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CONTINUED DEVELOPMENT OF THE COMBINED PULSED NEUTRON EXPERIMENT (CPNE) FOR LUNAR AND PLANETARY SURFACES

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FOREWORD

This is Report No. IITRI-V6121-2, a Final Report under Contract No. NASW-2258, entitled "Continued Development of the Combined Pulsed Neutron Experiment (CPNE) for Lunar and Planetary Surfaces." This report covers the period from June 15, 1971 through September 23, 1972.

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Respectfully submitted,
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ABSTRACT

The following document reports the current progress on the inelastic scattering, capture, and activation gamma-ray portions of the Combined Pulsed Neutron Experiment (CPNE). Experiments are described which have enabled us to reduce the weight of the experimental probe to 7.3 Kg. Parametric studies are described which have enabled us to optimize the experimental parameters (e.g., gate time settings, neutron pulse rate, etc.). Estimated detection sensitivities using this light weight probe and the optimized experimental parameters are discussed.

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1. INTRODUCTION, SUMMARY, AND CONCLUSIONS

The Combined Pulsed Neutron Experiment (CPNE) is a sophisticated analysis method capable of performing bulk major element composition analysis to depths of about 50 gm/cm^2. Its development has been predicated on the assumptions that such information for most of the major solar system bodies is important and that a system compatible with the usual requirements of space flight can be developed. The present project has dealt primarily with verifying the latter assumption.

The CPNE utilizes five different neutron analytical methods: capture gamma-ray analysis, fast neutron activation analysis, inelastic neutron scattering, thermal neutron die-away, and epithermal neutron die-away. Elemental compositions can be determined by three of these methods (capture gamma-ray analysis, fast neutron activation analysis, and inelastic neutron scattering); the presence of hydrogen can be determined by two of the methods (capture gamma-ray analysis and epithermal neutron die-away); and density information can be provided by the thermal neutron die-away technique.

As a result of extensive experimentation performed earlier, the feasibility of using the CPNE for elemental analysis has been established[1]. In particular, the five neutron techniques were successfully combined and data were collected on rock models using a first generation probe. However, until the present work was undertaken no serious attempt had been made to minimize the probe weight and to establish the final parameters of the experiment (e.g., neutron pulse rate, neutron output, etc.). The program discussed herein was initiated to investigate these areas. The major objectives were (1) to evaluate the sensitivity of the CPNE in low-weight configurations that are compatible with planetary mission restrictions, (2) to investigate the effect on sensitivity of experimental parameters such as pulse rate, gate times, neutron output, and count time with the goal of optimizing the values of these parameters, and (3) to collect a library of

component spectra to be used in the analysis of the spectral data.

These objectives have been largely attained. To accomplish the objective of weight reduction, the effects of the following changes in the probe design were investigated; (1) substitution of beryllium for molybdenum in the fast neutron shadow shield, (2) substitutuion of a 5.1 cm x 7.6 cm NaI(Tl) gamma-ray detector for the larger 7.6 cm x 7.6 cm NaI(Tl) detector, and (3) elimination of the neutron reflector. The studies indicated that the data will be unaffected by replacement of beryllium for molybdenum in the fast neutron shadow shield as long as the neutron generator is constructed of material that exhibits low cross sections for neutron capture, activation, and inelastic scattering (beryllium cannot adequately shield the detector from gamma rays originating in the neutron generator).

The only important effect observed when a 5.1 cm x 7.6 cm NaI(Tl) detector replaces the 7.6 cm x 7.6 cm NaI(Tl) detector is about a factor of two decrease in neutron efficiency i.e., the intensity of the spectrum is decreased by about a factor of two for a given neutron output. The spectral shape is somewhat altered due to the different ratio of full energy peak intensity to escape peak intensities for the two detectors, but the spectral quality (for a given intensity) is not significantly affected.

Our studies concluded that a neutron reflector is very beneficial to the capture gamma-ray analysis and under certain conditions (i.e., a dry sample) it is essential. For common earth type rock samples (e.g., basalt, dunite, granite) which contain several tenths of a percent water, a reflector enhances the capture gamma-ray intensity by more than a factor of two compared to the intensity measured using no reflector. No new capture gamma-ray peaks occur when a reflector is used. For dry samples, however, many capture gamma-ray peaks are observable only when a reflector is used.

The studies whose goal was to optimize the parameters of the experiment consisted of an extensive investigation of the effect on sensitivity of parameters such as neutron intensity, neutron pulse rate, gate times, count time, and geometry (i.e., distance between the probe and the sample and separations between the neutron source, shadow shield, and gamma-ray detector). Experimentation was used to optimize the geometry while computer programs were utilized for the optimization of the other experimental parameters.

Our computer studies resulted in the following conclusions. (1) It is best to use the highest neutron output that the electronics can handle. Although the dead time of the analyzer will cause a decrease in neutron efficiency, the gain in spectrum intensity for a given count time will be greater than the loss in neutron efficiency. (2) The optimum capture gamma-ray gate is about 20-520 μsec. (3) A pulse rate of about 500 pps is probably a practical optimum. A lower pulse rate results in a longer required count time. Although a higher pulse rate would allow a shorter count time, the separation of the capture and activation spectra would be more difficult (i.e., the activation spectrum would contain a significant capture component). (4) Using these optimized values for the experimental parameters, a capture gamma-ray spectrum with statistics sufficient to attain the sensitivities listed in Table 1 can be obtained in about 45 minutes.

The threshold given for each element (Table 1) represents that abundance which results in the error (one standard deviation) being equal to the abundance. With this definition of the detection threshold, elements whose abundances are greater than the detection threshold but less than about three times the threshold will be detected, although no meaningful quantitative results will be obtained. Elements whose abundances are greater than about three times the detection threshold will be detected and their abundances determined.

TABLE 1

DETECTION THRESHOLDS FOR THE CPNE

Element	Detection Threshold (weight percent)
H	0.01
O	2.
Na	0.2
Mg	0.8
Al	0.3
Si	0.3
K	0.15
Ca	0.4
Ti	0.04
Fe	0.15

The studies concerning the geometry indicated that a configuration with the probe centerline 9.5 cm above the sample provides a reasonable compromise between spectrum quality and neutron economy. Similarly, a 5.1 cm detector-to-shadow shield separation and a 2.5 cm neutron source-to-shadow shield separation were found to be the practical optimum distances.

As a result of these studies, the design of the probe can now be specified. The probe will consist of a basically cylindrical structure of length 91.5 cm and maximum diameter of 10.3 cm. The detector will be a 7.6 cm x 7.6 cm NaI(Tl) crystal. A beryllium shadow shield in the form of a truncated cone of length 14.3 cm and radii 2.5 cm and 7.6 cm is located between the neutron generator and the detector, and 4 cm thick x 11.3 cm long neutron reflector is located above the generator. The reflector is necessary in order to obtain good quality capture gamma-ray spectra from dry samples. The probe will weight about 7.3 kg. (compared to 29.5 kg for the first generation probe). If weight is of prime concern, a 5.1 cm x 7.6 cm detector can be used. The weight of the probe would then drop to about 5.5 kg, but the required counting time would be increased from 45 minutes to about 90 minutes.

Due to unanticipated difficulties, we were unable to collect a library of component spectra. Because of the dryness of the samples and as a result an underestimation of the required size of the models, the capture gamma-ray spectra were not of adequate quality to be used for the library.

As a result of the studies just concluded, the CPNE has reached a stage of development making it ready for field testing in the light weight probe configuration and assignment to a mission. Additional work in the area of data analysis, however, needs to be accomplished. This should include the collection of a library of component spectra using the light weight probe and further investigations concerning the effects of density and hydrogen content on spectral analysis.

The results of the present studies are the subject of this report and detailed discussions follow. In order to provide some perspective on the rationale for various aspects of the studies, a brief history of the development of the CPNE will be presented in the following section.

2. EVOLUTION OF THE CPNE

2.1 Origin of the CPNE

Prior to the initiation of the CPNE studies, capture gamma-ray analysis, fast neutron activation analysis, and inelastic neutron scattering were well established laboratory techniques, each requiring distinctly different experimental parameters. For example, capture gamma-ray analysis requires a source of thermal neutrons while fast neutron activation and inelastic scattering require fast neutrons. The various techniques, however, have many similarities (e.g., they are all neutron techniques which produce gamma radiations and all require a neutron source and a gamma-ray detector). These similarities lead to the suggestion that it may be possible to combine the techniques into a single package[2]. Specifically the idea of the CPNE was based on the assumptions that (1) the various techniques could all be performed using a source of 14 MeV neutrons, (2) a portable 14 MeV neutron generator consistent with the space mission requirements of small size and low power requirements could be built, and (3) the various techniques could be integrated into a single package utilizing the same neutron source, gamma-ray detector, and electronics. Since portable 14 MeV neutron generators existed at the time of the conception of the CPNE (1965), the design of one that would meet space mission requirements was expected to pose no problem. Also, since the radiations from the various techniques are emitted at different times when using a pulsed neutron source (i.e., gamma rays from inelastic neutron scattering are emitted only during the neutron burst, the capture gamma-ray intensity quickly builds up after the burst and then dies away, and the intensity of activation gamma rays builds up during the neutron burst and remains approximately constant between bursts), it was believed that appropriate timing of the neutron generator and spectrum recording could largely eliminate the overlap of spectral types. The questions that required investigation were (1) would the 14-MeV

neutrons thermalize close enough to the surface to permit observation of capture gamma rays, (2) can spectra of adequate quality be obtained from bulk samples, (3) can the techniques be integrated into a single package which is small enough and light enough to be considered in a space exploration mission, and (4) if the answers to the above are yes, what is the best way to combine the techniques and will the combined experiment provide sufficient sensitivity.

Since the CPNE utilizes five distinctly different neutron techniques, it was developed as a joint effort involving groups with experience in the various techniques. IIT Research Institute was given the responsibility of developing the capture and activation portions of the CPNE (recently IITRI was charged with the additional task of obtaining the neutron inelastic scattering data, with Dr. James Waggoner serving as a consultant); Mobil Research and Development Corporation became responsible for the thermal neutron die-away and epithermal neutron die-away techniques; Dr. James Waggoner was given the responsibility of developing the inelastic portion of the CPNE; and Sandia Corporation was given the job of designing and building the neutron generator. The following discussions will be primarily concerned with the capture and activation portions of the CPNE. The development of the inelastic and die-away portions of the CPNE can be found in References 3 and 4, respectively.

2.2 Feasibility Studies

The feasibility studies for the CPNE were accomplished in two stages. The initial studies were concerned with demonstrating that it was feasible to perform capture gamma-ray analysis using 14-MeV neutrons on a bulk sample. Once this was accomplished, the emphasis of the studies was placed on demonstrating that the five neutron techniques could successfully be integrated into a single package and that useful data could be obtained by the CPNE.

Initial feasibility studies were concerned with determining (1) whether the 14-MeV neutrons will thermalize close enough to the surface of the sample to permit observation of capture gamma rays and (2) whether spectra of adequate quality (i.e., intensity and resolution) can be obtained from a semi-infinite sample. Studies were undertaken to predict and experimentally measure the thermal neutron flux in a semi-infinite sample when a source of 14-MeV neutrons was located on the surface of the sample. A neutron transport computer code (DTK) was used to provide a qualitative understanding of the effects of various parameters (e.g., composition, density, hydrogen content) on the thermal neutron flux distribution.

Results from the computer studies indicated that for typical terrestrial rocks (e.g., basalt, granite, dunite) the thermal neutron flux reaches a maximum at an appreciable distance below the surface (about 90 gm/cm^2 for a dry sample), which implies that the bulk of the thermal neutron capture gamma rays must penetrate relatively large thicknesses of material if they are to be observed. Further results of these computer studies indicated that if a low-Z material is placed above the source to reflect into the sample some of the neutrons normally lost to the atmosphere, the thermal neutron flux near the surface of the sample will be greatly increased. In fact, if as little as 4 cm of polyethylene, paraffin, or water is located above the neutron source, the thermal flux distribution

will reach a maximum at the surface. This increase in the thermal neutron flux at the surface of the sample significantly increases the sensitivity of the neutron capture gamma-ray technique, and since this increase in thermal flux is accomplished without decreasing the flux of fast neutrons incident on the sample, the sensitivities of the activation analysis technique and inelastic scattering technique are not inherently affected.

The results of the computer studies were verified experimentally using a large 152 cm x 152 cm x 76 cm sand sample and neutron activation foils. Results indicated that, as predicted, the thermal flux, in the absence of any reflector material, peaked at about a depth of 95 gm/cm^2. With a 4 cm thick paraffin reflector above the 14-MeV neutron source, the thermal flux at the surface is increased by about a factor of 100 and has its maximum value there.

The next step was to show that capture gamma rays can actually be measured from a bulk sample when a 14-MeV neutron source is used. This was accomplished using large (152 cm x 152 cm x 76 cm) samples of sand and an iron-sand mixture. Subsequently, more complex samples (such as basalt, dunite, granite) were used. The observed capture gamma-ray spectra from these samples were of adequate intensity and quality to enable identification of gamma-ray peaks from the component elements.

After establishing the feasibility of using the capture gamma-ray technique on bulk samples with a 14-MeV neutron source, the next step was to demonstrate that the various neutron techniques can be integrated into a single package using the same neutron source, gamma-ray detector, and electronics. In particular, the assumption that mutual interferences of the techniques could be reduced by appropriate timing of the pulsed neutron generator and sample collection had to be demonstrated.

Figure 1 shows the time relationship of the gamma rays emitted from a bulk sample when it is irradiated by a pulsed source of fast neutrons. Gamma rays from inelastic neutron scattering are emitted only during the neutron burst (because the average neutron energy is reduced to a value below the inelastic scattering threshold within 0.1 μsec). The capture gamma-ray intensity quickly builds up after the burst (as the fast neutrons are thermalized in the sample) and then die away (as the thermal neutrons are captured or lost from the sample). The intensity of the gamma rays due to activation analysis builds up during the neutron burst and remains approximately constant between bursts (because the time between pulses is much shorter than the half lives of the radioactive nuclides produced).

This unique time realationship between the emission of the gamma rays from the various reactions enables the different techniques of the CPNE to be combined into a single experiment. Mutual interferences can be reduced to negligible (or at least tolerable) levels by allowing sampling only during selected intervals. The sample gates which will give optimum separation of the gamma rays due to the various reactions are shown in Figure 1. By using these gates the inelastic, capture plus activation, and activation contributions to the gamma-ray spectrum are separated.

A first generation probe was designed and fabricated at IITRI for use in demonstrating that the various techniques can be combined into a single package. Figure 2 shows a drawing of this probe. This probe was constructed with a molybdenum framework and shadow shield (see Section 2.3 for a discussion of the need for a shadow shield) and was about one meter in length. Gamma-ray detection was accomplished using a 7.6 cm x 7.6 cm NaI(Tl) detector and neutrons were produced by a prototype neutron generator (developed by the Sandia Corporation) of a type that could be used on a space mission. It was designed to provide neutron reflector thicknesses of 0, 4 cm, and 8 cm.

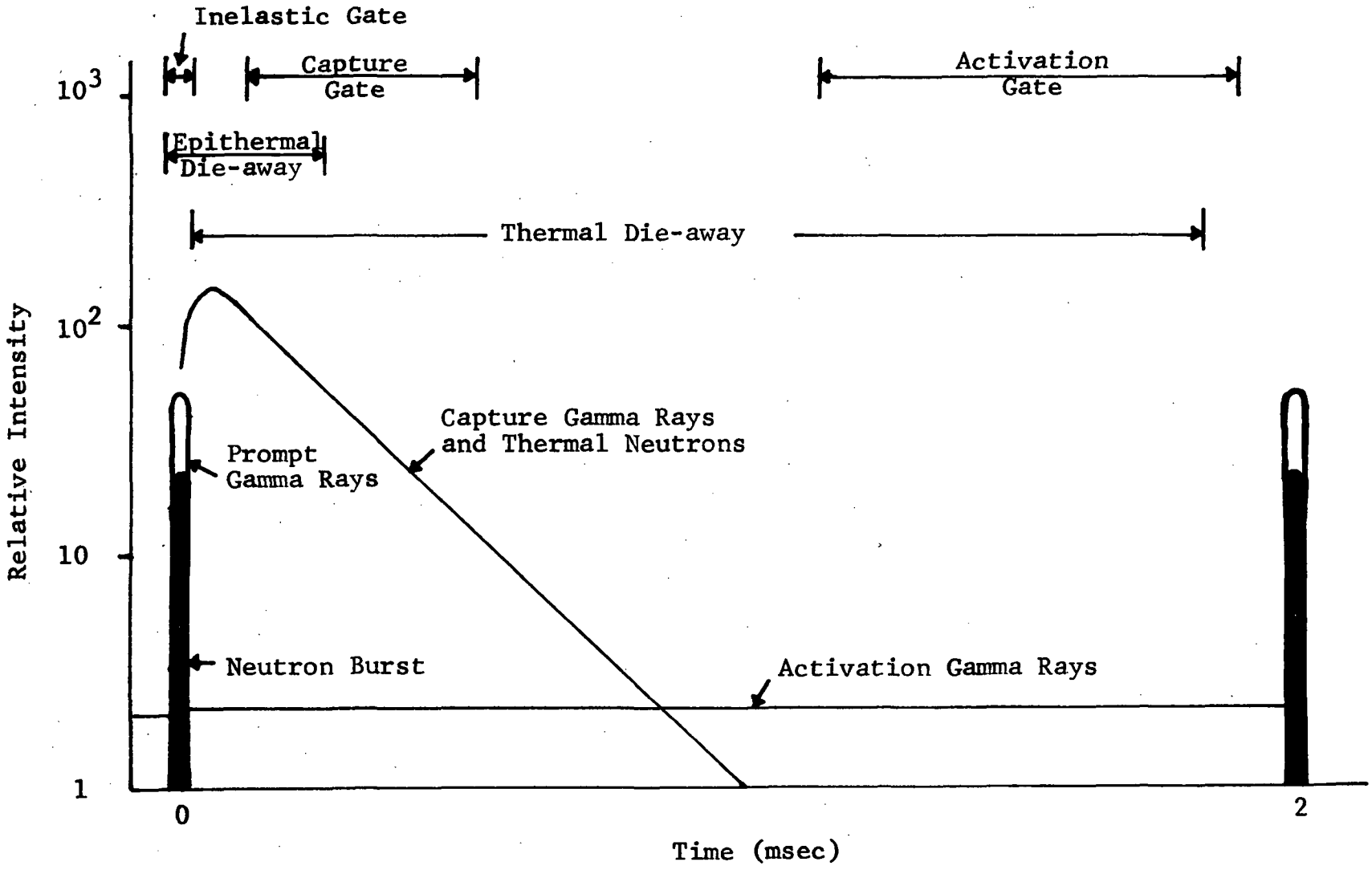


Figure 1 TIMING DIAGRAM FOR THE CPNE

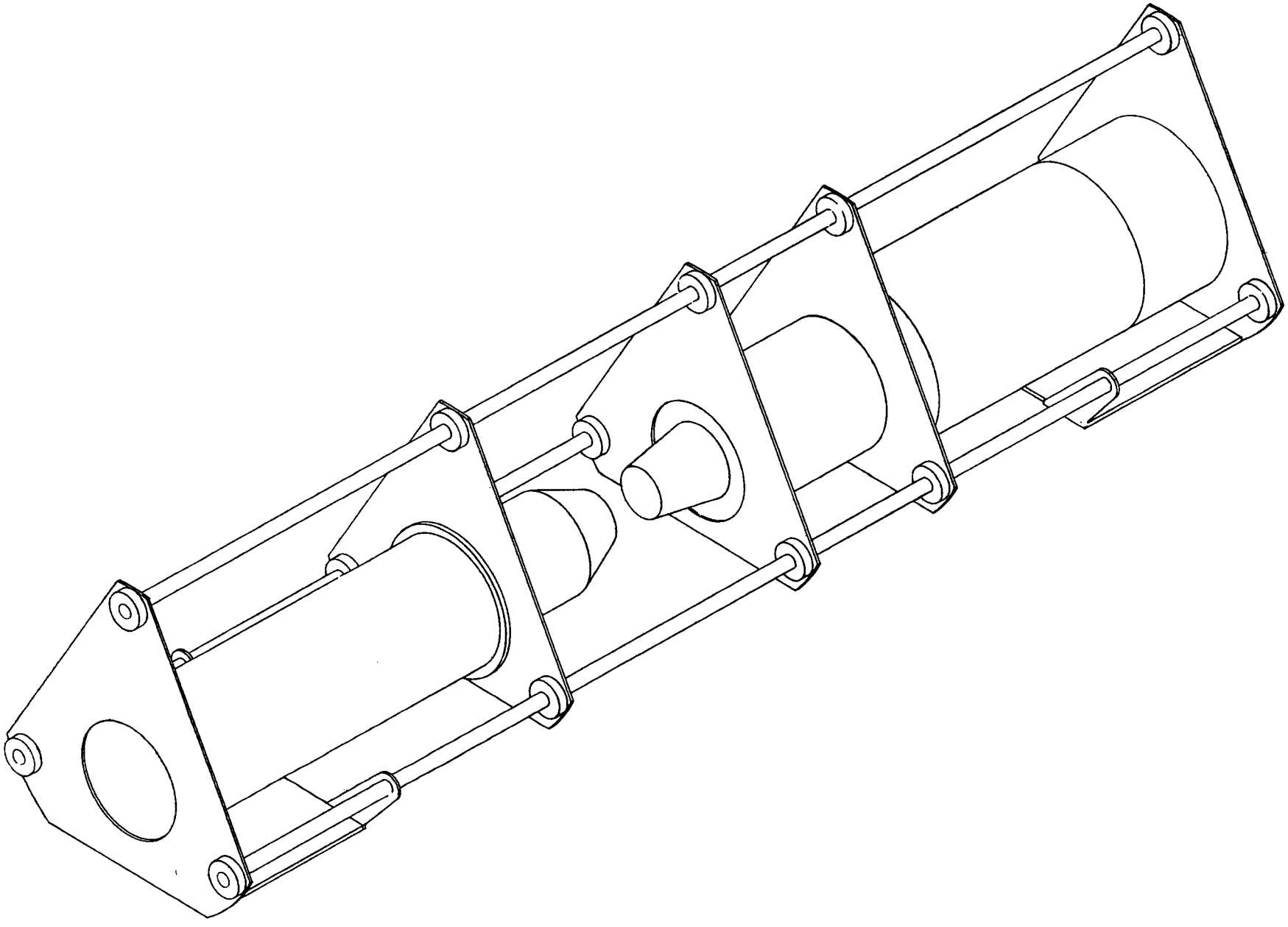


Figure 2 COMBINED NEUTRON EXPERIMENT DETECTOR PROBE (View with no Reflector)

The weight of the probe, including the neutron generator and 7.6 cm x 7.6 cm NaI(Tl) detector in the three configurations were: (1) 23.6 kg without reflector, (2) 29.5 kg with the 4-cm reflector, and (3) 39.1 kg with the 8-cm reflector. The centerline of the probe was located 6.7 cm above the sample to be analyzed.

The prototype probe was tested using the Mobil models of crushed basalt, dunite, and granite. Data for all the techniques (i.e., capture, activation, inelastic scattering, and die-away) was successfully obtained using the probe. Through these measurements the CPNE truly became a combined experiment.

2.3 Experimental Configuration

The gathering of the necessary capture and activation gamma-ray spectra for the studies of the CPNE required the use of large samples. A 152 cm x 152 cm x 76 cm plywood box filled with a crushed sample provided a very convenient approximation to a semi-infinite medium. Samples of basalt, dunite, granite, and sand were used.

The detection of the gamma rays for the CPNE has been accomplished primarily with a 7.6 cm x 7.6 cm NaI(Tl) detector. Recently, however, studies have been carried out to assess the feasibility of replacing the 7.6 cm x 7.6 cm NaI(Tl) detector with a 5.1 cm x 7.6 cm detector.

Early studies indicated that fast neutrons striking the gamma-ray detector can produce inelastic gamma rays which act as noise in the inelastic spectrum. Similarly, thermal neutrons can interact with the detector, producing capture gamma rays and gamma rays from the decay of radioactive daughters (e.g., I^{128}). These will contribute noise to the capture and activation spectra. Therefore, a fast neutron shadow shield was used between the neutron source and the detector to protect the detector from fast neutrons coming from the source. In addition, a thermal neutron shield was located around the detector to protect it from thermal neutrons scattered from the sample and surrounding structures. Studies of candidate materials for these shields resulted in the selection of Li^6 (in the form of Li^6F) for the thermal neutron sheild and molybdenum for the fast neutron shadow shield.

Various geometries were investigated for use in the CPNE. The angle between the source-detector axis and the sample surface was varied between 0° (horizontal) and 90° (vertical). As expected, the horizontal geometry with the source, shadow shield, and detector lying along a common axis parallel to the surface of the sample was found to be optimum for the capture

gamma-ray studies. A 9.5 cm spacing between this axis and the surface of the sample was found to provide a reasonable compromise between the spectrum quality and neutron economy. The source-to-shadow shield and shadow shield-to-detector spacings were also varied. Spacings of 2.5 cm and 5.1 cm, respectively, were selected, although these spacings were found not to be critical to the capture gamma-ray studies.

2.4 Electronics

Since one of the features of the CPNE is its ability to separate the gamma rays from the various reactions, a means of gating the multichannel analyzer on and off at the required times and routing the signal into the appropriate part of the memory was required. A gate generator and router (GGR) was designed and fabricated by IITRI. The GGR allows the analysis period to straddle the neutron burst (for collection of the inelastic scattering spectrum), occur just after the neutron burst (for collection of the capture gamma-ray plus activation spectrum), and occur just prior to the following neutron burst (for collection of the activation spectrum). In addition, the GGR provides the signals necessary to route the pulses detected during each of these intervals into different portions of the analyzer's memory. A schematic diagram of the GGR can be found in Reference 1.

During the development of the CPNE the electronics were continually updated. With the system used initially the neutron output was limited to about 7000 per pulse and sampling for capture gamma rays could not begin earlier than 100 μsec after the neutron burst. If these limits were violated, degradation of the gamma-ray spectrum resulted. Since it is desirable to collect the data from the CPNE as quickly as possible, the use of higher neutron outputs and earlier sampling for capture gamma rays was required. The electronics were upgraded in several steps, resulting in the system shown in Figure 3. This system (discussed in Section 3.2) allows neutron outputs in excess of 22,000 per pulse to be used and sampling for capture gamma rays to begin immediately after the neutron burst.

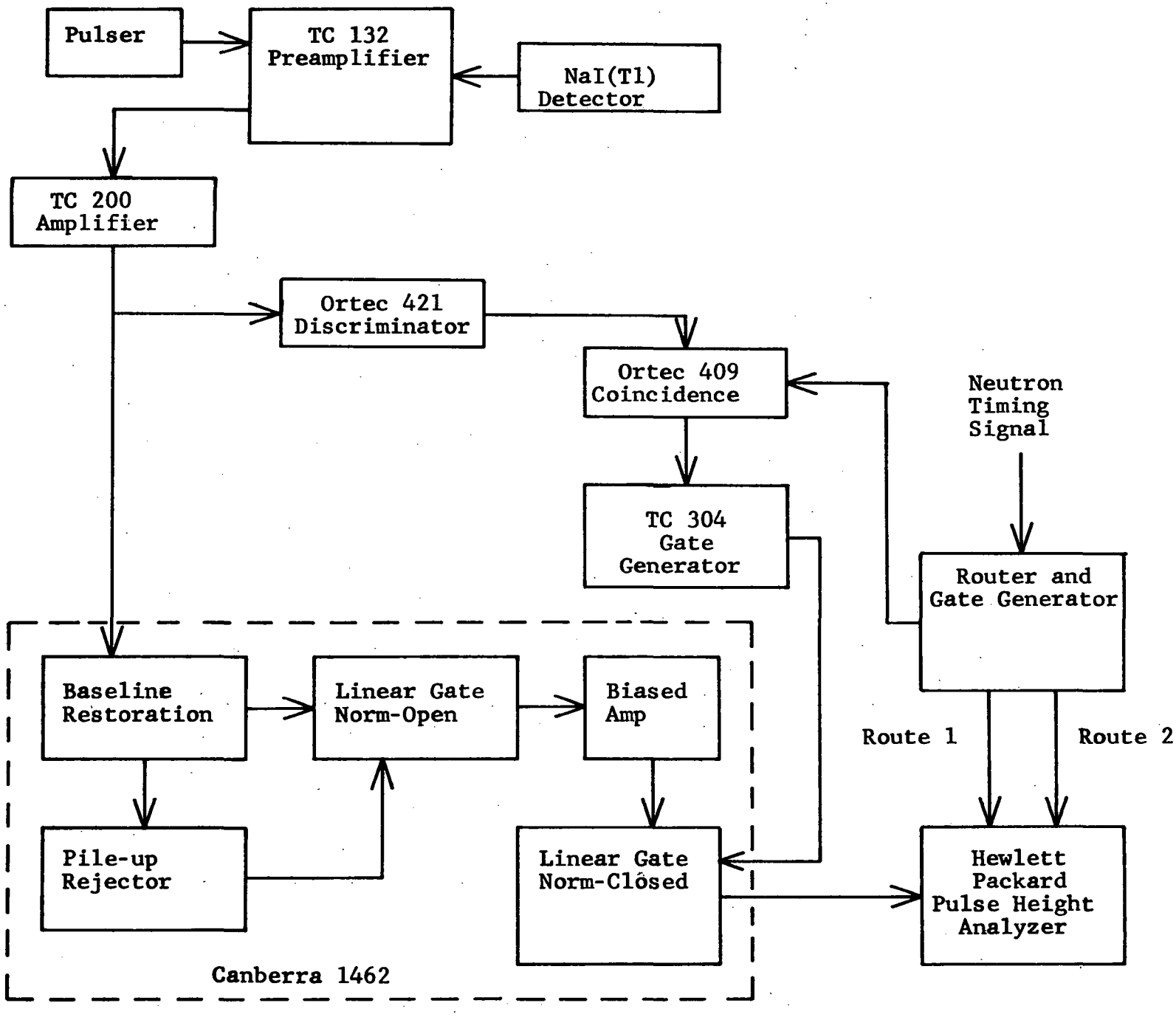


Figure 3 BLOCK DIAGRAM OF CURRENT DATA COLLECTION SYSTEM

2.5 Data Analysis

Neutron capture, inelastic neutron scattering, and activation produce complex gamma-ray spectra. Each spectrum contains peaks that are characteristic of the material being inspected. It is the purpose of an analysis scheme to determine the source (the element) and intensity (relative abundance) of the gamma rays which cause these peaks. If the sample is composed of many elements each of which emits a complex gamma-ray spectrum, the problem is compounded. In addition, if the sample is large causing the gamma-ray flux be attenuated before it reaches the detector, the problem is further compounded. This is the situation one is faced with when using the capture gamma-ray technique in the CPNE. In general, if a NaI(Tl) detector is used the capture gamma-ray spectrum from a compound sample will consist of many overlapping peaks on top of a complex structured background level.

There are several general approaches to the problem of analysis of gamma-ray spectra obtained using a NaI(Tl) detector. In one method (examplified by the computer program GAUST[5]) peak intensity and energy is determined by a least-squares fit of the data with a model. In a second method (commonly called the "library" method and examplified by the computer program ALPHA-M[6]) it is assumed that the spectrum to be analyzed is composed of a linear combination of a set of library spectra. The relative intensities of the components of the spectrum are then determined by a least-squares method. A third method (examplified by the approach developed by Trombka and his associates[7]) uses the response function of the detector (the pulse height distribution presented to the multichannel analyzer that results from the interaction of monoenergetic gamma rays in the detector) as a function of energy to calculate the gamma-ray flux incident on the detector.

After performing studies in which the merits of all the spectrum analysis approaches were compared (see Reference 1 for a detailed discussion of these studies), we selected the "library" method to be the most appropriate for the capture gamma-ray portion of the CPNE. The computer program ALPHA-M was modified to better serve our purposes and was used in further studies (e.g., optimization of experimental parameters and determination of sensitivities of the capture and activation techniques) of the CPNE.

2.6 Parametric Studies

Experimental parameters such as neutron pulse rate, sample gate times, and the neutron output are variable and must be optimized if the CPNE is to obtain the maximum information in the shortest period of time. Studies were performed to determine the optimum values for these parameters. These studies were largely computer studies based on specific experimental results. Since the collection of such a large series of experimental spectra for the various values of the parameters was not economically feasible, a computer program, called TIME, was written to calculate by statistical means the number of capture and background gamma rays expected to be detected as a function of the various experimental parameters. A sensitivity parameter was defined, and the values of the experimental parameters which maximized this sensitivity parameter were determined.

Although this study yielded parametric values which maximized the sensitivity parameter, the significance of the sensitivity parameter, was not yet known. For example, although it was assumed that if the value of the sensitivity parameter increased the sensitivity of the CPNE should increase, we did not know what the magnitude of this increase in sensitivity would be. To answer this question, a further study was performed. Since this study required the analysis of spectra obtained using various values of the experimental parameters, we wrote a computer program to generate simulated capture and activation spectra. Using these simulated spectra and the analysis program ALPHA-M, the significance of the sensitivity parameter was determined. In addition, sensitivities for elemental detection using the capture and activation techniques were also determined.

2.7 Studies Using a Germanium Detector

Some preliminary studies were carried out to determine the advantages of replacing the NaI(Tl) detector in the CPNE with a high resolution detector. A 40 cm^3 Ge(Li) detector and both isotopic and machine neutron sources were utilized in these studies. Results indicated the superiority of the Ge(Li) detector, both from the viewpoint of information obtained and the ease of analyzing and interpreting the data. There is essentially no interference among peaks, i.e., peaks rarely overlap in the Ge(Li) spectrum while overlapping of peaks is common in a NaI(Tl) spectrum. This makes data analysis easier, for each peak can then be fitted with a gaussian plus a background function using a least-squares method. The "library" approach (and its required set of component spectra) would no longer be used. Moreover, peaks (such as from magnesium and manganese) were detected which cannot be seen in the NaI(Tl) spectrum.

If a Ge(Li) detector is used the controlling electronics could also be simplified. Because the peaks would not overlap, the need for collecting separate capture, and activation spectra would no longer exist. Hence, the spectra could be acquired simultaneously, eliminating the need for gating.

As a result of these studies, it appears that it would be advantageous to use a high resolution Ge(Li) detector in the CPNE. Lithium drifted germanium detectors, however, require constant cooling. For storage a temperature of 200°K or less is required (dry ice), while a temperature of 100°K or less (usually liquid nitrogen at 77°K) is necessary during operation. Because of these severe temperature constraints, the use of a Ge(Li) detector on a space mission in the near future is improbable.

While NaI(Tl) continues to be the detector of choice for the CPNE, the appearance of intrinsic germanium detectors has renewed interest in the use of a high resolution detector. Intrinsic detectors may be stored at room temperature or above, a procedure that quickly destroys drifted detectors, and may be cycled at will. They require low temperatures for operation if high resolution is to be obtained; however, Martini and co-workers[8] have shown that only a modest loss of resolution is incurred at operating temperatures up to about 200°K. Hence, cryogenic temperatures would not be required during a long space voyage and cooling only to about 200°K would be required on a planetary surface. This in-situ cooling could be accomplished by radiation or a combination of radiation and thermoelectric cooling. In addition, large intrinsic detectors are now possible which attain resolutions only slightly poorer than for Ge(Li)[9]. We suggest that the use of an intrinsic germanium detector in the CPNE be investigated further.

3. CURRENT STUDIES

3.1 Objectives

The major objectives of the present investigation were (1) to evaluate the sensitivity of the CPNE in various low-weight configurations compatible with planetary mission requirements, (2) to optimize experimental parameters such as pulse rate, gate times, neutron output, and count time and to determine the sensitivity of the CPNE using the optimized parametric values, and (3) to collect a library of component spectra to be used in the analysis of the spectral data. The results of our studies aimed at accomplishing these goals are discussed in the following sections.

3.2 Improvements in the Electronics

Prior to the current program, IITRI's responsibility was restricted to the capture and activation data. The electronics that had been assembled for data collection were based on these requirements. In this phase of the project we have been charged with the additional task of obtaining the neutron inelastic scattering data, with Dr. James Waggoner serving as a consultant.

The assembled system proved to be inadequate for the inelastic work. Serious spectral degradation was present. One deficiency was traced to the manner in which the linear gate was being enabled by the IITRI fabricated gate generator and router (GGR). The GGR provides the logic signal that selects the time interval after, or associated with, each neutron burst for which a gamma-ray spectrum is being collected. To obtain the inelastic data the GGR is set to produce a fixed duration logic signal or output that coincides with the neutron production. The duration of this logic signal is typically 15 μsec. In the original electronics system the GGR directly controlled the linear gate and it was distinctly possible for the gate to shut down with a gamma-ray signal present, and hence alter the amplitude of the signal. In the present system, Figure 3, if the leading edge of a gamma signal occurs during the time interval established by the GGR, then an enable signal is generated to allow the pulse to pass through the linear gate.

In addition, the pile-up rejection, built into the Canberra unit, was made more sensitive by adjusting its input low level discriminator from 100 keV to 30 keV. Previously, the unit ignored the presence of gamma signals of less than 100 keV. Thus, significant pile-up could occur with resulting spectral degradation.

A change in preamplifier has improved the system's capture gamma capability. The TC 132 preamplifier is of the non-blocking type, intended for applications requiring quick recovery

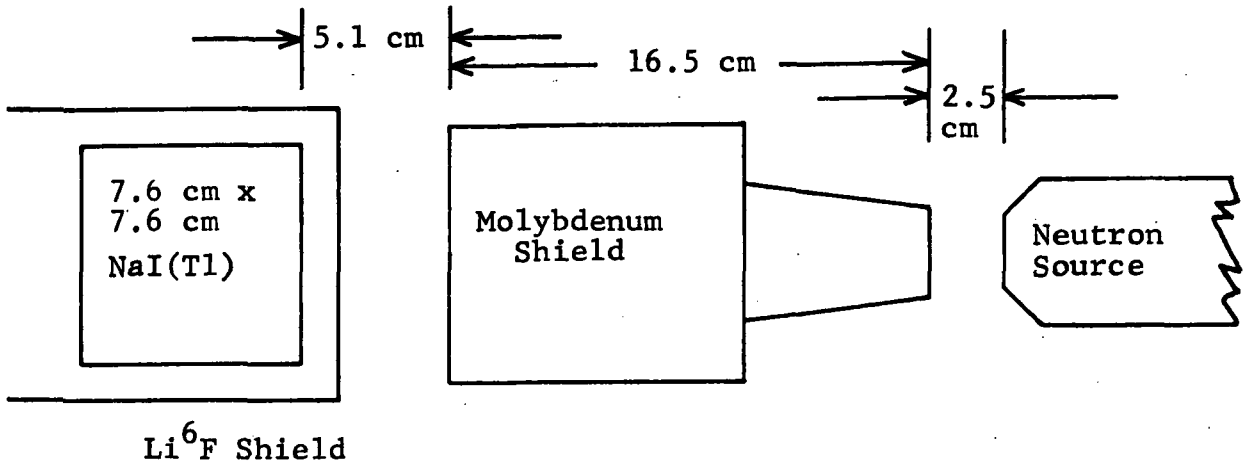
after an intense pulse. This is precisely the condition encountered in the collection of the capture gamma-ray spectra. This preamplifier has enabled us to open the linear gate immediately (within several microseconds) after the end of the neutron burst without degrading the resolution (previously we were restricted to about 50 μsec at a lower neutron burst intensity). In practice, however, because the neutron burst does not have a distinct cut off (the intensity exhibits a tail that lasts for 5-10 μsec) the linear gate is normally opened 20 μsec after the burst to ensure that no gamma rays from inelastic scattering are included in the capture spectrum.

New NaI(Tl) detectors have been acquired from the Bicron Corporation. The 7.6 cm x 7.6 cm thick detector is mounted on a SRC 75B01 photomultiplier tube and the 5.1 cm x 7.6 cm thick unit is mounted on a SRC 50B01. With the current system both detectors exhibit an on-axis resolution of 7.0 percent at 662 keV at a 15,000 cps count rate.

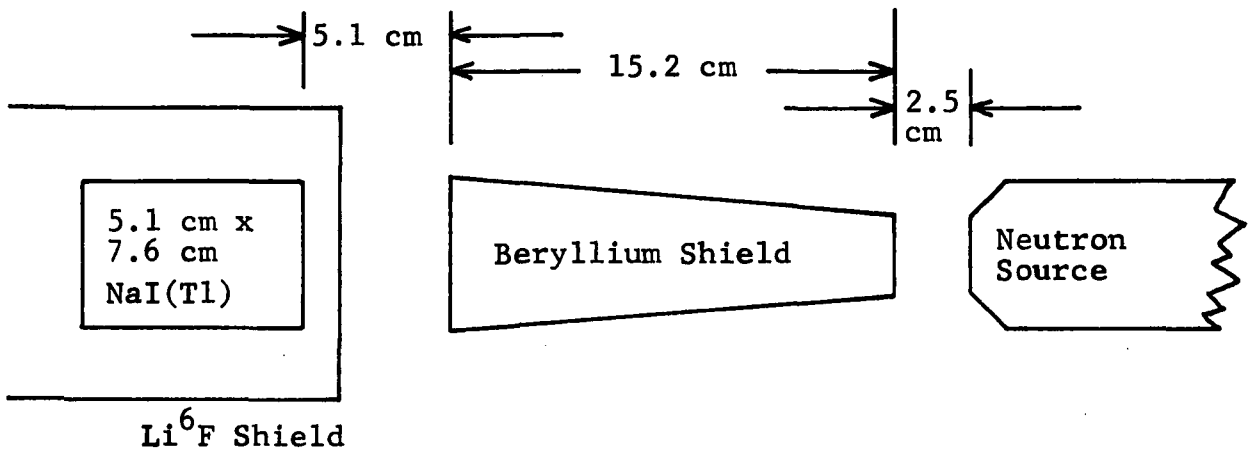
3.3 Low-Weight Configuration Studies

In order to minimize the mass of the CPNE, the effects of several weight-reducing changes in the probe design were studied. Capture gamma-ray, activation, and inelastic spectra were obtained and evaluated for each of these changes. The capture, activation, and inelastic measurements were performed using the 3 geometries of Figure 4. The IITRI Van de Graaff, pulsing at a rate of 500 pps, provided the source of neutrons for the capture and activation measurements. The neutrons for the inelastic scattering studies were provided by a prototype generator supplied by Sandia Corporation which operated at a rate of 5000 pps. The neutron outputs for all the measurements were monitored with a plastic scintillator biased to limit its sensitivity to neutrons with energies above 4 MeV.

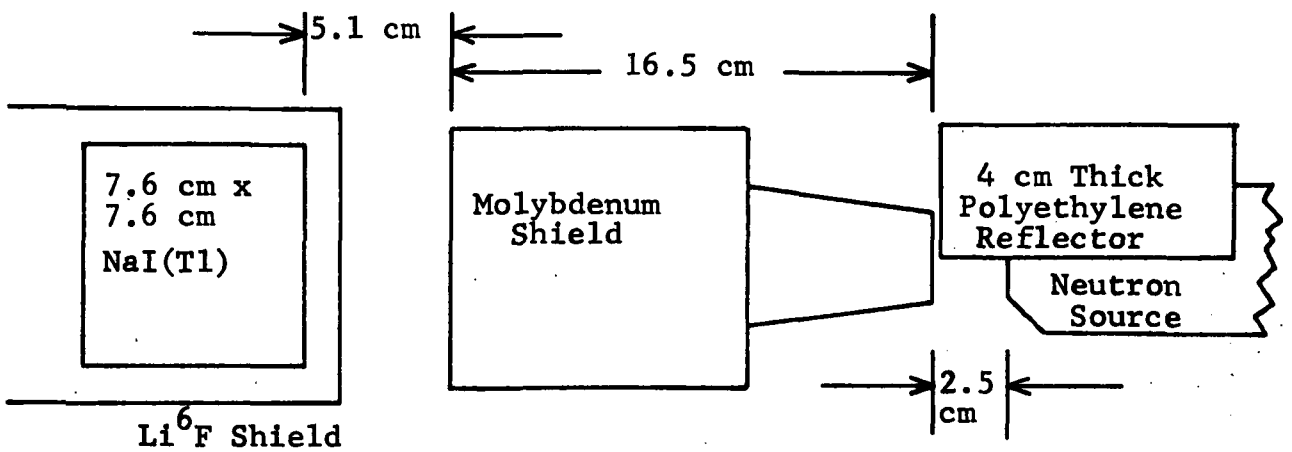
The first series of experiments were conducted to determine the optimum separations between probe and sample and between detector and shadow shield. We found that the inelastic spectra were significantly improved by raising the probe above the surface of the material rather than allowing it to rest directly on the sample. A configuration with the probe centerline 9.5 cm above the sample was selected as optimum. This separation provides a reasonable compromise between the spectrum quality and neutron economy. Experimentation also indicated that increasing the separation between the detector and the shadow shield beyond the standard 5.1 cm has a beneficial effect on the inelastic spectra. The improvement, however, is not adequate to warrant the lower neutron economy and the increase in the probe dimensions. An examination of the capture and activation spectra obtained for various separations indicated that neither raising the centerline of the probe to 9.5 cm above the sample nor increasing the detector-to-shadow shield separation beyond 5.1 cm had any noticeable effect. As a result of these measurements, a 9.5 cm separation between sample surface and probe centerline and a 5.1 cm detector-to-shadow shield separation were adopted as standard.



Geometry (a)



Geometry (b)



Geometry (c)

Figure 4 EXPERIMENTAL CONFIGURATIONS

The first weight reduction alteration that was studied was removal of the Li^6F thermal neutron shield. The Li^6F shield was found not to be necessary for the inelastic portion of the CPNE. Inelastic spectra obtained under identical conditions with and without the Li^6F shield are indistinguishable. A comparison of capture and activation spectra obtained under identical conditions, both with and without the Li^6F shield, indicated that the Li^6F shield is necessary when a neutron reflector (e.g., 4 cm polyethylene) is used above the neutron source. When the Li^6F shield is not used, the activation of the crystal is increased. This increase in crystal activation results in a higher background count rate in the low energy portions of the spectra (both capture and activation) together with a considerable degradation in the resolution of the 511 keV annihilation peak. When the neutron reflector is not used, the only observable effect removal of the Li^6F shield produces is a slight degradation in the resolution of the annihilation peak in the capture spectrum. Since the use of a neutron reflector was required in some of the later studies, (e.g., comparisons of spectra obtained both with and without a reflector) we decided to continue using the Li^6F shield for the capture and activation portions of the CPNE.

Capture, activation, and inelastic scattering spectra were obtained under identical conditions except that in one case a 7.6 cm x 7.6 cm NaI(Tl) detector was used and in the other case a 5.1 cm x 7.6 cm detector was used. Comparisons of the spectra obtained indicated that with the larger detector the full energy peaks are enhanced as compared to the escape peaks and the gamma-ray count rate for a given neutron output is approximately doubled (i.e., the neutron efficiency is about doubled). No new peaks were detected in either the capture, activation, or inelastic spectra when using the 7.6 cm x 7.6 cm detector that are not present when the 5.1 cm x 7.6 cm detector is used.

Spectra obtained with a molybdenum and with a beryllium shadow shield were also obtained. The only significant difference in the inelastic spectrum when beryllium is substituted for molybdenum is the addition of a 4.42 MeV gamma line (and its escape peaks) due to inelastic neutron scattering on carbon. These gamma rays originate in the neutron generator itself and are not significantly attenuated by the beryllium shadow shield (beryllium is a good neutron shield but a very poor gamma-ray shield). If beryllium is selected for the shadow shield in the CPNE probe, the neutron generator must contain as little carbon as possible (at present the main construction material of the prototype generator is plastic and the insulating medium is oil).

Capture and activation spectra obtained using the two shadow shield materials were virtually identical. The only observable difference was the appearance of a small activation peak in the low energy region when the beryllium shield was used. This peak was due to activation of the Van de Graaff target and since beryllium cannot adequately shield the detector from gamma rays (neither capture, activation, nor inelastic) originating in the neutron generator, great care therefore must be exercised in selecting the construction materials and insulating medium for the generator.

The effect on the spectra when a neutron reflector is used above the neutron source was also studied. Inelastic spectra obtained with and without a 4 cm thick neutron reflector were virtually identical. Therefore, the use of a neutron reflector would have little or no effect on the inelastic portion of the CPNE. Similarly, the presence of a neutron reflector has little or no observable effect on the activation spectra. On the other hand a 4 cm thick reflector enhances the capture gamma ray intensity by more than a factor of two compared to the intensity measured using no reflector. For common earth type rock samples (i.e., basalt, dunite, granite) which contain

several tenths of a percent water, no new capture gamma-ray peaks occur when a reflector is used. For dry samples, however, many capture gamma-ray peaks are observable only when a reflector is used. This is discussed in more detail in Section 3.5.

3.4 Optimization Studies

Another objective of the present investigation was the optimization of the experimental parameters to enable the CPNE to obtain the maximum information in the shortest period of time. Experimental parameters such as the neutron pulse rate, sample gate times, neutron output, and sample count times are variable and subject to optimization. In addition, uncontrolable parameters such as the background (both from natural activity and from the radiothermal generator) will affect the optimum values of the experimental parameters.

Although the best way to evaluate and optimize the experimental parameters would be to collect a series of experimental spectra for various values of the parameters, such an approach would be prohibitively costly and time consuming. Fortunately, it is possible with appropriate computer programs to simulate the experimental data sufficiently well to perform the parametric studies without actual experimentation.

The quality of the results obtained in analyzing gamma-ray spectra will depend on the intensity of the spectrum and the intensity of any interferences present. The CPNE is designed to obtain 4 gamma-ray spectra: the capture, activation, inelastic, and natural background spectra. The natural radioactivity or background spectrum can be determined without interference by counting prior to activating the neutron generator. The inelastic spectrum can be obtained without interference (except for a negligible contribution due to capture, activation, and natural background) by sampling only during the neutron bursts. Similarly, the activation spectrum can be determined without interference (except for natural or other continuous radiation) by sampling just prior to each neutron burst. The capture gamma-ray spectrum, on the other hand, cannot be completely separated from the activation radiation. Experimental parameters which tend to maximize the number of capture gamma rays collected and minimize the number of activation gamma rays must be determined.

Experimental parameters which influence the capture gamma-ray intensity include neutron intensity, gate times, neutron pulse rate and geometry. Other parameters such as the ratio of the effective number of capture gamma rays to background gamma rays produced per neutron pulse and the thermal neutron half-life are determined by the sample material and cannot be veried. Each of the variable parameters mentioned above (except geometry which was investigated experimentally) was studied individually using a computer program called TIME. TIME calculates, by statistical means, the number of capture gamma rays and background (activation plus natural activity plus activity from the radiothermal generator) gamma rays expected to be detected as a function of gate times, pulse rate of the neutron generator, neutron intensity, thermal neutron half-life, analyzer dead time, and the ratio of the effective number of capture gamma rays to background gamma rays produced per neutron pulse.

In order to compare the results obtained using different parametric values, a figure of merit is needed which provides a means of qualitatively observing the sensitivity as a function of the parameters. The ability to detect a gamma-ray peak in a spectrum is proportional to the ratio of the counts in the peak to the square root of the total counts. We defined a sensitivity parameter as the counts of interest (i.e., the capture gamma-ray counts) divided by the square root of the sum of the capture gamma-ray counts and the background counts. This sensitivity parameter was used to optimize the various experimental parameters, i.e., the neutron intensity, gate times, and neutron pulse rate.

Theoretically, the neutron intensity is variable and limited only by the neutron generator. However, in practice, it is limited by the electronics of the gamma-ray detecting system. The determining factors are the maximum count rate which can be processed by the electronics and the average analyzer dead time. The results from TIME indicate that if the

average analyzer dead time is 40 μsec (the average dead time of the Nuclear Data Model 130 pulse height analyzer determined during earlier experiments), neutrons are wasted for intensities above 10,000 neutrons per pulse. However, TIME also indicates that the data acquisition efficiency (i.e., the number of gamma rays detected per pulse) increases more rapidly than the neutron efficiency decreases.

The electronics currently being used can respond correctly to gamma-ray count rates equivalent to a neutron intensity of 22,000 neutrons per pulse. Under these conditions, we are able to obtain the same number of counts in about one-fifth the time while using only about 15 percent more neutrons than in previous experiments (on the Mobil samples). The dead time of the analyzer to be used for the CPNE is not yet known and count rates due to natural activity and the radiothermal generator have not yet been estimated. Thus, the neutron intensity cannot be finally determined at this time.

The gate times are parameters which must be optimized to enable the capture gamma-ray portion of the CPNE to obtain the maximum information from the sample being analyzed. Results from TIME indicate that the capture gamma-ray sensitivity parameter decreases as the delay before the gate opening for the capture is delayed. This was expected because the capture gamma-ray intensity decreases with time after the neutron pulse (see Figure 1). The capture gamma-ray gate should be opened as soon as possible after the neutron burst, consistent with the requirement that no inelastic gamma rays should be observed.

Figure 5 shows how the capture gamma-ray sensitivity parameter varies as a function of the length of the capture gate. The opening of the gate was set at 20 μsec after the neutron burst. A neutron pulse rate of 500 pps was assumed, together with an intensity of 3300 neutrons/pulse and a basalt sample. No neutron reflector was assumed. The curve indicates that the optimum capture sensitivity is obtained with a gate

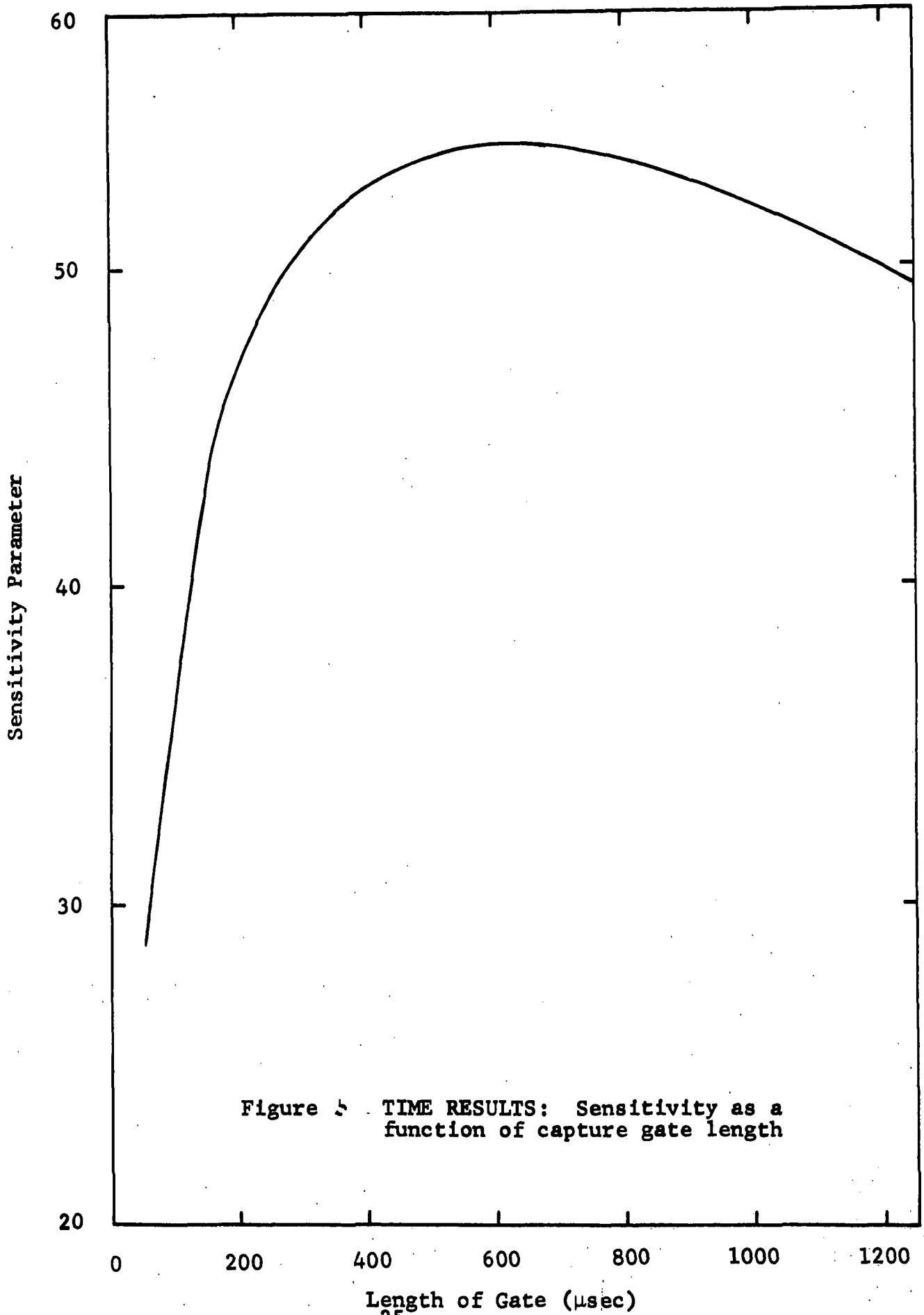


Figure 5 TIME RESULTS: Sensitivity as a function of capture gate length

length between 400 and 800 μsec. The precise value of the gate length is not critical in this interval.

It was also determined that the optimum activation sensitivity is obtained with a gate length of about 1600 μsec. Since a gate length of this magnitude would allow a significant number of capture gamma rays in the spectrum, thereby increasing the complexity of the analysis, it is unlikely that it would be used. A gate length of 1200 μsec would be better since the number of capture gamma rays detected is much smaller and the activation sensitivity parameter suffers only about a 10 percent decrease from its value for a gate length of 1600 μsec.

The results from these studies of gate length indicate that for a 500 pps neutron pulse rate and a basalt sample, appropriate gates would be 20 to 520 μsec for capture and 780 to 1980 μsec for activation.

The background from natural activity and from the radiothermal generator will affect the optimum values of the experimental parameters because the gamma count rate and the ratio of the effective number of capture gamma rays to background gamma rays produced per neutron pulse will change. Figure 6 contains curves which show how the capture sensitivity parameter varies with the length of the capture gate and the background count rate. The curves are for background count rates of 0, 361, 1986, and 5054 counts per second. The conditions were a 500 pps neutron pulse rate, a neutron intensity of 3300 n/pulse, and a capture gate opening at 20 μsec after the burst on a basalt sample with no reflector. The curves show that as the background intensity increases the capture sensitivity parameter decreases as does the optimum capture gate length. It is obvious from these results that the final selection of gate length cannot be made until the background intensity is known (or estimated satisfactorily).

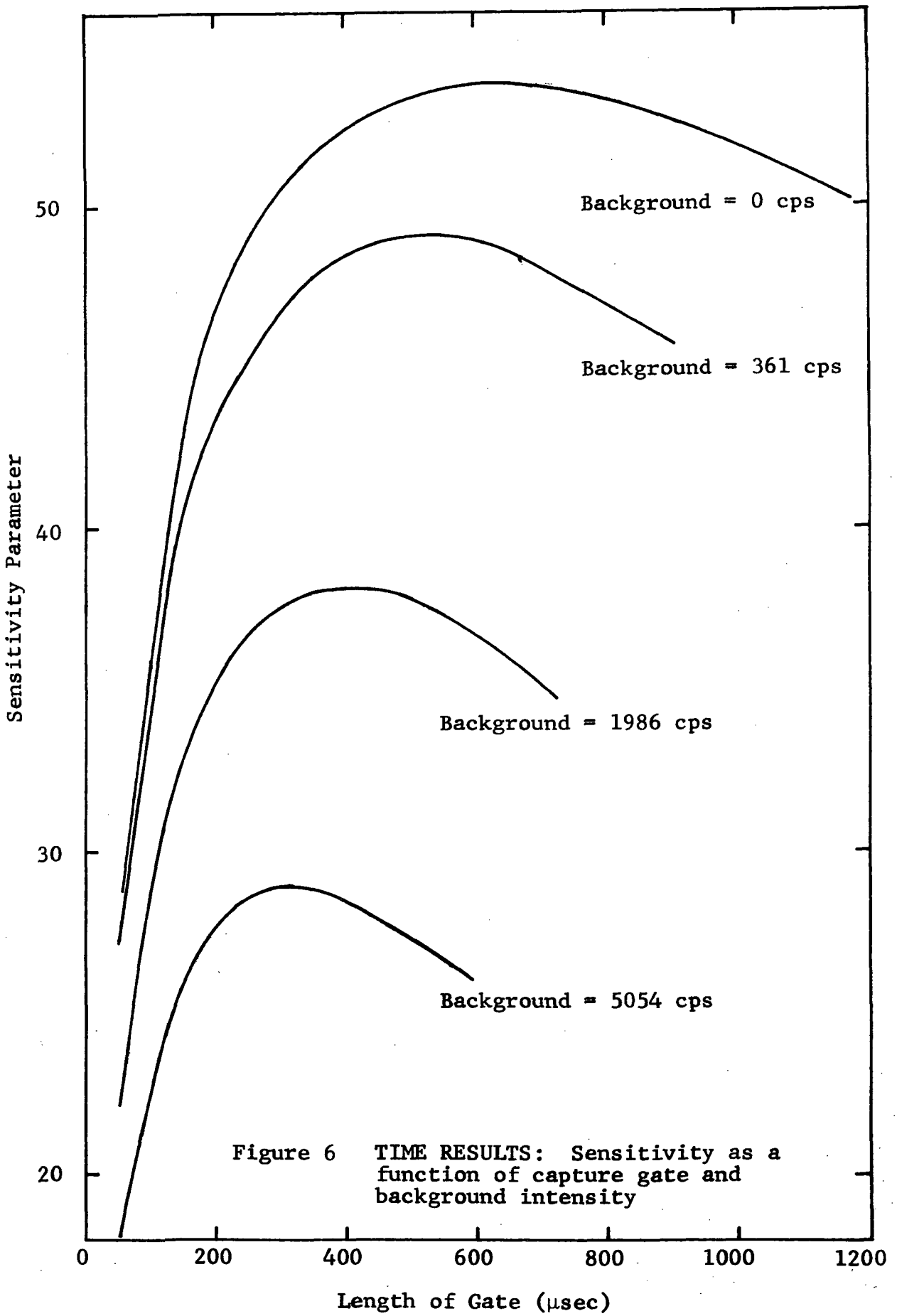


Figure 6 TIME RESULTS: Sensitivity as a function of capture gate and background intensity

The neutron pulse rate is another parameter which can be optimized for the capture sensitivity parameter. Results from TIME indicate that for a given total neutron output the capture sensitivity parameter decreases about 10 percent when the neutron pulse rate is raised from 500 pps to 1000 pps. This is accompanied by a factor of two reduction in the required counting time. As a result, for equal counting times, the capture sensitivity parameter actually increases by about 30 percent when the pulse rate is increased to 1000 pps (see Table 2).

In order to give the sensitivity parameter some quantitative significance, a study was carried out in which spectra were simulated using capture and background count rates predicted by TIME (see Section 3.5 for a discussion of spectrum simulation). These spectra were subsequently analyzed using ALPHA-M, and the calculated compositions and errors were analyzed and compared with the sensitivity parameter.

Spectra for all the cases shown in Table 2 (capture sensitivity parameters ranging between 44 and 112) were simulated. Several sets of random statistics were added to each spectrum and analysis was then accomplished using ALPHA-M. The results indicate that within the investigated range of the capture sensitivity parameter (i.e., 44 to 112) the fractional errors in the determinations of the elements varied approximately inversely with the sensitivity parameter. Thus, if the sensitivity parameter is doubled, the fractional errors in the determinations of the elemental concentrations will be approximately halved.

Since we have shown that the fractional errors in the measured elemental concentrations are approximately inversely proportional to the sensitivity parameter, we can use the sensitivity parameter to optimize the experimental parameters. First, we desire about a 4 cm neutron reflector above the neutron source, for the omission of a reflector is detrimental to the capture gamma-ray portion of the CPNE (see Section 3.6).

TABLE 2

COMPARISON OF EXPERIMENTAL PARAMETERS
FOR CAPTURE PORTION OF THE CPNE

Pulse Rate (pps)	Neutron Intensity (n/pulse)	Gate (μsec)	Sensitivity Parameter
500	3,900	20-520	44
500	7,800	20-520	60
500	11,800	20-520	74
500	15,600	20-520	80
500	18,000	20-520	85
500	18,000	20-120	57
500	18,000	20-320	79
500	18,000	20-520	85
500	18,000	20-720	86
500	18,000	20-920	84
500	18,000	20-1420	80
500	18,000	20-1920	74
250	18,000	20-320	59
250	18,000	20-520	64
250	18,000	20-720	65
1000	18,000	20-120	76
1000	18,000	20-320	104
1000	18,000	20-520	112
1000	18,000	20-720	108
1000	18,000	20-920	105
2000	18,000	20-120	101

Secondly, the neutron intensity should be the maximum consistent with the capabilities of the electronics and the analyzer (i.e., the dead time restrictions governed by the analyzer).

Table 2, which shows how the capture sensitivity parameter varies with neutron pulse rate, neutron intensity, and gate times, was used as an aid in determining the optimum values of the experimental parameters. The information in Table 2 was based on the assumptions that the sample was basalt and that a 4 cm thick neutron reflector was used together with a 1 hour count time. Neutron intensities only up to 18,000 neutrons/pulse were considered, for this intensity gives the highest gamma-ray count rate our present electronics can handle (without a reflector, 22,000 neutrons/pulse will give this maximum gamma-ray count rate).

Using the results from Table 2 we concluded that for either a 500 pps or 1000 pps pulse rate, the optimum capture gate would be about 20-520 pps. Moreover, the neutron intensity would be the one producing the maximum gamma-ray count rate which the electronics can properly handle. The only experimental parameter which has not yet been finally specified is the pulse rate. Although the sensitivity parameter indicates that the 1000 pps pulse rate is better for statistical reasons, the 1000 pps pulse rate may not be best for the CPNE. One of the objectives of the CPNE is the separation of the various radiations produced. With a 500 pps pulse rate, the activation spectrum will contain only a small capture component (less than about 10 percent). With a 1000 pps pulse rate, however, the activation spectrum will contain about a 20 percent contribution due to capture gamma rays. This increased capture contribution (or background) may make the detection of weak activation components more difficult. This becomes more important if the thermal neutron half-life in the sample is longer than that for basalt with 0.25 percent hydrogen.

One additional experimental parameter which must be specified is the sample count time. It is desirable to use as short a count time as possible consistent with obtaining a spectrum containing adequate statistics. As an aid in specifying the count time, we carried out an investigation of the effects of statistics. We wanted to discover how the intensity of the spectrum affects the accuracy of the analysis and how random statistics affect the results for a given intensity. We generated capture plus activation spectra from basalt with intensities of 1, 2, and 4 times the intensity of the experimental spectrum obtained from the Mobil model. Based on the intensity of each spectrum, random statistics were added to each. This was done seven independent times and the resulting 21 spectra were then analyzed using ALPHA-M.

Table 3 contains the results for a spectral intensity equal to the experimental values and shows how random statistics affect the measurement of the elemental concentrations. Examination of Table 3 indicates that random statistics can have a great effect on the accuracy of the determined intensities. For example, the fractional error for the determined intensity of silicon is 0.11 or less for 6 of the 7 determinations. For one case, however, it is 0.20 which is almost double the maximum for the other 6 cases. Similar occurrences appear in the results for the other elements.

The R.M.S. value of the family of determinations was selected as an appropriate measure of the fractional error in the determined elemental concentration. A comparison of the R.M.S. values for the elements (see Table 4) with the individual errors (Table 3) indicates that generally the individual error falls near or less than the R.M.S. value. The individual error greatly exceeds (by about a factor of 2) the R.M.S. value only about once in seven determinations. Furthermore, we found that, on the basis of the analysis of 21 spectra with different

TABLE 3

FRACTIONAL ERRORS IN MEASURED ELEMENTAL CONCENTRATIONS

Element	Fractional Error in Spectrum Number						
	1	2	3	4	5	6	7
Si	0.20	-0.067	-0.034	-0.017	-0.028	0.061	-0.11
Fe	0.048	0.056	0.017	0.0010	0.028	-0.011	0.0041
Al	0.31	-0.18	0.23	0.36	-0.13	0.028	-0.13
Ca	0.11	0.081	-0.029	-0.031	-0.058	-0.074	0.020
Mg	-0.89	0.026	-0.11	-0.22	0.22	-0.063	0.34
Na	0.11	0.084	-0.059	0.0099	0.030	-0.11	-0.18
K	0.019	0.49	0.093	-0.081	-0.19	0.14	0.075
Ti	0.071	0.036	0.010	-0.0051	0.010	0.025	-0.015
H	0.13	-0.18	-0.084	0.012	0.13	-0.020	-0.084

TABLE 4

COMPARISON OF ERRORS IN MEASURED CONCENTRATIONS
AS A FUNCTION OF SPECTRAL INTENSITY

	R.M.S. Fractional Error for Intensity		
Element	x1	x2	x3
Si	0.095	0.062	0.017
Fe	0.031	0.043	0.014
Al	0.22	0.13	0.040
Ca	0.065	0.070	0.043
Mg	0.38	0.20	0.090
Na	0.099	0.11	0.085
K	0.21	0.17	0.090
Ti	0.033	0.026	0.018
H	0.11	0.037	0.044

random statistics, the R.M.S. value closely approximates the standard deviation for a gaussian distribution. We, therefore, will use the R.M.S. value as the value of the standard deviation for our determinations of the concentration of an element in a sample.

Table 4 contains the R.M.S. values of the fractional error in elemental abundance as determined for several values of spectral intensity. In general, the accuracy of the measurements improves with increasing spectral intensity.

Table 5 shows how the time required to attain a capture sensitivity parameter equal to 72 varies as a function of pulse rate and gate times. A spectrum whose capture sensitivity parameter is 72 will have about the same statistics as the x4 results in Table 4. The times listed in Table 5 indicate that to attain the desired statistics a run should last 43 minutes with a 500 pps pulse rate or 25 minutes with a 1000 pps pulse rate. Therefore, if time is of the utmost importance, a 1000 pps pulse rate should be used. Otherwise, a 500 pps pulse rate would be better.

TABLE 5

COMPARISON OF EXPERIMENTAL PARAMETERS
FOR SENSITIVITY PARAMETER EQUAL TO 72

Pulse Rate (pps)	Neutron Intensity (n/pulse)	Gate (μsec)	Time (hour)
500	18,000	20-520	0.71
500	18,000	20-720	0.70
500	18,000	20-920	0.73
1000	18,000	20-320	0.47
1000	18,000	20-520	0.41
1000	18,000	20-720	0.45
1000	18,000	20-920	0.47

3.5 Spectrum Simulation

Our goal in these studies was to adequately simulate a complex gamma-ray spectrum from a bulk sample. Elemental capture gamma-ray spectra from small samples are available in Greenwood and Reed's[10] atlas of thermal neutron capture gamma-ray spectra. We accomplished our goal by first forming composite capture gamma-ray spectra for basalt, dunite, and granite from elemental spectra, and then determining a formula (or mathematical filter) which adequately transformed the composite small-sample spectra into spectra that closely resemble the CPNE experimental data.

Three mechanisms must be considered when spectra from bulk samples are simulated. These are smearing, attenuation, and build-up. The gamma-ray peaks suffer a loss in resolution (smearing) due to small angle scattering of the gamma rays in the sample. The gamma rays also are attenuated while traversing the sample. Finally, the lower energy portions of the spectrum exhibit build-up due to high energy gamma rays being scattered into the detector after undergoing a Compton collision. Formulas to simulate all three of these mechanisms were tested on the composite small-sample thermal neutron spectra to determine their effects. They were then combined to transform the initial data to spectra which resembled the CPNE data. The spectra from basalt, dunite, and granite obtained experimentally from the Mobil samples were used as the standard CPNE spectra in the simulation studies.

A normal attenuation function, $e^{-\rho x \sigma_t(E)}$, where ρ is the density of the sample, x is the average sample thickness traversed by the gamma rays (assumed to be 5 cm), and $\sigma_t(E)$ is the total attenuation cross section for gamma rays of energy E, was used to attenuate the gamma rays. The function which was found to best simulate the build-up in the spectrum is

$$C(I) = 0.25 \sum_{J=1}^{I} C_o(J) \left(1-e^{-\rho x \sigma_s(J)}\right),$$

where C(I) is the counts in channel I due to build-up, $C_o(J)$ is the initial number of counts in channel J, $\sigma_s(J)$ is the scattering cross section for gamma rays whose energy corresponds to channel J, and 0.25 is a normalization factor.

The usual gaussian smearing function was used to smear the data

$$S(E) = \frac{1}{C\sqrt{\pi}} \sum_{E'=0} F(E')\, e^{-(E-E')^2/C^2},$$

where S(E) is the smeared function, F(E) is the initial function, and $C = \sqrt{b^2-a^2}$ where a is the width of the initial gaussian and b is the desired width of the smeared gaussian function.

After undergoing smearing, attenuation, and build-up, each simulated spectrum was normalized to the number of counts contained in its corresponding experimental standard spectrum. The resulting spectra were simulations (without statistics) of only the capture gamma-ray spectra from the CPNE.

In addition to capture gamma rays spectra our library of simulated spectra requires components for the activation spectra from silicon and from oxygen. We obtained these spectra from experimental data using the following procedure. An activation spectrum from sand contains components from silicon, oxygen, detector activation, and natural background. Since the contribution to the activation spectrum from our sand sample due to natural background is small, it could be neglected. The contribution due to detector background, however, must be included. Since the detector activation is relatively long lived (it is due to 25 minute iodine-128) we could get an essentially pure spectrum by running the CPNE until equilibrium conditions were

attained, turning off the neutron source, waiting until the aluminum-28 (from activation of silicon) had decayed away (about 15 minutes), and then counting. This gives a spectrum of the detector activation with only a minor contribution due to natural background.

In order to obtain an activation spectrum from silicon we again ran the CPNE until equilibrium conditions were attained and then counted the aluminum-28 activity. After summing several of these spectra to get better statistics, we stripped out the crystal activation contribution, thereby obtaining an activation spectrum from silicon. Stripping the silicon and crystal activation contributions from the activation spectrum from sand yielded the pure oxygen activation spectrum. Finally, the three activation component spectra (silicon, oxygen, crystal activation) were smoothed.

Simulated capture plus activation spectra for basalt, dunite, and granite were generated by first forming a composite activation spectrum using our simulated components, adding the simulated capture and simulated activation spectra together, and then adding random statistics. Figures 7, 8, and 9 show plots of the resulting simulated capture plus activation spectra together with their corresponding counterparts.

An examination of Figures 7, 8, and 9 indicates that, in general, the simulated spectra give adequate approximations to the experimental spectra. The very low energy portions, however, are slightly underestimated. The only other discrepancies are that the titanium contribution is overestimated by approximately a factor of two in the simulated basalt spectrum and the hydrogen contribution is underestimated in all the simulated spectra. The titanium discrepancy may be due to either an erroneous measured value (the measured titanium content of the basalt is about double that for an average basalt rock) or an erroneous parametric value reported in Greenwood and Reed's atlas.

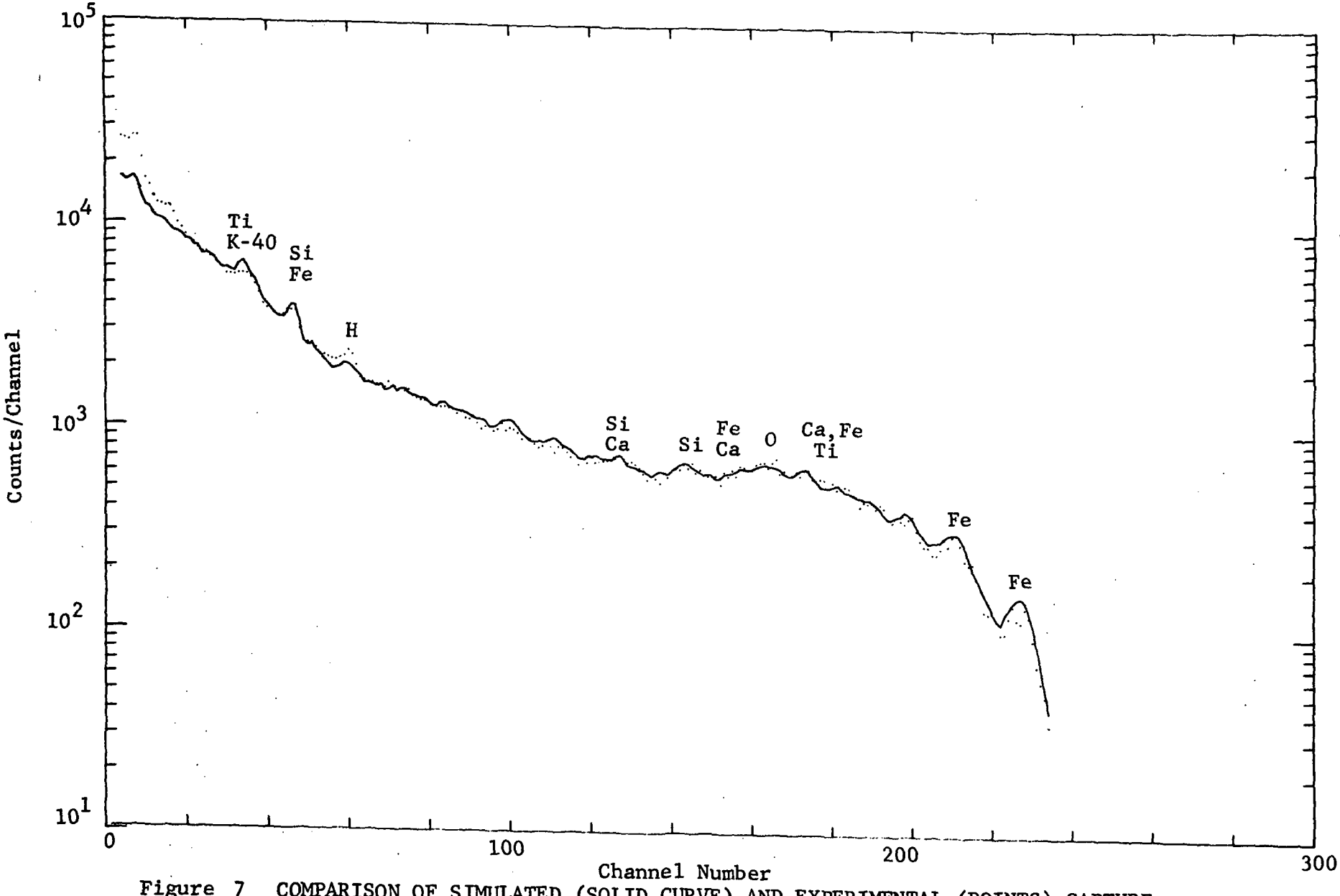


Figure 7 COMPARISON OF SIMULATED (SOLID CURVE) AND EXPERIMENTAL (POINTS) CAPTURE PLUS ACTIVATION SPECTRUM FROM BASALT

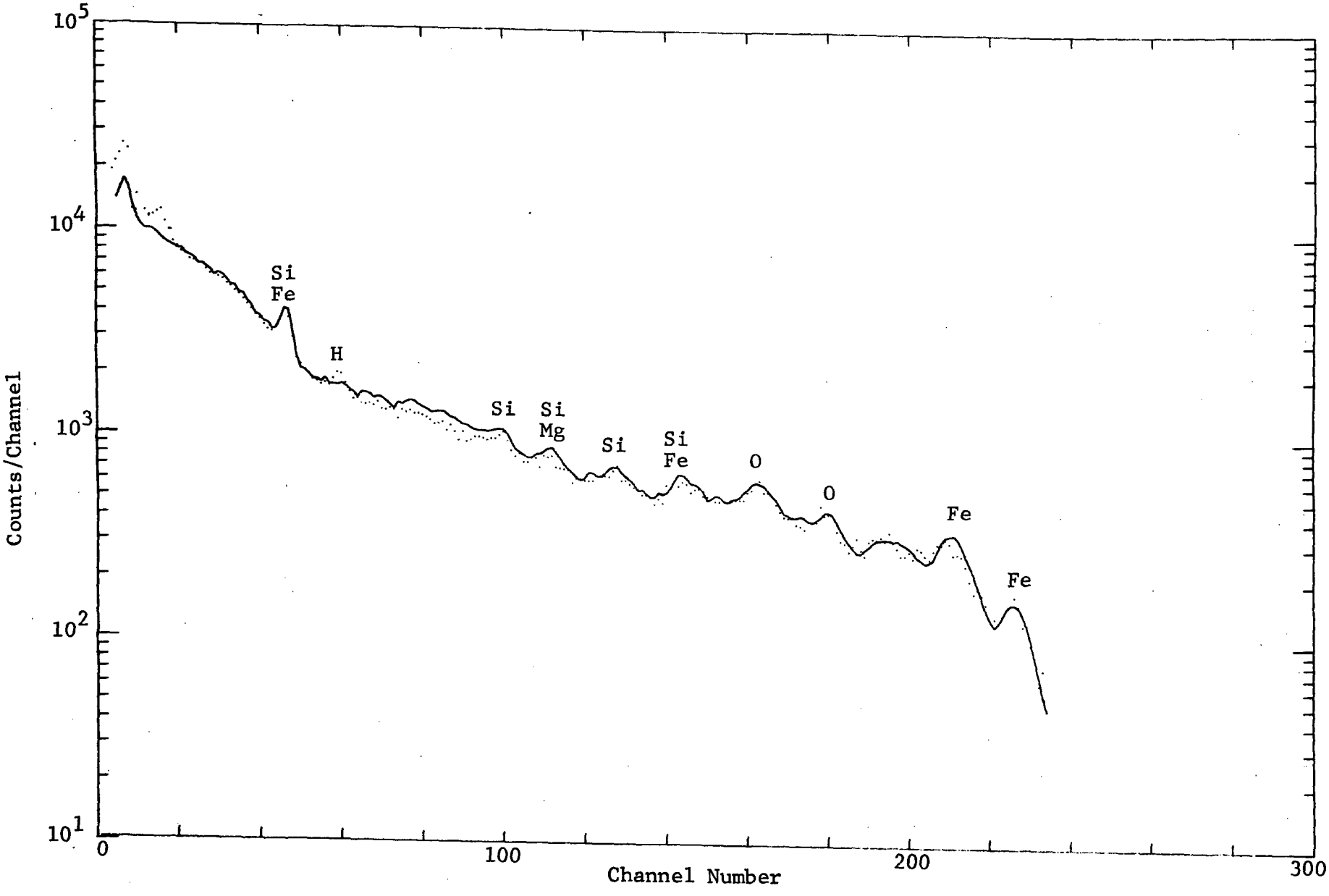


Figure 8 COMPARISON OF SIMULATED (SOLID CURVE) AND EXPERIMENTAL (POINTS) CAPTURE PLUS ACTIVATION SPECTRUM FROM DUNITE

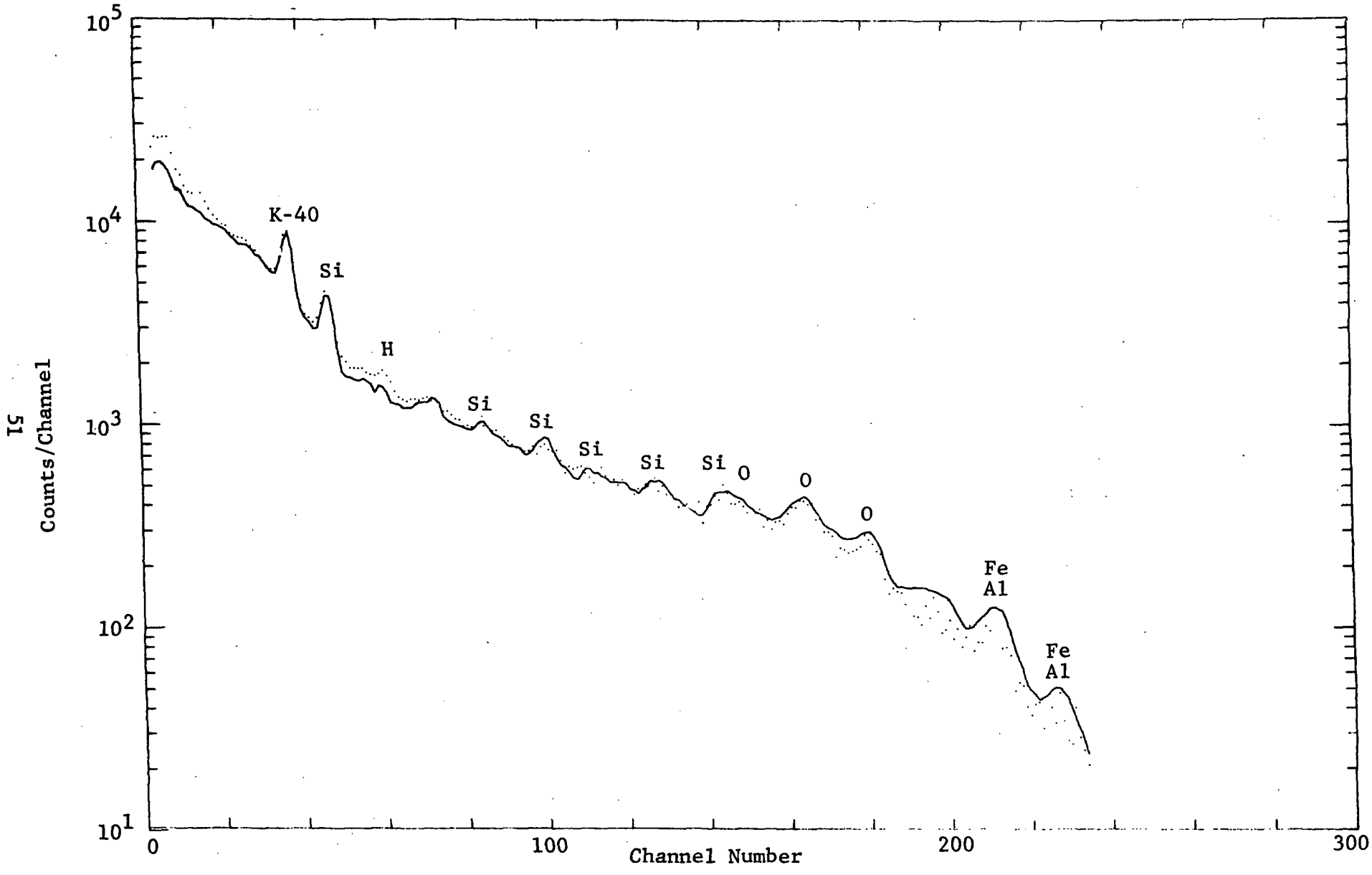


Figure 9 COMPARISON OF SIMULATED (SOLID CURVE) AND EXPERIMENTAL (POINTS) CAPTURE PLUS ACTIVATION SPECTRUM FROM GRANITE

Since the simulated spectra approximate the experimental spectra from the CPNE, we concluded that our method of spectrum simulation is adequate. We therefore transformed elemental small-sample capture gamma-ray spectra into simulated bulk sample capture gamma-ray spectra. To this collection of spectra we added our simulated activation spectra, (using ratios predicted by TIME) forming a complete library of simulated spectra for the capture and activation portions of the CPNE.

3.6 Collection of Library Spectra

A library of component capture gamma-ray, activation, and inelastic scattering spectra for silicon, calcium, iron, and titanium is needed for the analysis of spectra via the "library" method. Oxides of the elements (i.e., SiO_2, CaO, Fe_2O_3, and TiO_2) are required for these studies. Since it is important that the neutron flux distribution be the same within each sample, it was decided to use silicon dioxide as a matrix material in which each of the other three oxides are separately diluted. In this way the densities of the samples can be made comparable. Since the neutron distribution within a sample is controlled by the density, the neutron distributions will also be comparable as a consequence.

Previously, a sample size of 152 cm x 152 cm x 76 cm was used for the CPNE studies. This large volume would have required large amounts (about 2000 kg) of the various oxides to be used. Since some of the oxides are quite expensive, we wished to minimize the amounts of oxides used. This was accomplished by using an insert of the sand-oxide mixture in a larger sand matrix.

In order to properly specify the sample size, a knowledge of the active volume sampled by the CPNE is needed. A computer program, called GAMMAP, was written and implemented to provide this information. Given the neutron flux distribution and the gamma-ray energy, GAMMAP calculates for each incremental volume element within the sample the number of gamma rays of the given energy which strike the detector (i.e., the number of gamma rays which are emitted in the direction of the detector and reach the detector without being scattered by the sample itself or the fast neutron shadow shield). These results can then be intergrated to delineate the volumes from which any chosen fraction of the detected gamma rays originate.

Calculations were performed for a basalt sample with a bulk density of 1.75 gm/cm^3 and the thermal neutron flux distribution reported in Reference 11. The probe geometry assumed a 7.6 cm x 7.6 cm NaI(Tl) detector, a 15.2 cm long shadow shield (of molybdenum or beryllium), a 5.1 cm gap between the shadow shield and detector, and a 2.54 cm gap between the shadow shield and the neutron source. The neutron source was fixed at 5 cm above the sample surface and calculations were performed for cases where the angle between the probe axis and the sample surface was 0°, 30°, 60°, and 90°. Results for the 0° case (probe axis parallel to the sample surface) and a gamma energy of 6 MeV indicate that if no reflector is used 50 percent of the detected gamma rays have to traverse at least 50 cm of sample material. When a 4 cm thick neutron reflector is used the sample thickness traversed by 50 percent of the gamma-rays is reduced to 13 cm.

On the basis of these calculations, we fabricated a thin-walled steel cylinder 120 cm in diameter and 61 cm deep. This cylinder was used as an insert in a large sand matrix with the sample material to be studied placed in the insert. The calculations indicate that 85 to 90 percent of the detected capture gamma rays (for a terrestrial rock such as basalt with 0.25 percent hydrogen) should originate in this volume. To verify that the gamma rays being detected were indeed coming from this volume, the sand matrix was replaced by basalt, and sand was used as the sample in the insert. Capture gamma-ray spectra obtained from this sample exhibited only gamma rays from silicon and oxygen, indicating that most of the detected gamma rays came from the material in the insert.

Samples consisting of sand-oxide mixtures were obtained for use in the collection of the library spectra. The compositions of these samples is shown in Table 6.

TABLE 6

COMPOSITION OF LIBRARY SAMPLES

Library Component	Wt. % of Desired Oxide	Wt. % of SiO_2 Matrix	Captures Due to Desired Oxide
SiO_2	100	---	100%
Fe_2O_3	10	90	58%
MgO	35	65	23.5%
Al_2O_3	38	62	51%
CaO	28	72	52%
TiO_2	8.3	91.7	72%

Great care was exercised during the gathering of the library spectra to assure reproducible geometry, adequate statistics, and a known relative neutron output. Conditions selected for obtaining the inelastic spectra were: (1) a neutron pulse of approximately 15 μsec duration, (2) a 20 μsec long gate straddling the neutron burst, (3) a pulse rate of 5000 pps, and (4) a neutron intensity of 350 neutrons/pulse. No neutron reflector was used in the collection of inelastic spectra.

Capture and activation library spectra were obtained both with and without a 4-cm neutron reflector. Conditions used for obtaining the capture and activation spectra were: (1) a neutron pulse of approximately 15 μsec duration, (2) a 50-550 μsec capture gate (relative to the beginning of the neutron pulse) and a 1350-1850 μsec activation gate, (3) a pulse rate of 500 pps, and (4) a neutron intensity of 18,000 neutrons/pulse when the reflector was used and 22,000 neutrons/pulse when the reflector was not present.

Figure 10 shows the inelastic spectrum from SiO_2. Approximately 3×10^{10} neutrons were required to obtain this spectrum. The observed instantaneous gamma counting rates observed during the acquisition of this spectrum are tabulated in Table 7.

Figure 11 shows the capture plus activation spectrum from SiO_2 obtained without a reflector. Approximately 1.2×10^{11} neutrons were required to obtain this spectrum. Only weak capture gamma-ray peaks are evident in this spectrum. Figure 12 shows the capture plus activation spectrum from SiO_2 obtained using a 4 cm reflector. To obtain better statistics, the data collection time was approximately doubled (compared to that for Figure 11 and hence about 2.1×10^{11} total neutrons were required. Capture gamma-ray peaks are much more prominent in this spectrum. The observed instantaneous gamma-ray counting rates present during the collection of the capture spectra are listed in Table 8.

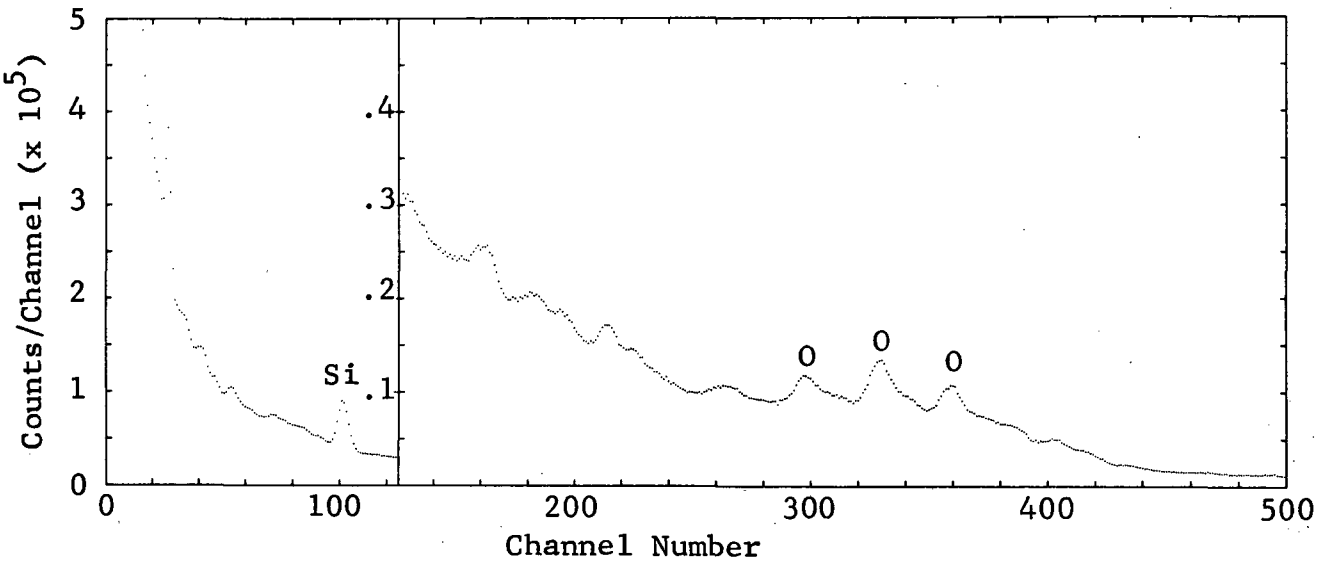


Figure 10 INELASTIC SPECTRUM FROM SAND OBTAINED USING 7.6 cm x 7.6 cm NaI(Tl) DETECTOR AND MOLYBDENUM SHIELD

TABLE 7

INSTANTANEOUS COUNT RATES FOR INELASTIC SCATTERING DATA

Time Interval (μ/sec)	Instantaneous Count Rate (c/sec)
0*- 5	200
5 - 10	11,400
10 - 15	27,400
15 - 20	13,400
20 - 25	32

* t = 0 at start of neutron burst.

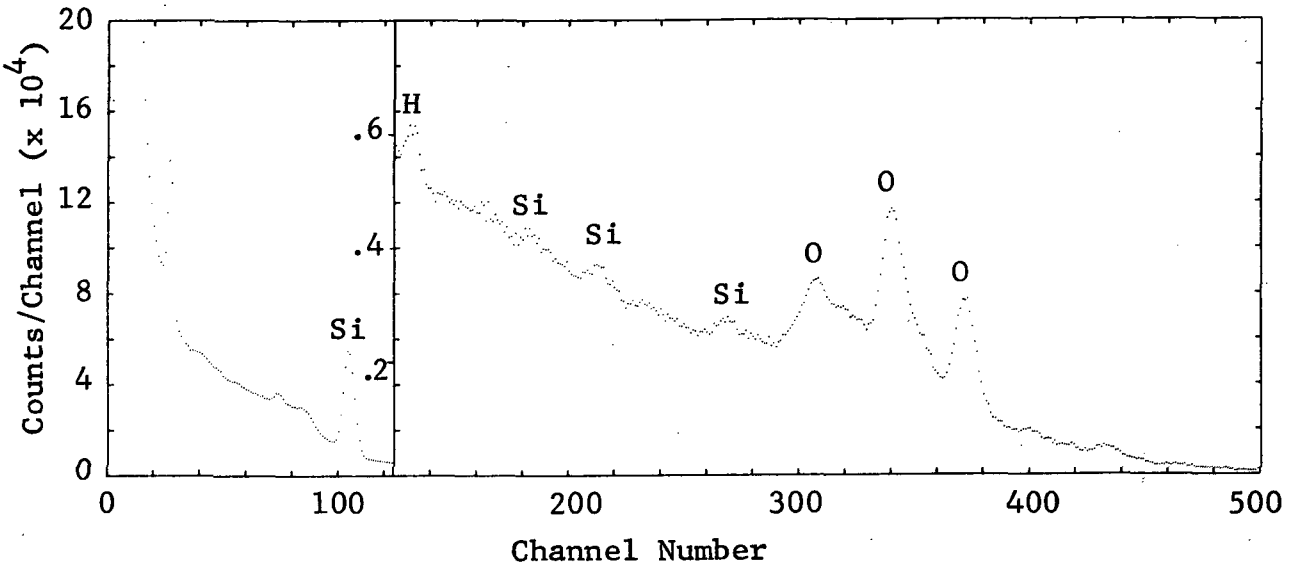


Figure 11 CAPTURE PLUS ACTIVATION SPECTRUM FROM SAND OBTAINED USING 7.6 cm x 7.6 cm DETECTOR WITHOUT A REFLECTOR

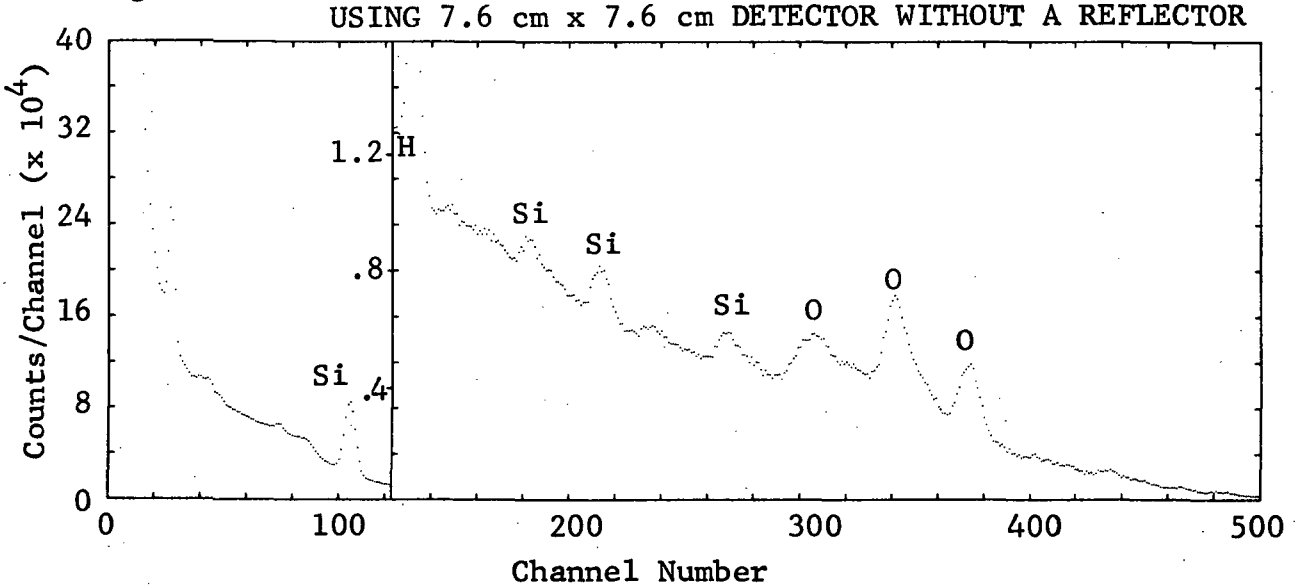


Figure 12 CAPTURE PLUS ACTIVATION SPECTRUM FROM SAND OBTAINED USING 7.6 cm x 7.6 cm NaI(Tl) DETECTOR WITH 4 cm REFLECTOR

TABLE 8

INSTANTANEOUS COUNT RATES FOR RADIATIVE CAPTURE DATA*

Time Interval (μsec)**	Instantaneous Count Rates (c/sec)
0 - 20	577,000
30 - 50	5,270
50 - 90	4,475
90 - 130	4,010
130 - 1930	3,980

* SiO_2 samples, 500 neutron bursts per second

** t = 0 at start of neutron burst

A comparison of Figures 11 and 12 indicates the need for a neutron reflector in the collection of capture gamma-ray spectra from a completely dry sample. Attempts to obtain the capture plus activation spectra from the TiO_2 - SiO_2 and CaO - SiO_2 samples reinforced this conclusion. For both TiO_2 - SiO_2 and CaO - SiO_2, capture gamma ray peaks from Ti and Ca, respectively, were masked by the silicon and oxygen peaks when no reflector was used. With a reflector weak capture gamma-ray peaks from Ti and Ca were evident in the respective spectra; however, these peaks were not of high enough intensity to enable a satisfactory stripping of the SiO_2 component. Adequate library of component spectra could not be obtained.

Previous experiments showed that satisfactory capture gamma-ray spectra can be obtained from basalt, dunite, and granite without a reflector. Since the conditions for the collection of the library spectra were optimized for capture gamma-rays better than the previous experiments (e.g., the gate opening was sooner and the gate length was optimized using the results of our parametric studies), we concluded that the experimental conditions were not the cause of the poor quality capture gamma-ray spectra. We suspect that the actual cause was the lack of water in the samples. The samples we obtained for the library studies were kiln dried and contained essentially no water while the basalt, dunite, and granite samples used previously contained 0.25, 0.059, and 0.054 weight percent of hydrogen, respectively.

Mobil[11] has measured the profile of the thermal flux distribution in both dry sand and sand containing 0.2 weight percent hydrogen. They found that the peak in the thermal flux distribution occurs at a depth of about 37 cm in the sample containing 0.2 percent hydrogen, but in the dry sample the depth of the peak is increased to about 66 cm. Moreover, they found that for a dry sample, a model whose radius is 70 cm is no longer a good approximation to a semi-infinite model. Fewer neutrons are

thermalized in the inadequately small model. The intensity of the thermal flux in the small model will be only about one sixth the intensity in a semi-infinite model.

Using the thermal flux distribution measured by Mobil, we calculated (using GAMMAP) that for a dry sample 50 percent of the detected 6 MeV capture gamma rays originate at a radius greater than 55 cm. For a sample containing 0.2 weight percent hydrogen, 50 percent of these gamma rays originate within a radius of 37 cm. We also found that for a dry sample the intensity of the detected capture gamma-rays decreases by about a factor of 5 if the radius of the sample is decreased from infinity to 70 cm. This is due to a large leakage of neutrons out of the 70 cm radius sample. The leakage reduces the thermal flux in the sample volume by about a factor of 5 which consequently reduces the flux of capture gamma rays.

We conclude that the low thermal neutron flux caused by both the dryness of the samples and their inadequately small sizes prevented the collection of the required library spectra. Using a sample which adequately approximated a semi-infinite medium would have reduced the neutron leakage thereby increasing the capture gamma-ray flux by about a factor of 5. Similarly, a reflector would have increased the capture gamma-ray flux about a factor of 2 by increasing the thermal neutron flux near the surface of the sample. Greater care must be exercised in selecting the models to be used for the library. First, the samples should be as simple as feasible, for complex samples necessitate spectrum stripping and the larger errors introduced by stripping. Secondly, either a neutron reflector must be used or the hydrogen content of the samples should be high enough to ensure an adequate thermal flux within the sample. Thirdly, the models must be of sufficient size to adequately approximate a semi-infinite medium.

3.7 Questions Not Answered

As a result of the studies just concluded, the CPNE has reached a stage where the experimental parameters have been finalized, the design of the light weight probe completed, and detection thresholds predicted. The CPNE must now be field tested in the light weight probe configuration and the predicted sensitivities verified using a complex sample. In addition to the field test, additional work in the area of data analysis needs to be accomplished. This study should include the collection of a library of component spectra using the light weight probe and further investigations concerning the effects of density and hydrogen content on spectral analysis.

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