3}}} \right]^n E_1^{n-1}
$$

[7.14]

Thus an entire decay series can be constructed with the correct relative peak heights and energy distributions, by summing the distribution of all the peaks of the series; the entire spectrum height being dependent only upon the count chosen for the peak channel of the first peak.
calculated. In practice the highest energy peak is the easiest peak for which to choose an arbitrary height since it stands out from the other peaks. The computer program developed to compute and plot the synthetic thick source alpha spectrum is given in Appendix A.

Figure 24 illustrates a synthetic alpha particle spectrum composed of the accumulated energy distributions for the entire U-238 decay series in radioactive equilibrium for an infinitely thick source. The spectrum was computed assuming a peak count of 125 counts in the energy channel containing the initial energy of the 7.68 Mev Polonium-214 alpha peak and a value of 1.5 was used for N. The energy increment used is 10 Kev the same as that used for the experimental spectra. Thus the envelope of the distributions in the figure shows the same as a pulse height analyzer over 900 channels from 0.0 Mev to 9.0 Mev with 10 Kev per channel.

Figure 25 shows the synthetic alpha spectrum composed of the accumulated energy distributions for the entire Th-232 decay series in radioactive equilibrium. In this case the 8.78 Mev Polonium-212 peak was chosen to contain 125 counts, and the rest of the spectrum is computed relative to that peak.

Before considering more complex spectra, the effect of the value of N (which theory predicts to be close to 1.5) should be determined. As an extreme illus-
SYNTHETIC ALPHA SPECTRUM
U-238 DECAY SERIES IN EQUILIBRIUM.

PEAK MEV ISOTOPE
1 4.13 U238
2 4.18 U238
3 4.59 Rn226
4 4.62 Th230
5 4.68 Th230
6 4.72 U234
7 4.77 U234
8 4.78 Rn226
9 5.30 Po210
10 5.48 Rn222
11 6.00 Po210
12 7.68 Po214

Figure 24
SYNTHETIC ALPHA SPECTRUM

TH-232 DECAY SERIES IN EQUILIBRIUM.

Figure 25

PEAK MEV ISOPOPE
1 3.95 TH232
2 4.01 TH232
3 5.43 TH232
4 5.42 TH232
5 5.45 R4224
6 6.09 R4224
7 6.09 R4224
8 6.28 R4224
9 6.28 R4224
10 6.76 R0232
11 9.0
tration of fluctuations in N, the energy distribution for three peaks at 4.01 Mev, 6.28 Mev, and 8.78 Mev are plotted in Figure 26 with N values of 1, 3, 5, 7, and 9. When N equals 1, the distribution is flat, since the equation for the distribution reduces to a constant. Each distribution rides on the remaining tail of all higher energy distributions. A value of N = 9 produces a very rapid fall off rate, and the lower energy peaks are almost riding on the zero count line. The relative peak heights are also smaller at lower energies due to the relative range factor in the equation.

The energy spectrum ranges from 2.0 Mev to 9.0 Mev in Figure 26 instead of 0.0 Mev to 9.0 Mev in order to make the figure larger showing more detail in the important region of interest. Since the lowest energy alpha particle is about 4 Mev, this range of display is used for all further spectra.

A more realistic possible variation in the value of N about a value "close to 1.5 " is shown in Figure 27 for alpha spectrum of the U-238 decay series.

The effect of the change in the value of N is primarily noticeable at low energies due to the cumulative effect of all peaks and is very small at high energies. For clarity, and future comparison purposes, Figures 28 and 29 show the synthetic alpha spectra of the U-238 decay series and Th-232 decay series respect-
ENERGY DISTRIBUTION FOR 3 PEAKS WITH N=1, 3, 5, 7 AND 9.

Figure 26
SYNTHETIC ALPHA SPECTRUM
U-238 DECAY SERIES IN EQUILIBRIUM. INFINITELY THICK SOURCE.

PEAK MEV ISOTOPE
1 4.13 U238
2 4.16 U238
3 4.93 RA226
4 4.62 TH230
5 4.68 TH230
6 4.72 U234
7 4.77 U234
8 4.78 RA226
9 5.30 PO210
10 5.46 RA222
11 6.00 PO218
12 7.68 PO214

Figure 28
ively, over a range from 2.0 Mev to 9.0 Mev, with the obscuring energy distributions under the spectral envelope removed. Both spectra are computed using \( N=1.5 \).

Figure 30 illustrates the complexity to be expected of a rock spectrum with thorium:uranium ratio of 3.5. This synthetic spectrum was generated assuming 125 counts in the 8.78 Mev Polonium-212 peak from the thorium series. In addition the relative activities of uranium and thorium had to be taken into consideration. The activity of the thorium series in equilibrium is about one third that of the uranium series (246/733 exactly; Hyde, Perlman, and Seaborg, 1964) and must be considered in addition to the thorium:uranium ratio as given. It should be noted that because of this, when thorium and uranium are present in equal amounts, and in equilibrium with their respective daughter products, the thorium will only contribute about one third of the counts obtained under the spectrum.

Although the U-235 decay series should be a non-important part of this equilibrium study, it is interesting to observe the U-235 infinitely thick spectrum as shown in Figure 31. The U-235 series contains a large number of different energies compared to either the U-238 and Th-232 series whose alpha decays usually result only in one or two energies at most.

Since the U-235 occurs in nature in the ratio of
SYNTHETIC ALPHA SPECTRUM
TH/UR-8S. INFINITELY THICK SOURCE.

URANIUM
PEAK MEV ISOTOPE
3 4.13  U238
4 4.19  U238
5 4.59  RA226
6 4.62  TH230
7 4.88  TH230
8 4.72  U234
9 4.77  U234
10 4.70  RA226
11 5.30  PO210
12 5.48  RN222
13 6.00  PO210
22 7.60  PO214

THORIUM
PEAK MEV ISOTOPE
1 3.65  TH232
2 4.01  TH232
12 5.34  TH230
13 5.42  TH228
14 5.46  RA224
16 5.66  RA224
18 6.04  BI212
19 6.05  BI212
20 6.26  RN208
21 6.78  PO216
23 6.76  PO212

Figure 30
SYNTHETIC ALPHA SPECTRUM

U-235 DECAY SERIES IN EQUILIBRIUM

INFINITELY THICK SOURCE

PEAK MEV ISOTOPE

1  4.22  U235
2  4.32  U235
3  4.37  U235
4  4.40  U235
5  4.42  U235
6  4.56  U235
7  4.60  U235
8  4.73  PA231
9  4.94  PA231
10 5.01  PA231
11 5.02  PA231
12 5.05  PA231
13 5.54  RA223
14 5.61  RA223
15 5.69  TH227
16 5.70  TH227
17 5.71  TH227
18 5.71  RA223
19 5.75  RA223
20 5.76  TH227
21 5.96  TH227
22 5.98  TH227
23 6.04  TH227
24 6.28  BI121
25 6.42  RN219
26 6.55  RN219
27 6.62  BI121
28 6.82  RN219
29 7.38  PO215

Figure 31
U-238/U-235 = 137.8 (Sentile et al., 1957) it will not normally be discernible in an average rock spectrum. A few abnormal spectra, however, do show some peaks related to the U-235 series and these will be discussed in Chapter 8.

7.3 Elimination of the Th-232 Series Contribution to a Rock Spectrum

In order to study the U-238 decay series for the presence of radioactive equilibrium, the contributions of the Th-232 decay series to the spectrum, if present, must be removed. A large number of Pitchblende uranium ores do not contain appreciable thorium and this is not a problem, but in general thorium is associated with uranium. The Th-232 decay series can be safely assumed to be in radioactive equilibrium since the largest half-life of all the daughters in the series is 6.7 years, that of Radium-228. Thus, even if the series were disturbed by a metamorphic event, it would cause only temporary disequilibrium, and would in geological terms be 'instantly' returned to equilibrium.

The U-238 decay series, on the other hand, contains daughters with half lives which are of appreciable length even geologically such as U-234 with half life equal to 250,000 years and Th-230 with a half life of 80,000 years. A disturbance resulting in loss of a
daughter would not quickly be restored to equilibrium. The assumption of equilibrium of the Th-232 series is supported by the fact also that the Radon-220 has a half life of 54.5 seconds, not long enough for it to escape, while U-238 series produces Radon-222 with a half life of 3.82 days, a significant time in which it could possibly escape. The assumption of equilibrium is further supported in the literature by Mero (1960).

Thus it should be possible to match the synthetic equilibrium alpha spectrum of the Th-232 series to the Th-232 contribution in the rock spectrum and to subtract it leaving the U-238 series as a residual. This can be accomplished by fitting the synthetic Th-232 spectrum to the rock spectrum at the high energy Po-212 peak of the Th-232 series. In this portion of the spectrum the U-238 is absent, and will not present a problem. Since the rock spectrum contains the statistical scatter of counting, fitting it to the synthetic spectrum is accomplished using the entire portion of the 8.78 Mev Po-212 peak energy distribution from 8.78 Mev down to 6.78 Mev, (the highest U-238 series peak). The least squares criterion is used in determining the best fit. This determines the factor by which the synthetic spectrum must be multiplied to match it to the rock spectrum. Figure 32 shows a theoretical example where a synthetic spectrum containing both thorium and uranium in a ratio
TH/U = 3.5. INFIN. THICK

PEAK MEV ISOTOPE
1 4.13 U238
2 4.18 U238
3 4.59 U238
4 4.62 TH230
5 4.68 TH230
6 4.72 U234
7 4.77 U234
8 4.78 U234
9 5.30 PO210
10 5.40 PO210
11 6.60 PO210
12 7.68 PO214

Figure 32
of $\text{Th:U} = 3.5$ has the Th-232 series contribution subtracted and the residual displayed beneath it is the remaining U-238 decay series. This residual is then examined to determine the state of equilibrium.

7.4 Determination of the Presence of Disequilibrium in a U-238 Alpha Spectrum

Consider the U-238 series residual after removal of Th-232 series components as described above, or the U-238 series spectrum obtained from a rock with no thorium, to be in radioactive equilibrium. Then, fitting the synthetic U-238 series alpha spectrum to the highest energy peak in the series and subtracting the synthetic spectrum should give a residual spectrum which simply reflects the statistical scatter present in the experimental spectrum, fluctuating about a value of zero.

If the experimental or rock spectrum were not in equilibrium it would show a departure from this scatter about the zero residual, and would reflect the location in the decay series and amount of disequilibrium present. From previous equilibrium studies using radiochemical and other techniques, carried out on various radioactive ores, several common cases of disequilibrium have been observed, along with a few relatively rare cases. A study of the theoretical results of the application of the present technique to these characteristic
cases described by other workers would give an insight into what to expect as a residual spectrum for each case.

7.5 Application of the Technique to Theoretical Disequilibrium Spectra

Subtraction of the synthetic spectrum from the rock spectrum was eventually adopted as the standard technique, mainly because it resulted in a positive residual spectrum if the rock contained additional components not contained in the synthetic spectrum. If the rock did not contain a particular peak that was in the synthetic spectrum, it showed as a negative residual, indicating a deficiency in the rock spectrum. So in effect, positive and negative residuals indicated excesses and deficiencies respectively, in the rock spectrum.

The total count in each channel of the alpha energy spectrum determines the standard deviation for that portion of the spectrum. Since counting statistics are Poisson distributed, one standard deviation equals the square root of the count. The 99% confidence limits equals 2.58 times the standard deviation. In all analyses of the state of equilibrium of samples under consideration, the residual spectrum is displayed with the 99% confidence limits above and below the zero residual line. This allows one to determine whether a particular positive or negative residual can be relied upon to be valid indications of
disequilibrium or a result of statistical scatter. The 99% confidence limits are computed and displayed even for the theoretical cases to be discussed below, although they really have no meaning for a theoretical case. They can be increased or decreased by increasing or decreasing the scale on the theoretical alpha spectrum. They are, however, instructive in that they illustrate some examples wherein disequilibrium is not detectable within the 99% confidence limits since the count was not sufficiently high.

Figure 33 shows the first theoretical case of disequilibrium wherein the U-238 isotope has suffered a 50% loss. The example is considered to be one in which recent leaching of uranium took place, the time being relatively short, and the remainder of the series is in equilibrium with the original amount of U-238 that was present. This could easily occur in nature since the first alpha emitting daughter below U-238 is U-234 with a half life of 250,000 years. The parent U-238 could be removed, for example, during recent exposure to weathering by glaciation, and the remainder of the series would not be significantly affected by the loss for several thousand years. The highest count in this theoretical example is about 650 counts and in this example of 50% U-238 loss the negative residual indicating U-238 deficiency is inside the 99% confidence limits and would be theoretically
undetectable. A full discussion on the total count needed to detect various percentages of loss of the various daughters within given confidence limits, is given at the end of this chapter.

Another possible case mentioned in Chapter 5 is that of preferential leaching of U-234. A theoretical example of this case with 50% U-234 is shown in Figure 34. This example is again considered an "instant" loss in geologic time. The 80,000 year half life of the Th-230 below it maintains the bottom of the series in equilibrium with the top for several thousand years before the loss affects it significantly. The result is similar to the previous case, in that the negative residual which occurs at the position of U-234 in the alpha spectrum, indicates a U-234 deficiency but is theoretically undetectable within 99% confidence limits for the given total count of the theoretical spectrum.

A comparison of this case to that shown in Figure 35 indicates that in theory the loss of Th-230 would be distinguishable from the loss of U-234, but in practice would require a detector of higher resolution than that used in this study. Figure 36 shows a 50% Ra-226 loss which again in theory would be distinguishable from Th-230 or U-234 losses. This last case would never occur in nature, since the immediate daughter to Ra-226 is Rn-222 with a half life of 3.82 days. This
50% U-234 LOSSES.

PEAK MEV ISOTOPE
1 4.13 U238
2 4.18 U238
3 4.59 RA226
4 4.62 TH230
5 4.68 TH230
6 4.72 U234
7 4.77 U234
8 4.78 RA226
9 5.30 PO210
10 5.48 RN222
11 6.00 PO216
12 7.68 PO214

99% CONFIDENCE LIMITS

Figure 34
50% TH-230 LOSSES.

PEAK MEV ISOTOPE
1 4.13 U238
2 4.18 U238
3 4.59 RA226
4 4.62 TH230
5 4.68 TH230
6 4.72 U234
7 4.77 U234
8 4.78 RA226
9 5.30 P0210
10 5.48 RN222
11 6.00 P0216
12 7.68 P0214

Figure 35
RA-226, 50% LOSSES.

Figure 36
would quickly show similar losses to the parent Ra-226, and the entire lower end of the series (the high energy end of the alpha spectrum) would reflect the Ra-226 losses. So the only ambiguity would be between U-234 and Th-230 losses.

One particularly interesting case to consider is that of disequilibrium introduced in the laboratory during sample preparation time. Figure 37 shows this case which results in a large positive residual at the low energy end of the spectrum. The residual departs from zero at the location of the 5.30 Mev Po-210 peak. If the Radon loss had occurred in the sample while it was in situ, the residual would depart from zero at the Ra-226 peak location at 4.78 Mev instead. The reason for this is that disequilibrium introduced by Rn-222 loss in the laboratory would result in a similar loss of Po-218 (6.00 Mev) and Po-214 (7.68 Mev), which are the immediate daughters of Rn-222 with half lives of 3.05 minutes and 160 milliseconds respectively. The last alpha emitter in the decay series is the Po-210 (5.30 Mev) which has a half life of 138.4 days and would continue to remain in equilibrium with the top members of the series. Thus the positive residual would occur at 5.30 Mev, showing the "excess" of Po-210 relative to the Rn-222, Po-218, and Po-214. In the case of Rn-222 loss in the field, it would more than likely have occurred at a time appreci-
ably greater than several half lives of 138.4 day Po-210, and the Rn-222 loss would be reflected in the Po-210 peak. Thus the residual would depart from zero at the 4.78 Mev, the Ra-226 peak, which would represent the lowest daughter still in equilibrium with the upper members of the decay series.

Two less straightforward cases of multiple losses will be considered next. The theoretical case of progressive losses in the decay series is shown in Figure 38. Here the losses are 50% Th-230, 75% Ra-226, 80% Rn-222, and the loss occurred a sufficiently long time ago that the Po-210 is in equilibrium with the radon and shows a similar 80% loss. All losses are relative to the equilibrium amount that would normally be associated with the amount of U-238 present. Since the last four alpha emitting daughters in the series Rn-222, Po-218, Po-214, and Po-210 are in equilibrium, the residual spectrum departs from zero at the 4.78 Mev peak of Ra-226. The upper part of the decay series is present in amounts greater than that necessary to support the bottom of the series in equilibrium. This is a difficult case in which a further fit of the synthetic spectrum is required to the portion of the 4.59 Mev Ra-226 peak lying between 4.18 Mev (U-238) and 4.59 Mev. A fit to this would indicate that there was a loss of either Th-230, Ra-226, or U-234 relative to the amount of U-238 present, but
PROGRESSIVE LOSSES:
TH-230, 50%,
RA-226, 75%,
RN-222, 80%,
PO-210, 80%.

PEAK MEV ISOPE
1 4.13 U238
2 4.10 U238
3 4.59 RA226
4 4.62 Th230
5 4.53 Th230
6 4.72 U234
7 4.77 U234
8 4.79 RA226
9 5.30 Pu239
10 5.48 RN222
11 6.09 PO210
12 7.68 PO214

Figure 38
which one was missing would be indeterminate due to their similar energies.

A similarly complex case is shown in Figure 39 which illustrates 80% Th-230 losses and 50% Ra-226 losses relative to the amount of U-238 present. The Rn-222 isotope and daughters are in equilibrium with the Ra-226 due to their short half lives. The residual spectrum departs positively from zero at the 4.77 Mev U-234 peak, indicating it is present in excess of the amount required to support the lower daughters in the series. Then at the 4.68 Mev Th-230 peak the residual drops back to a less positive position indicating the amount of Th-230 is less than that required to support the amount of U-234 present. The residual is then flat over to the 4.18 Mev peak of U-238 where it rises positively again indicating the U-238 is present in amounts greater than that required to support the Th-230 in equilibrium. This rather complex case is illustrative of the type of information it is possible to derive from the residual spectrum if the resolution is sufficiently high. The conclusions obtained from this residual spectrum would indicate U-238 $\rightarrow$ Th-230 $\rightarrow$ Ra-226 = lower daughters. This refers to the equilibrium amounts of the isotopes.

7.6 Total Count Required to Detect a Given Amount of Disequilibrium
LOSSES:
TH-230, 80%, RA-226, 50%.

PEAK MEV ISOTOPE
1  4.19  U238
2  4.18  U238
3  4.19  R226
4  4.62  TH230
5  4.68  TH230
6  4.72  U234
7  4.72  U234
8  4.78  R226
9  5.30  P0210
10 5.48  RN224
11 6.00  P0210
12 7.60  P0214

Figure 39
If we consider the example of disequilibrium shown in Figure 33 again, a method of pre-calculating the count required to guarantee detection of a given amount of disequilibrium can be developed. The reason why the 50% losses in U-238 cannot be seen as definitely above the 99% confidence limits set for the error in the residual is because the count rate is too low for any one individual alpha peak. For example, take a peak height of 100 counts, the maximum total peak height of the spectrum would be say 1000 counts. This has a possible error of \( \sqrt{1000} = 32 \) counts and 99% confidence limits = 2.58 x 32 which is approximately equal to 80 counts. Therefore the error limits will be \( \pm 80 \) counts while a 50% loss in the U-238 peak will show up as 50 counts (i.e. half of the normal equilibrium value of 100 counts). A peak height of double the above example (200 counts) would give an error of \( \sqrt{2} \) times the above, i.e. \( \sqrt{2000} \) counts or approximately 45 counts. The 99% confidence limit = 2.58 x 45 = \( \pm 115 \) counts. While the 50% loss is 100 counts (half of 200) and is getting close to being observable outside the confidence limits. Thus we can determine the limiting number of counts required in a peak to be able to detect a certain percentage loss due to disequilibrium within certain confidence limits. In Figure 33, for example, the highest energy peak has
a maximum count height of 125 counts. This predetermines the count to be expected at all other peaks. Thus for example, peak 9, the 5.30 Mev Po-210 peak is about 400 counts high. This includes the energy distribution 'tails' of all higher energy peaks. The Po-210 alone will have a peak height roughly equal to that of the highest energy peak which was 125 counts. Consider the ratio of the total count at the Po-210 position relative to the highest energy peak height, in this case 400/125. If we call this ratio \( R(1) \) i.e. the ratio of the total height of the peak 1 to the height of the highest energy peak which we will call \( N \) counts high, then to detect say a 50% loss in peak 1 with 99% confidence limits we have
\[
\frac{50}{100} \times N(1) \geq 2.58 \times \sqrt{R(1) \times N}
\]
In most cases the height of a given individual peak \( N(1) \) will be equal to the height of the highest energy peak \( N \). Then if

\[
P = \% \text{ losses desired to be detectable}
\]
\[
C = \text{confidence limits desired in terms of number of standard deviations.}
\]
\[
N = \text{count in highest energy peak}
\]
\[
R(1) = \text{ratio of total count at peak (1) to the count } N \text{ of the highest energy peak.}
\]
we have
\[
P \times N \geq C \times \sqrt{R \times N}
\]
When the equation is an equality, the residual will depart from zero by exactly an amount equal to the confidence limits chosen.
We can solve for $N$ and get

$$N = \frac{C^2 R}{P^2}$$

As an example suppose we wish to detect a 50% loss ($P = 0.5$) in a peak with $R = 5.0$ with 99% confidence ($C = 2.58$ standard deviations). Then

$$N = \frac{(2.58)^2 (5.0)}{(0.5)^2} = 126 \text{ counts}$$

as a check the total height at the peak will be $5 \times 126 = 630$, then 99% confidence limits $= 2.58 \times \sqrt{630} = 63 \text{ counts}$. Note that this is exactly equal to the 50% loss, i.e. 50% x 126 counts. Therefore, the 126 counts in the highest energy peak is just sufficient to detect a 50% loss with 99% confidence in the given peak.
CHAPTER 8

EXPERIMENTAL RESULTS

8.1 Introduction

Before the technique of disequilibrium detection described in previous chapters could be generally applied to any rock sample, the effect of variation of experimental parameters had to be determined. In order to simulate variations in concentrations of the radioelements, a number of synthetic rocks were produced by mixing pure silica sand with epoxy and known amounts of $\text{U}_3\text{O}_8$ powder. This enabled a control over the $\text{U}_3\text{O}_8$ concentrations which would not be possible with real rock samples. The synthetic rocks were prepared and allowed to harden in glass petri dishes, which gave them a thickness of about half an inch. When the mixture had hardened, the surface was ground flat to expose a fresh surface of the synthetic rock. The surface area was approximately 65 square centimeters.

8.2 Determination of the Effect of Variation of Experimental Parameters
8.2.1 Experiments on Variations of Source Area

The possibility of changes occurring in the shape of an infinitely thick source alpha spectrum with variations in source area, although unlikely, could not be discounted, and a series of spectra were measured from the same source, with varying surface areas exposed. The source was a 50% U\textsubscript{238} synthetic rock, maintained at 1 centimeter distance from the detector. The source area was controlled by shielding all but the desired area with a lead sheet. The areas were circular and varied from 1 square centimeter to 65 square centimeters. The results are shown in Figure 40 in the form of a comparison of seven spectra, with source areas of 1, 2, 4, 8, 16, 32, and 65 square centimeters respectively. Counting times were 800, 400, 200, 200, 200, 100, and 100 minutes respectively. The counting times were longer for the smaller source areas. The smoothed spectra as shown in the figure (9 point smooth with a 2nd degree polynomial) were more amenable to comparison than the original spectra with the associated statistical scatter. That the shape of the spectra is unaffected by variations in source area can be seen in the diagram. The effect of the Po-210 peak at 5.30 Mev in the background is becoming apparent for the 1 square centimeter area source since it had to be counted for over 13 hours. The conclusion
VARIATION IN SOURCE AREA
FOR
50% U3O8 SYNTHETIC ROCK

ALL SPECTRA ARE NORMALIZED TO BOTTOM SPECTRA
9 POINT SMOOTH WITH A POLYNOMIAL OF DEGREE 2

Figure 40
is that source area does not affect the spectral shape.

An interesting experiment to determine the relative effect on the shape of the spectrum of the center area of the source compared to the outer edge of the source was performed. A synthetic uraniferous rock was counted twice with an annular area of 32 square centimeters exposed (tape 164) and again with 32 square centimeters of center area exposed (tape 160). Counting times were 46 and 27 hours respectively. Figure 41 shows the normalized smoothed spectra for comparison. The large 5.30 Mev background peak has been truncated for clarity. The central area contributes a higher count rate than the annular area as is expected, but the spectral shape is unaffected by the location of the radioactive component in the sample.

8.2.2 Experiments on Variations in Source Concentration

A set of synthetic rocks with varying amounts of $\text{U}_3\text{O}_8$ was prepared and their alpha spectra were measured. This was to check on the possibility that the spectral shape would be dependent upon uranium concentrations in the rock samples. A comparison of five smoothed spectra from sources of 1%, 5%, 10%, 25%, and 50% $\text{U}_3\text{O}_8$ content respectively are normalized and shown in Figure 42. Counting times are 1500, 660, 300, 200, and 10 minutes respectively. The only difference between the spectra
Figure 1

All spectra are normalized to bottom spectrum 9 point smooth with a polynomial of degree 2.

T-164 CENTER SHIELD
T-160 ANNULAR SHIELD
(32 cm sq. exposed)
VARIATION IN CONCENTRATION
OF U308 FOR CONSTANT
SOURCE GEOMETRY

ALL SPECTRA ARE NORMALIZED TO BOTTOM SPECTRA
9 POINT SMOOTH WITH A POLYNOMIAL OF DEGREE 2

Figure 42
is the background component of the 5.30 Mev Po-210 peak. The counting time was inversely proportional to the uranium content, and the 1% U₃O₈ sample shows the large background peak obtained after counting for 25 hours. Source area was maintained constant, and source detector distance was 1 centimeter in all cases. In conclusion, spectral shape is unaffected by variation in source concentration.

8.2.3 Experiments on Variation of Source-Detector Separation

The 50% U₃O₈ synthetic rock was counted for source areas of 4 square centimeters and 65 square centimeters, at two different source-detector distances of 1.0 and 4.5 centimeters respectively. Figure 43 shows the normalized results of the smoothed spectra with the two 4 cm² area sources shown above the two 65 cm² area sources. The counting times for the spectra are from top to bottom 800, 200, 100, and 100 minutes respectively. The 800 minute count shows the 5.30 Mev background peak. It can be concluded that the source-detector separation has no effect on the spectral shape, but affects only the counting time.

8.3 Results of Application of the Present Technique to Rocks and Minerals
VARIATION IN SOURCE DISTANCE 
FOR TWO SOURCE AREAS OF 
50% U3O8 SYNTHETIC ROCK

ALL SPECTRA ARE NORMALIZED TO BOTTOM SPECTRA
3 POINT SMOOTH WITH A POLYNOMIAL OF DEGREE 2

Figure 43
The experimental spectra were first put through the gain shifting program to align them with the theoretical spectra which were generated with 10 Kev per channel and the zero in channel zero. Since the value of \( N \) in the equation generating the synthetic spectrum is a function of the effective atomic number of the source and has a value "close to 1.5" (Lapp and Andrews, 1963), an iterative technique was developed to determine the best value of \( N \). The best value was obtained when the residual from the fit of the experimental and synthetic spectra was a minimum. If the best value of \( N \) was found to lie between 1.43 and 1.57, the spectrum was arbitrarily assumed to be in equilibrium as the value of \( N \) is "close to 1.5". Otherwise, the value of \( N \) was set to exactly 1.50 and the computed residual spectrum indicated the location and amount of the disequilibrium in the decay series. The effect on the synthetic spectra of variation of the value of \( N \) was discussed in Chapter 7.

8.3.1 Results for Samples Containing U-238 Series Alone

These results can be subdivided into three sections dealing with the cases found to be in radioactive equilibrium; slightly out of equilibrium; and definitely in a 'state' of disequilibrium. In Chapter 7 the method of fitting the synthetic spectrum to the experimental spectrum was discussed. Since the height of
the synthetic spectrum is arbitrary, a value of 125 counts was chosen for the highest energy peak which provides a total spectral height which can be displayed over a range of about 700 counts for an equilibrium case. The 'fit' determines the normalization factor by which the experimental spectrum is multiplied to bring it into alignment with this synthetic spectrum. Thus all spectra are displayed over this same range making visual comparison easier.

Counting times were determined as the time required to obtain a peak height of about 100 counts in the highest energy peak. Although the counting time was very short for some extremely radioactive samples it was still determined by this criterion since increasing the time would also increase the detector contamination as discussed in Chapter 5. This would subsequently require a time-consuming background check to be made. For this reason a larger statistical scatter than absolutely necessary was tolerated. This amount of scatter has a negligible effect on the equilibrium determinations.

8.3.1.1 Cases in Equilibrium

Tape No.248 shown in Figure 44 is the alpha spectrum of Uraninite from Bolivia obtained from the Royal Ontario Museum (R.O.M.) in Toronto. The sample surface was a fresh cut with a source area of 8 sq.cm. and a counting time of 10 minutes. The normalization
factor by which the spectrum in Figure 44 is multiplied is 1.111. The iterated best value of N used for the synthetic spectrum is 1.568. The residual exhibits the statistical scatter to be expected, fluctuating about the zero residual line, within the 99% confidence limit lines.

An interesting effect visible in Figure 44 is the large residual at the locations of the peaks in the spectrum. This is the result of subtracting the synthetic spectrum which has a sharp theoretical rise at each peak, while the experimental spectrum exhibits a gaussian rounding of the edges of the peaks due to a combination of high energy stragglers, and the finite resolution of the instrumentation. It would be possible to eliminate this effect by including a gaussian rounding effect in the theoretical spectrum at each peak location. It was decided to retain the sharp rise in the theoretical spectra since the shape of the residual gives an indication as to whether the correct gain shift for the experimental spectrum was used. An incorrect gain shift is indicated by the presence of asymmetric residuals at the peak locations. The result is best seen by observing the highest energy peak and its associated residual. These residuals at peaks quite often extend outside the confidence limits but are in no way related to the equilibrium state of the sample.
Tape No. 250 Shown in Figure 45 is the alpha spectrum of a pitchblende sample originating from Katanga, Congo, also obtained from the R.O.M. The sample surface was a fresh cut of area 4 sq. cm., and a counting time of 8 minutes. The normalization factor is 0.981, and the iterated best N equals 1.5062. The residual is well within the confidence limits indicating the sample is in radioactive equilibrium.

Tape No. 252 shown in Figure 46 is the alpha spectrum of another pitchblende sample from Katanga, supplied by the R.O.M. The sample surface is a fresh cut of 2 sq. cm. area and the counting time was 20 minutes. The normalization factor is 0.920 and the iterated best N equals 1.4437. As in the previous Congo pitchblende sample the residual lies within the confidence limits, indicating a state of radioactive equilibrium.

Tape No. 204 shown in Figure 47 is the same Congo pitchblende sample described above as Tape No. 252, before the fresh surface was cut. A slight peaking effect is present due to the additional "thin-source" effect caused by the rough sample surface. The sample area was 4 sq. cm., and the counting time was 100 minutes. The normalization factor is 0.159 and the iterated best N equals 1.5687. The difference in the iterated best N values in the sample before and after cutting a fresh surface is 0.1250. This can be the result of several combined effects,
TAPE NO. 250.
KATANGA 99 FLAT
AREA (SQ. CM.) 4.0
TIME (MIN.) 8.0

Figure 45
Figure 46
namely (1) the additional thin source effect of the rough rock surface (Tape No. 204), (2) the change in effective atomic weight and density of the rough surface as seen by the alpha particles being emitted from the top 100 microns or so of infinitely thick surface and (3) the effect of a poor choice of the portion of the energy distribution of the highest energy peak for the least square fit to the synthetic spectrum. The latter is the most probable cause of the difference. Reference to Figure 27 in Chapter 7 indicates the change in shape of the spectrum to be expected from a change in the value of N of the order of 0.10. The same effect as a slight change in the value of N can be accomplished by choosing either a high or low portion of the energy distribution of the highest energy peak for the fit to the synthetic spectrum. The shape of the experimental results for the 7.68 Mev peak energy distribution is subject to this type of discrepancy. The residual shown in Figure 47 shows that the portion of the energy distribution between 6.25 Mev and 6.75 Mev is slightly higher than the portion between 6.75 and 7.25 Mev. The actual portion of the energy distribution used for the fit in this spectrum and the preceding spectra is between 6.50 and 7.50 Mev. A choice of 6.75 to 7.25 would result in a slightly lower value of N and a choice of 6.25 to 6.75 Mev would produce a slightly higher value of N. An example of the sensitivity
of this method to the choice of the portion of the energy
distribution used for the fit to the synthetic spectrum
is discussed at the end of this chapter.

Tape No.167 shown in Figure 48 is the alpha
spectrum of a pitchblende sample from Great Bear Lake,
N.W.T. The sample surface was a fresh cut of 8 sq.cm.
area, and a counting time of 300 minutes. The normalization
factor is 0.414 and the iterated best $N$ equals 1.5312.
The residual is within the confidence limits except for
the small peak at 5.30 Mev from the background which
extends outside the confidence limits due to the 300
minute counting time.

8.3.1.2 Cases of Slight Radioactive Disequilibrium

The following five samples represent cases wherein
the residual lies slightly outside the 99% confidence
limit lines, or inside the confidence limits but not
centered along the zero residual line. In this and all
succeeding spectra a value of $N = 1.5$ was used for the
synthetic spectra.

Tape No.214 shown in Figure 49 is the alpha
spectrum of a synthetic rock produced from a mixture
of powdered $U_3O_8$ with epoxy and silica sand as described
in an earlier section. The $U_3O_8$ powder represents 50%
by weight of the sample. The flat cut surface had an
area of 65 sq.cm. and was counted for 100 minutes. The
normalization factor is 0.231. Since this sample showed
indications of being out of radioactive equilibrium, a value of $N = 1.5$ was used in computing the synthetic spectrum. The residual is positive for the energy distribution below the 4.18 Mev U-238 peak which at first indicates an excess of U-238 relative to the daughters lower in the decay series, but further consideration changes the initial deduction as follows. The peculiar shape of the energy distribution for each peak is due to the decrease or absence of the surface portion of the $U_3O_8$ in the sample resulting from a combination of removal of surface grains of $U_3O_8$ in the cutting process, and partial melting of the epoxy during cutting which produces a thin epoxy film covering surface grains. Thus the highest energy particles, namely those emitted from near the surface, are present in a diminished amount, producing the slight negative slope in the energy distribution. The decrease of this component produces an over-subtraction when the synthetic spectrum is fitted to the experimental spectrum. The combined over-subtraction of the peaks of Ra-226, Th-230, and U-234 in the range 4.59 Mev to 4.78 Mev reduces the residual to a negative value slightly outside the lower confidence limit. This in effect artificially pulls the energy distribution between 4.18 Mev and 4.7 Mev down inside the confidence limits. The position at which disequilibrium occurs is
more likely at 4.7 Mev indicating the U-238, U-234, Th-230, and Ra-226 are all in excess of equilibrium amounts and that the losses are that of Rn-222 from the powdered U₃O₈ used in preparation of the synthetic rock. The next sample considered below has a rather dramatic similar effect only in the opposite direction.

Tape No.239 shown in Figure 50 is the alpha spectrum of a powdered carnotite sample from Colorado, obtained from the R.O.M. The sample was an infinitely thick layer of powder with a surface area of 30.0 sq.cm., a counting time of 400 minutes, and a normalization factor of 0.314. The additional thin source distribution characteristic of a powdered surface rides on top of the infinitely thick source distribution. Subtraction of the synthetic spectrum (N = 1.5) leaves the thin source spectrum as a residual. Since the residual decreases to negative values at low energies and does not entirely sit on the zero residual line, some indications of disequilibrium are present. If the infinitely thick source part of the spectrum had a deficiency of U-238, the thin source residual would drop to negative values below the lower confidence limit. It is also possible that the deficiency exists in the multiple peak instead of, or in addition to that in the U-238 peak at 4.18 Mev. Thus the only conclusion possible is that there is a deficiency in the top of the U-238 decay series. This is consistent
Tape No. 239
Carnotite Fine
Area (sq. cm.) 30.0
Time (min.) 400.0
.314 x

Peak MeV Isotope
1 4.13 U238
2 4.18 U238
3 4.59 RA226
4 4.62 TH230
5 4.68 TH230
6 4.72 U234
7 4.77 U234
8 4.78 RA226
9 5.30 PO210
10 5.48 RN222
11 6.00 PO210
12 7.68 PO214

Figure 50
with the published literature on Colorado carnotite ores which are often in a state of radioactive disequilibrium.

Tape No.233 shown in Figure 51 is the alpha spectrum of a pitchblende sample from Beaverlodge, Saskatchewan. The sample was a fresh cut of 8.0 sq.cm. with a counting time of 100 minutes, the normalization factor is 0.370. This is a case of slight losses of one of the three daughters in the multiple peak at 4.7 Mev, but the residual remains inside the 99% confidence limits at approximately the 68% confidence limits. Whether the loss is real is open to question, but in any case it would be relatively small.

Tape No.138 shown in Figure 52 is the alpha spectrum of a pitchblende sample from the Ace Mine in Beaverlodge, Saskatchewan. The sample was a fresh cut of 8.0 sq.cm. with an extremely long counting time of 2226.0 minutes. The normalization factor is 0.954. This is similar to the previous case with a slight negative residual below 4.18 Mev indicating U-238 losses, in a slight amount, being inside the 99% confidence limits. The long counting time explains the large peak of Po-210 at 5.30 Mev in the residual.

The three additional "thin-source-type" peaks above 6.0 Mev are caused by a very interesting phenomenon which occurs only in relatively porous or weathered
samples. The measured energies of the three peaks are 6.62 Mev, 6.82 Mev, and 7.38 Mev respectively. Since these do not correspond to any peaks in the U-238 series or the Th-232 series the unexpected source turns out to be the U-235 decay series. Normally the U-235 series contributes an undetectably small amount to the alpha spectrum since the U-235/U-238 ratio is about 1/137 (Sentfle et al, 1957, Hyde et al, 1964). In this spectrum the three peaks at 6.62, 6.82, and 7.38 Mev correspond to the Bi-211, Rn-219, and Po-215 peaks from the U-235 decay series respectively. The interpretation of these peaks is that they are caused by the out-gassing of the Rn-219 (from the U-235 series) from the porous rock. This Rn-219 in the vacuum chamber produces the thin source peak at 6.82 Mev along with less intense peaks at 6.42 and 6.55 Mev which are not discernible in this spectrum. The Rn-219 with a half life of 3.92 seconds, produces Po-215 with a half life of 1.83 milliseconds which produces the peak at 7.38 Mev. This further decays into Pb-211 with a half life of 36.1 minutes, emits a beta particle becoming Bi-211 with a half life of 2.16 minutes which produces the third "thin-source" peak at 6.62 Mev along with a less intense peak at 6.28 Mev which is not discernible. This same phenomenon does not occur with the out-gassing of Rn-222 from the U-238 decay series since it has a half life of 3.82 days and
would produce negligible thin source peaks. It does not occur with Rn-220 from the Th-232 series since this sample has no thorium present as is evidenced by the absence of the 8.78 Mev peak from the Th-232 series. Further substantiation of the validity of this interpretation is shown in Figure 53, the alpha spectrum of another sample from the same location (Tape No.129). The source area was 1 sq.cm. and the counting time was very long being 2880 minutes. The normalization factor is 2.306. Here even the low intensity peaks mentioned above at 6.28 and 6.42 Mev are visible, and the 6.55 Mev low intensity peak is seen as a broadening of the low energy side of the 6.62 Mev Bi-211 "thin-source" peak. The same conclusions regarding the state of equilibrium of this sample can be drawn as in the other sample. There appears to be a slight negative residual indicating losses in the upper end of the U-238 decay series, and the losses are small enough that they are undetectable with 99% confidence. These small losses are discussed at the end of the chapter.

8.3.1.3 Cases of Definite Disequilibrium

This section deals with the results of three uranium ore samples which showed visible oxidation and weathering (Tape 200, 201, and 202) and a fourth sample (Tape 208) which appeared to be a fresh unweathered
surface. All four samples had rough surfaces. One sample
(Tape 202) was subsequently cut and an alpha spectrum
was obtained from the fresh cut surface. This was dis-
cussed earlier in section 8.3.1.1 and was found to be
in radioactive equilibrium. The relative amounts of six
of the radioelements was estimated by x-ray fluorescence
for the fresh cut surface and the weathered surface,
and the results substantiate the interpretation given
below.

Tape No.200 shown in Figure 54 is the alpha
spectrum of a weathered pitchblende sample from Great
Bear Lake, N.W.T. The source area was 16.0 sq.cm., the
counting time was 100.0 minutes, and the normalization
factor was 0.356. The spectrum exhibits the three "thin-
source-type" peaks from the U-235 series discussed above
for Tape No.129 and Tape No.138. The positive residual
below 4.7 Mev indicates an excess at the high end of
the U-238 series which is interpreted as a loss of Rn-222
which itself is in radioactive equilibrium with its
daughter products. The loss did not occur in the lab
since 138 day half life Po-210 (peak No.9 at 5.30 Mev
in Figure 54) is in equilibrium with the series up to
Rn-222. The additional rise in the residual at U-238
peaks (4.18 Mev) indicates the U-238 is in excess rel-
ative to the daughters in the multiple peak containing
U-234, Th-230, and Ra-226.
Figure 54
Tape No. 201 shown in Figure 55 exhibits the same type of Rn-222 losses, but no U-238 excess relative to the three daughters in the multiple peak at 4.7 Mev. The sample is weathered pitchblende from Beaverlodge, Saskatchewan. The source area was 8 sq.cm., counting time was 200 minutes, and the normalization factor equals 0.063. The three "thin-source-type" peaks from the U-235 series are also present, indicative of the weathered, porous nature of the sample. A special peculiarity of this particular spectrum is the extra "thin-source-type" effect riding on the top of the multiple peak at 4.7 Mev. This "thin-source" effect is also indicated by the curved shape of the residual distribution below 4.7 Mev which should normally be linear.

The conclusion is that there is a slight excess of one of the three peaks (Ra-226, Th-230, or U-234) on the surface, sufficient to produce the slight "thin-source" effect. The effect is almost statistically insignificant but is supported by similar evidence in Tape No. 208 to be discussed later where the surface "thin-source" effect appears to be removed from the spectrum in this same peak location.

Before discussing Tape No. 208, it is interesting to point out another case related to that of Tape No. 201.

Tape No. 202 shown in Figure 56 is the alpha spectrum of a weathered sample of Uraninite from Bolivia
TAPE NO. 201.
PITCHBLEND 2824
AREA (SQ. CM.) 8.0
TIME (MIN.) 200.0
.063 X

PEAK MEV ISOTOPE
1 4.13 U238
2 4.16 U238
3 4.59 RA226
4 4.62 TH230
5 4.68 TH230
6 4.72 U234
7 4.77 U234
8 4.79 RA226
9 5.30 PO210
10 5.49 RN222
11 6.00 PO218
12 7.68 PO214

Figure 55
TAPE NO. 202.
BOLIVIA
AREA (SQ. CM.) 8.0
TIME (MIN.) 100.0
.092 X

PEAK MEV ISOTOPE
1 4.13 U238
2 4.18 U238
3 4.59 RA226
4 4.62 TH230
5 4.68 TH230
6 4.72 U234
7 4.77 U234
8 4.78 RA226
9 5.30 PO210
10 5.48 RN222
11 6.00 PO210
12 7.68 PO214

Figure 56
with surface area 8.0 sq.cm., counting time 100 minutes, and normalization factor 0.092. This weathered sample is the same as that discussed earlier as Tape No.248 which was a fresh cut from the sample showing no disequilibrium. The weathered sample, however, shows a loss of U-238, and one of the three daughters, Ra-226, Th-230, or U-234 in the multiple peak at 4.7 Mev. The most likely interpretation is that U-238 and U-234, being chemically similar were both lost.

An interesting feature carried over from the previous sample (Tape No.201) which was counted immediately before this sample is the small Bi-211 peak at 6.62 Mev. This is a result of the 36 minute half life of its parent Pb-211, a beta emitter. The supply of Rn-219 was removed when the porous rock was removed, but the Pb-211 continued to supply Bi-211 which emits the 6.62 Mev alpha particle. The peak is small since the half life is 36 minutes. The time between removal of sample recorded at Tape No.201, and the start of counting of Tape No.202 was about 40 minutes, one half life of Pb-211. A repeat of this spectrum taken at a later date showed no sign of this small peak.

A qualitative external check was made using x-ray fluorescence studies of both the weathered (Tape 202) and fresh (Tape 248) samples of this sample of Bolivian Uraninite by G. Renwick an undergraduate in geophysics
at the University of Western Ontario. Since the x-ray fluorescence unit was not calibrated for analysis of heavy minerals, only the relative counts obtained for the weathered and fresh surfaces were compared, for six radioelements. The count rate (counts per second) obtained for fresh and weathered surfaces were respectively: 72, 76 for bismuth; 394, 337 for radium; 527, 487 for thorium; 7006, 6322 for actinium; 7267, 8829 for lead; and 33691, 20141 for uranium. It should be pointed out that the various isotopes of individual elements are not distinguished by x-ray fluorescence. Thus, the uranium result, for example, includes U-238, U-234, and U-235. The results of x-ray fluorescence indicate the weathered surface has a decreased amount of radium, thorium, actinium, and uranium, and a slight increase of bismuth, although this may not be statistically significant (72 cps vs 76 cps). There is an increase in the amount of lead in the weathered zone relative to the fresh zone. How significant any of these results are, is unknown since the relation between elemental content and count rate is nonlinear for x-ray fluorescence, and this 20% change in count rate may not relate to a very large variation in the amount of lead. The 40% change in uranium count rate is the most significant difference, and supports the conclusions of uranium losses based on the alpha spectra, but further checks
of this nature would require a complete calibration and standardization of the x-ray fluorescence equipment for heavy element analysis.

Tape No. 208 shown in Figure 57 is the alpha spectrum of a sample of pitchblende from Great Bear Lake, N.W.T. obtained from the R.O.M. the surface did not appear to be weathered but was rough, of area 4.0 sq. cm., counting time was 100 minutes, and normalization factor 0.295.

This sample appears to be exhibiting an extra "thin-source" effect from the roughness of the surface, as postulated earlier for Tape No. 201 (Figure 55) and Tape No. 239 (Figure 50). The additional peaking can be seen in Figure 57 at the peak locations of peaks numbered 9, 10, 11, and 12. The peculiar rounded shape of the U-238 peak and the multiple peaks at 4.7 Mev indicates exactly the opposite effect, i.e. reduction at the surface of the amount of U-238 and one or more of U-234, Th-230, and Ra-226. The negative residual below 4.7 Mev is curved and extends back to zero residual by 2.0 Mev, indicating no disequilibrium, but a loss of the thin source portion of the energy distributions of these low energy peaks. A logical explanation that can be given is that this represents the result of a chemical oxidation of U-238 and U-234 producing an oxide film on the uranium isotopes decreasing the thin source effect normally expected
TAPE NO. 208
G.B.L. PITCH 10603
AREA (SQ. CM.) 4.0
TIME (MIN.) 100.0
.295 X

PEAK MEV ISOTOPE
1 4.13 U238
2 4.18 U238
3 4.59 R226
4 4.62 TH230
5 4.68 TH230
6 4.72 U234
7 4.77 U234
8 4.78 R226
9 5.30 P0210
10 5.48 R0222
11 6.00 P0218
12 7.68 P0214

Figure 57
from the rough surface, while peaks 9 to 12 have relatively short half lives and are freshly replenished by the production of new daughter material.

8.3.2 Rocks and Minerals Containing Th-232 and U-238

The elimination of the Th-232 series contribution to the alpha spectrum of a sample containing both uranium and thorium was discussed in Chapter 7. The following are the results of the application of the present radioactive equilibrium determination technique to six samples where:

(1) the amount of uranium is the same as that of thorium,
(2) the amount of uranium is greater than that of thorium,
(3) the amount of uranium is less than that of thorium, and
(4) a synthetic rock with Th/U = 0.5.

Finally two cases illustrating extreme "thin-source" peaking from out-gassing of Radon in the vacuum chamber are considered.

Tape No.169 shown in Figure 58 is the alpha spectrum of a fresh cut sample of Thorianite from Quebec, the source area was 8.0 sq.cm., counting time 1800 minutes, and normalization factor 0.906. The residual in this case is the alpha spectrum of the U-238 decay series and the 5.30 Mev Po-210 peak in the background. This Uranium residual is displayed again in Figure 59 with a normalization factor of 0.585. Here the residual
lies within the confidence limits indicating a state of radioactive equilibrium. The large Po-210 peak at 5.30 Mev extends outside the confidence limits as a positive residual as expected for a sample with a long counting time such as this one. The shape of this alpha spectrum is distorted slightly at 6.78 Mev as a result of the fact that this is a residual spectrum after subtracting the Thorium series component.

Tape No.174 shown in Figure 60 is the residual Uranium alpha spectrum of a sample of conglomeratic uranium ore from the Elliot Lake area of Ontario. The source was cut flat, of area 65 sq.cm., counting time 5735 minutes, and a normalization factor of 0.088. An inset on Figure 60 shows a portion of the original spectrum from 7.5 to 9.0 Mev, illustrating the highest energy thorium series peak, and the residual after its removal. The normalization factor is 9.885 for the inset, indicating that the thorium component in the spectrum was quite small, even after the long counting time involved. The final residual shows the sample to be in radioactive equilibrium within the confidence limits. The Po-210 peak at 5.30 Mev extends outside the confidence limits as a large positive residual as expected.

Tape No.172 shown in Figure 61 is the alpha spectrum of a sample of Allanite from Bancroft, Ontario. The surface was rough, of area 8.0 sq.cm., counting time
TAPE NO. 172.
ALLANITE 1805
AREA (SQ. CM.) 8.0
TIME (MIN.) 5610.0
1.880 X

PEAK MEV ISOTOPE
1 3.95 TH232
2 4.01 TH232
3 5.34 TH228
4 5.42 TH228
5 5.45 RA224
6 5.60 RA224
7 6.04 BI212
8 6.09 BI212
9 6.20 RN220
10 6.78 PO216
11 8.78 PO212

Figure 61
5610 minutes, and normalization factor 1.880. The residual is the alpha spectrum of the U-238 series after removal of the thorium component. This represents a sample with Thorium $\rightarrow$ Uranium, and the Uranium residual is actually too low in count height to have good counting statistics. The result of applying the present technique to this uranium residual is shown in Figure 62 where the normalization factor is 4.329. The residual indicates a possible loss of parent U-238 since the residual scatter is below the confidence limit but in this case the statistics were so poor for the highest energy uranium peak at 7.68 Mev that the fit of the synthetic spectrum to this peak was difficult and subject to error. Thus the negative residual below 4.5 Mev could be the result of obtaining an inaccurate normalization factor from the fit at the high energy peak. The average count in the energy distribution of this peak is only 25 counts and one standard deviation represents a 20% possible error. This particular sample was stretching the limits of maximum permissable counting time at 5610 minutes which is almost four full days of counting. The possibility of instrumental shifts becomes significant at this point.

The residual as expected contains a large 5.30 Mev Po-210 peak as a positive residual. The two spike-like peaks at 6.28 and 6.78 Mev in the Uranium spectrum are residuals at peak locations resulting from subtraction
of the thorium spectrum.

Tape No. 230 shown in Figure 63 is the residual uranium alpha spectrum after thorium subtraction for a synthetic rock with 10% U$_3$O$_8$ and 5% ThO$_2$ by weight. The source surface was cut flat, of area 65 sq.cm., counting time 1365 minutes, and normalization factor 0.092. An inset on Figure 63 illustrates the portion of the original spectrum between 7.5 and 9.0 Mev, showing the result of the thorium subtraction. The normalization factor for the inset is 1.076.

The result here is the same as for the synthetic rock of Tape No. 214 (Figure 49) discussed earlier where the thin source surface effect is missing, and the disequilibrium occurs as the result of radon loss, the U-238, U-234, Th-230, and Ra-226 being present in amounts in excess of that required to support the series below Ra-226.

Tape No. 165 shown in Figure 64 is the alpha spectrum of a split drill core sample of pegmatitic uranium ore from Bancroft, Ontario. The surface area was 32 sq.cm., counting time 2880 minutes (2 days), and normalization factor 4.611. The sample contains both thorium and uranium, and in addition several rather intense "thin-source peaks". These can be correlated with peaks from Rn-220 and its daughters in the Th-232 series, and Rn-219 and its daughters of the U-235 series.
TAPE NO. 165.
BMCRT 4-8 SPLIT CR9
AREA (SQ. CM.) 32.0
TIME (MIN.) 2880.0
4.611 X

PEAK MEV ISOTOPE
1 3.35 TH232
2 4.01 TH232
3 5.34 TH208
4 5.42 TH209
5 5.45 RA224
6 5.68 RA224
7 6.04 BI212
8 6.09 BI212
9 6.26 RN220
10 6.79 PO216
11 8.79 PO212

Figure 64
as discussed earlier with respect to Tape No.138 and Tape No.129 (Figures 52, 53). The rock sample is relatively porous and the Radon is out-gassed in the vacuum chamber, producing the "thin-source" effect for all daughters in the series below radon. The thin source peaks shown in Figure 64 are 6.04, 6.09 Mev (Bi-212), 6.28 Mev (Rn-220), 6.62 Mev (Bi-211 from the U-235 series), 6.78 Mev (Po-216), 6.82 Mev (Rn-219 from the U-235 series, undiscernible from the 6.78 Mev peak), 7.38 Mev (Po-215 from the U-235 series), and finally 8.78 Mev (Po-216). The short half lives of these daughters and the long counting time support this conclusion.

The residual uranium alpha spectrum (which includes the thin source peaks discussed above) is shown in Figure 65 with a normalization factor of 1.475. The final residual after uranium subtraction indicates the possibility of slight loss of U-238, since the residual is slightly negative at energies below 4 Mev, although not quite outside the confidence limits. The probability is that the fit is again affected by the poor counting statistics as in the case of Tape No. 172 (Figure 61). In any event the amount of disequilibrium would not be detectable with 99% confidence. The residual spectrum is primarily composed of the thin source spectrum and the 5.30 Mev Po-210 peak, all showing as positive residuals.
TAPE NO. 165.
URANIUM RESIDUAL
AREA (SQ. CM.) 32.0
TIME (MIN.) 2880.0
1.475 X

PEAK MEV ISOTOPE
1 4.13 U238
2 4.18 U238
3 4.59 Ra226
4 4.62 Th230
5 4.76 Th230
6 4.72 U234
7 4.77 U234
8 4.78 Ra226
9 5.39 Po210
10 5.46 Po222
11 6.00 Po218
12 7.66 Po214

Figure 65
Tape No. 143 shown in Figure 66 is the alpha spectrum of a sample of Thorite from Hybla, Ontario. The source was a relatively flat but porous surface, of area 4.0 sq.cm., a counting time of 870 minutes, and a normalization factor of 0.644. This sample also exhibits the thin source effect due to out-gassing of the Rn-220 from the thorium decay series. Subtraction of the thorium contribution produces a residual which indicates there is an insignificant amount of uranium in the sample. There is a small amount of U-235 present as is evidenced by the small peak at 7.38 Mev which can only be correlated with Po-215 of the U-235 series. This would indicate there is also U-238 present, but the only evidence of this is a slight discontinuity in the energy distribution at 7.68 Mev. The amount present would require much longer counting times than would be permissible in order to produce good counting statistics for a fit to the uranium residual.

8.4 Discussion

A note on the effect of the choice of the portion of the energy distribution used for the fit to the synthetic spectrum is useful at this point.

Tape No. 200 shown in Figure 54 is shown again here in Figure 67 with a different region used for the fit of the synthetic spectrum to the experimental spectrum.
TAPE NO. 143.
THORITE 1926
AREA (SQ. CM.) 4.0
TIME (MIN.) 870.0
.644 X

PEAK MEV ISOTOPE

<table>
<thead>
<tr>
<th>No.</th>
<th>MEV</th>
<th>Isootope</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>3.85</td>
<td>TH232</td>
</tr>
<tr>
<td>2</td>
<td>4.01</td>
<td>TH232</td>
</tr>
<tr>
<td>3</td>
<td>5.34</td>
<td>TH228</td>
</tr>
<tr>
<td>4</td>
<td>5.42</td>
<td>TH228</td>
</tr>
<tr>
<td>5</td>
<td>5.45</td>
<td>RA224</td>
</tr>
<tr>
<td>6</td>
<td>5.68</td>
<td>RA224</td>
</tr>
<tr>
<td>7</td>
<td>6.64</td>
<td>B1212</td>
</tr>
<tr>
<td>8</td>
<td>6.69</td>
<td>B1212</td>
</tr>
<tr>
<td>9</td>
<td>6.70</td>
<td>RN220</td>
</tr>
<tr>
<td>10</td>
<td>6.76</td>
<td>P0216</td>
</tr>
<tr>
<td>11</td>
<td>6.78</td>
<td>P0212</td>
</tr>
</tbody>
</table>

Figure 66
The fit in Figure 67 was over 100 channels from channel 650 to 750 (6.5 Mev to 7.5 Mev) which includes the effect of the three small "thin-source" peaks riding on the energy distribution of the infinitely thick source. Consequently, the normalization factor is too low (0.310). Thus the experimental spectrum is too low when multiplied by the normalization factor, resulting in oversubtraction and a negative residual and in this particular example reduces the effective amount of positive residual indicating disequilibrium as discussed for Tape No.200 earlier. In order to eliminate the effect of the three "thin-source" peaks, the fit should be made between channel 695 and 725 (6.95 Mev to 7.25 Mev), which represents a relatively flat portion of the energy distribution. This is the region used for the fit in Figure 54. Then the normalization factor becomes 0.356, and the series below radon, which in itself should normally be in equilibrium, produces a residual within the confidence limits except for the portion due to the thin source peak effects and their energy distribution tails. In general, a poor choice of region of fit is indicated by departure from equilibrium of the radon and immediate daughters which have such short half lives that equilibrium should be expected.

When losses are small, and are within the 99% confidence limits, they may be only apparent losses reflecting a poor fit of the synthetic spectrum. The poor
fit can be either the result of poor counting statistics or thin source peaking effects.
CHAPTER 9

SUMMARY, CONCLUSIONS, AND DISCUSSION

9.1 Introduction

The objective of this study was to develop a method of detecting the presence of disequilibrium in the U-238 decay series in rocks and minerals through the use of alpha particle spectrometry. One important condition to satisfy was that the method should involve a minimum of sample preparation time.

The present thick source alpha particle spectrometric technique satisfies this condition, and permits the detection of radioactive disequilibrium, and further identifies its location in the U-238 decay series within limitations which are primarily instrumental, and can be overcome with detectors of sufficiently high resolution.

9.2 Summary and Conclusions

9.2.1 The Disequilibrium Detection Technique

To summarize, the equilibrium study involves the following steps for a sample containing both thorium
and uranium:

A) Production of the experimental alpha particle spectrum from an infinitely thick source which is under investigation.

B) Computation of a synthetic equilibrium alpha spectrum for the Thorium-232 decay series.

C) Fitting the synthetic Thorium-232 equilibrium spectrum at the location of the highest energy peak to eliminate interference of the U-238 series for the fit.

D) Subtraction of the synthetic Thorium-232 series spectrum. The residual is the alpha spectrum of the Uranium-238 series component.

E) Computation of a synthetic equilibrium alpha spectrum for the U-238 series.

F) Fitting the synthetic U-238 equilibrium spectrum to the uranium residual obtained above in (D). Again the fit is made at the highest energy peak.

G) Subtraction of the synthetic U-238 equilibrium spectrum. The residual spectrum represents the statistical scatter present in all radioactive counting results, and should fluctuate about a residual count value of zero.

H) The departure of any residual spectrum from zero indicates a state of disequilibrium since the experimental and theoretical spectra differ. The location in the residual spectrum where the departure from zero
exists, indicates the location of the radioactive disequilibrium as discussed in Chapter 7. Steps B, C, D, can be eliminated if no thorium is present.

9.2.2 The Example Cases Considered for This Study

The method was applied to a variety of rock and mineral samples, nearly all comprising uranium ore from various parts of the world. The samples under study ranged from badly weathered surface exposures to fresh samples obtained from underground in active uranium mines. Some of the results were almost "text book" cases, while others required considerable explanation of peculiar behaviour. The out-gassing of radon and its "thin-source" effect on the spectrum of porous samples was discussed, as was the absence of the surface "thin-source" component from some synthetic rocks produced for this study. The main limitation of the technique seems to be the low emission rate of alpha particles from some samples which necessitates counting times beyond that permissible to retain good energy resolution without incurring instrumental variation. This limit is probably about 0.1% U₃O₈, the concentrations typical of Blind River conglomeratic uranium ore. Combining 2 or more detectors to provide a larger detector area for counting would enable lower concentrations to be studied.

The subtraction of the Thorium-232 decay series
component from the total alpha spectrum works well; the residual Uranium-238 spectrum being quite amenable to study by this technique, subject to the same limitations of minimum concentration mentioned above.

The samples considered here illustrated cases of radioactive equilibrium, slight disequilibrium, and definite disequilibrium. Disequilibrium was a result of losses of one or more radioelements in the U-238 decay series, and were representative of cases to be expected during routine application of the technique to uraniferous rocks and minerals. Sample preparation time averaged less than half an hour.

9.3 Discussion

The technique for detection and identification of radioactive disequilibrium developed here has applications to uranium mining and exploration. Further improvement in energy resolution would make it possible to do quantitative analyses of the disequilibrium samples, and leads to the possibility of computing the history of the sample with reference to the time of loss of the various daughter products (Rosholt, 1961A, Robinson and Rosholt, 1961). Advances in the 'state of the art' of semi-conductor detectors may result in the development of detectors combining high resolution with large area. Increasing the detecting surface area would result in being able to analyze samples of lower uranium content.
leading to the general application of the technique to any geologic material, even in situ lunar rocks and minerals. This would have special importance for ground-truth studies in the new field of remote sensing by airborne gamma-ray spectrometry, or 'airborne geochemistry' as it is sometimes optimistically called. Definition of geologic units by airborne gamma-ray spectrometric analysis of thorium, uranium, and potassium, and subsequent mapping of such ratios as Th/U or U/K require that the uranium be in equilibrium. The rocks under analysis are the top foot or so of surface, and as such are the most likely to be subject to possible disequilibrium, leading to incorrect results for uranium.

Random representative surface samples could be obtained for equilibrium analyses by this technique. Similarly, any ground gamma-ray spectrometric surveys for in situ analyses of Th, U, and K (Killeen and Carmichael, 1970; Carson, 1970; Doig, 1968) should have some of the survey area checked for equilibrium in the U-238 series to insure valid uranium analyses.
APPENDIX A

DISEQUILIBRIUM DETERMINATION AND PLOTTING PROGRAM

This Appendix combined with the following Appendices represents the complete program and subroutines required for the disequilibrium analyses. Programs are written in Fortran IV for the P.D.P.-10 computer. Total core storage required is 29 K words. This could be decreased somewhat with minor revisions. Typical execution time for complete analysis of 2 rock spectra is 1 minute. This includes compile time for the entire program and subroutines. Plotting is done on a Calcomp drum plotter. Typical plot times are 20 to 30 minutes depending on statistical scatter of the data and consequent pen travel distances.
C THIS T A R I T I U M D E T E R M I N A T I O N A N D P L O T T I N G P R O G R A M. P.G. K I L L E E N  (J A N / 1 9 7 1)
C
C. THE DATA REQUIRED TO GENERATE THE SYNTHETIC SPECTRUM IS READ AND SORTED
C. IN ORDER OF PEAKS OF INCREASING ENERGY. A 1024 CHANNEL THICK SOURCE ALPHA
C. PARTICLE ENERGY SPECTRUM IS THEN GENERATED.
C
C. NEXT THE EXPERIMENTAL SPECTRUM (ROCK DATA) IS READ AND APPROPRIATE GAIN AND
C. PROPORTIONAL SPECTRUM ARE MADE AS REQUIRED. THEN A LEAST SQUARES FIT IS MADE OVER THE
C. CHOSEN ENERGY REGION OF THE SPECTRUM. BETWEEN THE SYNTHETIC AND EXPERIMENTAL
C. SPECTRA. THIS DETERMINES THE VALUE OF THE RATIO BY WHICH THE EXPERIMENTAL
C. SPECTRUM IS MULTIPLIED TO BRING IT INTO COINCIDENCE WITH THE SYNTHETIC
C. SPECTRUM. THE RESIDUAL SPECTRUM IS THEN COMPUTED BY SUBTRACTING THE SYNTHETIC
C. SPECTRUM FROM THIS COINCIDENT SPECTRUM.
C
C. THE EXPERIMENTAL SPECTRUM IS SMOOTHED AND THE THREE SPECTRA ARE PLOTTED...
C. 11 EXPERIMENTAL
C. 21 SMOOTHED EXPERIMENTAL
C. 31 THE RESIDUAL SPECTRUM.
C
C. FINALLY THE PROGRAM CHECKS TO SEE IF MORE EXPERIMENTAL SPECTRA ARE TO BE READ
C. IN FOR DISASSOCIATIVE ANALYSIS.
C
C. MANY VARIABLES NAMES USED IN THE PROGRAM ARE SELF-EXPLANATORY. THOSE REQUIRING
C. EXPLANATION ARE MENTIONED IN COMMENTS THE FIRST TIME THEY APPEAR.
C
DIMENSION FINAL(1024), Y(1024), A(101), C(101)
DIMENSION HDIM(1024), DNXD(1024), X(1024), Y(1024), A(1024), C(1024)
DIMENSION PKENG(301), PKC(301), CONST(301), Y(1024)
DIMENSION NMAX(301), PERCENT(301), ORDER(301), PARENT(301), ISOTOPE(301)
DIMENSION TLIFE(301), T13(301), TLIFE2(301)
C CPINCH REFERS TO COUNTS PER INCH IN THE VERTICAL FOR PLOTTING PURPOSES.
CPINCH=100.0
C NSMITH, IPOLY ARE EXPLAINED IN SUBROUTINE SMOOTH.
NSMITH=0
IPOLY=2
1 FORMAT(4F10.4,A25,A3,F10.4)
2 FORMAT(10X,80H ENERGY PERCENT DECAY ORDER PARENT ISOTOPE
1 1 HALF LIFETIME IN AIR /
3 1 FORMAT(1H1,4X,4F10.4,2X4A5,A3,F10.4)
6 FORMAT(1H1,5X,1024)
9 FORMAT(1H1,22H THORIUM/URANIUM RATIO = F10.4,10X,5HMAX = F10.2)
11 FORMAT(1H1,10H READ SPIKES IN **************/
C
C CASE IS FOR A SHORT 10 LETTER DESCRIP TIVE TITLE.
READS(5,1109) CASE
FORMAT(5A5)
WRITE(6,1109) CASE
1109 FORMAT(10X,25)
C TITLE IS A LONG (40 LETTERS) TITLE.
C = E= FAX ENERGY OF THE ISOTOPE.
C PRECINT=NO. OF DECAYS PER 100 DECAYS OF PARENT IN EQUILIBRIUM.
C PARENT IS A CODE FOR THE DECAY SERIES. PARENT=1.0 FOR TH232. 0.0 FOR U238.
C NDFRR=DECAY ORDER (NOT IMPORTANT)
C TFIF1=TIFF= HALFLIFE OF THE ISOTOPE.
C RANGF=RANG IN &R IN CM.
100 1F(1),1F(2),1F(3),1F(4),1F(5),1F(6),1F(7),1F(8),1F(9),1F(10),1F(11),1F(12),1F(13),1F(14),1F(15)
47  I=I+1
48  GO TO 100
C THURAT=TH/H RATIO. USED AS A DUMMY IN THIS PROGRAM. SET=1.0.
C YFN=N. THE EXPONENT IN THE RANGE-ENERGY RELATION. N IS APPROX. =1.5.
C HCTOT= TOTAL NUMBER OF CHANNELS IN A SPECTRUM.
200 1F(1),1F(2),1F(3),1F(4),1F(5),1F(6),1F(7),1F(8),1F(9),1F(10),1F(11),1F(12),1F(13),1F(14),1F(15)
47  I=I+1
48  GO TO 100
C NUMALE=I-1
40  NUMALE=I-1
41  4 FORMAT(1H1,10X,7THURAT=E10.4,YEN= E10.4,7HCTOT= E10.4)
52  ICHTOT=GHTOT+0.5
C NPAX=NO. OF PAAXS IN THE SYNTHETIC SPECTRUM=NUMALE.
53  NPAX=NUMALE
54  WRITE(6,2)
55  DO 10 I=1,NUMALE
56  10 WRITE(6,3F(I),PRECNT(I),ORDER(I),PARENT(I),ISOTPE(I),TLIFE(I),TLIFE2(I)
C
C ********** ******* ******* ******* ******* SORT *******
C
C START IN ORDER OF INCREASING ENERGY.
60  INSIDE=NUMALE-1
61  DO 20 I=1,INSIDE
62  20 20 I=1,INSIDE
63  DO 60 J=I,NUMALE
64  IIFE(I)=RE(I)
67  TEMP=F(I)
70  F(I)=F(J)
71  F(J)=TEMP
72  TEMP=PRECNT(I)
73  PRECNT(I)=PRCNT(J)
74  PRECNT(J)=TEMP
75  TEMP=ORDER(I)
76  ORDER(I)=ORDER(J)
77  ORDER(J)=TEMP
100  TEMP=PARENT(I)
101  PARENT(I)=PARENT(J)
102  PARENT(J)=TEMP
103  TEMPTP=ISOTPE(I)
104  TEMPTP=ISOTPE(I)
105  ISOTPE(I)=ITEMP
106  TEMP=TIFF(I)
107  TIFF(I)=TIFF(I)
108  TIFF(I)=TEMP
109  TIFF(I)=TIFF(I)

C FORTRAN SOURCE LIST.
TSN  SMIRCF STATEMENT

111  TEMP=TLIFE2(I)
112  TLIFE2(I)=TLIFE2(J)
113  TLIFE2(J)=TEMP
114  TEMP=RANGFI(I)
115  RANGFI(I)=RANGFI(J)
116  RANGFI(J)=TEMP
117  20 CONTINUE

C
C
122  WRITE(6,41) THURAT,YEN,CHTOT
123  201 WRITE(6,21)
124  DO 30 T=1,NUMALE
125  PKENGY(I)=T(I)
126  30 WRITE(6,41)PKENGY(I),PRECENT(I),ORDER(I),PARENT(I),ISOTPE(I),TLIFE2(I),RANGFI(I)
C  IRIGPK=CHANNEL NUMBER CONTAINING THE HIGHEST ENERGY PEAK.
130  IRIGPK=PKENGY(NPEAKS)*100.0
131  WRITE(6,604)IRIGPK
132  604 FORMAT(10X,THIRGPK*,.1F)
C  SYNH150 IS THE SYNTHETIC SPECTRUM TO BE GENERATED BY SUBROUTINE SYNTCH.
133  CALL SYNTCH(YEN,CHTOT,NPEAKS,E,PRECENT,PARENT,RANGE,THURAT,
PENGY,SYNH150,IRIGPK)
134  CALL PRINTSYNH(SYNH150)
C  THE SYNTHETIC SPECTRUM IS SAVED AS ARRAY SAVE1.
135  45 45=1,CHTOT
136  45 SAVF(I)=SYNH150(I)
137  45 WRITE(6,12)
 **************************** READ ROCK DATA *****************************
C
C  NROCKS=NO. OF DECKS OF EXPERIMENTAL SPECTRA TO BE HANDLED.
C  EACH SPECTRUM IS ON 128 CARDS. 8 CHANNELS TO A CARD.
C
141  READ(5,N31NDFCKS
142  31 N5 FORMAT(19)
144  K10
C  NAPF=NO. OF THE ORIGINAL PAPER TAPE FROM THE PULSE HEIGHT ANALYZER.
C  T=4 LETTER TITLE.
C  DIST=SOURCE DETECTOR SEPARATION.
C  H=7 CHANNEL NO. FOR A 3 MEV PULSE BEFORE AND AFTER COUNTING THE SPECTRUM.
C  A=A=7 CHANNEL NO. FOR A 9 MEV PULSE BEFORE AND AFTER COUNTING THE SPECTRUM.
147  25 FORMAT(13,4A5,5A,F6.2,5F8.2,F8.1)
150  WRITE(6,6NAPET1),T(1),T(2),T(3),T(4),T(5),AREA,DIST,TIME,R3,A3,R9,A9
151  6 FORMAT(10X,13,4A5,5A,F6.2,5F8.2,F8.1/F)
C  GSHIFT=GAINE SHIFT DESIRED.
C  7SHIFT=ZERO SHIFT DESIRED.
152  READ(5,971GSHIFT,7SHIFT)
C  ISTART=CHANNEL AT WHICH THE LEAST SQUARE FIT STARTS.
C  NSOPT=NO. OF LEAST SQUARE POINTS. (MAX. OF 100 WITHOUT A DIMENSION CHANGE)
153  READS,971ISTART,NSOPT
154  971 FORMAT(25)
157  WRITE(6,9761ISTART,NSOPT)
160  976 FORMAT(10X,THIRGPK*,.1F,10X,THIRGPK*,.1F)
C  ****  S A V E  NSOPT CHANNELS OF SYNTCHETIC SPECTRUM.  *****
161  990 AT(I)=SYNH150(I),ISTART1
162  990 ** ** ** ** ** ** ** ** ** ** ** **
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*06/30/71*

164 22 FORMAT(1X,2F10.5)
165 WRITE(6,24)SHIFT,7SHIFT
166 24 FORMAT(15X,7HSHIFT=F10.5,7HSHIFT=F10.5)
167 *) XT=A COPY OF TITLE T
168 66 XT(I)=T(I)
169 C Y=THE COUNTS IN THE EXPERIMENTAL SPECTRUM.
170 READ(5,43)(Y(I),I=1,1,CHTOT)
177 43 FORMAT(F6.0,7F7.0)
200 CALL PRINTY(V)
201 C
202 C
203 C
204 C
205 C
206 C
207 C
208 C
209 C
210 C
211 C
212 C
213 C
214 C
215 C
216 C
217 C
218 C
219 C
220 C
221 C
222 C
223 C
224 C
225 C
226 C
227 C
228 C
229 C
230 C
231 C
232 C
233 C
234 C
235 C
236 C
237 C
238 C
239 C
240 C
241 C
242 C
243 C
244 C
245 C

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**P. KILLEN**

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**TSN**

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**SOURCE STATEMENT**
FORTRAN SOURCE LIST

246 FORMAT(1X,18H1 PRINT ERROR)  //1
247 CALL PRINT (ERROR)

********** ******** ** P L O T T I N G A R E A ******

C NTYP USE OF 30 INCH PAPER
250 CALL PLOT(60,60,0,29.5,3)
251 CALL PLOT(0,0,0,3)
C R F L I C A T E ORIGIN AT X=2.0, Y=12.75. THIS GIVES ROOM FOR NEGATIVE RESIDUAL PLOTS
252 CALL PLOT(2,0,12.75,3)
C X = DISTANCE ALONG PAPER IN INCHES.
253 X=0.02
C P L O T R A W D A T A
254 TAPS=NAPE
255 M=40 T=200.900
256 X=0.02
257 TEMP=1/11/CPINCH
C NTYP... 16.60 IS A SAFETY FACTOR TO PREVENT PLOTTING OFF THE PAPER.
260 IF(TEMP.GT.16.60) TEMP=16.60
262 IF(TEMP.LE.0.0) TEMP=0.0
266 CALL SYMNAT(X,TEMP,0.0,2.0,0.0,40)
267 RO CONTINUE
271 WRITE(6,6)NAPE,T(11),T(21),T(3),T(4),T(5),AREA,DIST,TIME,B3,A3,B9,A9
272 CALL SMOOTH(NAPE,Y,SMITH,IPOLY,FINAL)
273 X=0.02
C P L O T S M O O T H F I D A T A
274 M=81 T=900.200,1
275 X=0.02
276 TEMP=FINAL/11/CPINCH
277 IF(TEMP.GT.16.60) TEMP=16.60
278 IF(TEMP.LE.0.0) TEMP=0.0
280 CALL PLOT(X,TEMP,11)
286 RO CONTINUE
290 CALL PLOT(0,0,0,3)

******* TITLE PLOTTING AREA ******
C XSPOT, YSPOT = LOCATION FOR THE TITLE OF THE PLOT.
311 XSPOT=0.5
312 YSPOT=0.5
313 CALL PLOT(XSPOT,YSPOT,3)
314 HITF=0.28
315 CALL SYMNAT(XSPOT,YSPOT,HITF,8,TAPE NO.0.0,0.01)
316 XSHIFT=XSPOT+0.557,HITF=14.0,0.01
317 CALL NUMFRACT(XSHIFT,YSPOT,HITF,TAPE,0.0,0.01)
320 DNP=HITF+0.5
321 YSHIFT=YSPOT-DNP
322 XSPEC=XSPOT-0.1
323 CALL SYMNAT(XSPEC,YSHIFT,HITF,T0.0,0.24)
324 YSHIFT=YSHIFT-DNP
325 CALL SYMNAT(XSPEC,YSHIFT,HITF,14,TAPE (50 CM),0.0,0.14)
326 XSHIFT=XSPOT+0.557,HITF=14.0,0.01
327 CALL NUMFR(XSHIFT,HITF,TIME,0.0,0.11)
330 YSHIFT=YSHIFT-DNP
331 CALL SYMNAT(XSPEC,YSHIFT,HITF,11,TIME (MIN.),0.0,0.11)
332 XSHIFT=XSPOT+0.557,HITF=11,0.0,0.11
335 CALL NUMFR(XSHIFT,HITF,TIME,0.0,0.11)
CALL PLOT(0.0,0.0,3)

C $X_{\text{LENGTH}}$ is the length of the X axis in inches.

XLENGTH=14.0

YLENGTH=7.0

DELTAX=1.0

DELTAY=100.0

C $\text{DELTAY}$ is the increment to be added to zero as the Y axis numbers are plotted.

C $\text{XAXIS}$ prepares to plot residuals and error limits.

CALL XAXIS(XLENGTH,DELTAX)

CALL YAXIS(YLENGTH,DELTAY)

C $\text{CONTINUE}$

CALL PLOT(0.0,0.0,3)

X=X+0.02

N=100 I=200,500

X=X+0.02

CALL PLOT(X,ERROR(I),1)

N=102 I=900,200,-1

ERROR(I)=ERROR(I)*(-1.0)

IFERROR(I)*LT.-10.0 ERROR(I)=10.0

CALL PLOT(X,ERROR(I),2)

X=X+0.02

N=107 CONTINUE

CALL AXIS(0.0,-1.0,8RESIDUAL,8.2,0.0,0.0,-100.0,0.0,CPINCH,0,1.0,1.0)

CALL PLOT(0.0,0.0,3)

CALL PLOT(14.0,0.0,2)

CALL PLOT(14.0,0.0,3)

CALL PRINT

CALL END

NDECKS=NDECKS=1

IF(NDECKS.LE.01) GO TO 111

111 CONTINUE

GO TO 420

STOP

C $\text{RROUTINES}$

SUFFIX RSYNTHA,

C SUFFIX RLSY,

C SUFFIX RSMY,

C SUFFIX RPRNTA,

C SUFFIX RESLC,

C SUFFIX SHY,

C SUFFIX XAXIS, AND YAXIS.

END
0 NAME SYNTHA
1
define subroutine SYNTHA(YEN,ICHOT,NP,PEAK,F,PERC,PAN,RANGE,THIRAT,PKNY,SYN150,IRGT)
2
C
4 C CAN PRODUCE A SYNTHETIC INFINITELY THICK SOURCE SPECTRUM OF THE ENERGIES
5 C OF ALPHA PARTICLES OF THE HC-238 SERIES AND/OR THE TH-232 DECAY SERIES. A MAX.
6 C OF 30 PEAKS CAN BE HANDLED BUT THIS CAN BE EASILY INCREASED BY CHANGING THE
7 C DIMENSIONS. REF. APPOSINOV AND KOCHAROV,1962.
8 C THIRAT=TH/THOO WHEN DATA FROM BOTH SERIES IS ENTERED.
9 C PERCNT=1.0 FOR TH-232 SERIES = 0.0 FOR HC-238 SERIES.
10 C THE RATIO OF SPECIFIC ACTIVITIES OF TH/THOO=246,0/733,6.
11 C DNDE=COUNT DISTRIBUTION OVER RANGE OF ENERGIES FROM 0.01 MEV TO THAT OF THE
12 C PEAK CHANNEL.
13 C SYN150=SYNTHETIC SPECTRUM=SUM OF INDIVIDUAL PEAK ENERGY DISTRIBUTIONS.
14 DIMENSION F(30),PERC(30),PAN(30),RANGE(30),PKNY(30)
15 /SYN150/1 1 1 1 1 1 1
16 WRITE(6,1) 1 FORMAT(2X,31HNOW EXECUTING SYNTHETIC SYNTHA/)
17 DO 502 I=1,ICHOT
18 DNIT=0.0
19 507 SYN150(I)=0.0
20 DN 701 L=1,30
21 701 YEN(I)=YEN=1.0
22 IF(PERCNT(I).EQ.1.0) GO TO 501
23 PKNY(I)=PERC(I)
24 GO TO 503
25 501 PKNY(I)=PERC(I)*THIRAT*246.0/733.6
26 503 ACNST=PKNY(I)*PKNY(I)*CNST(I)
27 DO 500 K=1,PEAK
28 IF(PERCNT(K).EQ.1.0) GO TO 504
29 DN 504 CNST(K)=(RANGE(K)/RANGE(11))**ACNST*(PKNY(I)**CNST(I)**YN)
30 GO TO 500
31 504 CNST(K)=(RANGE(K)/RANGE(11))**ACNST*(PKNY(I)**CNST(I)**YN)
32 500 CONTINUE
33 DN 600 K=1,PEAKS
34 IPKCN(I)=PKNY(I)**100.0
35 DN 600 T=1,IPKCN
36 DALKPA=0.0
37 DN 600 T=1,IPKCHN
38 DALKPA=DALKPA+0.01
39 DN 600 K=1,PEAK
40 IF(DNDE(I).EQ.0.0) GO TO 703
41 DN 703 DNDE(I)=0.0
42 DN 704 SYN150(I)=SYN150(I)+DNDE(I)
43 704 CONTINUE
44 C NORMALIZE SO THAT HIGHEST ENERGY PEAK = 125 COUNTS HIGH.
45 C ARBITRARY. (SET FOR PLOTTING PURPOSES).
46 XNML(I)=125.0/SYN150(I) IRGT
47 DN 605 T=1,ICHTOT
48 605 SYN150(I)=SYN150(I)*XNML(I)
49 RETURN
50 END
0 $IRFC 1 SQ
1 SUBROUTINE LSQ(X,XTITLE,Y,YTITLE,N)
C
C    FAST SQUARE FIT.      P.G.KILEEN, GEOPHYSICS, U.W.O. 1971
C
C    SIMPLE LEAST SQUARES COMPARISON OF TWO SPECTRA, UP TO 100
C    CHANNELS AT A TIME.
C
C    R=CORRELATION COEFFICIENT
C    SERFST=STANDARD ERROR OF ESTIMATE.
C    XPLT=VAR=XPLTINFO VARIANCE, TOTVAR=TOTAL VARIANCE.
C    P=RATIO OF Y/X I.E. YVAR
C
C    N,N ARE THE CHANNELS BETWEEN WHICH THE CALCULATIONS ARE DONE.
2    DIMENSION X(101),Y(101),YCALC(101),RES(101)
3    DIMENSION XTITLE(5),YTITLE(5)
4    N=1
5    NLSOPT=N
6    WRITE(6,51)XTITLE(I),I=1,51
13   5 FORMAT(10X,45S,4/1)
14    WRITE(6,21)X(I),I=M,N
21   21 FORMAT(10X,4F8.1)
26    WRITE(6,25)YTITLE(I),I=1,51
25   25 FORMAT(10X,4F8.1)
33    2 FORMAT(10X,16F8.1)
40   50 SUMX=0.0
40   SUMXY=0.0
40   SIMKO=0.0
77   DO 10 I=M,N
40   SUMX=SUMX+X(I)
41   SUMXY=SUMXY*X(I)+Y(I)
42   SIMX=G*SUMXS+X(I)*X(I)
43   10 CONTINUE
47   10 CONTINUE
45   YVAR=SUMY/N
46   R=SUMXY/SUMXYS
47   XPLVAR=0.0
50   TOTVAR=0.0
51   SRES=0.0
52   DO 20 I=M,N
53   YCALC(I)=X(I)*R
54   RES(I)=Y(I)-YCALC(I)
55   SRES=SRES+RES(I)**2
56   XPLVAR=XPLVAR+YCALC(I)-YVAR)**2
57   TOTVAR=TOTVAR+(Y(I)-YBAR)**2
60   20 CONTINUE
62   4 FORMAT(10X,8F8.1)
63   WRITE(6,41)XPLVAR,TOTVAR
64   41 FORMAT(10X,8F8.1)
65   R=SORT(XPLVAR/TOTVAR)
66   SERFST=SORT(SRES/NLSOPT)
67   3 FORMAT(1H,2X,15.3H,N=15.3H YBAR=,F10.2,3H R=,F10.4,12H STD ER
1EST=,F10.4,3H R=,F10.4)
70   WRITE(6,21)RES(I),I=M,N
75   100 CONTINUE
76   RETURN
77   END

0 $IRFTC XAXIS
1 SUBROUTINE XAXIS(XLENSH,DELTAX)
   C
3 C X AXIS LABELLED ENERGY. DESIGNED TO OVERCOME LIMITATIONS ON SIZE OF LABELS
4 C IMPOSED ON THE USER BY CANNED AXIS ROUTINES OF THE COMPUTER CENTER.
5 CALL PLOT(0.0,0.0,0.3)
6 DIST=0.14
7 DELTA=2.0
8 300A CALL NUMRFT(-0.35,0.21,DELTAX,0.0,0.1)
9 DELTA=DELTAX+DELTAX
10 DIST=DIST+2.0
11 IF(DIST.GT.XLENSH) GO TO 3009
12 GO TO 300A
13 3009 TICK=-0.10
14 CALL PLOT(XLENSH,TICK,31)
15 DASH=XLENSH
16 CALL PLOT(DASH,0.0,21)
17 DASH=DASH+1.0
18 CALL PLOT(DASH,0.0,21)
19 DASH=DASH+1.0
20 IF(DASH.LT.0.0) GO TO 3010
21 GO TO 3007
22 3010 CONTINUE
23 TITLOC=XLENSH/2.0,1.0
24 CALL SYMN(TITLOC,-0.56,0.21,11,ENERGY(MEV),0.0,11)
25 RETURN
26 END

0 $IRFTC XAXIS
1 SUBROUTINE XAXIS(YLENSH,DELTAY)
2 CALL PLOT(0.0,0.0,0.3)
3 DIST=0.14
4 DELTA=0.0
5 200A CALL NUMRFT(-0.14,DIST,0.21,DELTAY,90.0,0.01)
6 DELTA=DELTA+DELTAY
7 DIST=DIST+1.0
8 IF(DIST,GT,YLENSH) GO TO 2009
9 GO TO 200A
10 2009 TICK=-0.10
11 CALL PLOT(TICK,YLENSH,31)
12 DASH=YLENSH
13 CALL PLOT(0.0,DASH,21)
14 DASH=DASH+1.0
15 CALL PLOT(DASH,0.0,21)
16 CALL PLOT(TICK,DASH,21)
17 CALL PLOT(DASH,0.0,21)
18 DASH=DASH+1.0
19 IF(DASH,LTE0.0) GO TO 2010
20 GO TO 2007
21 2010 CONTINUE
22 TITLOC=YLENSH/2.0,1.0
23 CALL SYMN(-0.42,TITLOC,0.21,13,NO. OF COUNTS,90.0,131)
24 RETURN
25 END
FORTRAN SOURCE LIST

06/30/71

0 SAVE RESCL
1 SUBROUTINE RESCL(YSAVE,RESSO,SYNR0,PAT10,RSMN)

C
C
C PAT10 IS RESULT FROM SUBROUTINE LSO.
2 DIMENSION YSAVE(1024), SYNR0(1024)
3 DIMENSION Y(1024), RSMN(1024)
4 WRITE(6,11)
5 1 FORMAT(2X,31HWNOW EXECUTING SUBROUTINE RESCL/)  
6 SRS=0.0
7 SRS0=0.0
8 N=400 I=200, 900
9 Y(I)=YSAVE(I)*RATIO
10 RSMN(I)=Y(I)-SYNR0(I)
11 SRS=SRES+RESI(I)*RESI(I)
12 RSMN=SRES/SRS*RSMN
13 RESO=ABS(SRS0-SRS)
14 44 44 FORMAT(10X,25HRS, RESMN, RESSO= )
15 RETURN
16 END

06/30/71

0 SAVE PRINTR
1 SUBROUTINE PRINTR(TEMP)

C
C
C PRINTS ANY 1024 CHANNEL SPECTRUM WITH EIGHT CHANNELS PER LINE, ASSIGNING A
C CONSECUTIVE NUMBER TO EACH LINE TO FACILITATE IDENTIFICATION OF ANY CHANNEL.
2 DIMENSION TEMP(1024)
3 1 FORMAT(10X,RF10.4,5X,14)
4 N=1
5 N=A
6 N=10 T=1:128
7 WRITE(6,11)TEMP(K),K=N,N+1
14 N=N+1
15 N=10
16 CONTINUE
20 RETURN
21 END
APPENDIX B
SPECTRUM SMOOTHING PROGRAM

```
06/30/71
P. M. KILLEN

FORTRAN SOURCE LIST

100  START: SMOOTH
110  DIMENSION FINAL(1024), COUNT(1024), SCOUNT(1024), W(1024)
120  COMMON SMOOTH(ICHOT, COUNT, NSMOTH, IPOLY, FINAL)
140  COMMON SAVITYKY AND OLAYE, 1964, ANAL. CHEM., VOL. 36, NO. 8, P. 1627.
150  COMMON THE WEIGHTING FACTORS USED IN THIS PROGRAM ARE FROM THE ABOVE REFERENCE.
160  COMMON COUNT IS THE ARRAY OF RAW DATA POINTS, SCOUNT IS SMOOTHED POINTS ARRAY.
170  COMMON ICHOT IS THE NUMBER OF RAW DATA POINTS (NO. OF CHANNELS)
180  COMMON NSMOTH IS THE NUMBER OF POINTS TO BE USED IN THE SMOOTH
190  COMMON IPOLY IS THE DEGREE OF THE POLYNOMIAL USED FOR THE SMOOTH
200  COMMON NTOTAL = COUNT(1) + SCOUNT(1) + SCOUNT(1024) - 1
210  COMMON CFRONT = COUNT(1) + SCOUNT(1) + SCOUNT(1024) - 1
220  COMMON WRITF = WRITE(6, 11NSMOTH)
230  COMMON I = 1
240  COMMON NYTOTAL = COUNT(1) + SCOUNT(1) + SCOUNT(1024) - 1
250  COMMON CFRONT = COUNT(1) + SCOUNT(1) + SCOUNT(1024) - 1
260  COMMON WRITF = WRITE(6, 11NSMOTH)
270  COMMON I = 1

100  IF(CFRONT.EQ.5) ASSIGN 111 TO KICK
110  IF(CFRONT.EQ.7) ASSIGN 222 TO KICK
120  IF(CFRONT.EQ.9) ASSIGN 333 TO KICK
130  GOTO KICK(111, 222, 333)
140  IF(CFRONT.EQ.1) ASSIGN 19 TO NURDLE
150  IF(CFRONT.EQ.4) OR. IPOLY.EQ.31 ASSIGN 29 TO NURDLE
160  GOTO 334
170  IF(CFRONT.EQ.2) OR. IPOLY.EQ.31 ASSIGN 39 TO NURDLE
180  IF(CFRONT.EQ.4) OR. IPOLY.EQ.51 ASSIGN 49 TO NURDLE
190  GOTO 334
200  IF(CFRONT.EQ.2) OR. IPOLY.EQ.31 ASSIGN 59 TO NURDLE
210  IF(CFRONT.EQ.4) OR. IPOLY.EQ.51 ASSIGN 69 TO NURDLE
220  NN=NSMOTH-1
230  GOTO 200 I=1,15SMOTH
240  I=I+NN
250  NN=3NN
260  KK=KK1
270  KSMOTH=COUNT(I)
280  GOTO NURDLE(19, 29, 39, 49, 59, 69)
290  SCOUNT(I(IPOLY)=I(I(IF(4)+6)+I(IF(3)+IF(5)))+3*I(IF(2)+IF(6)))-2*I(IF(1)+IF(17))/2
```

192
06/30/71
SMOOTH
FORTRAN SOURCE LIST

GO TO 200
112 49 SCOUNT(1)=131.0*(F(4)+15.0*(F(5)+30.0*(F(6)+5.0*(F(1)+F(7))
113 111)+231.0)
114 GO TO 200
115 59 SCOUNT(1)=59.0*(F(5)+54.0*(F(4)+61.0)+39.0*(F(3)+F(7))+14.0*(F(2)+
116 (F(1)+21.0)*(F(1)+F(9)+11)+231.0)
117 GO TO 200
118 69 SCOUNT(1)=179.0*(F(5)+135.0*(F(4)+F(6)+30.0*(F(3)+F(7))+55.0*(F(2)+
119 (F(1)+15.0)*(F(1)+F(9)+11)+429.0)
120 200 CONTINUE
121 1 FORMAT(1H1,10X,35HNUMBER OF POINTS FOR THE SMOOTH IS
122 /1H4,2X,6HPO
123 /1HNTS //
124 2 FORMAT(1H4,10X,20HILLEGAL VALUE OF /1H4,2X,4HPOINTS HAS BEEN
125 /1HCHosen FOR THE SMOOTHING. /56H I HAVE TAKEN THE LIBERTY OF USING
126 /2H5 POINTS INSTEAD. /61H NEXT TIME CHOOSE EITHER 5 POINTS
127 /2H, 7 POINTS, OR 9 POINTS. //
128 3 FORMAT(1H4,10X,20HILLEGAL VALUE OF /1H4,7HFOR THE DEGREE OF
129 /1HTHE POLYNOMIAL USED IN SMOOTHING HAS BEEN CHosen.
130 /1H200H I HAVE TAKEN THE OPTION OF USING A 3RD DEGREE POLYNOMIAL AS IT
131 /2HIS SOMEWHAT LESS TEDIOUS FOR ME. /7IH NEXT TIME PLEASE C
132 /2HHOSE EITHER 2ND, 3RD, 4TH OR 5TH DEGREE.
133 //
134 4 FORMAT(1H4,10X,40HTHE POLYNOMIAL USED FOR SMOOTHING IS A
135 /1H4,25H
136 /1H7DEGREE POLYNOMIAL. //
137 5 FORMAT(1H4,10X,10HUNFORTUNATELY I DO NOT HAVE THE CONVOLUTES FO
138 /1H4, 5 POINT SMOOTH WITH A 4TH OR 5TH DEGREE POLYNOMIAL
139 /281H (SEE SAVITZY AND GOLAY. 1964. IN ANAL. CHEM., VOL 36, NO.8
140 /3P31627 ). /1H90H I HAVE TAKEN THE OPTION OF GIVING YOU A 7
141 /4HPOINT SMOOTH WITH A 5TH DEGREE POLYNOMIAL.
142 //
143 C
144 /1H
145 /2H
146 /3H

C
FINAL DATA PREPARATION FOR PLOTTING AND PRINTING.

67 NN=NSMITH+1
126 CN=N2=NN/2
130 MM=M=ICHOT+NN2
131 JM=MM+1
132 66 NF=MM+1
133 69 FINAL(MM)=0
134 5 IF(MM.EQ.0,GO TO 66
137 677 GO TO 66
140 667 JACK=0
141 JM=MM+1
142 69 ON 50 IF(MM,M
143 677 JACK=JACK+1
144 69 FINA(L(I)=SCOUNT(JACK)
145 50 CONTINUE
146 MJ=MM+1
147 69 23 IF(MM.MM2)
148 69 21 IF(MM.EQ.ICHOT+GO TO 24
149 69 21 IF(MM.MM2)
150 69 23 GO TO 23
151 69 24 CONTINUE
152 577 WRITE(5,501)NSMITH,IPOLY
153 60 CALL PRINTF(FINAL)
154 501 FORMAT(1H,7X,25HFIRST SMOOTH OF RAW DATA /,7HIT IS A,14,45H P
155 75X 10 POINT SMOOTH WITH A POLYNOMIAL OF DEGREE
156 14 /1
157 RETURN
158 ENDD
APPENDIX C
SPECTRUM GAIN AND ZERO SHIFT PROGRAM

FORTRAN SOURCE LIST

0654 P KILLEN
06/30/71

00100 SOURCE STATEMENT

00200 FORTRAN SOURCE LIST

00300 SOURCE STATEMENT

00400

00500 0 START SHIFTV
00600 1 SUBROUTINE SHIFTV(COUNT,ICHOT,EZERO,GSHIFT,ZSHIFT)
00700 2 C
00900 4 C
01000 5 GAIN=(DIFFERENCE BETWEEN TWO ENERGIES)/(DIFFERENCE BETWEEN CHANNEL NUMBERS)
01100 6 ASSOCIATED WITH THE TWO ENERGIES).
01200 7 GSHIFT=M EASURED GAIN/DESIRED GAIN.
01300 8 D. B. SCHONFELD, et al., 1966, DETERMINATION OF NUCLIDE CONCENTRATIONS IN
01400 9 SOLUTIONS CONTAINING LOW LEVELS OF RADIOACTIVITY, BY LEAST SQUARES
01500 10 RESOLUTION OF THE GAMMA-RAY SPECTRA, NUCL. INSTRUM. AND M ETH., VOL. 45, PP. 1-21.
01600 11 C
01700 12 C NOTE THAT THE PROGRAMS THAT SCHONFELD REFERS TO HAVE ERRORS AND REQUIRE
01800 13 C TESTING AND REVISION.
01900 14 C
02000 29 DIMENSION COUNT(1024), CI(1024)
02100 30 00 I=1,ICHOT
02200 31 00 5 CI(1) = 0.0
02300 32 00 TF=7/FRM*(GSHIFT-1.0)
02400 33 00 7=ICHOT
02500 34 00 MX=6/GSHIFT*(7+ZSHIFT)+TF
02600 35 00 MX=MX+1
02700 36 00 WRITE(6,29)
02800 37 00 IF(ISTART.GT.0.1) ISTART=1
02900 38 00 IF(IEND.GT.ISTART) IEND=ICHOT
03000 39 00 DO 10 J=1,ICHOT
03100 40 00 7=J
03200 41 00 QI=GSHIFT*(7+ZSHIFT)+TF
03300 42 00 QJ=GSHIFT*(7+ZSHIFT)+TF
03400 43 00 IF(ISTART.LT.0.1) ISTART=1
03500 44 00 IF(IEND.GT.QJ) IEND=ICHOT
03600 45 00 IF(IEND.GT.ISTART) IEND=ICHOT
03700 46 00 IF(IEND.GT.ISTART) IEND=ICHOT
03800 47 00 IF(IEND.GT.ISTART) IEND=ICHOT
03900 48 00 IF(IEND.GT.ISTART) IEND=ICHOT
04000 49 00 IF(IEND.GT.ISTART) IEND=ICHOT
04100 50 00 IF(IEND.GT.ISTART) IEND=ICHOT
04200 51 00 IF(IEND.GT.ISTART) IEND=ICHOT
04300 52 00 IF(IEND.GT.ISTART) IEND=ICHOT
04400 53 00 IF(IEND.GT.ISTART) IEND=ICHOT
04500 54 00 IF(IEND.GT.ISTART) IEND=ICHOT
04600 55 00 IF(IEND.GT.ISTART) IEND=ICHOT
04700 56 00 IF(IEND.GT.ISTART) IEND=ICHOT
04800 57 00 IF(IEND.GT.ISTART) IEND=ICHOT
04900 58 00 IF(IEND.GT.ISTART) IEND=ICHOT
05000 59 00 IF(IEND.GT.ISTART) IEND=ICHOT
05100 60 00 IF(IEND.GT.ISTART) IEND=ICHOT
05200 61 00 IF(IEND.GT.ISTART) IEND=ICHOT
05300 62 00 IF(IEND.GT.ISTART) IEND=ICHOT
05400 63 00 IF(IEND.GT.ISTART) IEND=ICHOT
05500 64 00 IF(IEND.GT.ISTART) IEND=ICHOT
05600 65 00 IF(IEND.GT.ISTART) IEND=ICHOT
05700 66 00 IF(IEND.GT.ISTART) IEND=ICHOT
05800 67 00 IF(IEND.GT.ISTART) IEND=ICHOT
05900 68 00 IF(IEND.GT.ISTART) IEND=ICHOT
06000 69 00 IF(IEND.GT.ISTART) IEND=ICHOT
06100 70 00 IF(IEND.GT.ISTART) IEND=ICHOT
06200 71 00 IF(IEND.GT.ISTART) IEND=ICHOT
06300 72 00 IF(IEND.GT.ISTART) IEND=ICHOT
06400 73 00 IF(IEND.GT.ISTART) IEND=ICHOT
06500 74 00 IF(IEND.GT.ISTART) IEND=ICHOT
06600 75 00 IF(IEND.GT.ISTART) IEND=ICHOT
06700 76 00 IF(IEND.GT.ISTART) IEND=ICHOT
06800 77 00 IF(IEND.GT.ISTART) IEND=ICHOT
06900 78 00 IF(IEND.GT.ISTART) IEND=ICHOT
07000 79 00 IF(IEND.GT.ISTART) IEND=ICHOT
07100 80 00 IF(IEND.GT.ISTART) IEND=ICHOT
07200 81 00 IF(IEND.GT.ISTART) IEND=ICHOT
07300 82 00 IF(IEND.GT.ISTART) IEND=ICHOT
07400 83 00 IF(IEND.GT.ISTART) IEND=ICHOT
07500 84 00 IF(IEND.GT.ISTART) IEND=ICHOT
07600 85 00 IF(IEND.GT.ISTART) IEND=ICHOT
07700 86 00 IF(IEND.GT.ISTART) IEND=ICHOT
07800 87 00 IF(IEND.GT.ISTART) IEND=ICHOT
07900 88 00 IF(IEND.GT.ISTART) IEND=ICHOT
08000 89 00 IF(IEND.GT.ISTART) IEND=ICHOT
08100 90 00 IF(IEND.GT.ISTART) IEND=ICHOT
08200 91 00 IF(IEND.GT.ISTART) IEND=ICHOT
08300 92 00 IF(IEND.GT.ISTART) IEND=ICHOT
08400 93 00 IF(IEND.GT.ISTART) IEND=ICHOT
08500 94 00 IF(IEND.GT.ISTART) IEND=ICHOT
08600 95 00 IF(IEND.GT.ISTART) IEND=ICHOT
08700 96 00 IF(IEND.GT.ISTART) IEND=ICHOT
08800 97 00 IF(IEND.GT.ISTART) IEND=ICHOT
08900 98 00 IF(IEND.GT.ISTART) IEND=ICHOT
09000 99 00 IF(IEND.GT.ISTART) IEND=ICHOT
09100 100 FORMAT(10X,2F9.1) COMMENT: GOOD AFTER CHANNEL 16
09200 101 ON 11 I=1,ICHOT
09300 102 CONTINUE
09400 103 WRITE(6,100)
09500 104 FORMAT(10X,2F9.1)
09600 105 IF(IEND.GT.QI) GOTO 20
09700 106 CONTINUE
09800 107 RETURN
09900 110 END
10000

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APPENDIX D

PROGRAM TO READ SPECTRA FROM PAPER TAPE

NOTE: descriptive comments beginning with the
letter C are not part of the program.

C assign the name 'T' to dectape # 224, write enabled.
$ASSIGN DTA224 T WREN

C assign the name 'D' to the disc area
$ASSIGN DSK D

C assign the name 'R' to the paper tape reader
$ASSIGN PTR R

C print listing of all files on T
$PIE TTY:/L+T:

C request operator to load paper tape with name TAPE.247
C onto the paper tape reader
$PAUSE LOAD TAPE.247 INTO READER PLS

C SPECTR is the paper tape to dectape conversion
C program, on file on dectape 224
$RUN T SPECTR

C store this result on disc with file name TAPE.247
$PIE D:TAPE.247+D:SPECT.DAT/R

C copy this result onto the dectape
$PIE T:TAPE.247+D:TAPE.247

C at this point another paper tape may be loaded and
C the previous four instructions repeated using the
C name of the new paper tape (e.g. TAPE.248)
C
C print listing of all files on T
$PIE TTY:/L+T:
$EOJ

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PROGRAM TO PUNCH SPECTRA ONTO CARDS

$ASSIGN DTA224 T
   C assign the name 'B' to the interface buffer between
   C the dectape drives and the card puncher
$ASSIGN IFB B
   C copy TAPE.247 from the dectape onto disc
$PIP DSK:/XT:TAPE.247
   C at this point other tapes may be copied onto disc
   C using the previous statement with the new tape number
   C
   C run the special program which punches cards
$R PRRD
HELLO:010
   C punch TAPE.247 onto cards
CRD:DSK:TAPE.247
   C at this point additional files may be punched onto
   C cards by repeating the previous instruction with
   C the new file name (e.g.TAPE.248)
$EOJ

NOTE: the file names TAPE.247, TAPE.248 etc. refer to
individual alpha spectra which were given these
names to correspond with the paper tape number
assigned at the time the spectrum was transferred
from the pulse height analyzer memory to paper tape.
The file name consists of a main name and an ex-
tension of three characters separated by a dot. Thus
for example, the spectra could have been named ALPHA.92
or A247.PGK as desired.

On the other hand DECTAPE 224 is a fixed number
of a particular reel of magnetic tape kept in the
computer center tape library.
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