The seabed is envisaged to meet the increased future demands for minerals from the rapidly growing industrialized societies of the world. Shipboard analysis of cores can significantly reduce the cost and time spent at the exploratory drilling stage by obviating the need to go back to land for analysis. It can further speed the exploration process by enabling a quick modification of the exploration plan based on the results of the shipboard analysis.

A $^{252}$Cf-based analyzer utilizing the prompt gamma neutron activation analysis technique has been designed. The analyzer is a spherical iron shell with the source at its center. The seabed core is passed through a hollow composite tube which is positioned a short distance directly below the source and the resulting prompt gamma rays are collimated to a HPGe detector. The rest of the sphere is filled with paraffin. The gamma ray flux at the detector is converted into a count rate by using a semi-empirical detector response function. This count rate data are then used to determine the sensitivity and detection limits for the chosen elements (Mg, Al, Ti, Cr, Mn, Fe, Ni and Cu).
Monte Carlo simulations using the Monte Carlo neutron photon coupled transport code, MCNP, were carried out for a parametric study of important variables influencing the design of the analyzer. These parameters included the moderator type, source to sample distance and sample porosity. MCNP was then used to model the analyzer and to generate the neutron flux profiles in the sample and the prompt gamma flux at the detector.

Due to the non-availability of the prompt gamma data for most of the elements of economic interest in the ENDF/B-V cross section libraries associated with MCNP, the point kernel photon transport code ISOSHLD-II (modified for high energy gamma rays) was used to generate the gamma flux at the detector for specific elements. The ISOSHLD-II source term was calculated based on known gamma production data (thermal capture only) and the thermal neutron flux in the sample obtained from MCNP computations.

The sensitivity and detection limits obtained from the isotopic source based analyzer were compared for the case of aluminum with values reported from reactor facilities.

The results obtained indicate that the analyzer designed in this work could prove suitable for the on-line analysis of many elements of economic interest in seabed cores at the 1 weight percent level.
Monte Carlo Design and Simulation of a Shipboard $^{252}$Cf-based PGNAA Analyzer for the Sensitivity Analysis of Seafloor Cores

by

Ajay Anand

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Dean of Graduate School

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TABLE OF CONTENTS

1. INTRODUCTION

2. LITERATURE REVIEW OF PGNAA APPLICATIONS
   2.1 Elemental sensitivities and detection limits
   2.2 PGNAA as a laboratory analysis technique
      2.2.1 Reactor-based facilities
      2.2.2 Multi-element PGNAA capabilities of reactor-based facilities
      2.2.3 Isotopic source based facilities
      2.2.4 Multi-element PGNAA capabilities of isotopic source based facilities
   2.3 PGNAA field applications
      2.3.1 Bulk analysis systems
      2.3.2 In-situ analysis systems
         2.3.2.1 Terrestrial mineral prospecting
         2.3.2.2 Marine mineral prospecting
      2.3.3 PGNAA field applications
   2.4 Modelling studies
      2.4.1 Modelling with deterministic methods
      2.4.2 Monte Carlo simulation

3. REVIEW OF CONCEPTS IN PGNAA
   3.1 Neutrons
   3.2 Neutron-nuclear reactions
      3.2.1 Spontaneous disintegration
      3.2.2 Nuclear collision reactions
         3.2.2.1 Neutron scattering
         3.2.2.2 Neutron absorption
   3.3 Neutron transport
      3.3.1 Nuclear cross sections
         3.3.1.1 Energy dependence of neutron absorption cross sections
         3.3.1.2 Temperature dependence of neutron absorption cross sections
      3.3.1.2 Energy dependence of neutron scattering cross sections
         3.3.1.2.1 Low energy and temperature dependence of neutron scattering cross sections
   3.4 Neutron sources
      3.4.1 Isotopic sources
      3.4.2 Neutron generators
### LIST OF FIGURES

<table>
<thead>
<tr>
<th>Figure</th>
<th>Page</th>
</tr>
</thead>
<tbody>
<tr>
<td>3.1</td>
<td>Summary of gamma ray emitting processes caused by neutrons of various energies (Wylie, 1986).</td>
</tr>
<tr>
<td>3.2</td>
<td>Prompt gamma ray production</td>
</tr>
<tr>
<td>3.3 (a)</td>
<td>Propagation of gamma quanta under conditions of &quot;good geometry&quot; (Liepunskii et al., 1965).</td>
</tr>
<tr>
<td>3.3 (b)</td>
<td>Propagation of gamma quanta under conditions of &quot;bad geometry&quot; (Liepunskii et al., 1965).</td>
</tr>
<tr>
<td>3.4</td>
<td>Schematic of a gamma ray detection and analysis system.</td>
</tr>
<tr>
<td>4.1</td>
<td>Thermal neutron flux per source neutron in H₂O and D₂O as a function of distance from a $^{252}$Cf source. The source is surrounded by 5 cm of lead. The second Y-axis shows the ratio of the thermal neutron fluxes in the two moderators.</td>
</tr>
<tr>
<td>4.2</td>
<td>Thermal to non-thermal flux ratios for H₂O and D₂O as a function of distance from a $^{252}$Cf source. The source is surrounded by 5 cm of lead.</td>
</tr>
<tr>
<td>4.3</td>
<td>The variation of epithermal flux per unit thermal flux per unit lethargy interval at various distances into a water moderator from a $^{252}$Cf source. The source is surrounded by 5 cm of lead.</td>
</tr>
<tr>
<td>4.4</td>
<td>The variation of epithermal flux per unit thermal flux per unit lethargy interval at various distances into a heavy water moderator from a $^{252}$Cf source. The source is surrounded by 5 cm of lead.</td>
</tr>
<tr>
<td>4.5</td>
<td>Idealized representations of a well moderated (Spectrum 1) and an undermoderated (Spectrum 2) neutron energy spectrum.</td>
</tr>
<tr>
<td>4.6</td>
<td>Thermal neutron flux as a function of distance into a slab material of an average rock composition at various porosities for a well moderated incident neutron energy spectrum.</td>
</tr>
<tr>
<td>4.7</td>
<td>Thermal neutron flux as a function of distance into a slab material of an average rock composition at various porosities for an undermoderated incident neutron energy spectrum.</td>
</tr>
</tbody>
</table>
4.8 The variation of epithermal flux per unit thermal flux per unit lethargy interval at various distances into a slab material of an average rock composition at 10% porosity for a well moderated incident neutron energy spectrum.

4.9 The variation of epithermal flux per unit thermal flux per unit lethargy interval at various distances into a slab material of an average rock composition at 10% porosity for an undermoderated incident neutron energy spectrum.

4.10 The variation of epithermal flux per unit thermal flux per unit lethargy interval at various distances into a slab material of an average rock composition at 50% porosity for a well moderated incident neutron energy spectrum.

4.11 The variation of epithermal flux per unit thermal flux per unit lethargy interval at various distances into a slab material of an average rock composition at 50% porosity for an undermoderated incident neutron energy spectrum.

4.12 The variation of epithermal flux per unit thermal flux per unit lethargy interval at various distances into a slab material of an average rock composition at 90% porosity for a well moderated incident neutron energy spectrum.

4.13 The variation of epithermal flux per unit thermal flux per unit lethargy interval at various distances into a slab material of an average rock composition at 90% porosity for an undermoderated incident neutron energy spectrum.

4.14 The variation of the thermal to non-thermal neutron flux ratios as a function of distance into a slab material of an average rock composition at various porosities for a well moderated and an undermoderated incident neutron energy spectrum.

4.15 Cross sectional view of the irradiation and detection system as modeled in MCNP.

5.1 Construction material gamma ray background flux at the detector with associated errors as obtained from MCNP simulation. The values are per source neutron per unit energy interval.

5.2 Gamma ray flux at the detector from the sample material at 10% porosity, as obtained from MCNP simulation. The values are per source neutron per unit energy interval.

5.3 Gamma ray flux at the detector from the sample material at 50% porosity, as obtained from MCNP simulation. The values are per source neutron per unit energy interval.
5.4 Gamma ray flux at the detector from the sample material at 90% porosity, as obtained from MCNP simulation. The values are per source neutron per unit energy interval.

5.5 7.726 MeV aluminum line gamma flux at the detector from a range of aluminum concentrations in the sample material at three different porosities, normalized to unit weight of aluminum, as obtained from MCNP simulation. The values are per source neutron per unit energy interval.

5.6 The ratio of the 7.726 MeV aluminum line gamma flux at the detector for each concentration and porosity to the average flux from all concentrations and porosities.

5.7 Thermal neutron flux as a function of distance into the sample core at various porosities. Values are per source neutron.

5.8 Thermal to non-thermal neutron flux ratios as a function of distance into the sample core at various porosities.

5.9 The variation of epithermal flux per unit thermal flux per unit lethargy interval at various distances into the core sample at 10% porosity.

5.10 The variation of epithermal flux per unit thermal flux per unit lethargy interval at various distances into the core sample at 50% porosity.

5.11 The variation of epithermal flux per unit thermal flux per unit lethargy interval at various distances into the core sample at 90% porosity.

5.12 Construction material gamma ray background detector response spectrum.

5.13 Detector response gamma ray background spectrum for the sample material at 10% porosity.

5.14 Detector response gamma ray background spectrum for the sample material at 50% porosity.

5.15 Detector response gamma ray background spectrum for the sample material at 90% porosity.

5.16 Detector response gamma ray background spectra for three different porosities around the magnesium peak energy of 3.917 MeV.

5.17 Detector response gamma ray background spectra for three different porosities around the titanium peak energy of 6.760 MeV.
5.18 Detector response gamma ray background spectra for three different porosities around the manganese peak energy of 7.058 MeV.

5.19 Detector response gamma ray background spectra for three different porosities around the iron doublet at 7.631 and 7.645 MeV.

5.20 Detector response gamma ray background spectra for three different porosities around the aluminum peak energy of 7.726 MeV.

5.21 Detector response gamma ray background spectra for three different porosities around the copper peak energy of 7.915 MeV.

5.22 Detector response gamma ray background spectra for three different porosities around the chromium peak energy of 8.884 MeV.

5.23 Detector response gamma ray background spectra for three different porosities around the nickel peak energy of 9.000 MeV.

6.1 (a) Gamma ray flux at the detector between 3 and 5 MeV from the sample matrix at 10% porosity with the associated errors, as obtained from MCNP simulation.

6.1 (b) Gamma ray flux at the detector between 5 and 10 MeV from the sample matrix at 10% porosity with the associated errors, as obtained from MCNP simulation.

6.2 Peak intrinsic efficiency curve for the detector response function of Jin et al., 1986.
<table>
<thead>
<tr>
<th>Table</th>
<th>Description</th>
<th>Page</th>
</tr>
</thead>
<tbody>
<tr>
<td>2.1</td>
<td>Sensitivities and minimum detection limits of a number of elements in coal, basalt, and bovine liver, as achieved by reactor-based PGNAA facilities</td>
<td>11</td>
</tr>
<tr>
<td>4.1</td>
<td>Average rock composition at three different porosities</td>
<td>95</td>
</tr>
<tr>
<td>5.1</td>
<td>Selected gamma ray energies and their associated gamma production data</td>
<td>124</td>
</tr>
<tr>
<td>5.2</td>
<td>Uncollided flux (for 1 w% element) from ISOSHLD-II computations normalized to unit weight, i.e, per gram of element in sample</td>
<td>124</td>
</tr>
<tr>
<td>6.1</td>
<td>Count rates per gram of element at three different porosities</td>
<td>139</td>
</tr>
<tr>
<td>6.2</td>
<td>Minimum detection limits (interference free) in weight fraction of core sample for a 10 minute irradiation time</td>
<td>140</td>
</tr>
<tr>
<td>6.3</td>
<td>Comparison of ISOSHLD-II and MCNP sensitivity and detection limits for aluminum</td>
<td>147</td>
</tr>
</tbody>
</table>
Monte Carlo Design and Simulation of a Shipboard $^{252}$Cf-based PGNAA Analyzer for the Sensitivity Analysis of Seafloor Cores

1. INTRODUCTION

The discovery and potential utilization of our seabed and terrestrial resources are projected to be of increasing importance in helping fill the growing demands of raw material in our industrialized society. Presently ocean mineral utilization is limited to continental shelf areas. However, much of the mineral needs of the future may have to be met by the use of deposits on the deep ocean floor or from as yet undiscovered terrestrial resources (Gujar et al., 1988).

Many in-situ techniques have been used successfully in the exploration of the seabed surface. These techniques provide for a rapid preliminary identification of potential minable sites. At the assessment stage a knowledge of the vertical stratification of the potential mineral sites is indispensable. Core analysis is the only convenient method for obtaining such information. The analysis of cores is usually carried out in land based laboratories and can be very costly and time consuming.

Shipboard analysis of cores can significantly reduce the cost and time spent at the assessment stage of the exploration process by obviating the need to go back to land for analysis. It can further speed the
exploration process by enabling a quick modification of the exploration plan based on the results of the shipboard analysis.

The purpose of this work was to conduct computational investigations into developing a shipboard neutron capture gamma ray analysis tool for the elemental analysis of seafloor cores. The specific areas studied were the optimization of the experimental setup and the influence of porosity (and hence salt content) on the minimum detection limits of elements of economic interest.

Prompt gamma neutron activation analysis (PGNAA) was chosen as the elemental analysis technique for a shipboard seafloor core analyzer. Several features of PGNAA led to this choice.

1. The long range of neutrons and the long range of capture gamma rays, which have energies up to 11 MeV, makes PGNAA ideally suited for the analysis of large samples such as seafloor sediment cores.

2. The availability of prolific isotopic neutron sources, such as $^{252}$Cf, makes it possible to carry out PGNAA in the field, providing flexibility in sample size and type.

3. The on-line analysis capability of PGNAA, which can improve the sample turnaround time.
4. PGNAA can simultaneously measure most of the major and minor elements in various types of samples, a situation that can be troublesome for delayed gamma neutron activation analysis (DGNAA). As most elements of economic interest are required to be detected at percent levels, PGNAA becomes an attractive technique for this application.

5. Other advantages of PGNAA include

- Non-destructive instrumental technique
- Negligible residual activity
- Practicality of using the same equipment for subsequent DGNAA, which could be used to enhance the PGNAA analysis.

The design tool used was the Monte Carlo coupled neutron photon transport code (MCNP) developed by Los Alamos National Laboratories (Briesmeister, 1989). MCNP is a bulky computer code, but has the advantage of a generalized geometry which is extremely helpful in conducting a parametric study for optimizing the experimental setup. However a Monte Carlo code written for a specific application could prove faster and leaner in the final application, i.e., after the feasibility of the technique and the optimum geometry have been established.

A literature review of previous work on the application of nuclear techniques to mineral exploration is presented in Chapter 2.
Chapter 3 deals with basic theoretical concepts applicable to the study. These include neutron and gamma ray generation and transport, and their detection in semiconductor detectors. The Monte Carlo technique is also briefly discussed.

Chapter 4 describes the design parameter optimization process for the PGNAA analyzer. Computational investigations for the selection of moderator and for the influence of porosity on the neutron flux profiles in the sample are presented. The design of the analyzer based on the data generated by the computations is then described.

Chapter 5 describes the simulation of the PGNAA analyzer for the generation of the capture gamma ray spectra for the evaluation of elemental sensitivities and detection limits.

The results in the form of sensitivity and minimum detection limits are presented in Chapter 6. This is followed by a discussion of the results.

Chapter 7 documents the health physics aspects of the shipboard analytical facility.

Conclusions based on the present analysis and recommendations for future work are given in Chapter 8.
2. LITERATURE REVIEW OF PGNAA APPLICATIONS

Neutron capture gamma ray spectroscopy as a tool in chemical analysis has been in use for many years. The boost to the technique since the mid-1960's came from improvements in technology, viz. the development of high resolution Ge(Li) semiconductor gamma ray detectors, sealed-tube neutron generators, $^{252}$Cf sources and intelligent pulse-height analyzers. Three distinct areas of application of the prompt method have been (1) laboratory based elemental analysis (mainly using reactor neutrons), (2) field measurements in borehole logging for oil and mineral exploration (both at land and at sea), and (3) industrial process control, the latter two using neutron generators or isotopic neutron sources.

Extensive bibliographies of publications on the PGNAA method have been compiled by Gladney, 1979 and Glascock, 1984. Various review articles on PGNAA applications can be found in the literature (Greenwood, 1979; Clayton, 1982; Glascock, 1982; Chrien, 1984). An overview of prompt nuclear analysis is presented by Peisach, 1981. The potential of thermal neutron capture prompt gamma ray analysis is investigated by Herzenberg, 1983. The status and prospects of analytical capture gamma ray spectroscopy are discussed by Lindstrom and Anderson, 1985. Neutron capture gamma ray spectroscopy as a quantitative analytical method is discussed by Anderson et al., 1982. Noakes and Harding, 1982, Ranasinghe et al., 1988 and Noakes et al., 1989 have reviewed the nuclear techniques for seabed mineral exploration.
2.1 Elemental sensitivities and detection limits

The sensitivity, $S$, of an element is defined in terms of its signal strength, i.e., count rate under the photopeak per unit mass of the element (cps/mg). The capture reaction rate is given by $R = N\sigma_c\hat{\phi}$, where $N$ is the number of target nuclei, $\hat{\phi}$ is the neutron flux and $\sigma_c$ is the capture cross section. Hence since $N = N_A m/A$, other than the mass of the element in the sample and the neutron flux, the rate $R$ depends only on the ratio $\sigma/A$ (cross section/atomic mass). However, a better indication of the sensitivity of an element is the product, $I\sigma/A$, where $I$ is the intensity of the most favorable $\gamma$-ray, i.e., the number of favorable $\gamma$-rays generated per 100 neutrons captured.

It is important to realize that the capture gamma ray spectrum from each nuclide $^{AZ}$ consists of a series of lines with energies ranging up to the value of the neutron binding energy of the $^{A+1}_Z$ nucleus. As these neutron binding energies are comparable, typically from 3 to 11 MeV, the $\gamma$-ray spectra generated are quite complex and inter-element interferences can become a problem.

In the experimental determination of sensitivities, the count rate is obtained from samples of pure elements. The values reported are after background and interference (from contaminants) subtraction. Hence the sensitivity is independent of the sample (except for differences in self attenuation of the $\gamma$-rays, an effect which can usually be neglected for small samples) and depends only on the experimental setup, i.e., the
neutron flux, geometry, and the detection system.

The detection limit, however, is dependent on the inter-element interferences, on the background levels in the photopeak from the sample matrix elements, and on the counting times available. The minimum detection limit $C_{\text{MDL}}$ (mg/g) is defined as the smallest concentration that a particular measurement process can reliably detect (Currie, 1977).

It is worth noting here that it is not necessary to observe lines of an element in the $\gamma$-ray spectrum in order to calculate a $C_{\text{MDL}}$ of the element in that material. It is necessary only to obtain sensitivities

\[ C_D = 2 \times C_C = 2 \times Z_{95} \times \sigma_0 = 2 \times 1.645 \times \sigma_0, \]

where $\sigma_0$ is the standard deviation of the probability distribution of making a TYPE I error.

If it is assumed that the standard deviation at the minimum detection limit is determined solely by the Poisson distributed counting statistics in the background under the photopeak,

\[ \sigma_0 = (N_b)^{1/2}/(S \times t) = (R_b/t)^{1/2}/S \]

where
- $N_b$ = Background counts
- $R_b$ = is the background counting rate (counts/s)
- $t$ = counting time (seconds)
- $S$ = Sensitivity (counts/s-mg)
for various lines of the element from a standard of the element and the background rate in the appropriate regions of the sample spectrum.

The actual sensitivities and detection limits vary between experimental setups and depend upon:

1. Neutron flux at sample (source strength).

2. Sample-to-detector distance (solid angle factor).

3. Shielding materials and arrangement (background and interference).

4. Detector arrangement (Compton suppression).

Factors not depending on the design of the experimental facility also influence the detection limits, sometimes severely. These factors are:

1. Cross section of the element of interest.

2. Sample type.

3. Irradiation times available, $C_{MDL} \propto 1/t^{1/2}$.

Most of the data on element sensitivity and detection limits come from reactor-based facilities. In such facilities the non-destructive
determination of most major elements (elements present at concentrations greater than one percent) can be simultaneously performed in most matrices. A notable exception is oxygen, due to its low capture cross section. Also, Be, C, F and Li can usually be measured in concentrations greater than 5 percent. In addition to the major elements, a number of important minor elements (elements present at concentrations between 0.1 to 1.0 percent) and trace elements may also be measured.

A comparison of the sensitivities considering only primary $\gamma$-rays (Greenwood, 1979) shows that most of the elements lie within a narrow sensitivity range. This points to the observed fact that PGNAA is generally sensitive only to the major constituents of the sample.

2.2 PGNAA as a laboratory analysis technique

Laboratory applications of PGNAA, in various fields have been summarized by Glascock, 1982 and Chrien, 1984.

2.2.1 Reactor-based facilities

Many PGNAA facilities have been set up around the world. Two types of geometrical arrangements are utilized. They are the internal sample geometry and the external sample geometry. In the internal sample geometry the sample is placed near the neutron source beam and the resulting capture gamma ray flux is collimated toward a detector placed
at a workable distance from the neutron source beam. In the external sample geometry the neutron beam is collimated towards the sample with the detector placed close to the sample.

In the USA much work has been carried out at the internal sample facility at Los Alamos National Laboratory and at the external sample facilities at University of Maryland-NBS reactor and at the University of Missouri. All three installations have intense neutron sources and large Compton suppressed Ge gamma ray detectors. The sensitivities achieved by the three facilities are comparable. Characteristics of these and other reactor-based analytical PGNAA facilities have been tabulated by Anderson et al., 1982.

2.2.2 Multi-element PGNAA capabilities of reactor-based facilities

The multi-element capabilities of the technique for reactor-based facilities for a large variety of samples have been described by Failey et al., 1979; Anderson et al., 1982 and Hanna et al., 1981.

The reactor-based PGNAA facilities are capable of simultaneously determining the concentrations of up to 20 major, minor and trace elements, from the set of 35 elements shown in Table 2.1, in a variety of matrices. As the irradiation times used for most of the reactor-based analysis are relatively long (about 24 hours), some activation of the sample also takes place. Using delayed gamma neutron activation analysis (DGNAA) subsequent to the PGNAA in many cases can
Table 2.1  Sensitivities and minimum detection limits of a number of elements in coal, basalt, and bovine liver, as achieved by reactor-based PGNAA facilities.

<table>
<thead>
<tr>
<th>Element</th>
<th>Energy (MeV)</th>
<th>Sensitivity, $S_i$ (cps/mg)</th>
<th>Detection limits, $C_{MDL}$ (mg/g)</th>
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<tbody>
<tr>
<td></td>
<td></td>
<td></td>
<td>Coal</td>
</tr>
<tr>
<td>H</td>
<td>2.223</td>
<td>0.86</td>
<td>0.01</td>
</tr>
<tr>
<td>B</td>
<td>0.478</td>
<td>530</td>
<td>0.00005</td>
</tr>
<tr>
<td>C</td>
<td>1.262</td>
<td>0.00039</td>
<td>27</td>
</tr>
<tr>
<td>N</td>
<td>1.885</td>
<td>0.003</td>
<td>3.4</td>
</tr>
<tr>
<td>F</td>
<td>1.634</td>
<td>0.0011</td>
<td>6.0</td>
</tr>
<tr>
<td>Na</td>
<td>0.472</td>
<td>0.15</td>
<td>0.3</td>
</tr>
<tr>
<td>Mg</td>
<td>0.585</td>
<td>0.0085</td>
<td>1.2</td>
</tr>
<tr>
<td>Al</td>
<td>7.726</td>
<td>0.0013</td>
<td>0.86</td>
</tr>
<tr>
<td>Si</td>
<td>3.539</td>
<td>0.0066</td>
<td>0.6</td>
</tr>
<tr>
<td>P</td>
<td>0.637</td>
<td>0.0062</td>
<td>1.3</td>
</tr>
<tr>
<td>S</td>
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<td>Cl</td>
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<td>K</td>
<td>0.770</td>
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<td>Ca</td>
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<td>Ti</td>
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<tr>
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<td>1.434</td>
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</tr>
<tr>
<td>Cr</td>
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<tr>
<td>Mn</td>
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</tr>
<tr>
<td>Fe</td>
<td>0.352</td>
<td>0.046</td>
<td>0.3</td>
</tr>
<tr>
<td>Co</td>
<td>0.277</td>
<td>1.2</td>
<td>0.020</td>
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<tr>
<td>Ni</td>
<td>0.465</td>
<td>0.12</td>
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</tr>
<tr>
<td>Cu</td>
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<td>Se</td>
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<td>0.09</td>
</tr>
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<td>0.00006</td>
</tr>
<tr>
<td>In</td>
<td>0.162</td>
<td>2.9</td>
<td>0.014</td>
</tr>
<tr>
<td>Ba</td>
<td>0.627</td>
<td>0.55</td>
<td>0.02</td>
</tr>
<tr>
<td>Nd</td>
<td>0.697</td>
<td>1.3</td>
<td>0.007</td>
</tr>
<tr>
<td>Sm</td>
<td>0.334</td>
<td>640</td>
<td>0.00003</td>
</tr>
<tr>
<td>Gd</td>
<td>0.182</td>
<td>680</td>
<td>0.00005</td>
</tr>
<tr>
<td>Pb</td>
<td>7.368</td>
<td>0.0004</td>
<td>5.0</td>
</tr>
</tbody>
</table>

a. Flux at the sample about $2 \times 10^8$ n/cm$^2$-s, sample to detector distance 50 cm, compton suppression used. For further details see Anderson et al. 1982.

b. The gamma ray line is from the activation daughter product of the element.
enable a significant number of additional elements to be analyzed.

The multi-element analysis of biological materials by PGNAA has been shown by Failey et al., 1979, to be reliable for the elements H, C, N, Na, Mg, S, Cl, K, Ca, Mn and Cd. Samples analyzed included human liver, bovine liver and other tissues, and orchid leaves.

The value and potential of PGNAA in the geological area has been well demonstrated. Volcanic ash and lava, particulate on filters, dust, coal, oil shale, borehole samples and many other kinds of samples have been successfully analyzed (Anderson et al., 1982). Elemental analysis of masonry materials has been carried out by Anderson et al., 1983.

Graham et al., 1982; Anderson et al., 1985 and Olivier et al., 1988, have used PGNAA to characterize NBS, USGS, and IAEA geochemical standards.

2.2.3 Isotopic source based facilities

Isotopic source based PGNAA facilities have not been extensively utilized in the laboratory. Design considerations of such facilities have been discussed by Senftle et al., 1971; Greenwood, 1975; Lukander and Uusitalo, 1979 and Duffey and Wiggins, 1979 and 1987. The neutron source used in most of the applications is $^{252}$Cf. Trubert et al., 1989, have reported an Am-Be neutron source based facility.
It is seen that the design of the experimental setup depends quite heavily on the application, in particular, on the elements to be analyzed and the detection limits sought. As with reactor facilities, an isotopic source based PGNAA facility can have an internal sample geometry or an external sample geometry. The choice of the moderator and the construction materials strongly influences the geometry of the facility and the energy range of the capture gamma ray spectrum utilized. The placement of the detector is determined by the detector's tolerance to neutron damage (Section 3.9.1). The source strength used is also a compromise between the gain in sensitivity and the amount of shielding required and other safety and handling requirements. A parametric study (experimental and/or computer simulation) is usually carried out to find the geometry which gives the maximum sensitivity within the design constraints of the particular application.

Considerable effort may have to be spent in optimizing the geometry and materials in the construction of a PGNAA facility. Hence the construction of a facility may vary considerably depending on the application. A more detailed discussion of the optimization process and the factors that influence the choice of the geometry, as applied to the design of the PGNAA system of this work, are discussed further in Chapter 4. For the description of various isotopic source based PGNAA facilities the reader is referred to the references cited above.
2.2.4 Multi-element PGNAA capabilities of isotopic source based facilities

Wiggins et al., 1975 have shown the feasibility of using $^{252}$Cf isotopic source based PGNAA for Mn, Fe, Cu, Ni, V and Au in marine manganese nodules. The signature photon energies (above 3 MeV) obtained in their work compares well with the energies reported from research at reactor-based facilities. However, in some cases significant differences in the relative intensities of various lines in the capture gamma ray spectrum of some elements can be seen between the values obtained by Wiggins and the data reported from reactor facilities. This is thought to be due to the differences in the thermal to epithermal neutron flux ratio at the sample between the two kinds of facilities. The neutron energy dependence of the capture gamma ray spectra is discussed further in Section 3.6.

Hassan et al., 1983, have shown the feasibility of using a $^{252}$Cf isotopic source facility for analyzing boron (in Pyrex borosilicate glass mixed with sea sand), chlorine (in sodium chloride solution), and for the on-line analysis of phosphorous in industrial waste waters (in lime solution and $\text{Al}_2\text{O}_3$ phosphorous recovery filters).

Duffey and Wiggins, 1987, have shown the feasibility of using a $^{252}$Cf source facility for the elemental analysis of coal. Duffey and Wiggins observe that the choice of the gamma ray for analysis may depend on the type of coal, as different grades of coals may show different
concentration levels of major elements and contaminants. This shows that the sample matrix can have a significant influence on the PGNAA, at least as far as inter-element interferences are concerned. The influence of porosity on the prompt gamma ray analysis is further discussed and investigated in Chapter 4.

2.3 PGNAA field applications

The neutron capture gamma ray method is a commonly used procedure in the practice of geophysics (Földiák, 1986). Presently in-situ capture gamma ray analysis techniques are being increasingly utilized in coal, oil and mineral exploration. Systems employing PGNAA and other nuclear techniques (both in-situ and shipboard element analysis systems) have been developed for marine mineral exploration. These systems have been extensively tested in the laboratory environment and in the field. Based on the data generated by these trials, nuclear techniques, including PGNAA, have been shown to be feasible for the exploration of the seabed. However, they have not yet been commercialized.

2.3.1 Bulk analysis systems

Bulk analysis systems are usually utilized for continuous on-line multi-element analysis in the process industry (especially coal), geology (ocean bed sediment elemental analysis), and medicine (whole body in vivo and tissue in vitro analysis).
The geometrical setup of the analysis system may be generalized as a "conveyer belt" configuration. Here the sample is moved continuously or in batches on a conveyor belt or by some other suitable arrangement over the source, i.e. the irradiation region. Moderator is usually provided between the source and the sample for thermalization. A moderator also surrounds the source for reducing the radiation exposure to the workers. In cases of hydrogenous samples (coal, oil, etc.), moderation between the source and sample may not be necessary.

Industrial on-line PGNAA analyzers called "nucoalyzers" for coal and other materials have been developed by Science Applications International Corporation (SAIC) (Gozani, 1985). In the medical field several prompt gamma whole body neutron irradiators (Pu-Be multiple sources) have been built by Australia Nuclear Science Technology Organization (ANSTO) (NPG News, July 1990).

2.3.2 In-situ analysis systems

The main configuration utilized for in-situ analysis of land based oil, coal and deep mineral deposits is the borehole configuration. Here the sonde is lowered down a drilled borehole on coaxial cables. The cables serve as mechanical supports, power lines and as data carriers. A continuous log is obtained by moving the sonde up the borehole at an appropriate constant speed.

The sonde is a compact system having a neutron source (a neutron
generator or an isotopic source), a high resolution gamma ray detector (with its cooling system), and miniature electronics. The construction varies depending on the nuclear technique or techniques used, type and strength of neutron source, type of detector, the depth at which the sonde is designed to operate, and for deep sea operations, additionally on the static or towed mode of operation.

2.3.2.1 Terrestrial mineral prospecting

On land the \textit{in-situ} borehole logging technique has the advantage of analyzing a larger ore body. This makes the results of such \textit{in-situ} analysis compared to the analysis of smaller sample volumes in the laboratory, more representative of the chemical and mineral composition of the ore body. Numerous applications of PGNAA using $^{252}$Cf for coal, oil and mineral exploration in the field and also, by using simulated coal piles and boreholes in the laboratory, can be found in the literature (Fanger and Pepelink, 1975; Nargolwalla \textit{et al.}, 1977; Gozani \textit{et al.}, 1977; Gozani \textit{et al.}, 1978; Frank \textit{et al.}, 1978; Sohrabpour \textit{et al.}, 1979; Charbucinski \textit{et al.}, 1986).

2.3.2.2 Marine mineral prospecting

In the case of marine mineral prospecting the need for rapid and economical analysis of the seafloor makes \textit{in-situ} techniques indispensable. Nuclear techniques are well suited for \textit{in-situ} analysis of the seafloor which is a relatively rugged environment. The nuclear
techniques also provide for a rapid elemental analysis with a high degree of element specificity. Hence much of the work in the area of seabed mineral exploration is in the form of in situ analysis of sea bottom sediments (Noakes and Harding, 1982; Noakes et al., 1989; Ranasinghe et al., 1988). For the sediment analysis of marine minerals the sonde usually rests on the seafloor or penetrates a short distance into the seabed. Systems in which the sonde is towed by a surface vessel have also been developed (Miller et al., 1977).

The nuclear techniques considered for the in situ analysis of sea bottom sediments were natural radioactivity measurement, neutron activation analysis (prompt and delayed), and X-ray fluorescence (XRF). Neutron activation analysis was found to be best suited for the in-situ quantitative elemental analysis of ocean bottom sediments due to its ability to analyze many elements in a variety of matrices, feasibility of operation at greater depths, and its ability to analyze sediments to a greater vertical depth than XRF.

Earlier systems mostly employed the long established and proven DGNAA technique. Senftle et al., 1969, first suggested the possibility of using PGNAA for marine exploration. His conclusion was based on laboratory experiments; no in situ field tests were conducted. Moxham et al., 1976, first used PGNAA for the in-situ analysis of river bottom sediments. At the same time Senftle et al., 1976, reported a feasibility study for in-situ capture gamma ray analysis (PGNAA) for seabed exploration. More recently, detailed laboratory and computer
simulation studies of PGNAA in the marine environment have been conducted (Ranasinghe et al., 1988). Ocean bottom cores had not been previously analyzed by PGNAA using isotopic neutron sources.

Grab samples taken from the seafloor have been analyzed onboard ship (Noakes et al., 1975; Noakes and Harding, 1982). Elemental analysis of ocean and river bottom cores has also been carried out earlier using fast neutrons (Hayes and Peterson, 1975) and conventional NAA (Hayes and Peterson, 1975; Noakes et al., 1975). Only a few elements were analyzed which compared within 30% with other standard chemical analysis.

2.4 Modelling studies

Computer modelling studies have proven very useful since a suitable computational model for the analysis greatly reduces the number of calibration standards needed. It can also be used as a very cost and time effective method for optimizing the analyzer design. In such studies a model is used to predict the sensitivity of the analyzer as a function of various design parameters, without the need to make expensive and time consuming changes to an actual experimental setup. Such a process also provides additional insight into the physical processes involved and their interrelationships.

2.4.1 Modelling with deterministic methods

Development of theoretical models for simulating the multi-element
prompt gamma ray analysis of bulk material in the field is very
difficult. This is due to the complexity of the analyzer-sample
geometries and the energy dependence of neutron interactions. Most of
the models involve simplifications to the geometry. Multigroup
diffusion theory has been utilized to calculate the neutron flux
distribution in the region of interest from a point isotropic source.
The prompt gamma ray source term in the sample is then computed from the
neutron flux by using neutron capture cross sections and their
associated gamma ray production cross sections. The uncollided capture
gamma ray flux is then obtained by integrating over the sample region.
Such models have been derived for the borehole logging configuration of
neutron capture prompt gamma ray analyzers (Tittle, 1961; Tittle and
Allen, 1966; Doster et al., 1979 and O'Dell, 1985).

Doster et al., 1979, modelled a sonde-in-borehole configuration to
predict the photopeak detector response. The formation was modeled with
an ore containing three regions, each varying in its iron content. The
nuclear reactor code PDQ-5 was used to solve the four group neutron
diffusion equation for the flux distribution. The photopeak detector
response was obtained by a numerical integration of the prompt gamma ray
production kernel over the sample volume. Good agreement was obtained
between experimental and simulation results.

The discrete ordinate, one dimensional, neutron transport code ANISN was
used to determine the neutron flux distribution in a multi-layered coal
medium in a spherical geometry with a neutron source in the center
(Reynolds et al., 1977; Elias and Gozani, 1980). The effect of bulk density, sample uniformity, system size and hydrogen content on the prompt gamma ray flux in the detector was assessed. Results showed that PGNAA of coal can be sensitive to various design parameters.

### 2.4.2 Monte Carlo simulation

Monte Carlo simulation is considered very attractive as it can deal with complex geometries of actual interest unlike the deterministic models which are quite elementary and use approximations to the actual geometry. Monte Carlo models can also be used with fundamental, pointwise nuclear data instead of energy group data.

Sohrabpour and Bull, 1979, proposed a Monte Carlo model utilizing PGNAA for the calibration of in situ probes in the borehole geometry. Their model predicts the expected gamma ray emission rate of various elements in the host rock as a function of various parameters, viz., elemental concentration, geometry, detector type and efficiency, moisture content, neutron source strength, etc. Multigroup data were used and only the uncollided portion of the flux was calculated. Detector efficiency factors were used to generate the photopeak and the single and double escape peaks.

The computer simulation results were tested against data obtained from an actual experiment. The calculated count rates from the simulation were 30 to 40 percent higher compared to experimental values, but the
peak ratios showed good agreement for the two sets of data. The higher count rates from the simulation were attributed to some parametric uncertainties in the simulation.

Clark et al., 1982, developed a Monte Carlo model for in situ PGNAA probes in the borehole geometry. Their model predicted the response from a whole range of elemental concentrations in the host matrix simultaneously. Unlike the model of Sohrabpour and Bull, this model simulated the neutron and photon transport in the shielding components of the probe. ENDF/B-V (ENDF stands for the Evaluated Nuclear Data File, /B indicates files with completed evaluated sets of data and -V is version five) pointwise data were used. The simulation results agreed favorably with experiment. This model could also be used for optimizing the probe design parameters.

Sanders, 1983, used the Monte Carlo method to calculate the neutron flux distributions, and to some extent the neutron induced photon flux distributions, in the borehole geometry. Two approaches were used for the coupled neutron photon tracking. In the first approach the gamma rays produced were stored and then tracked according to known gamma ray interaction cross sections. In the second approach the neutron tracking was first completed to provide average neutron fluxes in chosen volume elements of the sample. The average neutron fluxes were then used to compute volume-distributed gamma ray sources according to known photon production data. The second approach, although not as accurate as the first, was appreciably less expensive, especially when the calculations
in the first approach were done for narrow energy bins.

Sanders further noted that converting the gamma ray flux in the detector to detector output would provide an even more realistic simulation, although at a still greater cost.

Schmidt and Rose, 1984, used a modified version of the SAM-CE Monte Carlo code, with ENDF/B-V cross sections (including photon production data) to calculate a complete gamma ray spectrum from a block of coal. The gamma ray fluxes were arbitrarily grouped into 10 keV bands for gamma ray source lines and 500 keV for the background. The weaker source lines were added to the background. A comparison with experimental results showed good agreement for the discrete lines. It was noted that the availability of all the \((n,\gamma)\) and \((n,n'\gamma)\) lines would significantly improve the analysis. Utilization of a detector response function was also suggested.

Rainbow, 1985, used the general purpose Monte Carlo code MORSE and nuclear data derived from ENDF/B-IV to simulate an iron ore analyzer in the conveyor belt configuration. User patches were used to modify the code to suit the special features of the problem, viz., source and detector modelling. Thermal neutron capture gamma rays from \(^{56}\text{Fe}\) were tallied. The results compared favorably with measurements from an actual system. A drawback of the simulation was that despite of the use of powerful variance reduction techniques, the efficiency of the calculations was low.
Jin et al., 1987, used specific purpose Monte Carlo models to simulate the complete pulse height spectral response of neutron capture prompt gamma ray analyzers for both the bulk media and the borehole configuration. Pointwise ENDF/B-V cross section data were used in the simulation. These models have been validated against experiment.

An important feature of their simulation was the incorporation of powerful variance reduction techniques not yet available or difficult to implement with general purpose Monte Carlo codes such as MCNP. These techniques included statistical estimation, use of a semi-empirical detector response function, and correlated sampling. Statistical estimation consists of forcing the desired detection result to occur at each interaction. The reaction type is sampled first and if it is a collision event, all gamma rays of interest from all elements in the sample are forced to be emitted at each such event. Further the scattered or emitted radiation is forced to occur in the direction of the detector. It is then transmitted without collision through the medium, and forced to be detected. Jin et al., reported that improved algorithms for forcing the gamma rays toward the detector resulted in a very efficient implementation of the statistical estimation variance reduction method.

The configurations tackled so far have been mainly borehole and bulk media. Almasoumi, 1989, has modeled a PGNAA analyzer for the elemental analysis of sea bottom sediment cores. Some work in validating the model remains to be done. Only the unscattered portion of the gamma ray
flux is modeled. Correlated sampling is used.

The need and usefulness of Monte Carlo computer simulations can further be recognized from the following brief discussion of the various aspects of formation analysis to which such simulations have been applied.

1. Basic studies on the effect of formation variables on the nuclear properties of the formation. These include the computation of neutron profiles and spectra as a function of the formation variables and the determination of characteristic lengths.

2. Evaluation of a PGNAA analyzer design by determining the sensitivity of various elements of interest with respect to the design constraints and expected formation perturbation.

3. Tool design calibration. Although some benchmark experiments are essential for validation and normalization of the computer simulation results, Monte Carlo computer simulations are relatively easy to use for an extensive parametric study which would be very difficult and expensive, if not impossible, by actual experiment.

Coupling a detector response function to the gamma ray transport simulation to compute the complete pulse height spectral response has been done by Jin et al., 1987. This additional step allows the
calculation of detection limits in addition to determining the sensitivities. Also interference from various contaminants can be studied more realistically.

In this work, the detector response function proposed by Jin et al., 1986, and extended to lower energies by Lee et al., 1987, is utilized. The manner of its utilization is given in Section 5.3. This response function is valid for the energy range between 60 keV and 6.2 MeV. However, considering the exploratory nature of this work and the essentially linear energy response of a HPGe detector, the same was used up to 10 MeV.
3. REVIEW OF CONCEPTS IN PGNAA

In this chapter general concepts relevant to this work are discussed. These include the generation of neutrons and gamma rays, the transport and interaction of neutrons and gamma rays in materials, the detection of gamma rays, and the interpretation of the detected gamma ray spectra for elemental analysis.

3.1 Neutrons

Neutrons are classified according to their energy. The type of reaction a neutron undergoes depends very strongly on its energy. Neutrons with energies above 0.1 MeV are called fast neutrons. Neutrons having the same kinetic energy as the molecules in their environment are called thermal neutrons. The energy of the thermal neutrons is characterized by the Maxwell-Boltzmann distribution

\[ f(E) = \frac{2\pi E^{1/2}}{(\pi kT)^{3/2}} e^{-E/kT} \]

where

- \( f(E) \) is the fraction of the neutrons of energy \( E \) per unit energy interval.
- \( k \) is the Boltzmann constant, \( 1.38 \times 10^{-23} \, \text{J/K} \) or \( 8.6 \times 10^{-5} \, \text{eV/K} \).
- \( T \) is the absolute temperature, K.

The most probable energy of the Maxwell-Boltzmann distribution, \( E_{mp} \), is equal to \( kT \). At room temperature (\( T=293 \, \text{K} \)), \( kT = 0.025 \, \text{eV} \). The
velocity of the neutron corresponding to this energy is 2200 m/s.

In the region of energy between thermal and fast lies the intermediate neutron energy range. The neutrons in this energy range are called epithermal, resonance or slow neutrons.

A commonly used experimental procedure to measure thermal and non-thermal neutrons is the cadmium-cutoff or the Cd/Au ratio. This ratio is considered an important parameter in PGNAA as the capture gamma ray spectra obtained can be significantly influenced by the hardness of the incident neutron energy spectrum (Section 3.6.2.1).

### 3.2 Neutron-nuclear reactions

#### 3.2.1 Spontaneous disintegration

Certain nuclei are unstable in the sense that they may spontaneously undergo a transformation into a different nuclide or nuclides, accompanied by the emission of energetic particles. If a spontaneous transformation results in the formation of two or more daughter nuclides, it is called spontaneous fission. Neutrons are among the energetic particles released.

\[
\begin{align*}
\text{A}_X & \rightarrow \text{A}_1X + \text{A}_2X + (2 \text{ to } 3) \text{ } n \\
& \quad + (\text{occasionally}) \text{ } ^3\text{H}
\end{align*}
\]
Californium-252 is an example of a nuclide which undergoes such spontaneous disintegration. The neutrons released can be used as a source of neutrons for inducing further neutron-nuclear reactions.

In radioactive decay the decaying nucleus does not split into two or more daughter nuclides, but is transformed into another close neighboring nuclide. Alpha, beta and gamma decay are the three most common types of radioactive decay reactions.

In alpha decay the parent nuclide emits a helium nucleus (alpha particle). The decay process is

\[
\frac{A}{Z}X_N^{\text{z}} \rightarrow \frac{A-4}{Z-2}X'_N^{\text{z}} + \frac{4}{2}\text{He}_2
\]

where \(X\) and \(X'\) represent the chemical symbols of the initial and final nuclei. The number of neutrons and protons are separately conserved in the alpha decay process.

A nucleus undergoes a beta decay to correct for a neutron or proton excess.

\[
\frac{A}{Z}X_N^{\text{z}} \rightarrow \frac{A-1}{Z+1}X'_N^{\text{z}+1}
\]

\(\beta^-\) decay

\[
\frac{A}{Z}X_N^{\text{z}} \rightarrow \frac{A+1}{Z-1}X'_N^{\text{z}-1}
\]

\(\beta^+\) decay

\[
\frac{A}{Z}X_N^{\text{z}} \rightarrow \frac{A}{Z-1}X'_N^{\text{z}+1}
\]

\(\varepsilon\) electron capture
In this process a neutron in the nucleus is converted to a proton or a proton into a neutron. In all three beta decay pathways an electrically neutral particle called the neutrino is also emitted. The emission of this particle is essential to conserve momentum. In beta decay $Z$ and $N$ each change by one unit, but the total mass number $A \ (A = Z + N)$ remains constant.

In gamma decay a nucleus decays from a higher excited state ($m_1$) to a lower (or possibly the ground state, $m_2$). This decay results in the loss of energy equal to the difference between the two excited states. This energy is carried away by a photon of gamma radiation. Gamma decay usually follows $\alpha$ and $\beta$ decay as these decays often lead to excited states in the daughter nucleus.

$$A^m_{ZN} \rightarrow A^{m_2}_{ZN} + \gamma \text{ ray}$$

### 3.2.2 Nuclear collision reactions

Neutrons at the time of their generation are mostly fast neutrons. The fast neutrons lose energy by collisions with the surrounding atoms. A collision can result in the scattering of the neutron or in its capture.

#### 3.2.2.1 Neutron scattering

Neutron scattering interaction may be visualized as a billiard ball type of collision in which kinetic energy and momentum are both conserved. Neutron scattering may be elastic or inelastic. For elastic scattering
the energy $E$ of the scattered neutron can be shown to be

$$E = E_0 \left( \frac{M - m}{M + m} \right)^2$$

where

$E_0$ = energy of incident neutron  
$m$ = mass of incident neutron  
$M$ = mass of scattering nucleus.

Elastic scattering is the most likely interaction between fast neutrons and low atomic number atoms.

In the case of inelastic scattering some of the kinetic energy that is transferred to the target nucleus excites the nucleus, and the excitation energy is emitted as a photon of gamma radiation. This interaction is best described by the compound nucleus model, in which the neutron is captured and then re-emitted by the target nucleus together with the gamma ray.

$$^1_0n + ^A_zX \rightarrow \text{Compound Nucleus} \rightarrow (^{A+1}_zX)^*$$

$^{A+1}_zX)^*$ \text{ elastic scattering} $\rightarrow ^1_0n + ^A_zX$

$^{A+1}_zX)^*$ \text{ inelastic scattering} $\rightarrow ^1_0n + (^{A}_zX)^*$

\[ \downarrow \]

$^A_zX + \gamma$
Inelastic scattering is a threshold phenomena. The neutron energy threshold varies from infinity for hydrogen (inelastic scattering cannot occur) to about 6 MeV for oxygen to less than 1 MeV for uranium.

3.2.2.2 Neutron absorption

In this type of interaction, the incident neutron is absorbed by the target nucleus to form a new nuclide with a unit increase in mass number. This new nuclide is formed in an excited state, best described as a compound nucleus. This compound nucleus can decay in many ways, each with a finite probability:

1. Charged particle emission, e.g., (n,α) or (n,p) reactions.

2. Photon emission, i.e. (n,γ) or the radiative capture reaction, in which the compound nucleus de-excites almost instantaneously by emitting a photon. This is usually followed by a beta particle emission.

3. Neutron producing reaction, i.e., (n,xn), where x ≥ 2.

4. Fission reaction, in which the compound nucleus splits into two large fragments (and one light fragment in the case of the rare ternary fission reaction) along with the instantaneous emission of generally one to three
free neutrons.

Of all the above neutron absorption reactions, the one of most significance to this study is the radiative capture reaction.

\[
\left( \frac{A+1}{Z}X \right)^* \xrightarrow{\text{Radiative Capture}} A+1 \xrightarrow{Z} X + \gamma
\]

The generation of capture gamma radiation is further discussed in Section 3.6.

3.3 Neutron transport

Neutron transport describes the motion of the neutrons as they stream about, and interact with, the nuclei of a medium. Any number of possible neutron-nuclear reactions can occur during this process. The scattering process results in the loss of kinetic energy of the neutron leading to its slowing down and thermalization. In an infinite medium the neutron will ultimately be absorbed in a capture reaction.

For a neutron source emitting neutrons at a constant rate, a time invariant neutron spatial and energy distribution will be established in the material surrounding the source. A knowledge of this distribution is essential for predicting the rate of neutron-nuclear reactions as a function of position within the surrounding material. The following is a discussion of some of the fundamental concepts in understanding the establishment of neutron profiles in a medium and determining the rate
of the resulting neutron-nuclear reaction.

3.3.1 Nuclear cross sections

For a monoenergetic beam of neutrons incident normally on a thin target the rate of neutron-nuclear reactions, \( R (\#/\text{cm}^2\text{-s}) \), is proportional to the incident neutron beam intensity, \( I (\#/\text{cm}^2\text{-s}) \), and the number of target atoms per unit area \( N_A (\#/\text{cm}^2) \), i.e.,

\[
R \propto I N_A
\]

or

\[
R = \sigma I N_A
\]

where \( \sigma \), the constant of proportionality, has the units of cross sectional area, i.e., \( \text{cm}^2 \), and is called the microscopic cross section. The microscopic cross section is not the actual (geometrical) cross sectional area of the nucleus, but can be thought of as an "effective" cross sectional area presented by a nucleus of the target material to an incident neutron beam. The cross section characterizes the probability for a neutron-nuclear reaction.

Since the nuclear radius is roughly \( 10^{-12} \text{ cm} \), the geometrical cross sectional area of the nucleus is roughly \( 10^{-24} \text{ cm}^2 \). Hence microscopic cross sectional areas are usually measured in units of this size, called barns \( (1 \text{ b} = 10^{-24} \text{ cm}^2) \). However, due to the quantum mechanical nature of the neutron and the atomic nucleus, the effective values of cross sections usually differ appreciably from their geometrical values and are also strongly dependent on the incident neutron energies.
Another useful quantity is the macroscopic cross section. To understand this, consider a thick target where nuclei deep within the target are shielded by the ones in front. At any position $x$ within the target, consider a thin section of thickness $dx$. Then for beam intensity $I(x)$ the total reaction rate within $dx$ will be equal to the reduction in the beam intensity; hence

$$dR(x) = -\sigma N dx \ I(x)$$

$$dR(x) = -dI(x) = -[I(x+dx) - I(x)]$$

$$dI(x) = -\sigma I(x) N dx$$

$$\frac{dI(x)}{dx} = -\sigma NI(x)$$

or,

$$I(x) = I_0 \exp(-N\sigma x)$$

where

$N =$ the number density of target nuclei, and $Ndx$ is the total number of nuclei per cm$^2$.

$\sigma =$ the total microscopic cross section, i.e., the probability of any type of reaction that effectively removes the neutron from the incident beam.

$I_0 =$ the incident neutron beam intensity at $x = 0$.

The quantity, $N\sigma$, is called the macroscopic cross section, and is
denoted by $\Sigma$. The macroscopic cross section has units of inverse length. It can be interpreted as the probability that a neutron will undergo a reaction within a unit path length in the target.

The total cross section characterizes the probability that any type of neutron-nuclear reaction will occur. The proportion in which this probability is distributed among the various possible reactions is fixed for a given nuclide and neutron energy. Hence, the total cross section for a sample material at neutron energy $E$ can be represented as

$$\sigma_t(E) = \sum_j \sum_i \sigma_{ji}(E)$$  \hspace{1cm} (3.4)

where the index $j$ represents all possible neutron-nuclear reactions, and the index $i$ represents all the isotopes making up the material of the sample.

Similar representation is followed for the macroscopic cross-sections by replacing $\sigma$ by $\Sigma$.

### 3.3.1.1 Energy dependence of neutron absorption cross sections

For low energies ($<2$ eV) the neutron absorption cross section tends to vary as $E^{-1/2}$ ($1/v$ behavior). For most of the lighter nuclei this $1/v$ behavior persists up to higher energies. However for intermediate and heavier nuclides there exists narrow bands of energies at which the cross sections show sharp increases. These peaks in the $\sigma(E)$ vs. $E$ curve are called resonances. A resonance is observed when the available
energy (i.e., the sum of the center of mass (CM) kinetic energy of the incident neutron, $E_C$, and the neutron binding energy of the product nucleus, $E_b$) matches an energy level (a metastable state) of the product nucleus. For resonances which are spaced widely apart, the Breit-Wigner single level resonance formula (Duderstadt and Hamilton, 1976) can be used to describe the energy dependence of the absorption cross section. At higher neutron energies (above a few hundred keV) the resonances become lower and broader and are closely spaced. In this region it is not possible to resolve the resonances by experimental measurements and the $\sigma(E)$ vs. $E$ curve takes on a smooth "unresolved resonance" appearance.

3.3.1.1.1 Temperature dependence of neutron absorption cross sections

In certain situations the target nuclei cannot be taken as stationary and their temperature dependent thermal motion (or vibrations) have to be taken into consideration. This effect is significant for thermal neutrons, their speed being comparable to the speed of the motion of the target nuclei. In this case the thermal cross sections of the target nuclei, at a particular neutron speed (and hence its corresponding energy), are averaged over the velocity distribution of the target nuclei. As the velocity distribution of the target nuclei is temperature dependent, cross sections must be used at the temperature of the experimental conditions.

Another situation where the thermal motion of the target nuclei has to
be considered is in the resonance region. This requirement is imposed due to the narrow energy bands of the resonances (~1 eV). The thermal motion of the target nuclei tends to change the relative speed of the incident neutron and hence broaden the neutron energy band at which the resonance is observed. This is called "Doppler broadening" of the resonance. This effect is important as an increase in temperature results in an increase in the resonance absorption. The Breit-Wigner single level resonance formula along with the Maxwell-Boltzmann distribution can be used to obtain the Doppler broadened cross sections in the vicinity of a resonance.

3.3.1.2 Energy dependence of neutron scattering cross sections

Since inelastic scattering is a threshold reaction, it usually occurs for relatively high neutron energies, say above 10 keV. The cross section for inelastic scattering is small for the low energy fast neutrons. The cross section increases with the neutron energy, approaching the geometrical cross section of the target nucleus.

Elastic scattering is of two types, elastic resonance scattering and potential scattering. Elastic resonance scattering, like the absorption reaction, also takes place through compound nucleus formation. The compound nucleus re-emits the neutron, with the target nucleus returning to its ground state. Potential scattering is essentially an energy independent, billiard ball type of collision reaction. The cross section for potential scattering is usually close to the geometrical
cross section of the target nucleus.

3.3.1.2.1 Low energy and temperature dependence of neutron scattering cross sections

For low energies the neutron no longer interacts with a free nucleus, but rather with an aggregate of bound nuclei. The neutron can interact with the internal modes of the target material, such as crystal lattice vibrations (in solids) and molecular rotations (in fluids). Also the effect of target nuclei thermal motion cannot be ignored for interactions with thermal neutrons. Hence, in general, the scattering cross sections depend on the physical and chemical forms and on the temperature of the scattering material.

The ENDF/B $S(\alpha,\beta)$ scattering law (Duderstadt and Hamilton, 1976) is the most rigorous treatment for thermal neutron scattering and is derived from the quantum mechanical theory of neutron scattering from a system of bound nuclei. The option of using this model for thermal neutron scattering (instead of the routinely used simpler free gas model) is available in MCNP and is used for light nuclei in this work.

3.3.2 Neutron moderation

The transport equation and its diffusion approximation were not required for this work as the flux distributions were calculated using the Monte Carlo code MCNP. However a study of the moderation process was
considered useful in gaining physical insight into the problem and is discussed briefly.

3.3.2.1 Fick's law

Fick's law is an important relationship between the current density, $J$, and the flux $\phi$. The current density is the net rate at which neutrons pass through a surface of unit area. Flux is the total rate at which neutrons pass through a surface of unit area, regardless of the direction in which they cross. Both have the same units, i.e., $#/\text{cm}^2\cdot\text{s}$.

Fick's law (also referred to as the diffusion approximation) implies that a spatial variation in the neutron flux (or density) will give rise to a current of neutrons flowing from regions of high to low density.

Mathematically Fick's law states that the current density vector is proportional to the negative spatial gradient of the flux.

$$J = -D \nabla \phi$$

where the proportionality constant $D$ ($D = \lambda_{tr}/3$), is called the diffusion coefficient, and $\lambda_{tr}$ is the transport mean free path. The transport mean free path is the net distance travelled by the neutron in its original direction. The inverse of the transport mean free path is the probability that the neutron will travel a unit distance in the material in its original direction. This then becomes the macroscopic transport cross section $\Sigma_{tr}$. Hence $\Sigma_{tr} = 1/\lambda_{tr}$. Also, by analogy to
previous definitions of the macroscopic cross section, $\Sigma_{tr} = N\sigma_{tr}$. In terms of the macroscopic cross sections, it can be shown that

$$D = \frac{\lambda_{tr}}{3} = \frac{1}{3\Sigma_{tr}} = \frac{1}{3 (\Sigma_t - \bar{\mu}\Sigma_a)} \quad (3.6)$$

where $\bar{\mu} = \frac{2}{3A}$ (A = mass number) is the average cosine of the angle through which a neutron will scatter from its original direction, in the case of isotropic scattering, in the CM system.

3.3.2.2 Characteristic lengths

The average distance travelled by neutrons from a localized source is characterized by certain lengths. These lengths provide a measure of the spatial spread of the neutrons, either to their being slowed down to a specified energy (slowing down length) or to their being absorbed during thermal diffusion (migration length). The characteristic length for neutrons at thermal energies is called the diffusion length.

Evaluation of the characteristic lengths can serve to identify regions which contribute importantly to the reactions of interest and assist in the study and assessment of the factors which would significantly perturb the flux and hence the reaction of interest. The fluxes can easily be generated in a simplified geometry (e.g., by the Monte Carlo method) in the material of interest and the characteristic lengths calculated. The effect of changes in material composition on the neutron spatial distribution can be studied in this way.
3.3.2.2.1 Slowing down length

If in some way (solution to the transport equation including the diffusion approximation, if valid) a neutron spatial flux distribution $\phi(z)$ is derived, then the $n^{th}$ moment of the flux is given by (Sanders, 1983)

$$
\bar{Z}^n = \frac{\int Z^n \phi(Z) dV}{\int \phi(Z) dV} \quad \text{(3.7)}
$$

where $Z$ is the distance coordinate from the source to a point in the material and $dV$ is a volume element at the distance $Z$. The first moment is the average crow flight distance a neutron travels from source to absorption and the second moment is the corresponding mean square distance.

For a plane source in an infinite medium the square of the slowing down length, $L_s(E)$, from some initial source energy to a final energy $E$, is $1/2$ times the mean square distance to slowing down. Hence

$$
2L_s^2(E) = \bar{Z}^2 = \frac{\int Z^2 \phi(E,Z) dV}{\int \phi(E,Z) dV} \quad \text{(3.8)}
$$
3.3.2.2 Migration and diffusion lengths

The migration length, $M$, can also be defined in a way similar to the slowing down length, by replacing $\Phi(E, Z)$ by $\Phi_{th}(Z)$ in the equation for the slowing down length. The relationship connecting the characteristic lengths is

$$M^2 = L_s^2(E_c) + L^2$$  \hspace{1cm} 3.9

where $E_c$ is a suitable neutron thermal energy cutoff and $L$, the diffusion length, is given by

$$L = \sqrt{\frac{D_{th}}{\Sigma_{a,th}}} = \frac{\lambda_{tr,th}}{3 \Sigma_{a,th}}$$  \hspace{1cm} 3.10

3.3.2.3 Neutron lethargy

The neutron lethargy is defined as

$$u = \ln \frac{E_0}{E}$$  \hspace{1cm} 3.11

where $E_0$ is the maximum energy of interest and $E$ is the neutron energy. It is noted from the definition that as the neutron energy $E$ decreases, its lethargy increases, i.e., as a fast neutron loses energy via scattering collisions, it moves more slowly, and becomes more "lethargic".
The average lethargy gain, $\xi$, (corresponding to the average logarithmic energy loss) of a neutron in a collision with a nucleus of mass $A$ is given by

$$\xi = 1 + \frac{\alpha}{1 - \alpha} \ln \alpha$$  \hspace{1cm} \text{3.12}$$

where $\alpha = \left(\frac{A - 1}{A + 1}\right)^2$.

3.3.2.4 Moderating power and moderating ratio

The average number of collisions required to thermalize a neutron from some higher energy $E_0$ is given by

$$\# = \frac{u}{\xi}$$  \hspace{1cm} \text{3.13}$$

Note from Equation 3.13 that the number of collisions required to thermalize the neutron is inversely proportional to $\xi$. Hence a good moderator will be characterized by a large value of $\xi$. Additionally, a good moderator must also be characterized by a large scattering cross section, since otherwise the probability of a scattering collision occurring will be small. Hence a more appropriate measure of the moderating or slowing down power of a material is the product $\xi \Sigma_s$. The moderating power (MP) is then defined as

$$\text{MP} = \xi \Sigma_s$$  \hspace{1cm} \text{3.14}$$
However, even the moderating power is not sufficient in itself to describe the effectiveness of a material for neutron moderation. It is also desirable to maintain a high population of thermal neutrons, which requires that the moderator have a small absorption cross section. Hence the moderating ratio (MR) defined as

$$MR = \frac{\xi \Sigma_s}{\Sigma_{a,2200}}$$

is chosen as a figure of merit for a moderator.

The MR of H$_2$O is 71, whereas D$_2$O has a MR of 5670, making D$_2$O a superior moderator for most applications.

3.4 Neutron sources

Presently the most prolific source of neutrons is the nuclear reactor. But for field applications, where compact neutron sources are essential, isotopic sources and neutron generators are used.

3.4.1 Isotopic sources

Many heavy (high atomic mass) radioisotopes decay by spontaneous fission and in the process emit neutrons. Among these is $^{252}$Cf, which is an alpha emitter. It undergoes 10 spontaneous fissions for every 313 alpha transformations. Its effective half life is 2.65 years (Cember, 1986). $^{252}$Cf emits neutrons at the rate of $2.31 \times 10^6$ neutrons per $\mu$g per second.
The emitted neutrons span a wide range of energies. The most probable energy is 1 MeV, while the average energy is 2.136 MeV. The energy distribution of the $^{252}$Cf source can be represented by a Watt spectrum (Briesmeister, 1989)

$$X(E) = C \exp(-E/a) \sinh(bE)^{1/2}$$

where, $a = 1.025 \text{ MeV}$ and $b = 2.926 \text{ MeV}^{-1}$

Among isotopic sources, $^{252}$Cf provides the highest specific neutron yield, the lowest mean neutron energy and a relatively low gamma ray emission rate. The low mean energy is important as the capture cross section of many elements are largest for thermal neutrons, and the low mean energy also reduces the moderator requirements. The above factors make $^{252}$Cf the most popular neutron source for PGNAA field applications.

All other neutron sources depend on nuclear reactions, such as ($\alpha$,n) and ($\gamma$,n), for the emission of neutrons. Sources using the ($\alpha$,n) reaction are available in which beryllium, $^9\text{Be}$, is bombarded with alpha particles. The source of the alpha particles could be any one of the alpha emitting isotopes, such as $^{210}\text{Po}$, $^{239}\text{Pu}$, $^{226}\text{Ra}$ or $^{241}\text{Am}$. The energy of an emitted neutron will depend on the mass defect as well as the kinetic energy of the initiating alpha particle and the recoil nucleus. As some of the energy of the alpha particles is dissipated due to self absorption in the source material, the energy spectrum of the emitted neutrons spans a wide energy distribution (0 to 11 MeV), with an average energy of about 4.5 MeV.
A table of various (α,n) and (γ,n) neutron sources with their associated mean neutron energies and specific yields may be found in the literature (Cember, 1986). The majority of the (γ,n) sources have very short half lives. The only (γ,n) source with a long half life (Ra + D₂O) has a very low specific yield of 10³ neutrons per second per curie.

3.4.2 Neutron generators

Copious neutron beams may also be produced in accelerators by many different reactions

\[ ^{3}_{1}T + ^{2}_{1}D = ^{4}_{2}He + ^{1}_{0}n \]
\[ ^{2}_{1}D + ^{2}_{1}D = ^{3}_{2}He + ^{2}_{0}n \]

In the former case neutrons can be produced with an energy of 14 MeV, and high yields can be obtained. These high energy neutrons are suitable for methods based on inelastic scattering, and the generator has the advantage that it can be turned off when not in use, thus reducing unnecessary exposure and irradiations. Pulsing the generator is also important and can be used for discriminating the inelastic scattering gamma rays from other capture related gamma rays and in neutron lifetime measurements. The neutron lifetime measurements are important in oil well technology for distinguishing between strata bearing fresh or saline water and for measuring the delayed neutron responses of uranium ores subject to delayed neutron irradiations.

The neutron generator has the disadvantage of being expensive,
delivering a somewhat unsteady neutron flux, and having a relatively short generator tube lifetime.

3.5 Neutron capture reaction rates

Let \( R(\vec{r}, E) dE \) be the number of capture reactions occurring per unit volume per unit time at position \( \vec{r} \) in the energy interval \( E \) to \( E + dE \). Then

\[
R(\vec{r}, E) dE = \Phi(\vec{r}, E) \Sigma_c(E) dE
\] 3.17

The flux \( \Phi(\vec{r}, E) \) can be written as \( n(\vec{r}, E) v(E) \), where \( n(\vec{r}, E) \) is the neutron density per unit energy in the energy interval \( E \) to \( E + dE \) and \( v(E) \) is the velocity of the neutron at energy \( E \).

It is useful to break up the energy dependent reaction rate into two parts, one for the thermal region (where the \( 1/v \) behavior is seen) and one for the resonance region.

\[
R(\vec{r}) = \int_0^E R(\vec{r}, E) dE = \int_0^{E_c} \Phi(\vec{r}, E) \Sigma_c(E) dE + \int_{E_c}^\infty \Phi(\vec{r}, E) \Sigma_c(E) dE
\] 3.18

where \( E_c \) is the thermal cutoff energy, usually set to 0.5 eV.

3.5.1 Thermal neutron capture rates

The thermal neutron interaction rate for \( 1/v \) absorbers may be expressed either as
where $E_0 = 0.0253$ eV, is equal to the most probable energy in a Maxwellian flux at $T = 293$ K. The corresponding speed of the neutron $v(E_0)$ is 2200 m/s. $\Phi_0$ (or $\Phi(E_0)$) is the 2200 m/s flux. $\Sigma_c$ is the cross section averaged over the thermal neutron distribution at temperature $T$, and $\Phi_{th}(\vec{r})$ is the thermal flux at the temperature of the medium. We note that $R_{th}(\vec{r})$ is a constant, independent of the velocity distribution of either neutrons or nuclei. Thermal cross sections are usually tabulated at $E_0$.

For non-1/$v$ absorbers (usually intermediate and heavy nuclei), a temperature dependent 1/$v$ correction factor is incorporated into the rate equation

$$R_{th}(\vec{r}) = g_c(T)\Sigma_c\Phi_{th}(\vec{r})$$

3.21

The values for the non 1/$v$ correction factor, $g_c$, are available in the literature.

3.5.2 Resonance capture

From Equation 3.18, the resonance capture contribution to the reaction rate is
\[ R_{\text{res}}(\vec{r}) = \int_{E_c}^{E} \Phi(\vec{r}, E) \Sigma_c(E) \, dE \] 3.22

If the flux of higher energy neutrons varies as \( 1/E \), then

\[ \Phi_{\text{epi}}(\vec{r}, E) = \frac{\lambda(\vec{r})}{E} \] 3.23

where \( \lambda(\vec{r}) \) is an energy-independent proportionality constant. Hence

\[ R_{\text{res}}(\vec{r}) = \lambda(\vec{r}) \int_{E_c}^{E} \frac{\Sigma_c(E)}{E} \, dE \] 3.24

\[ = \lambda(\vec{r}) \Sigma_{Ic} \]

where \( \Sigma_{Ic} = \int_{E_c}^{E} \frac{\Sigma_c(E)}{E} \, dE \) is the resonance integral for the capture reaction, values for which are available in the literature. Hence the capture rate at any position \( \vec{r} \) in a sample can be given by

\[ R(\vec{r}) = \Sigma_c \Phi_{\text{th}}(\vec{r}) + \Sigma_{Ic} \lambda(\vec{r}) \] 3.25

More insight can be gained by expressing the above equation in terms of the epithermal flux normalized to unit thermal flux

\[ \Phi_{\text{epi},n}(\vec{r}, E) = \frac{\Phi_{\text{epi}}(\vec{r}, E)}{\Phi_{\text{th}}(\vec{r})} = \frac{\lambda(\vec{r})}{\Phi_{\text{th}}(\vec{r}) E} \]

where the subscript \( n \) signifies the normalization by the thermal flux.
Hence the total reaction rate is given by

\[
R(\bar{r}) = \Sigma_c \Phi_{th}(\bar{r}) + \Sigma_{lc} \Phi_{th}(\bar{r}) \lambda_n(\bar{r})
\]

\[
= \Phi_{th}(\bar{r}) \left[ \Sigma_c + \Sigma_{lc} \lambda_n(\bar{r}) \right]
\]

3.27

If \( \lambda_n(\bar{r}) \) is known along with the thermal flux, the epithermal reaction contribution can be calculated from the tabulated values of the resonance integral, \( \Sigma_{lc} \). However, these values are difficult to find for small energy intervals in the epithermal region. Usually the value of the resonance integral is calculated assuming a \( 1/E \) behavior over the entire epithermal and fast flux. This assumption is reasonable for well moderated neutron spectra. However, due to changes in the material properties, as is often the case in field applications, a spectrum may be significantly perturbed, leading to uncertainty in the results.

3.6 Gamma rays

Gamma radiation are electromagnetic vibrations carried by photons. These \( \gamma \)-rays are produced either during nuclear transformations (radioactive decay, nuclear reactions, etc.), by the stopping of charged particles in the medium (bremsstrahlung), or during the annihilation of particles and anti-particles (e.g., an electron with a positron). In this work, prompt or capture gamma rays are of most significance. A summary of gamma ray emitting processes caused by neutrons of various energies is given in Figure 3.1.
The radiative neutron capture reaction is the prime source of capture prompt gamma rays. This reaction can take place at all incident neutron energies of interest here (thermal to 10 MeV), although the probability is highest at thermal energies.

3.6.1 Capture gamma rays

A schematic of the capture gamma ray production reaction is shown in Figure 3.2.
At low neutron energies, the capture reaction is best explained by the compound nucleus model. In this model the absorption of the neutron is followed by a rapid distribution of the available energy among the other nucleons. The system decays when sufficient energy, by chance, becomes concentrated in a nucleon or a group of nucleons. This lifetime is of the order of $10^{-16} - 10^{-18}$ seconds.

A compound nucleus may decay in a variety of different ways. The
relative probability of decay into any specific set of final products is independent of the means of formation of the compound nucleus. The decay probability depends only on the energy given to the nucleus, i.e., the available energy.

For heavy nuclei and slow neutrons, decay by γ-ray emission is the most probable decay mode (Krane, 1986). For thermal neutron capture the available energy is close to the binding energy of the neutron in the compound nucleus. This energy ranges from 2.2 MeV in hydrogen to 11 MeV in nitrogen (Garrett et al., 1973).

3.6.2 Capture gamma ray spectra

The spectra of most nuclides are fairly complex, with discrete energies ranging up to the value of the neutron binding energy. The γ-ray spectrum is complex since all excited states can be populated to some extent, limited only by the quantum mechanical selection rules for γ radiation. Due to the above quantum mechanical considerations, high energy transitions, i.e., those that populate the lower excited states are most likely. Thus the γ-ray spectrum shows two principal components: the primary component (consisting of radiation coming directly from the capture state to lower excited states, usually below 2 MeV), and the secondary component (consisting of low energy radiations between the low lying excited states).

The primary radiation is completely unselective in populating the lower
energy states. However, for a neutron capture reaction at a particular energy, each nuclide displays a unique spectrum.

3.6.2.1 Energy dependence of gamma ray spectra

The spectrum of any particular element is made up of the weighted contributions of the several naturally occurring isotopes of that element. At different incident neutron energies, the shape (relative intensities) of the prompt gamma ray line spectrum can change. Further, capture in the resonance region leads to a broadening of the capture \(\gamma\)-ray peaks and shifts the centroid of the peak towards higher energies (Broomhall, 1972 and Kenny, 1971). This is usually true for the primary \(\gamma\)-rays and hence primary photons are useful as signature photons if produced in thermal neutron capture. Hence it would be better to select those signature photons for the purpose of elemental analysis which do not have a significant production cross section from non-thermal neutron capture.

However, the relative intensity of such a primary photon in the capture gamma ray spectrum of a nuclide may still change for neutron capture at non-thermal energies. The effect of neutron energy on the relative intensities of various \(\gamma\)-rays in the capture \(\gamma\)-ray spectrum of various elements at thermal and higher energies can easily be seen in the keV neutron radiative capture data reported by Bird et al., 1973. Hence a variation in the spatial and energy distribution of the neutron field, due to the variation in the formation variables, can lead to a change in
the capture γ-ray spectrum generated.

Serious errors can result if the capture γ-ray intensities reported for thermal neutron captures are used for epithermal neutron captures. This error becomes significant if the proportion of epithermal neutrons is high. This makes radiative capture an elementally and energetically specific reaction. However, with proper calibration, this problem can be overcome.

3.7 Gamma ray interactions with matter

There are more than ten elementary processes of interaction of γ-rays with matter. But for photons of the energies considered here, only three processes occur with significant probability. They are photoelectric absorption, Compton scattering, and the pair production process.

As a result of these processes, a part or all of the γ-ray energy is transmitted to electrons, resulting in a photoelectron, a Compton electron or an electron-positron pair. The energy of the electron, as a result of any of the above primary events, is transferred to the material near the point of collision. However, part of this energy may subsequently be emitted in the form of radiation (fluorescence, bremsstrahlung of electrons and positrons, and annihilation radiation). The energy and intensity of this secondary radiation is considerably less than that of the primary radiation, so that its effect can usually
be neglected in comparison with the effect of the primary radiation (particularly for dose rate calculations).

3.7.1 The photoelectric effect

The ejection of photoelectrons from a surface as a result of photon absorption is called the photoelectric effect. The incoming $\gamma$-ray completely disappears in this process. This interaction occurs between a photon and an electron when the binding energy of the electron, $(BE)_e$, is less than or equal to the energy, $E$, of the incident photon.

The energy of the photoelectron, $E_{pe}$, is the difference between the energy of the incident $\gamma$-ray and the binding energy of the electron, i.e., $E_{pe} = E - (BE)_e$. The photoelectron transfers its energy to the material mainly by excitation and ionization. The binding energy of the electron is also deposited in the material by subsequent emission of fluorescence radiation leading to further photoelectrons. In this way all the energy, $E$, of the incident $\gamma$-ray may be deposited within a small region of the material.

The photoelectric effect is favored by low energy photons and high $Z$ absorbers. For low energies (less than 0.2 MeV) the cross section varies approximately as $Z^5 E^{-3.5}$ and as approximately $1/E$ for much higher energies. The cross section varies discontinuously at $\gamma$-ray energies equal to the binding energy of the electrons in the atom. Energy and momentum considerations imply that photoelectric absorption cannot take
3.7.2 Compton scattering

When the interaction of a γ-ray with an electron (orbital or free), results in the γ-ray losing part of its energy to the electron, the γ-ray is said to have undergone inelastic scattering. This process is called Compton scattering. The energy lost depends only on the initial γ-ray energy $E_0$ and the angle through which the incident γ-ray is scattered from its original direction. The final energy of the γ-ray, $E'$, is given by

$$E' = \frac{E_0}{1 + \left(\frac{E_0}{m_0c^2}\right)(1 - \cos\theta)}$$  \hspace{1cm} 3.28

where $m_0$ is the rest mass of the electron.

As the γ-rays transfer their energy mainly to electrons, it is convenient to measure the energy of the γ-rays in units of the electron rest mass energy, $m_0c^2$, which is equal to 0.511 MeV. The energy of γ-rays expressed in such units is denoted by $\alpha$. Hence

$$\alpha' = \left(\frac{E'}{m_0c^2}\right), \text{ and } \alpha = \left(\frac{E_0}{m_0c^2}\right)$$  \hspace{1cm} 3.29

Thus Equation 3.28 can be written in terms of $\alpha$ as
\[
\alpha' = \frac{\alpha}{1 + \alpha(1 - \cos\theta)}
\]

The Compton scattering cross section, \(\sigma_{cs}\), is proportional to the electron density, and hence to the atomic number of the interacting atoms. Above 0.5 MeV the Compton scattering cross section varies as \(1/E\). The angular distribution of the scattered photons is described by the Klein-Nishina equation (Garrett et al., 1973).

3.7.3 Pair production

Pair production is a threshold reaction in which the photon possessing at least twice the electron rest mass energy (\(\alpha = 2\)) gets converted into an electron-positron pair. This reaction can take place in the field of an atomic nucleus or in the field of an atomic electron. The probability for the reaction to occur in the field of an electron is considerably smaller and the threshold energy twice as much.

The excess of gamma ray energy above the threshold energy is the kinetic energy shared by the electron-positron pair. The positron slows down within a short distance of its production and annihilates, giving rise to two annihilation photons of 0.511 MeV each, moving in opposing directions to conserve momentum. The cross section for pair production increases rapidly above the threshold energy and varies as \(Z^2\).
3.8 Propagation of gamma rays in matter

Analogous to the concept of the macroscopic cross section for neutron interactions, the probability that a γ-ray interaction will take place per unit path length in the material is given by the linear attenuation coefficient, \( \mu \). Hence the total linear attenuation coefficient, \( \mu_t \), is given by

\[
\mu_t = \mu_{pe} + \mu_{cs} + \mu_{pp}
\]

where

- \( \mu_{pe} \) is the photoelectric absorption event,
- \( \mu_{cs} \) is the Compton scattering event, and
- \( \mu_{pp} \) is the pair production event.

Also similar to the microscopic cross sections for neutron interactions, the probability that an absorber atom of the material will interact with an incident photon is given by the atomic attenuation coefficient, or simply "cross section" \( \sigma \). Hence \( \mu = N\sigma \), where \( N \) is the number density of the absorber atoms. The linear attenuation coefficient \( \mu \) is often used in the form of \( \mu/\rho \), i.e., the mass attenuation coefficient, which has the advantage of being independent of the density state of the material.

By a procedure similar to the one used to define the macroscopic cross section in terms of a reduction in the incident neutron beam intensity, it can be shown that the γ-rays are also exponentially attenuated, i.e.,
where

\[ I(x) = I_0 \exp(-\mu x) \]

\[ 3.31 \]

I(x) is the beam intensity at position x into the material

and

\[ I(x) \]

\[ \mu \]

is the total linear attenuation coefficient.

As cross sections are functions of energy, so is the linear attenuation coefficient, \( \mu \). Hence for a non-monoenergetic source, the above equation can be applied separately at each energy to calculate the attenuation.

The intensity (or flux) given by the above equation is called the "uncollided" flux. The uncollided flux is a measure of the primary radiation, i.e., radiation which has not yet interacted or collided with the material atoms.

The total intensity at any point x would be higher due to the contributions from secondary radiations. To account for this contribution the uncollided flux is multiplied by a buildup factor B.

Such buildup factors have been calculated or measured for various \( \gamma \)-ray energies and for various absorbers for many of the common geometries (Liepunskii et al., 1965; Cember, 1986).

The buildup factor is used in shielding and dose calculations for a realistic estimate of the shield thickness and dose rates. In activation analysis it contributes to a higher background in the \( \gamma \)-ray
detector response spectra. As this is an undesirable feature, an experimental configuration with "good geometry" is usually sought (Figures 3.3 (a) and 3.3 (b)).

Figure 3.3 (a). Propagation of gamma quanta under conditions of "good geometry" (Liepunskii et al., 1965).

Figure 3.3 (b). Propagation of gamma quanta under conditions of "bad geometry" (Liepunskii et al., 1965).
In a good geometry the contribution from secondary radiations to the γ-rays incident on the detector is negligible. This assumption is reasonable only if the radius of the collimator used is much smaller than the mean free path of the secondary γ-rays (Liepunskii et al., 1965).

3.9 Detection of gamma rays

Detectors are used to measure the energy spectrum of the emitted γ-rays. As the capture gamma ray energy spectrum from a multi-element sample is highly complex, a detector with a high energy resolution is required. Along with this high energy resolution, a reasonable γ-ray interaction efficiency within the detector material is also necessary. Germanium semiconductor detectors have excellent energy resolution with up to 100 percent efficiency relative to a 3 × 3 inch NaI(Tl) scintillation detector.

3.9.1 Semiconductor junction detectors

The usefulness of semiconductors as electronic circuit elements and for radiation measurements stems from the special properties created at a junction where n-type (electron rich donor atoms, e.g., As in Ge) and p-type (electron deficient acceptor impurity atoms, e.g., Ga or In in Ge) semiconductors are brought into good thermodynamic contact.

When the n- and p-type semiconductors are brought into contact,
electrons move from the n-type to the acceptor p-type region. This leads to a separation of charge across the junction. This region is called the depletion region and forms the sensitive or active volume of a detector.

The depletion region acts like an ionization chamber, making it suitable for radiation detection. Ion pairs produced here migrate outwards with the electrons moving toward the electron deficient n-side and holes to the opposite side. A high reverse bias voltage (positive on the n-side, typically 1000 V/cm), applied to the ends of the semiconductor, helps in optimum charge collection and noise reduction.

When $\gamma$-rays enter a detector, any of the three primary interactions (Section 3.7) may take place. The interacting $\gamma$-ray may deposit some or all of its energy in the detector material. This energy may lead to charge production, i.e., the formation of electron-hole pairs, or it may be lost to other competing processes. The amount of charge collected is related to the energy deposited (assuming complete charge collection), through the parameter $\epsilon$ (3 eV/electron-hole pair for Ge). This factor represents the average energy (including that lost to competing processes) required to produce an electron-hole pair.

For a given energy, $E$, absorbed in the primary interaction, the total number of electron-hole pairs created is subject to statistical fluctuations, due to the random energy loss division between the useful ionization process and competing processes. This sharing is described
in terms of the Fano factor, $F$ (large germanium detectors have $F \approx 0.13$, Pehl, 1977).

The standard deviation in the average number of electron-hole pairs produced for a given energy $E$, is given by

$$
\sigma = \left( F \left( \frac{E}{\epsilon} \right) \right)^{1/2}
$$

This number determines the resolution of the detector. Hence important characteristics for a detector include:

1. A linear relation between the number of electron-hole pairs produced and the energy absorbed, i.e., $\epsilon$ independent of energy.

2. A small value of $\sigma$ relative to the number of electron-hole pairs produced.

3. Trapping defect-free detector material, as these defects could lead to incomplete charge collection and hence additional statistical fluctuations, resulting in a degradation of resolution.

The trapping defects mentioned above can be produced by particle radiation such as fast neutrons. It is observed that for germanium detectors a significant energy resolution degradation occurs after a
fluence of $10^9$ to $10^{10}$ 5.5 MeV neutrons per cm$^2$ (Pehl, 1977; Knoll, 1979). Annealing of the high purity germanium (HPGe) detectors is seen to restore the resolution (Stelson et al., 1972; Kraner et al., 1975).

3.9.2 Detection system

![Figure 3.4 Schematic of a gamma ray detection and analysis system.](image)

Schematic representation of a typical gamma ray detection and analysis system in given in Figure 3.4.

The energy deposited by a $\gamma$-ray is measured by the detection system in the form of a voltage pulse signal. The pulse height or magnitude of
the voltage pulse is linearly proportional to the energy deposited. The pulses are processed by a multichannel analyzer (MCA) which integrates the pulses in various pulse-height windows (called channels). The resulting pulse-height spectrum can be displayed as a graph in which the horizontal axis represents the pulse height and the vertical axis the number of pulses. This pulse-height spectrum can then be used to determine the energies of the emitted γ-rays (from the location of the peak centroids on the horizontal axis) and the relative intensities of the γ-rays (from the net area of the various peaks in the spectrum).

Time resolution and resolving time are important characteristics of the detection system. The time resolution is the overall timing uncertainty in the measurement system, and the resolving time is the time interval required by the detection system to uniquely process a single interacting quantum of radiation. The time resolution is usually within 10 to 100 ns for large volume Ge detectors, whereas the resolving time is a circuit parameter usually limited by the peak shaping time of the linear amplifier (which is about 5 μs).

Hence if two or more γ-rays were to enter a detector within less than the resolving time, the generated pulse becomes a "sum pulse" and is no longer representative of a single energy γ-ray from the source. This phenomena is called the pulse pile-up effect.

The γ-rays entering the detector within the resolving time may be either random or related to each other (such as two γ-rays from the same
cascade). Related $\gamma$-rays would result in a definite coincidence sum spectrum of their own. This type of coincidence usually results in a distortion or broadening of the high energy side of the peak, which persists for several channels before dropping down to the continuum. The random coincidences occur when $\gamma$-rays from different atoms in the irradiated material, by chance, enter the detector within the resolving time. As these are uncorrelated, the contribution from such random coincidences is not definite, but forms a continuum which increases the background, reduces the peak count and hence degrades the peak to background ratio (Section 3.9.3).

The effect of coincidence pulse pile-up is mainly dependent upon factors such as the resolving time, the solid angle subtended by the detector and the efficiency of the detector (which is a function of the $\gamma$-ray energy). The effect of random pulse pile-up, in addition, also depends upon the count rate which is related to the activity of the source. A high activity source increases the probability that two unrelated $\gamma$-rays will be emitted and enter the detector within the resolving time of the detector. Another important effect of high count rate is the "dead time". This occurs when counts are lost due to the MCA gate being closed while it is processing a previous pulse (i.e., the conversion of the voltage pulse into its digital equivalent and its subsequent storage in the proper memory location).

Hence for higher count rates faster electronics are usually required. Methods have been developed for correcting the pulse-height spectrum for
the pulse pile-up effect and recent activation analysis software packages incorporate the pulse pile-up correction (Mitchell et al., 1989). Although such methods can reduce the distortion, they cannot eliminate it.

3.9.3 Spectral features

3.9.3.1 Photopeak

When the γ-ray interacts by photoelectric absorption, all the γ-ray energy is usually deposited in the detector, and hence this type of interaction will result in a pulse being counted in a channel proportional to the full energy of the incoming photon (assuming no pulse pile-up effect). The pulse is said to have contributed to the photopeak or its full energy peak in the spectrum.

3.9.3.2 Compton continuum

If the γ-ray Compton scatters (one or more times), loses some of its energy and is ultimately absorbed in photoelectric absorption, it too would contribute to the full energy peak or the photopeak. However the γ-ray can escape from the detector after one or more scatterings. This would result in partial energy deposition. As the energy of the escaping γ-ray has a continuous distribution, it gives rise to a continuum in the energy response of the detector. This contribution to the spectrum is the Compton continuum, ranging from zero energy up to a
maximum energy called the Compton edge. The Compton edge is the maximum energy that a γ-ray can lose to an electron in a Compton scattering event. This happens when the γ-ray backscatters. Hence the energy of the Compton edge, $E_{ce}$, is given by

$$E_{ce} = E_\gamma \left( \frac{1}{1 + \left( \frac{2E_\gamma}{E_0} \right)} \right)$$

The shape of the Compton continuum is determined by the Klein-Nishina equation. The Compton continuum decreases with increasing energy.

3.9.3.3 Escape peaks

In pair production, both annihilation quanta may be absorbed, thus contributing to the photopeak, or one or both of the annihilation photons may escape. In the detector energy response spectrum, the escaping annihilation photons contribute to the single and double escape peaks, shifted down by 0.511 MeV and 1.022 MeV, respectively, from the photopeak.

3.9.3.4 Peak to background ratio

The relative magnitudes of the photopeak, the Compton continuum, and the escape peaks depend on the size and shape of the detector. In general the larger the detector, the smaller is the Compton continuum relative
to the photopeak. The pulse pile-up random coincidences also contribute to the continuum and hence increases the background. This reduces the peak to background (P/B) ratio. A higher P/B ratio enables small peaks to be visible over the background and hence is desirable.

One of the methods extensively utilized to improve the P/B ratio is to use a large NaI(Tl) crystal surrounding the semiconductor detector. The surrounding crystal in association with an electronic circuit is used in an anticoincidence mode to reject events which give simultaneous pulses in both the detectors. A spectrum collected in this way is said to be Compton suppressed. A reduction in the Compton background of 1.5 to 4 can be obtained.

Another mode of spectrum collection is called the pairs spectrum. This is done by recording only those pulses from the HPGe detector which have a coincidence signal proportional to 1022 ± 50 keV from the surrounding NaI(Tl) detector. The resulting spectra are generally simplified with only double escape peaks appearing over a very low background. This spectra is mainly used for qualitative identification purposes.

3.9.3.5 Photopeak resolution

As described in Section 3.9.1, statistical fluctuations in the net charge produced determine the resolution of the detector. Some non-statistical processes (incomplete charge collection and electronic noise) also influence the resolution. The resolution is expressed in
terms of $\Delta E$, the energy width of the photopeak at half its maximum height. This is called the full width at half maximum (FWHM). The resolution is expressed as a ratio of FWHM to the energy of the photopeak, i.e., as $\Delta E/E$. FWHM can be expressed as $2.35 \sigma$, where $\sigma$ is given by Equation 3.3.2. A typical value for a good Ge detector at 1332 keV is around 1.7 keV. As the FWHM has an $E^{1/2}$ dependence, $\Delta E$ increases with energy, but the resolution $\Delta E/E$ decreases (i.e., improves) with energy proportional to $E^{-1/2}$.

3.10 Spectrum analysis

In spectroscopy measurements, the goal is usually to determine the energy and intensity of the radiation, from which the nuclides and their concentrations can be inferred. To determine the area related to the energy of the centroid, the background over which the peak rests has to be subtracted. For a simple spectrum this can be done easily, but for complex spectra sophisticated fitting programs that can handle many parameters are required. Peak shaping functions and non-linear background fitting under the peak are important considerations for fitting spectra which may have overlapping peaks and multiplets (Westmeier, 1986). A built-in capacity to correct for pulse pile-up is highly desirable. Interferences can also degrade the resolution, and the correction for this is usually accomplished by having reference libraries of various nuclides and subtracting the contribution from the interfering nuclide using this standard spectra (this spectra should ideally be collected for the same experimental conditions as used in the
actual measurement). This process is called stripping. Peak shaping functions can also be used to unfold contributions from such overlapping peak interferences.

Another powerful analysis technique, called full spectrum analysis, fits the whole spectrum rather than specific features such as photopeaks. Although this is a powerful technique, spectral templates are needed for all the elements in the sample. Also, when the gamma ray source is either self attenuating or shielded, the photon flux (including the secondary radiation) at the detector (as calculated by the transport code) can then be fitted. The resulting data can then be analyzed for information pertaining to the intervening material.

3.11 Measurement of gamma rays

In order to visualize the effect of formation changes on the instrument response, a study of the equation linking the count rate obtained in a pulse height spectrum to the elemental composition of the irradiated sample material was considered helpful and is discussed below. The discussion is based on a similar analysis given by Sanders, 1983.

If the source neutron flux is constant (a reasonable assumption for a long-lived isotopic source and a short irradiation time), the relationship of the detected $\gamma$-ray yield to the elemental concentration may be expressed as
where

\[ Y_{ji} = \frac{N_i}{V} b_{ji} \int \int \Phi(\vec{r}, E)(\sigma_c(E))_i \Gamma(\vec{r}, E_{\gamma_{ji}}) d\vec{r} dE \]  

\[ 3.34 \]

\[ Y_{ji} = \text{detected } \gamma\text{-ray yield of } \gamma\text{-ray } j \text{ from element } i, \]
\[ \text{i.e., the yield of } \gamma_{ji}. \]

\[ N_i = \text{total number of atoms of element } i \text{ (assumed homogeneous and uniform).} \]

\[ V = \text{total volume of sample. } N_i/V \text{ is the atom number density of element } i. \]

\[ b_{ji} = \text{branching fraction of } \gamma_{ji}. \]

\[ \Phi(\vec{r}, E) = \text{neutron flux per unit energy as a function of position in the sample.} \]

\[ \sigma_c(E)_i = \text{energy dependent microscopic capture cross section of element } i. \]

\[ \Gamma(\vec{r}, E_{\gamma_{ji}}) = \text{transmission and detection probability for } \gamma_{ji}. \]

In complex geometries an accurate evaluation of this integral is difficult using deterministic methods. Hence such calculations are usually carried out using Monte Carlo simulations. However, Equation 3.34 may be simplified and solved approximately. This approach often
provides some valuable physical insight into the problem and may also help as a rough check for the more sophisticated Monte Carlo simulations.

If Equations from Section 3.5 are used and $\lambda_n(\vec{r})$ is assumed to be fairly constant with position (a reasonable assumption for well moderated neutron spectra), then

$$Y_{ji} = \left[ (b_{ji})_{\text{th}} \Sigma_c + (b_{ji})_{\text{epi}} \Sigma_c \lambda_n \right] \times \int_{\vec{r}} \Phi_{\text{th}}(\vec{r}) \Gamma(\vec{r}, E_{\gamma j_1}) d\vec{r} \quad 3.35$$

The $\gamma$-ray transmission and detection factor, $\Gamma$ (which gives the photopeak contribution from the uncollided flux reaching the detector), may be expressed as

$$\Gamma(\vec{r}, E_{\gamma j_1}) = S_\theta(\vec{r}, \vec{r}') A_{\mu}(\vec{r}, \vec{r}') F_\eta(\vec{r}, \vec{r}') \quad 3.36$$

where

$\vec{r}$ = the position vector of the point of origin of the $\gamma$-ray in the sample.

$\vec{r}'$ = the position vector for the mean coordinate of detector position.
\[ S_\theta(\vec{r}, \vec{r}') = \] the solid angle subtended by the detector from \( \vec{r} \). When the detector is not close to the sample and is reasonably small in size, the solid angle can be approximated by \( \frac{S_D}{4\pi|\vec{r} - \vec{r}'|^2} \), where \( S_D \) is the effective cross sectional area of the detector.

\[ A_\mu(\vec{r}, \vec{r}') = \] the \( \gamma \)-ray attenuation factor, which may be written as

\[
\exp\left(-\sum_m (\mu_m) x_m\right)
\]

where the subscript \( m \) denotes the materials through which the \( \gamma \)-ray travels, \((\mu_m)\) is the \( \gamma \)-ray linear attenuation coefficient at \( E_{\gamma_{ji}} \), and \( x \) is the distance through which \( \gamma_{ji} \) travels in the direction of \(|\vec{r} - \vec{r}'|\).

\[ F_\eta(\vec{r}, \vec{r}') = \] the detector intrinsic efficiency factor. The position vectors \( \vec{r} \) and \( \vec{r}' \) determine the point and angle of entrance into the detector. An average value can usually be assigned to \( F_\eta(\vec{r}, \vec{r}') \), which depends only on the detector characteristics. Hence the photopeak efficiency of the detector as a function of energy, may be written as \( \eta_{av}(E_{\gamma_{ji}}) \).
Hence Equation 3.35 becomes

$$Y_{ji} = \eta_{av}(E_{\gamma_{ji}}) \frac{S_0}{4\pi} \left[ (b_{ji})_{th} \bar{\Sigma}_c + (b_{ji})_{ep} \Sigma_c \lambda_n \right] \times$$

$$\exp \left[ -\sum_m (\mu_m \lambda_m) x_m \right]$$

$$\int_{\int} \Phi_{th}(\bar{r}) \frac{\exp \left[ -\sum_m (\mu_m \lambda_m) x_m \right]}{|\bar{r} - \bar{r}'|^2} d\bar{r}$$

3.37

The integral in Equation 3.37 may be converted into a summation over a finite number of volume elements (denoted by subscript k).

$$\int_{\int} \Phi_{th}(\bar{r}) \frac{\exp \left[ -\sum_m (\mu_m \lambda_m) x_m \right]}{|\bar{r} - \bar{r}'|^2} d\bar{r} = \sum_k (\Phi_{th})_k \frac{\exp \left[ -\sum_m (\mu_m \lambda_m) x_m \right]}{x_k^2} \Delta V_k$$

3.38

where

$$x_k = \text{the distance from the mean coordinate position vector of the } k^{th} \text{ volume element to the mean coordinate position vector of the detector.}$$

$$(\Phi_{th})_k = \text{the average flux in the } k^{th} \text{ volume element.}$$

$$\Delta V_k = \text{the volume of the } k^{th} \text{ volume element; the volume elements are chosen small to keep the flux fairly constant in each volume element.}$$

Hence the relationship between count rate $\dot{C}$ in the photopeak of $\gamma_{ji}$ to
the number of atoms of isotope i in the sample may be expressed as

\[ \dot{C} = Y_{ji} = \eta_{av}(E_{\gamma ij}) \frac{S_0}{4\pi} \left[ (b_{ji})_{th} \bar{\Sigma}_c + (b_{ji})_{epi} \bar{\Sigma}_2 \lambda_n \right] \times \]

\[ \sum_k (\Phi_{th})_k \frac{\exp \left( -\sum_m (\mu_j)_m x_m \right)}{\chi_k^2} \Delta V_k \]

3.39

The flux distribution, and hence \((\Phi_{th})_k\) and \(\lambda_n\), can be calculated in chosen volume elements using a suitable neutron transport code. The average fluxes can then be used along with known \(\gamma\)-ray production data to compute the volume distributed \(\gamma\)-ray source. Although this method can be used to calculate photopeak count rates, the background in the peak area is ignored. But for obtaining detection limits, a knowledge of the background in the photopeak channels is essential. Hence the use of this method is limited to exploratory studies of the formation perturbation effects and as rough checks on more sophisticated calculations.

A careful look at Equation 3.39 clearly shows the effect of a significant epithermal capture contribution through the parameter \(\lambda_n\). As the \(\gamma\)-ray production data are not very extensive in the epithermal region, it becomes clear that proper calibration would play a very important role. Also, a change in the value of \(\lambda_n\) or its variation with energy can lead to significant errors (see Sections 3.5 and 3.6), and hence calibration curves for all the parameters that can significantly
3.12 The Monte Carlo method

Tracking the transport of neutrons, γ-ray or other elementary particles through matter by Monte Carlo techniques is an attempt to simulate the actual statistical nature of the interaction process. A stochastic model (which may require a preliminary parametric study) is set up. Random numbers are used to select the values of the various variables in the problem from their respective probability density distributions (e.g., choice of interaction at a collision site, angle of emergence from a collision, etc.). This sampling along with known nuclear data is used to simulate the particle’s transport through its lifetime.

The tracking and record keeping of all the events (or of those chosen by the user) that take place in a particle’s lifetime is called a "history". If sufficient histories are simulated, events (or variables computed from various events) can be scored (tallied) with acceptable precision. Examples of such events would be particle fluxes (computed from track length or collision densities) or reaction rates. These events can be scored in chosen volume or surface elements of the experimental geometry (cells and surfaces, respectively) and also in chosen energy bands. Hence spectra and reaction rate energy dependence can be computed as spatial distributions.
3.12.1 Variance reduction

When a particle's random walk directly follow the physical processes, the Monte Carlo calculations are said to be analog. Such calculations are effective only for problems where substantial number of particles are in the tally region. For problems where the events of interest have a low probability of occurrence, a large number of histories have to be run to obtain acceptable precision. This is not only expensive, but in certain situations prohibitive. Hence non-analog schemes have to be used. These techniques, called variance reduction techniques, reduce the statistical errors associated with the tallies for a given amount of computation time. In many situations, as noted by Sanders, 1983, the success or failure of the simulation depends crucially on the techniques applied for variance reduction.

The following is a discussion of the various kinds of variance reduction techniques in Monte Carlo simulation with emphasis on the more sophisticated variance reduction techniques available in MCNP and utilized in this work (adapted from Forster et al., 1990; Briesmeister, 1989).

The various variance reduction techniques may be grouped into five classes.

1. Truncation methods: Parts of the phase space (geometry, energy ranges, times, etc.) which do not contribute significantly to
the solution are truncated. The simplest example is geometry truncation in which unimportant parts of the geometry are simply not modeled. An example of energy range truncation (energy cutoff), as used in this work, is the termination of a photon's history when its energy falls below 3 MeV.

2. Population control methods: These methods use splitting and Russian Roulette to preferentially populate the regions of interest with particles which are expected to contribute significantly to the desired result. In MCNP such methods are the geometry splitting, energy splitting, weight cutoff, and the powerful weight window technique.

A weight window is an upper and lower weight bound in a energy-geometry phase space. Particles above the upper bound are split, and Russian Roulette is played for particles below the lower bound. Although the weight window is a powerful variance reduction technique and can be very effective in controlling the weight fluctuations caused by other variance reduction techniques in the problem, it is very difficult to specify, specially in a problem with a large number of energy-geometry regions. This has led to the development of the weight window generator. The generator is a statistical bookkeeping operation in which a cell's importance is estimated as the ratio of the weight tallied due to the particle which enters the cell to the total weight that enters the cell.
The generator itself has some limitations. It is statistical in nature and hence is prone to bad estimates from regions of low importance. Consequently several iterations and significant user intervention may be required for good convergence. Further the generator is specific to a tally, and hence the optimization for a particular tally leads to the deterioration in the results for other tallies. Work in improving the generator is ongoing (Soran et al., 1990; Forster et al., 1990). For a more detailed discussion on the weight window and the weight window generator, see Booth and Hendricks, 1984; Booth, 1985; Briesmeister, 1989; and Soran et al., 1990.

3. Modified sampling methods: Such methods alter the statistical sampling of a problem to increase the number of tallies per particle. In MCNP such techniques include the exponential transform (path length stretching), implicit capture, forced collision, source biasing and neutron-induced photon production biasing. Exponential transform can provide effective penetration in preferred directions (should be used in conjunction with weight windows). The implicit capture technique adjusts the weight of the particle upon capture instead of terminating it. The forced collision technique splits particles into collided and uncollided parts in specified regions. Source biasing is used to preferentially increase the probability of selecting source parameters which
are expected to contribute more to the tallies. The neutron-induced photon production biasing technique is used to produce only the desired discrete γ-ray lines from neutron-nuclear collision reactions.

In calculations of neutron-induced photon spectra, for example, in elemental analysis, the intensity of a few signature photons is frequently required. If these lines have a low probability of being sampled or if the concentration of the desired element is low, accumulating acceptable statistics is very inefficient. To overcome this problem the neutron induced photon spectrum is biased to produce only the lines of interest. Collision biasing is included to produce the desired photons at each collision regardless of the isotope involved. Although this is a very powerful variation reduction technique, extensive data are not available for many isotopes.

4. Partially deterministic methods: The normal random walk of a particle is partially circumvented by using deterministic techniques. Examples in MCNP would be the point detector tally (flux estimator at a point), DXTRAN, and correlated sampling. In the point detector the contribution of particles from each source and collision event is estimated at the desired point. DXTRAN is similar to the point detector, but it is not a tally. A DXTRAN sphere is a user specified spherical region which encloses the part of the geometry where tallies are desired.
Upon particle collision (or exiting the source) outside the DXTRAN sphere, a pseudo particle is created. The pseudo particle is deterministically transported without further collision to the surface of the sphere. The random walk is then continued within the sphere. The collision itself is treated normally, producing the usual particle that is sampled in the normal way without reduction in weight, but is terminated if it tries to enter the DXTRAN sphere.

In MCNP correlated sampling is implemented by starting the \( n^{th} \) particle with the same random number. A user patch for perturbation (due to material and density changes) calculations is available which can run up to three slightly different problems simultaneously. However, more efficient correlated sampling techniques are increasingly being used, e.g., to estimate the response from a range of elemental concentrations from a single computation at a reference concentration (Jin et al., 1987; Almasoumi, 1989) in specific purpose Monte Carlo codes.

5. Deterministic methods: Part of the problem may be solved analytically. An example would be to use an analytical detector response function instead of using Monte Carlo modeling to get the detector response. However, the pulse height detector tally is now being included in version 4 of MCNP.
In MCNP the effectiveness of a variance reduction technique or a combination of techniques is measured in terms of a figure of merit (FOM). FOM = 1 / R^2T, where T is the computer time spent and R is the estimated relative error at the 1 σ level (i.e., \( \frac{s_x}{x} \)).
4. DESIGN OF A SEAFLOOR SEDIMENT CORE ANALYZER

The basic design objectives for the elemental analysis facility for seafloor sediment cores were to achieve the best possible sensitivity and detection limits within the engineering, radiation protection and economic constraints.

The neutron/gamma ray coupled transport equation (Equation 3.39) shows that to improve sensitivity and minimize detection limits we need to maximize neutron flux at the sample and minimize the sample to detector distance. The effect of the epithermal contribution is clear through the parameter $\lambda_n$, and hence a high thermal to fast flux ratio is also desirable. It is also desirable to minimize variability in the analyzer response due to formation perturbations. The detection limit (Section 2.1) is inversely proportional to the square root of the count rate under the peak, and hence a proper selection of construction materials is very important.

4.1 Design constraints

4.1.1 Counting time and source strength

Under field conditions, counting (irradiation) times are limited by the need to have fast sample turnaround time. The neutron flux (source strength) is also limited by shield weight and radiation dose rate constraints. The limitation of time and source strength dictates that
the best possible counting equipment (electronics, detector, etc.) be used to obtain the best possible resolution and counting rates. Further the above limitations make analyzer optimization an important design consideration.

4.1.2 Formation perturbation

Perturbations in the core formation are basically a design constraint of accuracy and repeatability or calibration. Sample properties are not under the control of the experimenter and hence have to be accounted for through calibration and supplementary measurements (e.g., measurement of porosity by gamma-gamma density gauges). Some of the more important disturbances which can seriously affect the tool response are:

1. Variation in the type and weight fractions of the major elements making up the sample matrix.

2. Changes in the physical characteristics of the sample, such as porosity, and hence density and salt content.

3. Presence of neutron poisons, which may depress the flux more than anticipated.

The above factors perturb the calibration by influencing the spatial and energy distribution of the neutron field and by altering the γ-ray attenuation factors. Inter-element interferences (e.g., strong Cl
interference observed in the spectra of seabed sediments, Senftle et al., 1976) can also lead to a loss in resolution and higher uncertainties in the results.

As epithermal neutron spectrum and associated capture gamma ray spectrum branching ratio data are either difficult to obtain or not available, it would be essential to keep the thermal to fast neutron flux ratio as high as reasonably possible and adjust the variable parameters of the design to minimize the perturbation in this ratio.

4.1.3 Detector damage

A germanium semiconductor detector loses its resolution substantially after being subjected to a fluence of $10^9$-$10^{10}$ neutrons/cm$^2$ (Section 3.9.1). This limits the gamma ray transmission factor by limiting the source to detector distance. Hence the detector placement is decided by the limiting neutron flux at the detector position and cannot be used as a variable parameter. This distance strongly depends on the source strength and moderator type.

4.1.4 Construction materials

The choice of construction materials mainly influences the inter-element interferences and background levels. This can limit the elements which can be detected and may also limit the concentration levels at which certain elements are detected. The choices are based on the criterion
of the interference parameter $S_T^2$. These have been calculated for some typical construction materials (Senftle et al., 1971). Elements with small $S_T$ values and without prominent peaks (small $S_{E_{\text{max}}}$ values) are the most desirable materials for construction.

Tin, magnesium, and aluminum are preferred for lower temperature applications and zirconium, niobium, or tantalum are better suited for high temperature applications. However if the best construction material also happens to be the element of interest in prospecting, the next best choice has to be considered. The above best choices are made on a unit weight basis. In actual construction the total amounts of the individual materials should be considered. Also to be considered are the mechanical and financial constraints and the neutron flux in the vicinity of the materials.

\[ S_T^2 = \sum_{3 \text{ MeV}}^{10 \text{ MeV}} S_E + \sum_{3 \text{ MeV}}^{10 \text{ MeV}} I_E \sigma/A \]

where

$S_E$ = sensitivity index for energy $E$.
$I_E$ = gamma photon yield, photons per 100 neutrons captured.
$\sigma^T$ = elemental thermal capture cross-section, barns
$A$ = atomic weight of element, grams.
4.2 Analyzer design optimization

4.2.1 Geometrical configuration and moderator selection

The moderator strongly influences the neutron flux and the neutron energy spectrum, and the choice of the experimental geometry greatly depends on the type of moderator used. The neutron flux is higher for the internal target geometry, whereas the gamma ray transmission factor is higher for the external sample geometry. The neutron flux and the gamma ray background at the detector are also somewhat affected by the choice of geometry. The source-sample-detector configuration may be chosen to minimize the neutron and background gamma ray fluxes at the detector. The source to sample distance also influences the neutron flux and the neutron energy spectrum at the sample position. It determines the extent of calibration necessary and the counting times needed for acceptable statistics.

4.2.1.1 Neutron moderation

The internal sample geometry is usually preferred for relatively small samples and the external target geometry for objects with large sizes and irregular shapes. As the sediment core is relatively large, the possibility of using the external target geometry was initially considered. The thermal neutron absorption in a hydrogenous moderator reduces the thermal flux drastically after about 10 cm (Greenwood, 1979), and hence the possibility of using a non-hydrogenous moderator
such as D$_2$O was considered for the external target geometry.

To compare the moderating properties of hydrogenous and non-hydrogenous moderators, H$_2$O and D$_2$O were chosen. A simple spherical geometry with a point source at the center was assumed to be adequate for this purpose. The source was surrounded with a 5 cm thick spherical lead shield which was considered essential for attenuating the gamma ray component of the $^{252}$Cf source. The aim of the study was to compare the behavior of the thermal and epithermal flux in the two types of moderators as a function of distance into the moderator.

The thermal neutron flux profiles for both moderators are shown in Figure 4.1. Figure 4.2 is a plot of the thermal to non-thermal neutron flux ratios. Figures 4.3 and 4.4 show the variation in the epithermal flux (characterized by the dimensionless parameter $\lambda_n(\bar{r})$) for the two moderators.

It is seen from Figure 4.1 that for distances less than about 14 cm from the source (including the 5 cm of lead shield around the source), the thermal flux is greater in H$_2$O, after which, due to the higher neutron absorption cross section of H$_2$O, the thermal neutron population in D$_2$O is higher.

Figures 4.3 and 4.4 show that in the case of H$_2$O rapid thermalization takes place and a reasonably well moderated neutron spectrum is achieved as close as 9 cm from the source. The same level of moderation is
Figure 4.1  Thermal neutron flux per source neutron in H$_2$O and D$_2$O as a function of distance from a $^{252}$Cf source. The source is surrounded by 5 cm of lead. The second Y-axis shows the ratio of the thermal neutron fluxes in the two moderators.

Figure 4.2  Thermal to non-thermal flux ratios for H$_2$O and D$_2$O as a function of distance from a $^{252}$Cf source. The source is surrounded by 5 cm of lead.
Figure 4.3  The variation of epithermal flux per unit thermal flux per unit lethargy interval at various distances into a water moderator from a $^{252}$Cf source. The source is surrounded by 5 cm of lead.

Figure 4.4  The variation of epithermal flux per unit thermal flux per unit lethargy interval at various distances into a heavy water moderator from a $^{252}$Cf source. The source is surrounded by 5 cm of lead.
achieved by D₂O at much greater distances. Furthermore Figure 4.2 shows that the non-thermal to thermal ratio for H₂O does not improve beyond about 15 cm from the source, at which point it has a value of about 4. Hence it seems that for relatively short distances, i.e., within distances for which a reasonable fraction of the source strength can be utilized, a thermal to non-thermal neutron flux ratio of about 4 is the best that can be achieved in water. Hence it seems that there will always be a significant contribution from the epithermal region of the neutron spectrum.

4.2.1.2 Porosity perturbation

To investigate the influence of porosity (and hence also salt content) on the thermal and epithermal flux profiles (characterized by $\lambda_n(\bar{r})$) in the sample, a planar geometry with a monodirectional source into a slab was chosen. The slab material composition was that of an "average rock" (Table 4.1). Calculations were done at three different porosities of 10, 50 and 90 percent.

As counting times are limited, the source-sample position becomes a compromise between maximizing the neutron flux and the thermal to fast ratio against minimizing its change due to perturbations. The source-sample distances determine the amount of moderator available between them and may be characterized by the neutron energy spectrum incident on the sample position. Hence to study the effect of the source-sample placement, in addition to the porosity effects, on the
neutron profiles in the sample material, calculations were done for two neutron energy spectra (Figure 4.5). Spectrum 1 and Spectrum 2 are idealized representations of a well moderated and an undermoderated neutron energy spectrum, respectively.

Table 4.1 Average rock composition at three different porosities.

<table>
<thead>
<tr>
<th>Element</th>
<th>Weight percent</th>
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</thead>
<tbody>
<tr>
<td></td>
<td>10% Porosity</td>
</tr>
<tr>
<td>Si</td>
<td>2.15E-01</td>
</tr>
<tr>
<td>Al</td>
<td>2.30E-02</td>
</tr>
<tr>
<td>Fe</td>
<td>1.00E-02</td>
</tr>
<tr>
<td>Na</td>
<td>7.48E-04</td>
</tr>
<tr>
<td>K</td>
<td>8.34E-03</td>
</tr>
<tr>
<td>Ca</td>
<td>1.21E-01</td>
</tr>
<tr>
<td>Mg</td>
<td>3.50E-03</td>
</tr>
<tr>
<td>Ba</td>
<td>5.17E-03</td>
</tr>
<tr>
<td>H</td>
<td>1.63E-02</td>
</tr>
<tr>
<td>O</td>
<td>5.62E-01</td>
</tr>
<tr>
<td>C</td>
<td>3.44E-02</td>
</tr>
<tr>
<td>Cl</td>
<td>8.34E-04</td>
</tr>
<tr>
<td>S</td>
<td>3.86E-05</td>
</tr>
<tr>
<td>Total</td>
<td>1.0000</td>
</tr>
<tr>
<td>Bulk density (g/cm³)</td>
<td>2.3795</td>
</tr>
</tbody>
</table>

Figures 4.6 and 4.7 are plots of the thermal flux profiles as a function of distance into the slab material for Spectrum 1 and Spectrum 2, respectively, for the three different porosities. Figures 4.8 to 4.13 are the corresponding plots for the epithermal component of the neutron flux. Figure 4.14 is the plot of the thermal to non-thermal ratio for Spectrum 1 and Spectrum 2.
Figure 4.5 Idealized representations of a well moderated (Spectrum 1) and an undermoderated (Spectrum 2) neutron energy spectrum.
Figures 4.6 and 4.7 show that the thermal flux is less perturbed by porosity changes for an undermoderated input neutron spectrum than for the well moderated case. Also it is worth noting that the thermal flux decreases appreciably within the sample.

A careful look at Figures 4.8 to 4.14 show the following trends common to both input neutron spectra:

1. The epithermal flux per unit lethargy interval normalized by the thermal flux (i.e., the parameter $\lambda_n(\tilde{F})$) is fairly constant with distance into the sample for the neutron energies below 0.1 MeV, but increases for higher energies.

2. The epithermal component above 0.1 MeV, and especially above 1 MeV, is much higher compared to the epithermal flux at lower energies. This difference increases with increasing porosity.

3. Both input neutron spectra are about equally affected by changes in porosity. An increase in porosity is seen to decrease the epithermal contribution. This can best be seen in Figure 4.14.

4. Figure 4.14 also shows that for both the input spectra the thermal to non-thermal neutron flux ratios start out higher for higher porosities and show a reversal in this trend with increasing distances into the sample material. This reversal
Figure 4.6 Thermal neutron flux as a function of distance into a slab material of an average rock composition at various porosities for a well moderated incident neutron energy spectrum.

Figure 4.7 Thermal neutron flux as a function of distance into a slab material of an average rock composition at various porosities for an undermoderated incident neutron energy spectrum.
Figure 4.8  The variation of epithermal flux per unit thermal flux per unit lethargy interval at various distances into a slab material of an average rock composition at 10% porosity for a well moderated incident neutron energy spectrum.

Figure 4.9  The variation of epithermal flux per unit thermal flux per unit lethargy interval at various distances into a slab material of an average rock composition at 10% porosity for an undermoderated incident neutron energy spectrum.
Figure 4.10 The variation of epithermal flux per unit thermal flux per unit lethargy interval at various distances into a slab material of an average rock composition at 50% porosity for a well moderated incident neutron energy spectrum.

Figure 4.11 The variation of epithermal flux per unit thermal flux per unit lethargy interval at various distances into a slab material of an average rock composition at 50% porosity for an undermoderated incident neutron energy spectrum.
Figure 4.12 The variation of epithermal flux per unit thermal flux per unit lethargy interval at various distances into a slab material of an average rock composition at 90% porosity for a well moderated incident neutron energy spectrum.

Figure 4.13 The variation of epithermal flux per unit thermal flux per unit lethargy interval at various distances into a slab material of an average rock composition at 90% porosity for an undermoderated incident neutron energy spectrum.
Figure 4.14 The variation of the thermal to non-thermal neutron flux ratios as a function of distance into a slab material of an average rock composition at various porosities for a well moderated and an undermoderated incident neutron energy spectrum.
takes place at shorter distances for Spectrum 1 than for Spectrum 2. This effect seems to be caused by the higher absorption of neutrons at higher porosities without much simultaneous thermalization of fast neutrons from the already well moderated input neutron spectrum.

5. The epithermal component is about twice as high for Spectrum 2 as compared to Spectrum 1.

The above analysis points to the fact that a detailed knowledge of the neutron flux profile is essential. However, as this cannot be determined experimentally in the sample, calibration by experiment and simulation is necessary for accurately predicting the concentration from the count rates.

Consideration of the source strength utilization and the thermal to fast ratio on the basis of equal source strength utilization shows that the internal target geometry with H$_2$O as the moderator would be the better choice. This choice was thought to be appropriate considering the regular shape of the core sample which would make it easy to design a convenient sample handling system.

Other favorable features of using H$_2$O as the moderator were:

1. Use of a lower source strength.
2. Reduced neutron shielding (and hence handling) requirements due to increased absorption of neutrons in H₂O and the use of a lower source strength.

3. A lower fast neutron flux at the detector. Hence the detector can be placed closer to the source, increasing the gamma ray transmission factor.

One drawback of using a hydrogenous moderator is the much higher gamma ray background from the hydrogen 2.223 MeV line, which makes detecting elements with signature lines below this energy difficult. However this was not considered a major problem as only the high energy (> 3 MeV) gamma ray spectrum was considered for this study.

On the basis of the above calculations the center of the core sample was placed at a distance of 12.5 cm from the source. Although perturbation effects could be further investigated for a range of source-sample distances in the actual geometry, this aspect was not undertaken due to time constraints. Moreover it seemed clear that sufficient thermalization is achieved within a few centimeters of a hydrogenous moderator, and considering that the calibration for a significant epithermal contribution is essential, the gain in the neutron flux by placing the sample close to the source outweighed the loss in accuracy that may be caused due to the somewhat larger perturbations that may be observed as a result of a harder input neutron spectrum.
4.3 Description of the analyzer

A cross sectional view of the irradiation and detection system as modeled into the geometry of MCNP is given in Figure 4.15. The container is a cast iron sphere. The cold finger and cryogen tank have not been modeled and hence have not been shown. The detector is shown as an active volume surrounded by a neutron and gamma ray shield.

The spherical container serves both as the irradiation and the storage system and provides the best neutron shielding per unit weight of the moderator. To get maximum utilization of the neutron moderator shield, the source was placed at the center. A 5 cm thick lead gamma ray shield was placed around the source to reduce the gamma component of the source.

The sample (seafloor sediment core) passes through a hollow composite tube through the container directly below the $^{252}$Cf source, with its central axis 12.5 cm from the center. The tube has a 10 cm air gap directly below the source for unhindered irradiation. The tube is made of 2 mm cadmium with 3 mm of lead. The cadmium reduces the thermal neutron flux seen by the sample sides (i.e., the sample portion outside the 10 cm long detection region). Lead is used in the composite tube mainly to reduce the gamma ray flux from the sample sides as seen by the detector. In an actual design sufficient thickness could be chosen to provide adequate structural strength to the tube (steel was not used due to inter-element interference considerations). Furthermore, the sample
Paraffin
Moderator
Iron Container
Composite Sleeve (Pb+Cd)
Pb Shield for Source Gamma Rays
Cf-252 Source
Sample core with Poly Casing
Collimator
Borated Paraffin Plug
Detector
Pb shielding for Detector

Figure 4.15 Cross sectional view of the irradiation and detection system as modeled in MCNP.
core handling system could be designed to rotate the core as it passes through the composite tube. This would help even out the effect of radial non-homogeneity in the distribution of the sample material.

Directly in line with the source but on the other side is a gamma ray collimator. This collimator is made from lead, bismuth and borated paraffin, and the design was taken from Greenwood, 1975. This design was seen to help reduce the neutron flux seen at the detector. The HPGe detector was placed on the far end of the collimator. The detector is surrounded by a 5 cm thick lead shield to reduce the $\gamma$-ray contributions from the sample sides, and a 10 cm borated paraffin neutron plug is placed inside the collimator at the detector end to reduce the neutron damage to the detector from neutrons streaming through the collimator.

The rest of the sphere is filled with the moderator. For modelling purposes the radius of the sphere was taken as 60 cm, which is sufficient for a 200 $\mu$g $^{252}$Cf source (Noakes et al., 1975). However the actual radius would depend upon the source strength. The source, sample, and detector are placed in a straight line, as this reduces the neutron flux in regions closer to the detector, hence reducing the background signal at the detector from the moderator. The fast neutron flux at the detector, placed at a source-detector distance of 70 cm, for a 1 mg $^{252}$Cf source in the present geometry, was found to be about 400 n/cm$^2$-s (from a MCNP calculation), which was considered acceptable.
5. SIMULATION OF THE PGNAA EXPERIMENT

The aim of the PGNAA experiment was to determine the sensitivities and detection limits achievable by the designed system for elements of economic interest. From among the elements of economic interest, the elements Mg, Al, Ti, Cr, Mn, Fe, Ni, and Cu were chosen for this study. This choice included hard to measure elements such as magnesium and the relatively easier to measure elements such as Ti and Mn. Mg was considered especially hard to measure due to its relatively low main signature photon energy of 3.917 MeV, low thermal neutron capture cross section of 0.603 barns, and strong interferences from the major elements Si and C (Table 5.1).

5.1 Modeling the geometry

The geometry of the system described in Section 4.3 was modeled using the combinatorial geometry capability of MCNP. The source was modeled as a point source, without its encapsulation. For the detector, only the active volume (covered with 0.5 mm of aluminum) was modeled, as a gallium (germanium cross sections were not available) crystal, with dimensions taken from the geometry of Jin et al., 1987. This seemed appropriate as the detector response function used by the above authors was used in this work, although the function is valid for any large volume HPGe detector. The cell to cell interface was assumed to be a perfect match.
The paraffin moderator/shield was modeled up to a thickness of 60 cm. This thickness, although not sufficient to bring the radiation dose below acceptable levels (Chapter 7), was considered sufficient for obtaining a fairly accurate representation of the neutron flux at the 10 cm central portion of the sample and of the construction material gamma ray background flux at the detector. The rest of the geometrical representation was quite accurate. A typical MCNP input file is shown in Appendix I.

5.2 Tallying the gamma ray spectra at the detector

5.2.1 MCNP computations

Three types of γ-ray spectra (all above 3 MeV) were computed using MCNP. These were the spectra from the construction material background (assumed unaffected by changes in sample porosities), the sample (sea bottom sediment core modeled as average rock) spectra at three different porosities, and the line γ spectra from a range of aluminum concentrations for each of the porosities.

The computations for the three kinds of spectra were done separately. This strategy was used throughout the simulation in the interest of computational efficiency. This was necessary as tallies from different space, energy and material phase space of the problem required a different set of variance reduction techniques or different parameters for the same variance reduction technique. Since, in many cases, the
improvement in a particular tally came at the expense of others, different tallies had to be obtained separately.

The construction material background was calculated in three different steps. In the first step contributions from thermal neutron capture gamma rays from carbon were collected. In the second step the fast neutron inelastic scattering contribution from carbon was tallied. In both cases the photon generation biasing feature of MCNP was used, and hence only the data in the peaks were considered reliable. In the third step contributions from the rest of the construction materials were collected in 10 keV energy bins. This was done by selectively turning off photon generation in the moderator region. Contribution from hydrogen was ignored as it was below the energy of interest. The three spectra were then added to get the total construction material background which is shown in Figure 5.1. Also shown in Figure 5.1 are the associated errors.

To get the spectra from the sample, the aluminum concentration in the average rock was replaced by silicon. This was done because line gamma ray data were not available for any other element of interest in the ENDF/B-V cross section. This also enabled a comparison between MCNP and ISOSHLD-II computations (Table 6.3).

Only the spectra from the 10 cm central irradiated portion of the sample were collected. It was assumed that the shielding of the detector and the collimator would be sufficient to make the contributions from the
Figure 5.1 Construction material gamma ray background flux at the detector with associated errors as obtained from MCNP simulation. The values are per source neutron per unit energy interval.
rest of the sample negligible. This assumption is not quite accurate as a test case showed significant contribution from the sides of the sample (see Section 6.3). This limitation of the simulation had to be tolerated in the interest of computational efficiency.

A coarse energy bin structure (10 keV energy bins) was used for the scattered contributions, and the fluxes for the peak energies were scored in relatively narrow 2 keV energy bins. The peak energies were taken from the prompt gamma ray cross section data files associated with MCNP and from Lone et al., 1981. The spectra from the 10 cm central portion of the sample at the three different porosities are shown in Figures 5.2 to 5.4.

A peculiar feature of these spectra was the high contribution in the energy range of 7.60 to 7.65 MeV. As the iron doublet peaks are at 7.631 and 7.645 MeV, the high contribution in the two 10 keV energy bins from 7.60 to 7.62 MeV could not be explained. No significant gamma rays from the elements of the average rock core sample could be found in this energy range in the data of Lone et al., 1981. The data were, however, used as such. The effect of this peculiarity is evident in the detector response spectra around the iron doublet energies (Figure 5.19). Similar broadening of the Fe peak can also be seen for the peak energy of 9.299 MeV in the sample background spectra at 10 percent porosity (Figure 5.2).

Gamma ray line intensities from aluminum were tallied at the detector
Figure 5.2  Gamma ray flux at the detector from the sample material at 10% porosity, as obtained from MCNP simulation. The values are per source neutron per unit energy interval.
Figure 5.3 Gamma ray flux at the detector from the sample material at 50% porosity, as obtained from MCNP simulation. The values are per source neutron per unit energy interval.
Figure 5.4  Gamma ray flux at the detector from the sample material at 90% porosity, as obtained from MCNP simulation. The values are per source neutron per unit energy interval.
for four gamma ray energies (4.261, 6.200, 7.695 and 7.726 MeV). The concentration was varied from 0.2 w% to 2 w% for each porosity. Again only the contribution from the 10 cm central irradiated portion of the sample was collected. The gamma ray line flux at the detector for the 7.726 MeV gamma ray per source neutron and normalized to unit weight of aluminum in the sample, is shown in Figure 5.5. It is seen that the response per unit weight of aluminum increases with an increase in porosity. The same trend is also seen for the other gamma rays.

To see the variation of the response per unit weight of an element with changes in porosity, the 7.726 MeV gamma ray response at the three different porosities was plotted as a single series and is shown in Figure 5.6. Figure 5.6 shows that for the present geometry, the increase in response is within about 10 percent for a wide porosity range of 10 to 90 percent porosity. Hence minor changes in porosity are not expected to affect the calibration appreciably. However, to better see the effect of porosity, the response at still higher concentrations will have to be studied. This would be essential for calibration purposes, but was not done as the main objective of this study was to obtain estimates of the detection limits.

5.2.2 ISOSHLD-II computations

The line gamma ray data for the chosen elements other than aluminum were not available in the ENDF/B-V data libraries of MCNP. Hence the uncollided gamma ray flux from the selected lines of elements of
Figure 5.5 7.726 MeV aluminum line gamma flux at the detector from a range of aluminum concentrations in the sample material at three different porosities, normalized to unit weight of aluminum, as obtained from MCNP simulation. The values are per source neutron per unit energy interval.
Figure 5.6 The ratio of the 7.726 MeV aluminum line gamma flux at the detector for each concentration and porosity to the average flux from all concentrations and porosities.
interest were tallied at the detector using ISOSHLD-II (Rittmann, 1987), a point kernel code.

To use ISOSHLD-II for high energy gamma ray computations, the data library associated with the code was modified to include three new materials (i.e., the sample at three different porosities), attenuation coefficients at higher energies, and the buildup factor was forced to unity. Further, to obtain the output in units of photons/cm²-s instead of exposure rates, the photon flux to exposure rate conversion factors in the program were all modified to unity.

A uniform volume distributed gamma ray source term was used, since an exponential source could not be coded into one of the standard geometries available in ISOSHLD-II. The gamma ray source term in the sample for a 1 mg $^{252}$Cf source for each of the selected gamma rays was calculated from known gamma ray production data and the thermal flux in the sample (obtained from MCNP computations). Based on the approximations being used in the ISOSHLD-II computations, only the response at a single concentration (1 w%) was calculated for each porosity.

The neutron flux profiles in the sample at the three different porosities are shown in Figures 5.7 to 5.11. Figure 5.7 shows the variation of the thermal flux with distance into the sample. It is seen that the thermal flux falls by a factor of about 2 for 90 percent sample porosity and somewhat less for lower porosities. Also the thermal flux
Figure 5.7  Thermal neutron flux as a function of distance into the sample core at various porosities. Values are per source neutron.

Figure 5.8  Thermal to non-thermal neutron flux ratios as a function of distance into the sample core at various porosities.
increases with porosity and is highest for 50 percent sample porosity. Figure 5.8 shows the variation in the thermal to non-thermal neutron flux ratios as a function of distance into the sample. This ratio shows the same trend as the thermal flux in the sample, i.e., the ratio increases with an increase in porosity. Also, the ratio increases with distances into the sample, indicating the contribution of the sample to neutron thermalization. However, at high porosities, due to higher thermal neutron absorption, the thermal to non-thermal ratio increases less rapidly with distance into the sample and falls below the ratio for the 50 percent sample porosity.

Figures 5.9 to 5.11 show the non-thermal neutron flux profiles. The trend is the same as that observed for the porosity perturbation studies of Chapter 4, with the maximum contribution to the non-thermal flux coming from neutron energies greater than 1 MeV.

Table 5.1 shows the gamma ray lines selected for the analysis along with their production data and major interferences. The selection of the gamma ray lines was based on their sensitivities (Section 2.1). Additionally if more than one line was available for analysis, then the most interference-free line was selected. Only lines above 3 MeV were considered. This compromise was evident in the case of Mn and Ti, where for Mn the most interference-free line of 7.058 MeV was selected over the stronger line of 7.244 MeV, and for Ti the lower intensity line at 6.760 MeV was chosen over the stronger 6.418 MeV line due to the close interference from the Ca line at 6.420 MeV. MCNP ENDF/B-V data did not
Figure 5.9 The variation of epithermal flux per unit thermal flux per unit lethargy interval at various distances into the core sample at 10% porosity.

Figure 5.10 The variation of epithermal flux per unit thermal flux per unit lethargy interval at various distances into the core sample at 50% porosity.
show a chlorine line at 7.790 MeV (as shown in the data of Lone et al., 1981) which, if present, could interfere with the 6.760 MeV peak of Ti through its double escape peak.

For Fe the higher intensity line at 7.645 MeV of the doublet peak was chosen, although the 9.299 MeV Fe line could prove a better choice due to the much lower gamma ray background expected at higher energies.

The uncollided flux from ISOSHLD-II computations for a 1 mg $^{252}$Cf source per gram of an element and considering only the thermal neutron flux in the sample are shown in Table 5.2. The fluxes from Ti, Mn and Ni are the highest followed by about half as much for Cu, Cr and Fe. Al and Mn fluxes are about an order of magnitude less than the other elements.
Table 5.1  Selected gamma ray energies and their associated gamma ray production data.

<table>
<thead>
<tr>
<th>Element</th>
<th>cross-sections (barns)</th>
<th>$\gamma$-Ray parameters for thermal neutron capture</th>
<th>Major Interferences</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>$\sigma$</td>
<td>$I_7$</td>
<td>$E_7$ (MeV)</td>
</tr>
<tr>
<td>Mg</td>
<td>0.063</td>
<td>0.038</td>
<td>3.917</td>
</tr>
<tr>
<td>Mn</td>
<td>13.3</td>
<td>14</td>
<td>7.058</td>
</tr>
<tr>
<td>Ti</td>
<td>6.1</td>
<td>2.9</td>
<td>6.760</td>
</tr>
<tr>
<td>Ni</td>
<td>4.5</td>
<td>2.2</td>
<td>9.00</td>
</tr>
<tr>
<td>Cu</td>
<td>3.8</td>
<td>4.1</td>
<td>7.915</td>
</tr>
<tr>
<td>Cr</td>
<td>3.1</td>
<td>1.6</td>
<td>8.884</td>
</tr>
<tr>
<td>Fe</td>
<td>2.56</td>
<td>1.4</td>
<td>7.645</td>
</tr>
<tr>
<td>Al</td>
<td>0.233</td>
<td>0.17</td>
<td>7.726</td>
</tr>
</tbody>
</table>

Table 5.2  Uncollided flux (for 1 w% element) from ISOSHLDFII computations normalized to unit weight, i.e., per gram of element in sample.

<table>
<thead>
<tr>
<th>Element</th>
<th>$E_7$ (MeV)</th>
<th>Uncollided flux at detector position for a 1 mg $^{252}$Cf source. (Photons/cm$^2$-s)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>10% Porosity</td>
</tr>
<tr>
<td>Mg</td>
<td>3.917</td>
<td>0.025</td>
</tr>
<tr>
<td>Ti</td>
<td>6.760</td>
<td>0.705</td>
</tr>
<tr>
<td>Mn</td>
<td>7.058</td>
<td>0.618</td>
</tr>
<tr>
<td>Fe</td>
<td>7.645</td>
<td>0.264</td>
</tr>
<tr>
<td>Al</td>
<td>7.726</td>
<td>0.055</td>
</tr>
<tr>
<td>Cu</td>
<td>7.915</td>
<td>0.435</td>
</tr>
<tr>
<td>Cr</td>
<td>8.884</td>
<td>0.387</td>
</tr>
<tr>
<td>Ni</td>
<td>9.00</td>
<td>0.697</td>
</tr>
</tbody>
</table>
As only thermal neutron capture is considered, the variation in the gamma ray flux with porosity follows the variation in the thermal flux with porosity. It is also noted that all the flux values are less than 1 photon/cm²-s.

5.3 Generating the detector response

The gamma ray spectra from MCNP were processed to distribute the weight from the broader 10 keV energy bins into 2 keV energy channels. The weight in an energy bin $E + \Delta E$ corresponded to the intensity of the gamma rays of energy $E + (\Delta E/2)$ incident on the detector. The response of the detector to each incident energy was calculated down to 3 MeV, in 2 keV energy channels, for the entire incident $\gamma$-ray spectra. The response for each incident energy added gave the total detector response.

The gamma ray flux incident on the front face of the detector was multiplied by the probability of any kind of interaction within the active volume. This multiplication factor was calculated as $1 - e^{-\mu x}$, where $\mu$ was the linear attenuation coefficient for germanium at the appropriate energy, and $x$ was the characteristic length of the detector, taken as the axial length of the detector, which is 6.15 cm. This factor is fairly constant over the energy range used (3 to 10 MeV) and is about 0.64. This total interaction probability is considered somewhat high, even for the 150 cm³ HPGe detector being used here.
The detector response spectra for the construction material background and for the sample matrix background at the three different porosities are shown in Figures 5.12 to 5.15. The complex nature of the prompt gamma ray spectra with many full energy peaks and their associated single and double escape peaks is evident from Figures 5.12 to 5.15. The background is seen to increase with porosity, which is mainly caused by an increase in the chlorine concentration. The magnitude of the background does not start falling until above the lead peak energy of 7.368 MeV. An interesting feature of the spectra is that the gamma ray background from the sample reduces with porosity beyond the 8.577 MeV peak of chlorine.

Figures 5.16 to 5.23 show the gamma ray background (including construction material contribution) under the peak energies being used for the analysis. The reduction in the background beyond the 8.577 MeV peak of chlorine is evident from Figures 5.22 and 5.23, which are the background spectra under the chromium and titanium peak energies of 8.884 and 9.000 MeV.

The ISOSHLD-II and MCNP data generated in this simulation were used to derive the sensitivities and detection limits for the elements shown in Table 5.1. The results are presented in the next chapter.
Figure 5.12 Construction material gamma ray background detector response spectrum.
Figure 5.13 Detector response gamma ray background spectrum for the sample material at 10% porosity.
Figure 5.14 Detector response gamma ray background spectrum for the sample material at 50% porosity.
Figure 5.15 Detector response gamma ray background spectrum for the sample material at 90% porosity.
Figure 5.16 Detector response gamma ray background spectra for three different porosities around the magnesium peak energy of 3.917 MeV.
Figure 5.17 Detector response gamma ray background spectra for three different porosities around the titanium peak energy of 6.760 MeV.
Figure 5.18 Detector response gamma ray background spectra for three different porosities around the manganese peak energy of 7.058 MeV.
Figure 5.19 Detector response gamma ray background spectra for three different porosities around the iron doublet at 7.631 and 7.645 MeV.
Figure 5.20 Detector response gamma ray background spectra for three different porosities around the aluminum peak energy of 7.726 MeV.
Gamma Ray Energy (MeV)

Cu peak energy of 7.915 MeV

Si, SEP of 8.473 MeV

Figure 5.21 Detector response gamma ray background spectra for three different porosities around the copper peak energy of 7.915 MeV.
Figure 5.22 Detector response gamma ray background spectra for three different porosities around the chromium peak energy of 8.884 MeV.
Figure 5.23 Detector response gamma ray background spectra for three different porosities around the nickel peak energy of 9.000 MeV.
6. RESULTS AND DISCUSSION

6.1 Results

Total count rates derived from Table 5.2 data are shown in Table 6.1, the sensitivity table. The count rates were obtained as the product of the uncollided flux (from ISOSHLD-II), the total interaction efficiency (Section 5.3), the peak intrinsic efficiency, and the surface area of the detector face. The photopeak intrinsic efficiency of the detector was obtained by using the peak to total ratio of the detector response function for the gamma ray energy under consideration.

Table 6.1 Count rates per gram of element at three different porosities.

<table>
<thead>
<tr>
<th>Element</th>
<th>$E_γ$ (MeV)</th>
<th>Counts/g-s for a 1 mg $^{252}$Cf source.</th>
<th>10% Porosity</th>
<th>50% Porosity</th>
<th>90% Porosity</th>
</tr>
</thead>
<tbody>
<tr>
<td>Mg</td>
<td>3.917</td>
<td>0.022</td>
<td>0.027</td>
<td>0.026</td>
<td></td>
</tr>
<tr>
<td>Ti</td>
<td>6.760</td>
<td>0.553</td>
<td>0.674</td>
<td>0.648</td>
<td></td>
</tr>
<tr>
<td>Mn</td>
<td>7.058</td>
<td>0.515</td>
<td>0.628</td>
<td>0.601</td>
<td></td>
</tr>
<tr>
<td>Fe</td>
<td>7.645</td>
<td>0.248</td>
<td>0.301</td>
<td>0.289</td>
<td></td>
</tr>
<tr>
<td>Al</td>
<td>7.726</td>
<td>0.053</td>
<td>0.065</td>
<td>0.062</td>
<td></td>
</tr>
<tr>
<td>Cu</td>
<td>7.915</td>
<td>0.432</td>
<td>0.525</td>
<td>0.504</td>
<td></td>
</tr>
<tr>
<td>Cr</td>
<td>8.884</td>
<td>0.454</td>
<td>0.549</td>
<td>0.529</td>
<td></td>
</tr>
<tr>
<td>Ni</td>
<td>9.00</td>
<td>0.839</td>
<td>1.019</td>
<td>0.977</td>
<td></td>
</tr>
</tbody>
</table>

Table 6.2 shows the minimum detection limits in weight fraction of element in the sample core. The minimum detection limits are calculated according to the method described in Section 2.1.
Table 6.2 Minimum Detection limits (interference free) in weight fraction of core sample for a 10 minute irradiation time.

<table>
<thead>
<tr>
<th>Element</th>
<th>$E_{\gamma}$ (MeV)</th>
<th>$C_{MDL}$ in weight fraction for a 1 mg $^{252}$Cf source</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>10% Porosity</td>
</tr>
<tr>
<td>Mg</td>
<td>3.917</td>
<td>0.545</td>
</tr>
<tr>
<td>Ti</td>
<td>6.760</td>
<td>0.020</td>
</tr>
<tr>
<td>Mn</td>
<td>7.058</td>
<td>0.023</td>
</tr>
<tr>
<td>Fe</td>
<td>7.645</td>
<td>0.010</td>
</tr>
<tr>
<td>Al</td>
<td>7.726</td>
<td>0.043</td>
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<tr>
<td>Cu</td>
<td>7.915</td>
<td>0.004</td>
</tr>
<tr>
<td>Cr</td>
<td>8.884</td>
<td>0.003</td>
</tr>
<tr>
<td>Ni</td>
<td>9.000</td>
<td>0.002</td>
</tr>
</tbody>
</table>

6.2 Limitations of the simulation

The above analysis is subject to the following limitations:

1. The statistical errors in the $\gamma$-ray spectra obtained at the detector are quite significant for some energy bins between prominent peaks. As an example, the gamma ray spectra from the sample matrix at 10% porosity is shown in Figures 6.1 (a) and 6.1 (b). These errors had to be tolerated as the work was done on a 80386/25 MHz personal computer, and sufficient computation power was not available to get good statistics for low probability events. This problem was more severe due to the wide range of both the gamma ray energies and their associated production cross sections. This made the effective use of the otherwise powerful
Figure 6.1 (a) Gamma ray flux at the detector between 3 and 5 MeV from the sample matrix at 10% porosity with the associated errors, as obtained from MCNP simulation.
Figure 6.1 (b) Gamma ray flux at the detector between 5 and 10 MeV from the sample matrix at 10% porosity with the associated errors, as obtained from MCNP simulation.
variance reduction technique, the weight window, difficult.

Another difficulty in effective computation of the gamma ray spectrum in MCNP was the availability of only one problem-wide photon production weight (PWT) card, the weight limits on which have to match the lower weight window bounds for the energy dependent photon weight window with the lowest weight bounds. This results in the generation of many gamma rays (a maximum of 10 per collision) for the events which already have a high probability. Additionally, the track termination card associated with the DXTRAN sphere (DD card) can be set only for a single problem-wide weight cut off value. If this value is lowered to accommodate the low probability events, too much weight fluctuation is introduced in the tallies, degrading the FOM.

Increasing the weight window lower bounds, the values associated with the PWT card, and the track termination criterion on the DD card does increase the FOM (by controlling the weight fluctuations), but the high energy gamma ray spectrum (above 6 MeV) is not sampled adequately, again necessitating longer running times.

2. The detector response function used is valid for energies between 60 keV and 6.2 MeV. It is seen that it increasingly overestimates the response at higher energies. At 10 MeV
the integral of the response function over the complete 
energy range rises to about 2.5 instead of 1.0 as would be 
expected for a function expressed as a probability density 
distribution. This suggests that the background calculated 
from this function will be somewhat higher compared to 
actual measurements if energies higher than 6.2 MeV are 
present in the incident spectrum. However, as this will 
make the results conservative, it was used without change.

3. The peak intrinsic efficiency calculated using the above 
detector response function for the selected gamma rays is shown 
in Figure 6.2. It is seen that this efficiency increases for 
energies above about 6 MeV. This behavior is attributed to the 
energy dependent normalization parameter used in the full 
energy peak function of the total response function. However, 
as the background is calculated using the same detector 
response function, the intrinsic efficiency is also used from 
the same function. This limitation is expected to give 
detection limits which are somewhat lower than would be 
expected in an actual experiment for energies higher than 6 
MeV, and higher than expected for energies lower than 6 MeV.

4. MCNP calculations with a gamma ray energy of 8.5 MeV showed 
that the contributions from the sides of the sample was about 
30% of the total contribution from the full sample (120 cm 
long). This contribution could easily be turned off in MCNP
Figure 6.2 Peak intrinsic efficiency curve for the detector response function of Jin et al., 1986.
computations, but more effective shielding may have to be used to reduce the contribution from the sample sides in the final design. As this contribution cannot be fully eliminated, it will also tend to degrade the detection limits.

6.3 Discussion of results

From Table 6.2, it is seen that the elements Ni, Cr and Cu can be detected at concentrations of less than 1 weight percent for a 10 minute irradiation time. Also the detection limits do not change much with porosity for these elements. This seems to be due to the much lower background at the higher energies, i.e., above the 7.368 MeV line from lead and especially above the 8.577 MeV line from chlorine. Furthermore, Fe, Mn and Ti also have fairly good detection limits, but to get detection at below the 1 weight percent level, longer counting times may be required. The possibility of measuring aluminum at the 1 weight percent level is marginal and will depend on the time available for counting and the reduction that can be achieved in the gamma ray background by a better shield design and by Compton suppression. Measuring magnesium which has the poorest response of all the selected elements would be even harder due to the presence of interferences from the major elements carbon and silicon.

A comparison of the above analysis with previous reactor data (Table 2.1) for aluminum (aluminum data used in this comparison is for a sample porosity of 50 percent) shows that the sensitivity achieved by the
reactor facility is about 20 times higher. The corresponding detection
limit in coal (Lindstrom and Anderson, 1985; counting time was not
reported and is assumed to be about 16 hours) is reported as 0.86 mg/g
(0.086 w%). The detection limit from this isotopic facility is of the
order of 10 w% (for a 10 minute irradiation). However the detection
limit on equal irradiation time basis are comparable. Further
improvement in the detection limits may be achieved with a different
design of the shielding arrangement and materials (to reduce the high
background from lead) and with Compton suppression.

Table 6.3 shows a comparison of the sensitivity and detection limits
obtained from ISOSHLD-II and MCNP computations for aluminum.

Table 6.3  Comparison of ISOSHLD-II and MCNP sensitivity and detection
limits for aluminum.

<table>
<thead>
<tr>
<th>Porosity</th>
<th>ISOSHLD-II calculations</th>
<th>MCNP calculations</th>
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</thead>
<tbody>
<tr>
<td></td>
<td>Sensitivity(^1) (counts/g-s)</td>
<td>(C_{\text{MDL}}) (^2) (w%)</td>
</tr>
<tr>
<td>10%</td>
<td>0.053</td>
<td>0.043</td>
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<tr>
<td>50%</td>
<td>0.065</td>
<td>0.054</td>
</tr>
<tr>
<td>90%</td>
<td>0.062</td>
<td>0.118</td>
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</tbody>
</table>

\(^1\)Sensitivities are per gram of aluminum in the 10 cm irradiated
portion of the sample.

\(^2\)The minimum detection limits are in grams of aluminum per gram of
sample and are for a counting time of 10 minutes. The \(C_{\text{MDL}}\) goes
down by the square root of the counting time.

The sensitivities obtained from ISOSHLD-II computations are higher by 15
to 35 percent, depending on porosity, when compared to the sensitivities
obtained from MCNP. The trend in the sensitivities, however, shows a
reasonable similarity. As various approximations were made in the
gometry and gamma ray transport in ISOSHLD-II computations, the results
obtained from MCNP are considered more representative. However, the
above results still have to be verified against experiment.

It is noted that the sensitivity is not very dependent on porosity, but
the associated detection limits increase with an increase in porosity
due to an increase in the background under the peak. For high energies,
i.e., above the 8.577 MeV gamma ray line of chlorine, the background
decreases with porosity and improves the detection limits.
7. HEALTH PHYSICS

Radiation protection considerations form an important part of any study connected with the use of radiation or radioisotopes. The use of a 1 mg $^{252}\text{Cf}$ source (0.535 Ci) requires that proper radiation protection measures be taken for limiting the radiation exposures from the source or any other activated material during their transportation, handling, use, storage and disposal.

7.1 Transportation

The regulations governing the packaging and transportation of radioactive material are given in the U.S. Code of Federal Regulations, Title 49, Part 173. The $^{252}\text{Cf}$ source is an encapsulated solid and is classified as a special form radioactive material for shipment purposes. Its specific activity is $5.4 \times 10^2$ Ci/g and the $A_4$ limit (activity limit for special form radioactive material for a type A package) is 2.7 Ci. Hence a 1 mg source (about 0.54 Ci) can be transported in a Type A package. Relevant regulations for the design, testing, approval and transportation of the package should be consulted.

7.2 Shield design for the $^{252}\text{Cf}$ source

The shield used in the present design of the analyzer is a composite of lead, paraffin and the iron. The 1 mg $^{252}\text{Cf}$ source is immediately surrounded by 5 cm of lead, 100 cm thickness of paraffin and then 5 cm
of iron. The paraffin also serves as the moderator, and the iron spherical shell containing the source and paraffin moderator also provides some shielding. This container also serves as the storage container. The above thicknesses are based on $^{252}$Cf prompt gamma ray production data as given in USAEC, 1971.

The gamma ray component of the dose rate at the surface of the container for the above shield design was about 0.45 mrem/hr, which was obtained from ISOSHLD-II computations. The neutron dose rates for paraffin thicknesses of about 100 cm are negligible (USAEC, 1971). For a 40 hour week and 13 weeks per quarter, the above shielding is enough to bring the dose rates at the surface of the container to well below the limit of occupational dose rate i.e., 1.25, rem per quarter (10 CFR Part 20).

It is advantageous to optimize the shield thicknesses due to the sharp rise in shield weight with increasing thickness. Further reduction in the thickness of the moderator may be made by the addition of 5 to 20 mg/cm$^3$ of natural boron to the paraffin shield (USAEC, 1971). However, boron should be added only to regions not significantly affecting the thermal neutron population at the sample core. More detailed calculations would be required to determine the optimal shield design thickness.

7.3 Licensing

$^{252}$Cf is classified as a byproduct material. Licensing requirements for
byproduct material are given in 10 CFR Parts 30 to 35 and 39. Although an NRC agreement state can license the use of byproduct material, the byproduct material comes under the jurisdiction of the NRC if it will be used in offshore waters (10 CFR Part 150). It is worth noting that if the license is for activity within the agreement state land boundaries and out to 3 miles offshore, the process of reciprocity grants a general license to conduct the same activity in offshore waters. Reciprocity is granted by the submission of Form-241 to the NRC along with a copy of the state license and providing the departure and return dates. As $^{252}\text{Cf}$ is a transuranic material, NRC agreement states no longer have the jurisdiction over its disposal. The $^{252}\text{Cf}$ source cannot be disposed of at sea.

7.4 Radiation protection during operation

A properly designed health physics program is essential and required of any facility using licensed radioactive material. This program is aimed at minimizing the radiation exposure to personnel and the spread of radioactive material. This philosophy is called "as low as reasonably achievable" (ALARA) and requires that doses be kept as far below the regulatory limits as possible. Briefly described, a typical radiation protection program for a facility using a sealed californium source will contain at least the following aspects:
7.4.1 Administrative structure and controls

For a facility using radiation or radioisotopes, technical support and supervision for the radiation protection aspects of the operations of the facility has to be provided. The requirement of trained personnel to provide the technical support and supervision will depend upon the type of facility and the nature of its operations. Administrative controls are used to ensure that people are aware of the hazards of working with radiation and the controls necessary to minimize their exposure to such hazards.

7.4.1.1 Notification

Notices, instructions and reports, and other mandatory information with which the workers have to be provided are specified in 10 CFR Part 19. This information ensures that the worker is aware of the license conditions for the facility, the operating procedures applicable to the facility, any violations of proper radiation protection practices, his radiation exposure, and his responsibilities and rights concerning working with radiation.

7.4.1.2 Orientation and training

The orientation and training of all personnel working with or around radiation is required by law. Such personnel are required to have a through understanding of the risk involved in working with radiation,
the proper operating procedures, radiation protection methods, and the equipment used for monitoring and protecting against radiation exposure. Hence an orientation and training program has to be developed and approved for any facility using radiation or radioisotopes. Periodic testing of worker knowledge and training is also included in a radiation protection program.

Operating and emergency procedures should be developed to minimize the doses received. These should include clear instructions for the actions required if high levels of radiation are detected or in case of an accident while handling the source.

### 7.4.1.3 Posting of signs and labels

Areas where there are radiological hazards must be posted with certain standard signs. 10 CFR Part 20 specifies the colors, wording, proportions and meaning of the signs. Each area or room in which the $^{252}$Cf source is used or stored should be posted with the "CAUTION RADIOACTIVE MATERIAL" sign. As the radiation dose rate from the $^{252}$Cf source at the surface of its container is less than 5 mrem/hr, a "CAUTION RADIATION AREA" sign is not needed. Proper labeling of the $^{252}$Cf source container which shows the type of isotope, activity, radiation level with distance, etc., should be provided. Furthermore, the source should be permanently marked for identification and inventory purposes.
7.4.1.4 Access control

Only authorized personnel should be provided with access keys to the $^{252}\text{Cf}$ facility, which should otherwise be locked. Any other person wishing to use the facility has to go through proper orientation and training in the procedures of the facility. Based on this training, work permits may be granted.

To provide additional physical security for the $^{252}\text{Cf}$ source, the source access port of the container should be kept locked.

7.4.2 Personnel monitoring

The main purpose of personnel monitoring is to obtain information about the radiation exposure of the individual worker. The type of radiation monitors worn should be compatible with the type and energy of the radiation being measured. Film badges and TLD dosimeters may be used for gamma radiation monitoring, and the use of track etch plastic dosimeters is suggested for neutron dose rate measurements, due to their uniform energy response.

7.4.3 Area monitoring

Routine surveys of the radiation levels around the isotopic source irradiator should be conducted. These surveys are helpful in showing any cracks or other leakage routes that might develop in the shield,
resulting in "hot spots".

7.4.4 Leak testing of the source

To prevent the spread of radioactive material, the sealed source should be checked for surface contamination or leakage prior to its use and then at periodic intervals. To check for contamination the surface of the source should be wiped with a solvent wetted filter paper or cotton swab (preferably done remotely with the help of tongs) and the alpha and beta activity measured. If the activity is less than 0.005 μCi (alpha) or less than 0.05 μCi (beta), the source may be considered free of contamination.

7.4.5 Equipment requirements

Instruments for measuring dose rates in and around the facility as well as for personnel monitoring will be required. At the very minimum these instruments would include several GM counters for the detection of the presence of radioactivity, cutie pie exposure rate meters for gamma (and, if needed, beta) dose rates, a "Bonner sphere" (with a $^{10}\text{BF}_3$ or a $^{6}\text{LiI}$ crystal neutron detector) neutron dose rate meter, direct reading pocket dosimeters, and gas flow proportional counters for alpha and beta counting of the smear samples.

An inventory of protective clothing, such as disposable gloves, lab coats and lead glass glasses, should be maintained. As a single cruise
could take months, calibration sources and equipment will also be required on board ship to periodically check the calibration of the other instruments. Furthermore, depending on the length of the cruise, readout equipment for the dosimeters used may be required.

7.4.6 Handling and storage of irradiated cores

Calculations show that for an irradiation period of 1 hr, the specific activity at end of bombardment (EOB) for the core sample of 10% porosity is about 980 pCi and for the core sample with 90% porosity about 660 pCi. After a week the activities reduce to about 40 fCi and 190 fCi, respectively.

As a first approximation, all the activity was assumed to be from $^{24}\text{Na}$. A length of one meter of irradiated core was considered sufficient to estimate the dose rates to personnel handling the cores. The doses at EOB from the higher activity 10% porosity core were calculated at the surface and at a distance of one meter from the surface, in both cases for points equidistant from the ends of the cylindrical sample core. The surface dose rate was 1.27 mrem/hr and at one meter the dose rate was 15.3 μrem/hr. These dose rates are considered conservative due to the assumptions and parameters chosen for the calculations.

Personal doses from the irradiated core sample can be kept negligible with proper implementation of the standard radiation protection principles of maximizing distance, minimizing time for handling, using
proper handling gear, and providing adequate shielding.

Due to the dose rates on the surface of the irradiated core, at EOB being about 1 mrem/hr, the possibility of storing the irradiated cores in an unrestricted environment should be considered. However, the decision will depend upon calculations or measurements, taking into consideration the operating and handling procedures, of the dose and dose rates that could be received to personnel near such storage areas. If such calculations show, to the satisfaction of the NRC, that the dose and dose rates do not exceed the limits specified in 10 CFR part 20.105 (i.e., total dose less than 0.5 rem per calendar year, a dose rate less than 2 mrem/hr, and a total dose less than 100 mrems for a person present in the storage area continuously for 7 days), then the irradiated cores could be stored in an unrestricted area.

In addition to the above, a radiation protection program will have to be approved by the NRC as having met all the requirements of the license agreement, including that of maintaining records, filing periodic reports, and meeting the ALARA requirement.
8. CONCLUSIONS AND RECOMMENDATIONS

8.1 Conclusions

The results tabulated in Section 6.1 indicate that the present method and design could prove suitable for the shipboard analysis of ocean cores. This conclusion is, however, subject to the limitations stated in Section 6.3. More detail in the gamma ray spectra generation and their verification by experimental measurements are necessary before the analyzer designed in this work could be considered for the PGNAA of seafloor cores.

8.2 Recommendations for future work

It is obvious from the limitations of the present analysis, as discussed in Section 6.3, that improvement in many areas can be made. More design studies are needed regarding optimizing the source sample placement and the optimum source strength. Due to the need for a reduction in the gamma ray background in general, and from lead in particular, design studies into a better shield design could prove helpful.

Experimentation for verification and normalization of simulation results is considered essential. Also further simulation with different sample types, neutron poisons such as samarium, and with more variability of the chlorine concentration for each sample porosity, needs to be done for generating a detailed calibration curve of counts versus
concentration in the sample. Generation of interference parameters by simulation and their verification by an experiment would also improve the detection of hard to measure elements such as magnesium.

The use of a faster computer or a faster specific purpose Monte Carlo code would be essential for accuracy in the generation of the gamma ray background. Also extending the range of the detector response function to cover gamma ray energies up to 10 MeV would make the results more reliable.
REFERENCES


APPENDIX
APPENDIX I

A Typical MCNP Input File

PROB.ALO1.IN ; AS MODIFIED ON ---> 02/21/91

To obtain aluminum line gamma flux from central sample portion

------------- Cell Descriptions -------------

1 0 -160 $ SPHERICAL SOURCE
2 2 -11.34 160 (-107 111 -4 5 -59 60) $ SQUARE LEAD SHIELD
10 1 -.95 105 -161 $ LARGE UPPER PARAFFIN CELL

-------------

40 1 -.95 -105 106 -4 5 -59 60
41 1 -.95 -105 106 -12 13 -63 64 (4:-5:59:-60)
42 1 -.95 -105 106 -20 21 -69 70 (12:-13:63:-64)
43 1 -.95 -105 106 -22 23 -73 74 (20:-21:69:-70)
44 1 -.95 -105 106 -24 25 -75 76 (22:-23:73:-74)
45 1 -.95 -105 106 -26 27 -77 78 (24:-25:75:-76)
46 1 -.95 -105 106 -28 29 -79 80 (26:-27:77:-78)
47 1 -.95 -105 106 -161 (28:-29:-80:79)

-------------

50 1 -.95 -106 107 -4 5 -59 60
51 1 -.95 -106 107 -12 13 -63 64 (4:-5:59:-60)
52 1 -.95 -106 107 -20 21 -69 70 (12:-13:63:-64)
53 1 -.95 -106 107 -22 23 -73 74 (20:-21:69:-70)
54 1 -.95 -106 107 -24 25 -75 76 (22:-23:73:-74)
55 1 -.95 -106 107 -26 27 -77 78 (24:-25:75:-76)
56 1 -.95 -106 107 -28 29 -79 80 (26:-27:77:-78)
57 1 -.95 -106 107 -161 (28:-29:-80:79)

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61 1 -.95 -107 110 -20 21 -69 70 (12:-13:63:-64)
62 1 -.95 -107 110 -22 23 -73 74 (20:-21:69:-70)
63 1 -.95 -107 110 -24 25 -75 76 (22:-23:73:-74)
64 1 -.95 -107 110 -26 27 -77 78 (24:-25:75:-76)
65 1 -.95 -107 110 -28 29 -79 80 (26:-27:77:-78)
66 1 -.95 -107 110 -161 (28:-29:-80:79)

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270  2  -11.34  -130 -161 -16 17 -69 70 (2: -3: 59: -60)
271  1  - .95  -130 -161 (16: -17.69: -70)
C
280  2  -11.34  161 136 -16 17 -69 70 (2: -3: 59: -60) $Pb. EXT OF COLL.
281  0  -117 131 -2 3 -59 60 $ AIR INSIDE COLLIMATOR
282  7  -1.0  -131 132 -2 3 -59 60 $ BORATED POLY PLUG
283  7  -1.0  -132 133 -2 3 -59 60
284  7  -1.0  -133 134 -2 3 -59 60
285  7  -1.0  -134 135 -2 3 -59 60
286  7  -1.0  -135 136 -2 3 -59 60
C
C
SAMPLE AND OTHER CELLS AROUND IT
C
320  0  -112 113 202 -203 -59 60  $ CENTRAL AIR
321  2  -11.34  -112 113 202 -203 ((59 -65):(-60 66))
322  2  -11.34  -112 113 202 -203 ((65 -69):(-66 70))
323  2  -11.34  -112 113 202 -203 ((69 -73):(-70 74))
324  2  -11.34  -112 113 202 -203 ((73 -75):(-74 76))
325  2  -11.34  -112 113 202 -203 ((75 -77):(-76 78))
326  2  -11.34  -112 113 202 -203 ((77 -79):(-78 80))
327  2  -11.34  -112 113 202 -203 ((79 -161):(-80 -161))
C
330  0  -112 113 201 -202 -59 60  $ CENTRAL AIR
331  9  -8.65  -112 113 201 -202 ((59 -65):(-60 66))
332  9  -8.65  -112 113 201 -202 ((65 -69):(-66 70))
333  9  -8.65  -112 113 201 -202 ((69 -73):(-70 74))
334  9  -8.65  -112 113 201 -202 ((73 -75):(-74 76))
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337  9  -8.65  -112 113 201 -202 ((79 -161):(-80 -161))
C
340  1  - .95  -112 113 200 -201 -59 60  $ CENTRAL CASING
341  1  - .95  -112 113 200 -201 ((59 -65):(-60 66))
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C
350 10 -2.38 -112 113 -200 -59 60 $ TOP CENTRAL SAMPLE PORTION
351 10 -2.38 -112 113 -200 ((59 -65):(-60 66))
352 10 -2.38 -112 113 -200 ((65 -69):(-66 70))
353 10 -2.38 -112 113 -200 ((69 -73):(-70 74))
354 10 -2.38 -112 113 -200 ((73 -75):(-74 76))
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420 0 -113 114 202 -203 -59 60 $ CENTRAL AIR
421 2 -11.34 -113 114 202 -203 ((59 -65):(-60 66))
422 2 -11.34 -113 114 202 -203 ((65 -69):(-66 70))
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431 9 -8.65 -113 114 201 -202 ((59 -65):(-60 66))
432 9 -8.65 -113 114 201 -202 ((65 -69):(-66 70))
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440 1 -.95 -113 114 200 -201 -59 60 $ CENTRAL CASING
441 1 -.95 -113 114 200 -201 ((59 -65):(-60 66))
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450 10 -2.38 -113 114 -200 -59 60 $ TOP CENTRAL SAMPLE PORTION
451 10 -2.38 -113 114 -200 ((59 -65):(-60 66))
452 10 -2.38 -113 114 -200 ((65 -69):(-66 70))
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520 0 -114 115 202 -203 -59 60 $ CENTRAL AIR
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Cells related to collimator, detector etc.

Detector

Pb. back cover

Void

Outside universe

Surface descriptions

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C

160 SO 0.1 $ Spherical source
161 SO 60.0 $ Inner surface of iron container
162 SO 63.0 $ Outer surface of iron container
163 SO 100 $ Outside universe

C

170 CZ 3.0 $ Detector
171 CZ 3.05 $ Al. cover
172 CZ 8.05

C

Sample related cells

C

200 C/Y 0 -12.5 3.30 $ End of sample, start of casing
201 C/Y 0 -12.5 4.00 $ End of sample poly casing
202 C/Y 0 -12.5 4.20 $.2CM AIR GAP (Changed to Cd)
203 C/Y 0 -12.5 4.5 $ .3cm pb shld

C

MODE N P

C

Sample related cells

C

Weight Windows

C

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WWN 1:N 0.5

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WWP: P 5 3 5

WWE: P 1.0000E+02

WWN 1: P 5.0000E-01 1.0E+6 1.0E+6

1.0E+6  1.0E+6  1.0E+6  1.0E+6
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1.0E+6  6R
1.0E+6  6R
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1.0E+6  7R
1.0E+6  7R
1.0E+6  7R
1.0E+6  7R
Only the irradiated portion of sample (not including the casing) are allowed to have photon production by using a PWT card value of -1.0E+06

C PWT
-1.0E+06 185R
-1.0E+06 7R
-1.0E+06 7R
-1.0E+06 7R
1.0E-06 -1.0E+06 6R
-1.0E+06 7R
-1.0E+06 7R
-1.0E+06 7R
1.0E-06 -1.0E+06 6R
-1.0E+06 7R
-1.0E+06 7R
-1.0E+06 7R
1.0E-06 -1.0E+06 6R
-1.0E+06 7R
-1.0E+06 7R
-1.0E+06 7R
1.0E-06 -1.0E+06 6R
-1.0E+06 6R

------------------------ Source specification ------------------------

C
C UNIFORM SPHERICAL VOLUME SOURCE AT (0,0,0)
C
SDEF CEL = 1 POS = 0 0 0 ERG D1 RAD D2 VEC = 0 0 -1
SC1 WATT ENERGY SPECTRUM FROM 0 TO 14.1 MEV.
S11 H 0 10.0
SP1 -3 1.025 2.926
S12 L 0 0.1
SP2 -21 2
C
C ------------------------ Material specification ------------------------
C
C NOTE:----> Use S(a,b) treatment else results are lower.
Sample material

Av. CLAY(0.3), OPAL(0.4), CALCITE(0.3), POROSITY 10%

Photon production biasing

COMMENT:--> Reaction 001, 002 and 034 are primary photons and their centroid would be expected to shift to higher energies with increasing energy of the neutrons. However, the XS for these line gamma rays is ZERO for non thermal captures. Hence this shift would not be expected and hence in this case we will not be able to observe it. Another interesting observation can be the changing branching ratio due to changes in the neutron energy spectrum. In an attempt to see this, the reaction 007 was added and Gaussian broadening tally function was used.

Neutron fluxes

FLUX IN SAMPLE CELLS
The following neutron tally may be used for computing the fast neutron flux at the detector.

As $S(a,b)$ is ON, no thermal contribution is included in this tally due to the tally region being surrounded by a DXTRAN sphere.

Fast flux for detector damage calculation

Code also calculates it.

Surrounds detector, no $S(a,b)$ contribution.

Don't turn off roulette. Fatal error due to very low weight transport.

Photon fluxes

Current on detector

Surrounds detector

Only sample contributions

Detector top surface

First cosine bin relevant (-1 to 0)

Converts to surface current density
No contribution is seen to the detector rear face except at low energy (< 1MeV). Hence this contribution is not tallied any more.

Gaussian broadening of 6.200 MeV peak. The Gaussian FWHM parameter was calculated as 2.35 $\sigma$, where $\sigma = F\Gamma/(E/\epsilon)$.