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The design, construction, and characterization of a facility
for neutron capture gamma ray analysis
of sulfur in coal using californium-252
by

James Robert Layfield

MS Thesis submitted to Iowa State University

Ames Laboratory, DOE

Iowa State University

Ames, Iowa 50011

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for neutron capture gamma ray analysis
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by

James Robert Layfield

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The design, construction, and characterization
of a facility for neutron capture gamma ray
analysis of sulfur in coal using californium-252¹

James Robert Layfield

Under the supervision of A. F. Voigt
From the Department of Chemistry
Iowa State University

A study of neutron capture gamma ray analysis of sulfur in coal using californium-252 as a neutron source is reported. Both internal and external target geometries are investigated. The facility designed for and used in this study is described. The external target geometry is found to be inappropriate because of the low thermal neutron flux at the sample location, which must be outside the biological shielding. The internal target geometry is found to have a sufficient thermal neutron flux, but an excessive gamma ray background. A water filled plastic facility, rather than the paraffin filled steel one used in this study, is suggested as a means of increasing flexibility and decreasing the background in the internal target geometry.

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I. INTRODUCTION

Political pressures (e.g. growing public dissension over the use of nuclear power and unrest in oil rich nations) have combined with economic pressures (e.g. increasing demands for decreasing supplies of oil and the effects of oil imports on a nation's balance of payments) to increase the use of coal as a source of energy in the United States. Its use, however, is not without disadvantages. Much of this country's coal, including nearly all that from Iowa, contains sulfur in sufficient concentrations to make its use environmentally unsound. America's energy needs, however, demand the use of all available grades of coal.

Physical methods are available for removing much of the mineral content of coal, including pyritic sulfur. Once processed, this coal can be used, either alone or mixed with a higher grade of coal, without violating federal restrictions on sulfur dioxide emissions. In order to evaluate the effectiveness of the cleaning process and to decide to what extent mixing may be necessary, a continuous knowledge of the sulfur content of the coal stream is necessary. Present methods for determining sulfur in coal require extensive sampling techniques and several hours to complete. These factors severely restrict their use in analyzing a moving process stream.

Prompt neutron gamma ray analysis using a portable neutron source like californium-252 has two significant advantages over the conventional methods of coal analysis. The continuous nature of the method makes possible the elimination of the delay between the time the samples are taken and the results are available. Current methods require the use of

complicated sampling procedures in order to eliminate problems associated with the inhomogeneous nature of coal. With prompt neutron gamma ray analysis, essentially the entire process stream can be examined and sampling errors are largely eliminated.

The goal of this work is to design, build, and characterize an off-line system for the analysis of sulfur in coal using californium-252 as a neutron source for prompt neutron gamma ray analysis.

II. THEORY

A. Californium-252

Element 98, called californium, was discovered in 1950 by G. T. Seaborg and coworkers (1) when they irradiated microgram quantities of ^{242}Cm with 35 MeV helium ions in the Berkeley 60-inch cyclotron. Each irradiation produced only about 5000 atoms of ^{245}Cf . Further experiments using cyclotrons produced other isotopes of californium, but all were on the neutron deficient side of the beta decay belt of stability. Experiments were begun in 1952 to produce usable quantities of neutron rich transplutonium elements. The process used involved the irradiation of ^{239}Pu in a neutron flux of 4×10^{14} neutrons/cm²-sec for several years. Using this method, californium isotopes 249 through 254 were eventually produced and identified.

Californium-252 was first isolated from the debris of the "Mike" thermonuclear explosion in 1952. Later studies, using samples from the reactor experiment described above, showed that it was produced from ^{239}Pu by 13 successive neutron captures. Using a thermal flux of 4×10^{14} n/cm²-sec it takes six years to produce one microgram of californium-252 from a gram of ^{239}Pu starting material.

The important nuclear characteristics of californium-252 are listed in Table 1. Prince (2) has presented a very complete review of these, along with the element's physical properties. It is the long half-life, along with the high output of moderately low energy neutrons, that makes californium-252 so useful. Sources of californium-252, unlike all other common isotopic neutron sources, produce neutrons by spontaneous fission.

Table 1. Nuclear properties of ^{252}Cf

Mode of decay	
Alpha emission	96.9%
Spontaneous fission	3.1%
Half-life	
Alpha decay	2.731 ± 0.007 yr
Spontaneous fission	85.5 ± 0.5 yr
Effective (α and SF)	2.646 ± 0.004 yr
Neutron emission rate	2.31×10^{12} n/(sec-g)
Neutrons emitted per spontaneous fission	3.76
Average neutron energy	2.348 MeV
Average alpha particle energy	6.117 MeV
Gamma emission rate (exclusive of internal conversion X-rays)	1.3×10^{13} photons/(sec-g)
Dose rate at one meter in air	
Neutron	2.2×10^3 rem/(hr-g)
Gamma	1.6×10^2 rads/(hr-g)
Decay Heat	
From alpha decay	18.8 w/g
From fission	19.7 w/g

The decay scheme of californium-252 is shown in Figure 1. Almost 97% of its decay is by alpha emission and the remainder by spontaneous fission. The decay constant for alpha emissions thus dictates the effective half-life, 2.65 years, of californium-252. The ratio of alpha decay to fission is rather small, 31.3, compared to 1260 for ^{250}Cf .

The fission neutron spectrum of californium-252 is similar to, though slightly more energetic than, that of neutron induced fission of ^{235}U . The fission process produces almost all of the neutrons emitted by californium-252. A small fraction result from (α, n) reactions on light elements in or near the source capsule. A list of the number of neutrons emitted at various energies by californium-252 is included in Table 2. The neutrons associated with this isotope can also be represented by equation 1 (3).

$$N(E) \propto 0.373 \exp(-0.88E) \sinh(2.0E)^{\frac{1}{2}} \quad (1)$$

where: $N(E)$ = probability of emission of a fission neutron with energy E

E = neutron energy in MeV.

The neutron dose rate from an unshielded californium-252 source can be calculated using equation 2.

$$D_N = \left[\frac{M}{4\pi R^2} \right] \left[\frac{2.31 \times 10^{12} \text{ neutrons}}{\text{sec-gram}} \right] \left[\left(\frac{1.22 \times 10^{-1} \text{ mrem-hr}^{-1}}{\text{neutrons-sec}^{-1}} \right) \text{cm}^2 \right] \quad (2)$$

where M = mass of californium-252 in grams

R = distance from source in cm.

Figure 1. Decay scheme for californium-252 (2)

Table 2. Spontaneous fission neutrons from Cf-252

Energy (MeV)	Abundance, neutrons/sec (g of nuclide) Cf-252 $\times 10^{10}$
0.3 - 0.4	9.4
0.4 - 0.6	19.
0.6 - 0.8	20.
0.8 - 1.0	17.
1.0 - 1.2	18.
1.2 - 1.4	17.
1.4 - 1.6	15.
1.6 - 1.8	14.
1.8 - 2.0	12.
2.0 - 2.2	11.
2.2 - 2.4	10.
2.4 - 2.6	10.
2.6 - 2.8	8.
2.8 - 3.0	6.8
3.0 - 3.2	6.2
3.2 - 3.4	6.1
3.4 - 3.6	5.9
3.6 - 3.8	4.6
3.8 - 4.0	5.0
4.0 - 4.4	6.4
4.4 - 4.8	5.4
4.8 - 5.2	3.9
5.2 - 5.6	3.1
5.6 - 6.0	2.3
6.0 - 6.4	1.8
6.4 - 6.8	1.3
6.8 - 7.2	0.93
7.2 - 7.6	0.69
7.6 - 8.0	0.58
8.0 - 8.0	0.62
8.8 - 9.6	0.18
9.6 - 10.4	0.19
10.4 - 11.2	0.13
11.2 - 12.8	0.088

The gamma ray activity of californium-252 originates in three processes, prompt gammas accompanying spontaneous fission, delayed gammas produced in the decay of the fission products, and the prompt gammas following alpha decay. The first two are more significant, and the number of photons of various energies produced by these processes are listed in Table 3. The total gamma ray dose rate from an unshielded californium-252 source in air can be calculated using equation 3.

$$D_G = \left[\frac{M}{4\pi R^2} \right] \left[\frac{2.31 \times 10^{12} \text{ neutrons}}{\text{sec-gram}} \right] \left[\left(\frac{8.58 \times 10^{-3} \text{ mrem-hr}^{-1}}{\text{neutrons-sec}^{-1}} \right) \text{cm}^2 \right] \quad (3)$$

where M = mass of californium-252 in grams

R = distance from source in cm.

In a typical source, on fabrication, only approximately 80% of the californium is californium-252, with 12% californium-250 and minor amounts of other neutron rich californium isotopes. As can be seen in Table 4, none of these other isotopes makes any significant contribution to the fission radiation (neutron and gamma) of the source. Californium-251 and californium-253 do not undergo spontaneous fission. The half-lives of californium-249 and californium-250 are so long as to preclude any appreciable contribution to the overall radiation count rate, and its short half-life means that small amounts of californium-254 present when the sources are fabricated decay quickly. Hence it is correct to assume that all source radiation originates from the spontaneous fission of californium-252, and to use the measured radiation levels to determine the weight of californium-252 present in the source.

Table 3. Gamma rays from spontaneous fission of Cf-252

Energy (MeV)	Abundance, Photons/sec (q of nuclide)		
	Prompt Gammas	Equilibrium Fission Product Gammas	Total
0 - 0.5	3.3×10^{12}	1.3×10^{12}	4.6×10^{12}
0.5 - 1.0	1.7×10^{12}	4.0×10^{12}	5.7×10^{12}
1.0 - 1.5	7.7×10^{11}	9.1×10^{11}	1.7×10^{12}
1.5 - 2.0	4.2×10^{11}	3.5×10^{11}	7.7×10^{11}
2.0 - 2.5	2.2×10^{11}		2.2×10^{11}
2.5 - 3.0	1.1×10^{11}		1.1×10^{11}
3.0 - 3.5	5.6×10^{10}		5.6×10^{10}
3.5 - 4.0	3.0×10^{10}		3.0×10^{10}
4.0 - 4.5	1.7×10^{10}		1.7×10^{10}
4.5 - 5.0	8.2×10^9		8.2×10^9
5.0 - 5.5	4.9×10^9		4.9×10^9
5.5 - 6.0	1.8×10^9		1.8×10^9
6.0 - 6.5	1.0×10^9		1.0×10^9

Table 4. Spontaneous fission half-lives of californium isotopes

Isotope	Spontaneous fission half-life
Cf-249	1.5×10^9 y
Cf-250	1.73×10^4 y
Cf-251	---
Cf-252	85.5 y
Cf-253	---
Cf-254	61.9 d

B. Neutron Shielding

Neutrons carry no charge, and hence do not interact with electrons. Instead, their interactions with matter are limited to nuclear effects. The neutron may be scattered elastically, captured by the nucleus, which then emits a particle or photon, or scattered inelastically. In the latter case the nucleus will emit a photon.

Fast neutrons lose large amounts of their energy through inelastic collisions, especially with heavy nuclei. This process is not, however, effective below intermediate energies, and will not slow neutrons to thermal energies. That process is accomplished by elastic collisions. Equation 4 shows the average energy retained by a neutron after an elastic collision with a nucleus of mass number A.

$$\bar{E} = E_0 \left[1 - \frac{2A}{(A + 1)^2} \right] \quad (4)$$

where $\bar{E}$ = average energy retained by neutron after one elastic collision

E_0 = initial energy of neutron

A = mass number of target nucleus.

It is evident from equation 4 that elastic collisions are most effective when A is small. Hence paraffin, water, or other hydrogenous materials are quite effective in slowing down neutrons.

The average number of elastic collisions necessary to lower the energy of a neutron from E_0 to E is given by equation 5.

$$\bar{n} = \frac{\ln (E_0/E)}{1 - \left[\frac{(A - 1)^2}{2A} \right] \ln \left[\frac{(A + 1)}{(A - 1)} \right]} \quad (5)$$

For collisions with hydrogen the denominator of equation 5 is equal to 1, and equation 5 reduces to equation 6.

$$E = E_0 \exp (-\bar{n}) \quad (6)$$

The average energy of a californium-252 fission neutron is 2.3 MeV and to reduce this to thermal energies, about 0.025 eV, requires approximately 18 collisions with hydrogen nuclei.

Once it has been thermalized, three possible fates await a neutron. It may be scattered, captured, or decay by the emission of a beta particle. The first two of these are more probable than the last, except in a vacuum. Whether the neutron is scattered or captured depends on the relative size of the cross sections for these processes in the material through which it passes. In a hydrogenous material the capture cross section for thermal neutrons is significantly lower than that for scattering thermal neutrons. As a result of this a thermal neutron will go through about 150 collisions before being captured. Other materials, such as D₂O and carbon (graphite) require more collisions than water to thermalize neutrons, but have still greater differences between their capture and scattering cross sections. Neutrons, therefore, require a greater thickness of these materials to reach thermal velocities but can also pass through greater thicknesses of the material after thermalization.

Often a hydrogenous material is doped with a material containing a nuclide with a very high capture cross section for neutrons. The nuclide most commonly used is ¹⁰B, usually in the form of boric acid, borax, or boron carbide. Its presence greatly reduces the number of collisions a

thermal neutron will have before it is captured. It also reduces the number of 2.23 MeV hydrogen capture gamma rays emitted by the shielding material, and replaces them with the 0.478 MeV boron prompt gamma ray. The ^{10}B reaction is $^{10}\text{B}(n,\alpha)^7\text{Li}$ rather than the more usual (n,γ) capture reaction and the high energy gamma rays usually seen in neutron capture are absent.

C. Gamma Ray Shielding

The stopping of gamma rays involves the conversion of electromagnetic energy into other forms of energy or matter. There are three important processes through which this is accomplished. These are pair-production, Compton scattering, and the photoelectric effect. Most often the first two of these reduce the energy of a gamma ray, and the remaining energy is dissipated through the third process.

Pair-production is a process in which 1.02 MeV of the energy of a photon is converted into a positron-electron pair. In order for momentum and energy to be conserved in this process it must occur in the field of a nucleus, which carries off some of the momentum and energy. Thus the larger the nucleus (one with the largest atomic charge Z), the greater the likelihood that this process will take place. In fact the cross section for this process is proportional to Z^2 . The cross section also increases with increasing photon energy, and this is the predominant process at high gamma energies.

The second process, Compton scattering, involves the transfer of a portion of a photon's energy to an electron with the reemission of a lower energy photon at an angle with the original. The electron,

originally considered at rest, moves with an energy equal to that lost by the photon. The relationship between the initial and final energies of the gamma ray and the angle between the original and final directions is given in equation 7.

$$\frac{1}{E_{\gamma'}} - \frac{1}{E_{\gamma}} = \frac{1 - \cos\phi}{E_0} \quad (7)$$

where E_{γ} = initial gamma ray energy

$E_{\gamma'}$ = final gamma ray energy

E_0 = rest energy of an electron (0.511 MeV)

ϕ = angle between original and final direction of gamma ray.

The Compton scattering per electron is independent of Z , and hence the scattering per atom is proportional to Z . The likelihood that this process will occur for photons above 0.5 MeV is approximately proportional to $E_{\gamma'}^{-1}$, and it is the predominant process for photons with moderate energies.

At low photon energies the photoelectric effect predominates. In this process energy is transferred completely, giving an electron ejected from an atom or molecule the energy described in equation 8.

$$E_e = E_{\gamma} - E_B \quad (8)$$

where E_e = energy of the ejected electron

E_{γ} = energy of the photon

E_B = binding energy of the electron.

Unlike the two previous processes, in the photoelectric effect the entire quanta of energy disappears. The probability of this process taking

place falls off rapidly as the photon energy increases. It is also approximately proportional to Z^5 . Except for heavy elements it is relatively unimportant for photons with energies above 1 MeV.

The likelihood that any of these processes will occur in a given thickness increases as Z increases. Hence heavy elements, usually lead, are used as gamma ray absorbers. In lead, the cross section for the photoelectric effect is greater than those of the other processes up to about 0.15 MeV. Between 0.15 MeV and 7.5 MeV, Compton scattering is the most likely mode of photon interaction. Above 7.5 MeV pair-production predominates (4).

D. Prompt Neutron Gamma Ray Analysis

The capture of a neutron by a nucleus results in that nucleus being elevated to an excited state. De-excitation may occur by the emission of one or more neutrons, a proton, an alpha particle, or a gamma ray. The most probable of these modes of de-excitation is the emission of one or more gamma rays. This process is a (n,γ) reaction, and the resulting gamma ray is referred to as a capture or prompt gamma. After the emission of the prompt gamma(s) the nucleus, now one neutron heavier than the original will frequently be radioactive, and decay by emitting a beta particle. This decay has a characteristic and measurable half-life, and may be accompanied by the emission of one or more gamma rays, which are referred to as delayed gammas.

Traditionally delayed gammas have been used to determine the elemental composition of samples in a method called delayed, or instrumental neutron activation analysis. Equation 9 describes the

relationship between the total disintegration rate from an isotope and the number of target atoms present in the sample.

$$R = N \phi \sigma (1 - \exp(-\lambda t_i)) \exp(-\lambda t_d) \quad (9)$$

where R = disintegration rate in counts per second

N = total number of target nuclei

ϕ = neutron flux in neutrons per square centimeter per second

σ = capture cross section in square centimeters

λ = decay constant ($0.693/t_{1/2}$)

t_i = irradiation time

t_d = decay time after irradiation period

$t_{1/2}$ = half-life.

When this equation is applied to capture gamma ray analysis, two important simplifications are possible. Because the half-life for prompt gamma emission is less than 10^{-12} sec, and the resulting decay constant greater than 10^{12} sec^{-1} , the $(1 - \exp(-\lambda t_i))$ term is equal to 1. Secondly, the $(\exp(-\lambda t_d))$ term is also equal to 1 because the measurements are made during the irradiation, and hence $t_d = 0$. The result of these simplifications is equation 10.

$$R = N \phi \sigma \quad (10)$$

Equation 11 shows the relationship between the number of target nuclei, N , and the more useful value W , the weight of the species of interest.

$$N = W N_0 / A \quad (11)$$

where W = weight in grams

N_0 = Avogadro's number

A = the atomic weight.

Substituting equation 11 into equation 10, and including an efficiency factor, E , for the detector used, one obtains equation 12.

$$\text{Count Rate} = \phi W E \sigma / A \quad (12)$$

One further term must be added to equation 12. The gamma ray yield, I , is the number of gamma rays of a certain energy emitted per neutron captured. The resulting expression, equation 13, relates the prompt gamma count rate to the weight of the element present. Since assignment of prompt gamma rays to particular isotopes has not been made, cross sections, σ , and gamma intensities, I , are on the basis of the element with normal isotopic abundance.

$$\text{Count Rate} = \phi W E (\sigma I / A) \quad (13)$$

Elkady (5) and Duffey et al. (6) have referred to the group of terms, $\sigma I / A$, as the "relative sensitivity index". The higher this value for a particular capture gamma ray the more sensitive the analysis. Duffey et al. (6, 7) have compiled an extensive list of these values, and have included them in tables of prompt gamma ray energies. A portion of those tables relevant to this work is reproduced in Table 5.

When samples and standards of similar composition are irradiated in equal fluxes, their prompt gammas counted using the same detector, and the same prompt gamma ray used for analysis of both of them, equation 13 simplifies to equation 14.

Table 5. Capture gamma-rays important to coal analysis

Element	Cross section σ , barns	Atomic weight A	E_{γ} MeV	I_{γ}^a	Relative sensitivity index ^b
H	0.33	1.008	2.223	100	32.7
C	0.0034	12.01	4.945	67.0	0.019
			3.684	31.8	0.009
Al	0.230	26.98	7.724	25.9	0.221
			7.695	5.10	0.043
			4.734	4.14	0.035
			4.259	4.82	0.041
S	0.512	32.06	8.641	2.66	0.042
			7.800	3.91	0.062
			5.420	59.1	0.944
			4.870	11.5	0.184
			3.221	27.1	0.433
			2.931	22.3	0.356
			2.380	44.5	0.711
Fe	2.62	55.85	7.647	23.9	1.12
			7.632	29.3	1.37
			6.018	8.71	0.409
			5.920	8.94	0.419
			4.219	4.33	0.203

^aPhotons per 100 neutrons absorbed.^bRelative yield per unit weight, $\sigma I/A$.

$$R \propto W$$

(14)

Prompt neutron gamma ray analysis is, therefore, usually done by measuring the prompt gammas from several standards and constructing a standard curve. Most often the count rate is replaced with the total number of gamma rays of a given energy collected over a certain length of time.

Prompt neutron gamma ray analysis is most useful for the analysis of process streams. The low neutron flux, typically six orders of magnitude lower than that inside of a reactor, associated with this method reduces its ability to compete with delayed neutron activation analysis when samples can be taken and then analyzed later. The few exceptions to this are those species, such as carbon, hydrogen, and sulfur, that produce isotopes upon neutron capture that do not yield usable delayed gamma rays. The method is, however, far more widely applicable to problems involving the continuous analysis of a moving process stream. In order to perform delayed activation analysis on such a stream two criteria must be met. First, the species being analyzed should have a high capture cross section for neutrons, so that a significant number of neutron captures will occur during the short residence time of any given nucleus in the irradiation chamber. Secondly, the half-life of the nuclide resulting from the neutron capture should be in the range of 1-5 minutes. If it is much shorter than this most of the delayed gamma activity will be gone before the sample can reach the detection chamber. If it is much longer than this too few disintegrations will take place before the sample flows out of the detection chamber. Few species of interest to industry meet both of these requirements.

Prompt neutron gamma ray analysis also requires a good "relative sensitivity index", but its continuous nature eliminates the problems associated with the half-life of the nuclide produced. Hence it is more widely applicable in industries which need constant information on the make-up of process streams.

III. LITERATURE REVIEW

A. Californium-252 Facilities

The wide variety of uses to which californium-252 sources have been or could be put were presented pictorially by Crandell (8) in the "Californium Use Tree" he introduced in 1968. The vast majority of these applications are based on the large number of neutrons that are emitted during the spontaneous fission of californium-252. These include most types of activation analysis, processes that rely on neutron scattering and thermalization, shielding studies, and tumor treatment. Other applications make use of the fission process itself. These include fundamental studies of fission, studies of materials and radiation damage, and heat sources.

Several good reviews of the development and applications of californium-252 have been published (9, 10, 11, 12). Permar (13) has attempted to evaluate the extent to which these applications will be utilized by industry, and what their economic impact may be in the near future. Californium-252 has found its way into almost all areas where research reactors, and neutron generators, as well as (α, n) and (γ, n) sources, have been used and its unique properties make it more widely applicable than any of them (14).

The numbers and types of californium-252 source facilities in existence are even greater and more diverse than the isotope's applications. They range from simple systems using common laboratory equipment to very complex structures that fill entire rooms. Many of these facilities are described in a recent review by Atkinson (15).

Perhaps the simplest system was described by Johnston (16), who has used 1-3 μg californium-252 sources contained in 500 ml beakers filled with paraffin for activation analysis. Samples of from 10 to 25 grams of several materials were placed in sample tubes cut out of the paraffin about 5 cm from the source. Sources as small as 1.0 μg have produced measurable activities in a wide range of materials, including ^{56}Mn in MnO_2 samples and ^{64}Cu in samples of copper. Liquid samples were irradiated by simply placing the source in a quartz tube that was then immersed in a beaker filled with the sample. Trombka et al. (17) have developed a system for calibration of thallium activated sodium iodide ($\text{NaI}(\text{Tl})$) detectors that is based on about 1.0 μg of californium-252. The source and the sample are set on a plastic frame, which is placed inside a large bottle that is filled with water. This bottle is then set within, and toward one side of, a cylindrical container about three times its volume filled with a water solution of borax. This solution shields the detector, as well as personnel, by thermalizing and capturing the source neutrons. The lack of boron in the inner vessel allows thermal neutrons to reach the sample. These neutrons cause the emission from the sample of prompt gamma rays which are measured with a $\text{NaI}(\text{Tl})$ detector mounted outside the container. Attempts to shield the detector from the very large fission gamma ray background proved to be impractical. Secondary gamma rays caused by neutron interactions with the shielding actually decreased, rather than increased, the signal to noise ratio.

At the other extreme, MacMardo and Bowman (18, 19) have described the californium-252 absolute activation analysis facility at the Savannah River Laboratory. This facility is capable of holding up to

100 mg of the isotope. It consists of a steel cylinder about four feet in diameter and five feet tall. This cylinder contains the sources and a pneumatic sample delivery system passes through it. The tank is placed in the center of a water filled pool that is 12 feet deep and 4 feet wide. The walls of this pool are made of one foot thick concrete. The pneumatic system is under automated, preset control and is capable of cyclic irradiation/decay/counting sequences while unattended. This allows analysis for species that produce short-lived isotopes upon activation. The system also makes use of the predictable and constant neutron flux produced by californium-252 to allow automatic calculations of the absolute concentrations of sample constituents directly from gamma ray spectra.

The facility at the Kossuth University in Debrecen, Hungary is similar in design to the Savannah River facility, but it is smaller and less sophisticated (20). Designed to hold up to 50 mg of californium-252, this system is simply a water filled steel cylinder 7.5 feet deep and 2.5 feet in diameter set in a 7.0 feet deep hole in the ground below the laboratory floor. The source is located in a lead block at the base of the tank during storage, and can be raised through an aluminum tube to the irradiation station about 2.5 feet above the base of the tank. Samples are lowered by hand into two other aluminum tubes parallel to the source tube. The distance between the sample and the source can be varied by moving the sample tubes horizontally along their cross-beam support guides. This system is limited to standard activation analysis methods, but does allow the analyst to tailor the neutron spectrum used to irradiate the sample by moving the sample tubes.

Despite the differences in their size and complexity, all of the facilities described thus far have several characteristics which allow them to be grouped together. Such "tank" facilities are generally cylindrical, with the source located along the central axis with several layers of shielding surrounding it. They are most often designed for flexibility and can be used for several types of experiments. For this reason many of the californium-252 facilities used for research fit into this category. The components of such a facility can be illustrated using the one designed by Aron (21) for the analysis of manganese in steels. The 1.0 mg source is contained in a rod that is located on the central axis of two concentric, cylindrical, steel tanks. The inner tank is 36 inches in diameter and 29 inches tall, while the same dimensions of the outer tank are 52 and 60 inches. The inner tank is filled with a water moderator. A sample tube runs through this tank parallel to, and one inch away from, the source rod. The outer tank is filled with a water solution of boric acid, and is surrounded by about 12 inches of concrete block. It is this pattern of source, moderator, neutron shield, and gamma shield, along with the familiar cylindrical, or nearly cylindrical, shape, that defines a "tank" facility.

Peterson (22) has described a similar facility that can hold up to 5 mg of californium-252. A plastic tank 5 feet in diameter and 4 feet tall is filled with water. The source is once again fixed in the center of the tank, and the cylinder is surrounded with 24 inches of concrete. In this system neutron shielding is provided by the water and concrete rather than boron. This system also allows the tailoring of the neutron flux and spectrum at the sample by moving the sample tube.

Three common modifications of the basic tank configuration are illustrated by the facility at the University of Texas at Austin designed by Gage et al. (23). This facility uses a solid neutron shield to increase structural integrity. The material used is a water extended polyester resin that is 30% polyester and 70% water. In order to increase the number of experiments that can be performed with the system, four beam ports of various sizes were included. These open ports pass from the outer surface of the facility to a central cavity about 20 inches tall and 16 inches in diameter with the source in its center. Various assemblies ranging from neutron collimators to pneumatically controlled sample holders are then fitted into these ports when they are needed. The cavity allows for the introduction of one or more additional layers of material between the source and the neutron shielding. These materials may include a moderator, a cadmium thermal neutron shield, additional gamma shielding, or some combination of these. Placement of an additional gamma shield between the source and the neutron shielding is common in tank facilities. The inner gamma shield is used to stop the source gamma rays, while the outer gamma shield stops the gamma rays from neutron capture.

NuTex Corporation has designed a facility that can be used for shipping as well as for experimentation (24). It is a cylinder 3 feet in diameter and 3 feet tall with an axial port that traverses the entire facility. The source is located in the center of this port, inside a cylinder of lead that fills the center one-third of the port. The rest of the facility is filled with Borotex^{tm}, which is the NuTex formula for a water/glycol extended polyester resin. The glycol is included to

depress the freezing point of the water, and hence prevent damage caused by the expansion of water as it freezes during shipping. This resin is doped with enough borax to yield 2 mg/cm^3 of ^{10}B and with enough Pb_3O_4 to yield 5% lead by volume. In this facility neutron and gamma shielding are provided by a single layer of material.

A similar facility has been described by Ricci and Handley (25, 26). The shielding material is concrete, and the facility has cubic, rather than cylindrical geometry. It is a dry box, 42 inches on a side, and contains two irradiation devices. The source may be placed in a 12 inch high density polyethylene moderator cube for a thermal neutron flux, or in a simple steel holder for a fast neutron flux. Gamma shielding is provided by 36 inches of concrete block on the sides and 12 inches on the top. Samples are delivered to the irradiation devices through a pneumatic system. The inside of the dry box is visible through a ZnBr_2 solution window, and the source is moved to the desired irradiation station using master-slave manipulators.

The shield designed by Dahlke (27) combines the typical layered shielding of a tank facility with the structural integrity and portability of the NuTex shield. The source is contained in a rotation mechanism that allows the operator to move it from its storage location to the beam port. In the storage position it is surrounded by a gamma shield of depleted uranium metal. The remainder of the nearly spherical shield is filled with polyethylene doped with 30% B_4C near the source, and 5% B_4C from about one foot from the source out to the surface of the shield. A steel shell provides the secondary gamma ray shielding. When rotated into

the beam port position, the source may be either bare, or surrounded by a polyethylene moderator.

Several factors must be considered when a californium-252 facility is being designed. The shielding must be sufficient to meet occupational safety standards. The experiments for which it will be used, as well as whether or not it will be used as a shipping container, also effect its design. The cost of materials, their weight and strength, and the ease with which they can be formed and shaped are also important considerations.

Several guides have been published to assist in the selection of shielding materials that best meet the needs of a given researcher, and help him decide on the necessary dimensions of his facility (28, 29, 30, 31, 32). These describe the various neutron and gamma shielding materials commonly used in californium-252 facilities. They also provide graphs or tables showing the dose rate of various forms of radiation at a given distance from the source through several single and multiple layer shields. In most cases these graphs or tables are theoretical, and are produced using computer codes such as ANISN (33) and DOT (34). Stoddard and Moyer have compared the predicted doses with experimentally determined ones, and have found that the measured values are often lower than expected (32). Browne et al. (35) examined the neutron attenuation properties of several materials using a Po-Be point source, and determined that a saturated $\text{Cd}(\text{NO}_3)_2$ solution offered the greatest stopping power.

Auxier et al. (36) have used a Po-Be source to compare the shielding properties of fiberboard, a polyethylene mixture made up of polyethylene resin, boric acid and fibrous strengthening agents, Bakelite, and paraffin borated with B_4C . They found that paraffin provided the best overall

shielding, but noted that it lacked the structural strength to meet impact requirements, and would not meet fire resistance requirements for source transportation containers. Bakelite offers slightly less shielding than paraffin, and it is also heavier and more difficult to shape, but it does meet government safety requirements. The reinforced polyethylene mixture is about the same weight as the paraffin, but the radiation level at the shield surface is 44% greater. This mixture is, however, somewhat easier to shape than Bakelite. Schamberger et al. (37) have investigated the secondary gamma ray production in shielding materials using the same type of source. Their results for iron, lead, and stainless steel compare well with those predicted using computer modeling.

Sharma and Hood (38) have studied the possibility of using polyethylene pellets as neutron shielding in californium-252 storage and transportation casks. The pellets used are about 3 mm in diameter, and have an effective thickness equal to 85% of that of solid polyethylene. It is, of course, much easier to produce a shield of the desired shape by filling a shell with these pellets than to machine pieces of solid polyethylene. Pellets impregnated with B_4C were also studied, and found to yield a radiation dose 40% of that from the same thickness of pellets without boron.

Ammonium metatungstate, $[(NH_4)_6H_{12}W_{12}O_{40} \cdot xH_2O]$, contains a substantial amount of both hydrogen and tungsten. This fact, along with its high solubility, suggests that it would be an effective shield for neutrons and gamma rays when mixed with water or paraffin. Senftle and Philbin (39) have studied this material, and have found that, as expected,

the presence of large amounts of the heavy metal tungsten made a nearly saturated solution (9.35 lbs/gal) of the metatungstate far superior to water as a gamma shield. They also found that the solution was a better attenuator of both slow and fast neutrons than water alone. The possibility of using solid ammonium metatungstate was also investigated. Although the solid contains less hydrogen, and is hence poorer neutron shields, it does not suffer from the possibility of evaporation or leakage. The radiation properties of high density concrete, a more common neutron and gamma shielding material, were examined by Northup (40). An aggregate that would retain 11.8-14.4 lb/ft³ of chemically bound water at 100°C was prepared. This water content is far above the average of 2 lb/ft³ for concrete.

The special shielding requirements and radiation properties of californium-252 therapy needles have been described by Wright (41). The californium-252 distribution in these needles result in their acting as line sources, rather than point sources. This, along with the fact that only 10% of californium-252's radiation is useful in therapy, greatly increases the shielding requirements over those of the more commonly used radium or cobalt sources.

When a californium-252 facility is designed for uses that require thermal neutrons, the selection of a moderator is quite important. The ideal moderator has two important characteristics, a large slowing down power, and a large moderating ratio (42). These are measures of how well the material slows down the neutrons, and how effectively this is done without neutron loss by capture. Unfortunately, no ideal moderator exists but they fall into two classes, each with one of the desired qualities.

Moderators that make use of the highly effective collisions with hydrogen to slow down the neutrons, such as water, paraffin and metal hydrides, suffer from hydrogen's moderate capture cross section. This results in a maximum in the plot of thermal flux versus moderator thickness at about 3-5 cm, and a rapid drop in the flux as the thickness is increased, since the neutrons are captured at a rate greater than the rate at which other neutrons are slowed. Materials with heavier atoms, e.g. carbon, beryllium, and heavy water are less effective in slowing down the neutrons, but capture fewer of the thermal neutrons. The result is a flux that never reaches the maximum seen for the hydrogenous materials, but falls off at a much slower rate as the thickness of the material is increased.

Shaw and Carson (43, 44, 45, 46) have evaluated several moderator materials for their usefulness in a californium-252 neutron radiography camera. Theoretical plots of thermal neutron flux versus moderator thickness for water, paraffin, carbon, beryllium, ZrH_2 , TiH_2 , and BeH_2 were generated using the ANISN (25) code, and are shown in Figure 2. As expected, thin layers of hydrogenous materials yielded the greatest thermal neutron flux. For this reason paraffin and ZrH_2 were studied in detail, and ZrH_2 was found to yield a flux approximately twice as large as paraffin in the optimum case. Forster and Menlove (42, 47) studied several moderators for use in a californium-252 facility for assaying fissionable material. Because the method used involves counting the fission neutrons emitted when the thermalized source neutrons strike the fissionable material, it is necessary to thermalize nearly all of the source neutrons, so that they are not counted by the detector. A

Figure 2. Thermal neutron flux from a californium-252 point source as a function of moderator thickness for several materials (43). Data were generated using the ANISN code (25)

multilayer moderator assembly was fabricated. A 2 cm thick tungsten shell was used to slow the neutrons by inelastic scattering, and as a gamma ray shield. Then a 4 cm thick layer of ZrH_2 , followed by a 6 cm layer of ZrH_2 with 20% paraffin was used to thermalize as many neutrons as possible. The final layer was 14 cm of D_2O , which allows many of the already thermalized neutrons to pass, while slowing down the remaining ones.

Longworth and Tobias (48) have studied the moderating properties and the flux distribution in a system made up of a lead sphere containing an Am-Be source and surrounded by water or polyethylene. This information was used to design the low gamma thermal neutron stack. Keller et al. (49) have studied the thermal neutron flux distribution 0-10 cm from the center of a californium-252 source using several common moderator-reflector combinations. The thermal flux distribution of a like source in a water moderator was measured by Magnuson (50), and was found to be in excellent agreement with the flux predicted by the ANISN (25) code. The angular dependence of the neutron emissions of a californium source has been reported by Kooi et al. (51). Kok et al. (52) have found that the material chosen to encapsulate the californium-252 has an appreciable effect on the thermal neutron flux distribution of the source.

Aside from its large slowing down power, several other properties make zirconium hydride a desirable material to use for neutron moderation. In its massive form it possesses more structural strength than any commonly used moderator. The term "massive" is used to differentiate these solid pieces from ones made using pressed powders. It also has a low thermal coefficient of expansion, and is stable at a much higher temperature than

hydrocarbon moderators. Its high density allows it to double as a gamma ray shield as well. Its major disadvantage is the great deal of effort necessary to produce it in a massive form.

A good early review of the physical and chemical properties of zirconium hydride, along with a description of how it is produced, was written by Vetrano (53). Beck and Mueller (54) have also thoroughly reviewed its properties and nuclear applications. Vetrano (55) and Merten (56) were both awarded early patents on procedures for the production of massive zirconium hydride. Most studies involving the production of massive zirconium hydride, especially in large pieces and pieces with complicated shapes, has been carried out by researchers at General Electric's Nuclear Propulsion Laboratory (57, 58, 59). This work has led to the development of small nuclear reactors for use in space vehicles and the like that are moderated with zirconium hydride or fueled with hydrided uranium-zirconium alloys.

The flux distribution within slabs of zirconium hydride, as well as the secondary gamma ray spectrum of the material, have been investigated by Western (60). He found that the material yields two prominent gamma rays on neutron capture, a hydrogen peak at 2.23 MeV and a zirconium peak at 6.30 MeV. Young et al. (61, 62, 63) have extensively studied the neutron thermalization process in zirconium hydride.

When a californium-252 facility is to be used for prompt neutron gamma ray analysis, neutron radiography, or other experiments that require neutron beams, the design of its beam port becomes especially important. The materials from which the port itself is built, as well as the collimators that are fitted into them, are determined by the planned applications of

of the facility, and the desired nuclear characteristics of the beam. Collimators for beams of thermal or fast neutrons and gamma rays have been used.

The use of collimators in prompt neutron gamma ray analysis with a californium-252 source has been investigated by Greenwood (64, 65, 66). He has described a neutron collimator for prompt neutron gamma ray analysis using the external target geometry, and a gamma ray collimator for use in the internal target geometry. Both consist of layered cylinders four inches in diameter with a two inch hole through most of their longitudinal axes. The source is two inches above the beam port and sealed in a polyethylene moderator. The external target geometry collimator begins below the center of the source, and the first six inches are made of lead with the axial hole increased to three inches in diameter. This is followed by four inches of bismuth, which keeps source gamma rays from reaching the sample and detector without attenuating the neutrons, and does so without adding to the gamma ray background. The lead prompt neutron gamma ray spectrum was found to add significantly to the background. Four inches of borated polyethylene, three cycles of two inches of lead and two inches of borated polyethylene, and finally four inches of polyethylene follow the bismuth. The lead keeps unwanted source gamma rays from reaching the detector, and the borated polyethylene prevents neutron streaming through the collimator. The internal target geometry collimator is similar in design. The initial layer with the enlarged hole is polyethylene to increase the thermalization of source neutrons. The gamma ray collimator is three inches longer than the neutron collimator so that the sample can be placed below the source. Following a layer of bismuth

are eight inches of borated polyethylene, four inches of lead, and ten inches of polyethylene. A four inch long borated polyethylene plug is fitted into the outer end of the axial hole. The purposes of these materials are the same as those in the neutron collimator. Far less lead is used, especially close to the outer end of the collimator, because of the secondary gamma rays that it produces. The borated polyethylene plug protects the detector from neutrons that have been able to pass through the collimator.

Greenwood's studies have shown that the large distance between the source and the sample in the external target geometry, and the resulting low thermal neutron flux at the sample location, makes prompt neutron gamma ray analysis using such a system difficult. He found that the internal target geometry, although less applicable to on line analysis, offered considerably better sensitivity.

In order to improve the sensitivity of the external target geometry, Greenwood proposed a californium-252 facility with the source surrounded by two inches of lead, twenty inches of D_2O , and then ten inches of light water. A converging collimator, twice as wide at its widest point as the distance between the source and the start of the collimator, would pass through both liquid layers. This design would make use of the high moderating ratio of the moderator, rather than its high slowing down power.

In order to improve the sensitivity of the external target geometry the form and characteristics of the neutron beam from isotopic sources must be understood. Ponomarev et al. (67) have studied the effects of several geometrical factors, including the location of the source relative

to the collimator, as well as the effects of several collimator materials, on the characteristics of the neutron beam. The neutron reflectivity of nickel has been investigated by Bandopadhyay (68), who found that a collimator lined with nickel will increase the neutron flux at the sample because of the high neutron reflectivity of nickel. Olsen and Mortensen (69) have designed a collimator for neutron radiography made of plastic drinking straws. The result is a beam that diverges slowly and provides easier focussing.

The problem of secondary gamma rays caused by the presence of fast neutrons in a thermal neutron beam has been considered by Brockhouse (70). He has shown that at low temperatures a perfect crystal composed of atoms which do not capture neutrons or scatter them incoherently can be used as a filter to discriminate in favor of slow neutrons. The total cross section of a crystal for slow neutrons consists of four principal components: neutron capture, incoherent scattering, Lave-Bragg scattering, and thermal diffuse scattering. The first two of those can be reduced by selecting the proper material. Lave-Bragg scattering has a negligible effect on the spectrum transmitted through a large perfect crystal. Thermal diffuse scattering is temperature and energy dependent, and approaches zero for low temperatures and neutron energies. Thus, at low temperatures, a properly selected perfect crystal will preferentially pass slow neutrons. Five single crystals of quartz with a total volume of 16.4 cm^3 and about 28 cm long, kept at liquid nitrogen temperature, were used in a neutron beam from a research reactor. Twenty percent of the neutron spectrum passed through the crystals with a cadmium ratio of 2400. The thermal neutron flux was reduced by a factor of 2.5, and the

fast flux by factors of between 25 to 2000, depending on the initial neutron energy.

B. General Analysis Applications

A majority of the research efforts with californium-252 involve its use in on-line and process control systems. Kirvan (71) has reviewed non-reactor activation analysis, including activation analysis using isotopic sources. The applications of delayed gamma neutron activation analysis have also been reviewed by Rhodes (72, 73, 74). O'Fallon et al. (75) have discussed instruments based on isotopic sources and their application to flowing streams.

Much of the research into the theory and mathematical basis of neutron activation analysis of a moving stream has been done by workers at the All-Union Scientific Research Institute on Radiation Technology in the Soviet Union. Ivanov et al. (76) have studied the problem of activation analysis of materials on a transporter from a mathematical standpoint. This analysis was used by Akopov et al. (77) to determine the optimum irradiation time to be used in activation analysis of flowing streams. These models have been used to establish detection limits for several elements in flowing streams (78, 79) and improvements in the selectivity of these systems have been described (80).

Until the advent of strong isotopic sources, such as californium-252, on-line neutron activation analysis was somewhat limited in scope. Few process streams flow sufficiently close to a nuclear reactor to make its use feasible. Neutron generators are sufficiently portable, but their use is limited by the high energy of the neutrons that they produce.

Gray and Metcalf (81) have described a system for the analysis of oxygen in process streams using a neutron generator. The problems associated with the use of neutron generator based activation analysis systems in the field have been considered by Iddings (82). A versatile system for on-line thermal neutron activation analysis of solutions, using a 6000 curie Sb-Be source and built by the Atomic Energy Commission of Canada, (83) has been used to determine over twenty elements in streams flowing from 0.5 gal/min to 8.0 gal/min.

Several systems using a californium-252 source for on-line or in situ delayed neutron activation analysis have been developed. The System for Investigations of Slurries Through Activation by Californium (SUSAC) was designed and tested by Braun and Riffel (84). This compact and portable facility can be connected directly to a slurry product stream and used as an open loop analyzer. The effects of flow rate, slurry density, and irradiation time have been investigated, and the system has been used to determine fluorine in calcium fluoride slurries. Rhodes (85) has described a closed loop slurry analyzer, which would first sample a process stream, then pass the sample through its irradiation and counting loop for an extended period of time. This system has been used to determine cobalt in solution. Meier et al. (86), Al-Jobori et al. (87), and Orange and Larson (88), have described systems for the determination of vanadium in various processed and crude oils. The choice of vanadium as a test element lies in its relatively large capture cross section for thermal neutrons and the half-life of the product nuclide ^{52}V , 3.76 min. This half-life is sufficiently long to allow the irradiated vanadium to flow from the

source to the detector before its activity is depleted, and short enough to result in a significant count rate and allow short counting times.

In situ delayed activation analysis of minerals, on land and under the oceans, is another widespread use of californium-252. Senftle et al. (89, 90) have used this isotopic source, as well as a neutron generator, to prospect for silver deposits on land. Perkins and Wagman (91, 92, 93) have developed an ocean floor mineral analyzer. This self-contained system can be controlled from a surface ship. The irradiation housing is set on the ocean floor to activate the minerals. Then it is rotated out of the way and the gamma ray detector assembly is rotated into position to measure the induced activity. Noakes et al. (94) have also used californium-252 for in situ analysis on the sea floor, but their system was operated by divers or from a submersible vehicle.

The use of prompt neutron gamma ray activation analysis has increased steadily since the mid 1960's. The introduction of californium-252 sources into widespread use has only served to speed up this increase. Typical of the prompt neutron gamma ray analysis applications using a reactor are the biological studies done by Comar (95). He observed chlorine, sodium, potassium, boron, sulfur, nitrogen, phosphorus, and calcium in samples of bone, blood and hair. Elkady and Duffey (5, 96) used this method to determine gold and other elements in ore-like mixtures. Najam et al. (97) have determined Fe, Cr, Ni, Mn, and B in stainless steels using a reactor neutron beam.

Prompt neutron gamma ray analysis has also been performed using (α, n) and (γ, n) sources. Jordan and Schierling (98) compared the gamma ray background of Pu-Be and Am-Be sources. They found that the Pu-Be

source has only one-fourth the gamma ray background of the Am-Be source. A Po-Be source was used by Allan et al. (99) to obtain the prompt gamma ray spectra of nineteen elements and compounds.

The application of prompt neutron gamma ray analysis using a californium-252 source to process streams has been discussed by Duffey et al. (100, 101). They described applications in the mining and processing of iron, copper, and nickel, and proposed an experimental arrangement that could be applied to these types of analyses. Wiggins et al. (102, 103, 104, 105) have used prompt neutron gamma ray analysis to study metallurgical and mineral samples and have also analyzed manganese nodules on a simulated sea floor. The elemental composition of the surface of the earth has been studied using prompt neutron gamma ray analysis and a californium-252 source by Trombka et al. (106). A 20 μ g source was placed directly into the earth's crust, and the resulting gamma ray spectrum collected. The data were used to calculate elemental abundances.

C. Nuclear Sulfur Analysis

There are several nuclear reactions that can be used to analyze for sulfur. Early attempts to do so involved using the $^{36}\text{S}(n,\gamma)^{37}\text{S}$ reaction with thermal neutrons (107). This reaction suffers from the low isotopic abundance (0.017%) of ^{36}S and its low neutron capture cross section, but the half-life of the product, 5.07 min, is favorable. The sensitivity of this method can be improved by using photoneutron counting (108). Following irradiation, the samples were placed at the center of a one liter polyethylene bottle containing heavy water and surrounded by a ring

of neutron detectors. The 3.1 MeV gamma ray from ^{37}S interacts with the heavy water producing a photoneutron. The detection of these neutrons is easier than the detection of the original gamma ray because the neutrons are not buried in a large and complex background spectrum. Despite its low sensitivity, this reaction has been used to analyze for sulfur in crude oil by several research groups (109, 110, 111, 112).

Another thermal neutron reaction used in sulfur analysis is $^{34}\text{S}(n,\gamma)^{35}\text{S}$ (113). Sulfur-35 is a weak beta emitter and the presence of any other beta emitters is likely to interfere with its determination. The isotopic abundance of ^{34}S (4.2%) is an improvement over that of ^{36}S , but is still rather low. Gruverman and Henninger (114) and Nadkarni and Haldar (115) have used the $^{34}\text{S}(n,\gamma)$ reaction to determine sulfur in steel.

Most delayed neutron activation analysis for sulfur uses the $^{32}\text{S}(n,p)^{32}\text{P}$ fast neutron reaction. While the cross section for this reaction is low, the isotopic abundance of ^{32}S is high (95%). The major problems associated with this method are that ^{32}P is not a gamma emitter, and that the $^{31}\text{P}(n,\gamma)^{32}\text{P}$ interferes. Despite such problems, the reaction has found wide application. It has been used to analyze for sulfur in iron (116), high purity selenium (117), water (118), snow (119), fluorocarbons (120), polysaccharides (121), and even beer (122). Less common nuclear reactions used to analyze for sulfur include ^3He activation ($^{35}\text{S}(^3\text{He},p)^{34m}\text{Cl}$) (123, 124) and proton activation ($^{32}\text{S}(p,n)^{32}\text{Cl}$) (125).

Sulfur can also be determined using prompt neutron gamma ray analysis. Greenwood et al. have measured the energy of the capture gamma rays from

sulfur (126, 127). Profio and Pouraghabager (128) used this method with a californium-252 source to determine sulfur in oil and Maerker and Muckenthaler (129) used a reactor neutron beam to determine sulfur in stainless steels.

D. Nuclear Coal Analysis

The use of nuclear techniques for the analysis of coal began in the early sixties. Most of this early work was done by researchers at the Texas Nuclear Corporation. Early experiments were based on fast neutron inelastic scattering and delayed fast neutron activation analysis using a 14 MeV neutron generator (130, 131). These first experiments provided moderately useful information about the oxygen, carbon, iron, silicon, and aluminum content of coal. They also indicated that it might be possible to determine sulfur in a coal process stream using the $^{34}\text{S}(n,p)^{34}\text{P}$ reaction. Further experiments showed that this method was useful only for order of magnitude estimates of sulfur content, but was accurate within 3% for carbon, oxygen, silicon, and aluminum (132, 133, 134). Gorin and Yavorsky (135) made a critical evaluation of the status of neutron activation analysis methods for coal analysis in mid 1964. They compared the American Society of Testing Materials' minimum allowed tolerances, as the standard deviation of a single analysis from the true average, with the standard deviations achieved at Texas Nuclear Corporation. They concluded that while the method held promise it had not yet met these minimum requirements. Further research led to a pilot plant facility (136, 137, 138) which was used to analyze for carbon and oxygen in coal using prompt neutron gamma ray analysis and for aluminum and silicon using delayed

activation analysis. The analysis of carbon in coal was further improved by the use of a split annulus of a plastic phosphor as an anti-coincidence shield to reduce the signal to noise ratio (139). As late as 1968 the attempts to determine sulfur in a moving coal stream using the (n,p) reaction showed little promise, having resulted in an average error of 100% (140).

The low neutron flux produced by the Am-Be source studied by Martin et al. (139) for the prompt neutron gamma ray analysis of coal, combined with the low sensitivity of this method for sulfur, precluded its use. Pak and Starchik (141) used a Po-Be source to determine the iron content of coal using capture gamma ray analysis. They attempted to correlate this with the sulfur content of coal, because much of the sulfur in coal is in the form of pyrite. The first successful measurement of the sulfur content of coal by capture gamma ray analysis was made by Rasmussen and Hukai (142) using the thermal neutron beam from a research reactor. Fay et al. (143, 144) have also measured the sulfur content as well as the iron, silicon, and moisture content of coal using a reactor.

The inapplicability of a reactor to on-line analysis, along with the low thermal neutron output of induced reaction neutron sources e.g. (α ,n), (γ ,n), (d,n), has limited their use in coal analysis. The result of this has been the widespread use of californium-252 in such systems. After his initial studies using a reactor (142), Rasmussen (145) used a small californium-252 source placed inside 55 gallon drums of coal. The prompt neutron gamma ray spectra were collected with a GeLi detector outside the drum, and were used to determine the sulfur content of coal using its 5.42 MeV gamma ray.

Duffey et al. have used a similar experimental arrangement. In early experiments a 20 μg source was placed in the center of a 9 inch by 9 inch annular aluminum can filled with a synthetic coal (146, 147). This synthetic "coal", made up of a polystyrene bead base and powders of Al_2O_3 , SiO_2 , S, $\text{Ca}(\text{OH})_2$, Fe, MgO, NaCl, K_2CO_3 , NH_4NO_3 , and TiO_2 , was used in order to eliminate problems of heterogeneity encountered in natural coals. The capture gamma spectra were collected with a GeLi detector about three feet away from the can, and shielded by 15 inches of heavy water. Further tests were performed using the same facility and several real coals (148).

T. Gozani et al. (149, 150, 151) have also studied this problem, using 60-100 Kg samples of coal contained in wooden boxes. The resulting capture gamma ray spectra were collected for 14 hours using a large GeLi detector and a 5" x 6" NaI(Tl) detector. The NaI(Tl) data was then unfolded using the MAZE (152) code and compared with the GeLi spectra. The information gained from these studies, along with that gained from a series of computer modeling studies, has been used in an attempt to determine realistic accuracy limits for the determination of concentrations of various elements in coal process streams using prompt neutron gamma ray analysis (153, 154). The result of this is a proposed prototype for a Continuous Nuclear Analyzer for coal (CONAC).

By far, the most successful research into the analysis of coal using nuclear techniques has been done by the workers at the Bureau of Mines Morgantown Energy Research Center. Their initial work in this field did not involve neutron activation analysis, but rather measured the moisture content of coal by counting neutrons from an Am-Be source

that had been thermalized while passing through a bin of flowing coal (155, 156). Another important undertaking of this group was the development of a nuclear sulfur meter for analyzing coal streams. Based on previous studies of the feasibility of determining sulfur in coal using capture gamma ray analysis a pilot plant was built (157). Coal is circulated through a conical bin via a conveyer belt. A californium-252 source is located at the center of this bin. With two single channel analyzers data from overlapping regions of the capture gamma spectra are collected in two channels (158) and the ratio of the counts in the channels is related to the percent sulfur in the sample. This method of data collection allows for the determination of sulfur without interferences from iron, aluminum, silicon or moisture (159, 160). The success of this pilot plant led to the installation of such a sulfur meter in a coal processing plant (161, 162). Coal from the process stream was diverted into the meter and passed through it at a rate of 7-10 ton per hour. The results obtained by the meter were compared with those obtained when samples collected every 2.4 minutes were analyzed using wet chemical methods. The tests showed that such a system had adequate precision to meet process control needs.

Other investigations in this area have centered on methods to reduce the gamma ray background and eliminate the need for the ratio method used by the Morgantown sulfur meter. Parsignault et al. (163) measured the gamma ray spectrum produced when a 12 μg californium-252 source was placed in a bin of coal with a 5" x 6" NaI(Tl) crystal as detector surrounded by a plastic scintillator. By counting only NaI(Tl) pulses not in coincidence with those from the plastic scintillator the

background due to the Compton process is reduced. Journey et al. (164) also used anti-coincidence spectrometry to determine sulfur in coal and other matrices. They used a GeLi detector surrounded by a NaI(Tl) annulus. Unlike all previous workers, they used small samples (300-600 mg) inserted directly into a reactor thermal column, and the lower energy sulfur capture gamma rays (at 841 and 2380 keV) for analysis.

IV. DESIGN AND CONSTRUCTION

A. Californium-252 Analysis System

An overhead view of the entire californium-252 system is shown in Figure 3. It consists of the storage-irradiation facility (SIF), the sample irradiation housing (SIH) and associated detector rack, concrete shielding, and an electronics rack. The system is located in the staging area of the Ames Laboratory Research Reactor (ALRR) building, and is surrounded on two sides by reactor airlock walls.

B. Storage-Irradiation Facility

The storage-irradiation facility (SIF) consists of three cylindrical casks fitted inside one another. Vertical and horizontal cross sectional views of it are shown in Figures 4 and 5 respectively. The outermost cask is called the storage cask, and provides most of the biological shielding. Its shell, and those of the other two casks, is constructed of 0.25 inch thick steel. The cask consists of two concentric cylinders, open at the top, and held together with steel supports so that their tops are even. The area between these cylinders is filled with paraffin impregnated with B_4C .

A beam port made of steel pipe passes through the paraffin in the storage cask. The major portion of the port is 11 inches long and 3 inches in diameter. The same dimensions of the interior portion of the port are 4 and 2 inches. A plug of high purity bismuth metal is fitted into this portion of the port. When not in use the rest of it is filled with a paraffin and steel plug. The internal target geometry detector housing is located above the beam port, and is described in section IV-H.

Figure 3. Overall view of californium-252 analysis system from above

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Figure 4. Vertical cross sectional view of the californium-252 Storage-Irradiation Facility (SIF) with the source in the storage position

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Figure 5. Horizontal cross sectional view of the californium-252 Storage-Irradiation Facility (SIF) with the source in the storage position. The internal geometry detector housing has been omitted for clarity

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The travel cask fits inside the central cavity of the storage cask. It makes it possible to transport the source short distances without excessive radiation dose rates or the bulk of the storage cask. The shell of the travel cask also consists of two cylinders open at the top. The smaller diameter cylinder is placed inside and along the edge of the larger one. The area around the outside of the smaller cylinder, and near the center of the larger one, is filled with lead shot. An aluminum sample tube, 1.5 inches in diameter, passes vertically through the travel cask. Except on the side facing the inner cylinder, the sample tube is surrounded by a ring of lead shot two feet high. The area inside this ring is filled with paraffin, while the remainder of the cask is filled with paraffin impregnated with B_4C .

The source rotation cask fits into the cavity of the travel cask, and pivots on a pin in the center of the cavity's base. Its shell is a single cylinder sealed at both ends. A pipe, 3 inches in diameter, passes vertically through the cask along its side. The source rod assembly described in section IV-E fits into this pipe. The area surrounding the base of the pipe is filled with lead shot, and the rest of the cask is filled with paraffin impregnated with B_4C .

When the source rotation cask is aligned as shown in Figures 4 and 5, the source is surrounded by lead shot and is in the storage position. When rotated 45° clockwise it can be used to irradiate samples for delayed gamma neutron activation analysis. When rotated 180° it can be used for prompt neutron gamma ray analysis or other experiments requiring a neutron beam.

C. Californium-252 Source

The californium-252 source used in the system was borrowed from the Oak Ridge National Laboratory. It is designated NSD-89 (89th neutron source that was doubly encapsulated in stainless steel), and contained 5.71 mg of californium-252 on May 1, 1978. The NSD-type encapsulation has been described by Van Cleve et al. (165) and the resulting capsule is shown in Figure 6.

The californium in this source is 85.2% californium-252, and 99.8% of its neutrons are emitted by that isotope. It is believed to be in the form of the oxysulfate, $\text{Cf}_2\text{O}_2\text{SO}_4$, as a result of the thermal decomposition of the Dowex-50 resin on which the californium was sorbed.

D. Zirconium Hydride Moderator

The cylinder of massive zirconium hydride used in the system was provided by the Advanced Energy Programs division of the General Electric Company. It was analyzed by the U. S. Geological Survey Laboratory in 1972 and found to be 2.0347% hydrogen by weight with a density of 5.632 g/cm^3 . This corresponds to 6.85×10^{22} hydrogen atoms/cm³ and a stoichiometry of $\text{ZrH}_{1.9}$.

The cylinder is 7.3 cm in diameter and 27.4 cm long with a 2.5 cm hole centered through its length. A hole 2.5 cm in diameter was drilled perpendicular to this axial hole from the outside of the cylinder to its center, to be used as a port for fast neutrons. This was done in a nitrogen atmosphere because of the explosive nature of finely divided zirconium hydride in air.

Figure 6. Californium-252 sources produced by the Oak Ridge National Laboratory: (a) Model NSS,
(b) Model NSD. ORNL Drawing No. 71-1678-R-3

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E. Source Rod Assembly

A stainless steel rod containing the californium-252 source, and the zirconium hydride moderator are the important components of the source rod assembly (SRA), which is shown from two different perspectives in Figures 7 and 8. The source rod was constructed in two sections, and a hole was drilled into one end of the top section. The source was sealed in the resulting cavity, with its threaded end pointing towards the top of the rod, when the bottom section was screwed into the hole.

Stainless steel pipes with 1/8 inch thick walls were fitted over the zirconium hydride cylinder and inside its axial hole, and joined together with endplates. The resulting "can" was longer than the zirconium hydride moderator and the area above that cylinder was filled with borated paraffin. The source rod is located in the hole that passes through the center of this moderator assembly, and its position within the source rotation cask is fixed. The moderator assembly is suspended from the top of the source rotation cask by two threaded rods, which can be used to raise it so the source will shine through the fast neutron port.

F. Sample Irradiation Housing

The sample irradiation housing (SIH) is employed when prompt neutron gamma ray experiments using the external target geometry are performed. Figure 9 presents an overhead view of the SIH with the top of the cave removed.

The frame of the SIF is a wooden cube two feet on a side. Wood was chosen to limit the background due to capture gamma rays from construction

Figure 7. Vertical cross sectional view of the source rod assembly (SRA)

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Figure 8. Horizontal cross sectional view of the source rod assembly (SRA)

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Figure 9. Overhead view of the external target geometry sample irradiation housing (SIH) with the cover removed

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materials. The faces of the cube are $\frac{1}{4}$ inch sheets of boral (a boron and aluminum composite) lined on the inside with $\frac{1}{2}$ inch sheets of Lexan plastic. A hole 5 inches in diameter was cut through the center of the face of the cube perpendicular to the beam and closest to the SIF. The inside of this face was then lined with lead bricks, again leaving a passage for the beam. The face nearest the detector rack has a nine inch hole at its center through which the detector housing is inserted into the cave. The portion of this face between the center of the hole and the SIF is lined with lead bricks, and the remainder with polyethylene bricks impregnated with B_4C . Both the inside and outside of the face perpendicular to the beam and farthest from the SIF are lined with borated polyethylene bricks. The inside of the remaining vertical face was lined with borated polyethylene bricks and the outside with high density concrete blocks.

A sample rotator, consisting of a small electric motor and a plastic turntable, sits in a hole at the center of the bottom face. Shielding at the top of the cave is provided by two layers of 1 inch thick polyethylene sheets impregnated with B_4C and containing 10% by weight of lead stacked above the boral sheet.

The open area between the cubic SIH and the cylindrical SIF was covered with several layers of the borated polyethylene-lead composite cut into the necessary shape. Bricks of this material were used to construct the base on which the cube rests. A lead beam stop and more concrete blocks outside the face of the cube opposite the beam complete the shielding.

Figure 10 is a side view of the external target geometry detector rack with the detector pulled back for clarity. The rack has three main components: the track assembly, the trolley, and the detector housing. The track assembly is constructed of wood supported by a steel frame. The space below the track is filled with lead bricks to keep the assembly stationary and prevent it from tipping when the heavy detector housing is located near one end of the track.

The trolley is constructed almost entirely of wood, with only the wheels and axles made of steel. A trench was cut in the bed of the trolley to keep the cylindrical detector housing from moving side to side. When the detector housing (described in section IV-H) is fully inserted into the SIH the front end of the trolley is flush against the SIH and the large endplate of the detector housing is flush against the other end of the trolley. In this position the center of the sample is 8.25 inches from the center of the NaI(Tl) detector. This distance can, of course, be increased by pulling the detector housing back as far as necessary.

G. Neutron Collimator

A neutron collimator was designed to fit inside the beam port to be used for prompt neutron gamma ray analysis in the external target geometry. It is a multilayer cylinder 2.95 inches in diameter held together with epoxy. Moving out from the source the collimator is constructed of 7 inches of masonite, 0.25 inches of boral and 4 inches of lead.

Figure 10. Side view of the external target geometry detector rack

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A port through the collimator increases from 0.75 inches to 1.25 inches in diameter as shown in Figure 11, and is off center. At its widest point the edge of this port reaches the central axis of the collimator. The cylinder is fitted into the beam port so that the hole through it is below the californium-252 source.

H. Internal Target Geometry

The internal target geometry detector housing shown in Figure 4 was not part of the original SIF. Initially the area above the beam port was filled with borated paraffin. The first internal target geometry arrangement used is shown in Figure 12. A vertical hole 2 inches in diameter was cut through the paraffin from the top of the SIF to the beam port. A 22 inch tall stack of lead bricks, 12 inches square with a 4 inch square void through its center, was placed above this hole. The detector housing was positioned on top of this structure. In this arrangement the sample is placed inside the beam port, and its center is 56 inches from the center of the detector.

The second internal target geometry arrangement was more permanent, and reduced the distance between the sample and detector. Figure 13 is a vertical cross sectional view of this arrangement.

The portion of Figure 13 inside the $\frac{1}{4}$ inch boral sheet is the same detector housing used in the external target geometry detector rack and the first internal target geometry arrangement. The NaI(Tl) crystal and photomultiplier tube are set inside a cylinder of lead, and are held in place by a wooden ring. When a 3" x 3" crystal is used, as in Figure 13, a ring of Lexan surrounds the detector to keep it stationary.

Figure 11. Longitudinal section of the external target geometry neutron collimator

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Figure 12. Side view of the initial internal target geometry arrangement

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Figure 13. Vertical cross sectional view of the final internal target geometry arrangement

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The rest of the second internal target geometry arrangement is a lead filled aluminum structure called the internal detector support (IDS). The IDS surrounds the detector housing and the hole through which the capture gamma rays pass with from 1.5 to 2.0 inches of lead. The detector housing is set into the central cavity of the IDS above as many lead plates as it takes to reduce the gamma ray background to a usable level. These plates are rings of lead 8.9 inches in diameter with a 3.0 inch hole through their centers, and replace the lead bricks used in the first internal target geometry arrangement. A piece of aluminum pipe with an internal diameter of 2.75 inches, is permanently fitted into the center hole of each ring. A 2.75 inch hole was cut through the top of the beam port so that there is no steel between the sample and the detector. The shortest distance between the centers of the sample and detector in this arrangement is 14 inches, when no lead plates are used.

1. Instrumentation

Two detectors were used to collect capture gamma ray spectra. A 4" x 4" Harshaw Chemical Company NaI(Tl) crystal coupled to a matching photomultiplier tube was used for most of this work. This detector had an experimentally measured resolution of about 150 KeV full width at half maximum (FWHM) at 5 MeV. Some measurements involving the second internal target geometry arrangement were made with a 3" x 3" Harshaw NaI(Tl) crystal and matching photomultiplier tube. Its resolution was much poorer than that of the 4" x 4" crystal.

The signals from these detectors were amplified and fed into one of two pulse height analyzers, usually a Technical Measurements Corporation Model 1001 pulse height analyzer with 1024 channels. The data from this analyzer were transferred to punched paper tape for computer reduction. The capture gamma ray spectra included in this thesis were collected using a 4096 channel Nuclear Data Incorporated Model 50/50 pulse height analyzer. Data from this system were transferred to magnetic tape for computer reduction.

V. CHARACTERIZATION

The purpose of this work was to design, construct, and characterize a system capable of determining the sulfur content of coal by prompt neutron gamma ray analysis using californium-252. In addition to an external and two internal target geometry arrangements for this purpose, a sample tube to be used for delayed gamma activation analysis was included in the facility. Characterization of the system consisted of measuring the radiation dose rates around the facility, the neutron flux distribution at the various sample irradiation positions and the capture gamma ray spectra at those locations.

A. Radiation Survey

The radiation dose rates measured at the locations near the californium-252 system marked in Figure 14 are listed in Table 6. The beta and gamma ray dose rates were measured with a Geiger-Müller counter and the neutron dose rates with a BF_3 detector. A BF_3 detector responds only to neutrons with intermediate to thermal energies. When it is covered with a borated polyethylene sleeve, the thermal neutrons present are captured before reaching it, while the fast neutrons are slowed and then counted. When no sleeve is used, the intermediate dose rate is measured.

The low dose rates outside the face of the SIH opposite the detector were expected because it is covered with a concrete block wall. The high beta and gamma ray dose rate near the center of the detector face of the SIH is due to the lack of any shielding outside of the boral sheet. The dose rates at locations 4 through 9 show radiation leaks around the

Figure 14. SIH radiation survey locations: A. face opposite detector, B. detector face,
C. top face, D. rear face

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Table 6. Radiation dose rates in the californium-252 system

Survey location number ^a	Radiation dose rate (mR/hour)		
	Fast neutrons	Intermediate neutrons ^b	betas and gammas
1	15.0	27.0	15.0
2	36.0	46.0	12.0
3	--	--	125.0
4	28.0	34.0	50.0
5	13.0	55.0	60.0
6	50.0	55.0	60.0
7	32.0	40.0	80.0
8	45.0	50.0	68.0
9	28.0	34.0	50.0
10	65.0	90.0	19.5
11	10.0	8.0	13.0
12	5.8	8.0	12.5
13	10.0	10.0	7.5
14	5.8	8.0	13.5

^aNumbers refer to locations marked in Figure 14.

^bThermal and epithermal neutrons.

edge of the SIH which are due to the difficulty associated with connecting a cube to a cylinder. The lack of any shielding inside the Lexan sheet at the top face of the SIH explains the neutron dose rates seen at location 10.

B. External Target Geometry

Measurement of the neutron flux distribution in the beam port was the first step in the characterization of the external target geometry arrangement for prompt neutron gamma ray analysis. These and all other neutron flux measurements were made using the gold foil activation method. Small gold foil disks sealed in aluminum or cadmium were irradiated for one hour. The aluminum cover passes the entire neutron spectrum while only those neutrons with energies above about 0.6 eV pass through the cadmium cover. The nuclear reaction involved is $^{197}\text{Au}(n,\gamma)^{198}\text{Au}$. The ^{198}Au decays by beta emission with a 2.698 day half-life, and these betas were measured using a gas flow proportional counter. The measured count rate is then compared with that produced by the known flux of a National Bureau of Standards reactor using equation 15.

$$F = \left[\frac{C k_a k_s}{m} \right] \left[\frac{1}{1 - N_i} \right] \left[\frac{1}{N_w} \right] \quad (15)$$

where F = neutron flux

C = beta count rate in counts/sec

k_s = constant set by foil position in counter (equals 1 for top shelf)

m = mass of gold foil in mg

k_a = proportionality constant found by irradiating foils in known flux at NBS (1.13×10^4)

$1/1 - N_i$ = buildup factor (93.9 for a one hour irradiation)

$1/N_w$ = decay factor (taken to be 1 for decays of less than 15 minutes).

The "Cadmium Ratio" is a ratio of the flux measured with an aluminum cover to that measured with a cadmium cover. It approaches 1 for a purely fast neutron flux and infinity for a purely thermal flux.

The fluxes measured at various locations in the beam port with the source lined up with the zirconium hydride are listed in Table 7. The thermal neutron flux was calculated by subtracting the flux measured with a cadmium cover from that measured with an aluminum cover. It is apparent from Table 7 that the flux in the beam port decreases at a rate somewhat faster than that explained by the increase in the distance from the source. This additional decrease is due to the scattering of neutrons by the air in the beam port. The cadmium ratio also decreases as the distance from the source increases because those neutrons already thermalized are being scattered out of the port and are not being replaced by thermalization of the fast neutrons. The flux, as expected, is greatest in the center of a cross section of the port.

The flux at the sample location of the external target geometry arrangement could not be measured effectively with the gold foils, as they are not sufficiently sensitive. Using the flux at the center of the port 16.5 inches from the source to estimate the source strength, and assuming no further scattering, the total flux at the sample location, 28.5 inches from the source, would be 3.4×10^4 n/cm²-sec and the thermal

Table 7. Neutron flux distribution in the californium-252 storage-irradiation facility beam port

Distance from source, in	Location ^a	Total neutron flux ^b n/cm ² x sec	Thermal neutron flux ^b n/cm ² x sec	Cadmium ratio
5.5 ^c	left	2.5×10^6	1.5×10^6	2.4
5.5	center	3.1×10^6	1.9×10^6	2.5
5.5	right	2.5×10^6	1.5×10^6	2.5
12.5	left	3.6×10^5	2.0×10^5	2.2
12.5	center	4.0×10^5	2.2×10^5	2.2
12.5	right	3.7×10^5	1.9×10^5	2.1
16.5 ^d	center	1.0×10^5	4.2×10^4	1.7

^aPosition on the horizontal diameter of the beam port as seen when looking towards the source.

^bNeutrons/cm²-sec.

^cPoint in 3 inch diameter section of the beam port closest to the source.

^dPoint in 3 inch diameter section of the beam port farthest from the source.

flux 1.4×10^4 n/cm²-sec. As seen in equation 15, this would correspond to less than 1 count/sec, which is less than the background and not measurable by this method.

The gamma ray spectrum shown in Figure 15 was collected for 10^4 seconds of live time with an empty polyethylene bottle in the sample location of the external target geometry and the detector housing inserted as close to the sample as possible. Four peaks are visible, and these are identified as coming from structural steel and aluminum in the SIF. Figure 16 shows an expanded view of the same spectrum below 1 MeV and collected for 3600 seconds of live time. The only peak that can be assigned to any material in the SIF combines the 0.476 MeV boron peak and the 0.511 MeV annihilation peak. The other features are part of the complex californium-252 source spectrum. Figure 17 shows the gamma ray spectrum collected for 10^4 seconds of live time with a polyethylene bottle filled with sulfur in the sample location. It is essentially identical to Figure 15, and no sulfur peaks are visible. The low thermal neutron flux at the sample location of the external target geometry arrangement makes the determination of sulfur by prompt neutron gamma ray analysis essentially impossible using that arrangement.

In order to determine whether the large background spectrum at the sample location was due primarily to source gammas or fast neutron interactions with structural materials, a steel plate was placed across the SIH perpendicular to the neutron beam and passing through the sample location. Spectra collected with and without the plate showed that the background increases considerably when the extra steel was present but the shape of the background spectrum did not change. This would indicate

Figure 15. Neutron capture background spectrum (0 - 9 MeV) collected using the external target geometry arrangement

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Figure 16. Neutron capture background spectrum (0 - 1 MeV) collected using the external target geometry arrangement

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Figure 17. Neutron capture spectrum of sulfur (0 - 9 MeV) collected using the external target geometry arrangement

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that much of the background is due to fast neutron interactions with the steel in the SIH. Most of the source gammas are removed by the bismuth plug in the beam port. When bottles filled with paraffin wax were placed in the beam port the fast neutron flux was greatly reduced, and so was the gamma ray background.

C. Neutron Collimator

The neutron collimator was designed to reduce the gamma ray background at the sample position in the external target geometry arrangement. It was expected that by using a port offset from the source the gamma ray background from the source would be reduced. The gamma ray background produced by fast neutron interactions with structural materials would also be reduced because most of the neutrons that approach the sample location would be moderated while passing through the masonite.

The flux data shown in Table 8, when compared with that in Table 7, show that the fast neutron flux at the sample and of the port has been reduced. The thermal neutron flux, however, has been reduced to an even greater extent, and the cadmium ratio has therefore been reduced. The thermal flux has, in fact, been reduced so much that it approached the lower limits of detection by gold foil measurements. Many of the neutrons thermalized by the zirconium hydride would have to pass through the borated paraffin that surrounds the narrow end of the beam port in order to reach the offset port of the collimator. Their capture by the boron explains the observed reduction in the thermal flux.

In order to be effective the collimator would have to drastically reduce the gamma ray background in the SIH without lowering the already

Table 8. Flux characteristics of the neutron collimator

Total neutron flux ^a	2.1×10^4 neutrons/cm ² -sec
Fast neutron flux ^a	1.8×10^4 neutrons/cm ² -sec
Thermal neutron flux ^a	2.9×10^3 neutrons/cm ² -sec
Cadmium ratio	1.2

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^aMeasured in the center of the port of the neutron collimator
16.7 inches from the source.

marginal thermal neutron flux. Prompt neutron gamma ray analysis of sulfur in coal is, therefore, impossible using the external target geometry arrangement of this system and this type of offset neutron collimator.

D. First Internal Target Geometry Arrangement

The first internal target arrangement was designed to examine the usefulness of such an arrangement for the analysis of sulfur in coal. As such, it was simple in design and temporary in construction. It is important that the distance between the sample and detector be kept as small as possible while keeping the detector live time at a usable level. The intensity of the capture, and all other, gamma rays at the detector decreases as the square of the distance they must travel to reach it. A large dead time increases the length of time necessary to collect a usable spectrum. In addition, the large background that causes the large dead time obscures the gamma rays of interest.

The live times measured with various thicknesses of lead between the detector housing and the top of the SIF are listed in Table 9. Initial measurements were made with the source in the storage position and before the 2 inch diameter hole was drilled through the wax above the beam port. In this position the source is surrounded by lead shot and, as seen, the gamma ray background is relatively small. The detector live time decreased dramatically when the source was moved into the beam position, as the source is both closer to the detector and no longer surrounded by lead. A considerable increase in the live time is seen when the lead thickness is increased from 10 to 12 inches. The number of gamma rays observed, not their energy, determines the live time of the detector. It

Table 9. Effect of lead thickness on detector live time in the first internal target geometry arrangement

Height of lead ^a	Live Time		
	Source in storage position Through solid wax	Source in beam port position	
		Through solid wax	Through 2" hole
2	80%		
4	90%		
6	94%		
8	99%	42%	
10		58%	
12		82%	
14		85%	
16		92%	82%
18			86%
20			90%
22			94%

^aHeight of lead structure above the SIF in Figure 12 in inches.

is apparent that 12 inches of lead is sufficient to stop most of the background gammas while 10 inches reduces the energy of most of these gammas but is not able to stop them completely. A decrease in detector live time of about 10% is seen when the hole is drilled in the wax. The poor gamma ray shielding characteristics of low Z materials is evidenced by the fact that 6 additional inches of lead more than replaces the 30 inches of wax that were removed.

The center of the sample location in the first internal target geometry arrangement is 3 inches from the outer end of the bismuth plug. Its neutron flux characteristics, when the source is looking through the hydride, are listed in Table 10. The thermal flux is comparable to that used by Fay (144) to measure the sulfur content of coal, but the fast flux is much greater and the cadmium ratio much lower than he encountered. Fay's detector was 4.5 inches from the sample, while they are 56 inches apart in this arrangement.

Capture gamma ray spectra were collected with an empty polyethylene bottle and a bottle filled with sulfur in the sample location. No sulfur peaks were seen, but small changes were apparent in the spectra. The addition of sulfur caused small increases in the intensity of the spectra over a broad energy range below the energy of the expected peaks. It is apparent that the sulfur gamma rays are being scattered while traveling the 56 inches to the detector, and that this distance must be decreased in order to make use of the internal target geometry to determine sulfur in coal.

Table 10. Flux characteristics of the internal target geometry sample location

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Total neutron flux	1.3×10^6 neutrons/cm ² -sec
Fast neutron flux	5.2×10^5 neutrons/cm ² -sec
Thermal neutron flux	7.8×10^5 neutrons/cm ² -sec
Cadmium ratio	2.5

E. Second Internal Target Geometry Arrangement

The second internal target geometry arrangement is more complex in design and permanent in construction than its predecessor. It decreases the distance between the detector and the sample and surrounds the entire length of that pathway with lead. Its sample position is the same as the first internal target geometry arrangement, and hence Table 10 applies equally to both of them.

The live time of the detector as a function of the thickness of the lead plates between the sample and the detector is listed in Table 11. These plates were cast and machined until smooth, which accounts for their apparent random thicknesses. In the first internal target geometry arrangement 18 inches of lead yielded 86% live time. The second internal target geometry arrangement brings the detector closer to the source. The expected increase in the intensity of the gamma ray background is evidenced by the fact that 18.4 inches of lead (i.e. 16.4 inches of lead plates and the 2 inches of lead in the support structure) provides only 64% live time in this arrangement. When 17.8 inches of lead plates were used, the detector housing is nearly the same distance from the source, 27 inches, as it would be if no lead were used in the first internal target geometry arrangement.

Figure 18 shows the gamma ray background at the sample location of the second internal target geometry arrangement collected for 10^4 seconds of live time. Six peaks are visible, and all are attributed to the structural iron and aluminum in the SIF. The flux near the sample location of this arrangement is greater than that at the sample position of the external target geometry arrangement. This accounts for the

Table 11. Effect of lead plate thickness on detector live time in the second internal geometry arrangement

Lead plate thickness (inches)	Live time	
	Source in storage position	Source in beam port position
0	45%	11%
3.6	70%	20%
6.9	80%	29%
9.6	84%	37%
11.4	86%	42%
13.1	87%	46%
14.8	92%	53%
16.4	99%	64%
17.8	100%	70%

Figure 18. Neutron capture background spectrum (0 - 9 MeV) collected using the second internal target geometry arrangement

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appearance of less sensitive peaks in the background spectrum of the second internal arrangement. Despite this increase in flux the overall intensity of the background spectrum collected using the internal target geometry arrangement is about one-sixth that observed when the external arrangement is used. The more extensive shielding of the detector is responsible for this improvement. Figure 19 is the background spectrum below about 1 MeV collected for 3600 seconds of live time. Again the only peak that can be attributed to materials in the SIF combines the 0.476 MeV boron peak and the 0.511 MeV annihilation peak. The capture gamma ray spectrum of sulfur using the second internal target geometry arrangement is shown in Figure 20. No sulfur peaks are evident, but the intensity of the spectrum is somewhat greater than that of the background.

The thermal neutron flux at the sample location of the internal target geometry arrangement is sufficient to analyze for sulfur in coal using prompt neutron gamma ray analysis. The high gamma ray background of the source, however, makes it impossible to reduce the sample to detector distance enough to carry out such analysis using the present set up.

F. Sample Tube

The sample tube in the SIF was designed to irradiate samples for delayed gamma activation analysis, and its flux characteristics are listed in Table 12. When the zirconium hydride is "down" the neutrons must pass through it and 4.5 inches of paraffin to reach the sample. When it is raised all of the way "up", or far enough so that the source is in line

Figure 19. Neutron capture background spectrum (0 - 1 MeV) collected using the second internal target geometry arrangement

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Figure 20. Neutron capture spectrum of sulfur (0 - 9 MeV) collected using the second internal target geometry arrangement

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Table 12. Neutron flux distribution in the sample tube

Height ^a (inches)	Moderator position	Total flux ^b	Thermal flux ^b	Cadmium ratio
4.5	up ^c	6.7×10^6	6.2×10^6	14.3
4.5	port ^d	7.3×10^6	6.6×10^6	9.48
4.5	down ^e	7.1×10^5	1.7×10^5	1.31
6.5	up	5.9×10^6	5.5×10^6	13.7
6.5	down	6.0×10^5	1.1×10^5	1.23

^aDistance from the bottom of the sample tube in inches.

^bNeutrons/cm²-sec.

^cHydride cylinder raised as far as possible.

^dHydride cylinder raised 1.44 inches above the bottom of the source rod assembly.

^eHydride cylinder resting on the bottom of the source rod assembly.

with the fast neutron port, the neutrons pass through the paraffin only. The extra thickness of moderating material present in the "down" configuration reduces the neutron flux and the cadmium ratio by capturing thermal neutrons faster than they can be replaced.

The neutron flux distribution in the sample tube with the hydride cylinder in both the "up" and "down" configurations is shown in Figure 21. The flux is greatest 4.5 inches from the bottom of the sample tube, but even there it is six or seven orders of magnitude below that available in an average research reactor.

Figure 21. Total neutron flux distribution in the sample tube

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VI. CONCLUSION

The californium-252 system for prompt neutron gamma ray analysis described in this work has fallen short of expectations. The thermal neutron flux at the sample location of the external target geometry arrangement is simply too low to be useful, while the gamma ray background remains high. Little improvement can be expected for such an arrangement. A larger source would simply increase the necessary biological shielding and the distance between the source and sample. A smaller source would decrease this distance but its lower neutron output would yield a flux that is little improved over that now available. Improved shielding might reduce the gamma background but it is doubtful that a useful flux could be obtained using an external target.

The internal target geometry arrangement has a much greater potential for success. The thermal neutron flux at the sample location is usable, but the gamma ray background requires that the detector be positioned so far from the sample that it is impossible to collect usable spectra. The background might be reduced by increasing the thickness of the lead surrounding the detector. Replacing the steel and aluminum in the SIF with structural plastics would eliminate the peaks in the background and lower its overall intensity by eliminating the gamma rays that result from fast neutron interactions with those materials. The lower flux yielded by a smaller source might even be tolerated in order to reduce the background and the required sample to detector distance.

The major drawback to the present system is inflexibility. Safety requirements led to the use of rigid shielding materials. A system using

a plastic shell and water as a moderator and shielding would offer much greater flexibility. The sample location and detector could be moved quite easily and a far greater number of geometric variations could be examined.

It is apparent that no advantages are gained by using such a large source. Anything gained from the increased neutron flux is more than outweighed by the disadvantages caused by the increased background.

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