

INSTRUMENTAL NEUTRON ACTIVATION
ANALYSIS OF COAL AND COAL FLY ASH

by

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1.0 Introduction

The purpose of this work was to develop procedures for determining the elemental concentration of coal and coal fly ash using the technique of instrumental neutron activation analysis, INAA.

In recent years, the concentration of various elements in coal and coal fly ash have become of interest primarily because of the the increased use of coal as an energy source. As shown in Fig. 1.1 the annual coal consumption in the United States increased from around 434 million short tons in 1960 to 669 million short tons in 1980.¹ The approximate fly ash release to the environment in 1980, assuming 15% conversion to ash upon coal combustion and 99% precipitator efficiency,² is 1.0 million tons while the remaining 99.3 million tons of ash must be disposed of as solid waste. In terms of the state of Kansas for 1980, 8.7 million tons of coal was burned, resulting in approximately 13.1 thousand tons of fly ash being released to the atmosphere, while 1.3 million tons of ash needed to be disposed of as waste.

The first and foremost reason for wanting to determine the elemental concentration of the coal and coal fly ash is the environmental impact of releasing fly ash to the atmosphere, of storage or utilization of the bottoms ash and the impact of coal storage and transport before combustion. Considering the large tonnages involved in each of these phases even the trace elements become important.

Another reason for determining elemental concentrations of coal fly ash is the recent interest in reclamation of metals from fly ash. An accurate accounting of the metals present is required for any economic decision concerning a reclamation project.³ The elemental

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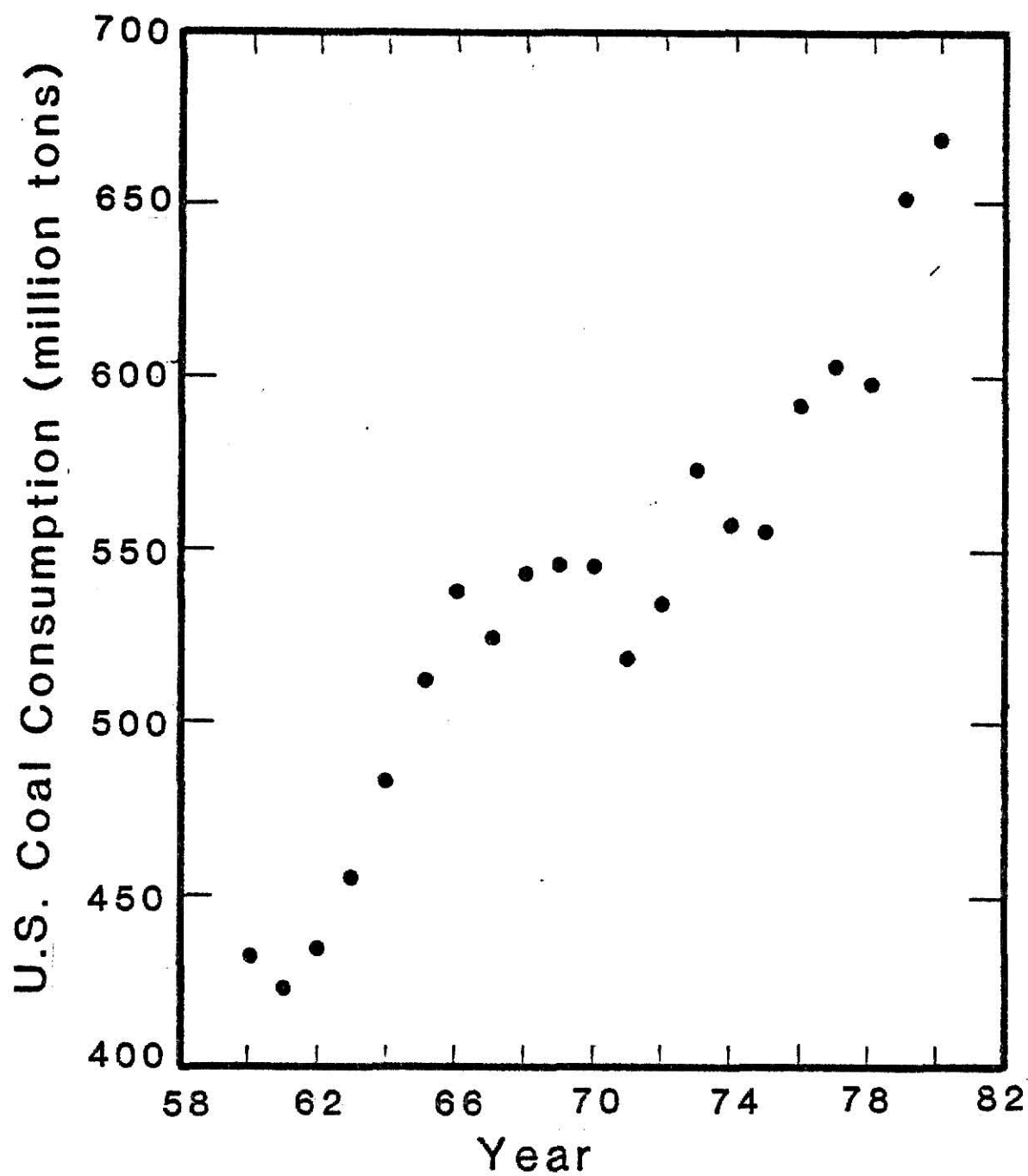


FIG. 1.1. United States Annual Coal Consumption

concentrations may also be important in the field of basic coal combustion research.

The reasons for using instrumental neutron activation analysis have often been stated in the literature.^{2,4} INAA requires no chemical separation or preparation of the sample. Thus, there is no chance of reagent contamination of the sample or loss of volatile elements during analysis. INAA can identify a large number of elements while remaining highly selective even when measuring low concentrations (ppm or lower). And finally, INAA is unaffected by matrix interference.

The main thrust of this work was to develop a detailed 'cookbook' procedure for determining minor and trace element concentrations of coal and coal fly ash. This procedure is intended to be used by a technician who has minimal experience in INAA. The second purpose of this work was to measure the elemental concentrations of 5 coals presently used in Kansas to generate electricity and to analyze 2 fly ash samples.

The five types of coals analyzed came from three power plants in Kansas. The first coal was a low sulfur Wyoming coal which is used at the Jeffrey Energy Center and is labeled WY-1. The next two coals are used at a plant near Lawrence, Kansas. These are a Colorado coal, CO and another Wyoming coal, WY-2. The LaCyne Power Plant uses two types of coal. One is a high sulfur Missouri/Kansas coal, MK and the other is from a mine in Illinois, IL6. Another coal, used as a standard, was purchased from the National Bureau of Standards, Standard Reference Material 1632a, Bituminous Coal.

The first ash sample was another NBS reference material -- SRM 1633a Coal Fly Ash. The second ash sample was sent to Kansas State University by A. Anderson, a graduate student at Purdue University.

2.0 Theoretical Development and Calculation Description

Instrumental Neutron Activation Analysis, INAA, is a well established technique for determining the mass of an element in an unknown sample. Basically this involves exposing an unknown sample to neutrons, some of which are absorbed by various isotopes of the sample leading to the transmutation of the isotope to a radioactive species. By measuring the emission rate of specific energy gamma rays and having some knowledge of activation reactions involved, the parent element can be identified and quantified.

What follows is the derivation of the model used to describe the method of measuring the concentration of a given element in an unknown sample. All equations below apply to the i^{th} element of the sample and the element must activate to a daughter product that emits gamma rays, only one of which is of interest.

The number of activated daughter products at some time, t , during an irradiation can be found by solving the rate equation which describes the change in the number of those atoms. If N represents the number of daughter product atoms at time t then

$$\text{change in } N = \text{production of } N - \text{loss of } N. \quad (1)$$

The production of N is the number of parent atoms present at time t , $n(t)$, multiplied by the probability of activating the parent atom upon interaction with a neutron, multiplied by the number of neutrons striking the target, ϕ . The assumption has been made that the flux varies so slowly with time that it is essentially a constant. Also, any energy or scattering angle dependence in the cross section is applied as required by the neutron flux, i.e., $\sigma(\Omega, E)\phi(\Omega, E) = \text{constant}$. Self shielding

effects are neglected. Mathematically, the production of N is expressed as the product $\eta(t)\sigma_p\phi$, where σ_p is the absorption cross section of the parent isotope. The number of parent atoms present at time t, is the initial number, N_0 , minus the numbers that are activated, which implies that the parent is a stable isotope so,

$$\eta(t) = N_0 - \int_0^t N(t')dt' .$$

Thus

$$\text{production of N} = \sigma_p\phi\left(N_0 - \int_0^t N(t')dt'\right) \quad (2)$$

The loss of N is the number that decay plus the number that are converted to another isotope. The decay of N is the number of daughter product atoms multiplied by the probability for decay λ .

$$\text{decay of N} = \lambda N \quad (3)$$

The number of N lost by activation is

$$\text{activation loss} = N\sigma_d\phi , \quad (4)$$

where σ_d is the cross section of the daughter and thus

$$\text{Loss of N} = \lambda N + N\sigma_d\phi . \quad (5)$$

Combining Eqs. (5), (2) and (1) yields,

$$\frac{dN}{dt} = \sigma_p\phi\left(N_0 - \int_0^t N(t')dt'\right) - \lambda N - N\sigma_d\phi . \quad (6)$$

Taking the LaPlace transform of Eq. (6) with the initial condition $N(0)=0$, yields

$$s\bar{N}(s) + \lambda\bar{N}(s) + \sigma_d\phi\bar{N}(s) - \frac{\sigma_p\phi N(s)}{s} = \frac{N_o\sigma_p\phi}{s} .$$

Solving for $\bar{N}(s)$ gives

$$\bar{N}(s) = \frac{N_o\sigma_p\phi}{s^2 + (\lambda + \sigma_d\phi)s + \sigma_p\phi} . \quad (7)$$

So that the roots of the denominator are found using the quadratic equation

$$s = \frac{-(\lambda + \sigma_d\phi) \pm \left[(\lambda + \sigma_d\phi)^2 - 4\sigma_p\phi \right]^{\frac{1}{2}}}{2} .$$

If $(\lambda + \sigma_d\phi)^2 - 4\sigma_p\phi$ is positive, the roots are real and equal to

$$R_1 = \frac{-(\lambda + \sigma_d\phi) - \left[(\lambda + \sigma_d\phi)^2 - 4\sigma_p\phi \right]^{\frac{1}{2}}}{2} \quad (8)$$

$$R_2 = \frac{-(\lambda + \sigma_d\phi) + \left[(\lambda + \sigma_d\phi)^2 - 4\sigma_p\phi \right]^{\frac{1}{2}}}{2} . \quad (9)$$

Substitution of Eqs. (8) and (9) into Eq. (7) yields

$$N(s) = \frac{N_o\sigma_p\phi}{(s-R_1)(s-R_2)} .$$

Taking the inverse LaPlace transform of the expression gives the time dependent solution

$$N(t) = \frac{N_o\sigma_p\phi}{(R_1-R_2)} \{ e^{R_1 t} - e^{R_2 t} \}$$

or

$$N(t) = \frac{-N_o \sigma_p \phi}{[(\lambda + \sigma_d \phi)^2 - 4\sigma_p \phi]^{\frac{1}{2}}} \exp\left\{\frac{-(\lambda + \sigma_d \phi) - [(\lambda + \sigma_d \phi)^2 - 4\sigma_p \phi]^{\frac{1}{2}}}{2}\right\} t - \exp\left\{\frac{-(\lambda + \sigma_d \phi) + [(\lambda + \sigma_d \phi)^2 - 4\sigma_p \phi]^{\frac{1}{2}}}{2}\right\} t \quad (10)$$

If we assume there were no significant losses due to the activation of the daughter, i.e., $\sigma_d \phi \ll \lambda$, then Eq. (10) becomes,

$$N(t) = \frac{-N_o \sigma_p \phi}{[\lambda^2 - 4\sigma_p \phi]^{\frac{1}{2}}} \exp\left\{\frac{-\lambda - [\lambda^2 - 4\sigma_p \phi]^{\frac{1}{2}}}{2}\right\} t - \exp\left\{\frac{-\lambda + [\lambda^2 - 4\sigma_p \phi]^{\frac{1}{2}}}{2}\right\} t.$$

Further, by neglecting depletion of the parent isotope during activation, i.e., $4\sigma_p \phi \ll \lambda^2$, then

$$N(t) = \frac{N_o \sigma_p \phi}{\lambda} \{1 - e^{-\lambda t}\} \quad (11)$$

At this point, it is desirable to express the number of parent atoms in terms of the mass of the parent element present initially, i.e.,

$$N_o = \frac{M A_b N_a}{A} \quad (12)$$

where M is the mass of the parent element, A_b is the natural abundance of the parent isotope, N_a is Avagadro's number and A is the isotope atomic weight. Substituting Eq. (12) into Eq. (11) yields,

$$N(t) = \frac{M A_b N_a \sigma_p \phi}{A \lambda} \{1 - e^{-\lambda t}\}.$$

The number of daughter atoms at the end of the irradiation time, t_r is

$$N(t_r) = \frac{M \text{ Ab } N_a \sigma_p \phi}{A\lambda} \{1 - e^{-\lambda t_r}\} . \quad (13)$$

After irradiation, the rate equation, Eq. (1), simplifies to

$$\text{change in } N = -\text{loss of } N$$

where the only loss is through radioactive decay so

$$\frac{dN}{dt} = -\lambda N$$

for $t > t_r$. Solving this equation by direct integration yields

$$N = C e^{-\lambda t} . \quad (13a)$$

To determine the integration constant C , we apply the initial condition

$N_R = N(t_r)$ as given by Eq. (13).

$$C = N_R e^{\lambda t_r} .$$

Then substituting into Eq. 13a to get the number of daughter atoms at time t after insertion yields,

$$N(t) = N_R e^{\lambda(t_r - t)} . \quad (14)$$

The sample is then placed adjacent to a gamma ray detector. At time t_s the detector system begins to count the number of gamma-ray detector interactions. The counts are stored as a function of energy. At time, t_e the detector system stops collecting. The number of atoms that decay during that interval is defined as N_D and is given by

$$N_D = N(t_s) - N(t_e) . \quad (15)$$

Substituting Eq. (14) into Eq. (15) yields

$$N_D = N_R \left[e^{\lambda(t_r - t_s)} - e^{\lambda(t_r - t_e)} \right]. \quad (16)$$

The number of gamma rays emitted by the decayed atoms during the counting interval is the number of atoms that decay multiplied by the gamma-ray yield (gamma rays of each energy per disintegration), Y . The number of counts recorded by the detector system is the number of gammas emitted multiplied by the absolute efficiency, E_f , of the detector. The total number of counts recorded during the time interval is defined to be the peak area P_a and thus

$$P_a = N_D E_f Y. \quad (17)$$

Substitution of Eq. (16) into Eq. (17) yields

$$P_a = N_D E_f Y \left[e^{\lambda(t_r - t_s)} - e^{\lambda(t_r - t_e)} \right].$$

Substituting Eq. (12) for N_R

$$P_a = \frac{M A_b N_a \sigma_p \phi E_f Y}{A \lambda} \left[1 - e^{-\lambda t_r} \right] \left[e^{\lambda(t_r - t_s)} - e^{\lambda(t_r - t_e)} \right].$$

Finally solving for the mass of the parent element gives

$$M = \frac{P_a A \lambda}{A_b N_a \sigma_p \phi E_f Y \left[1 - e^{-\lambda t_r} \right] \left[e^{\lambda(t_r - t_s)} - e^{\lambda(t_r - t_e)} \right]}. \quad (18)$$

The peak area for each energy gamma-ray is measured with the detector system and the time values t_r , t_s and t_e are recorded for each pulse height distribution measured. The natural abundance and atomic

number of the parent isotope are values easily found in the literature. The literature also provides values of the daughter product gamma-ray yield and decay constant. The detector efficiency is strongly dependent on the energy of the incident gamma rays. If needed, such an efficiency can be measured. However, the values of the neutron flux ϕ , and the cross section σ_p , are difficult to determine. To evaluate M using Eq. (18), the neutron flux level and cross sections must be given or measured.

To circumvent the problem of knowing the values of ϕ , σ_p , and E_f , we now introduce the use of a standard sample and fluence monitors. The standard sample is to be encapsulated separately from the unknown sample and the mass of each parent element in the standard must be a known quantity. The fluence monitors are metal wires of known mass wrapped around the sample containers and they are used to measure the value of $\int \phi dt$.

Equation (18) is applied to both the unknown and the standard samples. In the resulting two equations, a presubscript of u is used for the unknown sample equation and s for the standard sample equation. These equations are

$$M_u = \frac{P_u A_u \lambda}{A_b N_a \sigma_p \phi E_f Y \left(1 - e^{-\lambda t_r} \right) \left(e^{\lambda(t_r - t_s)} - e^{\lambda(t_r - t_e)} \right)} \quad (19)$$

$$M_s = \frac{P_s A_s \lambda}{A_b N_a \sigma_p \phi E_f Y \left(1 - e^{-\lambda t_r} \right) \left(e^{\lambda(t_r - t_s)} - e^{\lambda(t_r - t_e)} \right)} \quad (20)$$

but $A_b = A_u$; $\sigma_p = \sigma_p$; $N_a = N_a$.

The samples should be homogeneous, contained in the same counting geometry and of the same gross activity to allow equality between the system dead times so

$$s^E_f = u^E_f \quad . \quad (21)$$

Dividing Eq. (19) by Eq. (20) gives

$$\frac{u^M}{s^M} = \frac{u^P_a s^\phi \left(1 - e^{-\lambda s^t_r} \right) \left(e^{\lambda(s^t_r - s^t_s)} - e^{\lambda(s^t_r - s^t_e)} \right)}{s^P_a u^\phi \left(1 - e^{-\lambda u^t_r} \right) \left(e^{\lambda(u^t_r - u^t_s)} - e^{\lambda(u^t_r - u^t_e)} \right)} \quad (22)$$

The ratio s^ϕ/u^ϕ is found from information supplied by the fluence monitor wires. Eq. (18) is applied to both the unknown and standard sample wires and the resulting two equations are ratioed, using the superscript w to designate wire values. Solving for the flux ratio s^w_ϕ/u^w_ϕ ;

$$\frac{s^w_\phi}{u^w_\phi} = \frac{u^M_s w^P_A \left(1 - e^{-\lambda w^t_r} \right) \left(e^{\lambda(w^t_r - w^t_s)} - e^{\lambda(w^t_r - w^t_e)} \right)}{s^M_s w^P_A \left(1 - e^{-\lambda w^t_r} \right) \left(e^{\lambda(w^t_r - w^t_s)} - e^{\lambda(w^t_r - w^t_e)} \right)} \quad . \quad (23)$$

Assuming $s^w_\phi/u^w_\phi = s^\phi/u^\phi$, Eq. (23) can be directly substituted into Eq. (22).

Ultimately we wish to have a concentration number for each element in $\mu\text{g/g}$ or ppm. If we know the mass of the i^{th} isotope in the standard, s^M_i , we can multiply it by the mass ratio given by Eq. (22) to find the mass of the i^{th} isotope in the unknown.

$$\left(\frac{u^M}{s^M} \right)_i = \left(\frac{u^M}{s^M} \right)_i \left(\frac{s^M}{s^M} \right)_i \quad (24)$$

Then to convert to concentration units, let M_a be the measured mass of the unknown. The concentration of the i^{th} element would be

$$\text{ppm} = \frac{(M)_i}{M_a} \times 10^6 \quad (25)$$

Let M_u/M_s from Eq. (22) be the mass ratio for the i^{th} isotope, MR_i , then substitute Eq. (24) into Eq. (25), to find the concentration of the i^{th} isotope

$$\text{ppm} = \frac{MR_i \cdot (M)_i}{M_a} \quad (26)$$

In Eq. (26), the parameters sample masses or decay times can be measured directly. Other parameters such as the decay constants and elemental masses of standards can be found in the literature, but the peak areas are calculated values arising from measured detector-gamma-ray interactions. The calculation of the peak area is as follows.

Each gamma ray interaction with the detector results in a count being recorded for specific gamma-ray energy ranges called channels. As more of these counts are measured, a peak develops above some background level in the pulse height distribution of counts versus channel number as shown in Fig. 2.1. The area of the peak above the background is found by summing the counts between some channel to the left of the peak, A and another channel to the right of the peak B. Points A and B are k channels from either side of the peak. This summation is the integral counts as given by

$$\text{Integral} = \sum_{j=A}^B X_j ,$$

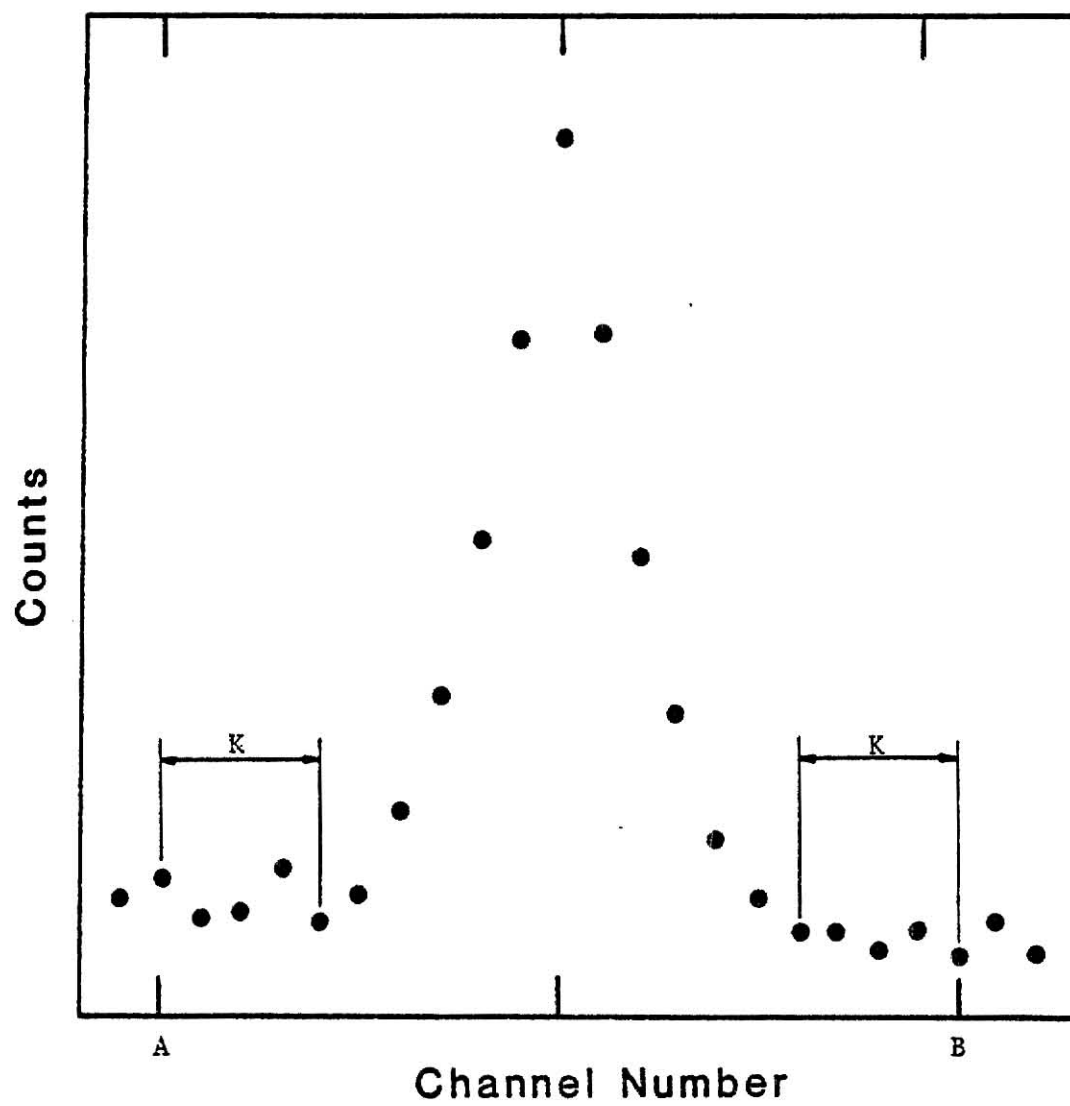


FIG. 2.1. Defination of Spectrum Background Parameters for Peak Area Calculation

where X_j is the number of counts in the j^{th} channel. The area is the area of the background trapezoid subtracted from the integral. The background area is given by

$$\text{Background} = \frac{(B-A+a)}{2k} \left(\sum_{j=A}^{A+k-1} X_j + \sum_{i=B-k+1}^B X_i \right)$$

and combining these equations gives the peak area

$$P_a = \sum_{i=A}^B X_i - \frac{(B-A+1)}{2k} \left(\sum_{j=A}^{A+k-1} X_j + \sum_{m=B-k+1}^B X_m \right). \quad (27)$$

The uncertainty of the concentration calculated for the i^{th} isotope given by Eq. (26) depends on the uncertainty of each of the measured values used in the calculation. The uncertainties of the measured values can be determined quantitatively and combined to estimate the uncertainty of the calculated value. The uncertainty of the calculated concentration is described by the standard deviation of the population of all measured values of that concentration. This deviation, σ_{ppm} , is related to the measured parameter uncertainties, σ_{x_j} by the general expression¹⁴

$$\text{VAR}(\text{ppm}) \cong \text{VAR}(X_1) \left(\frac{\partial f}{\partial x_1} \right)^2 + \text{VAR}(X_2) \left(\frac{\partial f}{\partial x_2} \right)^2 + 2\text{COV}(X_1, X_2) \left(\frac{\partial f}{\partial x_1} \right) \left(\frac{\partial f}{\partial x_2} \right) + \dots,$$

where f is the functional relationship used to calculate the contribution given the n measured parameters x_j , $j = 1, 2, \dots, n$, VAR is the variance of the specified parameter and COV is the covariance of x_1 and x_2 . As long as the parameters are independent of one another then the covariance term $\text{COV}(x_1, x_2) \left(\frac{df}{dx_1} \right) \left(\frac{df}{dx_2} \right)$ disappears, leaving the expression

$$\text{VAR}(\text{ppm}) \cong \sum_{j=1}^n \text{VAR}(x_j) \left(\frac{\partial f}{\partial x_j} \right)^2. \quad (28)$$

This equation is known as the error propagation formula¹⁵.

To calculate VAR(ppm), apply Eq. (28) to Eq. (26) treating the mass ratio as an independent variable. The result is

$$\text{VAR}(\text{ppm}) \approx \text{VAR}(\text{MR}) \left(\frac{s^M}{u^M_a} \right)^2 + \text{VAR}(s^M) \left(\frac{\text{MR}}{u^M_a} \right)^2 + \text{VAR}(u^M_a) \left(\frac{-\text{MR}(s^M)}{(u^M_a)^2} \right)^2 \quad (29)$$

and estimates of VAR(MR), VAR(s^M) and VAR(u^M_a) are needed.

The measured mass of the unknown, u^M_a will belong to a population of measured masses described by some mean value and some variance VAR(u^M_a). This variance is estimated by measuring the weight of standard masses of the same approximate weight of the coal samples to be used. The results are assumed to fit a normal distribution and the variance is calculated. This value is used to estimate VAR(u^M_a).

The mass of the i^{th} isotope in the standard, s^M is found by multiplying the mass of the standard, smass, by the concentration of the i^{th} isotope, conc. Application of Eq. (28) to this multiplication yields

$$\text{VAR}(s^M) \approx \text{VAR}(\text{smass}) (\text{conc})^2 + \text{VAR}(\text{conc}) (\text{smass})^2, \quad (30)$$

where VAR(conc) is known from the reference literature on the standard and VAR(smass) is assumed to VAR(u^M_a) since smass and u^M_a are from the same population.

To find the variance of the mass ratio VAR(MR), Eq. (22) is rewritten as

$$\text{MR} = \frac{u^P_a}{s^P_a} \left(\text{FR} \right) \left(\text{TR} \right), \quad (31)$$

where FR is the flux ratio given by Eq. (23) and TR is the time ratio given by

$$TR = \frac{\left[\frac{-\lambda_{sr}}{1-e} \right] \left[\frac{\lambda_{sr} t_s - t_s}{e} \quad \frac{\lambda_{sr} t_r - t_r}{-e} \right]}{\left[\frac{-\lambda_{ur}}{1-e} \right] \left[\frac{\lambda_{ur} t_r - t_r}{e} \quad \frac{\lambda_{ur} t_s - t_s}{-e} \right]} \quad (32)$$

For reasons of simplification, any errors associated with the time ratio are neglected. Applying Eq. (28) to Eq. (31) yields the variance of the mass ratio,

$$VAR(MR) \approx VAR(FR) \left[\frac{(TR)(P_a)}{s_a} \right]^2 + VAR(P_a) \left[\frac{(TR)(FR)}{s_a} \right]^2 + \quad (33)$$

$$VAR(s_a) \left[\frac{-(P_a)(FR)(TR)}{(s_a)^2} \right]^2.$$

To find VAR(FR), Eq. (28) is applied to Eq. (23) to give

$$VAR(FR) \approx VAR(u_M) \left[\frac{(TR_w)(P_a)}{s_M(u_a)^2} \right]^2 + VAR(s_a) \left[\frac{(TR_w)(u_M)}{s_M(u_a)^2} \right]^2 +$$

$$VAR(s_M) \left[\frac{-(TR_w)(P_a)}{(u_M)^2(u_a)} \right]^2 + VAR(u_a) \left[\frac{-(TR_w)(u_M)(P_a)}{s_M(s_M(u_a)^2)} \right]^2, \quad (34)$$

where TR_w is the wire time ratio similar to Eq. (32). The variances $VAR(u_M)$ and $VAR(s_M)$ are assumed to equal $VAR(u_a)$.

The variances $VAR(s_a)$ and $VAR(u_a)$ from Eq. (34) and $VAR(s_a)$ and $VAR(u_a)$ of Eq. (33) are not equal and can be calculated from the photo-peak areas. To find the variance of the calculated peak area, $VAR(P_a)$, apply the error propagation formula to the peak area equation, Eq. (27).

Carrying out the differentiation and some algebra, the following equation results:

$$\begin{aligned} \text{VAR}(P_a) = & \left(1 - \frac{B-A+1}{2k}\right)^2 \text{VAR}(X_A) + \dots + \left(1 - \frac{B-A+1}{2k}\right)^2 \text{VAR}(X_C) + \\ & \text{VAR}(X_{C+1}) + \dots + \text{VAR}(X_{D-1}) + \left(1 - \frac{B-A+1}{2k}\right)^2 \text{VAR}(X_D) + \\ & \dots + \left(1 - \frac{B-A+1}{2k}\right)^2 \text{VAR}(X_B) \quad . \end{aligned} \quad (35)$$

It has been shown in the literature^{14,15} that the variance of the counts in a given channel is the counts, so Eq. (35) becomes

$$\begin{aligned} \text{VAR}(P_a) = & \left(1 - \frac{B-A+1}{2k}\right)^2 X_A + \dots + \left(1 - \frac{B-A+1}{2k}\right)^2 X_C + X_{C+1} + \dots + X_{D-1} \\ & + \left(1 - \frac{B-A+1}{2k}\right)^2 X_D + \dots + \left(1 - \frac{B-A+1}{2k}\right)^2 X_B \quad . \end{aligned} \quad (36)$$

Rearranging Eq. (36) and reintroducing the summation notation yields the following equation for the variance of the peak area

$$\text{VAR}(P_a) = \sum_{i=A}^B X_i + \left(\frac{B-A-4k+1}{2k}\right) \left(\frac{B-A+1}{2k}\right) \left[\sum_{j=A}^{A+k+1} X_j + \sum_{m=B-k+1}^B X_m \right] \quad . \quad (37)$$

Equations (37) and (34) are used to evaluate Eq. (33). Equation (33) is then used with Eq. (30) to evaluate Eq. (29) which gives the error associated with the measured concentrations.

3.0 Description of Materials and Equipment

3.1 Standard Sample

The standard sample must contain known quantities of each element of interest, the elemental concentrations in the standard must approximate those of the unknown and the physical characteristics of the two must be similar to equalize the effects of self shielding.

The literature^{2,4,5,6} reports the preparation of a standard by pipetting given quantities of desired material onto various filler materials. These standards are used to estimate the elemental concentrations of the unknown. A new standard is then prepared to better approximate those concentrations. An iteration process is continued until the samples are matched, thus preventing detection system dead time problems and ensuring that the absolute efficiencies of Eq. (21) are equal. Naturally, some range exists for the matching of concentrations. The major constituents provide the greatest contribution to the final activity, so trace element concentrations in the standard can be increased to improve counting statistics without significantly affecting the overall activity.

For this work however, it was decided not to spend the effort preparing standard samples from the reagents but to use the NBS standard reference material 1632a as the standard. This Pennsylvania bituminous coal⁷ is certified by NBS for 18 elements and NBS supplies information for another 12 elements. Preliminary qualitative analysis of the coal samples revealed that a number of elements could be identified using INAA but concentration information for these elements is not included in the NBS Certificate of Analysis. This problem was resolved by utilizing the results of an analysis done at the University of Maryland.⁵

The University of Maryland analysis utilized INAA, neutron capture prompt gamma activation analysis, PGAA and instrumental photon activation analysis, IPAA to measure the concentrations of 51 elements in the NBS coal 1632a. The NBS and Maryland concentration values are given for a selected list of 47 elements in Table 3.1. As pointed out by the authors of the article, only 4 elements, Ti, Co, Rb and Cs, have NBS concentration values outside the given intervals of uncertainty given by the University of Maryland.

Since agreement exists between the concentrations reported by the University of Maryland and the NBS certified values, the values reported by the University of Maryland shall be used as the standard concentrations in this work.

The elemental concentrations of the standard are assumed to approximate those of the unknowns to be studied and since both standard and unknown are coal samples, their physical characteristics are similar.

3.2 Sample Encapsulation

The coal and fly ash samples are encapsulated in polyethylene vials shown in Fig. 3.1. The vial sizes vary depending on the amount of material or size of vial to be contained. Two samples of each coal or fly ash are prepared. The first sample is prepared by filling the smallest vial, 2/27 dram (approximately 1 ml), with coal or coal fly ash. The largest vial, 2 dram, is used to encapsulate the first sample during irradiation. The second sample is the 2/5 dram vial (approximately 7 ml) filled with sample material. This vial is enclosed in a 2" x 3" polyethylene bag during irradiation.

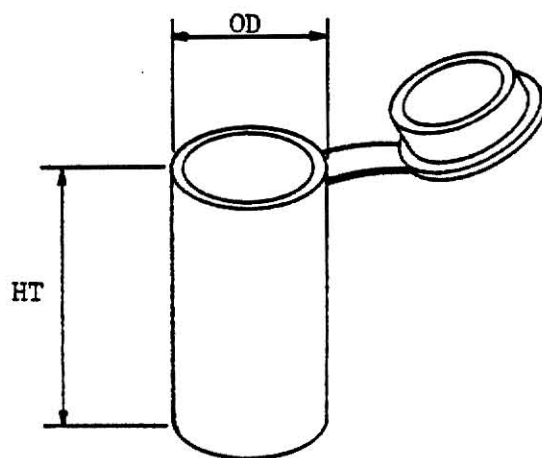
TABLE 3.1. Elemental Concentrations of NBS Standard Reference Material 1632a as Reported by the National Bureau of Standards and the University of Maryland.

element	University of Maryland	NBS
Na (%)	0.085 ± 0.004	0.084 ± 0.004
Mg (%)	0.13 ± 0.03	0.1
Al (T)	2.94 ± 0.13	3.1
Si (%)	5.8 ± 0.1	-
Cl (ppm)	790 ± 20	-
K (%)	0.42 ± 0.02	0.42 ± 0.02
Ca (%)	0.32 ± 0.02	0.23 ± 0.03
Si (ppm)	6.8 ± 0.6	6.3
Ti (%)	0.161 ± 0.004	0.18
V (ppm)	44 ± 3	44 ± 3
Cr (ppm)	34 ± 2	34.4 ± 1.5
Mn (ppm)	32 ± 3	28 ± 2
Fe (%)	1.16 ± 0.03	1.11 ± 0.02
Co (ppm)	6.5 ± 0.2	6.8
Ni (ppm)	26 ± 4	19.4 ± 1
Zn (ppm)	31 ± 6	28 ± 2
Ga (ppm)	8.0 ± 0.8	8.5
As (ppm)	10.2 ± 0.5	9.3 ± 1
Se (ppm)	2.6 ± 0.3	2.6 ± 0.7
Be (ppm)	41 ± 4	-
Rb (ppm)	24 ± 1	31
Sr (ppm)	84 ± 9	-
Y (ppm)	5.8 ± 0.5	-
Zr (ppm)	53 ± 5	-
Cd (ppm)	0.21 ± 0.03	0.17 ± 0.02
In (ppb)	36 ± 4	-
Sb (ppm)	0.60 ± 0.09	0.6
I (ppm)	1.8 ± 0.2	-
Cs (ppm)	2.0 ± 0.3	24
Ba (ppm)	122 ± 11	-
La (ppm)	18 ± 2	-
Ce (ppm)	32 ± 4	30
Nd (ppm)	12.1 ± 0.4	-
Eu (ppb)	550 ± 30	500
Gd (ppm)	1.95 ± 0.03	-
Tb (ppm)	0.36 ± 0.12	-
Dy (ppm)	2.2 ± 0.3	-
Yb (ppm)	0.98 ± 0.08	-
Lu (ppb)	180 ± 30	-
Hf (ppm)	1.55 ± 0.08	1.6

TABLE 3.1 (cont.)

element	University of Maryland	NBS
Ta (ppb)	400 \pm 30	-
W (ppm)	0.6 \pm 0.2	-
Pb (ppm)	15 \pm 4	12.4 \pm 0.6
Th (ppm)	4.8 \pm 0.2	4.5 \pm 0.1
U (ppm)	1.2 \pm 0.1	1.28 \pm 0.02
Cu (ppm)	-	16.5 \pm 1.0
Hg (ppm)	-	0.13 \pm 0.03

^aValues with no limits are non-certified NBS values



<u>Vial Use</u>	<u>OD</u>	<u>HT</u>	<u>dram</u>
short lived sample	7.11 mm	12.70 mm	2/5
long lived sample	11.94 mm	12.70 mm	2/27
irradiation container for short lived sample	16.76 mm	57.15 mm	2

FIG. 3.1 Polyethylene Vials Used for Sample Encapsulation

3.3 Irradiation Facility

The neutron irradiation of the samples utilizes the rotary specimen rack, RSR, of the TRIGA Mark II nuclear reactor. The TRIGA is a zirconium hydride-8.5 wt% uranium fueled reactor licensed to a power level of 250 kW thermal. The thermal flux, energy <0.21 keV, at 250 kW in the RSR is 1.8×10^{12} neutrons/(cm²·s) while the fast flux, energy >10 keV, is 1.5×10^{12} neutrons/(cm²·s).

The rotary specimen rack is a circular aluminum ring supporting 40 cylindrical containers which hold the samples during irradiation. The rack assembly is encased in a water tight, air filled aluminum housing, embedded in the graphite reflector surrounding the core, as shown in Fig. 3.2. The samples are loaded using a plastic loading vial attached to a nylon cord, then lowered through a tube, see Fig. 3.3, into an irradiation position.

3.4 Detector Analyzer System

This section will introduce the basic concept behind the detector analyzer system, then the system will be described in detail.

The activation daughter products emit characteristically energetic gamma rays and the detection system must be capable of measuring the number of gamma rays interacting with the detector as well as their incident energy. The detection system, shown in the block diagram of Fig. 3.4, basically consists of a solid state germanium lithium drifted detector coupled to a multichannel analyzer, MCA. Upon gamma ray interaction with the detector, voltage pulses with amplitudes proportional to the incident gamma ray energy are sent to the analog to digital converter (ADC), which is a component of the MCA system.

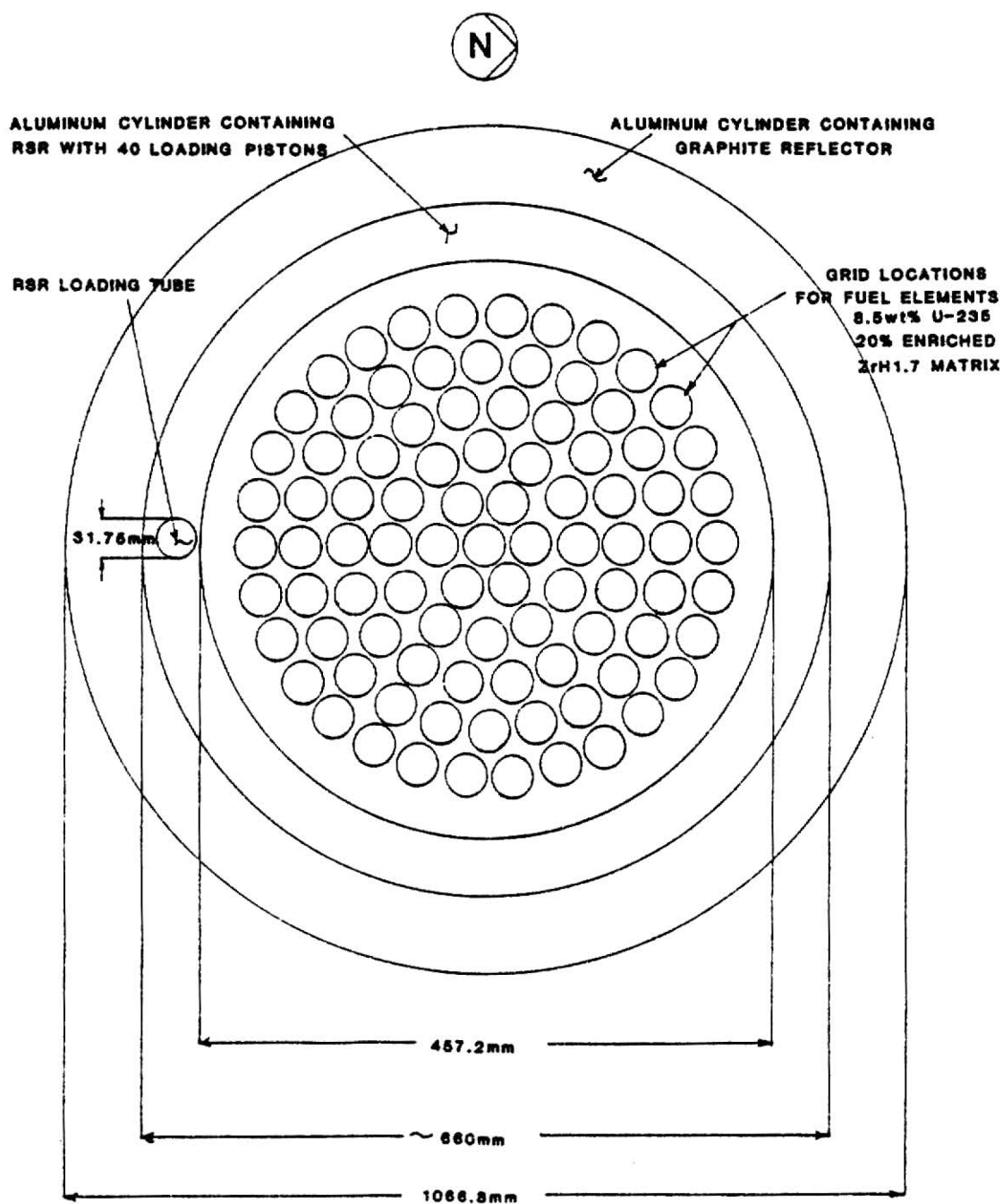


FIG. 3.2. Location of Rotary Specimen Rack, RSR, With Respect to the Reactor Core

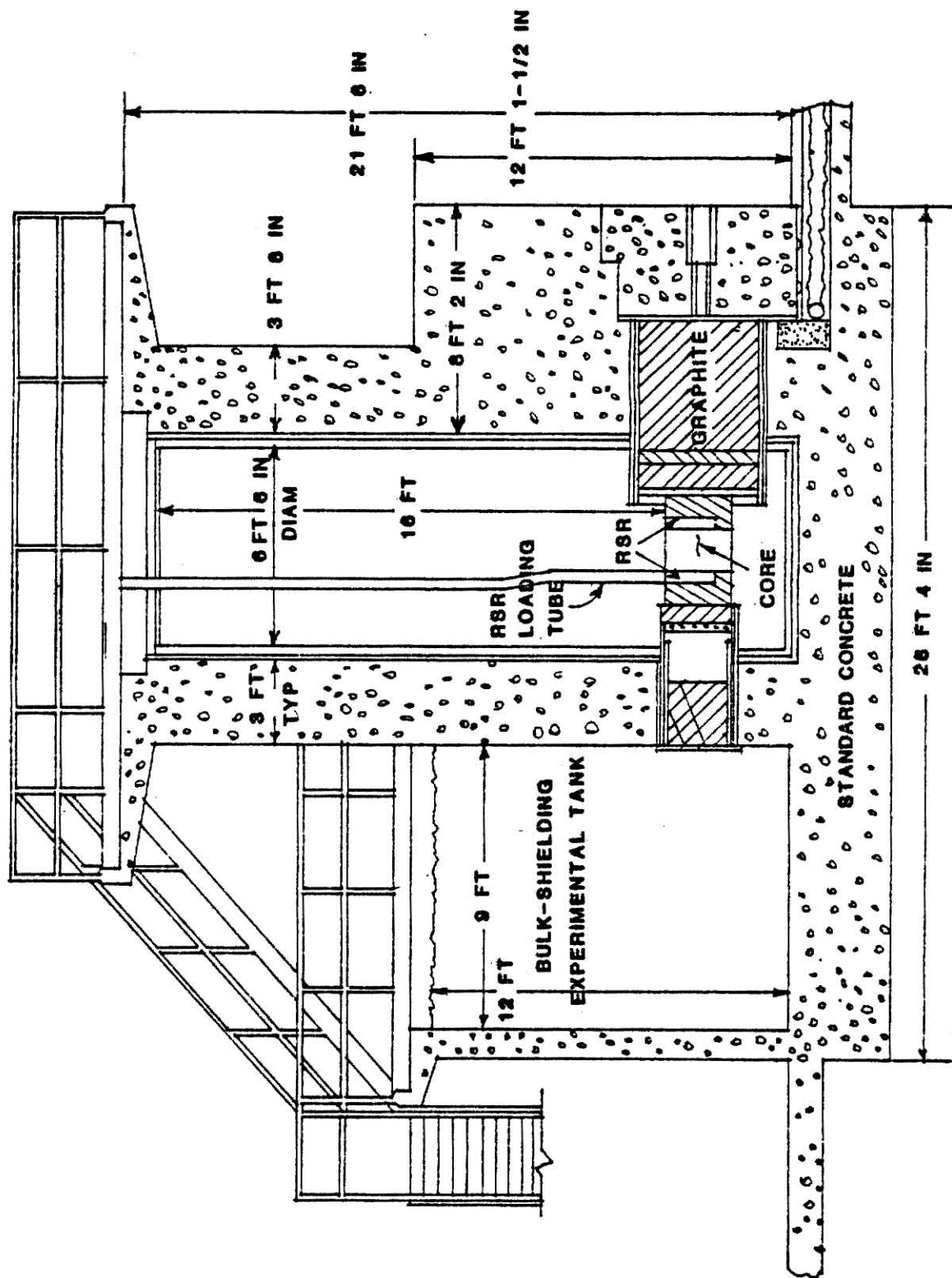


FIG. 3.3. Location of Rotary Specimen Rack Loading Tube in Reactor

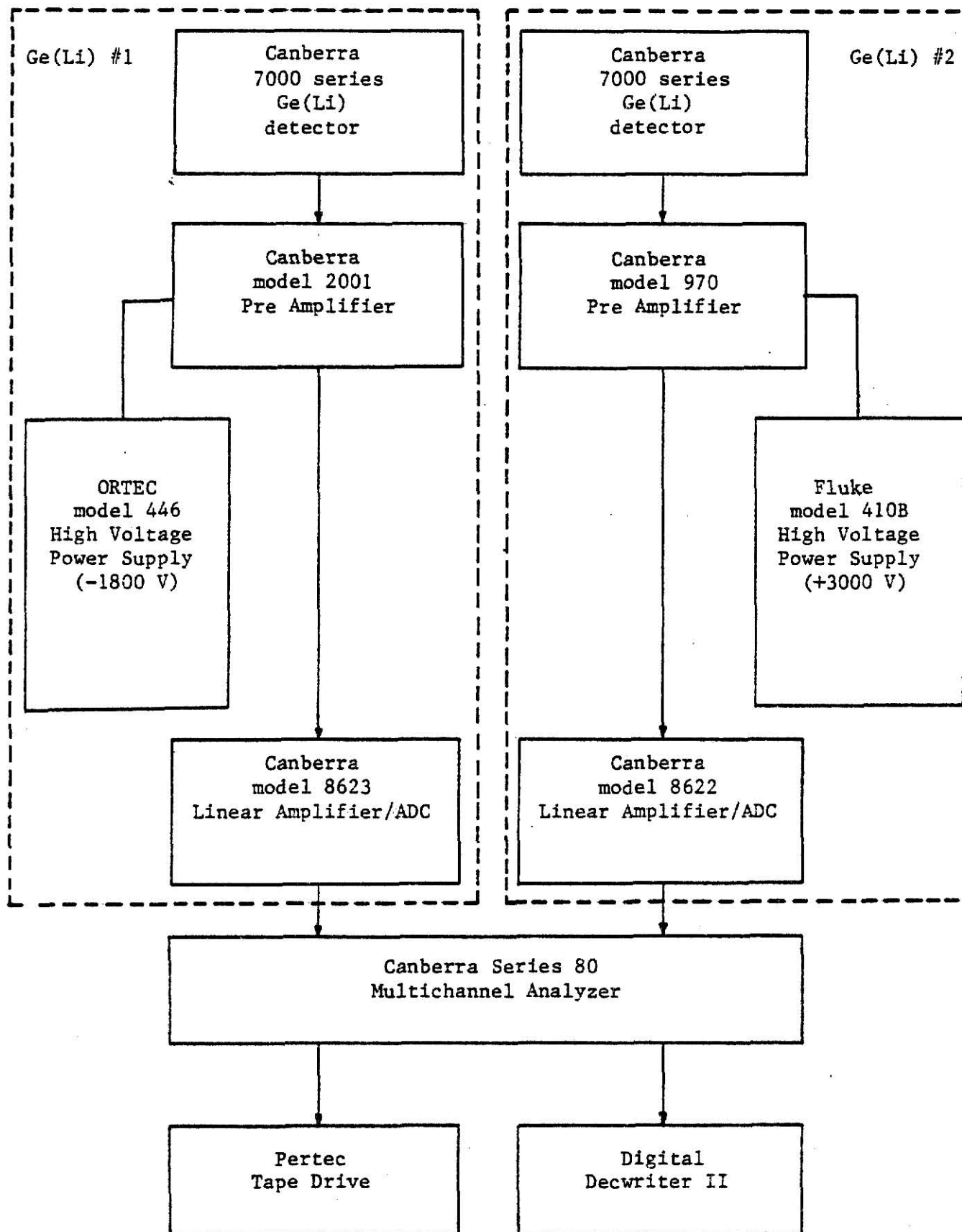


FIG. 3.4 Block Diagram of Gamma-Ray Detection System Showing Both Ge(Li) Detectors

The ADC then produces digital values that are proportional to the amplitudes of the incoming pulses. The digital information is stored in the MCA memory using a simple counting scheme. This scheme involves equally dividing the range of possible values into a group of subintervals called channels. Usually the range is divided into 4096 channels for this work. Whenever a digital signal of given value is sensed by the analyzer, a count is recorded in the corresponding channel. The number of counts versus channel number is defined as the pulse height distribution.

In order for the pulse height distribution data to be useful, the experimenter must know in which channel a count will be recorded if all the energy of a gamma-ray is deposited in the detector. In other words, he must have some way of knowing the detector, ADC and amplifier proportionality constants. The process of relating a channel number to a gamma-ray energy is defined as energy calibration.

The energy calibration is determined by placing a gamma-ray source of known energies near the detector, and observing in which channels counts are being recorded. Generally, the peaks in the pulse height distribution correspond to the energy of the incident gamma-ray. By fitting a curve through the gamma-ray energy versus peak channel data, an energy calibration equation is generated. This equation is later used to determine the energy of gamma rays emitted from the activated samples.

The analyzer, Series 80 has the capability of identifying the peak energies in a spectrum then comparing those values with a library of gamma-ray energies to identify the isotope present in the sample. The library is a list of isotopes, and the emission energies of the

gamma-rays with the corresponding percent gamma-ray yields. This library is input into the Series 80 following the procedure outlined in Appendix B.

The collection of a pulse height distribution for a coal sample begins with the placement of the sample near the detector. This placement must be constant for each sample and standard measurement because variation in this counting geometry will lead to differences in the absolute efficiencies and thus violation of the assumption of Eq. (21). Fig. 3.5 shows a Lucite rack that is placed atop the detector casing to hold samples at specified distances above the detector. The rack also has three 3.2 mm thick lucite plates that can be placed between the sample and detector to minimize the beta particle exposure. Fig. 3.6 shows the detector attached to a Dewar containing liquid nitrogen. The nitrogen is required to cool the detector to prevent thermal migration of lithium from the detector. Also shown is the location of the detector pre-amplifier which contains the high voltage connection for the detector as well as the signal output.

Figure 3.4 shows two Ge(Li) detectors and their associated preamplifiers, linear amplifier and high voltage power supply systems connected to the Canberra Series 80 Multichannel Analyzer. Ge(Li) no. 2 is used to collect sample spectra because of its better energy resolution when compared with Ge(Li) no. 1. Ge(Li) no. 1 is used to measure the fluence monitors. The Series 80 MCA is capable of storing data from both detectors simultaneously by partitioning the ADC memory properly. Users should refer to the Series 80 operators manual for details.

Adjustable settings for the detector analyzer system, such as memory partitions, amplifier gain settings, etc. can be recorded on the standard forms given in Appendix A.

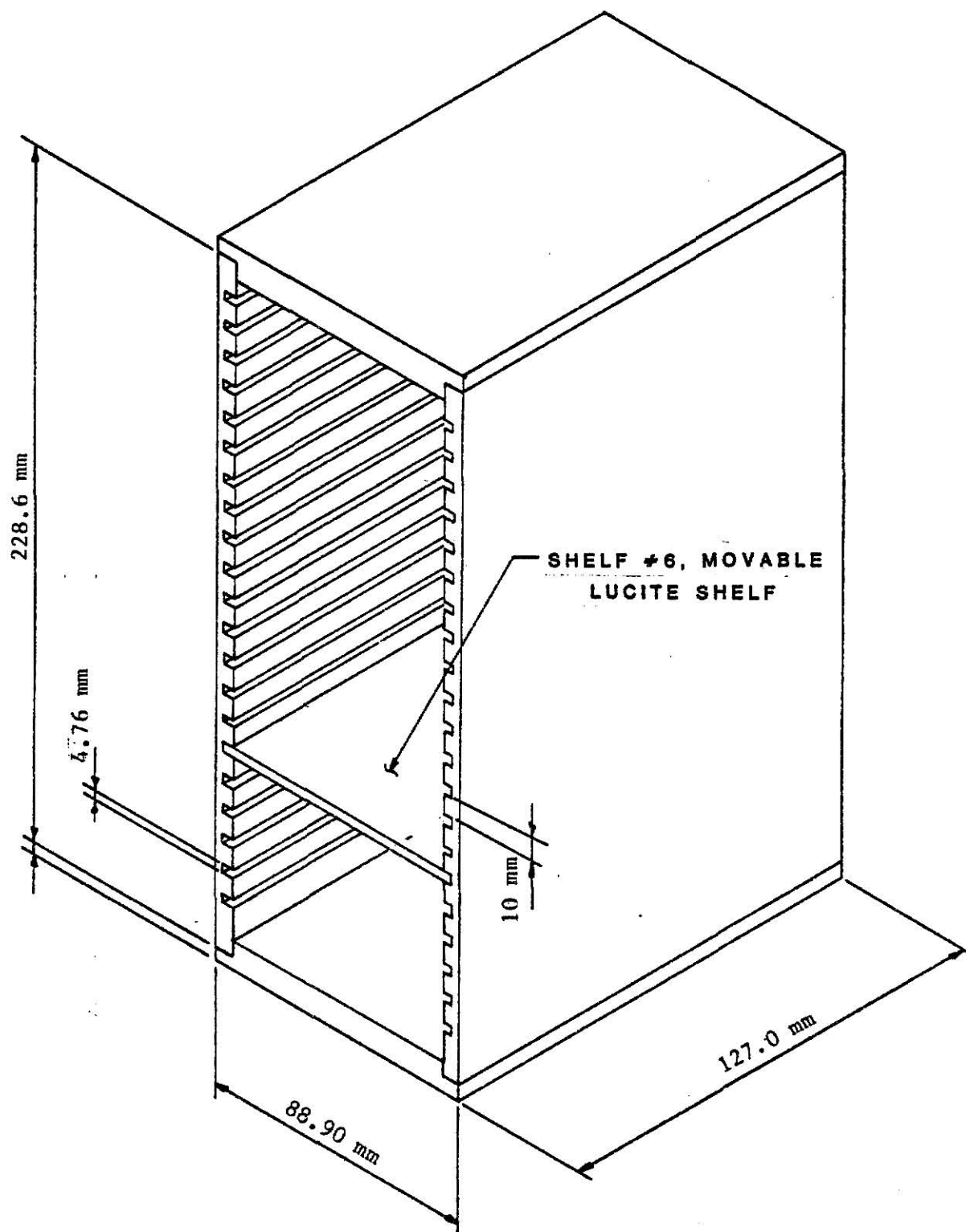


FIG. 3.5 Lucite Rack for Sample Positioning During Counting

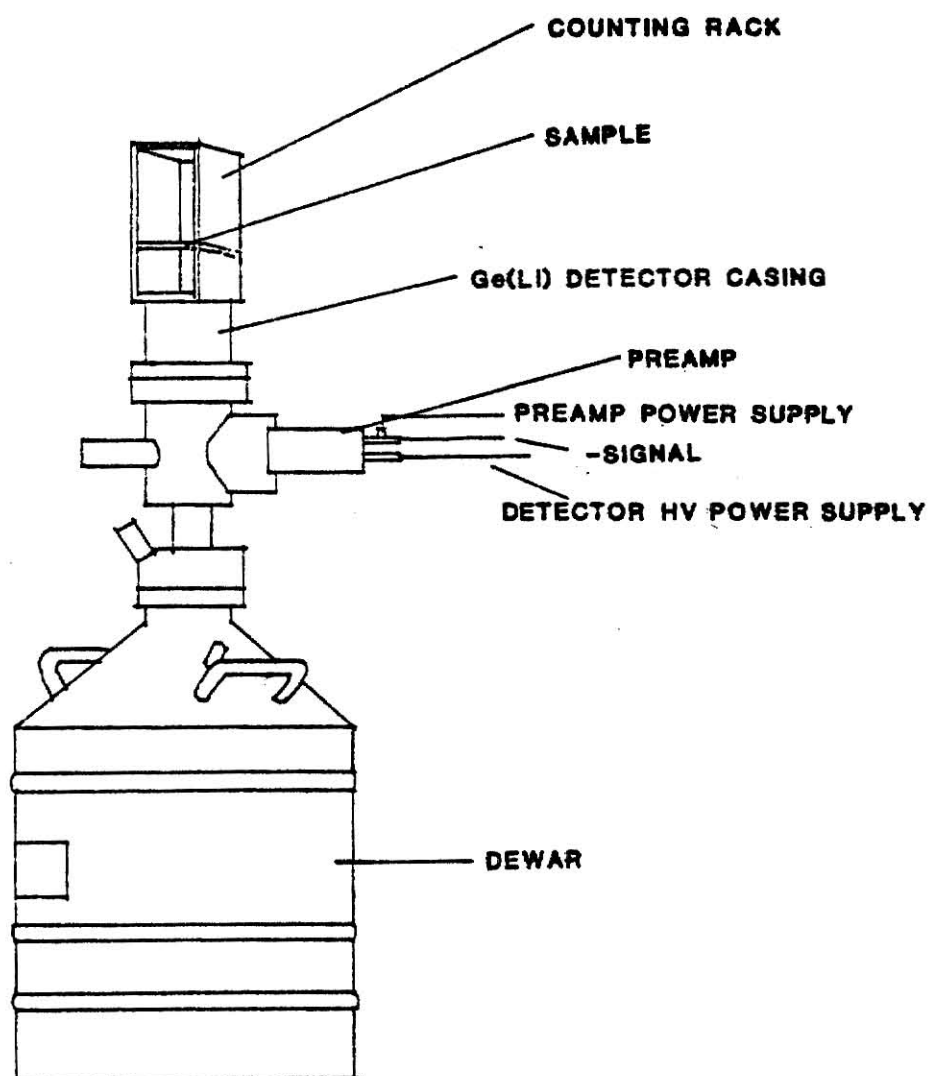


FIG. 3.6. Positions of Counting Rack, Sample, Detector and Associated Electronics

After each sample count, the energy spectrum is stored on magnetic tape using the Pertec tape drive and is later returned to the analyzer for analysis. The Digital Decwriter is used to provide hard copy output of such parameters as peak area and error.

4.0 Experimental Procedures

4.1 Preliminary Analysis

4.1.1 Encapsulation

A review of the literature reveals four methods of encapsulating the samples during irradiation and counting. The first involves simply placing the sample inside a polyethylene vial.⁸ The second places the sample inside a polyethylene bag which is then heat sealed.² The third places the coal or ash in filter paper which is then pelletized.⁴ The last method is to fold the sample in Al foil then pack into a polyethylene vial or cadmium box.⁶ The amount of material encapsulated by the above methods ranges from 50 mg to 500 mg.

The encapsulation method should contain the sample during irradiation and/or counting, provide minimal activation interference from packaging material, and provide a quick, simple way of encapsulating the sample.

The aluminum foil polyethylene vial method is eliminated from consideration because the aluminum activation presents an exposure problem during short reactor runs and the additional work to prepare the sample provide no advantage over simply placing the sample into a polyethylene vial. The Nuclear Engineering Department does not readily have the equipment necessary to pelletize samples so this method is eliminated. Rectangular polyethylene bags, 50.8 mm by 76.2 mm, were used to encapsulate some samples but the time required to heat seal the bags and the inadequacy of the seal forced this method to be eliminated. The method of placing the sample in a polyethylene vial best meets the encapsulation criteria.

4.1.2 Irradiation Times and Flux Levels

Selection of reactor flux levels and sample irradiation times were based primarily on the experience of the experimenter. Each type of coal or coal fly ash was irradiated in two separate reactor runs. The first was for an irradiation time of 5 min at a reactor thermal flux level of approximately 1.6×10^{12} n/cm²/s. This run was used to identify short lived, $T_{1/2} < 24$ h, activation products. The second irradiation was for 8 h at a thermal flux level of 1.6×10^{12} n/cm²/s. This was used to identify the longer lived isotopes, $T_{1/2} > 24$ hr. The flux level is the highest normally available in the KSU Triga Mk II reactor. Table 4.1 provides a list of the irradiation times and flux levels reported in the literature by various experimenters. The flux level available for this work was most closely approximated by those available to Black and Dams⁽⁴⁾, as are the irradiation times.

4.1.3 Counting Scheme

The counting scheme is a combination of decay and count intervals which have been selected to minimize the error in the quantification of isotopes. The quantification calculation is largely dependent on the experimenter's ability to measure the area of the full energy peaks in the spectrum. The major obstacle in measuring these areas comes when the activities of other isotopes in the sample are so high, relative to the isotope of interest, that the peak of interest is lost in the statistical noise of the more active isotope. Generally this increased activity is due to the relatively short half life of the products involved. The counting scheme approach allows the interfering isotopes to decay to a point where they themselves are lost in the statistical noise of the peak

TABLE 4.1 Irradiation Times and Flux Levels used by Various Experimenters to Identify Elements in Coal and Coal Fly Ash.

Experimenter	Irradiation Time	Flux Levels*
Germani, et. al.	5 min	$2-5 \times 10^{13}$
	4 h	$2-5 \times 10^{13}$
Abel and Rancitelli	0.5 - 1 min	1×10^{13}
	6 - 8 h	5×10^{12}
Block and Dams	5 min	2.6×10^{12}
	7 h	1.6×10^{12}
Obrusnik, et. al.	10 min	1×10^{13}
	6 h	1×10^{13}
Ondov, et. al.	30 s	1×10^{13}
	5 min	1×10^{13}
	4 h	1×10^{13}
Row and Steinnes	1 min	1.5×10^{13}
	4 h	1.5×10^{13}
Ruch, et. al.	15 min	2×10^{12}
	2 h	4.1×10^{12}

* (n/cm²/sec)

of interest. A counting scheme is determined for both the short and long reactor runs.

To determine the count scheme for the short reactor run, a sample was activated then counted at various times during the sample decay. The selection of when to begin a count and the length of the count interval was based on the count schemes reported by various experimenters as shown in Table 4.2. An NBS coal sample was prepared and activated for 5 minutes in the RSR at a reactor power of 225 kW. The sample was then allowed to decay for 10 minutes after which a 300 second count was begun with the sample on shelf 6 of the rack, shown in Fig. 3.5. Twenty minutes after the end of the irradiation another 300 second count was begun. After moving the sample to shelf 1 of the rack, a count of 1800 seconds was started at a decay time of 35 minutes and another at 81 minutes. The values of the decay times were selected by the experimenter as being possible break points where the highly active isotopes may have decayed significantly.

Isotopes are identified in the four spectra and the peak areas of the predominant photopeaks were calculated. The areas were then normalized to counts per unit isotope mass per unit count time. The maximum of the normalized count rates would yield the results of greatest accuracy. In Table 4.3 are listed the normalized count rates for each isotope identified in each spectrum.

Using the results presented in Table 4.3 as a guideline, the short run counting scheme was a 5 minute post irradiation decay followed by a 600 second count with the sample on shelf 10. After a 35 minute post irradiation decay an 1800 second count was started with the sample on shelf 1. Finally, an 8 hour count was started 4 hours after irradiation.

TABLE 4.2 Sample Counting Schemes Used by Various Experimenters

Experimenter	Irradiation No.	Count No.	Decay Interval	Count Time
Germani, et. al.	1	1	6 - 12 min	-
		2	15 - 20 min	-
	2	1	3 - 5 d	4 h
		2	20 - 30 d	8 - 24 h
Abel and Rancitelli	1	1	20 min	5 min
	2	1	4 - 7 d	10 min
		2	30 d	2 - 17 h
Block and Dams	1	1	3 min	6 min
		2	15 min	20 min
	2	1	1 d	15 min
		2	20 d	1 h
Obrisnik, et. al.	1	1	10 min	5 min
		2	20 min	10 min
		3	12 - 36 h	10 min
	2	1	24 - 36 h	-
		2	14 - 21 d	-
Rowe and Steinnes	1	1	20 min	10 min
		2	1 d	10 min
	2	1	6 - 7 d	1 h
			20 - 25 d	1 h
Ruch, et. al.	1	1	30 min	5 min
		2	3 h	33 - 50 min
	2	1	24 h	1.1 - 1.9 h
		2	30 d	6 - 10 h

TABLE 4.3. Normalized count rates for a short reactor run of NBS coal counted at specified times during decay after activation (counts/s/g)

Isotope (gamma-ray used to identify (keV))	Decay Time After Irradiation			
	10 m	20 m	35 m	81 m
^{24}Na (1368.6)	4.12	3.88	26.33 ^a	26.09
^{27}Mg (1014.4)	2.46 ^a	0.88	b	b
^{28}Al (1778.7)	152.6 ^a	4.90	b	b
^{38}Cl (1642.2)	2.09	1.96	8.25 ^a	2.69
^{51}Ti (319.7)	7.98 ^a	1.96	b	b
^{52}V (1434.1)	10.9 ^a	1.33	b	b
^{55}Mn (1810.7)	1.4	1.51	9.27 ^a	7.16
^{79}Br (616.2)	2.93	2.02	6.84 ^a	2.18

^a maximum normalized count rate

^b photopeak not observed

The determination of the count scheme for the long reactor run was carried out in the same manner except this time the peak area error was used as the discriminating factor instead of the normalized count rate. The activities of the various isotopes generally do not exhibit the large changes seen in the short run results because of the longer half lives of the isotopes remaining. But as time progresses, the degree of interference from spectrum structure tends to vary the counts in the background channels described in the paragraph preceeding Eq. (27). For this reason the peak area error values fluctuate. In Table 4.4 the peak area error values as a function of decay time for isotopes identified in the coal are listed.

It is desired to select the decay interval which yields the largest number of isotopes with the smallest possible error in the peak areas. To guide in the selection of this decay interval from the data of Table 4.4, the count times were ordered by increasing error for each isotope. Then a frequency distribution was generated for how many times a given decay time was ranked first in peak area error then second and finally third. This distribution is given in Table 4.5.

From Table 4.5, the 18.04.31 decay interval count yielded 11 isotopes which ranked the lowest or second lowest in peak area error. This decay interval was therefore selected for the long run count scheme. The only problem with the 18.04.31 decay interval is that three isotopes ^{175}Yb , ^{181}Hf , ^{140}La are not optimized. Thus, it is decided to include a decay interval of 6 days followed by a count to identify these isotopes.

When the data of Table 4.4 were collected, the 18 day decay intervals used a count time of 16 hours and the 6 day decay interval used a time of 4 hours. These count times are included to complete development of the long run count scheme.

TABLE 4.4. Percent peak area error values for a long reactor run of NBS coal and counted at specified times during decay after irradiation

Isotope	Decay Time After Irradiation (d.hr.m)						
	6.10.45	10.2.37	13.5.57	16.1.44	18.4.31	25.6.1	29.21.17
¹⁶⁵ Dy	a	a	12.7	a	a	a	a
¹⁴¹ Ce	13.9	8.4	8.6	a	6.3	a	a
¹⁷⁷ Lu	24.7	23.5	60.	21.	29.	44.	54.
²³³ Pa	7.8	a	10.6	5.7	5.4	a	7.0
¹⁷⁵ Yb	28.0	22.	34.	38.	68.	a	a
¹⁸¹ Hf	5.8	6.9	16.2	20.	27.	19.	20.
¹³⁴ Cs	43.	29.	27.5	20.	17.	19.	16.
⁴⁵ Sc	1.9	1.2	1.3	a	0.9	0.9	0.9
⁸⁶ Rb	a	31.	36.	25.	27.	31.	39.
⁵⁹ Fe	7.1	3.9	4.0	2.6	2.7	2.9	3.0
⁶⁰ Co	21.	9.8	10.4	6.6	7.	6.4	6.
¹⁸² Ta	a	80.6	a	33.5	30.	39.	42.
²⁴ Na	3.7	50.	a	a	a	a	a
¹⁵¹ Eu	a	40.	31.	17.	17.	18.	14.
⁴² K	82	a	a	a	a	a	a
¹⁴⁰ La	2.0	3.1	7.2	9.9	a	a	a
¹²⁴ Sb	85.	51.	a	34.	31.	30.	a
⁷⁵ Sc	a	a	a	48.	a	a	70.
¹⁶⁰ Tb	a	a	a	a	45.	79.	62.
⁶⁵ Zn	a	a	a	a	0.8	a	0.9
¹³¹ Ba	a	a	a	53.	53.	60.	a

^aValue not available because of species decay, interference, or insignificance.

TABLE 4.5. Frequency distribution of how often the decay time is ranked first, second or third by peak area error of each isotope

rank	Decay Time After Irradiation (d.hr.m)						
	6.10.45	10.2.37	13.5.57	16.1.44	18.4.31	25.6.1	29.21.17
1	2	1	1	5	6	1	3
2	1	4	0	3	5	2	3
3	1	1	3	2	1	5	2

4.1.4 Isotope Identification and Quantification

The majority of activation products emit gamma rays of various energies as well as varying intensities. The photopeak energies and intensities for isotopes expected to be found in coal and fly ash are given in Appendix B. By calibrating the detection system to read out in energy units, the measured spectrum provides the photopeak energies. The isotopes are identified by comparing the measured peak energies with the values given in Appendix B. After an isotope has been identified, one photopeak is selected to provide information required for quantitative analysis. The peak selected must have the minimum of interference from other spectrum structure such as other peaks, escape peaks, etc. This selection process is detailed in the articles by Abel and Rancitelli,² Block and Dams,⁴ Obrušník, Stáková and Blazek⁹ and Rowe and Steinnes.¹⁰ Using this information as a guideline, Table 4.6 is constructed to identify the gamma-ray to be used to quantify a specific element. Table 4.6 also provides information concerning the daughter isotope half life and the parent elemental concentration in the NBS Standard Reference Material 1632a.

Before attempting to set up some analysis procedure, a preliminary identification run is made on both the NBS coal and the NBS coal fly ash. Two samples of approximately 700 mg of each material are prepared in polyethylene vials. One sample of each is irradiated in the RSR for 5 minutes at a reactor power of 225 kW, is allowed to decay for 15 min, then is counted for 422 seconds. The resulting coal spectrum is shown in Fig. 4.1 and the fly ash spectrum in Fig. 4.2. The second sample of each is irradiated at the same power for eight hours, allowed to decay for 11

TABLE 4.6. Nuclear Data for Possible Activation Products in NBS Coal and Coal Fly Ash

Parent Isotope	Daughter Isotope	γ -Energy ¹¹ Used (keV)	Half Life	Fractional ⁵ Concentration of element	Percent error in fractional conc
Dy-164	Dy-165	94.7	2.334 h	2.2 E-6	13.6
Sm-152	Sm-153	103.2	46.8 h	2.8 E-6	10.7
Ce-140	Ce-141	145.4	32.55 d	3.2 E-5	12.5
Lu-177	Lu-177	208.4	6.71 d	1.8 E-7	16.7
Nd-147	Nd-147	211.3	1.73 h	1.0 E-5	20.0
Se-75	Se-75	264.6	118.45 d	2.6 E-6	11.5
U-238	Np-239	277.6	2.346 d	1.21 E-6	8.3
Th-232	Pa-233	311.9	26.95 d	4.8 E-6	4.2
Ti-50	Ti-51	319.7	5.80 m	1.63 E-3	4.3
Cr-50	Cr-51	320.1	27.70 d	3.4 E-5	5.9
Yb-175	Yb-175	396.3	4.19 d	9.8 E-7	8.2
In-115	In-116m1	416.9	54.12 m	3.6 E-8	11.1
I-128	I-128	443.0	24.99 m	1.8 E-6	11.1
Hf-181	Hf-181	482.0	42.45 d	1.55 E-6	5.2
Ba-130	Ba-131	496.3	12.0 d	1.22 E-4	9.0
Sr-84	Sr-85	514.0	64.85 d	8.4 E-5	10.7
As-75	As-76	559.1	26.32 h	1.1 E-5	18.2
Br-79	Br-80	616.2	17.68 m	4.1 E-5	9.8
W-186	W-187	685.8	23.85 h	6.0 E-7	33.3
Zr-94	Zr-95	743.4	16.9 h	4.7 E-5	12.8
Br-81	Br-82	776.5	35.34 h	4.1 E-5	9.8
Cs-133	Cs-134	795.8	2.06 y	2.0 E-6	15.0
La-139	La-140	815.9	40.27 h	1.8 E-5	11.1
Ga-71	Ga-72	833.9	14.12 h	8.0 E-6	10.0
Tb-159	Tb-160	879.4	72.1 d	3.2 E-7	15.6
Sc-45	Sc-46	889.3	83.80 d	6.8 E-6	8.8
Mg-26	Mg-27	1014.4	9.462 m	1.3 E-3	23.1
Rb-85	Rb-86	1077.2	18.82 d	2.9 E-5	3.5
Fe-58	Fe-59	1099.2	44.56 d	1.16 E-2	2.6

TABLE 4.6. (con.)

Parent Isotope	Daughter Isotope	γ -Energy ¹¹ Used (keV)	Half Life	Fractional ⁵ Concentration of element	Percent error in fractional conc
Zn-64	Zn-65	1115.5	244.0 d	3.1 E-5	19.4
Co-59	Co-60	1173.2	5.272 y	6.5 E-6	3.1
Ta-181	Ta-182	1221.4	155.0 d	4.0 E-7	7.5
Ca-46	Ca-47	1297.0	4.54 d	2.4 E-3	8.3
Na-23	Na-24	1368.6	15.03 h	8.5 E-4	4.7
Eu-151	Eu-152	1408.1	13.2 y	5.5 E-7	5.5
V-51	V-52	1434.1	3.746 m	4.4 E-5	6.8
K-41	K-42	1524.6	12.36 h	4.2 E-3	4.8
Cl-37	Cl-38	1642.2	37.29 m	8.0 E-4	8.8
Sb-123	Sb-123	1691.0	60.20 d	6.0 E-7	15.0
Al-27	Al-28	1778.7	2.241 m	2.9 E-2	10.3
Mn-55	Mn-56	1810.7	2.571 h	3.2 E-5	9.4

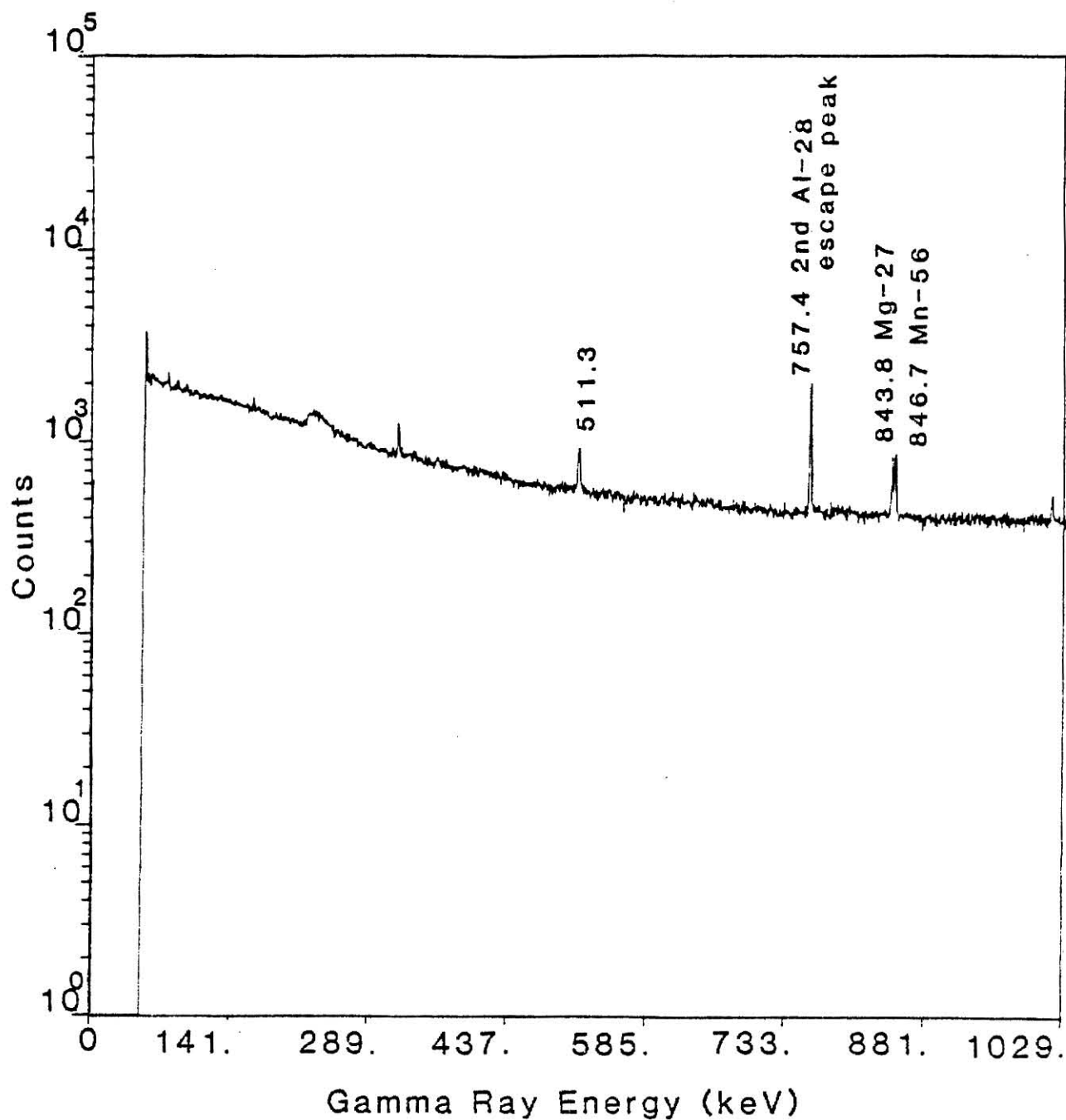


FIG. 4.1. Isotope Identification for NBS Coal after 5 Minute Irradiation at a Reactor Power of 225 kW

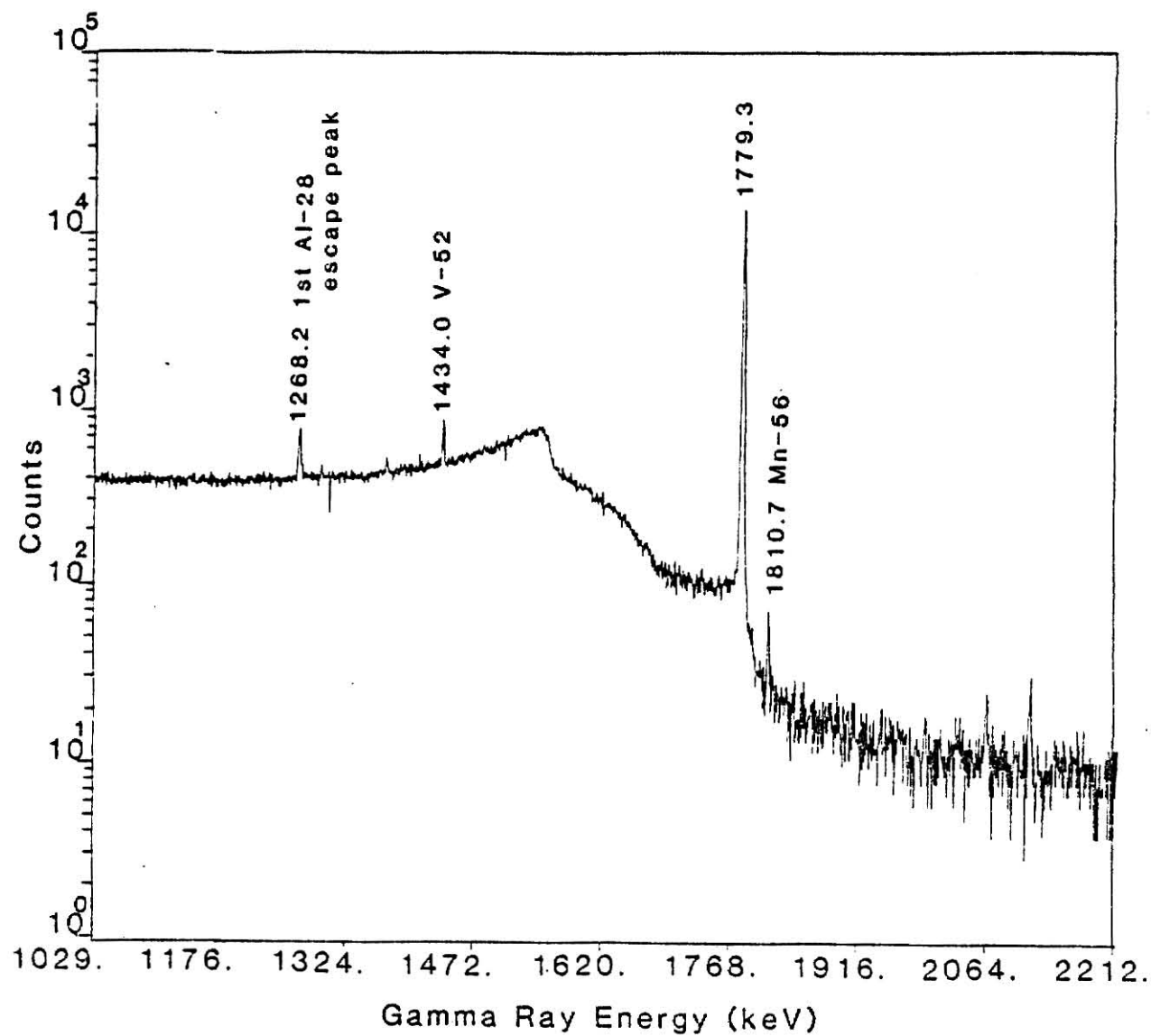


FIG. 4.1 (con.). Isotope Identification for NBS Coal after 5 Minute Irradiation at a Reactor Power of 225 kW

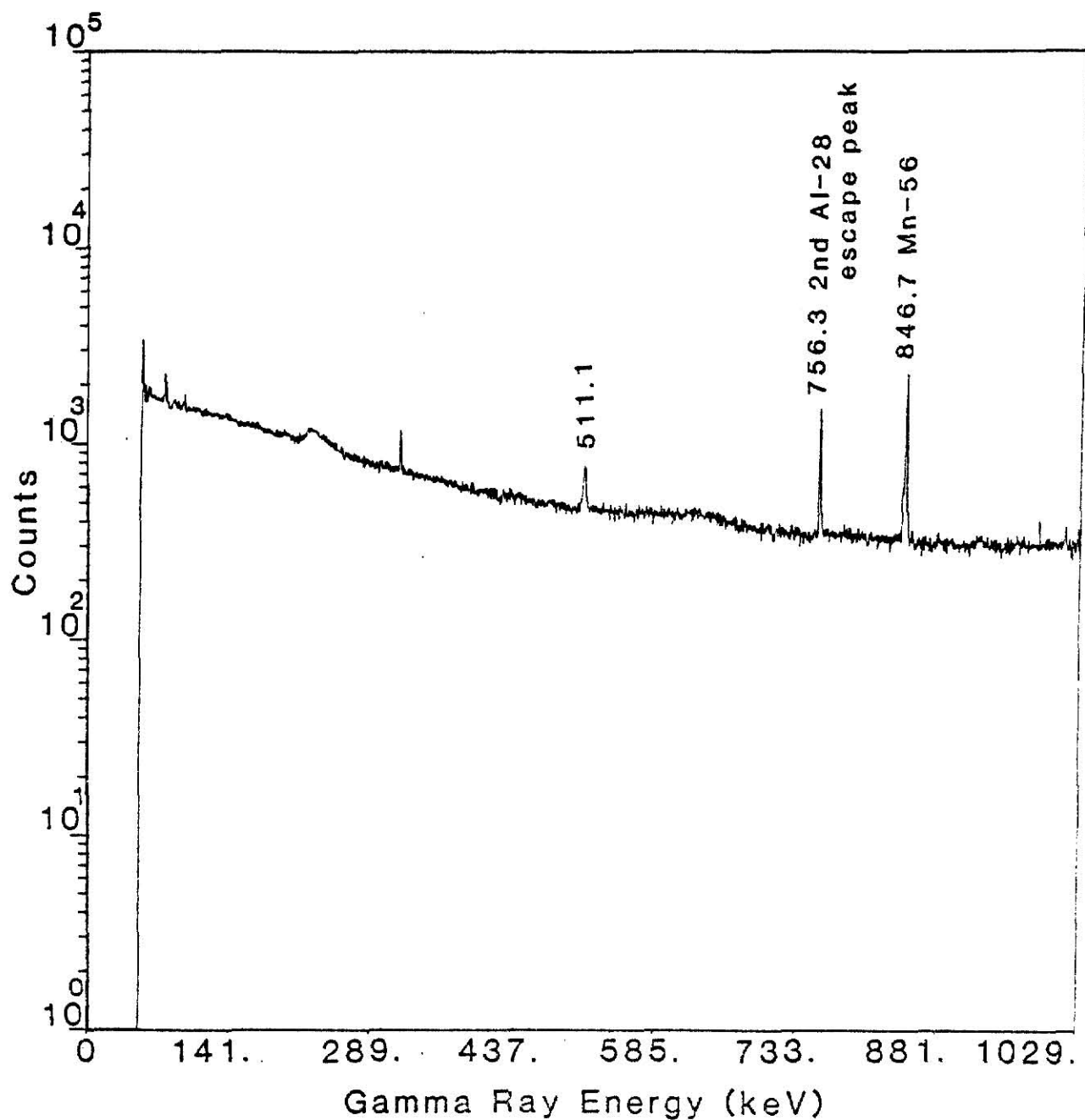


FIG. 4.2 Isotope Identification for NBS Fly Ash after 5 Minute Irradiation at a Reactor Power of 225 kW

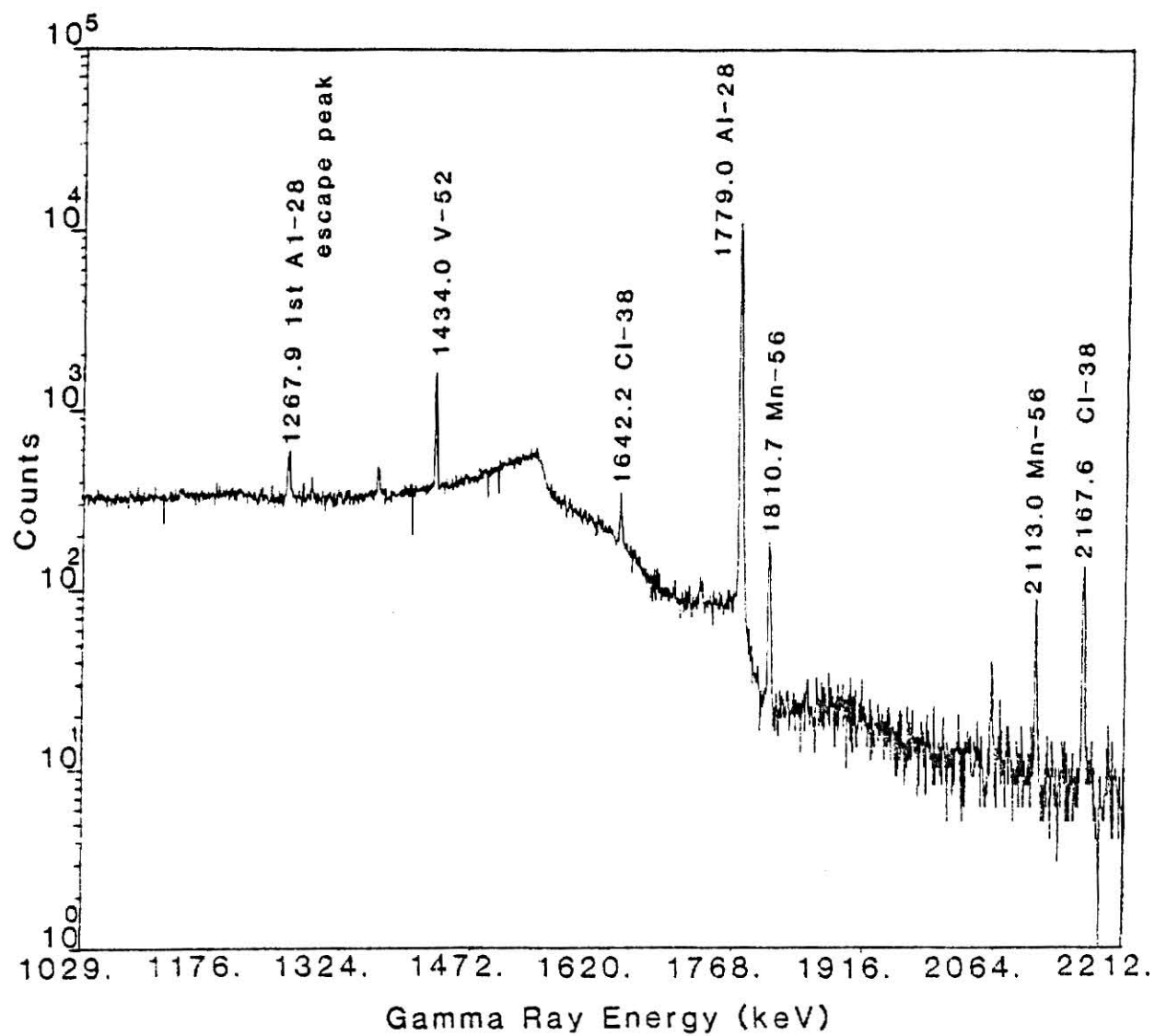


FIG. 4.2 (con.). Isotope Identification for NBS Fly Ash after 5 Minute Irradiation at a Reactor Power of 225 kW

days and is counted for 2 hours. The resulting spectra are shown in Figs. 4.3 and 4.4. The isotopes identified by these 4 runs are listed in Table 4.7.

4.2 Optimized Procedure

4.2.1 Sample Preparation

Polyethylene vials shown in Fig. 3.1 are used to encapsulate the coal and ash. Two samples of each coal and ash are prepared. The first is used to determine the concentration of elements that produce activated daughter products having half lives greater than twenty four hours. The second is used to measure the short lived activation products, i.e., those with half lives less than 24 hours.

To prepare the long lived sample, a 7 ml vial is cleaned by rinsing with warm tap water, then rinsing with methynol and finally using a distilled water rinse. The vials are then placed on "Kimwipes" and allowed to air dry. All further handling of the vials is done with bone tipped tweezers or with hands covered with surgical gloves. The vial is then weighed on the Mettler balance.

Coal is transferred to the vial using a stainless steel spatula and a glass funnel. The vial is filled to a line molded on the vial which marks the lowest penetration of the cap. Extreme care is taken, so that no coal is put on the outside or on the top edge of the vial. The cap is then shut and the vial-sample combination is weighed. An electric soldering iron is used to heat seal the cap onto the vial. The iron is also used to engrave an identifying code name onto the side of the vial. Care is taken not to penetrate the vial.

An iron wire is wrapped around the middle of the vial and the ends are twisted together. Excess wire is trimmed off using wire cutters.

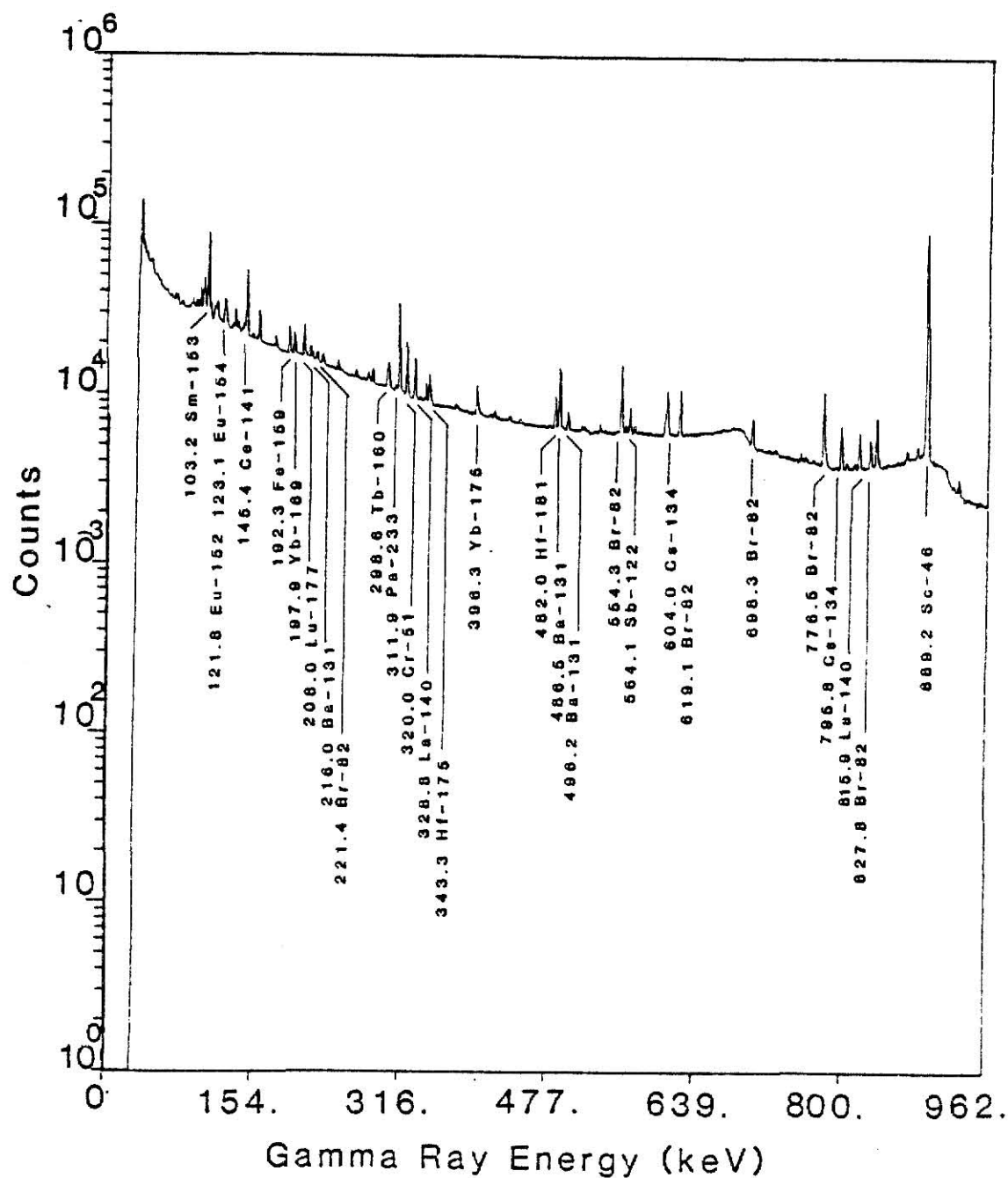


FIG. 4.3. Isotope Identification for NBS Coal after 8 Hour Irradiation at a Reactor Power of 225 kW

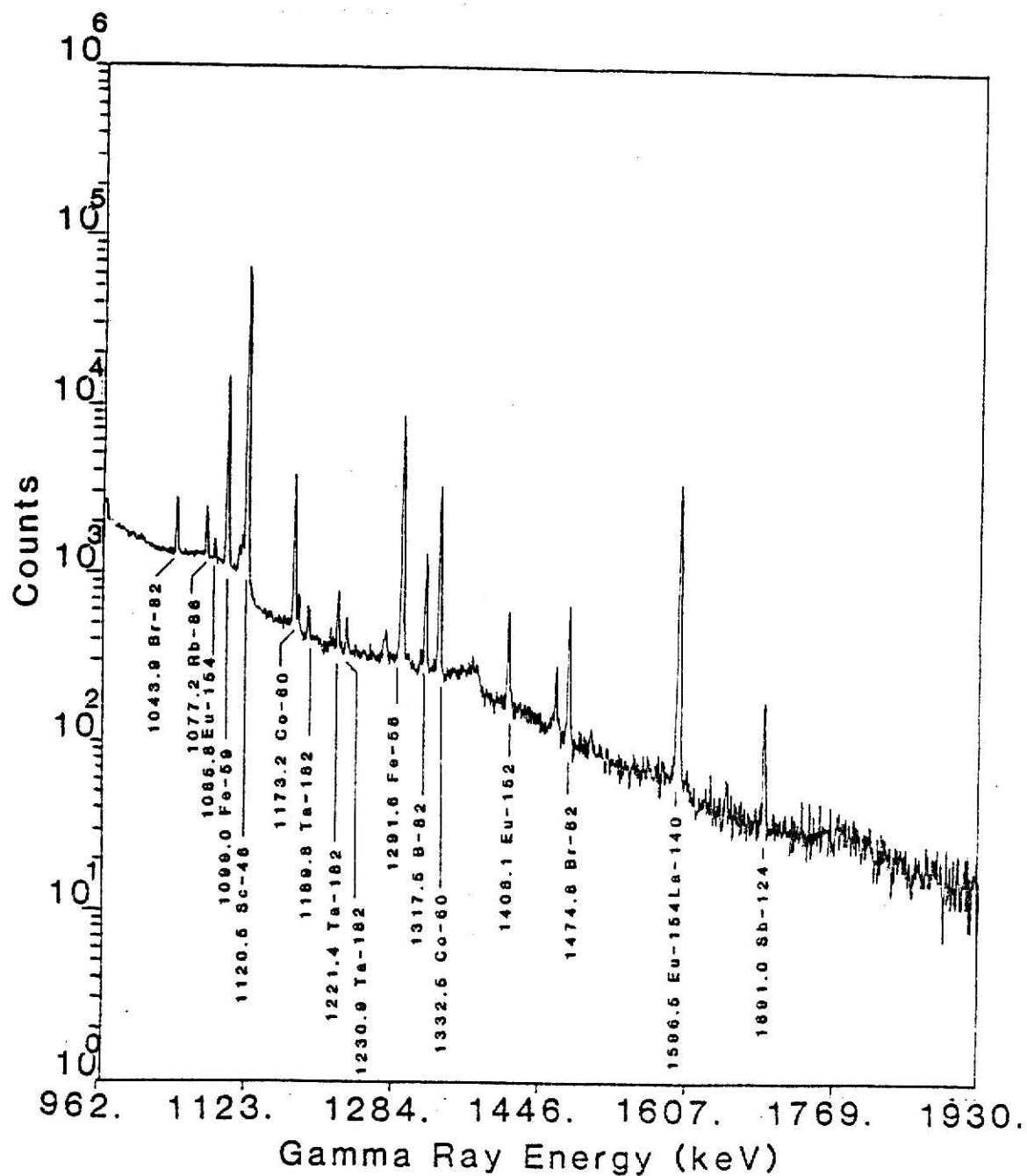


FIG. 4.3 (con.). Isotope Identification for NBS Coal after 8 Hour irradiation at a Reactor Power of 225 kW

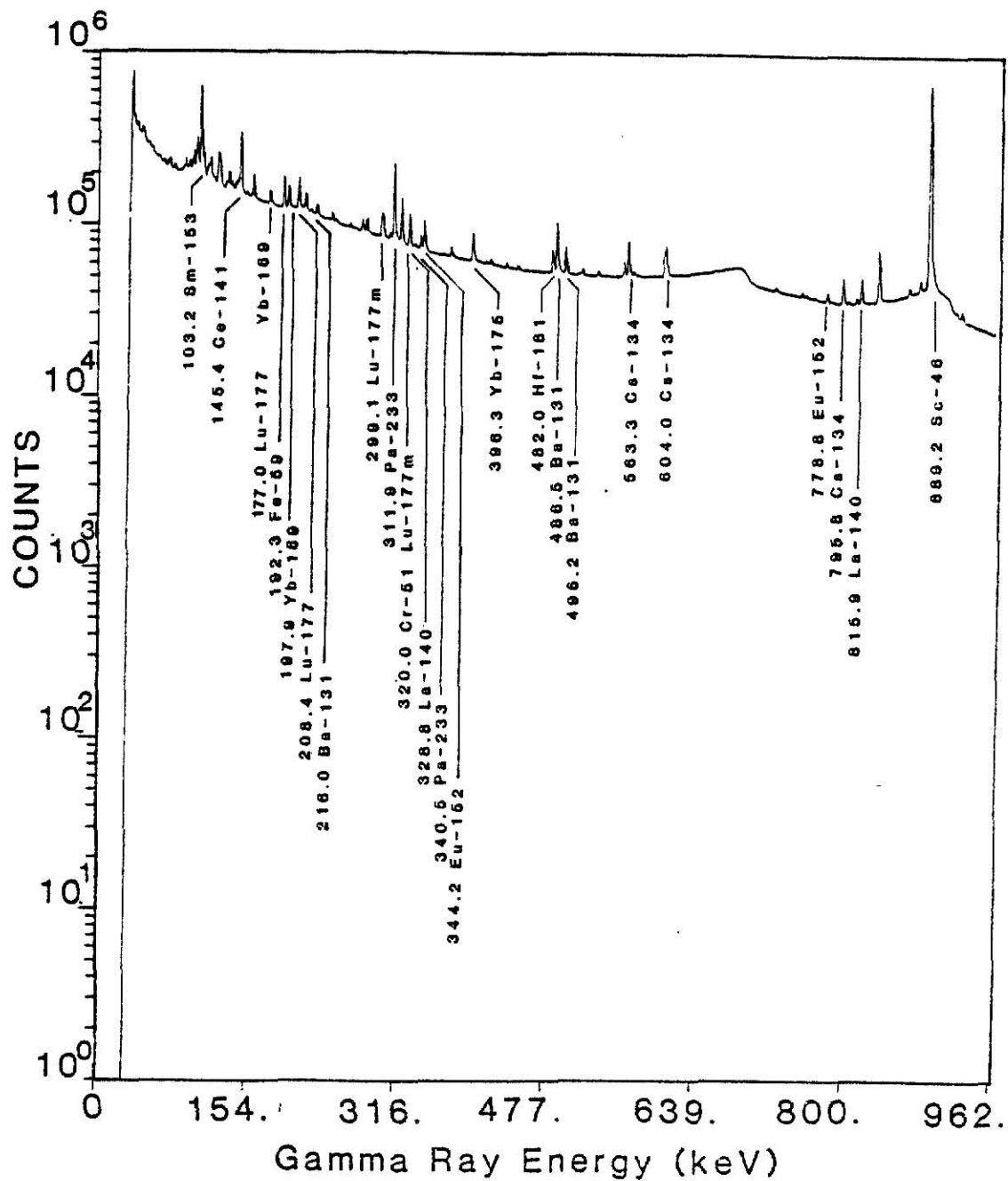


FIG. 4.4. Isotope Identification for NBS Fly Ash after 8 Hour Irradiation at a Reactor Power of 225 kW

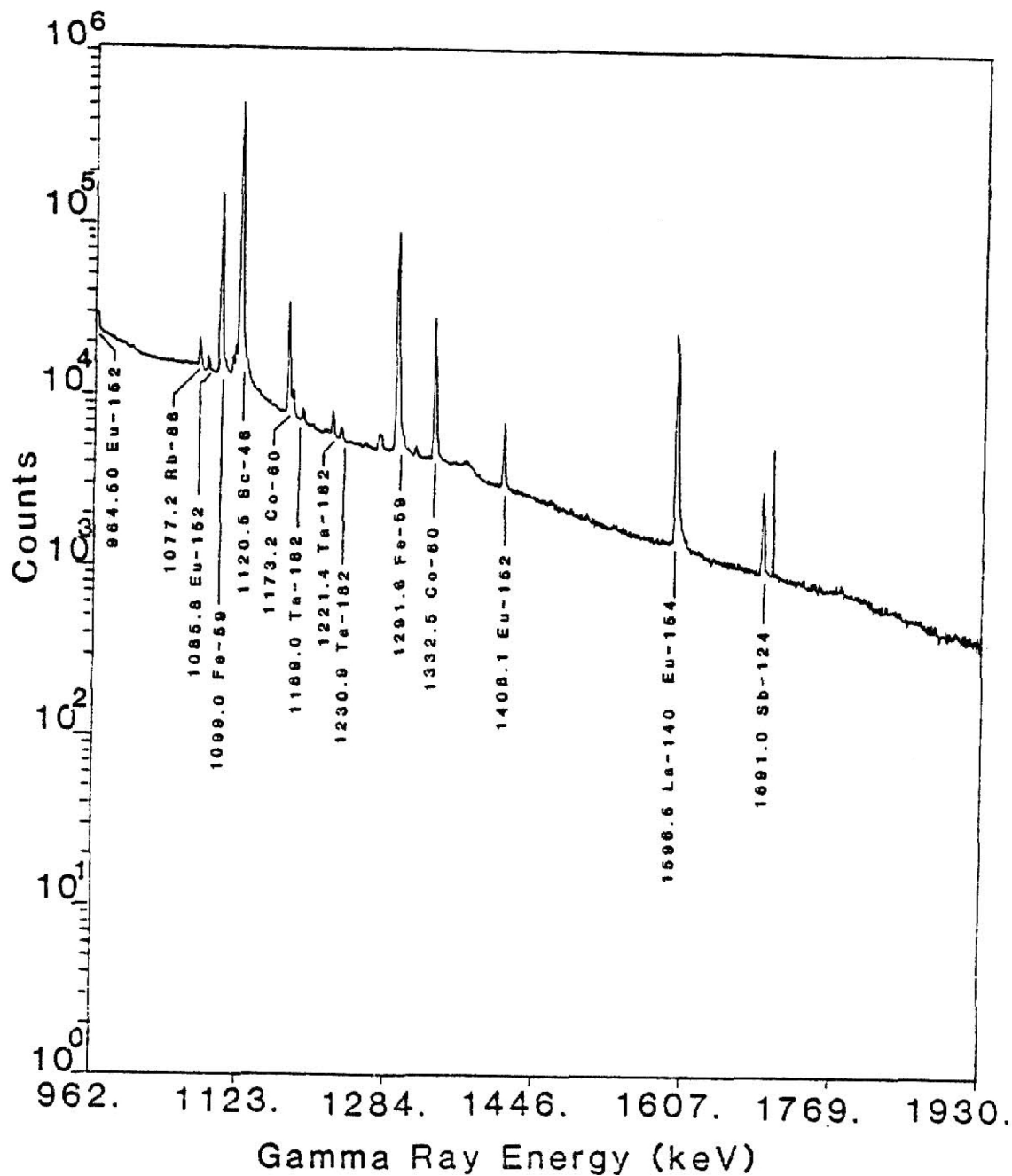


FIG. 4.4 (con.). Isotope Identification for NBS Fly Ash after 8 Hour Irradiation at a Reactor Power of 225 kW

TABLE 4.7. Isotopes Identified in NBS Coal and Coal Fly Ash Standards

<u>Isotope</u>	<u>Coal</u>	<u>Coal Fly Ash</u>	<u>Identified in Long or Short run</u>
Sm-153	x	x	L
Ce-141	x	x	L
Lu-177	x	x	L
Th-233	x	x	L
Cr-51	x	x	L
Yb-175	x	x	L
Hf-181	x	x	L
Ba-131	x	x	L
Br-82	x		L
Cs-134	x	x	L
La-140		x	L
Sc-46	x	x	L
Mg-27	x		S
Rb-86	x	x	L
Fe-59	x	x	L
Co-60	x	x	L
Ta-182	x	x	L
Fu-152	x	x	L
V-52	x	x	S
Cl-38		x	S
Sb-123	x	x	L
Al-28	x	x	S
Mn-56	x	x	S

The vial is placed in a 2 in. x 3 in. polyethylene bag to prevent contamination of the exterior of the vial. Excess air is removed from the bag and the bag is heat sealed. The sample is ready for irradiation.

The short lived sample is to be placed into a 1 ml polyethylene vial. The vial is cleaned using the same method used for the long lived samples. The vial is then weighed. The vial is filled to the molded line with coal or ash using a spatula. The cap is closed and the vial-sample is weighed. The cap tab is cut off using a stainless steel blade. The vial and sample are placed inside a clean 7 ml vial. The outer vial is then heat sealed and an identifying code etched on its surface. A tungsten wire is wrapped around the outer vial at a level equal to the middle of the coal sample. The wire ends are twisted and the excess trimmed.

4.2.2 Sample Irradiations

Two neutron irradiations are required for each coal or ash. The first is used to determine the concentrations of elements whose activated daughters have half lives greater than 24 hours. The second determines the elemental concentrations of the parent isotopes of short lived activation products.

The first sample is inserted into the Rotary Specimen Rack (RSR) of the TRIGA Mark II Reactor. The reactor is brought to a steady state power of 225 kW ($\phi_{th} \sim 1.6 \times 10^{12}$ nv). To maintain as much uniformity as possible during irradiation, it is recommended that the shim and pulse rods be fully withdrawn and the reg rod used to maintain the power level for 8 hours. At the end of that time the reactor is shut down and a timer is started to record the decay time of the sample.

The second sample is placed inside a reactor loading vial which is attached by string to the "fishing pole" loading device. With the reactor at 225 kW, insert the sample into the RSR and irradiate for 5 minutes. Upon removal of the sample from the RSR start a timer to measure decay times.

4.2.3 Energy Calibration

In order to determine the energy of the spectrum photopeaks, the Series 80-Ge(Li) system needs to be calibrated to readout in energy units instead of channel numbers. This is accomplished by measuring the spectrum of some known gamma-ray emitters and recording what peak channel number corresponds to the incident gamma-ray energy. The Series 80 can then determine a functional relationship between energy and channel number which it can use to solve for the energy corresponding to any intermediate channel number.

The energy calibration is a two step process. The first step is to collect a spectrum from a ^{137}Cs and ^{60}Co source. This spectrum provides three energy-channel number data points to which a linear fit is made. The second step is to place the NBS mixed Radionuclide point source on the detector along with the ^{137}Cs and ^{60}Co sources that are already present. A spectrum is collected. The linear energy calibration allows for identification of the peak to be used in the final calibration. A region of interest is established around each of the peaks specified in Table 4.8. The energy calibration is cleared and a second order fit is made to the peaks of Table 4.7 and the corresponding channel numbers. This calibration spectrum is stored on magnetic tape. The second order calibration is used to identify peaks in the coal and coal fly ash spectra.

TABLE 4.8. Gamma Ray Energies
Used for 2nd Order
Energy Calibration
of the Series 80
MCA¹³

Isotope Description	Gamma-ray Energy (keV)
¹⁵⁵ Eu	86.6
¹⁵⁴ Eu	123.1
¹²⁵ Sb	176.4
¹²⁵ Sb	427.9
¹³⁷ Cs	661.6
¹⁵⁴ Eu	723.3
⁶⁰ Co	1173.2
¹⁵⁴ Eu	1274.4
⁶⁰ Co	1332.5
⁶⁰ Co sumpeak	2505.7

4.2.4 Post-irradiation Sample Handling and Counting

To maximize the areas of the photopeaks, a specified counting scheme involving various decay and counting times is used. These schemes are detailed in Table 4.9.

After the long lived sample has decayed at least 5 days, the coal is to be transferred to another vial for counting. This removes any contribution from trace element activation of the vial. The person transferring the coal should be wearing surgical gloves on both hands to minimize skin contamination and the transfer should be accomplished inside a fume hood to minimize exposure to airborne radiation. The heat seal is broken with a stainless steel spatula. The coal is poured through a glass funnel into a clean 7 ml vial which has been weighed. The vial is capped, the sample-vial combination is weighed then the vial is heat sealed. The difference between the vial mass and the sample and vial mass is the sample mass. The irradiation vial is disposed of as waste.

After 6 days of decay the transferred sample is placed on shelf 1 of the rack shown in Fig. 3.5. The sample is placed upright above the detector center with $\frac{1}{2}$ inch of lucite between the sample and detector surface. The sample is counted for 4 hours. After 18 days of decay, the sample is placed in the same counting geometry and is counted for 16 hours. After each count the spectrum is stored on magnetic tape.

The short lived sample is counted three times. Since there is insufficient time between irradiation and counting to transfer the sample to a clean vial, the counting is accomplished in the irradiation vial. The first count begins after 5 minutes decay with the sample placed upright above the detector center on shelf number 10 of the rack. The

TABLE 4.9. Counting and Decay Times for Long and Short Lived Samples

Sample Description	Count Number	Decay Time	Count Time
Long lived sample	1	6 days	4 hours
(8 hour irradiation)	2	18 days	16 hours
Short lived sample	1	5 minutes	600 second ^(a)
(5 minute irradiation)	2	35 minutes	1800 second
	3	4 hours	8 hour

(a) count on shelf 6 of rack all others on shelf 1.

count is for 600 seconds. The sample is lowered to shelf number 1 and a count of 1800 seconds is started after 35 minutes of post-irradiation decay. The last count occurs 4 hours after irradiation and is for a period of 8 hours in the same counting geometry as count number 2.

Both the long and short lived samples have fluence monitors wrapped around the vials. Before sample transfer or counting, the wires are removed and weighed on the Mettler balance. To identify them, a piece of transparent tape is folded in half over the wire rings and an identifying label is written on the tape.

The wire is counted on Ge(Li) No. 1. It is placed flat on 3/8 inch of lucite which is then placed on the detector and adjusted so the wire ring is above the detector center. The count is begun and allowed to continue until a peak area of 20,000 counts is measured for the ^{59}Fe peak at 1099 keV or for the ^{187}W peak at 685.8 keV. The clock and live times for this count are recorded.

4.2.5 Data Reduction

As shown in Chapter 2, there are a substantial number of data values to be recorded for each sample before the concentration of some element can be calculated. These values include peak area, decay times, sample masses, etc. To organize this data and to help ensure a value is not omitted, some data forms were prepared. Copies of these forms are given in Appendix A. Even though the forms should be self explanatory, some instructions concerning the completion are included in Appendix A.

As various steps in the analysis procedure are completed, values should be recorded onto the forms. For example, when preparing the standard sample, record the sample mass as the fourth entry of the only

line in data set no. 6. The wire mass for the standard would be the third entry on that line. The form identifies the different samples to be analyzed as different case numbers. If the Colorado coal is case number 1 and you begin a count at 8:53 a.m., ten days after irradiation you would record this information on line 3 of data set no. 5 as 10,8,53,0.

When analyzing the spectrum for this count, you would set regions of interest about the photopeaks corresponding to the elements you wish to quantify. For example, to identify thorium go to Table 4.6 to find the energy of the gamma ray to be used. Locate that peak on the spectrum, set a region of interest, and record the peak area and peak area error from the analyzer onto the line corresponding to that isotope and case no. in data set no. 9. As explained in Appendix A, you identify each isotope with the number assigned in data set no. 2. An example analysis is presented in Appendix D.

The data forms combine to form an input file for a Watfiv computer program which then calculates the elemental concentrations. A complete data form set is required for each spectrum to be analyzed. Thus, there should be 5 input files, each corresponding to one of the counts required by Table 4.9.

5.0 Results and Conclusions

Before applying the cookbook procedure of Section 4.2 to the coal and fly ash samples, an experiment is designed to test the procedure's accuracy. Since the elemental concentrations of the NBS Coal Standard are known, a sample of it is prepared and analyzed as an "unknown" sample. Comparing the measured concentrations with the known concentrations of the standard allows one to make a judgement concerning the accuracy to be expected when measuring specific elemental concentrations. The resulting concentrations are compared with the known (expected) values in Table 5.1.

The elements in Table 5.1 are ordered so that the absolute value of the deviation between the measured and expected concentrations increases as one moves down the table. The table shows the cerium yielded the smallest deviation and hence is the element that can be measured to the highest degree of accuracy. Proceeding down the list, Lanthanum had a deviation of 5.6 percent, Cesium, 9 percent, Tanalum, 20 percent, Uranium, 39 percent and Samarium was the worst at 103 percent. The error bands for the measured and expected values overlap for all but Iron and Rubidium however the deviations from the expected concentrations are 8.6 percent and 22 percent respectively. Thus, the only element that cannot be measured with a reasonable degree of confidence is Samarium.

The large deviation for Samarium probably results from spectrum interference because of the relatively low energy gamma-ray, 103 keV, used to identify Samarium. The 103-keV gamma-ray is the highest energy, highest yield gamma ray for the Samarium activation product ^{153}Sm . The other Samarium activation product ^{155}Sm is short lived, and its

TABLE 5.1 Comparison of Measured Elemental Concentrations for NBS Standard Reference Material 1632 A with Expected Concentrations - Elements Ordered by Deviation of Measured Mean from Expected Mean

Element	Standard Concentration (ppm)			Measured Concentration (ppm)		
	Lower ^c Limit	Expected Value	Upper ^c Limit	Lower ^c Limit	Measured Value	Upper ^c Limit
Ce	28.0	32.0	36.0	27.74	32.07	36.40
V	41.00	44.0	47.0	30.40	44.23	58.06
Al	2.60 ^a	2.90 ^a	3.20 ^a	2.51 ^a	2.81 ^a	3.11 ^a
Br	37.00	41.00	45.00	34.10	39.73	45.36
Na	810.0	850.0	890.0	777.4	822.8	868.2
Ba	111.0	122.0	133.0	64.79	118.0	171.3
Eu	0.52	0.55	0.58	0.32	0.57	0.82
Ga	7.20	8.00	8.80	4.77	7.65	10.53
K	4000.	4200.	4400.	3625.9	3975.1	4324.2
Dy	1.90	2.20	2.50	1.77	2.08	2.39
La	16.0	18.0	20.0	12.46	17.00	21.54
Ti	1560.0	1630.0	1700.0	777.8	1511.2	2244.5
Th	4.60	4.80	5.00	4.05	4.45	4.85
Cr	32.00	34.00	36.00	28.65	31.52	34.39
Co	6.30	6.50	6.70	5.64	6.02	6.40
Hf	1.47	1.55	1.63	1.43	1.67	1.91
Sc	6.20 _b	6.80 _b	7.40 _b	5.64 _b	6.25 _b	6.86 _b
Fe	1.13 _b	1.16 _b	1.19 _b	1.00 _b	1.06 _b	1.12 _b
Cs	1.70	2.00	2.30	1.46	1.82	2.18
Lu	0.15	0.18	0.21	0.12	0.16	0.20
Tb	0.27	0.32	0.37	0.18	0.36	0.54
Cl	730.0	800.0	870.0	588.78	696.16	803.54
Sb	0.51	0.60	0.69	0.39	0.52	0.65
Ta	0.37	0.40	0.43	0.25	0.32	0.39
Rb	28.00	29.00	30.00	17.44	22.56	27.68
Se	2.30	2.60	2.90	1.14	1.99	2.84
Yb	0.90	0.98	1.06	0.96	1.22	1.48
U	1.11	1.21	1.31	0.36	0.86	1.36
Sm	2.50	2.80	3.10	2.96	5.67	8.38

^aValues are weight percent - measurements of Al concentration are very sensitive to Si concentration of sample

^bValues are weight percent

^cOne standard deviation about the mean

relatively low gamma ray energies are not detectable in the short run spectra. Since the deviation is so large for Samarium, this isotope is not reported as one that can be quantitatively identified using the procedure developed in this thesis.

The elements that can be quantitatively identified by the procedure of Section 4.2 are listed in Table. 5.2.

The analysis procedure was applied to the five coals identified as Illinois-6, Wyoming-1, Wyoming-2, Missouri-Kansas and Colorado. The measured elemental concentrations and the expected error for each element are presented in Table 5.3.

The Missouri Kansas coal tends to have higher concentrations than the other coals of almost all the trace and minor elements and the difference is more pronounced in the metal concentrations, for example Cr, Fe, Ti, Th, U and V. As for the two Wyoming coals, Wyoming-2 tends to have higher concentrations in the metals. The Wyoming-1 coal has concentrations more comparable to the Colorado coal than the Wyoming-2 coal. The Illinois-6 element concentrations do not follow any trend when compared with the other coals.

The elemental concentrations of the NBS Fly Ash and the ash sample from A. Anderson were measured and the results are presented in Tables 5.4 and 5.5, respectively.

The NBS Fly Ash elemental concentrations are certified for some of the elements in Table 5.4. NBS reports a concentration of 9.40 ± 0.10 weight percent for iron, 196 ± 6 ppm for chromium, 131 ± 2 ppm for rubidium, 10.3 ± 0.6 for selenium and 24.7 ± 0.3 for thorium. Comparison of these values with Table 5.4 shows reasonable agreement but with a trend towards under-prediction for the measured values. This trend is

TABLE 5.2. Elements in Coal and Coal Fly Ash that are Quantitatively Identifiable using the Procedure Developed in this Thesis

Aluminum	Lutecium
Antimony	Potassium
Barium	Rubidium
Bromine	Samarium
Cerium	Scandium
Cesium	Selenium
Chlorine	Sodium
Chromium	Tantalum
Cobalt	Terbium
Dysprosium	Thorium
Europium	Titanium
Gallium	Uranium
Hafnium	Vanadium
Iron	Ytterbium
Lanthanum	

TABLE 5.3. Elemental Concentrations for Various Coal Samples.

Element	Illinois-6 Coal		Wyoming-1 Coal		Wyoming-2 Coal		Missouri-Kansas		Colorado Coal	
	Conc. (ppm)	Error (ppm)	Conc. (ppm)	Error (ppm)	Conc. (ppm)	Error (ppm)	Conc. (ppm)	Error (ppm)	Conc. (ppm)	Error (ppm)
As	2.85	0.95	b	-	b	-	41.54	11.26	1.66	0.61
Ba	43.59	24.74	257.7	69.3	253.1	71.5	170.3	60.3	199.1	54.0
Br	3.38	0.62	b	-	b	-	5.43	3.27	b	-
Ce	8.56	1.25	10.36	1.44	24.32	3.30	26.01	3.57	15.82	2.15
Cl	1594.7	192.	b	-	b	-	1634.	234.	b	-
Co	2.38	0.19	1.48	0.13	2.40	0.21	12.22	0.74	1.13	0.12
Cr	16.56	1.58	4.06	0.77	10.82	1.35	30.32	2.87	5.35	0.77
Cs	0.80	0.18	b	-	0.97	0.21	2.40	0.48	0.36	0.10
Dy	2.07	0.42	2.51	0.49	3.23	0.63	6.02	1.09	2.16	0.42
Eu	0.20	0.03	0.21	0.03	0.25	0.04	0.48	0.06	0.23	0.03
Fe	1.034 ^a	0.05 ^a	1904.	114.	6754.	352.	3.68 ^a	0.18 ^a	2907.	158.
Ga	7.11	3.03	b	-	7.88	3.49	16.76	7.10	9.76	2.90
Hf	0.47	0.11	0.54	0.09	0.57	0.13	1.43	0.23	1.48	0.18
La	5.19	0.94	5.85	1.91	0.12	0.03	16.85	4.26	9.64	1.31
Lu	0.07	0.02	0.06	0.02	0.08	0.03	0.21	0.05	0.09	0.02
Na	488.9	54.7	b	-	b	-	b	-	321.2	44.7
Rb	10.42	3.08	b	-	8.41	3.44	34.59	7.05	6.19	1.89
Sb	0.54	0.12	0.19	0.06	0.37	0.10	5.20	1.03	0.19	0.06
Sc	2.38	0.23	1.61	0.16	2.33	0.23	4.90	0.48	1.72	0.17
Se	2.01	0.73	0.54	0.38	0.70	0.46	4.32	1.46	0.77	0.40
Sm	1.09	0.26	1.26	0.35	2.87	0.76	3.19	0.69	1.29	0.22
Ta	0.12	0.05	b	-	0.12	0.06	0.36	0.09	0.18	0.04
Tb	0.14	0.09	0.12	0.07	0.25	0.18	0.31	0.19	0.16	0.08
Th	1.58	0.11	1.39	0.10	3.40	0.22	3.49	0.24	2.57	0.16
Ti	1501.9	604.	1773.9	605.3	1388.	758.	5241.	1413.	1565.	704.
U	2.19	0.42	0.47	0.25	1.87	0.51	6.17	1.21	0.72	0.17
V	139.3	19.16	86.8	13.7	107.1	18.3	211.8	31.8	48.8	13.5
Yb	0.40	0.10	0.31	0.10	0.52	0.24	0.94	0.37	0.45	0.09

^a weight percent^b not detected

TABLE 5.4. Elemental Concentrations for NBS Fly Ash

Element	Concentration (PPM)	Expected Error (PPM)	Element	Concentration (PPM)	Expected Error (PPM)
Ba	1136.7	316.3	Rb	125.1	19.3
Ce	183.4	24.5	Sb	15.76	3.16
Co	40.8	2.4	Sc	40.40	3.93
Cr	182.2	15.0	Se	7.15	2.48
Cs	8.18	1.49	Ta	1.69	0.3
Dy	37.98	5.73	Tb	2.65	1.04
Eu	3.83	0.38	Th	22.81	1.41
Fe	9.2 ^a	0.46 ^a	Ti	2.73 ^a	0.8 ^a
Hf	7.44	0.89	V	1451.4	179.6
Lu	1.09	0.25	Yb	7.12	2.52

^a weight percent

due to the higher absolute activity of the ash sample when compared with the NBS coal reference material. This higher activity leads to a higher detection system dead time and hence a systematic error in the efficiency ratio of Eq. (21). The increase in dead time decreases the absolute efficiency which decreases the mass ratio calculated by Eq. (22) and hence results in an underprediction of concentration. Since no relationship between system dead time and resultant loss of sensitivity is determined, the only way to correct for this under prediction is to re-analyze the sample so that the absolute activities are more equally matched. The A. Anderson ash sample is truly an unknown sample since no information about its history or intended use was provided. Hence, the results were reported as shown in Table 5.5.

TABLE 5.5. Elemental Concentrations of A. Anderson's Coal
Fly Ash Sample

Element	Concentration (PPM)	Expected Error (PPM)	Element	Concentration (PPM)	Expected Error (PPM)
Ba	0.307 ^a	0.066 ^a	K	0.246 ^a	0.033 ^a
Ce	9.07	1.22	Lu	0.93	0.18
Co	24.11	1.43	Na	1.13 ^a	0.06 ^a
Cs	0.73	0.35	Rb	12.79	6.32
Dy	8.44	0.75	Sb	0.94	0.24
Eu	2.96	0.29	Sc	26.08	2.56
Fe	6.77 ^a	0.35 ^a	Sm	13.04	1.60
Ga	10.75	4.31	Ta	1.59	0.27
Hf	15.23	1.60	Yb	3.36	0.70

^aweight percent

6.0 References

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APPENDIX A: DATA FORMS

The following data forms are used to record information required by the computer program MASCAL which calculates elemental concentrations. The program MASCAL is a Watfiv program that reads all its input in free format. The data forms are constructed so that the information is ordered as required by the program. Each line on the data sheet represents a data card and the values to be typed on the card are underlined. For example, the first card of data set No. 3 would read:

44.56, 'D'

for an iron fluence monitor.

The data are divided into a series of data sets. The first data set consists of a single card containing information concerning how many cases, NCASES, or samples are to be compared to the standard and how many isotopes, NISO, are to be quantified.

Each isotope to be quantified is identified by the number assigned in data set no. 2. The data set will consist of NISO cards each containing a single isotope name no longer than twenty characters.

Data set no. 3 is a set of NISO cards listing the numeric value of the half life of the fluence monitor and its corresponding unit of either year (Y), day (D), hour (H), minute (M), or second (S).

Data set no. 4 records the time information for the standard. Each value consists of the day, hour, minute and second components. The clock time and live time values are given in seconds and are measured in channels 0 and 1 of Series 80 spectrum.

Data set no. 5 consists NCASES groups of data where each group contains the time information for a specific case.

Data set no. 6 is a single card with information about the standard sample fluence wire as well as the sample mass of the standard.

Data set no. 7 consists of NISO cards with each containing the peak area, error in the peak area, the fractional concentration and the error in the fractional concentration information for a specific isotope.

Remember the isotopes are ordered in data set no. 2 and the same ordering should apply here. The concentration values for a number of isotopes are given in Table 4.6.

Data sets no. 8 and 9 are repeated for each case. So there will be NCASES groups of data sets 8 and 9. Data set no. 8 is a single card consisting of the fluence wire data and the mass of a specific case. The corresponding data set no. 9 is a group of NISO cards each containing the peak area and the error in peak area for the isotopes. The ordering of the groups must be consistent with the ordering of the information in data set no. 5. The input for MASCAL is now complete.

A sample analysis is given in APPENDIX D showing the completed forms the corresponding card input for MASCAL and a copy of the output.

The last form in this appendix is not input for MASCAL but is a method of recording instrument setting used during data collection. The settings must always be consistent with those used during measurement of the standard the analysis is based upon.

DATA RECORD
(FORMAT FOR INPUT TO MASCAL)

DATA SET NO. 1

NCASES, NISO _____, _____

DATA SET NO. 2

<u>ISOTOPE NO.</u>	<u>DESCRIPTION</u>
1	_____
2	_____
3	_____
4	_____
5	_____
6	_____
7	_____
8	_____
9	_____
10	_____
11	_____
12	_____
13	_____
14	_____
15	_____
16	_____
17	_____
18	_____
19	_____
20	_____

DATA RECORD
(FORMAT FOR INPUT TO MASCAL)

DATA SET NO. 3

ISOTOPE NO. FLUENCE MONITOR	HALF LIFE
	_____, ' ____'
1	_____, ' ____'
2	_____, ' ____'
3	_____, ' ____'
4	_____, ' ____'
5	_____, ' ____'
6	_____, ' ____'
7	_____, ' ____'
8	_____, ' ____'
9	_____, ' ____'
10	_____, ' ____'
11	_____, ' ____'
12	_____, ' ____'
13	_____, ' ____'
14	_____, ' ____'
15	_____, ' ____'
16	_____, ' ____'
17	_____, ' ____'
18	_____, ' ____'
19	_____, ' ____'
20	_____, ' ____'

DATA SET NO. 4 TIME VALUES FOR THE STANDARD

	DAY		HOUR		MINUTE		SECOND
TSIRR (IRRADIATION TIME)	_____	,	_____	,	_____	,	_____
STR (TIME REMOVED FROM RX)	_____	,	_____	,	_____	,	_____
STS (START TIME FOR COUNT)	_____	,	_____	,	_____	,	_____
STSM (START OF FLUENCE MON.)	_____	,	_____	,	_____	,	_____
CLOCK AND LIVE TIME FOR COUNT			_____	,	_____		
CLOCK TIME FOR FLUENCE COUNT			_____				

DATA SET NO. 5 TIME VALUES FOR THE UNKNOWN

CASE NO.	DAY		HOUR		MINUTE		SECOND
TUIRR (IRRADIATION TIME)	_____	,	_____	,	_____	,	_____
UTR (TIME REMOVED FROM RX)	_____	,	_____	,	_____	,	_____
UTS (START TIME FOR COUNT)	_____	,	_____	,	_____	,	_____
UTSM (START OF FLUENCE MON.)	_____	,	_____	,	_____	,	_____
CLOCK AND LIVE TIME FOR COUNT			_____	,	_____		
CLOCK TIME FOR FLUENCE COUNT			_____				

CASE NO.	DAY		HOUR		MINUTE		SECOND
TUIRR (IRRADIATION TIME)	_____	,	_____	,	_____	,	_____
UTR (TIME REMOVED FROM RX)	_____	,	_____	,	_____	,	_____
UTS (START TIME FOR COUNT)	_____	,	_____	,	_____	,	_____
UTSM (START OF FLUENCE MON.)	_____	,	_____	,	_____	,	_____
CLOCK AND LIVE TIME FOR COUNT			_____	,	_____		
CLOCK TIME FOR FLUENCE COUNT			_____				

DATA SET NO. 5 TIME VALUES FOR THE UNKNOWN

CASE NO.	DAY	HOUR	MINUTE	SECOND
TUIRR (IRRADIATION TIME)	_____	,	_____	,
UTR (TIME REMOVED FROM RX)	_____	,	_____	,
UTS (START TIME FOR COUNT)	_____	,	_____	,
UTSM (START OF FLUENCE MON.)	_____	,	_____	,
CLOCK AND LIVE TIME FOR COUNT	_____		,	_____
CLOCK TIME FOR FLUENCE COUNT	_____			

CASE NO.	DAY	HOUR	MINUTE	SECOND
TUIRR (IRRADIATION TIME)	_____	,	_____	,
UTR (TIME REMOVED FROM RX)	_____	,	_____	,
UTS (START TIME FOR COUNT)	_____	,	_____	,
UTSM (START OF FLUENCE MON.)	_____	,	_____	,
CLOCK AND LIVE TIME FOR COUNT	_____		,	_____
CLOCK TIME FOR FLUENCE COUNT	_____			

CASE NO.	DAY	HOUR	MINUTE	SECOND
TUIRR (IRRADIATION TIME)	_____	,	_____	,
UTR (TIME REMOVED FROM RX)	_____	,	_____	,
UTS (START TIME FOR COUNT)	_____	,	_____	,
UTSM (START OF FLUENCE MON.)	_____	,	_____	,
CLOCK AND LIVE TIME FOR COUNT	_____		,	_____
CLOCK TIME FOR FLUENCE COUNT	_____			

DATA SET NO. 6

Standard Sample	Wire Peak		
Wire Peak Area	Error	Wire Mass	Sample Mass
_____	_____	_____	_____

DATA SET NO. 7

ISOTOPE NO.	PEAK AREA	PEAK AREA ERROR (%)	FRACTIONAL CONCENTRATION	ERROR IN FRACTIONAL CONCENTRATION (%)
1	_____	_____	_____	_____
2	_____	_____	_____	_____
3	_____	_____	_____	_____
4	_____	_____	_____	_____
5	_____	_____	_____	_____
6	_____	_____	_____	_____
7	_____	_____	_____	_____
8	_____	_____	_____	_____
9	_____	_____	_____	_____
10	_____	_____	_____	_____
11	_____	_____	_____	_____
12	_____	_____	_____	_____
13	_____	_____	_____	_____
14	_____	_____	_____	_____
15	_____	_____	_____	_____
16	_____	_____	_____	_____
17	_____	_____	_____	_____
18	_____	_____	_____	_____
19	_____	_____	_____	_____
20	_____	_____	_____	_____

DATA SET NO. 8

UNKNOWN SAMPLE WIRE PEAK AREA	WIRE PEAK ERROR	WIRE MASS	SAMPLE MASS
_____	_____	_____	_____

DATA SET NO. 9

ISOTOPE NO.	PEAK AREA	PEAK ERROR (%)
1	_____	_____
2	_____	_____
3	_____	_____
4	_____	_____
5	_____	_____
6	_____	_____
7	_____	_____
8	_____	_____
9	_____	_____
10	_____	_____
11	_____	_____
12	_____	_____
13	_____	_____
14	_____	_____
15	_____	_____
16	_____	_____
17	_____	_____
18	_____	_____
19	_____	_____
20	_____	_____

CONTROL SETTINGS FOR SERIES 80 MCA

STATUS

CRT = () TIME = () DATE = ()

PRESET

ADC #

LIVE TIME = TRUE TIME =

PRESET LIMITS:

___ NONE			
___ COUNTS	COUNTS =	IN CH#	
___ INTEGRAL	INTEGRAL =	FROM CH#	TO CH#
___ AREA	AREA =	FROM CH#	TO CH#
___ % ERROR	% ERROR =	FROM CH#	TO CH#

ADC

ADC #

MODE = (UNAS) (PHA+) (LIST)

MEMORY = B() GRIN = OFFSET

MEMORY

UNIT = #5B() #6B() #7B() #8B()

UNIT = #1B() #2B()

I/O DEVICE

DEVICE USED()

DEVICE 1 TTY Lines/Pages=66 Lines/Block=8 Parity=2

DEVICE 5 X - Y Plotter

DEVICE 10 Mg Tape Mode BCD Block Size = 256

AMPLIFIER SETTINGS

COARSE GAIN FINE GAIN

LLD ULD

GATE INPUT POLARITY ADC IN

Ge(Li) #

APPENDIX B: Procedures for Entering Gamma-Ray Energy Libraries into the Series 80 and Listings of the Libraries for Isotopes Found in Coal and Coal Fly Ash.

To input a user gamma-ray energy library into the Series 80, the system must first be energy calibrated and the decwriter should be turned on with only the "110" control key depressed. Then set the PROGRAM rotary switch to LIB IN and press D0. Device #1 is the decwriter to be used during input of the library, so press YES to select this device. The library is to be typed in instead of read in so press NO until the asterisk is immediately before the word TYPE, then answer YES to the Series 80 question NEXT? Select the energy units to be used in the library by pressing NO until the asterisk is before the desired units then press YES. Generally, the energy units are keV, so the format 6.1 is adequate. Press YES to select this format. The input editor is now activated and the library can be entered using the decwriter.

There are only two input editor commands. "A" means to add the entry to the bottom of the library and "E" means input is complete and the editor is exited. Two types of entries are possible. One describes the isotope with its half life and the other gives the gamma-ray energy and percent yield.

The format for command or data input is straight forward. All entries are made following the editor prompt "*". So to add an isotope to the library, an "A" is typed after the prompt. The editor will then ask for the name of the isotope, its half life and the units of the half life value. All three are entered using the format:

* Isotope name, half life, unit of half life.

The only valid units for half life are Y, D, H, M and S which obviously

are abbreviations for year, day, hour, minute and second respectively. The editor will then prompt for the gamma-ray energy and yield. The format for entry is:

* gamma-ray energy, percent yield.

For cases when the isotope has multiple emissions, the energies and yields are entered by order of increasing energy. Failure to order entries will result in an input error.

Fig. B.1 is an example listing for input of a library consisting of the isotopes ^{76}As , ^{131}Ba , ^{47}Ca and ^{60}Co . The last two entries give examples of possible input errors.

Once the library is in the Series 80 memory, it can be output in two ways. One is a paper copy using the decwriter, as shown in Fig. B.2, and the other is onto magnetic tape. The magnetic tape can later be used to input the library back into the Series 80.

The Series 80 has a resident library in its memory in addition to the User's defined library. To print the user library on paper, press the ENTER FACTOR control on the Series 80, use the NO to select the ISO-ID option and answer YES to the question NEXT? Select the user library then set the window to some value normally 1 keV. A final YES ends the ENTER FACTOR routine. Set the PROGRAM rotary switch to LIB out, press DO, select device #1 and a find YES to print will begin the output on the decwriter.

To output the library to magnetic tape, set the FUNCTION rotary switch to I/O device, press the select button. To identify the magnetic tape as the I/O device, enter device number 10 and store it with the STORE key. Use the MODE option by entering YES and select a binary

format with a block size of 256. After selecting the I/O device, set the PROGRAM rotary switch to LIB OUT, press D0, select device #10, enter a tag number and begin output by pressing YES.

To read the library back into the Series 80 memory, set the PROGRAM rotary switch to LIB IN, press D0, identify I/O device number 10, enter the tag number of the library desired and begin the read in with a final YES.

For the work in this thesis, the possible activation products are divided into two libraries. The first library consists of the isotopes whose half lives are less than 24 hours and the second contains those greater than 24 hours. Tables B.1 and B.2 list the isotopes in the respective libraries.

TABLE B.1. Gamma-Ray Energy Library for Long Lived Isotopes,
Half Lives Greater than 24 Hours, Present in Coal.

Isotope	Half Life	Gamma-Ray Energy (keV) and Percent Yield							
As-76	26.32 h	559.0	45%	657.0	6%	1212.7	1%		
		1216.0	3%	1228.5	1%				
Ba-131	12.00 d	123.7	28%	133.5	1%	216.0	21%	239.4	2%
		249.3	3%	373.1	13%	486.5	1%	496.2	42%
Ca-47	4.54 d	488.8	6%	807.8	6%	1297.0	77%		
Ce-141	32.55 d	145.4	47%						
Co-60	5.27 d	1173.2	100%	1332.4	100%				
Cr-51	27.70 d	320.0	10%						
Cs-134	2.06 y	475.3	1%	536.2	8%	569.2	15%	604.6	97%
		795.7	85%	801.8	8%	1038.5	1%	1167.8	1%
		1365.1	3%						
Eu-151	13.20 y	121.7	30%	244.6	7%	344.2	27%		
		411.0	2%	778.8	12%	867.3	4%		
		964.0	14%	1085.7	10%	1089.8	1%		
		1112.0	13%	1212.8	1%	1299.1	1%		
		1408.0	20%						
Fe-59	44.56 d	142.6	1%	192.3	2%	1099.2	55%		
		1291.0	52%						
Hf-181	42.45 d	133.0	40%	136.2	6%	136.8	1%	345.8	13%
		476.0	1%	482.0	81%				
Hg-202	46.76 d	276.1	81%						
In-114m	49.51 d	190.2	15%	558.0	3%	725.2	3%		
La-140	40.27 h	328.7	18%	432.5	2%	487.0	42%		
		751.8	4%	815.8	22%	867.8	5%	919.6	2%
		925.2	6%	1596.4	95%	2521.6	3%		
Lu-177	6.71 d	112.9	6%	208.3	11%				
Co-58	70.78 d	810.7	99%	863.7	1%	1674.7	1%		

TABLE B.1. (con.)

Isotope	Half Life	Gamma-Ray Energy (keV) and Percent Yield							
Rb-86	18.82 d	1077.1	8%						
Sb-124	60.21 d	602.7	98%	645.8	7%	709.3	1%	713.8	2%
		722.7	11%	968.2	1%	1045.2	1%	1325.4	1%
		1368.2	2%	1436.0	1%	1691.0	49%		
		2099.0	5%						
Sb-122	2.68 d	564.0	70%	692.7	3%	1256.8	1%		
Sc-45	83.80 d	889.2	100%	1120.5	100%				
Se-74	118.45 d	121.0	17%	135.9	58%	198.5	1%	264.6	58%
		279.4	24%	303.8	1%				
Sm-153	46.80 h	97.4	1%	103.1	28%				
Sr-85	64.85 d	513.9	99%						
Ta-182	115.00 d	100.1	2%	1121.2	35%	1189.0	16%		
		1221.4	27%	1230.9	11%	1257.4	1%		
		1289.1	1%						
Tb-160	72.10 d	197.0	5%	215.6	3%	298.5	27%	309.0	57%
Tb-160	721.09 d	197.0	5%	215.6	4%	298.5	27%	309.5	1%
		392.4	1%	765.2	2%	879.3	30%	962.3	9%
		966.1	24%	1002.8	1%	1115.1	1%		
		1177.9	15%	1199.8	2%	1271.8	7%		
		1312.1	2%						
W-187	23.85 h	72.4	10%	134.2	8%	479.5	21%	511.7	1%
		551.5	4%	625.5	1%	685.8	26%	772.8	3%
Pa-233	26.95 d	300.1	6%	311.8	37%	340.4	4%	398.4	1%
		415.7	1%						
Yb-175	4.19 d	113.7	1%	282.5	2%	396.3	6%		
Zn-65	244.00 d	1115.5	50%						

TABLE B.2. Gamma-Ray Energy Library for Short Lived Isotopes,
Half Lives Less than 24 Hours, Present in Coal.

Isotope	Half Life	Gamma-Ray Energy (keV) and Percent Yield							
Al-28	2.24 M	1778.7	100%						
As-76	26.32 H	559.1	45%	657.0	6%	1212.7	1%	1216.0	3%
		1228.5	1%						
Ba-135m	28.70 H	268.2	16%						
Ba-139	82.90 M	165.8	22%						
Br-80	17.68 M	616.2	7%	639.2	0%				
Ca-49	8.72 M	3084.4	92%	4071.9	7%				
Cl-38	37.29 M	1642.2	31%	2167.6	42%				
Cu-66	5.10 M	1039.2	8%						
Dy-165	2.33 H	94.6	4%	279.8	1%	361.7	1%	633.4	1%
		715.3	1%						
Dy-165M	1.26 M	108.2	3%	361.7	1%	515.5	2%		
Ga-72	14.12 H	600.9	6%	629.9	24%	786.5	3%	810.2	2%
		833.9	96%	894.2	10%	970.6	1%	1050.7	7%
		1230.9	1%	1260.1	1%	1276.8	2%	1464.0	4%
		1596.8	4%	1861.1	5%	2109.5	1%	2201.6	26%
		2491.0	7%	2507.7	13%				
I-128	24.99 M	443.0	16%						
IN-116m ₁	54.12 M	138.3	3%	416.9	32%	818.7	12%	1097.3	56%
		1293.5	85%	1507.4	10%	1752.4	2%	2112.1	15%
K-42	12.36 H	1524.6	19%						
Lu-176M	3.68 H	88.3	9%						
Mg-27	9.46 M	170.7	1%	843.8	73%	1014.3	29%		
Mn-56	2.58 H	846.7	98%	1810.7	27%	2113.0	14%	2522.8	1%
		2657.4	1%						
Na-24	15.03 H	1368.6	100%	2754.1	100%				

TABLE B.2. (con.)

Isotope	Half Life	Gamma-Ray Energy (keV) and Percent Yield							
Nd-149	1.73 H	74.3	1%	97.0	2%	114.3	21%	188.6	2%
		198.9	2%	208.1	3%	211.3	31%	240.2	5%
		245.7	1%	267.7	7%	270.2	12%	326.6	5%
		349.2	2%	423.6	11%	443.6	2%	540.5	9%
		556.6	1%	654.8	8%				
Nd-151	12.44 M	69.2	1%	85.3	3%	89.9	2%	138.8	8%
		171.0	4%	175.0	8%	255.6	17%	300.6	2%
		402.2	2%	423.5	7%	460.6	1%	677.9	3%
		736.4	7%	739.4	22%	797.5	6%	841.1	1%
		914.1	1%	1016.4	3%	1122.1	4%	1180.6	15%
Rb-88	17.78 M	898.0	14%	1382.4	1%	1836.0	21%	2677.9	2%
Sm-155	22.40 M	104.3	69%	141.4	23%	245.7	4%		
Sr-87M	2.81 H	388.3	82%						
Ti-51	5.80 M	319.7	93%	609.4	1%	929.1	7%		
V-52	3.75 M	1332.6	1%	1434.0	100%				
W-187	23.85 H	72.4	10%	134.2	8%	479.5	21%	511.7	1%
		551.5	4%	625.5	1%	685.8	26%	772.8	3%
Zn-71	2.45 M	121.5	2%	390.0	3%	511.6	30%	910.3	7%
		1120.0	2%						
Zr-97	16.90 H	355.4	2%	507.6	5%	602.4	1%	743.4	92%
		1021.2	1%	1147.9	2%				

TYPE ENERGY IN KEV, YIELD IN %

*A

NAME,T1/2,U *AS-76,26.32,H

PEAK,Y *559.1,45.

PEAK,Y *657.03,6.1

PEAK,Y *1212.72,1.3

PEAK,Y *1216.02,3.4

PEAK,Y *1228.52,1.2

PEAK,Y *

General Format

*Isotope Name, half life, unit
of half life

*gamma-ray energy

*A

NAME,T1/2,U *BA-131,12.0,D

PEAK,Y *123.76,28.

PEAK,Y *133.54,1.9

PEAK,Y *216.04,21.7

PEAK,Y *239.45,2.6

PEAK,Y *249.34,3.1

PEAK,Y *373.15,13.1

PEAK,Y *486.52,1.8

PEAK,Y *496.26,42.1

PEAK,Y *

*A

NAME,T1/2,U *CA-47,4.54,J

ERROR 4

NAME,T1/2,U *CA-47,4.54,D

PEAK,Y *488.9,6.78

PEAK,Y *807.9,6.78

PEAK,Y *1297.0,77.

PEAK,Y *

incorrect half life unit

*A

NAME,T1/2,U *CO-60,5.27,5.2719,Y

ERROR 4

NAME,T1/2,U *CO-60,5.2719,Y

PEAK,Y *1332.47,100.

PEAK,Y *1332.47,100.

PEAK,Y *

ERROR 7

*E

error in typing half life value

repetition of a previous entry
does not lead to an editor
error

FIG. B.1. Decwriter Listing for Input of Gamma-Ray Energy Library into the Series 80. The last two entries show common input errors.

USER LIBRARY: ENERGY IN KEV				5 NOV 82	10:45
1) AS-76	26.32 H				
	559.0	45%	657.0	6%	1212.7 1% 1216.0 3% 1228.5 1%
2) BA-131	12.00 D				
	123.7	28%	133.5	1%	216.0 21% 239.4 2% 249.3 3%
	373.1	13%	486.5	1%	496.2 42%
3) CA-47	4.54 D				
	488.8	6%	807.8	6%	1297.0 77%
4) CO-60	5.27 Y				
	1332.4	100%	1332.4	100%	

FIG. B.2. Series 80 Output of the User Library Defined in FIG. B.1.

APPENDIX C: MASCAL-WATFIV PROGRAM TO CALCULATE THE MASS OF ELEMENTS
IN AN UNKNOWN SAMPLE USING INAA RESULTS.

ILLEGIBLE DOCUMENT

**THE FOLLOWING
DOCUMENT(S) IS OF
POOR LEGIBILITY IN
THE ORIGINAL**

**THIS IS THE BEST
COPY AVAILABLE**

```

$JOB          JFH,PAGES=75
COMMON/TIME/UTR(20),STR(20),UTS(20),UTE(20),STS(20)
?,STE(20)
COMMON/RMASS/UPA(20,20),SPA(20),RLAM(20)
COMMON/FTIME/UTSM(20),UTEM(20),STSM(20),STEM(20)
COMMON/FMASS/UMM(20),SHM,UPAM(20),SPAM,FLAM(20)
COMMON/RAT/RMA(20,20),FR(20,20),SIGFR(20,20),SIGMR(20,20)
COMMON/SMA/SM(20),UM(20)
COMMON/SIGMA/SIGSPA(20),SIGUPM(20),SIGSM(20),SIGUPA(20,20),SIGSPM
?,SIGM
COMMON/TITLE/TITNSO(5,20)
DIMENSION PPM(20,20),SIGPPM(20,20)
C*****
C*
C*  MASCAL - MASS CALCULATOR
C*
C*  WRITTEN BY  JACK F. HIGGINBOTHAM  8/82
C*
C*  PURPOSE:
C*    THIS PROGRAM TAKES PEAK AREA, DECAY TIME AND IRRADIATION
C*    TIME DATA FROM INAA RUNS ON SAMPLES AND CALCULATES THE
C*    MASS OF ELEMENTS IN UNKNOWN SAMPLES
C*
C*  VARIABLE DEFINATION:
C*
C*    NISO  - NUMBER OF ISOTOPES OR ELEMENTS TO BE EVALUATED
C*    NCASES - NUMBER OF CASES CONSIDERED
C*    SM    - ISOTOPE MASS IN STANDARD SAMPLE
C*    UM    - ACTUAL MASS OF UNKNOWN SAMPLE
C*
C*****
C  PRINT5
C  5  FORMAT('1',25X,'DATA SET 81'/'1')
C     CALL INPUT(NCASES,NISO)
C     DO 10 J=1,NCASES
C       CALL INIT(NISO,J)
C       DO 20 I=1,NISO
C         CALL RATIO(I,J)
C         CALL RATMA(I,J)
C         PPM(I,J)=RMA(I,J)*SM(I)/UM(J)
C         T=SM(I)/UM(J)
C         T1=T*T*SIGMR(I,J)*SIGMR(I,J)
C         T=RMA(I,J)/UM(J)
C         T2=T*T*SIGSM(I)*SIGSM(I)
C         T=RMA(I,J)*SM(I)/UM(J)/UM(J)
C         T3=T*T*SIGM*SIGM
C         SIGPPM(I,J)=SQRT(T1+T2+T3)
C  20  CONTINUE
C     CALL OUTPUT(NISO,J,PPM,SIGPPM)
C  10  CONTINUE
C     PRINT11
C  11  FORMAT('1')
C     STOP
C     END

```

```

      FUNCTION FEXP(X)
C*****
C*
C*   WRITTEN BY JACK F. HIGGINBOTHAM  8/82
C*
C*   PURPOSE:
C*   TO EVALUATE THE EXPONENTIAL FUNCTION OF THE VALUE X. THE
C*   SUBPROGRAM LIMITS THE UNDERFLOW ASSOCIATED WITH EXPONENTIALS
C*
C*****
      IF(X.LT.-73.) GO TO 10
      FEXP= EXP(X)
      RETURN
10  CONTINUE
C*
C*   UNDERFLOW WILL OCCUR SO SET TO ZERO
C*
      FEXP=0.
      RETURN
      END

```

```

      SUBROUTINE DAYCAL(XMON, DAY, XHR, XMIN, SEC, DATE)
C*****
C*
C*   SUBROUTINE DAYCAL(XMON, DAY, TIME, DATE)
C*
C*   WRITTEN BY: JACK F. HIGGINBOTHAM
C*   USING HP-19C/HP-29C APPLICATIONS BOOK AS A GUIDELINE
C*
C*   PURPOSE:
C*   THIS SUBROUTINE CONVERTS A TIME IN THE FORMAT MONTH, DAY
C*   CLOCK TIME(HRS) INTO A TIME WITH UNITS OF DAYS
C*
C*****
      IF(XMON.EQ.0.) GO TO 30
      IF(XMON.GT.2.) GO TO 10
      MPRIME=XMON+13.
      GO TO 20
10  CONTINUE
      MPRIME=XMON+1.
20  CONTINUE
      IM=30.6601*MPRIME
      GO TO 31
30  CONTINUE
      IM=XMON
31  CONTINUE
      TIME=XHR/24.+XMIN/1440.+SEC/86400.
      DATE=IM+DAY+TIME
      RETURN
      END

```



```

      SUBROUTINE INIT(NISO,J)
      COMMON/RAT/RMA(20,20),FR(20,20),SIGFR(20,20),SIGMR(20,20)
C*****
C*
C*   SUBROUTINE INIT
C*
C*   WRITTEN BY JACK F. HIGGINBOTHAM  8/82
C*
C*   PURPOSE:
C*
C*   THIS SUBROUTINE INITIALIZES VARIABLES TO BE USED IN
C*   COMMON BLOCK STATEMENTS.  VARIABLES ARE DEFINED IN THE
C*   SUBROUTINES
C*
C*****
C*
C**  INITIALIZE
C*
      DO 10 I=1,NISO
      RMA(I,J)=0.0
      FR(I,J)=0.0
      SIGFR(I,J)=0.
      SIGMR(I,J)=0.
10  CONTINUE
      RETURN
      END

```

```

SUBROUTINE INPUT(NCASES,NISO)
COMMON/TIME/UTR(20),STR(20),UTS(20),UTE(20),STS(20),STE(20)
COMMON/FTIME/UTSM(20),UTEM(20),STSM(20),STEM(20)
COMMON/SMA/SM(20),UM(20)
COMMON/RMASS/UPA(20,20),SPA(20),RLAM(20)
COMMON/FMASS/UMM(20),SMM,UPAM(20),SPAM,FLAM(20)
COMMON/SIGMA/SIGSPA(20),SIGUPM(20),SIGSM(20),SIGUPA(20,20),SIGSPM
7,SIGM
COMMON/TITLE/TITNSO(5,20)
DIMENSION TUIRR(20)
CHARACTER U*1

```

```

C*****
C*
C* SUBROUTINE INPUT
C*
C* WRITTEN BY JACK F. HIGGINBOTHAM 8/82
C*
C* PURPOSE:
C*   TO READ IN THE MEASURED DATA FOR CALCULATION OF UNKNOWN MASSES
C*
C* VARIABLE DEFINATION:
C*
C*   NCASES = NUMBER OF CASES CONSIDERED
C*   NISO = NUMBER OF ISOTOPES OR ELEMENTS EVALUATED
C*   FLAM = FLUX MONITOR DECAY CONSTANT
C*   DLAM = DUMMY VALUE FOR FLAM
C*   RLAM = ISOTOPE DECAY CONSTANT
C*   TSIRR= STANDARD SAMPLE IRRADIATION TIME
C*   STR = TIME STANDARD SAMPLE REMOVED FROM IRRADIATION FIELD
C*   STS = STANDARD SAMPLE START TIME FOR COUNT
C*   SCLOCK = STANDARD SAMPLE CLOCK TIME (SEC)
C*   SLIVE = STANDARD SAMPLE LIVE TIME (SEC)
C*   STE = STANDARD SAMPLE END OF COUNT
C*   SFCLCK = STANDARD SAMPLE FLUENCE MONITOR CLOCK TIME (SEC)
C*   STSM = STANDARD SAMPLE START TIME FOR FLUX MONITOR COUNT
C*   STEM = STANDARD SAMPLE END TIME FOR FLUX MONITOR COUNT
C*   TUIRR= UNKNOWN SAMPLE IRRADIATION TIME
C*   UTR = TIME UNKNOWN SAMPLE REMOVED FROM IRRADIATION FIELD
C*   UTS = UNKNOWN SAMPLE START TIME FOR COUNT
C*   UCLOCK = UNKNOWN SAMPLE CLOCK TIME (SEC)
C*   ULIVE = UNKNOWN SAMPLE LIVE TIME (SEC)
C*   UTE = UNKNOWN SAMPLE END OF COUNT
C*   UFCLCK = UNKNOWN SAMPLE FLUENCE MONITOR CLOCK TIME (SEC)
C*   UTSM = UNKNOWN SAMPLE START TIME FOR FLUX MONITOR COUNT
C*   UTEM = UNKNOWN SAMPLE END TIME FOR FLUX MONITOR COUNT
C*   SPAM = STANDARD SAMPLE FLUX MONITOR PEAK AREA
C*   SIGSPM = PERCENT ERROR IN SPAM
C*   SMM = STANDARD SAMPLE FLUX MONITOR MASS
C*   SMASS= ACTUAL MASS OF ENTIRE STANDARD SAMPLE
C*   SPA = STANDARD SAMPLE PEAK AREA
C*   SIGSPA = PERCENT ERROR IN SPA
C*   SCONC= CONCENTRATION OF ISOTOPE IN THE STANDARD SAMPLE(WT%)
C*   SIGCON = PERCENT ERROR IN SCONC
C*   UPAM = UNKNOWN SAMPLE FLUX MONITOR PEAK AREA
C*   SIGUPM = PERCENT ERROR IN UPAM
C*   UMM = UNKNOWN SAMPLE FLUX MONITOR MASS
C*   UM = ACTUAL MASS OF ENTIRE UNKNOWN SAMPLE

```

```

C*          UPA = UNKNOWN SAMPLE PEAK AREA
C*          SIGUM = PERCENT ERROR IN UPA
C*          FR = FLUX RATIO STANDARD TO UNKNOWN SAMPLE
C*          SM = ISOTOPE MASS IN STANDARD SAMPLE
C*          SIGSM = ERROR IN SM
C*          SIGM = ERROR IN MASS VALUE
C*
C*  NOTE: THE SUFFIXES MN, DY, AND TM ADDED TO THE TIME
C*  VARIABLE NAME DESIGNATES THE MONTH, DAY AND TIME COMPONENT
C*  OF THAT VARIABLE. THESE COMPONENTS WILL LATER BY USED TO
C*  DETERMINE TIME IN DAYS WITH THE BEGINNING OF THE IRRADIATION
C*  BEING THE REFERENCE POINT.
C*
C*  ALL ERROR VALUES ARE INPUT IN TERMS OF PERCENT. THE PROGRAM
C*  CORRECTS TO THE APPROPRIATE UNITS.
C*
C*****
C      READ,NCASES,NISO
C      PRINT1,NCASES,NISO
C      1 FORMAT('1'/'-' ,10X,'NCASES=',I4,5X,'NISO=',I4)
C      DO 70 I=1,NISO
C      READ(5,71) (TITNSO(K,I),K=1,5)
C      71 FORMAT(5A4)
C      70 CONTINUE
C*****
C*
C*          READ HALF LIFE VALUES AND CALCULATE DECAY CONSTANTS
C*
C*****
C      READ,THALF,U
C      PRINT2,THALF,U
C      2 FORMAT('0' ,10X,'FLUX MONITOR HALF LIFE =',E15.7,2X,A1)
C      CALL UNIT(THALF,U,DFLAM)
C      PRINT4
C      4 FORMAT('0' ,T7,'ISOTCPE',T40,'HALF'/' ' ,T9,'NO.',T20,
C      7'DESCRIPTION',T40,'LIFE')
C      PRINT6
C      6 FORMAT(' ' ,T7,40(' '))
C      DO 10 I=1,NISO
C      FLAM(I)=DFLAM
C      READ,THALF,U
C      PRINT3,I,(TITNSO(K,I),K=1,5),THALF,U
C      3 FORMAT(' ' ,T9,I2,5X,5A4,F6.2,2X,A1)
C      CALL UNIT(THALF,U,RLAM(I))
C      10 CONTINUE
C      PRINT6
C*****
C*
C*          READ TIME VALUES
C*
C*****
C*
C*          STANDARD SAMPLE
C*
C*****
C      PRINT61
C      61 FORMAT('1' ,15X,'TIME VALUES FOR STANDARD SAMPLE')

```

```

      PRINT 60
60 FORMAT('---',T35,'DAY',T44,'HOUR',T53,'MINUTE',T63,
?'SECOND')
      PRINT62
62 FORMAT('+',T35,'---',T44,'---',T53,6(' '),T63,6(' '))
C*
C*   READ TSIRR
C*
      READ,DY,HR,XMIN,SEC
      PRINT11,'TSIRR ',DY,HR,XMIN,SEC
      CALL DAYCAL(0.,DY,HR,XMIN,SEC,TSIRR)
C*
C*   READ STR
C*
      READ,DY,HR,XMIN,SEC
      PRINT11,'STR ',DY,HR,XMIN,SEC
      CALL DAYCAL(0.,DY,HR,XMIN,SEC,STRR)
C*
C*   READ STS
C*
      READ,DY,HR,XMIN,SEC
      PRINT11,'STS ',DY,HR,XMIN,SEC
      CALL DAYCAL(0.,DY,HR,XMIN,SEC,STSS)
C*
C*   READ STSM
C*
      READ,DY,HR,XMIN,SEC
      PRINT11,'STSM ',DY,HR,XMIN,SEC
      CALL DAYCAL(0.,DY,HR,XMIN,SEC,STSM)
C*
C*   READ SCLOCK AND SLIVE   CALCULATE STE
C*
      READ,SCLOCK,SLIVE
      PRINT13,'STAN','DARD',' CLO','CK T','IME ',SCLOCK
      PRINT13,'STAN','DARD',' LIV','E TI','ME ',SLIVE
      CALL DAYCAL(0.,0.,0.,0.,SCLOCK,XSC)
      STEE=STSS+XSC
      DEADTS=((SCLOCK-SLIVE)/SCLOCK)*100.
      PRINT14,'DEAD',' TIM','E ST','ANDA','RD ',DEADTS
C*
C*   READ SFCLCK   CALCULATE STEMM
C*
      READ,SFCLCK
      PRINT13,'STAN','D. F','LUEN','CE C','LOCK',SFCLCK
      CALL DAYCAL(0.,0.,0.,0.,SFCLCK,XSC)
      STEMM=STSM + XSC
C*
      PRINT5
5 FORMAT('1'/'-',LOX,'TIME VALUES FOR UNKNOWN SAMPLES')
      PRINT60
      PRINT62
C*****
C*
C**   UNKNOWN SAMPLES
C*
C*****
      N=1

```

```

      DD 20 J=1,NCASES
      PRINT67,J
67  FORMAT('0',10X,'CASE NO.=',I2)
      IHIGH=5*N
      IF(J.LE.IHIGH) GO TO 58
      PRINT69
69  FORMAT('1')
      N=N+1
68  CONTINUE

C*
C*  READ TUIRR
      READ,DY,HR,XMIN,SEC
      PRINT11,'TUIRR ',DY,HR,XMIN,SEC
      CALL DAYCAL(0.,DY,HR,XMIN,SEC,TUIRR(J))

C*
C*  READ UTR
      READ,DY,HR,XMIN,SEC
      PRINT11,'UTR ',DY,HR,XMIN,SEC
      CALL DAYCAL(0.,DY,HR,XMIN,SEC,UTR(J))

C*
C*  READ UTS
      READ,DY,HR,XMIN,SEC
      PRINT11,'UTS ',DY,HR,XMIN,SEC
      CALL DAYCAL(0.,DY,HR,XMIN,SEC,UTS(J))

C*
C*  READ UTSM
      READ,DY,HR,XMIN,SEC
      PRINT11,'UTSM ',DY,HR,XMIN,SEC
      CALL DAYCAL(0.,DY,HR,XMIN,SEC,UTSM(J))

C*
C*  READ UCLOCK AND ULIVE  CALCULATE UTE(J)
C*
      READ,UCLOCK,ULIVE
      PRINT13,'UNKN','OWN ','CLOCK','K TI','ME ',UCLOCK
      PRINT13,'UNKN','OWN ','LIVE',' TIM','E ',ULIVE
      CALL DAYCAL(0.,0.,0.,0.,UCLOCK,XSC)
      UTE(J)=UTS(J)+XSC
      DEADTU=(((UCLOCK-ULIVE)/UCLOCK))*100.
      PRINT14,'DEAD',' TIM','E U','NKND','WN ',DEADTU

C*
C*  READ UFCLCK  CALCULATE UTEM(J)
C*
      READ,UFCLCK
      PRINT13,'UNKN',' FL','UENC','E CL','OCK ',UFCLCK
      CALL DAYCAL(0.,0.,0.,0.,UFCLCK,XSC)
      UTEM(J)=UTSM(J)+XSC

C*
11  FORMAT(' ',10X,A8,T36,F3.0,6X,F3.0,7X,F3.0,7X,F3.0)
13  FORMAT(' ',10X,5A4,T63,F7.0)
14  FORMAT(' ',10X,5A4,T63,F5.1,'%')

C*
      REF=TUIRR(J)
      UTR(J)=UTR(J)+REF
      UTS(J)=UTS(J)+REF
      UTE(J)=UTE(J)+REF
      UTSM(J)=UTSM(J)+REF
      UTEM(J)=UTEM(J)+REF

```

```

      REF=TSIRR
      STR(J)=STRR+REF
      STS(J)=STSS+REF
      STE(J)=STEE+REF
      STSM(J)=STSM+REF
      STEM(J)=STEM+REF
20  CONTINUE
C*****
C*
C*      READ STANDARD MASS, ISOTOPE CONCENTRATIONS, AND PEAK AREAS
C*
C*****
      SIGM=1.26E-4
      READ, SPAM,SS,SMM,SMAS
      PRINT21,SPAM,SMM,SS,SMAS
21  FORMAT('1','-',T23,'* * * STANDARD SAMPLE INPUT * * */
      ?'-',T17,'WIRE PEAK AREA =',F10.0,T51,'WIRE MASS =',F10.4/
      ?'0',T17,'WIRE PEAK ERROR=',F10.2,T51,'SAMPLE MASS=',F10.4)
      SIGSPM=SPAM*SS/100.
      PRINT23
23  FORMAT('0',T13,'I',T20,'SPA(I)',T34,'SIGSPA',T47,'SCONC',T58,
      ?'SIGCON'/'+',T13,'_',T20,6(' '),T34,6(' '),T47,5(' '),T58,6(' '))
      DO 50 I=1,NISO
      READ, SPA(I),SP,SCONC,SC
      SIGSPA(I)=SPA(I)*SP/100.
      SIGCON=SCONC*SC/100.
      IF(SCONC.GT.0.01)GO TO 75
      SPC=SCONC*1.E6
      PRINT22,I,SPA(I),SP,SPC,SC
22  FORMAT('0',10X,I2,3X,F10.0,7X,F5.2,3X,F10.2,' PPM',3X,F5.2)
      GO TO 76
75  CONTINUE
      SPC=SCONC*100.
      PRINT77,I,SPA(I),SP,SPC,SC
77  FORMAT('0',10X,I2,3X,F10.0,7X,F5.2,3X,F10.2,' WT %',2X,F5.2)
76  CONTINUE
      SM(I)=SCONC*SMAS
      SIGSM(I)=SQRT((SIGM)**2.*(SCONC)**2.+SIGCON**2.*(SMAS)**2.)
50  CONTINUE
C*
C**
C*
      DO 40 J=1,NCASES
      PRINT41
41  FORMAT('1',10X,' J',7X,'UPAM(J)',8X,'SIGUPM(J)',8X,'UMM(J)',8X,'UM
      ?(J)')
      READ, UPAM(J),SU,UMM(J),UM(J)
      PRINT42,J,UPAM(J),SU,UMM(J),UM(J)
42  FORMAT(' ',10X,I2,E15.5,8X,F5.2,5X,E15.5,E13.5/'-')
      SIGUPM(J)=UPAM(J)*SU/100.
      DO 30 I=1,NISO
      READ,UPA(I,J),SU
      PRINT32,I,J,UPA(I,J),SU
32  FORMAT(' ',1CX,'UPA(',I2,',',I2,')= ',1X,F10.0,8X,'SIGUPA =',1X,F6
      ?2)
      SIGUPA(I,J)=UPA(I,J)*SU/100.
30  CONTINUE

```

```

      40 CONTINUE
      RETURN
      END
      SUBROUTINE UNIT(T,U,X)
C*****
C*
C*   PURPOSE:  TO CONVERT THE HALF LIFE UNITS TO DAYS THEN TO
C*             CALCULATE THE DECAY CONSTANT WITH UNITS OF
C*             INVERSE DAYS.
C*
C*****
      CHARACTER U*1
      IF(U.EQ.'S') GO TO 10
      IF(U.EQ.'M') GO TO 20
      IF(U.EQ.'H') GO TO 30
      IF(U.EQ.'D') GO TO 40
      IF(U.NE.'Y') GO TO 50
C*   UNITS ARE YEARS
      Y=T*365.
      X=ALOG(2.)/Y
      RETURN
      10 CONTINUE
C*   UNITS ARE SECONDS
      Y=T/86400.
      X=ALOG(2.)/Y
      RETURN
      20 CONTINUE
C*   UNITS ARE MINUTES
      Y=T/1440.
      X=ALOG(2.)/Y
      RETURN
      30 CONTINUE
C*   UNITS ARE HOURS
      Y=T/24.
      X=ALOG(2.)/Y
      RETURN
      40 CONTINUE
C*   UNITS ARE DAYS
      X=ALOG(2.)/T
      RETURN
      50 CONTINUE
      PRINT*1
      51 FORMAT('1','INPUT ERROR IN HALF LIFE UNITS')
      RETURN
      END

```

```

SUBROUTINE OUTPUT(NISO,J,PPM,SIGPPM)
COMMON/SIGMA/SIGSPA(20),SIGUPM(20),SIGSM(20),SIGUPA(20,20),SIGSPM
7,SIGM
COMMON/TITLE/TITNSQ(5,20)
DIMENSION SIGPPM(20,20),PPM(20,20),SIGC(20),CONC(20),NIT(20)
DIMENSION SIGPER(20)
INTEGER A/'PPM '/,B/'WT %'/
C*****
C*
C* SUBROUTINE OUTPUT
C*
C* WRITTEN BY JACK F. HIGGINBOTHAM 10/82
C*
C* PURPOSE:
C* THIS SUBROUTINE PRINTS THE MASS CONCENTRATIONS
C*
C*****
DO 10 I=1,NISO
IF(PPM(I,J).GT.0.01) GO TO 20
C*
C** UNITS ARE PPM
C*
CONC(I)=PPM(I,J)*1.E6
SIGC(I)=SIGPPM(I,J)*1.E6
IF(CONC(I).EQ.0.) GO TO 70
SIGPER(I)=(1.-((CONC(I)-SIGC(I))/CONC(I)))*100.
GO TO 71
70 CONTINUE
SIGPER(I)=0.
71 CONTINUE
NIT(I)=A
GO TO 10
20 CONTINUE
C*
C** UNITS ARE WEIGHT PERCENT
C*
CONC(I)=PPM(I,J)*100.
SIGC(I)=SIGPPM(I,J)*100.
IF(CONC(I).EQ.0.) GO TO 72
SIGPER(I)=(1.-((CONC(I)-SIGC(I))/CONC(I)))*100.
GO TO 73
72 CONTINUE
SIGPER(I)=0.
73 CONTINUE
NIT(I)=B
10 CONTINUE
C*
C**
C*
PRINT1,J
1 FORMAT('1'/'-',6X,'TABLE ISOTOPE CONCENTRATION ',
?'FOR CASE NUMBER',I3)
PRINT2
PRINT2
2 FORMAT(' ',6X,51(' '))
PRINT3
3 FORMAT(' ',8X,'ISOTOPE',T21,'CONCENTRATION',

```



```

710X,'EXPECTED ERROR')
PRINT2
DO 40 I=1,NISO
IF (NIT(I).EQ.A) GO TC41
PRINT4,(TITNSO(K,I),K=1,2),CONC(I),SIGC(I),NIT(I),SIGPER(I)
4 FORMAT('0',T10,2A4,T20,F9.3,T32,'+/-',2X,F7.2,2X,A4,2X,'(',F4.1,
?'%')')
GO TO 40
41 CONTINUE
PRINT5,(TITNSO(K,I),K=1,2),CONC(I),SIGC(I),NIT(I),SIGPER(I)
5 FORMAT('0',T10,2A4,T20,F9.2,T32,'+/-',2X,F7.2,2X,A4,2X,'(',F4.1,
?'%')')
40 CONTINUE
PRINT2
RETURN
END

```

```

SUBROUTINE RATIO(I,J)
COMMON/RMASS/UPA(20,20),SPA(20),RLAM(20)
COMMON/TIME/UTR(20),STR(20),UTS(20),UTE(20),STS(20),STE(20)
COMMON/FTIME/UTSM(20),UTEM(20),STSM(20),STEM(20)
COMMON/FMASS/UMM(20),SMM,UPAM(20),SPAM,FLAM(20)
COMMON/RAT/RMA(20,20),FR(20,20),SIGFR(20,20),SIGMR(20,20)
COMMON/SIGMA/SIGSPA(20),SIGUPM(20),SIGSM(20),SIGUPA(20,20),SIGSPM
?,SIGM
C*****
C*
C* SUBROUTINE RATIO(I,J)
C*
C* WRITTEN BY JACK F. HIGGINBOTHAM 8/82
C*
C* PURPOSE:
C* TO EVALUATE THE FLUX RATIO WHEN DETERMINING THE MASS OF AN
C* ELEMENT IN AN UNKNOWN SAMPLE
C*
C* VARIABLE DEFINATION
C*
C* UTR = UNKNOWN SAMPLE IRRADIATION TIME
C* UTSM = UNKNOWN SMAPLE START TIME FOR FLUX MONITOR COUNT
C* UTEM = UNKNOWN SAMPLE END TIME FOR FLUX MONITOR COUNT
C* STR = STANDARD SAMPLE IRRADIATION TIME
C* STSM = STANDARD SAMPLE START TIME FOR FLUX MONITOR COUNT
C* STEM = STANDARE SAMPLE END TIME FOR FLUX MONITOR COUNT
C* UMM = UNKNOWN SAMPLE FLUX MONITOR MASS
C* SMM = STANDARD SAMPLE FLUX MONITOR MASS
C* UPAM = UNKNOWN SAMPLE FLUX MONITOR PEAK AREA
C* SPAM = STANDARD SAMPLE FLUX MONITOR PEAK AREA
C* FR = FLUX RATIO STANDARD TO UNKNOWN SAMPLE
C* FLAM = FLUX MONITOR DECAY CONSTANT
C*
C*****
C*
C** EVALUATE NUMERATOR
C*
C* A=FLAM(I)*(UTR(J)-UTSM(J))
C* B=FLAM(I)*(UTR(J)-UTEM(J))
C* C=FEXP(A)-FEXP(B)
C* C1=C
C* D=-FLAM(I)*UTR(J)
C* E=1.-FEXP(D)
C* E1=E
C* F=UMM(J)*SPAM*E*C
C*
C** EVALUATE DENOMINATOR
C*
C* A=FLAM(I)*(STR(J)-STSM(J))
C* B=FLAM(I)*(STR(J)-STEM(J))
C* C=FEXP(A)-FEXP(B)
C* D=-FLAM(I)*STR(J)
C* E=1.-FEXP(D)
C*
C*
C** SOLN
C*
C* G=SMM*UPAM(J)*E*C

```

```

      FR(I,J)=F / G
C*
C** ERROR CALCULATION
C*
      TRW=E1/E*(C1/C)
      T=TRW*SPAM/SMM/UPAM(J)
      T1=T*T*(SIGM)**2.
      T=TRW*UMM(J)/SMM/UPAM(J)
      T2=T*T*SIGSPM*SIGSPM
      T=TRW*SPAM*UMM(J)/UPAM(J)/SMM/SMM
      T3=T*T*(SIGM)**2.
      T=TRW*SPAM*UMM(J)/SMM/UPAM(J)/UPAM(J)
      T4=T*T*SIGUPM(J)*SIGUPM(J)
      SIGFR(I,J)=SQRT(T1+T2+T3+T4)
      RETURN
      END

```

```

SUBROUTINE RATMA(I,J)
COMMON/TIME/UTR(20),STR(20),UTS(20),UTE(20),STS(20)
?,STE(20)
COMMON/RMASS/UPA(20,20),SPA(20),RLAM(20)
COMMON/RAT/RMA(20,20),FR(20,20),SIGFR(20,20),SIGMR(20,20)
COMMON/SIGMA/SIGSPA(20),SIGUPM(20),SIGSM(20),SIGUPA(20,20),
?SIGSMP,SIGM
C*****
C*
C* SUBROUTINE RATMA(I,J)
C*
C* WRITTEN BY JACK F. HIGGINBOTHAM
C*
C* PURPOSE:
C*   TO CALCULATE THE MASS RATIO
C*
C* VARIABLE DEFINATION:
C*
C*   UTR = UNKNOWN SAMPLE IRRADIATION TIME
C*   UTS = UNKNOWN SAMPLE START TIME FOR COUNT
C*   UTE = UNKNOWN SAMPLE AEND TIME FOR COUNT
C*   STR = STANDARD SAMPLE IRRADIATION TIME
C*   STS = STANDARD SAMPLE START TIME FOR COUNT
C*   STE = STANDARD SAMPLE END TIME FOR COUNT
C*   UPA = UNKNOWN SAMPLE PEAK AREA
C*   SPA = STANDARD SAMPLE PEAK AREA
C*   RLAM = DECAY CONSTANT FOR ACTIVATED ISOTOPE
C*
C*****
C*
C** EVALUATE NUMERATOR
C*
C*   A=RLAM(I)*(STR(J)-STS(J))
C*   B=RLAM(I)*(STR(J)-STE(J))
C*   C=FEXP(A)-FEXP(B)
C*   C1=C
C*   D=-RLAM(I)*STR(J)
C*   E=1.-FEXP(D)
C*   E1=E
C*   F=UPA(I,J)*E*C
C*
C** EVALUATE THE DENOMINATOR
C*
C*   A=RLAM(I)*(UTR(J)-UTS(J))
C*   B=RLAM(I)*(UTR(J)-UTE(J))
C*   C=FEXP(A)-FEXP(B)
C*   D=-RLAM(I)*UTR(J)
C*   E=1.-FEXP(D)
C*   G=SPA(I)*E*C
C*
C** CALCULATE MASS RATIO
C*
C*   RMA(I,J)=F / G *FR(I,J)
C*   TR=E1/E*C1/C
C*   T=FR(I,J)*TR/SPA(I)
C*   T1=T*T*(SIGUPA(I,J))**2.
C*   T=UPA(I,J)*FR(I,J)*TR/SPA(I)/SPA(I)

```

```
T2=T*T*(SIGSPA(I))**2.  
T=UPA(I,J)*TR/SPA(I)  
T3=T*T*(SIGFR(I,J))**2.  
SIGMR(I,J)=SQRT(T1+T2+T3)  
RETURN  
END
```

APPENDIX D: EXAMPLE ANALYSIS

The pages that follow represent a set of five coals that were analyzed. As the procedure of Section 4.2 was followed, the necessary values were recorded on the data forms. The following data forms are a listing of the input cards used by the MASCAL program to calculate the elemental concentrations. The program was run and the output listing is the last section of this chapter.

The example problem can be used as a benchmark to verify the experimenter's ability to run the computer analysis.

DATA RECORD
(FORMAT FOR INPUT TO MASCAL)

DATA SET NO. 1

NCASES, NISO 5, 13

DATA SET NO. 2

ISOTOPE NO.	DESCRIPTION
1	<u>Sm-153 103.2 keV</u>
2	<u>Ce-141 145.4 keV</u>
3	<u>Se-75 264.6 keV</u>
4	<u>Cr-51 320.0 keV</u>
5	<u>Hf-181 482.0 keV</u>
6	<u>Ba-131 496.2 keV</u>
7	<u>Cs-134 795.8 keV</u>
8	<u>Tb-160 879.3 keV</u>
9	<u>Sc-46 889.2 keV</u>
10	<u>Rb-86 1077.2 keV</u>
11	<u>Co-60 1173.2 keV</u>
12	<u>Ta-182 1221.4 keV</u>
13	<u>Sb-124 1691.0 keV</u>
14	_____
15	_____
16	_____
17	_____
18	_____
19	_____
20	_____

DATA RECORD

(FORMAT FOR INPUT TO MASCAL)

DATA SET NO. 3ISOTOPE
NO.
FLUENCE
MONITORHALF LIFE

	<u>44.56</u>	, ' D '
1	<u>46.80</u>	, ' H '
2	<u>32.55</u>	, ' D '
3	<u>118.45</u>	, ' D '
4	<u>27.70</u>	, ' D '
5	<u>42.45</u>	, ' D '
6	<u>12.00</u>	, ' D '
7	<u>2.06</u>	, ' Y '
8	<u>72.10</u>	, ' D '
9	<u>83.80</u>	, ' D '
10	<u>18.82</u>	, ' D '
11	<u>5.27</u>	, ' Y '
12	<u>115.0</u>	, ' D '
13	<u>60.20</u>	, ' D '
14	<u> </u>	, ' ' '
15	<u> </u>	, ' ' '
16	<u> </u>	, ' ' '
17	<u> </u>	, ' ' '
18	<u> </u>	, ' ' '
19	<u> </u>	, ' ' '
20	<u> </u>	, ' ' '

DATA SET NO. 4 TIME VALUES FOR THE STANDARD

	DAY	HOUR	MINUTE	SECOND
TSIRR (IRRADIATION TIME)	<u>0</u>	, <u>8</u>	, <u>0</u>	, <u>0</u>
STR (TIME REMOVED FROM RX)	<u>0</u>	, <u>0</u>	, <u>0</u>	, <u>0</u>
STS (START TIME FOR COUNT)	<u>17</u>	, <u>6</u>	, <u>58</u>	, <u>53</u>
STSM (START OF FLUENCE MON.)	<u>15</u>	, <u>1</u>	, <u>13</u>	, <u>35</u>
CLOCK AND LIVE TIME FOR COUNT	<u>57857</u> , <u>57600</u>			
CLOCK TIME FOR FLUENCE COUNT	<u>2413</u>			

DATA SET NO. 5 TIME VALUES FOR THE UNKNOWN

CASE NO. 1	DAY	HOUR	MINUTE	SECOND
TUIRR (IRRADIATION TIME)	<u>0</u>	, <u>8</u>	, <u>0</u>	, <u>0</u>
UTR (TIME REMOVED FROM RX)	<u>0</u>	, <u>0</u>	, <u>0</u>	, <u>0</u>
UTS (START TIME FOR COUNT)	<u>18</u>	, <u>4</u>	, <u>8</u>	, <u>20</u>
UTSM (START OF FLUENCE MON.)	<u>15</u>	, <u>3</u>	, <u>53</u>	, <u>7</u>
CLOCK AND LIVE TIME FOR COUNT	<u>57695</u> , <u>57600</u>			
CLOCK TIME FOR FLUENCE COUNT	<u>1303</u>			

CASE NO. 2	DAY	HOUR	MINUTE	SECOND
TUIRR (IRRADIATION TIME)	<u>0</u>	, <u>8</u>	, <u>0</u>	, <u>0</u>
UTR (TIME REMOVED FROM RX)	<u>0</u>	, <u>0</u>	, <u>0</u>	, <u>0</u>
UTS (START TIME FOR COUNT)	<u>18</u>	, <u>21</u>	, <u>1</u>	, <u>32</u>
UTSM (START OF FLUENCE MON.)	<u>15</u>	, <u>4</u>	, <u>29</u>	, <u>16</u>
CLOCK AND LIVE TIME FOR COUNT	<u>577.14</u> , <u>57600</u>			
CLOCK TIME FOR FLUENCE COUNT	<u>1350</u>			

DATA SET NO. 5 TIME VALUES FOR THE UNKNOWN

CASE NO. <u>3</u>	DAY	HOUR	MINUTE	SECOND
TUIRR (IRRADIATION TIME)	<u>0</u>	, <u>8</u>	, <u>0</u>	, <u>0</u>
UTR (TIME REMOVED FROM RX)	<u>0</u>	, <u>0</u>	, <u>0</u>	, <u>0</u>
UTS (START TIME FOR COUNT)	<u>20</u>	, <u>1</u>	, <u>16</u>	, <u>40</u>
UTSM (START OF FLUENCE MON.)	<u>15</u>	, <u>5</u>	, <u>2</u>	, <u>51</u>
CLOCK AND LIVE TIME FOR COUNT	<u>57909</u> , <u>57600</u>			
CLOCK TIME FOR FLUENCE COUNT	<u>1320</u>			

CASE NO. <u>4</u>	DAY	HOUR	MINUTE	SECOND
TUIRR (IRRADIATION TIME)	<u>0</u>	, <u>8</u>	, <u>0</u>	, <u>0</u>
UTR (TIME REMOVED FROM RX)	<u>0</u>	, <u>0</u>	, <u>0</u>	, <u>0</u>
UTS (START TIME FOR COUNT)	<u>20</u>	, <u>18</u>	, <u>26</u>	, <u>42</u>
UTSM (START OF FLUENCE MON.)	<u>15</u>	, <u>5</u>	, <u>26</u>	, <u>20</u>
CLOCK AND LIVE TIME FOR COUNT	<u>57671</u> , <u>57600</u>			
CLOCK TIME FOR FLUENCE COUNT	<u>1604</u>			

CASE NO.	DAY	HOUR	MINUTE	SECOND
TUIRR (IRRADIATION TIME)	<u>0</u>	, <u>8</u>	, <u>0</u>	, <u>0</u>
UTR (TIME REMOVED FROM RX)	<u>0</u>	, <u>0</u>	, <u>0</u>	, <u>0</u>
UTS (START TIME FOR COUNT)	<u>21</u>	, <u>16</u>	, <u>23</u>	, <u>44</u>
UTSM (START OF FLUENCE MON.)	<u>15</u>	, <u>16</u>	, <u>23</u>	, <u>52</u>
CLOCK AND LIVE TIME FOR COUNT	<u>57694</u> , <u>57600</u>			
CLOCK TIME FOR FLUENCE COUNT	<u>1683</u>			

DATA SET NO. 6

Standard Sample	Wire Peak		
Wire Peak Area	Error	Wire Mass	Sample Mass
<u>20008</u> ,	<u>2.20</u> ,	<u>0.0148</u> ,	<u>0.6023</u>

DATA SET NO. 7

ISOTOPE NO.	PEAK AREA	PEAK AREA ERROR (%)	FRACTIONAL CONCENTRATION	ERROR IN FRACTIONAL CONCENTRATION (%)
1	<u>23235</u>	<u>6.2</u>	<u>2.8E-6</u>	<u>10.71</u>
2	<u>68186</u>	<u>2.2</u>	<u>32.0E-5</u>	<u>12.50</u>
3	<u>5514</u>	<u>25.4</u>	<u>2.6E-6</u>	<u>11.54</u>
4	<u>32599</u>	<u>3.6</u>	<u>34.0E-5</u>	<u>5.88</u>
5	<u>10829</u>	<u>8.9</u>	<u>1.55E-6</u>	<u>5.16</u>
6	<u>3903</u>	<u>24.0</u>	<u>122.0E-4</u>	<u>9.02</u>
7	<u>10421</u>	<u>8.4</u>	<u>2.0E-6</u>	<u>15.00</u>
8	<u>2401</u>	<u>34.2</u>	<u>0.32E-7</u>	<u>15.63</u>
9	<u>349792</u>	<u>0.4</u>	<u>6.80E-6</u>	<u>8.82</u>
10	<u>4205</u>	<u>11.6</u>	<u>29.0E-5</u>	<u>3.45</u>
11	<u>16883</u>	<u>2.7</u>	<u>6.5E-6</u>	<u>3.08</u>
12	<u>1665</u>	<u>13.5</u>	<u>0.40E-7</u>	<u>7.5</u>
13	<u>663</u>	<u>12.1</u>	<u>0.60E-7</u>	<u>15.0</u>
14	_____	_____	_____	_____
15	_____	_____	_____	_____
16	_____	_____	_____	_____
17	_____	_____	_____	_____
18	_____	_____	_____	_____
19	_____	_____	_____	_____
20	_____	_____	_____	_____

 DATA SET NO. 8

UNKNOWN SAMPLE WIRE PEAK
 WIRE PEAK AREA ERROR WIRE MASS SAMPLE MASS
10009, 3.1, 0.0139, 0.6942

DATA SET NO. 9

ISOTOPE NO.	PEAK AREA	PEAK ERROR (%)
1	<u>8920</u>	<u>10.6</u>
2	<u>37571</u>	<u>2.6</u>
3	<u>1910</u>	<u>44.2</u>
4	<u>5700</u>	<u>12.0</u>
5	<u>11571</u>	<u>4.9</u>
6	<u>6873</u>	<u>7.9</u>
7	<u>2135</u>	<u>22.7</u>
8	<u>1367</u>	<u>32.9</u>
9	<u>99480</u>	<u>0.8</u>
10	<u>987</u>	<u>27.7</u>
11	<u>3337</u>	<u>8.5</u>
12	<u>865</u>	<u>17.7</u>
13	<u>234</u>	<u>22.5</u>
14	<u> </u>	<u> </u>
15	<u> </u>	<u> </u>
16	<u> </u>	<u> </u>
17	<u> </u>	<u> </u>
18	<u> </u>	<u> </u>
19	<u> </u>	<u> </u>
20	<u> </u>	<u> </u>

 DATA SET NO. 8

UNKNOWN SAMPLE WIRE PEAK
 WIRE PEAK AREA ERROR WIRE MASS SAMPLE MASS
10014, 3.0, 0.0140, 0.5983

DATA SET NO. 9

ISOTOPE NO.	PEAK AREA	PEAK ERROR (%)
1	<u>4841</u>	<u>20.3</u>
2	<u>16558</u>	<u>6.0</u>
3	<u>3969</u>	<u>22.9</u>
4	<u>14333</u>	<u>5.3</u>
5	<u>2987</u>	<u>20.7</u>
6	<u>1195</u>	<u>50.5</u>
7	<u>3924</u>	<u>14.5</u>
8	<u>976</u>	<u>55.2</u>
9	<u>113313</u>	<u>0.8</u>
10	<u>1339</u>	<u>26.7</u>
11	<u>5803</u>	<u>5.9</u>
12	<u>450</u>	<u>38.1</u>
13	<u>546</u>	<u>12.0</u>
14	<u> </u>	<u> </u>
15	<u> </u>	<u> </u>
16	<u> </u>	<u> </u>
17	<u> </u>	<u> </u>
18	<u> </u>	<u> </u>
19	<u> </u>	<u> </u>
20	<u> </u>	<u> </u>

 DATA SET NO. 8

UNKNOWN SAMPLE WIRE PEAK
 WIRE PEAK AREA ERROR WIRE MASS SAMPLE MASS
9998, 3.1, 0.0137, 0.5497

DATA SET NO. 9

ISOTOPE NO.	PEAK AREA	PEAK ERROR (%)
1	<u>8960</u>	<u>17.2</u>
2	<u>47210</u>	<u>3.4</u>
3	<u>8152</u>	<u>18.5</u>
4	<u>24510</u>	<u>5.1</u>
5	<u>8608</u>	<u>11.9</u>
6	<u>4196</u>	<u>24.1</u>
7	<u>11293</u>	<u>8.9</u>
8	<u>2022</u>	<u>47.9</u>
9	<u>222443</u>	<u>0.6</u>
10	<u>4093</u>	<u>15.9</u>
11	<u>28645</u>	<u>2.0</u>
12	<u>1331</u>	<u>20.7</u>
13	<u>5025</u>	<u>2.8</u>
14	<u> </u>	<u> </u>
15	<u> </u>	<u> </u>
16	<u> </u>	<u> </u>
17	<u> </u>	<u> </u>
18	<u> </u>	<u> </u>
19	<u> </u>	<u> </u>
20	<u> </u>	<u> </u>

 DATA SET NO. 8

UNKNOWN SAMPLE	WIRE PEAK		
WIRE PEAK AREA	ERROR	WIRE MASS	SAMPLE MASS
<u>9992</u> ,	<u>3.1</u> ,	<u>0.0132</u> ,	<u>0.7191</u>

DATA SET NO. 9

ISOTOPE NO.	PEAK AREA	PEAK ERROR (%)
1	<u>3051</u>	<u>25.</u>
2	<u>20604</u>	<u>3.9</u>
3	<u>1137</u>	<u>63.9</u>
4	<u>4066</u>	<u>14.6</u>
5	<u>3572</u>	<u>13.6</u>
6	<u>6778</u>	<u>7.1</u>
7	<u>0.</u>	<u>0.</u>
8	<u>906.</u>	<u>43.5</u>
9	<u>80961</u>	<u>0.8</u>
10	<u>0.</u>	<u>0.</u>
11	<u>3863</u>	<u>6.5</u>
12	<u>0.</u>	<u>0.</u>
13	<u>198.</u>	<u>22.5</u>
14	<u> </u>	<u> </u>
15	<u> </u>	<u> </u>
16	<u> </u>	<u> </u>
17	<u> </u>	<u> </u>
18	<u> </u>	<u> </u>
19	<u> </u>	<u> </u>
20	<u> </u>	<u> </u>

 DATA SET NO. 8

UNKNOWN SAMPLE WIRE PEAK
 WIRE PEAK AREA ERROR WIRE MASS SAMPLE MASS
10008 , 3.2 , 0.0133 , 0.5735

DATA SET NO. 9

ISOTOPE NO.	PEAK AREA	PEAK ERROR (%)
1	<u>3824</u>	<u>23.1</u>
2	<u>36121</u>	<u>2.6</u>
3	<u>1388</u>	<u>59.3</u>
4	<u>7115</u>	<u>9.5</u>
5	<u>2856</u>	<u>19.1</u>
6	<u>4809</u>	<u>11.1</u>
7	<u>3838</u>	<u>12.8</u>
8	<u>1391</u>	<u>33.2</u>
9	<u>88440</u>	<u>0.8</u>
10	<u>794</u>	<u>38.8</u>
11	<u>4770</u>	<u>6.3</u>
12	<u>381</u>	<u>42.8</u>
13	<u>295</u>	<u>17.3</u>
14	<u> </u>	<u> </u>
15	<u> </u>	<u> </u>
16	<u> </u>	<u> </u>
17	<u> </u>	<u> </u>
18	<u> </u>	<u> </u>
19	<u> </u>	<u> </u>
20	<u> </u>	<u> </u>

Listing of Input for MASCAL Run of Example Problem

```

5,13
SM-153 103.2 KEV
CE-141 145.4 KEV
SE-75 264.6 KEV
CR-51 320.0 KEV
HF-181 482.0 KEV
BA-131 496.2 KEV
CS-134 795.8 KEV
TB-160 879.3 KEV
SC-46 889.2 KEV
RB-86 1077.2 KEV
CO-60 1173.2 KEV
TA-182 1221.4 KEV
SB-124 1691.0 KEV
44.56, 'D'
46.8, 'H'
32.55, 'D'
118.45, 'D'
27.70, 'D'
42.45, 'D'
12.0, 'D'
2.06, 'Y'
72.1, 'D'
83.80, 'D'
18.82, 'D'
5.272, 'Y'
115.0, 'D'
60.20, 'D'
0,8,0,0
0,0,0,0
17,6,53,53
15,1,13,35
57857,57600
2413
0,8,0,0
0,0,0,0
18,4,8,20
15,3,53,7
57695,57600
1303
0,8,0,0
0,0,0,0
18,21,1,32
15,4,29,16
57714,57600
1350
0,8,0,0
0,0,0,0
20,1,16,40

```

15,5,2,51
57909,57600
1320
0,8,0,0
0,0,0,0
20,18,26,42
15,5,26,20
57671,57600
1604
0,8,0,0
0,0,0,0
21,16,23,44
15,16,23,52
57694,57600
1683
20008,2.2,0.0148,.6023
23235,6.2,2.8E-6,10.71
68186,2.2,3.2E-5,12.5
5514,25.4,2.6E-6,11.54
32599,3.6,3.4E-5,5.88
10829,8.9,1.55E-6,5.16
3903,24,1.22E-4,9.02
10421,8.4,2.0E-6,15.
2401,34.2,3.2E-7,15.63
349792,0.4,6.8E-6,8.82
4205,11.6,2.9E-5,3.45
16883,2.7,6.5E-6,3.08
1665,13.5,4.E-7,7.5
663,12.1,6.E-7,15
10009,3.1,.0139,.6942
8920,10.6
37571,2.6
1910,44.2
5700,12.
11571,4.9
6873,7.9
2135,22.7
1367,32.9
99480,.8
987,27.7
3337,8.5
865,17.7
234,22.5
10014,3,.0140,.5983
4841,20.3
16558,6.
3969,22.9
14333,5.3
2987,20.7
1195,50.5
3924,14.5

976,55.2
113313,0.8
1339,26.7
5803,5.9
450,38.1
546,12.
9998,3.1,.0137,.5497
8960,17.2
47210,3.4
8152,18.5
24510,5.1
8608,11.9
4196,24.1
11293,8.9
2022,47.9
222443,0.6
4093,15.9
28645,2.
1331,20.7
5025,2.8
9992,3.1,.0132,.7191
3051,25.
20604,3.9
1137,63.9
4066,14.6
3572,13.6
6778,7.1
0,0
906,43.5
80961,0.8
0,0
3863,6.5
0,0
198,22.5
10008,3.2,.0133,.5735
3824,23.1
36121,2.6
1388,59.3
7115,9.5
2856,19.1
4809,11.1
3838,12.8
1391,33.2
88440,0.8
794,38.8
4770,6.3
381,42.8
295,17.3

**THIS BOOK IS OF
POOR LEGIBILITY
DUE TO LIGHT
PRINTING
THROUGH OUT IT'S
ENTIRETY.**

**THIS IS AS
RECEIVED FROM
THE CUSTOMER.**

MASCAL Output for Example Problem

NCASES= 5 NISO= 13

FLUX MONITOR HALF LIFE = 0.4456000E 02 D

ISOTOPE NO.	DESCRIPTION		HALF LIFE	
1	SM-153	103.2 KEV	46.80	H
2	CE-141	145.4 KEV	32.55	D
3	SE-75	264.6 KEV	118.45	D
4	CR-51	320.0 KEV	27.70	D
5	HF-181	482.0 KEV	42.45	D
6	BA-131	496.2 KEV	12.00	D
7	CS-134	795.8 KEV	2.06	Y
8	TB-160	879.3 KEV	72.10	D
9	SC-46	889.2 KEV	83.80	D
10	RB-86	1077.2 KEV	18.82	D
11	CO-60	1173.2 KEV	5.27	Y
12	TA-182	1221.4 KEV	115.00	D
13	SB-124	1691.0 KEV	60.20	D

TIME VALUES FOR STANDARD SAMPLE

	DAY	HOUR	MINUTE	SECOND
TSIRR	0.	8.	0.	0.
STR	0.	0.	0.	0.
STS	17.	6.	58.	53.
STSM	15.	1.	13.	35.
STANDARD CLOCK TIME				57857.
STANDARD LIVE TIME				57600.
DEAD TIME STANDARD				0.4%
STAND. FLUENCE CLOCK				2413.

TIME VALUES FOR UNKNOWN SAMPLES

	DAY	HOUR	MINUTE	SECOND
CASE NO. = 1				
TUIRR	0.	8.	0.	0.
UTR	0.	0.	0.	0.
UTS	18.	4.	8.	20.
UTSM	15.	3.	53.	7.
UNKNOWN CLOCK TIME				57695.
UNKNOWN LIVE TIME				57600.
DEAD TIME UNKNOWN				0.2%
UNKN. FLUENCE CLOCK				1303.
CASE NO. = 2				
TUIRR	0.	8.	0.	0.
UTR	0.	0.	0.	0.
UTS	18.	21.	1.	32.
UTSM	15.	4.	29.	16.
UNKNOWN CLOCK TIME				57714.
UNKNOWN LIVE TIME				57600.
DEAD TIME UNKNOWN				0.2%
UNKN. FLUENCE CLOCK				1350.
CASE NO. = 3				
TUIRR	0.	8.	0.	0.
UTR	0.	0.	0.	0.
UTS	20.	1.	16.	40.
UTSM	15.	5.	2.	51.
UNKNOWN CLOCK TIME				57909.
UNKNOWN LIVE TIME				57600.
DEAD TIME UNKNOWN				0.5%
UNKN. FLUENCE CLOCK				1320.
CASE NO. = 4				
TUIRR	0.	8.	0.	0.
UTR	0.	0.	0.	0.
UTS	20.	12.	26.	42.
UTSM	15.	5.	26.	20.
UNKNOWN CLOCK TIME				57671.
UNKNOWN LIVE TIME				57600.
DEAD TIME UNKNOWN				0.1%
UNKN. FLUENCE CLOCK				1604.
CASE NO. = 5				
TUIRR	0.	8.	0.	0.
UTR	0.	0.	0.	0.
UTS	21.	16.	23.	44.
UTSM	15.	15.	23.	52.
UNKNOWN CLOCK TIME				57694.
UNKNOWN LIVE TIME				57600.
DEAD TIME UNKNOWN				0.2%
UNKN. FLUENCE CLOCK				1685.

* * * STANDARD SAMPLE INPUT * * *

WIRE PEAK AREA = 20008. WIRE MASS = 0.0148
 WIRE PEAK ERROR = 2.20 SAMPLE MASS = 0.6023

I	SEAL11	SIGSPA	SCONG	SIGCON
1	23235.	6.20	2.80 PPM	10.71
2	68186.	2.20	32.00 PPM	12.50
3	5514.	25.40	2.60 PPM	11.54
4	32599.	3.60	34.00 PPM	5.83
5	10829.	8.90	1.55 PPM	5.16
6	3903.	24.00	122.00 PPM	9.02
7	10421.	8.40	2.00 PPM	15.00
8	2401.	34.20	0.32 PPM	15.63
9	349792.	0.40	6.30 PPM	8.82
10	4205.	11.60	29.00 PPM	3.45
11	16883.	2.70	6.50 PPM	3.08
12	1665.	13.50	0.40 PPM	7.50
13	663.	12.10	0.50 PPM	15.00

J	UPAM(J)	SIGUPM(J)	UMM(J)	UM(J)
1	0.10009E 05	3.10	0.13900E-01	0.69420E 00

UPA(1, 1)=	8920.	SIGUPA =	10.60
UPA(2, 1)=	37571.	SIGUPA =	2.60
UPA(3, 1)=	1910.	SIGUPA =	44.20
UPA(4, 1)=	5700.	SIGUPA =	12.00
UPA(5, 1)=	11571.	SIGUPA =	4.90
UPA(6, 1)=	6373.	SIGUPA =	7.90
UPA(7, 1)=	2135.	SIGUPA =	22.70
UPA(8, 1)=	1367.	SIGUPA =	32.90
UPA(9, 1)=	99480.	SIGUPA =	0.80
UPA(10, 1)=	987.	SIGUPA =	27.70
UPA(11, 1)=	3337.	SIGUPA =	8.50
UPA(12, 1)=	865.	SIGUPA =	17.70
UPA(13, 1)=	234.	SIGUPA =	22.50

J	UPAH(J)	SIGUPM(J)	UMM(J)	UM(J)
2	0.10014E 05	3.00	0.14000E-01	0.59830E 00

UPA(1, 2)=	4841.	SIGUPA =	20.30
UPA(2, 2)=	16558.	SIGUPA =	6.00
UPA(3, 2)=	3969.	SIGUPA =	22.90
UPA(4, 2)=	14333.	SIGUPA =	5.30
UPA(5, 2)=	2987.	SIGUPA =	20.70
UPA(6, 2)=	1195.	SIGUPA =	50.50
UPA(7, 2)=	3924.	SIGUPA =	14.50
UPA(8, 2)=	976.	SIGUPA =	55.20
UPA(9, 2)=	113313.	SIGUPA =	0.80
UPA(10, 2)=	1339.	SIGUPA =	26.70
UPA(11, 2)=	5803.	SIGUPA =	5.90
UPA(12, 2)=	450.	SIGUPA =	33.10
UPA(13, 2)=	546.	SIGUPA =	12.00

J	UPAM(J)	SIGUPM(J)	UMM(J)	UM(J)
3	0.99980E 04	3.10	0.13700E-01	0.54970E 00

UPA(1, 3)=	8960.	SIGUPA =	17.20
UPA(2, 3)=	47210.	SIGUPA =	3.40
UPA(3, 3)=	8152.	SIGUPA =	18.50
UPA(4, 3)=	24510.	SIGUPA =	5.10
UPA(5, 3)=	8608.	SIGUPA =	11.90
UPA(6, 3)=	4196.	SIGUPA =	24.10
UPA(7, 3)=	11293.	SIGUPA =	8.90
UPA(8, 3)=	2022.	SIGUPA =	47.90
UPA(9, 3)=	222443.	SIGUPA =	0.60
UPA(10, 3)=	4093.	SIGUPA =	15.90
UPA(11, 3)=	23645.	SIGUPA =	2.00
UPA(12, 3)=	1331.	SIGUPA =	20.70
UPA(13, 3)=	5025.	SIGUPA =	2.80

J	UPAM(J)	SIGUPM(J)	UMM(J)	UM(J)
4	0.99920E 04	3.10	0.13200E-01	0.71910E 00

UPA(1, 4)=	3051.	SIGUPA =	25.00
UPA(2, 4)=	20604.	SIGUPA =	3.90
UPA(3, 4)=	1137.	SIGUPA =	63.90
UPA(4, 4)=	4066.	SIGUPA =	14.60
UPA(5, 4)=	3572.	SIGUPA =	13.60
UPA(6, 4)=	6778.	SIGUPA =	7.10
UPA(7, 4)=	0.	SIGUPA =	0.00
UPA(8, 4)=	906.	SIGUPA =	43.50
UPA(9, 4)=	80961.	SIGUPA =	0.80
UPA(10, 4)=	0.	SIGUPA =	0.00
UPA(11, 4)=	3862.	SIGUPA =	6.50
UPA(12, 4)=	0.	SIGUPA =	0.00
UPA(13, 4)=	198.	SIGUPA =	22.50

J	UPAM(J)	SIGUPM(J)	UMM(J)	UM(J)
5	0.10008E 05	3.20	0.13300E-01	0.57350E 00

UPA(1, 5)=	3824.	SIGUPA =	23.10
UPA(2, 5)=	36121.	SIGUPA =	2.60
UPA(3, 5)=	1388.	SIGUPA =	59.30
UPA(4, 5)=	7115.	SIGUPA =	9.50
UPA(5, 5)=	2856.	SIGUPA =	19.10
UPA(6, 5)=	4809.	SIGUPA =	11.10
UPA(7, 5)=	3838.	SIGUPA =	12.80
UPA(8, 5)=	1391.	SIGUPA =	33.20
UPA(9, 5)=	88440.	SIGUPA =	0.30
UPA(10, 5)=	794.	SIGUPA =	23.80
UPA(11, 5)=	4770.	SIGUPA =	6.30
UPA(12, 5)=	381.	SIGUPA =	42.80
UPA(13, 5)=	295.	SIGUPA =	17.30

TABLE ISOTOPE CONCENTRATION FOR CASE NUMBER 1

ISOTOPE	CONCENTRATION		EXPECTED ERROR		
SM-153	1.29	+/-	0.22	PPM	(16.8%)
CE-141	15.82	+/-	2.15	PPM	(13.6%)
SE-75	0.80	+/-	0.42	PPM	(52.4%)
CR-51	5.35	+/-	0.77	PPM	(14.4%)
HF-181	1.48	+/-	0.18	PPM	(12.1%)
BA-131	199.09	+/-	54.00	PPM	(27.1%)
CS-134	0.36	+/-	0.10	PPM	(28.8%)
TB-160	0.16	+/-	0.08	PPM	(50.1%)
SC-46	1.72	+/-	0.17	PPM	(9.7%)
RB-86	6.19	+/-	1.89	PPM	(30.5%)
CO-60	1.13	+/-	0.12	PPM	(10.2%)
TA-182	0.18	+/-	0.04	PPM	(23.8%)
SB-124	0.19	+/-	0.06	PPM	(29.9%)

TABLE ISOTOPE CONCENTRATION FOR CASE NUMBER 2

ISOTOPE	CONCENTRATION		EXPECTED ERROR		
SM-153	1.09	+/-	0.26	PPM	(24.1%)
CE-141	9.56	+/-	1.25	PPM	(14.6%)
SE-75	2.01	+/-	0.73	PPM	(36.3%)
CR-51	16.56	+/-	1.58	PPM	(9.5%)
HF-181	0.47	+/-	0.11	PPM	(23.4%)
BA-131	43.59	+/-	24.74	PPM	(56.8%)
CS-134	0.80	+/-	0.18	PPM	(22.0%)
TB-160	0.14	+/-	0.09	PPM	(66.9%)
SC-46	2.38	+/-	0.23	PPM	(9.7%)
PB-86	10.42	+/-	3.08	PPM	(29.6%)
CO-60	2.38	+/-	0.19	PPM	(8.2%)
TA-182	0.12	+/-	0.05	PPM	(41.3%)
SB-114	0.54	+/-	0.12	PPM	(23.0%)

TABLE ISOTOPE CONCENTRATION FOR CASE NUMBER 3

ISOTOPE	CONCENTRATION		EXPECTED ERROR		
SM-153	3.19	+/-	0.69	PPM	(21.6%)
CE-141	26.01	+/-	3.57	PPM	(13.7%)
SE-75	4.32	+/-	1.46	PPM	(33.7%)
CR-51	30.32	+/-	2.87	PPM	(9.5%)
HF-181	1.43	+/-	0.23	PPM	(16.2%)
BA-131	170.27	+/-	60.30	PPM	(35.4%)
CS-134	2.40	+/-	0.48	PPM	(19.8%)
TB-160	0.31	+/-	0.19	PPM	(61.0%)
SC-46	4.90	+/-	0.48	PPM	(9.7%)
FB-86	34.59	+/-	7.05	PPM	(20.4%)
CD-60	12.22	+/-	0.74	PPM	(6.1%)
TA-182	0.36	+/-	0.09	PPM	(26.1%)
SB-124	5.20	+/-	1.03	PPM	(19.9%)

TABLE ISOTOPE CONCENTRATION FOR CASE NUMBER 4

ISOTOPE	CONCENTRATION		EXPECTED ERROR			
SM-153	1.26	+/-	0.35	PPM	(28.2%)	
CE-141	10.36	+/-	1.44	PPM	(13.9%)	
SE-75	0.54	+/-	0.38	PPM	(69.8%)	
CR-51	4.60	+/-	0.77	PPM	(16.6%)	
HF-181	0.54	+/-	0.09	PPM	(17.5%)	
BA-131	257.68	+/-	69.33	PPM	(26.9%)	
CS-134	0.00	+/-	0.00	PPM	(0.0%)	
TB-160	0.12	+/-	0.07	PPM	(57.6%)	
SC-46	1.61	+/-	0.16	PPM	(9.7%)	
RB-86	0.00	+/-	0.00	PPM	(0.0%)	
CO-60	1.48	+/-	0.13	PPM	(8.7%)	
TA-182	0.00	+/-	0.00	PPM	(0.0%)	
SB-124	0.19	+/-	0.06	PPM	(29.9%)	

TABLE ISOTOPE CONCENTRATION FOR CASE NUMBER 5

ISOTOPE	CONCENTRATION		EXPECTED ERROR		
SM-153	2.87	+/-	0.76	PPM	(26.5%)
CE-141	24.32	+/-	3.30	PPM	(13.6%)
SE-75	0.88	+/-	0.58	PPM	(65.7%)
CR-51	10.92	+/-	1.35	PPM	(12.4%)
HF-181	0.57	+/-	0.13	PPM	(22.1%)
BA-131	253.13	+/-	71.47	PPM	(28.2%)
CS-134	0.97	+/-	0.21	PPM	(21.8%)
TB-160	0.25	+/-	0.13	PPM	(50.3%)
SC-46	2.33	+/-	0.23	PPM	(9.8%)
RB-86	3.41	+/-	3.44	PPM	(40.8%)
CO-60	2.40	+/-	0.21	PPM	(8.6%)
TA-162	0.12	+/-	0.06	PPM	(45.7%)
SB-124	0.37	+/-	0.10	PPM	(26.2%)

INSTRUMENTAL NEUTRON ACTIVATION
ANALYSIS OF COAL AND COAL FLY ASH

by

Jack F. Higginbotham
B.S., Kansas State University, 1981

AN ABSTRACT OF A MASTER'S THESIS

submitted in partial fulfillment of the
requirements for the degree

MASTER OF SCIENCE

Department of Nuclear Engineering
KANSAS STATE UNIVERSITY
Manhattan, Kansas

1983

ABSTRACT

The procedures are developed for using the analysis technique of instrumental neutron activation, INAA, to determine the minor and trace element composition of bituminous coals and fly ashes. These procedures are to be used by a technician with minimal experience with INAA but with some background in radiation physics and detection. The procedures require the use of a National Bureau of Standards Bituminous Coal, SRM 1632a, as a reference material and twenty-eight elements can be quantitatively identified relative to this coal.

The accuracy of the procedure was determined by analyzing a sample of the reference and comparing the results with the given concentrations. Nineteen elements were measured within 10 percent of the expected concentration and all twenty-eight were measured within 30 percent.

The elemental concentrations of 5 different coals used in Kansas for electrical generation were determined. Also, the composition was measured for an NBS Fly Ash, SRM 1633a, as well as a fly ash from Mr. A. Anderson of Purdue University.