

TK9008
M41
N96
no. 125

MITNE-125

Archives



PROMPT ACTIVATION ANALYSIS
OF COAL AND IRON ORE

by

Robert W. Schaefer Jr.

Norman C. Rasmussen

Massachusetts Institute of Technology
Department of Nuclear Engineering
77 Massachusetts Avenue
Cambridge, Massachusetts 02139

January 1971

MITNE-125

Prepared For

U.S. Department of the Interior
Bureau of Mines
Morgantown, West Virginia 26505

Contract Number HO180895



Room 14-0551
77 Massachusetts Avenue
Cambridge, MA 02139
Ph: 617.253.2800
Email: docs@mit.edu
<http://libraries.mit.edu/docs>

DISCLAIMER OF QUALITY

Due to the condition of the original material, there are unavoidable flaws in this reproduction. We have made every effort possible to provide you with the best copy available. If you are dissatisfied with this product and find it unusable, please contact Document Services as soon as possible.

Thank you.

Missing pages 17, 19, & 21

The images contained in this document are of the best quality available.

ABSTRACT

The capture gamma spectra from iron ore and various types of coal were measured with a 30 cc. Ge(Li) detector, a 4096 channel analyzer, and a 4 mCi. Cf-252 source. The iron spectrum was examined for the presence of silicon peaks and some parameters of those peaks were determined. The concentrations of three elements in each of three coal samples were determined by comparison of the spectra with that of a known fourth sample. Sulfur concentration, averaging about 1.6%, was measured to within 0.1%, hydrogen averaging 5.8% was found to within 1% and carbon averaging 66% was determined to within approximately 3%.

TABLE OF CONTENTS

	<u>Page</u>
Abstract	2
Table of Contents	3
List of Figures	4
List of Tables	5
Acknowledgements	6
I. Introduction.....	7
II. Experimental Procedure and Equipment	12
2.1 The Detector System	12
2.2 Experimental Procedure	13
III. Data Analysis and Results	23
3.1 GAMANL	23
3.2 The Coal Calculations	25
3.3 The Coal Results	26
3.4 Coal Spectrum Using a Pu-Be Source	29
3.5 The Iron Analysis	30
IV. Conclusion	35
Appendix A The Spectra.....	37
Appendix B Multiplets in the Coal Spectrum	45
References	47

LIST OF FIGURES

<u>Number</u>	<u>Title</u>	<u>Page</u>
2.1	Block Diagram of the Detector System.....	14
2.2	The Coal Setup.....	17
2.3	The Coal Setup.....	18
2.4	Arrangement of Detector, Lead Shield, and Source for Coal.....	19
2.5	The Iron Setup.....	21
2.6	The Iron Setup.....	22
A.1	Robena Spectrum (Cf-252 Source).....	38
A.2	Low Ash Spectrum (Cf-252 Source).....	39
A.3	Lignite Spectrum (Cf-252 Source).....	40
A.4	Arkwright Spectrum (Cf-252 Source).....	41
A.5	Arkwright Spectrum (Pu-Be Source).....	42
A.6	Dry Sintered Iron Ore Spectrum.....	43
A.7	Water-Saturated Sintered Iron Ore Spectrum.....	44

LIST OF TABLES

<u>Number</u>	<u>Title</u>	<u>Page</u>
3.1	Elemental Concentrations in Coal Samples.....	27
3.2	Silicon Multiplet Parameters Listed by GAMANL..	32
3.3	Suspected Components of Silicon Multiplets.....	33

Acknowledgements

I am indebted to several people for help with this work. I would like to express my appreciation for the aid and guidance of my supervisor, Prof. Norman C. Rasmussen. The assistance of others should be acknowledged including Prof. Franklyn Clikeman for aid with the electronics and Mr. Chang S. Kang for help with experimental equipment. I wish to thank Mrs. Stella Warshauer for typing this thesis.

Acknowledgement is made to the U. S. Bureau of Mines for financially supporting the project. Gratitude is also expressed to the U. S. Atomic Energy Commission for fellowship assistance.

Chapter I

INTRODUCTION

There are many industrial processes which could be optimized if the exact composition of process streams were known. This work investigates the possible use of prompt activation analysis in such applications. The method uses gamma rays produced upon neutron capture to measure elemental concentrations. Part of this effort is to determine if this process can be done with sufficient accuracy and efficiency to be useful in industrial applications.

Various other techniques have been applied in industry for elemental analysis, but there is a lack of methods suitable for on line analysis of large inhomogeneous process streams. Because methods such as chemical analysis and X-ray scattering are only suitable for small, thin samples, their results can be unrepresentative of inhomogeneous mixtures. Ordinary activation analysis has a time response which is usually too slow for on line work. There are other disadvantages of these methods including the high neutron flux required for ordinary activation analysis and the long time period necessary for chemical analysis.

Stewart, at the U. S. Bureau of Mines, has been investigating prompt activation analysis (ref. 1). This technique examines gammas emitted from inelastic scattering and thermal capture, as opposed to ordinary activation

analysis which measures the decay of induced activity. The method overcomes the problem of sample inhomogeneity because both the source neutrons and emitted gammas are quite penetrating. Stewart has described a method for bulk carbon analysis in iron ore sinter. He examined inelastic scattering peaks using an Am-Be source and a sodium iodine detector NaI. Such detectors have good efficiency but their energy resolution does not permit separation of gamma rays closely spaced in energy. The potential of the prompt activation analysis technique has been discussed by Rasmussen in reference 2. Rasmussen and Hukai examined coal samples (ref. 3). They used a lithium-drifted germanium detector, Ge(Li), which has good energy resolution but poorer efficiency than NaI. As a neutron source, a high thermal flux from a reactor was used. This type of neutron source would not be practical for industrial applications. Simonson and Hui (ref. 4, 5) studied inelastic gammas from various elements using a Ge(Li) detector and a Pu-Be source. The present work involves the examination of thermal capture peaks from coal and iron ore using a Ge(Li) detector and a Cf-252 neutron source.

The physical process employed in the measurements is the capture of neutrons followed promptly ($<10^{-12}$ sec.) by the emission of gamma rays characteristic of the absorbing elements. The number of captures by an element is proportional to the elemental concentration and the neutron capture cross section so the gamma ray intensity can be used to

infer the elemental concentration in the sample.

For neutrons at the source energy, the probability of neutron capture is extremely low. This probability becomes large enough to make the reaction useful, only when the neutrons have been slowed down or thermalized. Coal is well suited for this analysis because the considerable hydrogen concentration (~ 5 weight %) effectively thermalizes the fast source neutrons.

The elemental concentrations in four different coal samples were determined chemically by the Bureau of Mines. We chose one sample as a standard and by comparing the intensities of the characteristic capture gamma peaks, the concentrations of some elements in the other three were found.

The coal was examined for its sulfur content. The concentration of this element is of interest in two particular applications. A considerable amount of coal is used by the steel industry. In this use, the amount of sulfur in the coal affects the economics of the steel-making process. Metallurgical coal contracts specify the sulfur content required, and this analysis technique could be used to aid in meeting these specifications. The second application in which the sulfur level is important is its use as a fuel. When coal is burned the sulfur is released as toxic sulfur dioxide. This SO_2 emission is already such a serious pollution problem that an increasing number of cities have banned the burning of high sulfur coal.

The hydrogen concentration in coal can also be measured. The extent of this element's presence is important in coal processing. At one processing stage, it is washed with H_2O and then dried. If dried too much, the coal will be too dusty and present an explosive hazard. However, if it is too wet it interferes with the bulk handling procedures. Knowledge of the hydrogen concentration could be used to infer the water content and hence could be used to regulate the drying process.

Finally, the carbon concentration in the coal samples was measured. The calorific value of coal is related to the carbon content, and this may be of some interest to coal buyers.

Iron ore sinter was also examined by this technique. Of particular interest in this work was the silicon impurity. The amount of charge added to remove impurities during the iron processing can be optimized if the levels of impurities existing in the ore are known. It was sought to determine if this analysis technique can be used to adequately reveal the silicon content in the iron ore.

The lack of sufficient amounts of low atomic weight elements in iron ore results in inadequate slowing down of neutrons. As mentioned above, this means that relatively little neutron capture occurs. To overcome this, water was added and its effectiveness in thermalization was noted. The addition of water is a practical step since during some stages of processing, the iron ore is handled in the form

of a water slurry.

Chapter II contains an explanation of the equipment and method. In Chapter III the data analysis and results are covered. Conclusions and recommendations are made in Chapter IV.

Chapter II

EXPERIMENTAL PROCEDURE AND EQUIPMENT

2.1 The Detector System

The detector used throughout this work is a coaxial type Ge (Li) crystal with an active volume of approximately 30 cc. A detailed description of detectors of this type produced at M.I.T. has been presented by Orphan and Rasmussen (ref. 6). The detector is characterized by extremely good resolution. The full width at half maximum (FWHM) was approximately 8 keV in our work, in contrast to resolution a factor of ten worse for NaI crystals. High resolution allows better separation of closely spaced peaks. The detector efficiency of 0.1 to 1.0% is not so favorable however. This is about a factor of 10 less than that of NaI. As a consequence of this, the length of time required to collect data is relatively long. The maximum count rate that the system will tolerate is determined by the crystal and its associated electronics. Above the limit of approximately 3×10^5 c.p.s., the resolution deteriorates badly. This upper limit on the count rate implies a lower limit on the time period possible for data accumulation.

Considerable work has been done at this laboratory using a triple coincidence system. This system allows more precise determination of peak energies and intensities. Because the Ge(Li) crystal in free mode has a much more favorable efficiency and sufficient energy precision for

our purposes, we used the free mode.

In addition to the detector crystal, the system contains equipment for amplification, analysis, storage and readout (Fig. 2.1). A high voltage supply (Canberra model 3001) provides the 680 volt bias applied to the crystal. A detector head and preamplifier (Canberra model 1408C) receive the detector signal. The output is fed to the main amplifier (Canberra model 1417) which also contains pulse shaping circuitry. The amplified pulse then goes to a 4096 channel analyzer (Nuclear Data models 161F and 160M) which analyses the pulses and stores them in channels corresponding to their energy. A spectrum stabilizer (Nuclear Data model 502) is used to correct for system drift of all preceding components by applying a compensating voltage. A readout control and paper tape perforator (Nuclear Data model 160R and Talley model 406) punch the memory contents onto paper tape. The tape is converted to computer cards which are analyzed by a computer code (section 3.1). An oscilloscope is used to visually monitor the memory contents during and after data accumulation.

2.2 Experimental Procedure

In determining the components and geometry, the following factors were considered:

- (1) thermalization of the fast source neutrons
- (2) minimization of background neutrons and source gammas entering the detector

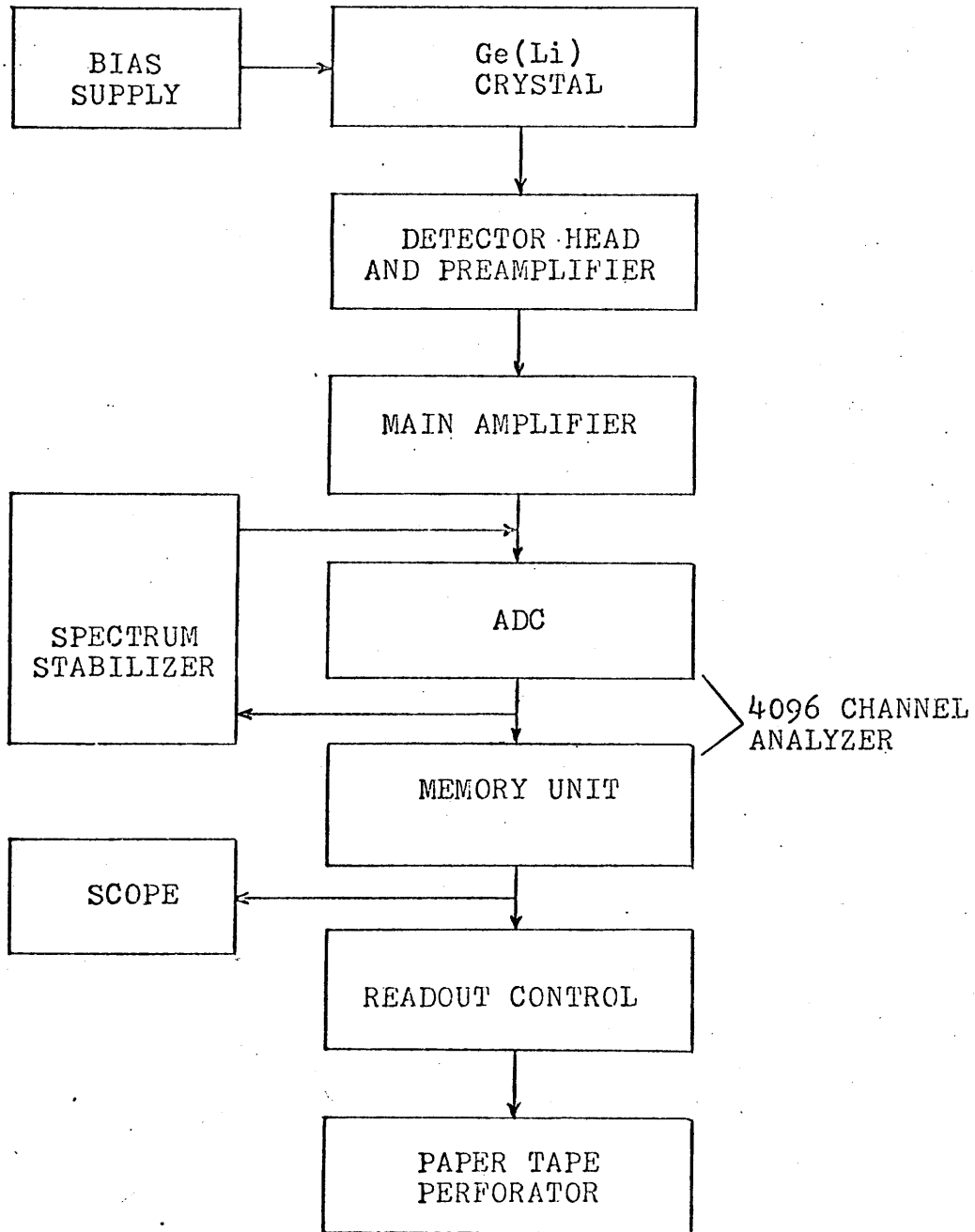


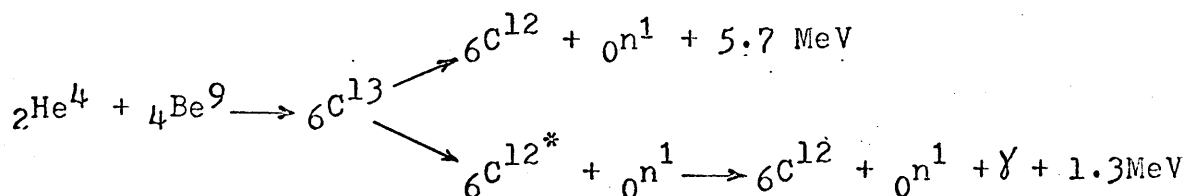
FIGURE 2.1 BLOCK DIAGRAM OF THE DETECTOR SYSTEM

(3) achieving a reasonably high count rate

(4) establishing easily and accurately reproducible conditions

The source used is Cf-252 emitting 1.7×10^7 neutrons per second. The principle source gammas are prompt and fission product gammas resulting from spontaneous fission.

For comparison, two coal runs were made using a 5 Ci. Pu-Be source. The source emits 8×10^6 neutrons per second and has a strong gamma ray line at 4445 keV. These gammas occur with any neutron source employing the (α, n) reaction of beryllium,



The 4445 keV gamma results from the de-excitation of the excited C^{12} nucleus formed as shown above. Note that this is the same gamma that appears from inelastic scattering collisions with carbon.

The coal samples are four different types, Arkwright, Robena, Lignite and Low Ash. Each sample is contained in a steel barrel 18 in. in diameter and filled to a height of about 26 in. with 200 lbs of coal. The mean distance a neutron travels from birth to capture in coal is 21 cm. (ref. 2). Thus the samples are large enough to insure that significant capture in the coal occurs and that the gammas are representative of the inhomogeneous mixture.

The components for the coal measurement and their arrangement are shown in figures 2.2 to 2.4. Location of the detector near the source results in a large count rate due to the high thermal flux there, but this is also the region of high background from source gammas and neutrons. In addition, at close separations the count rate is very sensitive to small changes in the source or counter position.

The background is effectively reduced by two components. The lead plug shields against direct source gammas and helps somewhat in suppressing fast neutron interference. The boral shield cuts down the thermal neutron background.

Referring to figure 2.3, the count rate is highly sensitive to the detector-to-source distance, D . For a fixed D , we found that small changes in A , B or C did not have large effects. After finding $D = 8$ in to be acceptable, B and C were arranged so that they could be reliably reproduced. $B = 0$ was chosen for convenience and was set by pulling the plug against the detector with nylon cord. The source to plug distance, C , was set at 3 in. by adjusting a nylon line. $A = 8$ in. was maintained by resting the detector on an aluminum plate supported by an adjustable frame. This arrangement was found superior to placing the detector just outside the sample. Our chosen arrangement more than doubled the peak to background ratio and reduced run length by about 25% over having $A = 0$ and no boral.



Room 14-0551
77 Massachusetts Avenue
Cambridge, MA 02139
Ph: 617.253.2800
Email: docs@mit.edu
<http://libraries.mit.edu/docs>

DISCLAIMER

MISSING PAGE(S)

17 ?

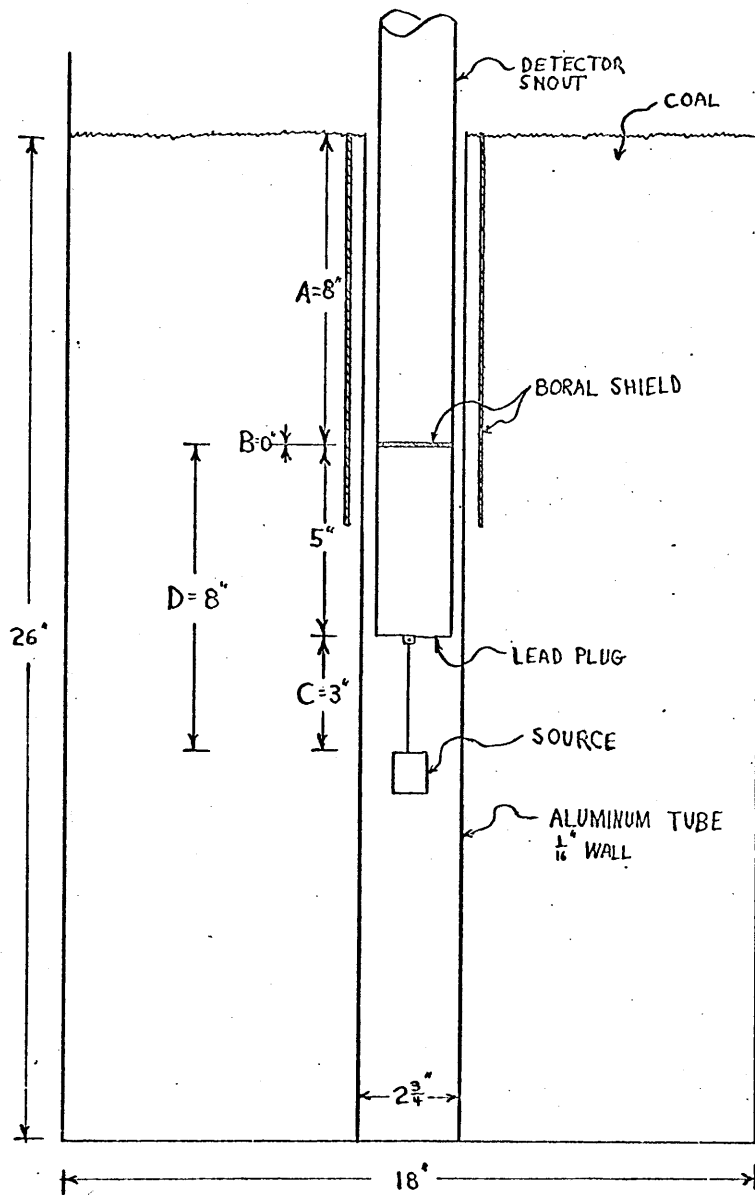


FIGURE 2.3 THE COAL SETUP



Room 14-0551
77 Massachusetts Avenue
Cambridge, MA 02139
Ph: 617.253.2800
Email: docs@mit.edu
<http://libraries.mit.edu/docs>

DISCLAIMER

MISSING PAGE(S)

19 ?

For these coal measurements the amplifier gain was set such that there was 1.2 keV per channel yielding a full range of about 5 MeV. Runs were of about 15 hour duration.

The arrangement of components for the iron measurements is shown in figures 2.5 and 2.6. The iron sample was a 5 gallon pail containing about 100 lbs. of sintered iron ore. This smaller size was chosen for managability. The sample rested on a 10 in. thick paraffin block. This very slightly softened the neutron spectrum at the sample by the reflection, back into the sample, of some neutrons which leaked into the moderating paraffin.

Since the sample size prohibited internal location of the detector, we placed it axially at the sample's surface. No boral shield was used because the thermal flux is quite low outside the sample. Using the coal sample notation, the geometry was $A = 0$, $B = 0$, $C = 2.5$ in.

A run totaling 32.8 hours was made with an additional data readout after 23.2 hours. Water was then added. By mixing thoroughly, we were able to add enough water to have 1.4 weight % hydrogen at saturation. A run was made for 23.7 hours with this mixture. Gain was set such that the energy range was 7 to 8 MeV.



Room 14-0551
77 Massachusetts Avenue
Cambridge, MA 02139
Ph: 617.253.2800
Email: docs@mit.edu
<http://libraries.mit.edu/docs>

DISCLAIMER

MISSING PAGE(S)

21 ?

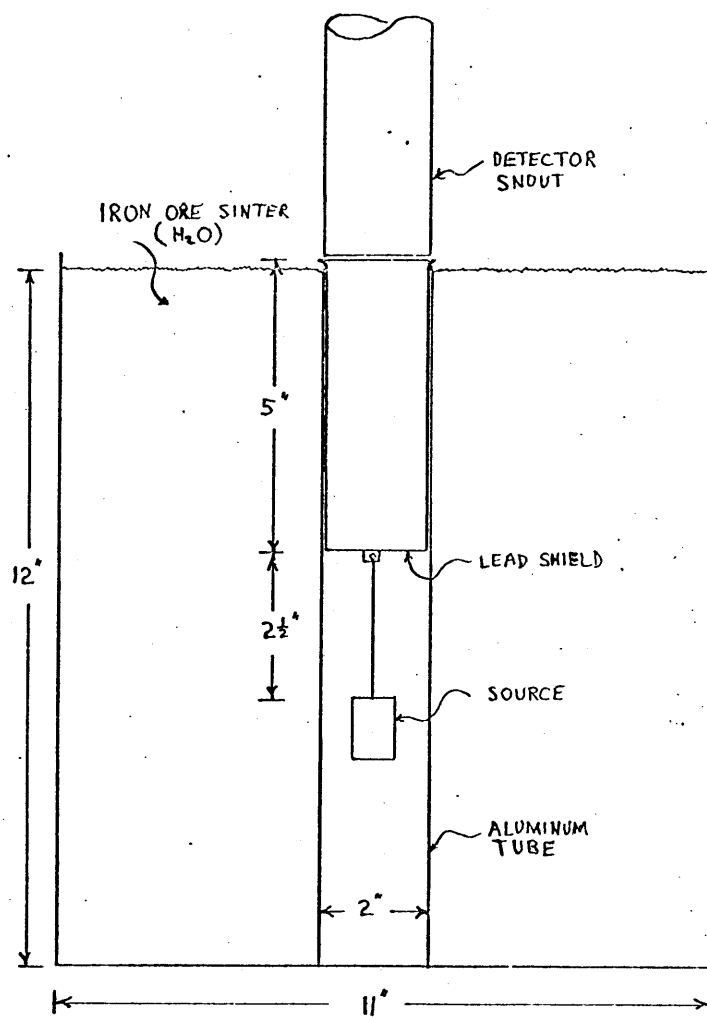


FIGURE 2.6 THE IRON SETUP

Chapter III

DATA ANALYSIS AND RESULTS

3.1 GAMANL

The raw data of counts vs. channel number is analyzed by a computer code, GAMANL. This program, developed at M.I.T., is described in detail in reference 7. The code first applies a smoothing filter function to the Fourier transformed data in order to eliminate the random fluctuations. The background is then determined and subtracted from the smoothed data. This is followed by the location of the peaks. It then analyzes the peaks and determines the following parameters: peak number, energy, center channel (channel number), height in counts, height to background ratio, area by summation method, area by Gaussian method, intensity, percent error in area by Gaussian method, FWHM measured, FWHM calculated by second order equation, width at base, and multiplet order of the peak.

Two areas are mentioned above; area by summation method and area by Gaussian method. The first is a straightforward summation of counts in the background subtracted peak. For the second, an equation of width vs. energy is evaluated by a least squares fit to strong singlets in the spectrum. The Gaussian areas corresponding to the peaks are then determined from the peak heights and calculated widths.

The intensity listed by GAMANL is the Gaussian area divided by constant factors and by the detector efficiency at the peak energy. The percent error in both types of area and in their ratio is a function of peak height and height to background ratio (ref. 8). The percent error in both areas increases as these parameters decrease. This is because when the peak rises only slightly above background, it is difficult to determine how many counts are actually due to the peak and how many are just background contributions. The ratio of Gaussian area error to summation area error becomes greater than 1 only for strong peaks with large height to background ratios. It is only for these strong peaks that the summation area is preferable.

GAMANL performs the energy calibration using two reference peaks and linearity data which are included in the input. Determination of peak origin is accomplished by comparing the energy from GAMANL with published values (ref. 9, 10). Some gammas entering the detector with sufficient energy interact via a pair production reaction. In these cases, one or both of the annihilation gammas may escape the detector without reacting. The result is the presence of peaks, called single escape (se) and double escape (de) peaks, which are degraded by 511 and 1022 keV respectively from their full energy values. Since the published energies are full energy values, 511 or 1022 keV must be added to some GAMANL peaks to identify their origin.

The small crystal size results in the double escape peaks being dominant above approximately 3 MeV.

3.2 The Coal Calculations

The area under a peak due to a particular element is given by,

$$\text{AREA} = N\sigma\phi f\epsilon g$$

where N = atom density of the element

σ = capture cross section

ϕ = neutron flux

f = fraction of gammas from the de-exciting nucleus which are of the energy considered

ϵ = efficiency of the system at that energy

g = solid angle factor

An absolute measurement was not attempted. Rather, the concentrations in three samples were determined by comparing the areas of relevant peaks to areas from the standard, whose concentrations are assumed known from chemical analysis supplied by the U. S. Bureau of Mines.

Measurements of the four samples were made and Arkwright was chosen as the known standard. Plots of spectra from all the samples are given in Appendix A, figures A.1 to A.4. The ordinate scales have been adjusted to account for differences in run length and sample density. Thus the plots can be compared directly.

The choice of peaks for the calculations had to be

made subject to at least two considerations. One is that the error due to statistical fluctuations is minimized by choosing as many peaks from an element as possible. However, another consideration is the limit on reliability of peak areas. It has been found (ref. 8) that only peaks with area error less than about 20% are reliable for quantitative analysis. Using this criteria, the peaks were chosen for the area comparisons. For sulfur, only the 4398 keV (de) peak was used. For carbon, the 2662 and 3923 keV double escape peaks were chosen. The Gaussian area was preferable for both elements. The 2223 keV (fe) hydrogen peak was so strong that using it alone presented negligible statistical error. The summation area was used in determining the intensity of this line.

Areas for each run were normalized to account for different run times and corrections were made for density differences. The concentration of an element in an unknown sample is then the value in the known sample times the ratio of the unknown's area to known's area.

3.3 The Coal Results

The results for coal are given in Table 3.1. The actual values, obtained from chemical analysis, are also tabulated there for comparison. The disagreement between the two methods of measurement of hydrogen content may not be as large as this table would indicate. The hydrogen concentration is not fixed, since it fluctuates with water

(27)

COAL TYPE	SULFUR WEIGHT %		HYDROGEN WEIGHT %		CARBON WEIGHT %	
	THIS WORK	CHEMICAL ANALYSIS	THIS WORK	CHEMICAL ANALYSIS	THIS WORK	CHEMICAL ANALYSIS
LIGNITE	0.4	0.4	7.8	6.8	46	42
ARKWRIGHT *	3.1	3.1	5.3	5.3	64.5	64.5
LOW ASH	1.1	1.1	6.4	5.5	82	80
ROBENA	1.8	1.8	5.9	5.4	74	76

* STANDARD

TABLE 3.1 ELEMENTAL CONCENTRATIONS IN COAL SAMPLES

content of the coal which was not carefully controlled in these measurements. Therefore part of the discrepancy in the two hydrogen analyses may be due to real differences in the amount present.

The error in Gaussian area listed by GAMANL is probably the largest single source of error. As mentioned in Section 3.1, this error is related to the uncertainty in separating counts due to a line from counts due to background. Statistical error is included in this error calculation. A large fraction of the discrepancy between the results of the two analyses can be accounted for by this area error; 0.1 wt. % sulfur, 4.8 wt. % carbon and 0.1 wt. % hydrogen. The problem of determining the true origin of counts is particularly difficult in cases where the peaks are contained in multiplets. In such cases there is uncertainty in the division of the multiplets' area among its components. Peaks from all three elements were in multiplets. A description of these multiplets, including suspected origin of the components, is given in Appendix B.

Another source of error is geometrical differences from run to run. Small differences in the arrangement of components occur when the system is disassembled and a new sample is set up. For example the source to detector distance, which strongly affects the count rate, was subject to inconsistencies. The nylon cord used to adjust this distance was slightly elastic, leading to small variations each time this distance was measured. The error contribu-

tion due to such factors is believed to be less than area error listed by GAMANL.

From the results and the above error evaluations, it is estimated that the technique can measure sulfur to 0.1%, carbon to 3% and hydrogen to at least 1%.

3.4 Coal Spectrum Using a Pu-Be Source

In order to evaluate the effect of having a different type of neutron source, some measurements were made of Arkwright using a Pu-Be source. The spectra resulting from the two different sources are shown in figures A.4 and A.5. The electronics was not performing as well during the Pu-Be run, leading to poorer resolution in this case. This is the reason for the somewhat lower and broader peaks in figure A.5.

The presence of the 4445 keV source gamma is quite prominent in the spectrum with Pu-Be. The multiplet containing the 3423 keV double escape is several times larger than with the Cf-252, and the single escape and full energy source peak are also apparent. They appear significant despite the presence of the 5 in. lead plug between the source and detector. These source gammas should not affect the sulfur or hydrogen measurements. Some problem from the 3934 keV (se) peak may arise from interference with the 3923 keV (de) carbon peak. It probably would not be a serious complication however, because they are 11 keV apart and the single escape peak is not strong.

3.5 The Iron Analysis

Measurements of both the dry and water-saturated iron ore sinter samples were made. Figures A.6 and A.7 display the two resulting spectra and show the thermalizing effect of the water. Because there were relatively few counts from the dry sample despite a 38% longer run, the ordinate scales are not the same. There are initially 1.5 times as many counts per inch on the wet plot's ordinate, and the differences in scaling factors are noted on the plots.

Note that with the water;

- (1) the time required to obtain a good spectrum is considerably less
- (2) the intensities of the capture peaks are greatly increased
- (3) the height to background ratios of these peaks are increased

A good example of the difference in the neutron spectrum is the prominence of the inelastic scattering oxygen peak (5108 keV (de)) in figure A.6 and its virtual absence in figure A.7.

The raw data from the water saturated sample was analyzed by GAMANL. There are three silicon capture peaks that appear; 2517, 3912 and 5358 keV double escape peaks. All three are in multiplets or have significant iron interference. Most of the iron peaks presenting difficulties would ordinarily be considered weak, but they are important here because of two factors. First, the iron is very much

more abundant than the silicon, and second, iron has a capture cross section 16 times larger than silicon (2.62 b. vs. 160 mb.).

Table 3.2 gives some parameters of the pertinent multiplets as listed by GAMANL. There is more interference than is immediately apparent from this table. That is, it is believed that there are cases where two sources contribute to the same multiplet component. Listed in table 3.3 are the suspected components of these multiplets, their source, energy and intensity. The meaning of the intensities listed in these tables should be clarified. The intensity column from GAMANL, as explained in Section 3.1, is proportional to the number of counts observed in the peak corrected for detector efficiency. The intensity of table 3.4 is the number of photons of the corresponding full energy emitted per unit flux per gram of the element. It should be kept in mind that with our system and in this energy region, double escape peaks are about four times as intense as their corresponding single escape or full energy peaks. This factor is not reflected in the intensities of table 3.4, since it lists the gamma ray emission probability not detection intensities.

Dual origin of the 2516 keV component in the first multiplet is suspected. A 2518 keV (de) Fe peak exists as well as the 2517 keV (de) Si peak. The 3912 peak in the second multiplet is believed to result purely from silicon

MULTIPLY ORDER	ENERGY (keV)	H TO BG RATIO	GAUSSIAN AREA (COUNTS)	RELATIVE INTENSITY	AREA % ERROR	FWHM (keV)
3	2516.0	0.055	5728	11.88	11.41	10.24
	2525.0	0.037	3907	8.06	16.48	10.24
	3RD. COMPONENT TOO WEAK FOR GAMANL TO ANALYZE					
3	3895.6	0.023	1557	1.72	33.75	10.99
	3913.4	0.077	5068	5.58	11.05	10.99
	3927.3	0.126	8276	9.08	7.28	10.99
1	5359.8	0.124	5919.7	6.28	8.62	11.85

TABLE 3.2 SILICON MULTIPLY PARAMETERS LISTED BY GAMANL

SILICON MULTIPLY	ENERGY (keV)	ELEMENT	INTENSITY PHOTONS (gm)(n/cm ²)
FIRST	2517.3 (de)	Si	2.73 x 10 ⁻³
	2518.1 (de)	Fe	2.83 x 10 ⁻⁵
	2527.7 (fe)	Fe	1.19 x 10 ⁻⁴
SECOND	3895.8 (se)	Fe	3.70 x 10 ⁻⁴
	3912.3 (de)	Si	2.42 x 10 ⁻³
	3918.8 (fe)	Fe	1.98 x 10 ⁻⁵
	3927.1 (de)	Fe	1.44 x 10 ⁻⁴
THIRD	5358.1 (de)	Si	4.33 x 10 ⁻⁴
	5358.7 (de)	Fe	1.81 x 10 ⁻⁴

TABLE 3.3 SUSPECTED COMPONENTS OF SILICON MULTIPLETS

since no iron peaks of significant intensity lie at that energy. The 5359 keV peak is listed as a singlet but is thought to really be two peaks, a silicon peak (de) at 5358 keV and an iron peak (de) at 5359 keV.

In order to approximately determine what fraction of the areas are attributable to silicon, the intensities of the 3912 keV (de) Si peak and the 4899 keV (de) Fe peak were compared. Each peak was assumed to be generated purely by the single element. The ratio of intensities from GAMANL was compared with the ratio of intensities per gram per unit flux listed in reference 10. From this, it appears that about 88% of the 3539 and 15% of the 5358 keV peaks are produced by silicon. This interference makes the usefulness of the two peaks doubtful, especially the 5358 keV peak.

Chapter IV

CONCLUSION

The results of the coal measurements indicate that this analysis technique can give reasonably accurate results. It was possible to measure the sulfur concentration averaging 1.6% to better than 0.1 wt. %, the carbon content averaging 66% to about 3 wt. %, and the hydrogen level of about 6% to at least 1 wt. %. As discussed in Section 3.3, the hydrogen accuracy may be better than this. Improved accuracy for all elements could probably have been obtained if repeated measurements of the standard sample were made, yielding average values.

The comparison of spectral data resulting from the Pu-Be and Cf-252 sources indicates that the Pu-Be could also be used for this analysis. This source has the advantages of being readily available and relatively inexpensive, but it has the drawback of a limited strength ($<10^9$ neutrons/sec.).

The application of this analysis technique to on line analysis can not be accomplished without considerable modifications and improvements. To analyze a moving process stream, the time for data collection would have to be cut from the present 15 hours down to a few minutes. To help achieve this, both improved detector efficiency and a stronger neutron source would be required. These are now becoming available. Rasmussen discusses the limitations

and requirements for such applications in reference 2. NaI detectors would be advantageous because of their high efficiency, but their relatively poor energy resolution limits their usefulness. In cases where the peaks of interest are relatively isolated, NaI detectors could be successfully employed. The sulfur and hydrogen lines in coal are peaks for which an NaI detector is probably useful.

It is thought that the problems mentioned above are solvable and that the technique can be successfully applied to some industrial problems. Some further work will however be required to solve the problems.

The iron results are not as promising as the coal measurements. The significant iron interference makes questionable the utility of the silicon peaks for quantitative analysis. The area errors are below the 20% limit even though the peaks are not strong above background. The 3912 peak appears most useful since it is relatively free of iron interference. The 2517 keV peak may be of value if the iron concentration is approximately constant from sample to sample. The 5359 keV peak is thought to be too dominated by iron to be useful.

Further work, similar to our coal analysis, could be done to test the reliability of the 3912 and 2517 keV peaks for quantitative silicon measurements.

Appendix A

THE SPECTRA

This appendix contains a plot of counts vs. energy (or channel number) for each of the coal samples examined. For one sample two spectra are given, one using a Pu-Be neutron source and one with Cf-252. Also included are spectra from both dry and water-saturated iron ore sinter.

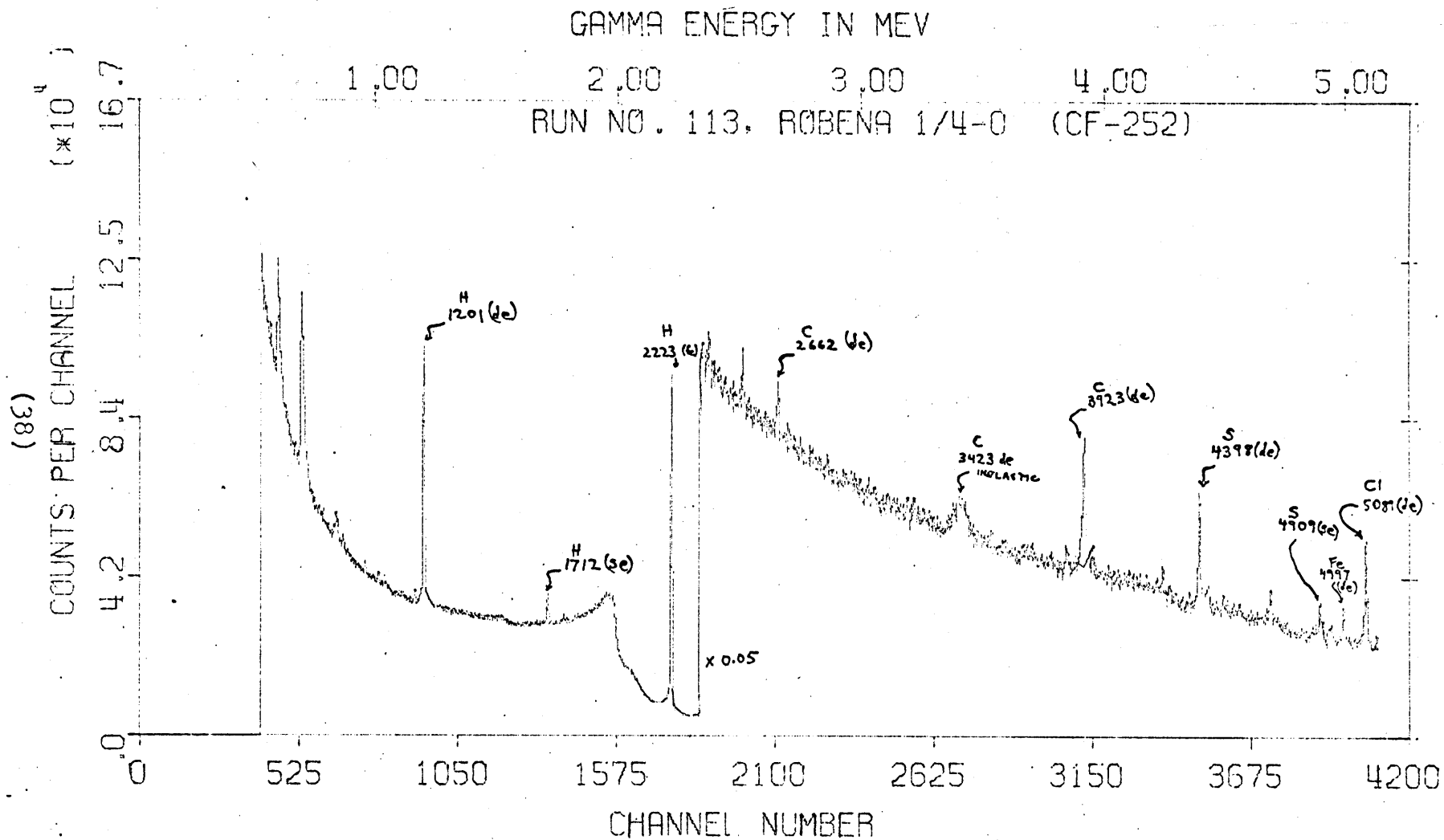


FIGURE A.1 ROBENA SPECTRUM

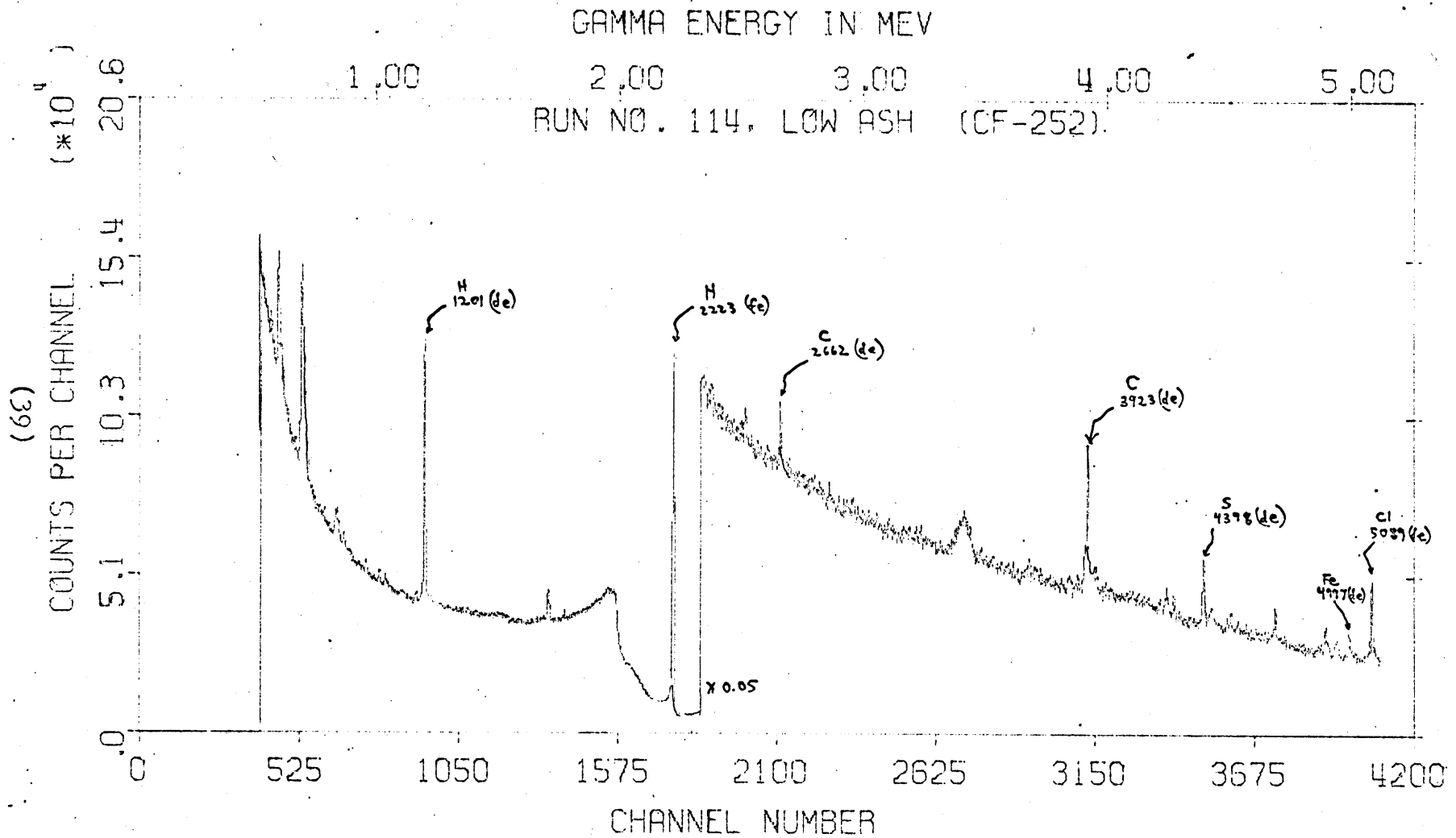


FIGURE A.2 LOW ASH SPECTRUM

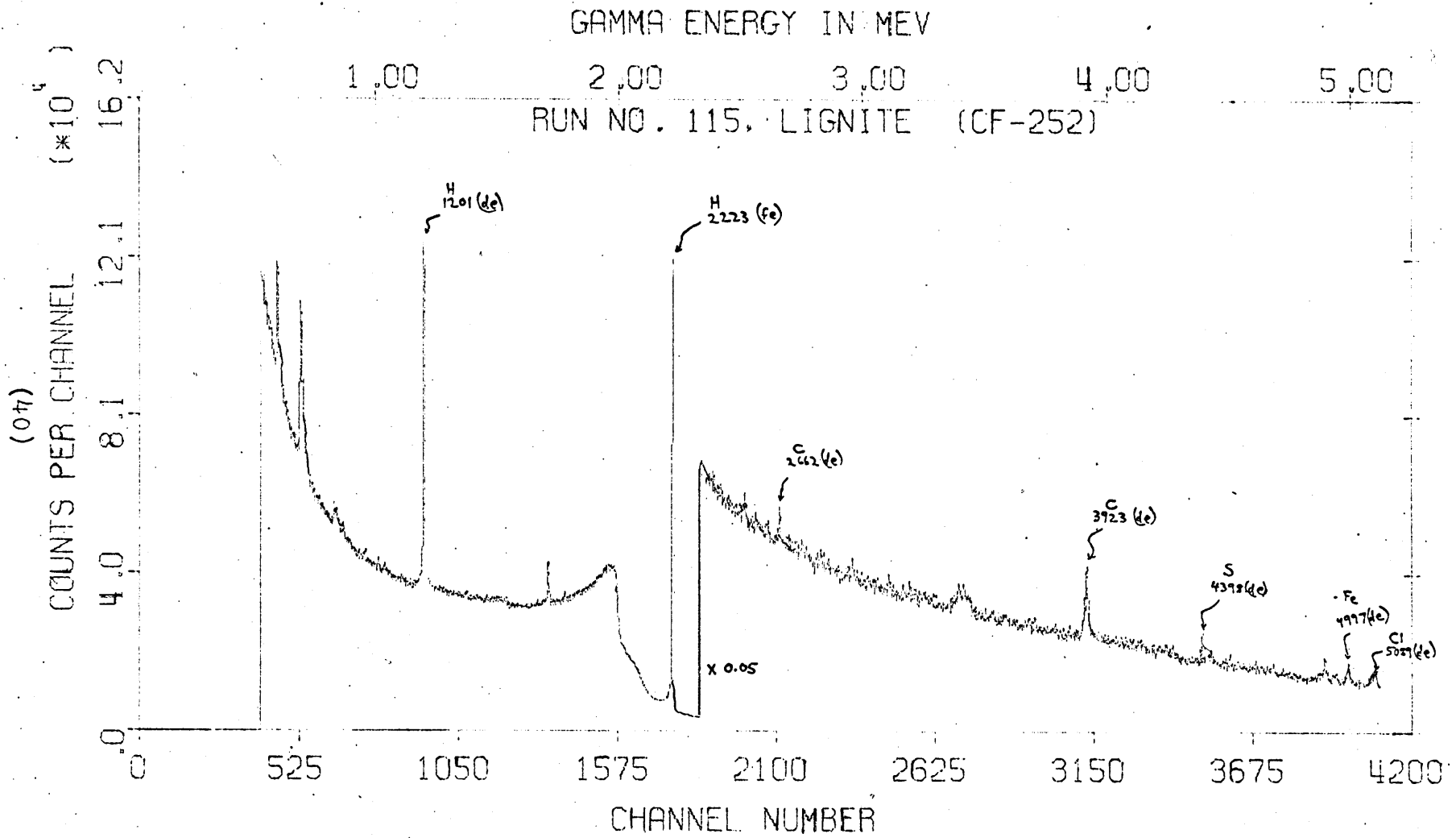


FIGURE A.3 LIGNITE SPECTRUM

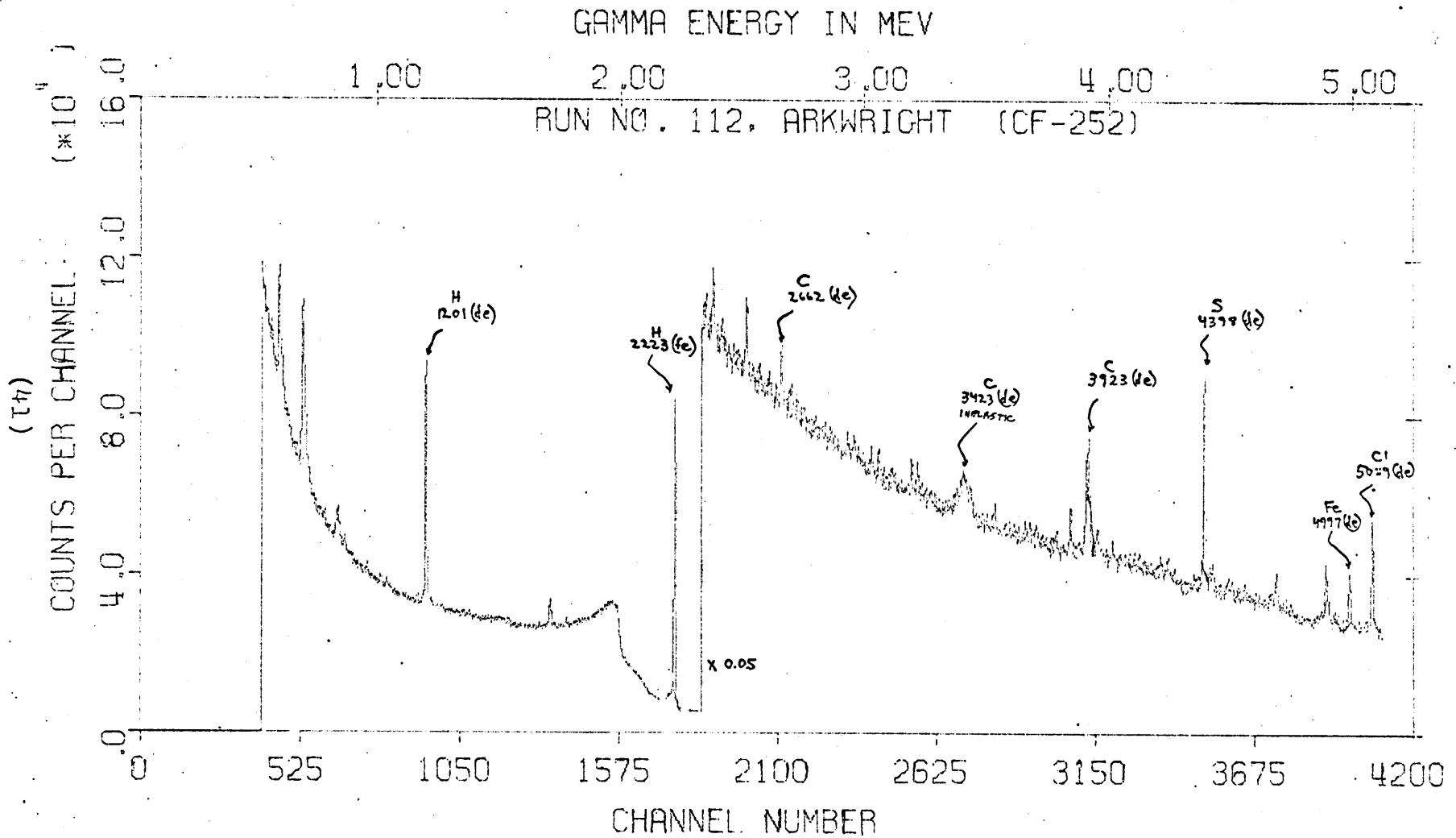


FIGURE A.4 ARKWRIGHT SPECTRUM

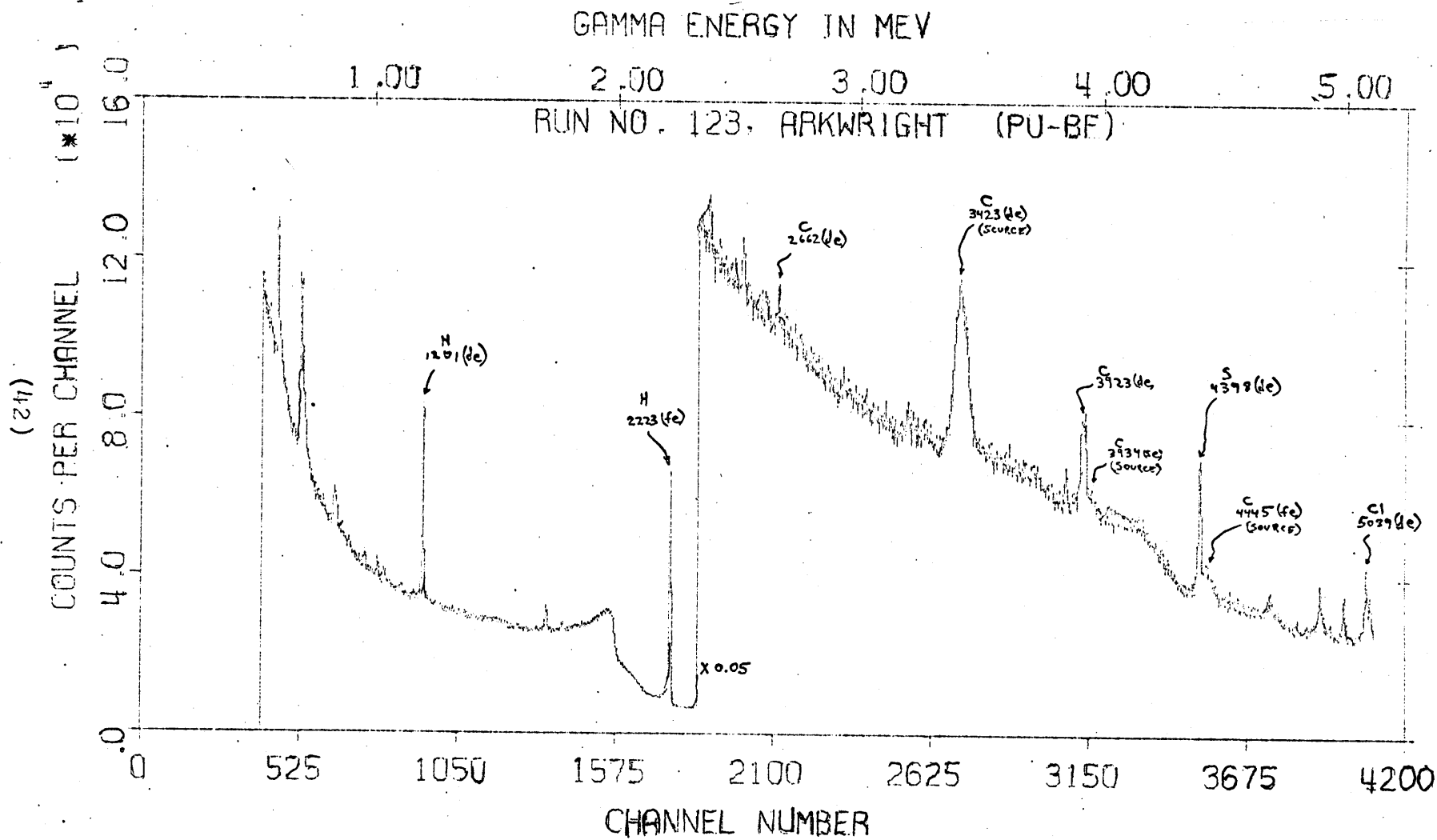


FIGURE A.5 ARKWRIGHT SPECTRUM (Pu-Be SOURCE)

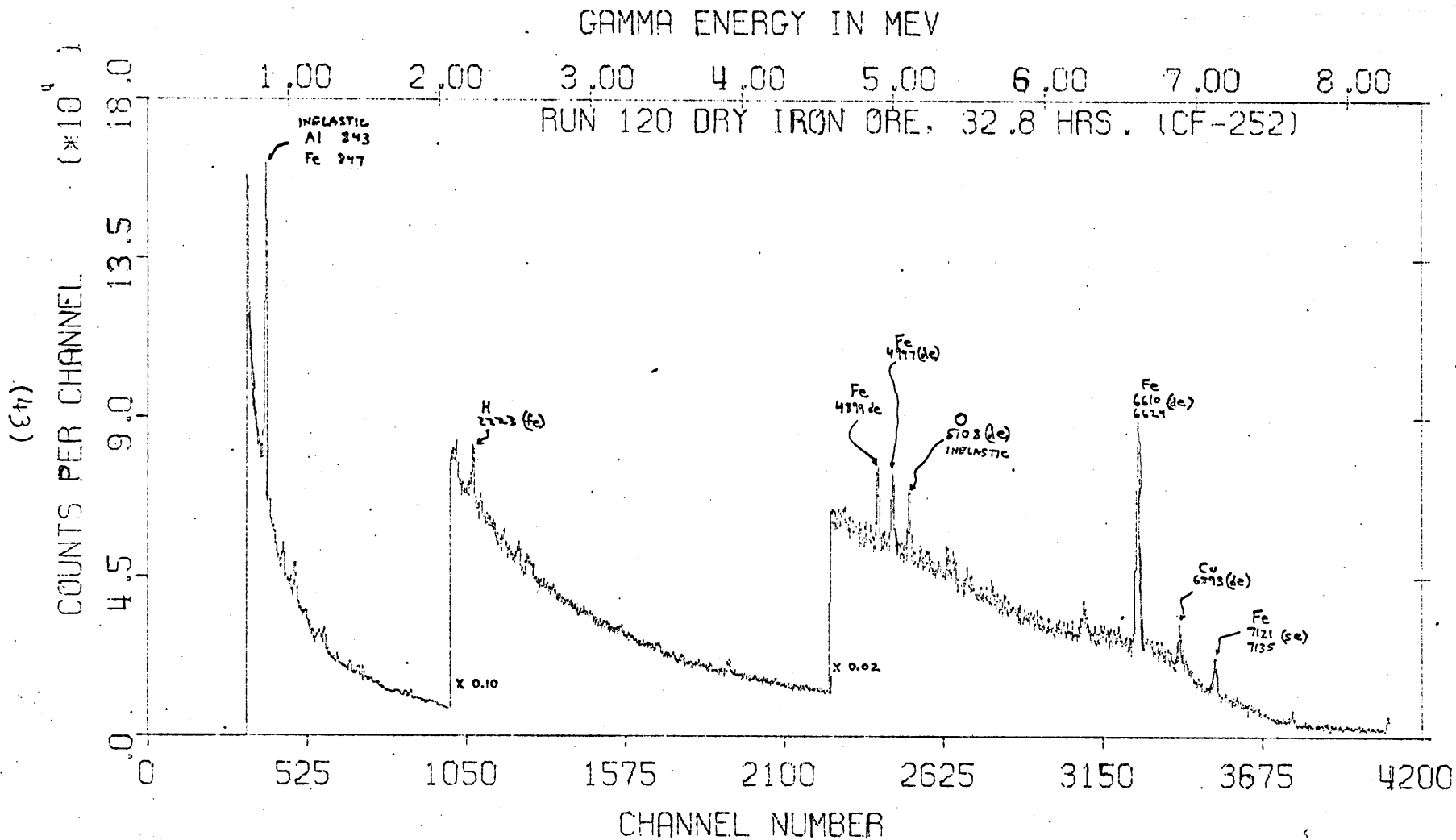


FIGURE A.6 DRY SINTERED IRON ORE SPECTRUM

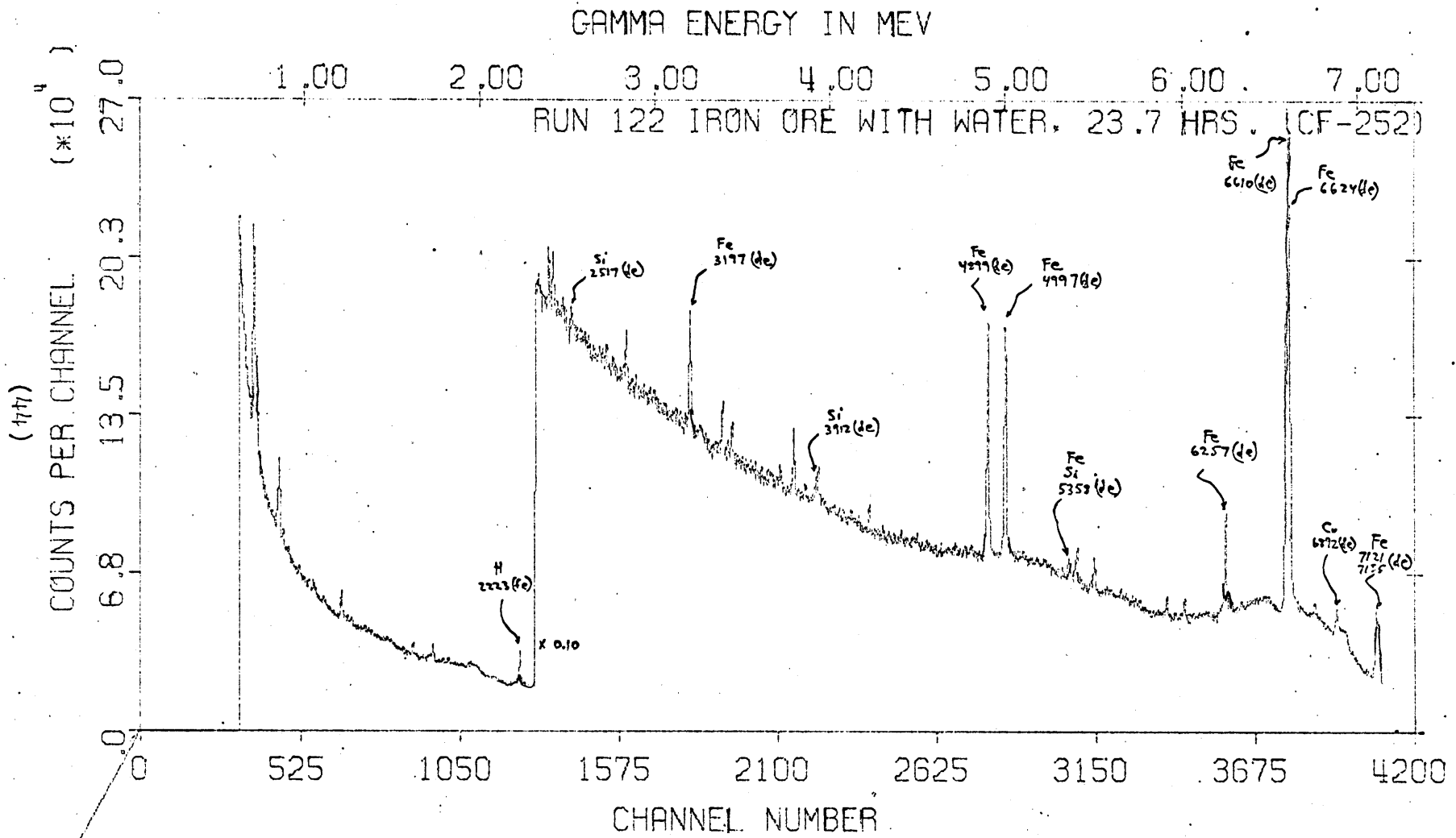


FIGURE A.7 WATER-SATURATED SINTERED IRON ORE SPECTRUM

Appendix B

MULTIPLETS IN THE COAL SPECTRUM

The interference with peaks of interest by nearby lines significantly affects the accuracy of the determination of how many counts are attributable to peaks of interest (Section 3.3). The extent of the interference also is a factor in determining whether a Ge(Li) detector is necessary or whether an NaI crystal, with poor energy resolution but high efficiency, can be used (Chapter 4). The multiplets containing the selected sulfur hydrogen and carbon peaks are now examined.

The 4398 keV (de) sulfur peak was interpreted as a singlet by GAMANL in all but two cases. In these, a very weak peak at 4384 keV was claimed, but a search through reference 10 showed no element which can account for it. Area errors greater than 30% constitute unreliable detection, and since the error for this peak was 33%, it is not believed to be real. Further support of this contention is that the peak did not appear in a GAMANL analysis of a different run using the same sample. The only other peak nearby is a single escape silicon line at 4423 keV. It only appears in the two types with the highest ash content, Arkwright and Robena, and is too weak and far removed to cause interference.

The 3923 keV (de) carbon peak appears in a doublet. The interfering peak is the 3912 keV (de) silicon peak with

an intensity about one half that of the carbon. A problem might be encountered if a source employing the (α , n) reaction of beryllium is used (Section 3.4). The 3934 keV (se) source line could interfere somewhat with this 3923 keV peak. The other carbon peak used, the 2662 keV double escape peak, appears as a singlet.

The 2223 keV (fe) hydrogen peak is very strong, but not completely isolated. In the two samples containing the largest amounts of sulfur, a 2199 keV (de) sulfur peak appears. It is only about 2% as intense as the 2223 keV peak. GAMANL also listed a peak at 2187 keV in the multiplet containing hydrogen. We have not identified its origin but it is so weak, only 0.7% of the 2223 keV intensity, that it is not important.

The peaks used for all three elements are prominent enough in their multiplets to make their analysis reasonably accurate. From the above, it appears that sulfur and hydrogen are sufficiently isolated to make possible the use of an NaI detector for them.



References

1. R. F. Stewart, "Nuclear Measurements of Carbon in Bulk Materials", ISA Transactions, vol. 6, no. 3, pp 200-208 (1967)
2. N. C. Rasmussen, "The Potential of Prompt Activation Analysis in Industrial Processing", Analysis Instrumentation, vol. 7, pp 186-192 (1969), available from Instrument Society of America. Presented at the 15th. AID symposium.
3. N. C. Rasmussen and Y. Hukai, "The Prompt Activation Analysis of Coal Using Neutron Capture Gamma Rays", Transactions of the ANS, vol. 10 (1), pp 29-30, (1967)
4. D. P. Simonson, "Prompt Gamma Ray Spectra Produced by Neutrons From a Pu-Be Source", S.M. Thesis, M.I.T., August 1968
5. B. H. Hui, "Study of Gamma Rays From Neutron Inelastic Scattering", S.M. Thesis, M.I.T., September 1969
6. V. J. Orphan and N. C. Rasmussen, "Study of Thermal Capture Gamma Rays Using a Lithium-Drifted Germanium Spectrometer", Report No. MITNE-80, January, 1967
7. T. Harper, T. Inouye, N. C. Rasmussen, "GAMANL, a Computer Program Applying Fourier Transformers to the Analysis of Gamma Spectral Data", Report No. MITNE-97, August, 1968
8. J. N. Hamawi, "Investigation of Elemental Analysis Using Neutron-Capture Gamma-Ray Spectra", Ph.D. Thesis, September, 1969
9. N. C. Rasmussen, Y. Hukai, T. Inouye, V. J. Orphan, "Thermal Neutron Capture Gamma-Ray Spectra of the Elements", Report No. MITNE-85, January, 1969
10. J. N. Hamawi and N. C. Rasmussen, "Neutron Capture Gamma Rays of 75 Elements Listed in Terms of Increasing Gamma-Ray Energy", Report No. MITNE-105, October, 1969