Californium-252 Neutron Capture and Decay Methods for Elemental Analysis

Final Report

to

National Aeronautics and Space Agency

from

U. S. Geological Survey

Washington, D. C. 20242

April 25, 1972

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INTRODUCTION

The following report covers the two year period from April 1970 to April 1972 during which this project was in force. The general objective of the project was to test the feasibility of using a ^{252}Cf neutron source in conjunction with a capture and/or decay gamma ray method for elemental analysis in lunar or planetary missions.

The project is a cooperative effort between the U. S. Geological Survey (F. Senftle) and Goddard Space Flight Center (J. Trombka). The work of each group was delineated so as to compliment each other. The primary responsibility of the USGS team was to work out the general problems of using a ^{252}Cf neutron source for both decay and capture gamma ray analysis in terrestrial environments. This work included the determination of the capture gamma ray spectra by neutron absorption in various metals used for the space hardware, ^{252}Cf source encapsulation materials, shielding, geometry, optimum source size for a space mission, etc. The responsibility of the GSFC team was to investigate the computer data reduction and data transmission techniques. The original plans allowed for some overlap of the primary missions of each team. The results of this investigation have led to the publication of eight scientific papers and a nineth paper is in preparation.

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PROJECT OBJECTIVES

The broad objective of the project is to evaluate the radiative neutron-capture gamma-ray and decay-gamma methods of neutron activation using a californium-252 neutron source as a feasible analytical technique to be used on lunar and/or planetary missions. To develop practical operating techniques basic information was lacking in a number of areas. It was therefore desired to:

(1) Determine optimum shielding and encapsulation materials for the californium source required for a planetary mission.

(2) Compare Ge(Li) and NaI(Tℓ) as detectors for capture gamma rays.

(3) Determine source-to-detector geometry for optimum operation.

(4) Determine the effect of the hydrogen concentration in the environment on the spectrum obtained.

(5) Develop computer data reduction systems to increase the sensitivity and obtain the best possible analytical data.

(6) Develop both NaI(Tℓ) and Ge(Li) probes for possible shallow borehole analysis below the planetary or lunar surface.

Over the past two years most of these objectives have been accomplished.

THE CAPTURE GAMMA RAY SPECTRUM

When an atomic nucleus absorbs a neutron, it is raised to an excited state. De-excitation generally takes place within a fraction

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of a millisecond by the emission of one or more radiative capture gamma rays which have energies up to the binding energy of the neutron, i.e. up to about 11 MeV. If the resulting nucleus is unstable, further emission of decay gamma rays will take place at a later time depending on the half-life of the nucleus. Both of these processes are shown in Figure 1. Conventional activation analysis uses the decay gamma rays. However, as the capture gamma rays are also diagnostic of the parent element, they also can be used for analytical purposes. Contrary to the radiative capture method, problems associated with using the decay gamma method are well known and will not be discussed further. Ideally, for a lunar or planetary mission it would be advantageous to use a gamma ray spectrometer in both the decay and capture gamma ray modes. Some elements are more easily detected by the decay method while others are more easily observed by the radiative capture method.

Comar et al[1], using a reactor source of neutrons and a Ge(Li) detector, have shown the advantages of using capture gamma rays for the analysis of biological samples, whereas Christell and Ljunggren[2] have used an isotopic source of neutrons with a NaI(Tℓ) detector to determine iron in ores. More recently, Senftle et al[3] and Wiggins et al[4] have used a ^{252}Cf as an isotopic source of neutrons with a Ge(Li) detector to analyze Ni and Ti respectively in ores by the capture γ ray method. In the latter four investigations, high energy capture gamma rays have been used to determine a given element. For space application it is desireable to use ^{252}Cf as a source of neutrons because of its small size and weight.

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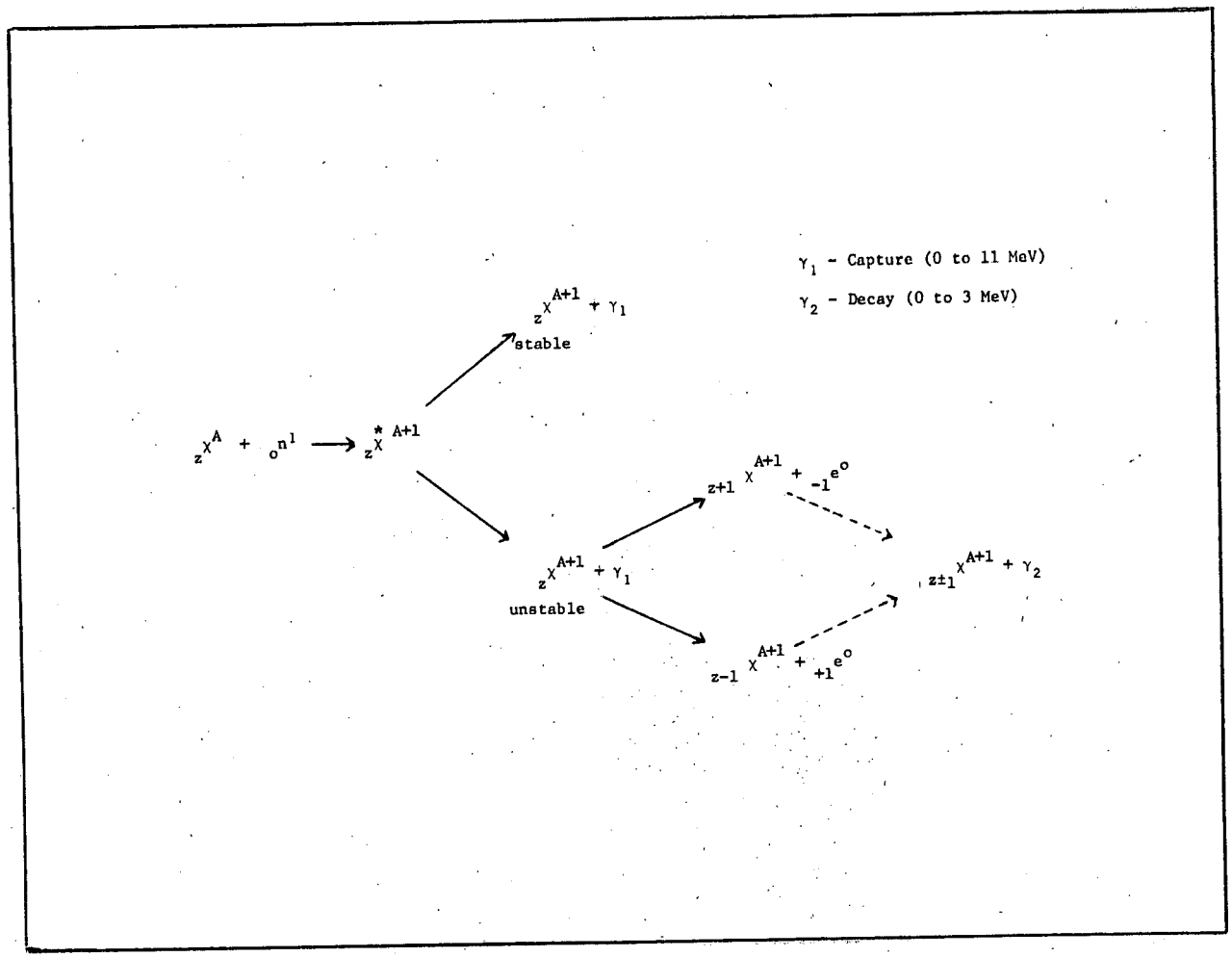
γ_1 - Capture (0 to 11 MeV)
γ_2 - Decay (0 to 3 MeV)
$_zX^A + {}_0n^1 \rightarrow {}_z^*X^{A+1}$
$_zX^{A+1} + \gamma_1$
stable
$_zX^{A+1} + \gamma_1$
unstable
$_{z+1}X^{A+1} + {}_{-1}e^0$
$_{z-1}X^{A+1} + {}_{+1}e^0$
$_{z\pm1}X^{A+1} + \gamma_2$

Figure 1

As in a reactor, radiative neutron-capture gamma rays can be produced with a ^{252}Cf neutron source by placing the target (sample) either close to the source (internal geometry) or in a neutron beam extracted from a massive shield surrounding the ^{252}Cf (external geometry). As pointed out by Garbrah and Whittey[5] the latter technique is generally most desirable when using a reactor. Practically, however, a ^{252}Cf source would have to be in the multigram weight range to furnish enough neutrons to produce a beam of sufficiently high flux for this purpose. ^{252}Cf sources of this size are not common and are difficult to manipulate safely except in special laboratory facilities. To use a smaller ^{252}Cf source of 500 μg or less, however, one must use an internal geometry because of the low initial neutron yield. Several variations of an internal geometry can be constructed, but, in general, the sample and ^{252}Cf source are placed in close proximity, and are surrounded with a moderator. The Ge(Li) detector is placed at some distance commensurate with the protection of the crystal from neutron radiation damage.

Under these conditions a number of background effects present themselves which interfere with the high-energy capture gamma-ray spectra:

(a) The material from which the ^{252}Cf capsule is fabricated emits a capture gamma-ray spectrum.

(b) The moderator surrounding the source and sample emits a characteristic capture gamma-ray spectrum.

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(c) Although in a relatively low neutron flux, the rather massive hardware associated with the detector will emit a characteristic capture gamma-ray spectrum.

(d) ^{252}Cf is a fission-type neutron source, and hence, there will be a substantial continuum underlying any capture gamma-ray spectrum.

By analogy with a reactor and as suggested by Hammermesh and Hummel[6] these background interferences can in theory be appropriately subtracted out. However, to do this one must count over long periods of time to obtain adequate counting statistics. Such long counting times are not always practical and thus, it is desirable to reduce these effects to a minimum by proper choice of materials, geometry, etc.

INTERFERENCES FROM SOURCE ENCAPSULATION MATERIALS

It is clear that the radiative capture gamma rays from the materials in the source capsule will be superimposed on the background spectra. Thus, the choice of encapsulation material will be important to minimize background interference from the source. Interference parameters for thirty-eight elements have been calculated (Senftle et al, ref. 7), most of which are important as construction materials. It was concluded that zirconium or a high-zirconium alloy would be most suitable for source encapsulation and would contribute least to the background. As a result of this study the U. S. Atomic Energy Commission was asked to supply two californium neutron sources of

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about 100 μg, one encapsulated in the usual stainless steel capsule, the other in a Zircaloy-2 capsule. The two sources were carefully compared in an oil moderator (Philbin et al, ref. 8). The iron, chromium, and nickel peaks in the spectrum of the stainless steel source were absent in the Zircaloy-2 clad source. The background of the Zircaloy-2 source was significantly lower above 6 MeV. Although there was a definite improvement in the high energy part of the spectrum, the background due to the fission gamma rays from the californium dominated the background spectrum below 6 MeV. For most capture-gamma-ray analyses Zircaloy-2 is the best material to be used for source encapsulation.

INTERFERENCE FROM THE EXTERNAL MODERATOR

The neutrons emitted from californium range from about 0.2 MeV to 14 MeV, but the energy distribution peaks at about 1 MeV with the overall average energy being 2.3 MeV. Generally, the probability of neutron capture (the cross section) increases as the energy of the neutron decreases. Therefore, to obtain maximum neutron capture the average neutron energy must be reduced by scattering in a low-Z medium of proper thickness generally interposed between the source and the sample. Of course this scattering material or moderator will also be a source of gamma radiation and will tend to interfere with the sample spectrum. Hydrogen is the best moderator and yields a strong capture gamma ray at 2.22 MeV. It was initially thought that an organic moderator such as polyethylene could be used to reduce the

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energy and scatter the neutrons into the lunar surface. Although the hydrogen in the polyethylene serves as a moderator, the carbon and oxygen will contribute their own spectrum and will also be a source of background interference. To determine the general effect of these elements on the continuum the Zircaloy-2 clad source was suspended successively in a tank of oil (carbon) and then in a tank of water (oxygen). Figure 2 shows the resulting spectra. The background is higher in oil in the region of the full, single and double escape peaks of carbon. Likewise, in water the background is higher in the vicinity of the oxygen peaks. The copper and iron peaks are due to impurities in the water. In both cases, but not shown in the figure, the background near 2 MeV and at lower energy is increased by the presence of the hydrogen peak at 2.22 MeV In addition to the hydrogen interference one can expect to obtain some interference in the spectrum from 3.50 MeV to 6.25 MeV due to carbon and oxygen, if an organic moderator is used on the lunar surface. These interferences can probably be minimized by computer techniques (discussed below).

Other materials such as ZrH_2, and, ammonium metatungstate were also tested.

The hydrogen and tungsten content, and the high solubility of ammonium metatungstate suggest that this salt may make a good neutron-gamma shield for ^{252}Cf. Comparative attenuation experiments with a saturated aqueous solution and pure water show that the ammonium metatungstate solution is a significantly better shielding material for gamma rays and slow neutrons, and is also better than water for fast

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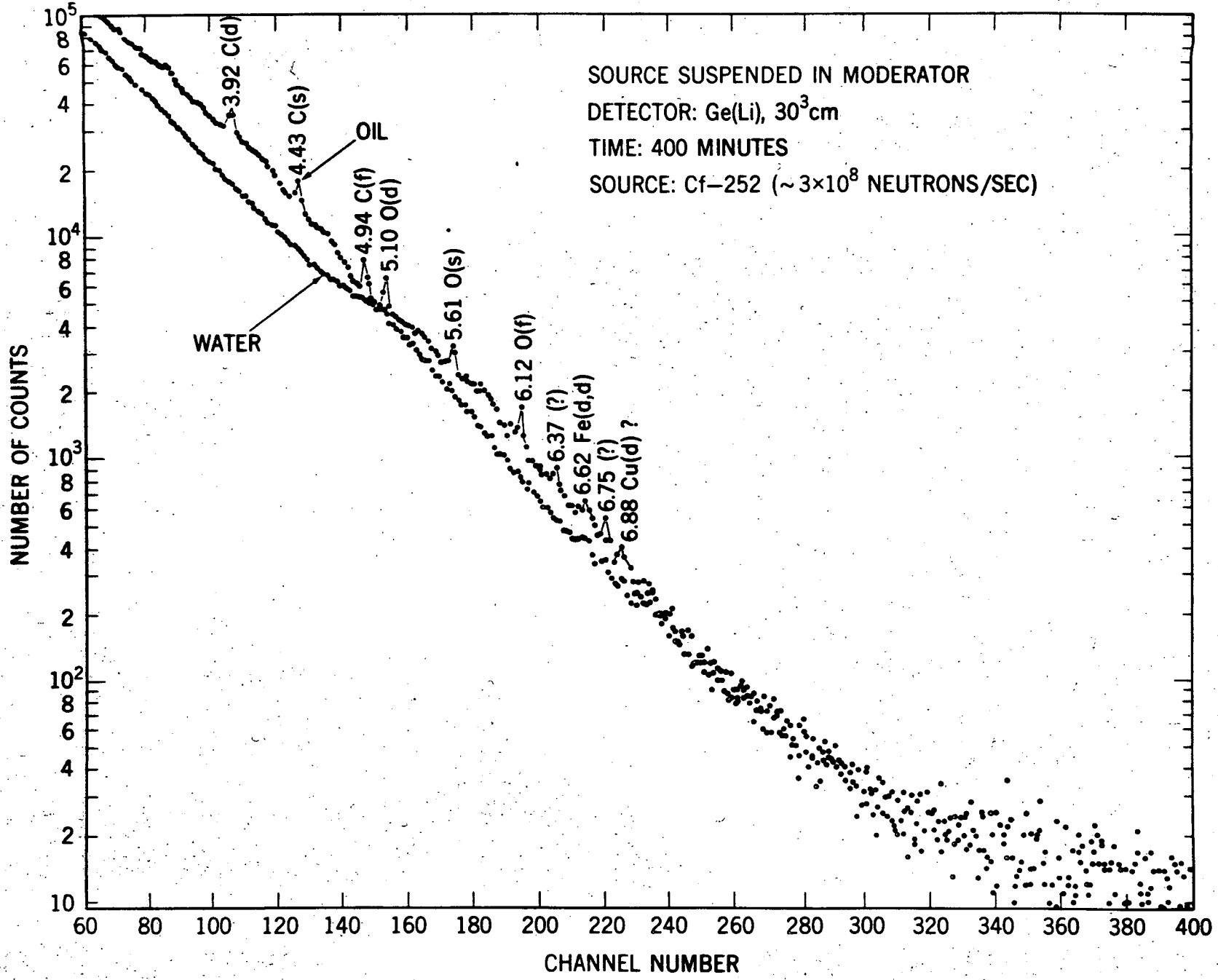
SOURCE SUSPENDED IN MODERATOR
DETECTOR: Ge(Li), 30³cm
TIME: 400 MINUTES
SOURCE: Cf–252 (~3×10⁸ NEUTRONS/SEC)
3.92 C(d)
4.43 C(s)
OIL
4.94 C(f)
5.10 O(d)
5.61 O(s)
6.12 O(f)
6.37 (?)
6.62 Fe(d,d)
6.75 (?)
6.88 Cu(d) ?
WATER
NUMBER OF COUNTS
CHANNEL NUMBER
10⁵
10⁴
10³
10²
10
60
80
100
120
140
160
180
200
220
240
260
280
300
320
340
360
380
400

Figure 2

neutrons. ^{252}Cf sources of less than 500 μg, a shield consisting of a thick paste of ammonium metatungstate was found to be useful as a shielding material in place of a saturated solution to reduce the hazard in case of container leakage. A detailed study of this report has been published (Senftle and Philbin, ref. 9).

INTERFERENCES FROM THE DETECTOR HARDWARE

To obtain the best counting geometry it will be necessary to place the detector as close as possible to the sample. It is clear that the detector and its associated hardware will thus necessarily be exposed to neutrons. Neutrons captured by the detector and hardware will be a source of background capture gamma rays. Using a 125 μg ^{252}Cf source, various thicknesses of water, oil or paraffin moderator were used to test the magnitude of this source of background interference. Even with 26 inches of oil moderator between the source and detector a substantial background was observed. To determine how much of this was due to slow neutron interaction with the hardware, the Ge(Li) detector and its associated liquid nitrogen reservoir were completely enclosed with 3 inches of Borax to reduce the slow neutron flux. Figure 3 shows a comparison of the background continuum with and without the protective borax shield. While this source of interference is not serious, it is significant even with this substantial amount of moderator. It is certainly desireable to bring the detector closer to the source to improve the geometry, and under these conditions the hardware interference will become more acute.

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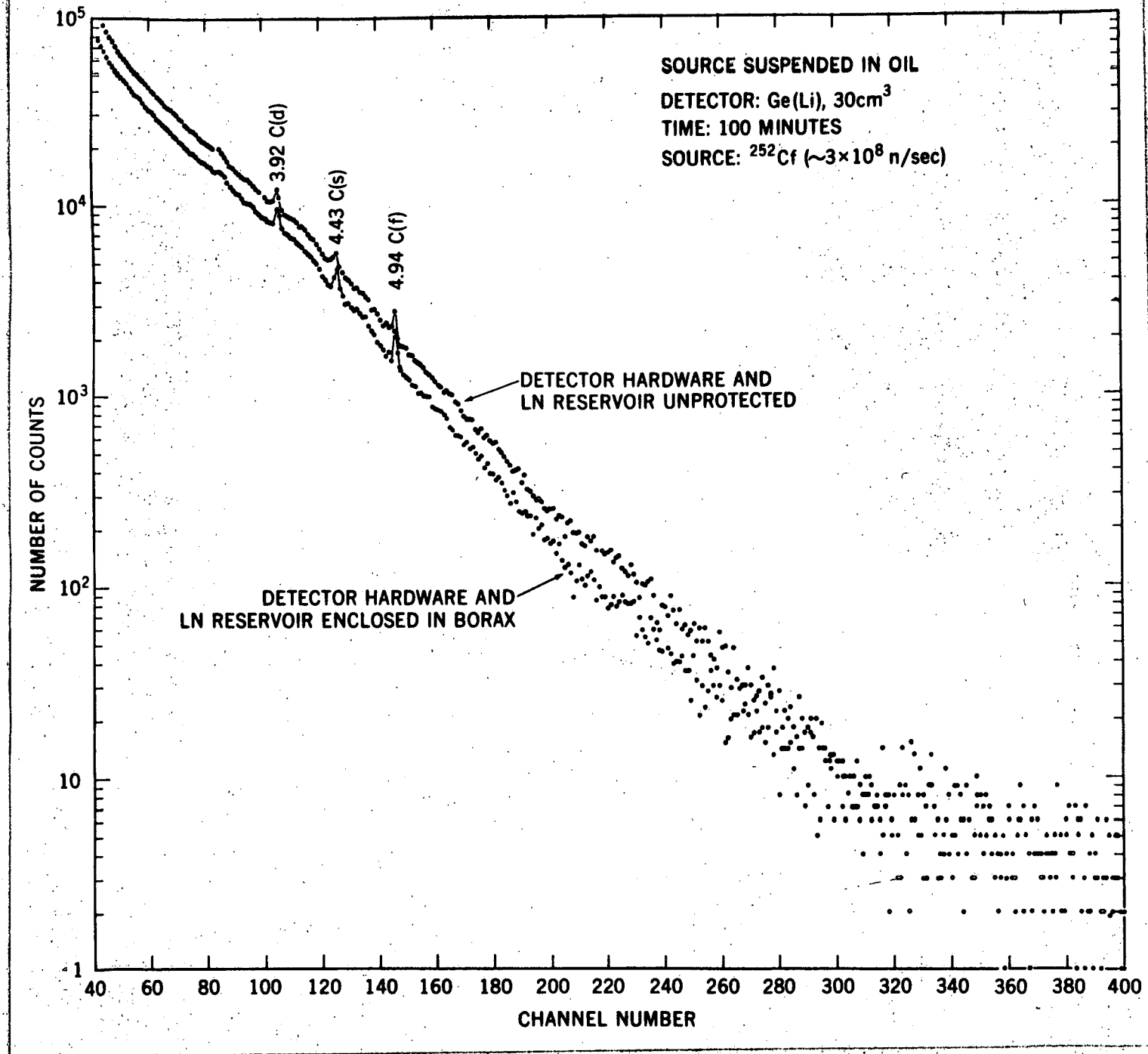
SOURCE SUSPENDED IN OIL
DETECTOR: Ge(Li), 30cm³
TIME: 100 MINUTES
SOURCE: ²⁵²Cf (~3×10⁸ n/sec)
3.92 C(d)
4.43 C(s)
4.94 C(f)
DETECTOR HARDWARE AND LN RESERVOIR UNPROTECTED
DETECTOR HARDWARE AND LN RESERVOIR ENCLOSED IN BORAX
NUMBER OF COUNTS
CHANNEL NUMBER
10⁵
10⁴
10³
10²
10
1
40
60
80
100
120
140
160
180
200
220
240
260
280
300
320
340
360
380
400

Figure 3

To minimize this effect for smaller source-to-detector distances one can (a) use a smaller ^{252}Cf source, (b) specially construct all hardware with low yield capture gamma ray elements[7,8], and (c) enclose the detector and its associated hardware with a light weight slow neutron shielding material such as boron. Laboratory experiments with powered boron and spectra taken of about 30 elements indicate that a thin boron coating on the outside of the detector hardware package will significantly reduce the interference. In addition, if Zircaloy-2 can be used to replace aluminum and stainless steel a further reduction can be achieved.

INTERFERENCES FROM ^{252}Cf FISSION GAMMA RAYS

Unlike other isotopic neutron sources, ^{252}Cf is a fission source and emits its own fission gamma ray continuum which is itself a source of interference. This can be minimized by use of a shadow shield between the source and detector. Experiments were made with various shadow shields including uranium and lead. Although uranium has the higher density, its natural gamma emission is high, and was not as good as lead for this purpose. Figure 4 shows the effect of using a 3/4 x 1-1/2 x 4 inch lead shadow shield. This shield substantially reduced both the fission gamma radiation. The fission gamma rays are perhaps the most serious source of interference when using a ^{252}Cf neutron source. If a point source is used these gamma rays can be substantially reduced with a shadow shield. However, if a distributed or broad source is used these gamma rays may be a serious source of

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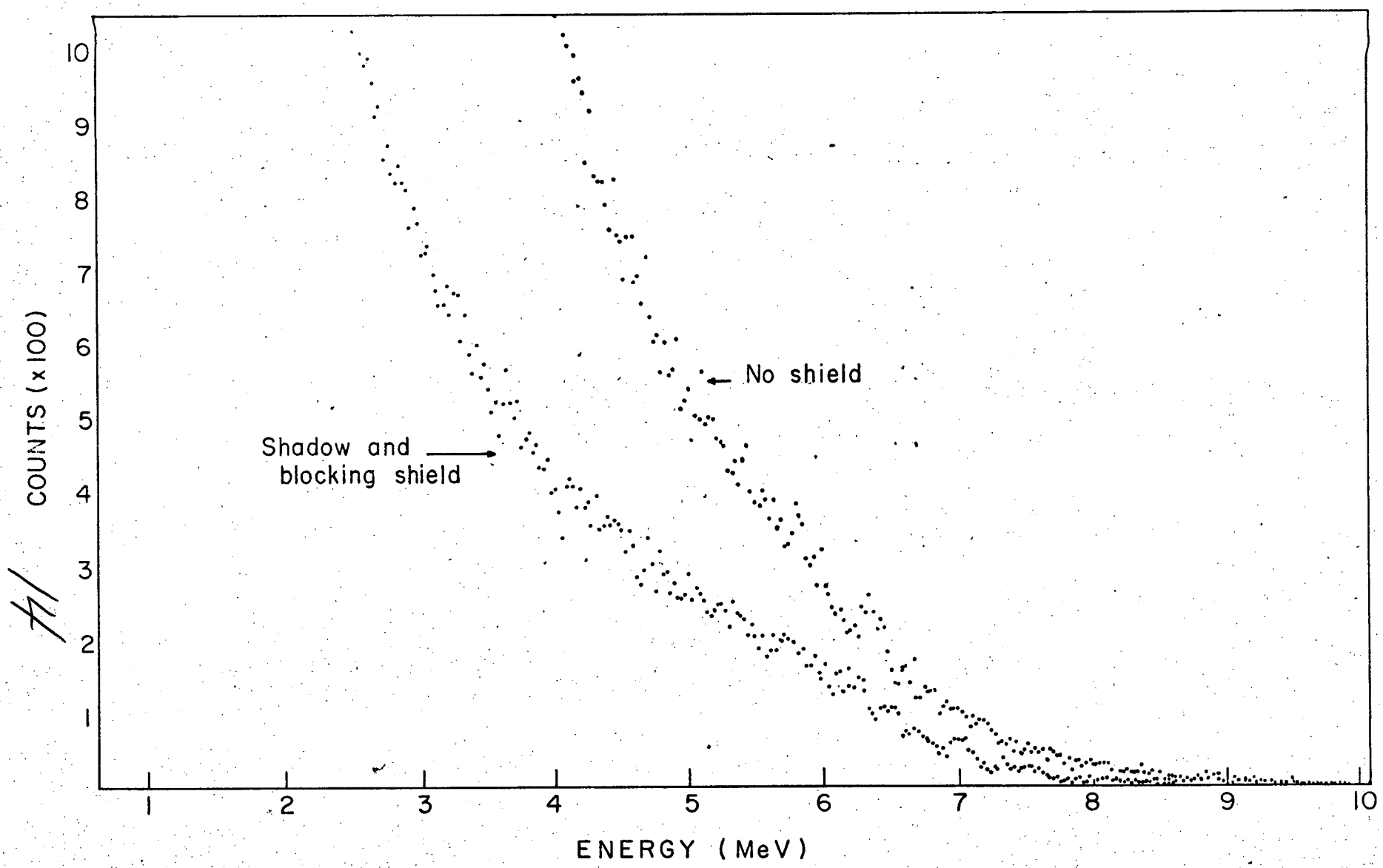
COUNTS (x100)
ENERGY (MeV)
No shield
Shadow and
blocking shield

Figure 4

interference.

THE SOURCE-TO-DETECTOR DISTANCE

To determine the response of the detector with the source-to-detector distance, preliminary experiments were made with a sample of titanium ore (∿8 lbs) in which the ^{252}Cf source was placed centrally within the ore sample. The sample and source were immersed in a water tank and the Ge(Li) detector was placed in a fixed position just outside the tank. The source-sample assembly was arranged so that it could be moved with respect to the detector. Using the double escape peak of titanium at 5.74 MeV, the detector response was measured as a function of source-to-detector distance as shown in Figure 5 for five ^{252}Cf sources from 1 μg to 100 μg. Using the 20 μg and 100 μg sources optimum distances of 16 and 20 inches, respectively, were found. Experiments showed that the sharp drop in counting rate as the source-sample assembly was moved closer to the detector was due to electronic blocking of the detector by low energy gamma rays from both the source and the moderator (mostly hydrogen). By placing a 0.5 inch thick piece of lead in front of the detector the dashed curve in Figure 5 was obtained. Blocking of the detector was reduced so that the source-sample assembly could be brought closer to the detector before the counting rate again started to drop. The resulting improvement of the geometry also resulted in enhancement of the 5.74 MeV peak to an optimum value at a source-to-detector distance of 17 inches. Experiments with smaller sources confirmed this explanation. Thus, the

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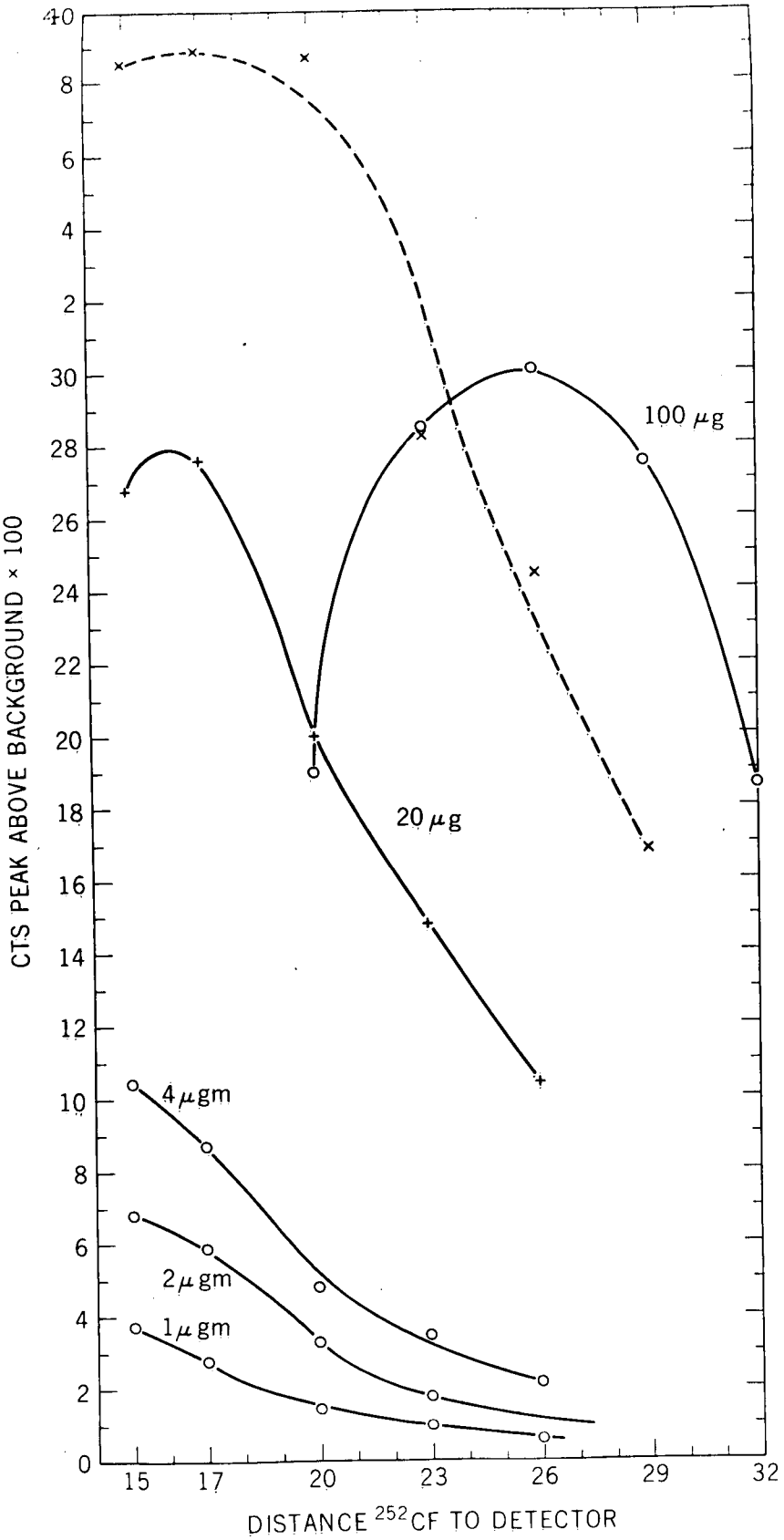
CTS PEAK ABOVE BACKGROUND × 100
DISTANCE 252CF TO DETECTOR
100 μg
20 μg
4 μgm
2 μgm
1 μgm
0
2
4
6
8
10
12
14
16
18
20
22
24
26
28
30
40
15
17
20
23
26
29
32

Figure 5

optimum source-to-detector distance is a function of the size of the source used.

The above experiments were repeated with a NaI(Tℓ) probe in the USGS Borehole Test Facility. The ^{252}Cf source was lowered down a borehole surrounded with lowgrade (∿1%) nickel ore to a fixed position. The NaI(Tℓ) detector was then dropped stepwise toward the source, and counts taken in a window encompassing the 8.489 MeV single escape peak of nickel. Figure 6 shows how the optimum source-to-detector distance and height of the nickel peak vary with strength of ^{252}Cf source. Considering the difficulties in handling large sources, the data suggests that sources larger then 50 μg may not be warrented for capture gamma ray analysis.

SAMPLE POSITION

If one fixes the source-to-detector spacing and assuming an infinite homogeneous medium, what is the position of the sample with respect to the detector? The sample immediately around the source receives the highest flux of neutrons of all energies, but the sample an inch or two from the source receives the highest thermal neutron flux. However, because of the absorption of the emitted gamma rays and the relatively poor geometry a sample closer to the detector, although it receives a lower neutron flux, may contribute proportionatly more gamma rays to the observed spectrum. To determine the position of the sample for high energy capture gamma rays a probe in which the ^{252}Cf was rigidly fixed with respect to the detector was

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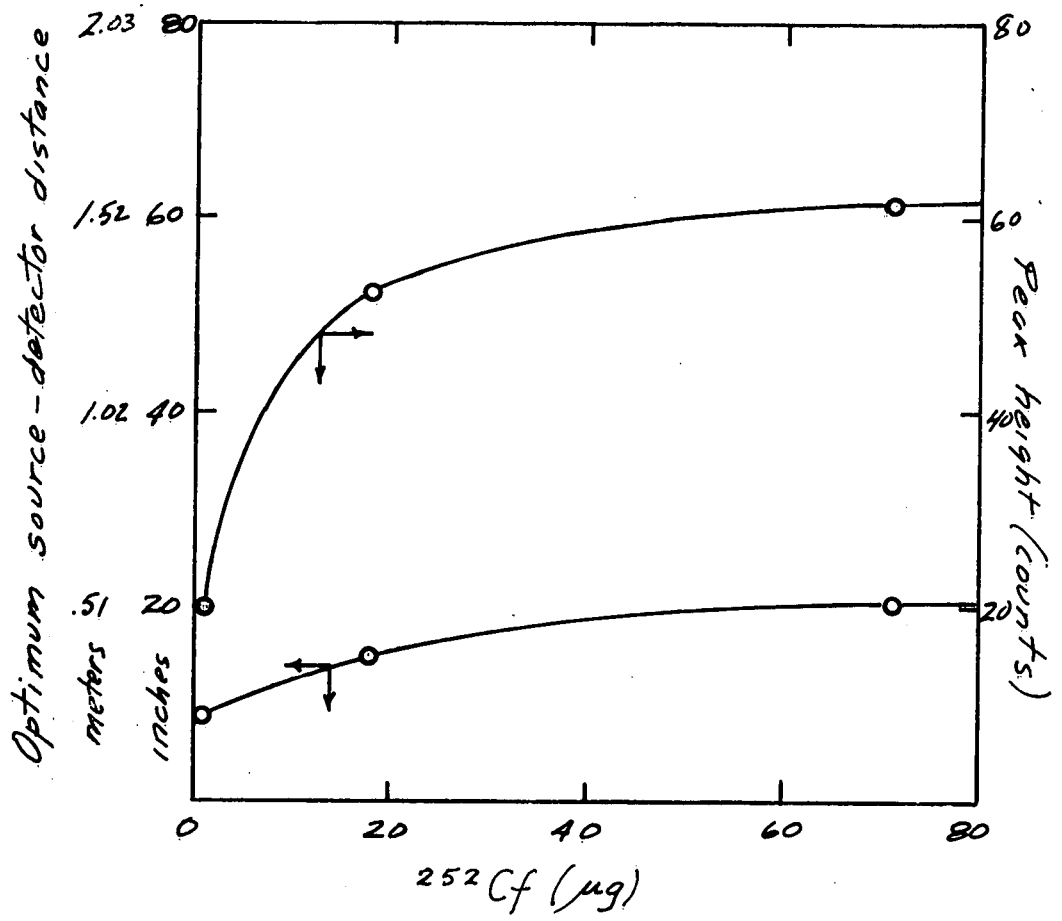

Figure 6 - The optimum source-detector distance and the height of the 8489 keV single escape peak of nickel at this distance, for a 3 x 3 inch NaI(Tℓ) crystal in a borehole configuration as a function of the strength of the ^{252}Cf.

used. Nominally 1 and 20 μg ^{252}Cf sources were used with both NaI(Tℓ) and Ge(Li) detectors. A half inch thick layer of nickel powder was sandwiched between thick layers of damp sand in the USGS Borehole Facility. The probe was lowered a few inches at a time down the borehole so that the source approached and passed by the nickel layer. Measurements were made of the single and double escape peaks of nickel at 8.489 and 7.978 MeV respectively. With a 1 μgm source it was found that the sample was 3/4 of the distance from the source to the detector when the source-to-detector distance was 16" regardless of the type of detector used. When dry sand was used the sample was about 2" closer to the detector.

HARDWARE CONFIGURATION

In order to use the neutron capture gamma ray method for elemental analysis on a planetary mission, the system must be constructed (a) with the ^{252}Cf source and detector as separate units which can be deployed subsequent to landing or (b) the source and detector can be rigidly mounted with respect to each other. Initial thinking favored system (a) in order to remove the ^{252}Cf source from close proximity to the NaI(Tℓ) crystal. However, from the experiments performed during the past year it is now clear that system (b) must be given serious consideration. If the ^{252}Cf is to be driven a few inches below the surface it would be extremely difficult to shadow shield the neutron source using system (a). In addition, if the source and detector are rigidly mounted as a probe, which would be driven a few inches into

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the ground, the geometry would be fixed and could be calibrated with greater ease.

For lunar exploration and possibly for future planetary work, it is desireable to have the capability of logging a shallow hole for a variety of elements. Thus, several probes have been built in the laboratory using both NaI(Tℓ) and Ge(Li) detectors and a 100 μg ^{252}Cf source. As determined in the other laboratory experiments, it was found that in order to keep the counter from blocking and also to prevent activation of the crystal, a 32 inch separation was needed between the source and detector. However, based on the experiments with a 1 μg ^{252}Cf source and a NaI(Tℓ) detector, a prototype probe using a 10 inch separation between detector and source has been constructed and successfully operated.

It is felt that experiments similar to those described above should be tried in a vacuum and high pressure gas environment using both NaI(Tℓ) and Ge(Li) detectors. Such experiments will require sealed detectors. Because of the liquid nitrogen cryogen needed for the Ge(Li) detector, it is not possible to operate such a detector in a vacuum; the constant low temperature is maintained by the latent heat of vaporization of the liquid nitrogen which must be vented. It occurred to us that to use the latent heat of melting of some substance might be used which would preclude the need to vent the cryostat. Such a probe has been built and tests have been quite successful. The details of this work have been prepared as a short paper[11]. This probe will allow us to test the Ge(Li) detector in a vacuum as well as

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down boreholes.

DATA REDUCTION

During the proposal phase of the project it was decided that the USGS team would work out the laboratory development and field testing of the technique and that the GSFC team would handle the data reduction and transmission methods. It soon became apparent that some overlap was not only unavoidable, but desireable. The data reduction techniques as described in the paper by Trombka et al[13] was already under investigation by the GSFC team and its development has since been progressing. At the same time the USGS team had been using a different data reduction technique for x-ray fluorescence spectra[4] which also appeared applicable to neutron capture gamma-ray analysis after some minor changes and alterations in the computer program. Tests of this method of data reduction is currently in progress.

The technique consists of (1) taking the spectrum of an element to be analyzed; (2) taking spectra of many substances that would interfere, if present, with analysis for the element sought; (3) computing a weighting function to minimize effects of the various interferences; and (4) applying the weighting function to spectra of unknown materials to obtain a figure of merit for each material, indicative of the extent to which it contains the element sought. The technique is novel in that the weighting function is computed without specific knowledge of the unknown material; all that is required is that interfering elements that it contains be represented in step (2)

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above, but not necessarily at the same concentration level. The weighting function can be made quite general without undue loss of sensitivity, and once it has been computed it can be used for prompt spectral interpretation with computers of very limited capacity or even with programmable electronic calculators. Concurrent determination of a dozen or so elements, using a precomputed weighting function for each, is quite feasible with 1 K of computer memory. The single numerical output for each element permits convenient recording by strip chart or contouring.

A first test of the linear combination technique using this system and capture gamma-ray analysis has been completed. Calcium, one of the more difficult elements to measure by the capture gamma-ray method, has arbitrarily been chosen as the element sought. Six 72-keV-wide "windows" containing the more important calcium peaks, are examined for spectra of calcium ($CaCO_3$) and of ten other elements or compounds having peaks within at least one of the six windows. The weighting functions have been computed for several different groupings of raw data and for counting times of 4-1/2 hours and of 17 minutes. Tests of the weighting function against the various interferences, notably titanium, indicate the ability to discriminate between calcium and individual interferences in ratios of from 1 in 41 to 1 in several hundred; between calcium and an aggregate of all interferences, calcium can be discriminated in a ratio of 1 in 12. Compared with two popular methods of peak area integration, the linear combination technique was much better at rejecting interferences. For analysis involving

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mixtures of unknown composition, the technique offers improved sensitivity. Details of the method are in press[10].

INTERNAL MODERATION

Most laboratory experiments are performed with a moderator external to the sample. An alternative method which is more applicable to planetary exploration is to use internal moderation, i.e. placement of the isotopic neutron source within the sample. To demonstrate the feasibility of using this technique, the effect of moderation in dry and water-saturated samples was tested in the following manner. An annular-shaped sample holder containing about 25 lbs. of rock sample was buried flush with the surface of the ground which was in a water saturated condition. The detector was placed about 2 feet away on the surface and the californium source (125 μg) dropped into the center of the annulus (see Figure 7). Figure 8 shows a spectrum taken with the sample holder filled with both dry and wet gold ore. Although water is a good moderator, the spectrum with the higher counting rate was obtained with the dry sample. Because of the high scattering cross-section of hydrogen, many of the high energy neutrons which passed through the sample were scattered in the water saturated ground and were thus returned to the sample at near thermal energies and captured. In this configuration many of the γ rays originated in the outer part of the annularily shaped sample. The sample was then saturated with water and rerun. The total number of spectral counts was depressed as shown in the figure. Although more neutrons were thermalized within

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^{252}Cf SOURCE Ge(Li) DETECTOR EXPERIMENT CONFIGURATION.

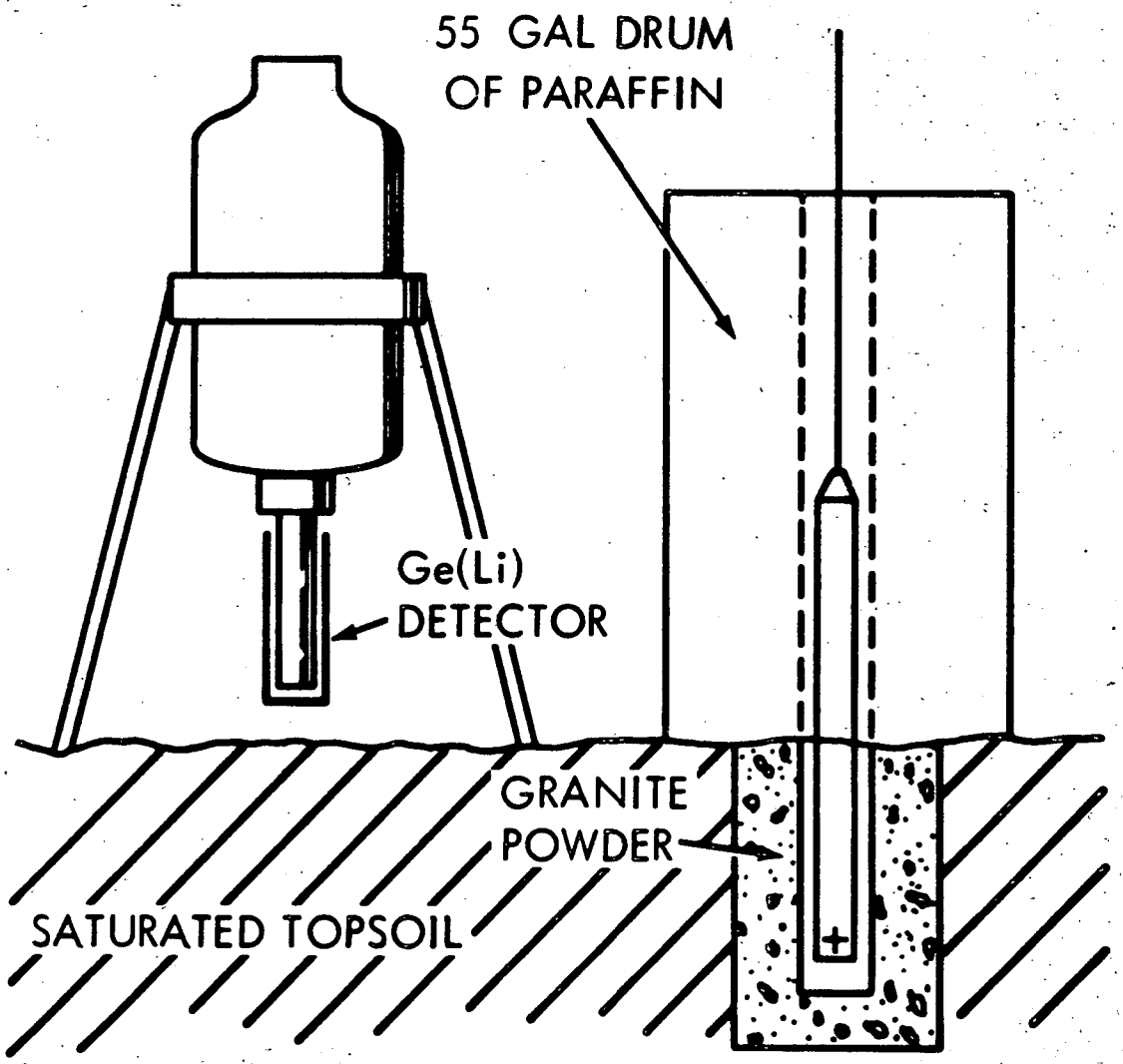
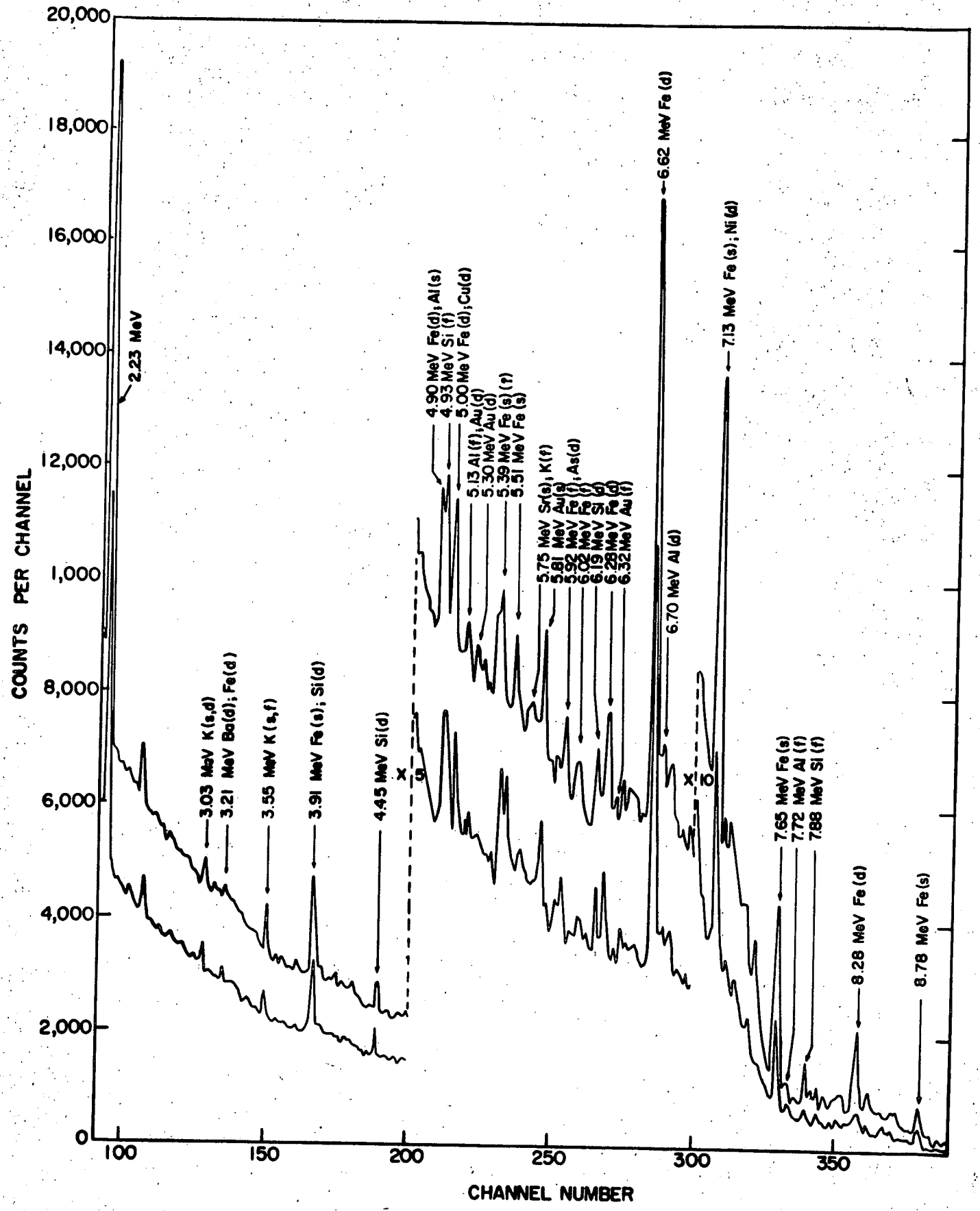


Figure 7


20,000
18,000
16,000
14,000
12,000
1,000
8,000
6,000
4,000
2,000
0
COUNTS PER CHANNEL
100
150
200
250
300
350
CHANNEL NUMBER
2.23 MeV
3.03 MeV K(s,d)
3.21 MeV Ba(d); Fe(d)
3.55 MeV K(s,f)
3.91 MeV Fe(s); Si(d)
4.45 MeV Si(d)
4.90 MeV Fe(d); Al(s)
4.93 MeV Si (f)
5.00 MeV Fe(d); Cu(d)
5.13 Al(f); Au(d)
5.30 MeV Au(d)
5.39 MeV Fe(s)(f)
5.51 MeV Fe(s)
5.75 MeV Sr(s); K(f)
5.81 MeV Au(s)
5.92 MeV Fe(f); As(d)
6.02 MeV Fe(f)
6.19 MeV Si(d)
6.28 MeV Fe(d)
6.32 MeV Au(f)
6.62 MeV Fe(d)
6.70 MeV Al(d)
7.13 MeV Fe(s); Ni(d)
7.65 MeV Fe(s)
7.72 MeV Al(f)
7.88 MeV Si(f)
8.28 MeV Fe(d)
8.78 MeV Fe(s)
X 5
X 10

25

Figure 8

the sample, the total number was not as large as the number scattered into the dry sample by the external water as shown by the previous measurement.

This is an interesting result which is not obvious when working with small samples in the laboratory. For planetary application it would be best to eliminate an external moderator from the point of view of weight. The question arises as to whether or not sufficient moderation can be obtained in a sample without the presence of hydrogen.

Figure 9 shows a comparison of two spectra of a one pound iron ring (3-1/2" pipe coupling, I.D. = 5") with a 1 μgm ^{252}Cf source placed inside taken with a NaI(Tℓ) detector at a distance of 15". A reasonably good spectrum showing the full, single and double escape peaks of iron is observed when the assembly was immersed in water (solid line). However, the iron lines have all but dissappeared when the measurements were made in dry nitrogen (dashed line).

When approximately the same amount of Fe_2O_3 was placed in a thin walled aluminum cylinder (2" diameter, 6.75" long) immersed in water a spectrum similar to the solid line in Figure 9 was obtained. When the same cylinder of Fe_2O_3 was then immersed in CO_2 at one atmosphere of pressure, the single escape peak of iron was just barely visible. The Fe_2O_3 was then mixed with Al_2O_3 to provide additional low mass scattering matrix. This mixture was then made up into a cylinder of the same height but 6.5 inches in diameter and weighed 7 lbs. When this second cylinder was immersed in an atmosphere of CO_2, the spectrum was essentially a continumum and was devoid of iron peaks. The importance

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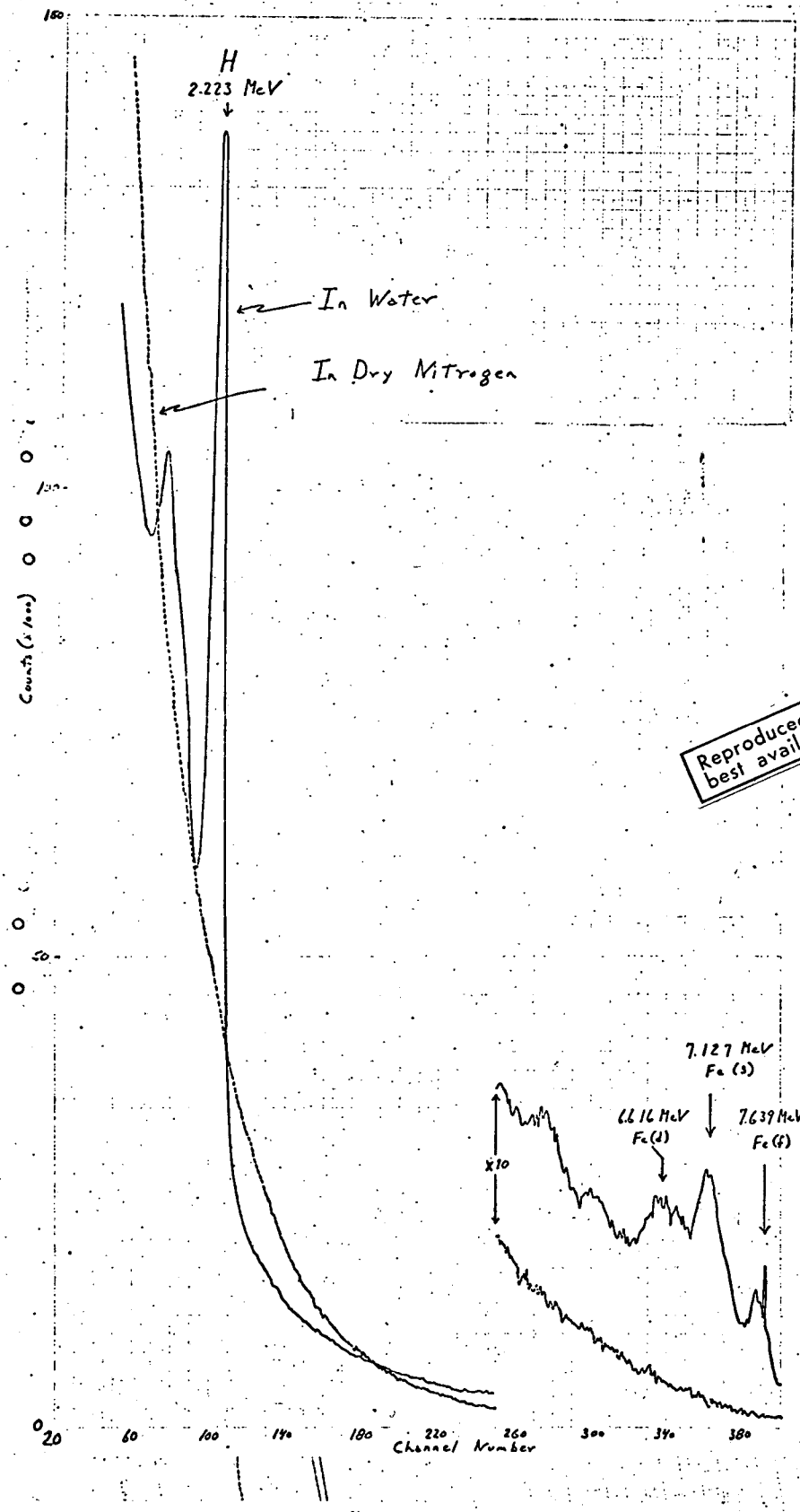

Figure 9

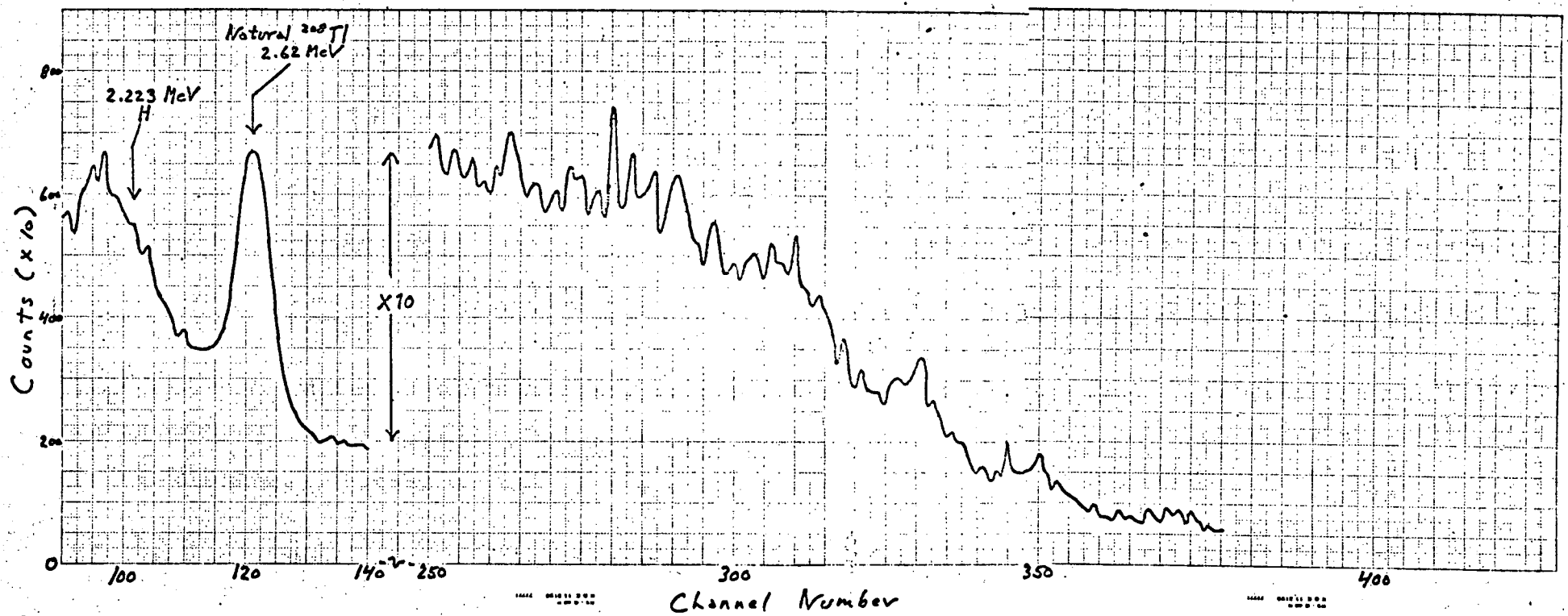
Natural ^{208}Tl
2.62 MeV
2.223 MeV
H
X10
Counts (x10)
8
6
4
2
0
100
120
140
250
300
350
400
Channel Number

Figure 10

of the presence of hydrogen is clear. For relatively small samples one must have hydrogen present to perform any serious spectral analysis.

We then ask if one had a semi-infinite sample would the high energy neutrons be slowed down sufficiently for capture and in a small enough volume to obtain a measurable spectrum? Large non-hydrogeneous samples are not easily available and hence an ideal experiment could not be made. However, a barrel of very dry heavy mineral sand concentrate (magnetite, monazite, rutile) was available which could be used to approximate the situation. The 1 μgm ^{252}Cf source was buried about 6-8 inches below the surface. The NaI(Tℓ) detector was placed beneath the barrel so that there was 20 inches of high density material between the source and detector. The spectrum in Figure 10 was obtained. The hydrogen peak is barely visible. Although the ore was quite dry some hydrogen was obviously present. The large peak at 2.02 MeV is due to $^{208}Tℓ$, a decay product of thorium in the monazite. The iron peaks are visible but poorly developed. The data again indicate that if there is no hydrogen present one can expect but poor capture gamma ray spectra. It is planned to repeat the latter experiments on more extensive samples which would more properly represent a planetary surface. However, it is felt that the above experiments point out a serious difficulty in obtaining good radiative capture gamma ray spectra on the lunar or planetary surfaces.

Some experiments were performed in which the ^{252}Cf source was surrounded with an inch or more of plastic as an external moderator. The plastic shrowded source and the iron ring described above were

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buried several inches deep in a large barrel of very dry fire clay. The resulting spectrum was exceedingly poor and the iron lines were barely visible. The use of an external moderator is certainly an improvement but more moderator and a better geometrical arrangement will be required to make its use on a planetary mission a feasible procedure.

CONCLUSIONS

Either a radiative capture or a delayed gamma ray method can be used for elemental detection and analysis. Theoretical calculations have been made to show which method is most sensitive for a given element. As the radiative capture method using a ^{252}Cf neutron source has not been fully developed, experiments were performed to test this technique using a ^{252}Cf source. The various causes of interferences have been evaluated and where possible, methods of reducing the interference have been investigated. The source-sample-detector geometry is critical to the capture gamma ray method, and optimum geometrical configurations have been examined. For practical use on a planetary lander the ^{252}Cf source will have to be embedded in or laid on the surface. This is most easily done without the use of an external neutron moderator. As there will be many cases where there will be essentially no hydrogen present experiments were made in relatively dry materials to determine the effects on the spectrum. With a point source of neutrons it was found virtually impossible to get good capture gamma ray spectra. As the delayed gamma ray spectra

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also depend on thermal neutron capture, it is doubtful whether either technique using a point source of neutrons can be used for planetary elementary analysis. Use of an external moderator will help but it is our experience that sufficient moderator to slow down the required number of neutrons will be too heavy and thus impractical.

Some hope lies in the utilization of a distributed source. Experiments in a dry environment with multiple sources are planned and will be carried out during the coming months.

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