APPLICATION
OF GAMMA RAY SPECTROMETRY
AS A SUPPLEMENTARY MIST TECHNIQUE

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MASTER

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Introduction

The determination of neutron exposure or fuel burnup by nondestructive gamma scanning techniques has the potential of being the only practical safeguards check against diversion of plutonium during the time following discharge of fuel from the reactor until it is chemically processed. The information one ultimately hopes to obtain from the application of the technique to power reactor fuels is the plutonium content of a given fuel bundle, thus providing a check on numbers quoted by the chemical processor of the fuel. The technique is also useful for identifying and sorting nuclear fuel.

The use of gamma scanning to determine fuel burnup is envisioned as a supplementary MIST technique wherein a redundant check on the plutonium content of fuel is obtained. The MIST technique, when applied to the chemical reprocessing data, will indicate whether the chemical processor has falsified his numbers. However, by also having determined the plutonium content via nondestructive gamma scanning techniques, there is a redundant check on this information and both methods are enhanced. For example, the number of mass spectrometric remeasurements of dissolved fuel samples required by the safeguards agency may be greatly reduced due to the confidence gained from gamma scanning.

An additional advantage of gamma scanning the reactor fuel is that it provides an essentially absolute check on the fuel during transportation. If one were to gamma scan the fuel before and after transportation one would expect to see essentially reproducible results. Hence one could very accurately ascertain whether fuel was substituted or diverted during trans-

portation. This in itself may be reason enough to further develop the technique.

There have been scattered attempts in the literature to apply this technique to the determination of exposure of irradiated fuels, but the existing data is insufficient to demonstrate the feasibility of the technique or to put an absolute error bar on the exposure so obtained. However, the research that has been done to date leads one to conclude that there is a potential to determine burnup of irradiated fuel to within 5%, and that the technique is indeed a feasible safeguards method.

In this paper, we will first present a basic description of the method itself. Then we will review two research efforts which have been performed at Battelle-Northwest with regard to the gamma scanning tec hnique and, finally, we will present how we feel the method could be applied directly as a supplementary MIST technique in the safeguards system.

NON-DESTRUCTIVE DETERMINATION OF BURNUP

The following will briefly summarize what is involved in the application
of a non-destructive gamma scanning technique[1] to the determination of
absolute flux and exposure as well as what may be necessary to demonstrate
its potential and reliability. Consider first, for simplicity sake, the
determination of exposure at a point on a fuel pin. In applying the technique,
one utilizes measured and calculated fission product activity ratios. The
measured activity ratios are inferred from photopeak areas in the gamma ray
spectra of the fission products as observed with a high resolution Ge(Li)
gamma ray spectrometer.

Having determined the photopeak area ratios, we must now convert these
to activity ratios. To develop the relationship necessary for this conversion,
consider the relationship between the number of gamma rays of energy i from
isotope j that are detected (full energy absorption in the crystal) per unit
time, i.e., the photopeak area, A_{ij}, relative to the number of decay events
per unit time of the radioactive isotope, i.e., the activity, $N_j \lambda_j$. Since
on the average only a given fraction of the $N_j \lambda_j$ decay events result in a
gamma ray of energy i, the number of gamma rays of energy i per decay event
must be known. Of this number only a certain fraction are traveling in the
direction of the detector and only the fraction which escape removal by
absorption or scattering between the source and detector actually reach the
detector. Of those gamma rays that do reach the detector, only a fraction are
detected, i.e., those whose total energy is absorbed in the crystal. These
gamma rays produce the photopeak at energy i (whose area is A_{ij}). In equation
form we have:

$$A_{ij} = \left\{ N_j \lambda_j \right\} \cdot \left\{ BR_{ij} \right\} \cdot \left\{ e_i \right\} \cdot \left\{ G \right\} \cdot \left\{ \exp \left(-\sum_1^k \mu_i \rho_k x \right) \right\} \qquad (1)$$

where

A_{ij} Area of photo-peak of energy i from decay of isotope j

$N_j \lambda_j$ $\equiv$ Decay rate of isotope j

BR_{ij} $\equiv$ Branching ratio: Unconverted gamma rays of energy i per data event of isotope j

e_i $\equiv$ Efficiency of detector for gamma rays of energy i

G $\equiv$ Geometry factor for the source-detector geometry used

$\exp \left(-\sum_1^k \mu_i \rho_k x \right) \equiv$ Removal of gamma rays of energy i in material k between source and detector

The last three factors in this equation yield the absolute efficiency of the detector system for gamma rays of energy i. Since we are interested in activity ratios rather than absolute activities, it is only necessary to determine relative efficiencies under actual counting conditions. To illustrate how these relative efficiencies are obtained consider the area ratio of two photopeaks resulting from gamma rays at energyes 1 and 2 emitted during the decay of a single isotope j. From (1) we have

$$\frac{A_{1j}}{A_{2j}} = \cdot \frac{BR_{1j}}{BR_{2j}} \cdot \frac{e_1}{e_2} \cdot \frac{\exp \left(-\sum_1^k \mu_1 \rho_k x \right)}{\exp \left(-\sum_1^k \mu_2 \rho_k x \right)}$$

Letting $e_i \exp \left(-\sum_1^k \mu_i \rho_k x \right) \equiv \epsilon_i$ we have

$$\frac{\epsilon_2}{\epsilon_1} = \frac{A_{2j}}{A_{1j}} \frac{BR_{1j}}{BR_{2j}} \qquad (2)$$

PARAMETERS THAT INFLUENCE THE FORMATION AND DETECTION OF RADIOACTIVE FISSION PRODUCT NUCLIDES BY GAMMA SCANNING

$$A_{ij} = \left\{ N_j \lambda_j \right\} \cdot \left\{ BR_{ij} \cdot \left\{ e_i \right\} \cdot \left\{ G \right\} \cdot \left\{ \exp\left(-\sum_1^k \mu_i \rho_k x \right) \right\} \right.$$

where

A_{ij} $\equiv$ AREA OF PHOTO-PEAK OF ENERGY i FROM DECAY OF ISOTOPE j

$N_j \lambda_j$ $\equiv$ DECAY RATE OF ISOTOPE j

BR_{ij} $\equiv$ BRANCHING RATIO: UNCONVERTED GAMMA RAYS OF ENERGY i PER DECAY EVENT OF ISOTOPE j

e_i $\equiv$ EFFICIENCY OF DETECTOR FOR GAMMA RAYS OF ENERGY i

G $\equiv$ GEOMETRY FACTOR FOR THE SOURCE-DETECTOR GEOMETRY USED

$\exp\left(-\sum_1^k \mu_i \rho_k x \right)$ $\equiv$ REMOVAL OF GAMMA RAYS OF ENERGY i IN MATERIAL k BETWEEN SOURCE AND DETECTOR

RELATIVE EFFICIENCY DETERMINATION

$$\frac{A_{1j}}{A_{2j}} = \cdot \frac{BR_{1j}}{BR_{2j}} \cdot \frac{e_1}{e_2} \cdot \frac{\exp\left(-\sum_1^k \mu_1 \rho_k x\right)}{\exp\left(-\sum_1^k \mu_2 \rho_k x\right)}$$

LETTING $e_i \exp\left(-\sum_1^k \mu_i \rho_k x\right) \equiv \varepsilon_i$ WE HAVE

$$\frac{\varepsilon_2}{\varepsilon_1} = \frac{A_{2j}}{A_{1j}} \frac{BR_{1j}}{BR_{2j}}$$

Equation (2) gives the detector efficiency for gamma rays of energy 2 relative to that for gamma rays of energy 1. Thus if we have a fission product isotope in the fuel emitting several gamma rays whose branching ratios or relative intensities are well known we can obtain the relative efficiency curve from the measured photopeak areas.

Having determined the relative efficiencies from one of the gamma ray spectra we are now in a position to get the desired activity ratios from the photopeak area ratios. Again using equation (1) we have for the area ratio of two photopeaks at different energies from different isotopes

$$\frac{A_{ij}}{A_{\ell m}} = \frac{N_j \lambda_j}{N_m \lambda_m} \cdot \frac{BR_{ij}}{Br_{\ell m}} \cdot \frac{\epsilon_i}{\epsilon_\ell}$$

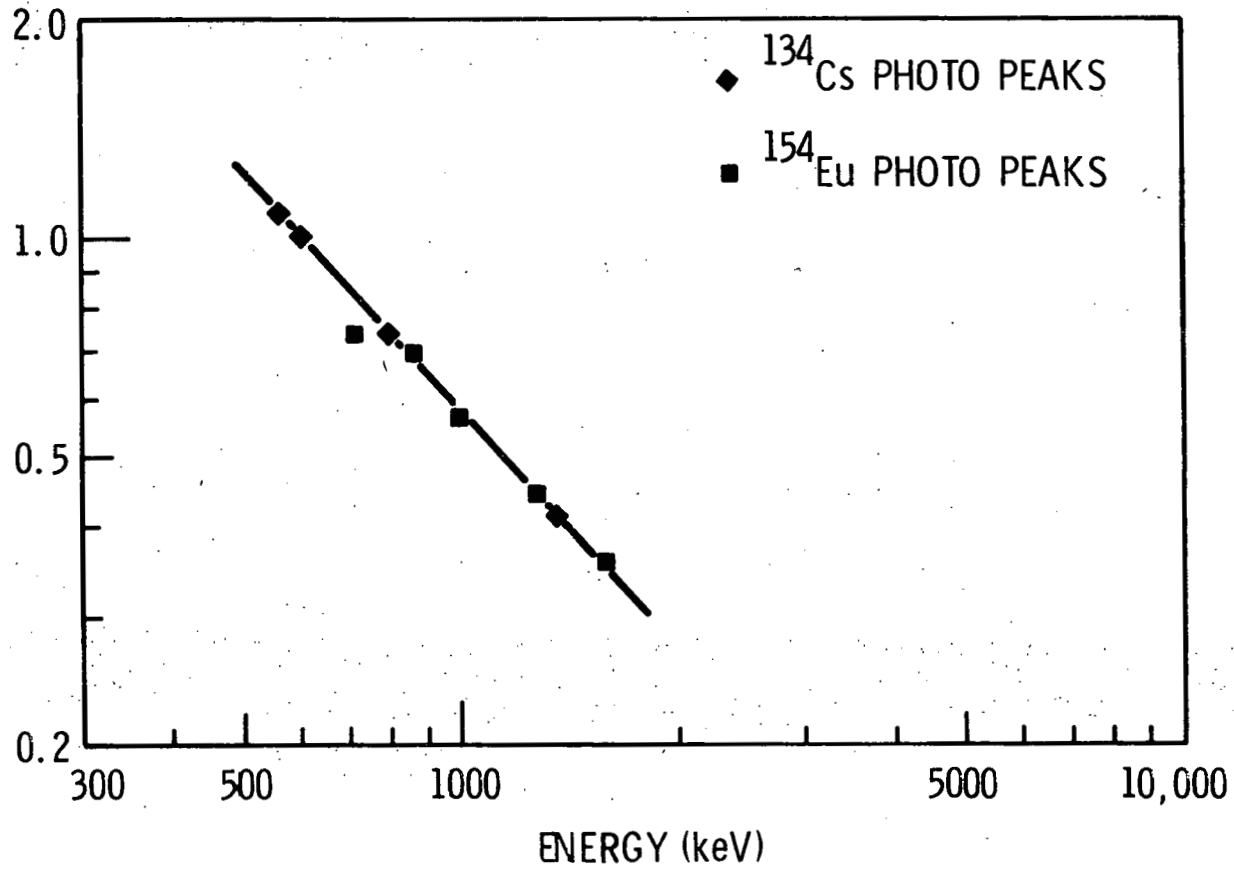Solving for the desired activity ratios we have

$$\frac{N_j \lambda_j}{N_m \lambda_m} = \frac{A_{ij}}{A_{\ell m}} \cdot \frac{BR_{\ell m}}{BR_{ij}} \cdot \frac{\epsilon_\ell}{\epsilon_i} \tag{3}$$

Thus the desired activity ratios are obtained as per equation (3) from the measured photopeak area ratios, the branching or relative intensity ratios tabulated in the literature and the measured relative efficiency ratios.

We see that the conversion of photopeak area ratios measured at a given cooling time to activity ratios at reactor shutdown requires the following information:

1. Cooling time.
2. Half-lives of fission products.
3. Branching ratios of gamma rays in the decay scheme.
4. Relative efficiency of the Ge(Li) detector.

RELATIVE EFFICIENCY CALIBRATION FOR THE Ge(Li) SPECTROMETER

ACTIVITY RATIOS FROM
MEASURED PHOTOPEAK AREA RATIOS

$$\frac{A_{ij}}{A_{\ell m}} = \frac{N_j \lambda_j}{N_m \lambda_m} \cdot \frac{BR_{ij}}{Br_{\ell m}} \cdot \frac{\varepsilon_i}{\varepsilon_\ell}$$

SOLVING FOR THE DESIRED ACTIVITY RATIOS WE HAVE

$$\frac{N_j \lambda_j}{N_m \lambda_m} = \frac{A_{ij}}{A_{\ell m}} \cdot \frac{BR_{\ell m}}{BR_{ij}} \cdot \frac{\varepsilon_\ell}{\varepsilon_i}$$

REQUIRED INFORMATION
FOR CONVERSION OF PHOTOPEAK AREA RATIOS

1. COOLING TIME.

2. HALF-LIVES OF FISSION PRODUCTS.

3. BRANCHING RATIOS OF GAMMA RAYS IN THE DECAY SCHEME.

4. RELATIVE EFFICIENCY OF THE Ge(Li) DETECTOR.

Items (2) are well known, while items (3) are generally not as well known. Item (4) is measurable with reasonable accuracy. It may be possible to get the cooling time from the gamma ray spectra, but for present purposes we will assume it is known from fuel discharge records. We now have a set of ratios, R_i^m, of measured fission product reactivity ratios at reactor shutdown.

It is also possible to calculate the set of ratios, R_i^c, with relatively simple analytical equations based on the basic processes involved. There are however, a considerable number of "constants" involved in these equations which have varying degrees of influence on the results. Optimum subsets of these can be arrived at by a systematic variation and evaluation, e.g., for consistency between and within different sets of data. The constants needed in the analytical determination of the ratios are:

1. Initial nuclei densities of all fissile and fertile fuel isotopes.

2. One group microscopic cross sections for absorption, capture and fission for all fissile and fertile fuel isotopes.

3. Fission product yields for each fission product from each fissionable isotope.

4. Microscopic absorption and capture cross section for each fission product of interest.

5. Half-life of each fission product of interest.

In the above items (1) and (5) are usually well known. Item (3) is thought to be well known for ^{235}U and ^{239}Pu. Items (2) can be obtained in a number of ways and an optimum set could be constructed for a particular reactor from the analysis of a few sets of data from that reactor. Item (4) is usually either relatively unimportant (^{137}Cs, ^{95}Zr) or very important (^{133}Cs, ^{134}Cs, ^{153}Eu and ^{154}Eu) but generally poorly known.

CONSTANTS NEEDED IN
THE ANALYTICAL DETERMINATION OF RATIOS

1. INITIAL NUCLEI DENSITIES OF ALL FISSILE AND FERTILE FUEL ISOTOPES.

2. ONE GROUP MICROSCOPIC CROSS SECTIONS FOR ABSORPTION, CAPTURE AND FISSION FOR ALL FISSILE AND FERTILE FUEL ISOTOPES.

3. FISSION PRODUCT YIELDS FOR EACH FISSION PRODUCT FROM EACH FISSIONABLE ISOTOPE.

4. MICROSCOPIC ABSORPTION AND CAPTURE CROSS SECTION FOR EACH FISSION PRODUCT OF INTEREST.

5. HALF-LIFE OF EACH FISSION PRODUCT OF INTEREST.

Additional information required for the calculation is the operating history of the reactor, i.e., relative power versus time histograms. The precision to which this must be known depends on the fission products of interest which is in turn related to the cooling time at which the gamma scan measurements are made.

With the above information as input for given fuel material, the computer program DRAFT[2] calculates information concerning fission events such as fuel isotopic concentrations and fission product disintegration rates at selected times during and after irradiation. Irradiation history of the fuel is input in the form of a histogram to allow an accurate description of fission product formation. The following information is calculated and printed at as many as 100 values of time as specified by the user in the input through the power history.

- Accumulated exposure time
- Time averaged flux and accumulated exposure
- Fuel isotope (^{235}U, ^{238}U and ^{239}Pu) concentrations,
- Accumulated fissions in each fuel isotope (^{235}U, ^{238}U and ^{239}Pu), accumulated fissions in all fuel isotopes, and the fraction of accumulated fissions for each fuel isotope
- Disintegration rate for each fission product
- Ratios of selected fission product disintegration rates.

The above information can be generated for as many as 100 input exposure averaged fluxes, 20 fission products and 20 fission product activity ratios in a single pass. A plotting routine has been added to the program to allow for any or all of the following output to be plotted:

1. Ratio of final to initial fuel concentrations versus exposure.

CALCULATED INFORMATION

- ACCUMULATED EXPOSURE TIME

- TIME AVERAGED FLUX AND ACCUMULATED EXPOSURE.

- FUEL ISOTOPE (^{235}U, ^{238}U AND ^{239}Pu) CONCENTRATIONS.

- ACCUMULATED FISSIONS IN EACH FUEL ISOTOPE (^{235}U, ^{238}U AND ^{239}Pu), ACCUMULATED FISSIONS IN ALL FUEL ISOTOPES, AND THE FRACTION OF ACCUMULATED FISSIONS FOR EACH FUEL ISOTOPE.

- DISINTEGRATION RATE FOR EACH FISSION PRODUCT.

- RATIOS OF SELECTED FISSION PRODUCT DISINTEGRATION RATES.

2. Disintegration rate versus exposure or time, between any two specified times, for any fission product.

3. Disintegration rate ratio versus exposure or time between any two specified times and for any two fission products.

The code is used mainly for two purposes. First, it is used to determine which fission products and fission product activity ratios will be most useful in determining such quantities as average flux, exposure, fraction of fissions in each fuel isotope and total fissions for a particular reactor type. Secondly, the actual determination of these quantities is made by correlating the selected calculated fission product activity ratios with measured values obtained by nondestructive gamma scanning techniques.

With Program DRAFT, then, calculated ratios, R_i^c, are determined assuming different values of the absolute neutron flux. For each value of the flux a plot of a given ratio vs. exposure results in a family of curves for the R_i^c. (See Figure 1). If a horizontal line is drawn which represents the measured value R_i^m , this line will intersect each curve in the family once. (Again see Figure 1). Each point of intersection determines a (ϕ, E) point for the R_i. A curve is then constructed of ϕ vs. E for each R_i. (See Figure 2). The "intersection" of these curves then yields the absolute flux and exposure. This flux is then inserted back into DRAFT and the end of life fuel isotopics and atom densities determined.

The larger the number of ratios used the better the exposure and flux can be determined and the easier it is to pick out inconsistent data and assign an uncertainty to the exposure. Which ratios can be used is a function of cooling time, among other things. It remains to be determined which ratios
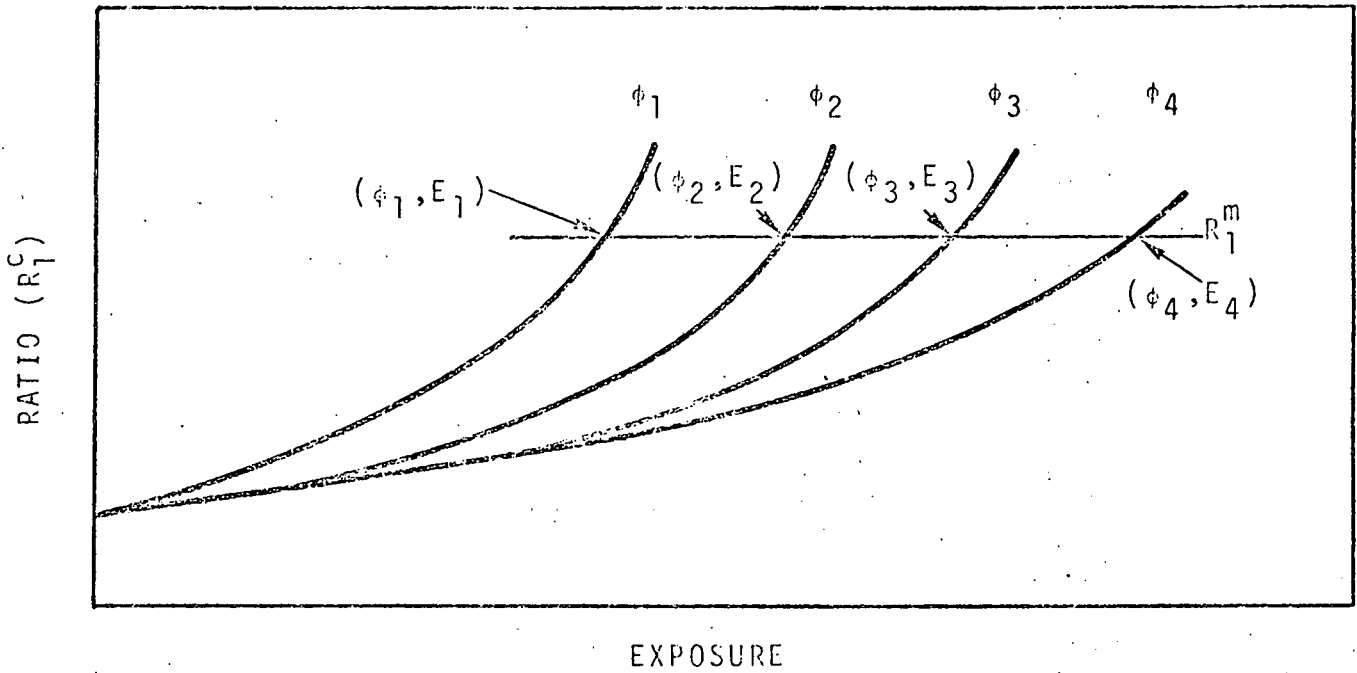
FIGURE 1. Example of a Calculated Fission Product Activity Ratio (R_1^c) as a Function of Neutron Exposure and Flux.
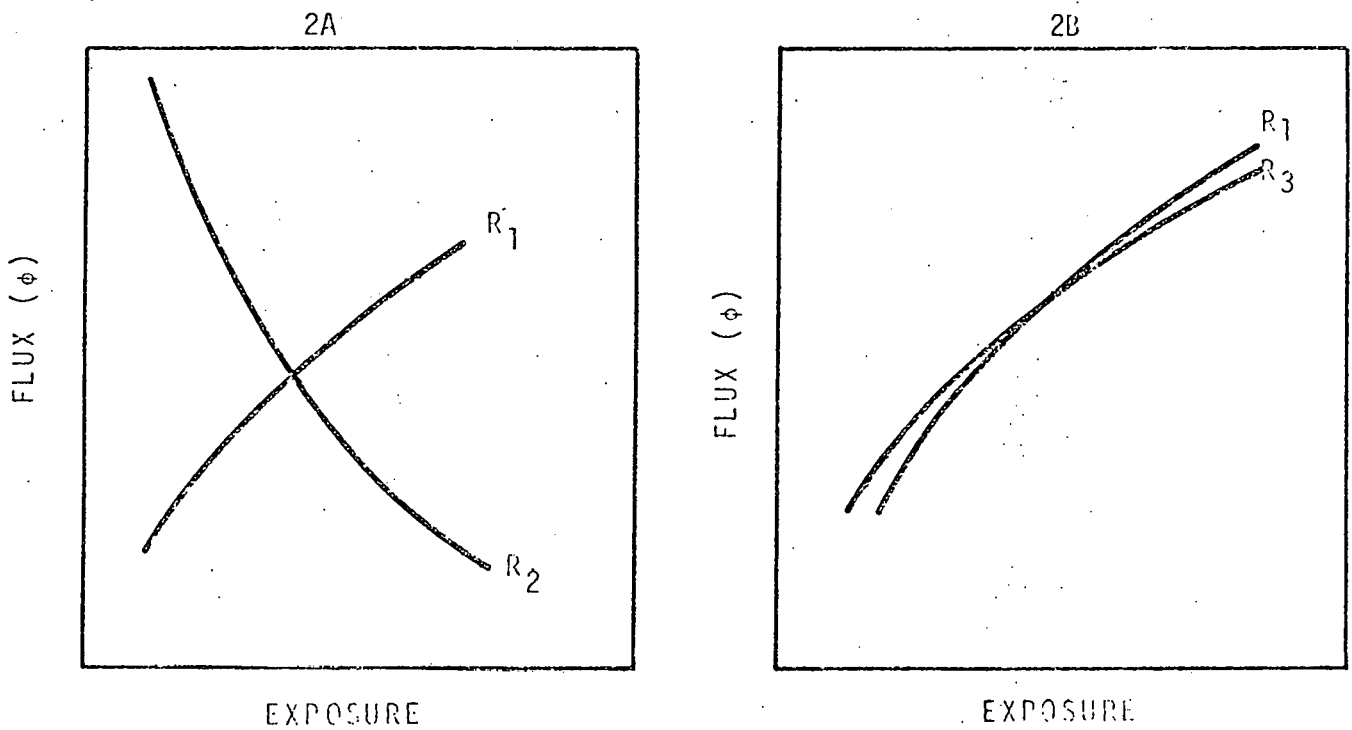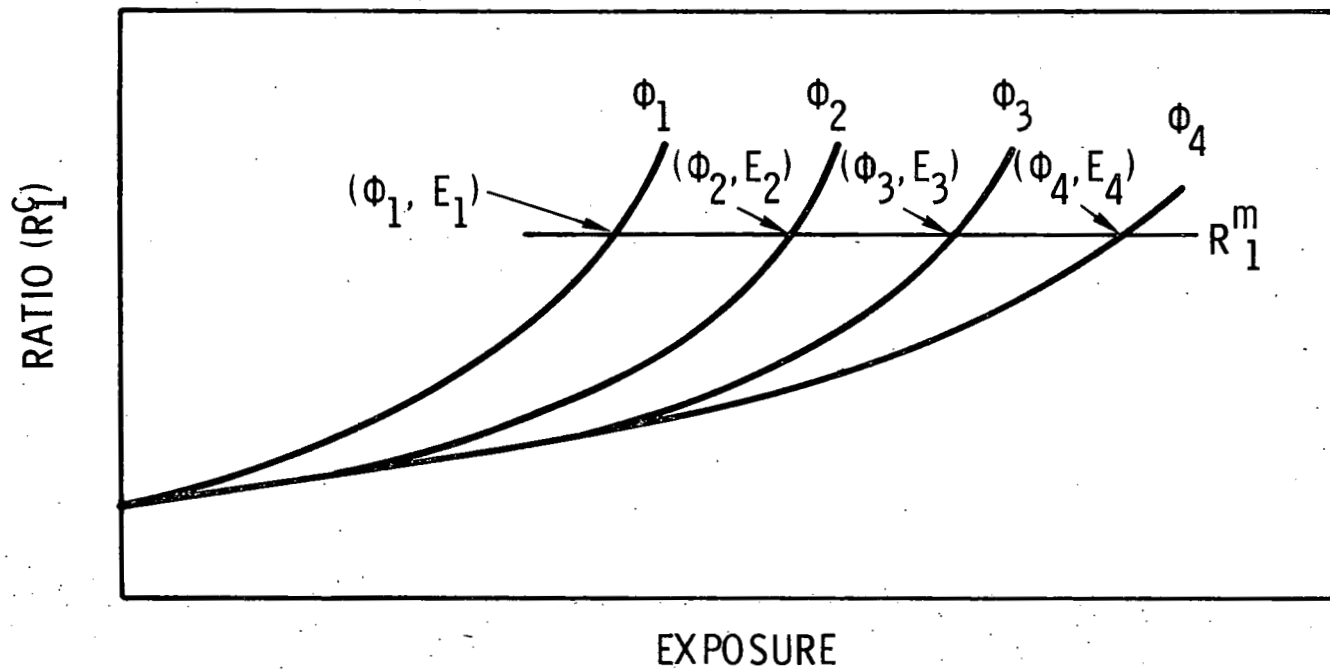

FIGURE 2. Example of the Determination of Flux and Exposure [A]. From Ratios One of Which is Flux Sensitive (R_1) and the Other Exposure Sensitive (R_2) and [B]. From Two Ratios Which are Flux Sensitive (R_1 and R_3).

EXAMPLE OF A CALCULATED FISSION PRODUCT ACTIVITY RATIO (R_1^c)
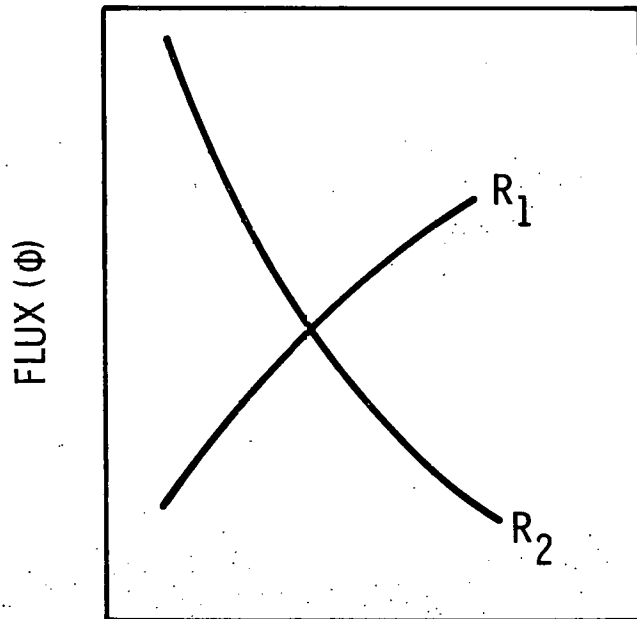
AS A FUNCTION OF NEUTRON EXPOSURE AND FLUX

EXAMPLE OF THE DETERMINATION OF FLUX AND EXPOSURE [A],

FROM RATIOS ONE OF WHICH IS FLUX SENSITIVE (R_1)
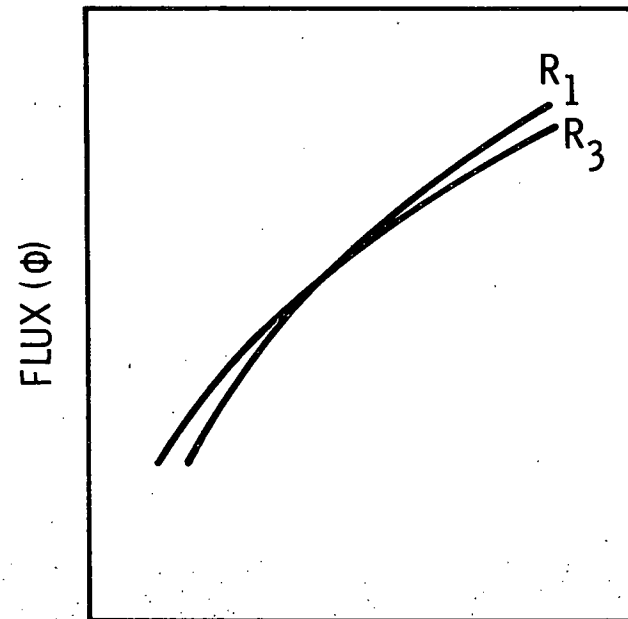
AND THE OTHER EXPOSURE SENSITIVE (R_2) AND [B].

FROM TWO RATIOS WHICH ARE FLUX SENSITIVE (R_1 AND R_3)

are the best to use. The graphical solution for ϕ and E indicated previously works best when one ratio is flux sensitive (e.g., $^{144}Ce/^{137}Cs$) while the other is exposure sensitive (e.g., $^{134}Cs/^{137}Cs$ or $^{154}Eu/^{137}Cs$) since this case yields curves which intersect almost perpendicularly (See Figure 2A). If only ratios of one type are used, the curves are nearly parallel at intersection and therefore make it difficult to get ϕ and E accurately for the general case (See Figure 2 B).

The extension of the above analysis technique to a completely automatic computerized system is straightforward and could be accomplished after the usefulness and adequacy of the method for burnup determination has been demonstrated.

NONDESTRUCTIVE DETERMINATION OF BURNUP OF YANKEE ROWE FUEL

The feasibility of non-destructive determination of the exposure or burnup of spent fuel has been investigated by comparing non-destructive and destructive measurements of burnup from two Yankee Rowe fuel rods irradiated in cores I, II and IV. Good agreement would indicate the desirability of attempting to extend the non-destructive techniques to determine Pu content of a spent fuel assembly. The relatively non-intrusive techniques have the potential of providing a practical safeguards measurement prior to fuel dissolution which is independent of subsequent measurements and amenable to automatic determination.

The non-destructive measurements were based on the utilization of a high resolution Ge(Li) spectrometer to measure fission product gamma ray activity. Data were collected in the BNW Underwater Gamma Scan Facility located in the Plutonium Recycle Test Reactor water basin.[3] To apply a technique which has been described by J. A. Sovka,[1] measured fission product activity ratios representing 13 photopeaks available from ^{106}Ru-^{106}Rh, ^{134}Cs, ^{137}Cs, ^{144}Ce-^{144}Pr and ^{154}Eu, were compared to calculated activity ratios obtained from relatively simple analytical burnup equations programmed at BNW as computer code DRAFT.[2] The ^{134}Cs and ^{154}Eu do not result directly from fission but are formed from neutron capture in ^{133}Cs and ^{153}Eu, respectively, which are formed directly in fission. The flux and exposure were determined by comparing measured and calculated activity ratios. Values of the flux which produced ratios identical with measured ratios are presented in Table I. The values represent four sets of measurements from the two rods, two measurements which have been averaged from one rod designated as an inner rod and two

THE UNDERWATER GAMMA SCAN FACILITY

CRYOSTAT

DETECTOR
HOUSING

CARRIAGE

RAILS

COLLIMATOR

SHUTTER HOUSING

FUEL TRAY
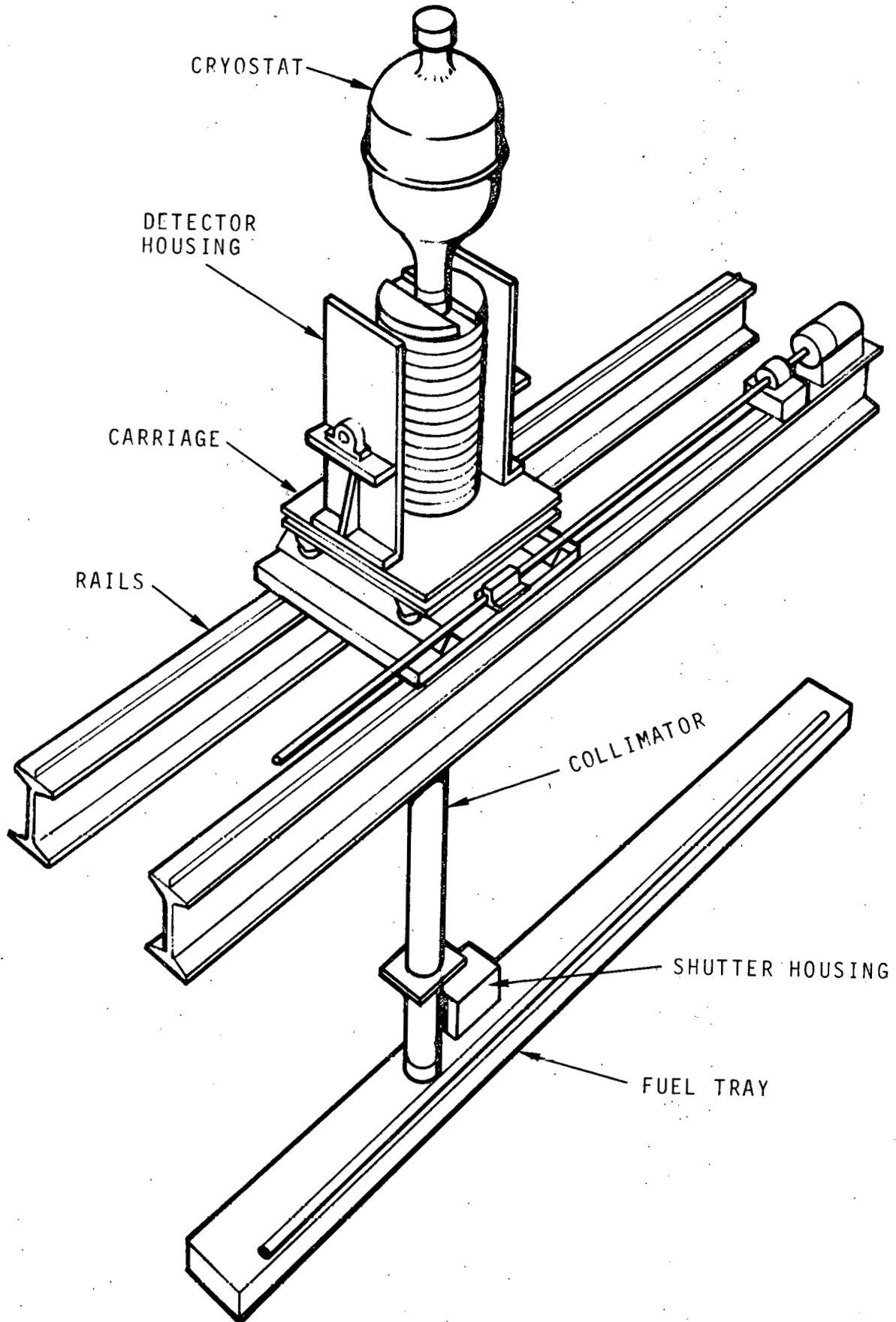
TYPICAL GAMMA RAY SPECTRUM FROM THE YANKEE FUEL
(COOLING TIME = 5.5 YEARS)

YANKEE SEGMENT I-4

SPECTRUM NO.   I-4B
LIVE TIME   1200MIN.
DATA TAKEN   10/5/70
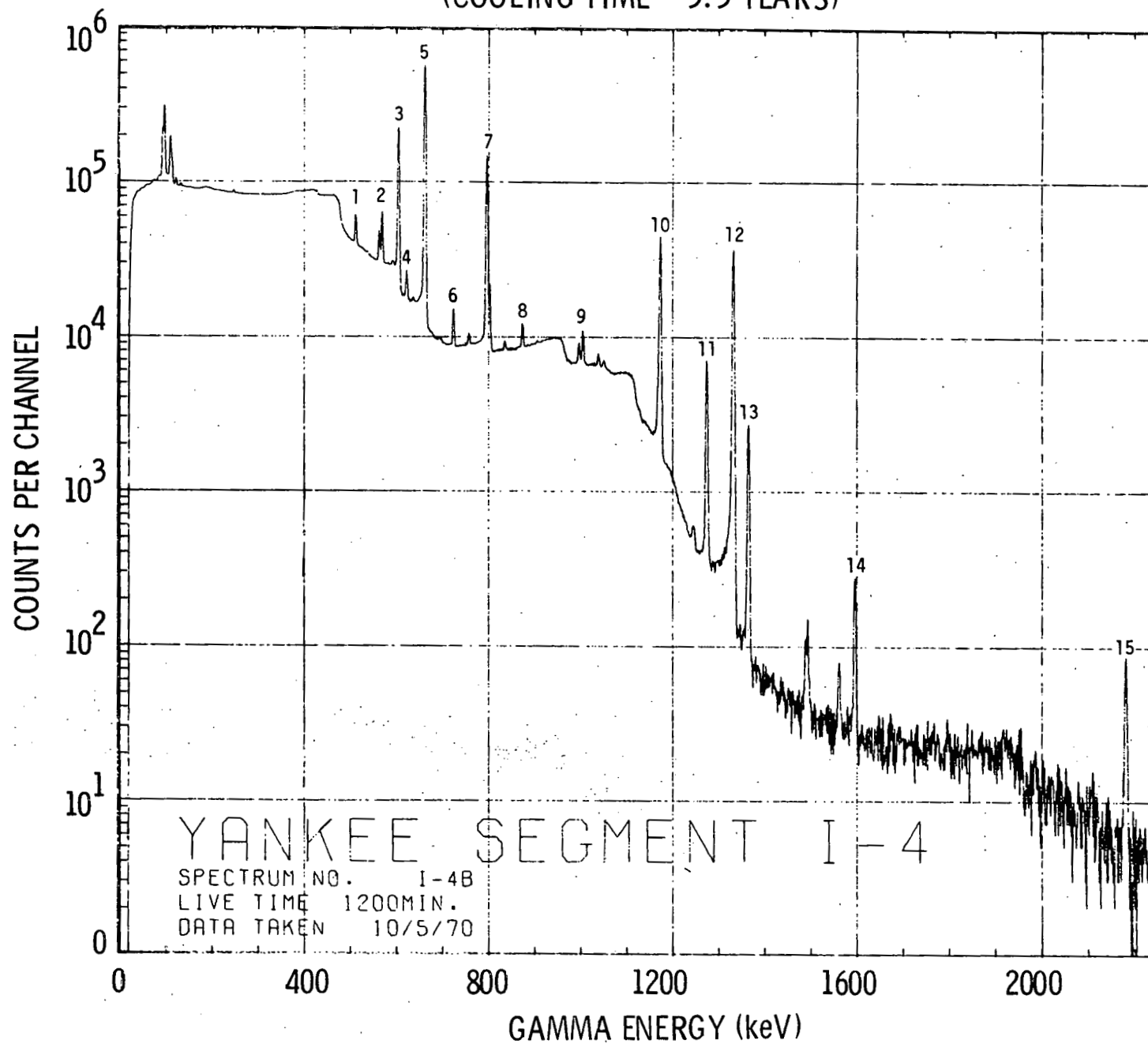
PHOTO-PEAK IDENTIFICATION
FOR YANKEE SPECTRUM I-4 B

PEAK NO.	ENERGY (KeV)	SOURCE
1	512	$^{106}Ru/^{106}Rh$
2	564, 569	^{134}Cs
3	605	^{134}Cs
4	622	$^{106}Ru/^{106}Rh$
5	662	^{137}Cs
6	723	^{154}Eu
7	796, 802	^{134}Cs
8	873	^{154}Eu
9	996, 1005	^{154}Eu
10	1173	^{60}Co (BACKGROUND)
11	1275	^{154}Eu
12	1332	^{60}Co (BACKGROUND)
13	1365	^{134}Cs
14	1594	^{154}Eu
15	2186	^{144}Ce

PROGRAM DRAFT ✳ YANKEE CORES 1,2 AND 4 FOR ACDA STUDY

Plot axes:
- Y-axis: RU-106/CS-137, with gridlines at 10^0 and 10^{-1}
- X-axis: TIME (SECONDS), $* 10^6$, values 0.00, 100.00, 200.00, 300.00, 400.00, 500.00, 600.00

Legend:
- $\Phi = 5 \times 10^{14}$ n/cm^{2}sec
- $\Phi = 3 \times 10^{14}$ n/cm^2/sec
- $\Phi = 1 \times 10^{14}$ n/cm^2/sec
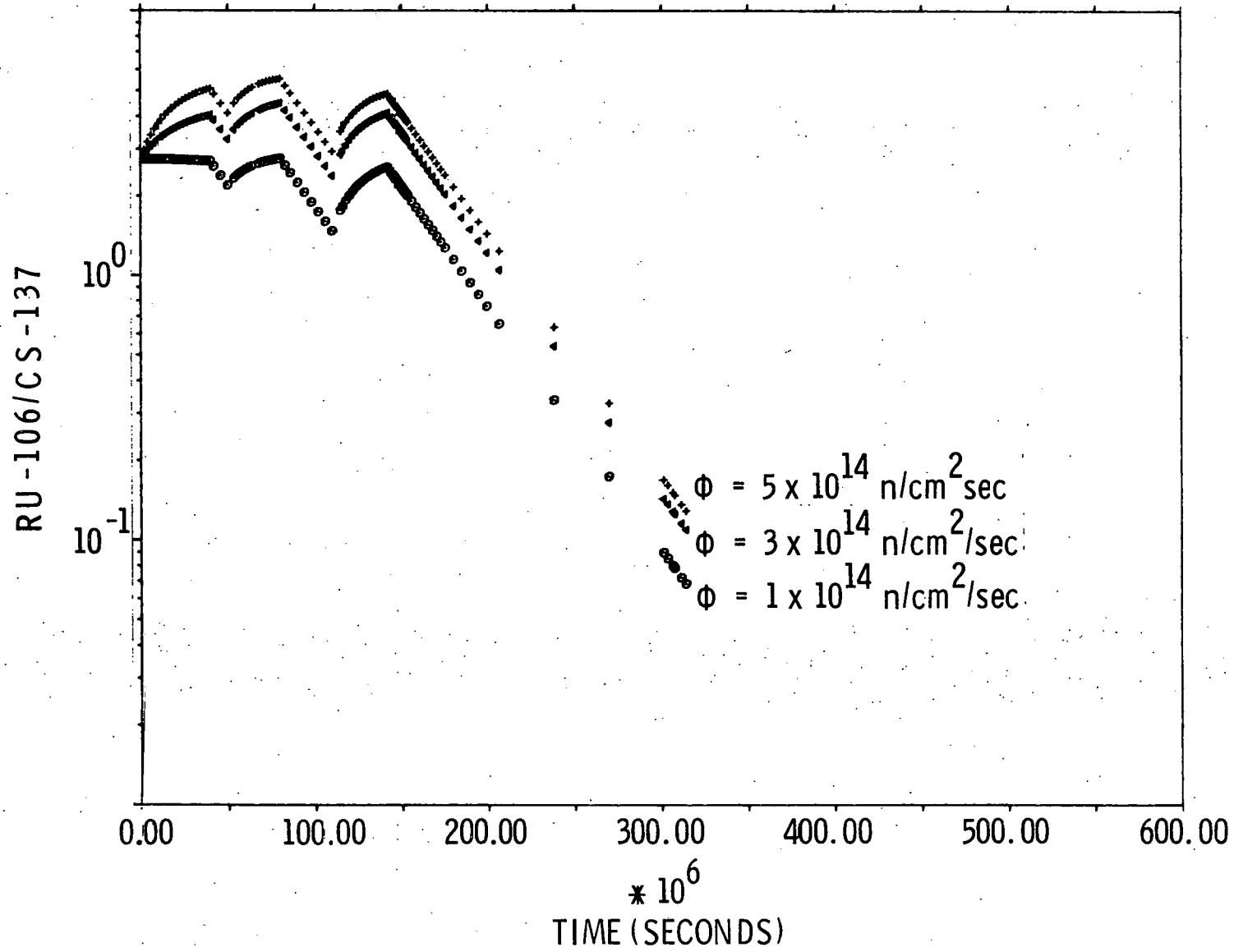
TABLE I

ABSOLUTE NEUTRON FLUX DETERMINED BY
NON-DESTRUCTIVE AND DESTRUCTIVE METHODS

	Absolute Flux* (10^{14} n/cm^2/sec)	
Method	Inner Rod	Outer Rod
Non-destructive Measurements		
^{106}Ru/^{137}Cs Activity Ratio	2.94 ± 0.06[1]	2.75 ± 0.06[1]
	2.00 ± 0.06[2]	1.70 ± 0.06[2]
^{144}Ce/^{137}Cs Activity Ratio	$3.9 \; ^{+2.0}_{-1.4}$	$5.4 \; ^{+2.6}_{-1.9}$
^{134}Cs/^{137}Cs Activity Ratio	3.99 ± 0.15	3.35 ± 0.06
^{154}Eu/^{137}Cs Activity Ratio	$\ll 1.0$	$\ll 1.0$
Destructive Measurements		
^{148}Nd Content	3.87 ± 0.07	3.73 ± 0.07
^{137}Cs Activity	3.98 ± 0.09	3.86 ± 0.09

*Uncertainties quoted are standard deviations propagated from uncertainties in measured concentrations or activities. In the latter case only random statistical errors were considered.

(1) 512 KeV Photopeak

(2) 622 KeV Photopeak

ABSOLUTE NEUTRON FLUX DETERMINED BY NONDESTRUCTIVE AND DESTRUCTIVE METHODS

METHOD	ABSOLUTE FLUX (10^{14} n/cm^2/sec)	
	INNER ROD	OUTER ROD
NONDESTRUCTIVE MEASUREMENTS		
^{106}Ru/^{137}Cs ACTIVITY RATIO	2.94 ± 0.06[1]	2.75 ± 0.06[1]
	2.00 ± 0.06[2]	1.70 ± 0.06[2]
^{144}Ce/^{137}Cs ACTIVITY RATIO	$3.9\ ^{+\ 2.0}_{-\ 1.4}$	$5.4\ ^{+\ 2.6}_{-\ 1.9}$
^{134}Cs/^{137}Cs ACTIVITY RATIO	3.99 ± 0.15	3.35 ± 0.06
^{154}Eu/^{137}Cs ACTIVITY RATIO	$\ll 1.0$	$\ll 1.0$
DESTRUCTIVE MEASUREMENTS		
^{148}Nd CONTENT	3.87 ± 0.07	3.73 ± 0.07
^{137}Cs ACTIVITY	3.98 ± 0.09	3.86 ± 0.09

(1) 512 KeV PHOTOPEAK
(2) 622 KeV PHOTOPEAK

measurements which have been averaged from one rod designated as an outer rod. These results were compared to results obtained using destructive measurements from cut fuel samples that were taken from rod positions adjacent to and in between the two gamma ray measurement positions on each rod. The samples were chemically analyzed for isotopic and gamma activity burnup data. With the destructive measurements for ^{148}Nd content and ^{137}Cs activity as a basis, flux values were then determined by comparing destructively measured values and calculated values from program DRAFT. These values are shown in Table I.

The general agreement between fluxes as determined using non-destructive and destructive measurements is good since the discrepancies observed, although large in some cases, resulted from variables extraneous to measurement variations. The measured gamma ray spectra data which were integrated to obtain photopeak areas from which the activity ratios were then determined, were found to be consistent and generally exhibited only the expected small random variations. The variations seen in Table I resulted from the step in the procedure of determining corresponding flux values. Limiting variables include the following:

a. Poorly known constants such as capture cross sections (particularly in the case of ^{133}Cs, ^{134}Cs, ^{153}Eu and ^{154}Eu), branching ratios (for ^{106}Ru-^{106}Rh) and fission yields which are a necessary part of the calculations. The ^{241}Pu fission contributions have not been included in program DRAFT, resulting in a discrepancy for ^{106}Ru production. The fission yield of ^{106}Ru from ^{241}Pu is greater than that from ^{239}Pu which itself is 11 times greater than the fission yield from ^{235}U.

b. Lack of consistent cross section generation techniques for the fission product isotopes involved.

c. Low activity in the case of ^{144}Ce due to 5.5 year cooling time such that an eight percent counting uncertainty in the flux values. The uncertainty was large due to the necessity to match measured and calculated ratios in a region where the calculated ratio had little variation with flux.

As the limiting factors of a. and b. are overcome, non-destructive gamma scanning techniques to determine burnup of shorter cooled fuel will be increasingly more reliable. This is due to the fact that the step of converting gamma ray activity values to flux values is subject to uncertainty moreso than any other step in the procedure. The last step in the procedure is to convert flux values to corresponding burnup values. This step is not hindered by poorly known nuclear constants and cross sections. To illustrate this, flux values as determined using measured ^{137}Cs activity and ^{148}Nd content have been utilized to determine the corresponding burnup from calculations of program DRAFT. The calculated burnup values are presented in Table II along with measured burnup values determined using both ^{137}Cs and ^{148}Nd. The values in Table VII compare favorably indicating that the technical problem to be resolved lies with the factors a. and b. The fission product measurements that are presently possible with high resolution spectrometers are apparently adequate at least for fuel which has not cooled for a substantial period of time.

A second consideration is that to date determinations have concentrated on point burnup. While this is adequate for monitoring fuel, it is necessary to determine how one extrapolates point burnup determinations to the average fuel bundle exposure, in order to relate the information to that from the thermal processing.

The analysis of the Yankee fuel indicated that the measurement itself was quite easy to perform and that even though one was handicapped by longer cooler time the results were very encouraging. A significant part of this analysis was that the nondestructive determinations were compared to destructive determinations of burnup for the fuel rods. This represents a very unique analysis in that respect.

TABLE II

BURNUP VALUES FROM PROGRAM DRAFT
COMPARED TO MEASURED VALUES

The Values from Program DRAFT Correspond to
Flux Values as Determined from Measured ^{137}Cs and ^{148}Nd.

Inner Rod				Outer Rod			
^{137}Cs		^{148}Nd		^{137}Cs		^{148}Nd	
Flux (x 10^{14} n/cm^2/sec)							
3.98		3.87		3.86		3.73	
Burnup (MWd/MTM)							
DRAFT	Measured	DRAFT	Measured	DRAFT	Measured	DRAFT	Measured
38,552	38,283	37,593	37,110	37,494	35,375	36,342	35,777

CONCLUSIONS AND RECOMMENDATIONS

Results from the non-destructive determination of burnup indicated that
the state of the art for obtaining high resolution gamma ray spectra and
determining reliable photopeak areas from these data is further advanced than
the state of the art on known fission product constants, i.e., cross sections
and branching ratios necessary to transform photopeak area values to flux values.
The subsequent step of determining corresponding burnup values appeared to be
straightforward and indicated that the fuel isotope cross sections and other
constants necessary to calculate burnup are adequately known. Thus, the
feasibility of non-destructive measurements depends to a large degree on whether
or not fission product constants are known accurately. However, the potential
utility of the gamma scanning technique was demonstrated by the ease with which
measurements were made. It is recommended that the next phase demonstrating
the utility of the non-destructive technique be to measure irradiated fuel at

BURNUP VALUES FROM PROGRAM DRAFT
COMPARED TO MEASURED VALUES

INNER ROD		OUTER ROD	
^{137}Cs	^{148}Nd	^{137}Cs	^{148}Nd
FLUX (x 10^{14} n/cm^2/sec)			
3.98	3.87	3.86	3.73
BURNUP (MWd/MTM)			
DRAFT	DRAFT	DRAFT	DRAFT
38,552	37,593	37,494	36,342
MEASURED	MEASURED	MEASURED	MEASURED
38,283	37,110	35,375	35,777

THE VALUES FROM PROGRAM DRAFT CORRESPOND
TO FLUX VALUES AS DETERMINED FROM
MEASURED ^{137}Cs AND ^{148}Nd.

several different exposures in the cooling time range of 1 month to 1 year. Concurrently, it is recommended that more accurate fission product constants be obtained by direct measurement involving separated amounts of each fission product or be determined by an experiment which is designed to derive a consistent set of constants from high resolution fission product gamma ray spectra of irradiated fuel. Further, it is recommended that DRAFT be expanded to include ^{240}Pu, ^{241}Pu and ^{242}Pu isotopes.

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DEVELOPMENT OF AGE MONITOR AT NPD

A second research effort concerned installing a spent fuel age monitor in the spent fuel bay at the NPD reactor, Rolphton, Ontario, Canada, for the purpose of determining cooling time of NPD irradiated fuel assemblies from gamma ray measurements. The operation of the monitor is a safeguards experiment planning by the joint Canada/USA Working Group on Safeguards Instrumentation as part of the Canada/USA Tamper-resistant Unattended Safeguards Techniques Porgram.

The spent fuel monitor, which is primarily a gamma ray spectrometer, was designed, fabricated and tested by Battelle-Northwest to demonstrate the age determination of spent fuels. The demonstration was preceded by testing of the monitor by analysis of irradiated fuel from the Hanford K East Reactor (KE).

As a result of the measurements made on the KE fuel, a standard in the form of a graph of the activity ratios as a function of cooling time was derived for cooling times between 10 and 90 days. The ^{140}Ba - ^{140}La to ^{95}Zr - ^{95}Nb ratio proved to be relatively insensitive to exposure thus providing a useful age index. By use of the standard it was possible to make an age determination to within an uncertainty of $\pm$ 3 days. The demonstration experiment at KE Reactor was thus successful and was the basis to proceed with a like demonstration using a sodium iodide detector at NPD. Results to date at NPD indicate that the definition of cooling time should include the time an element may reside in a zero flux position while in the NPD reactor.

The results of these analyses indicate good reason for optimism that gamma scanning can indeed provide a useful redundant safeguards technique

for determination of plutonium content of irradiated reactor fuel. We
will now discuss how one would implement such a technique into the safe-
guards system.

IMPLEMENTATION OF GAMMA RAY SPECTROMETRY AS
A SUPPLEMENTARY MIST TECHNIQUE

Development of the fundamental method and equipment needed to utilize gamma ray spectrometry as a safeguards tool has progressed to the point that highly automated, transportable assay systems are presently feasible. Several areas in the fuel cycle are especially appropriate for the utilization of this nondestructive method of material assay. These are: the output of the fuel fabrication plant; the receipt of fuel assemblies at the reactor site; the shipment of the irradiated fuel from the reactor site; and the input of the irradiated fuel into the fuel reprocessing plant. In all these cases the fissile material is contained within cladding material and is not amenable to any type of destructive analyses.

To demonstrate the feasibility of this technique, it is recommended that a prototype measuring system be developed for use by inspectors at either a reactor site or a fuel reprocessing plant. The aim is to demonstrate the feasibility of measuring plutonium content using a calibrated gamma scanning system. The measurement would thus provide a safeguards team with a means of ascertaining the resonableness of reported values for the nuclear materials content of spent fuel by examination of fission product gamma ray spectra emitted by the spent fuel.

The hardware system required is essentially well known and development of it could be initiated immediately. The development work required would be to provide the software systems required to analyze the data provided by the gamma scanning hardware system and ultimately to determine the fissile material content. Typically computer programs are required to analyze the experimental data and to provide the required calculational data which predicts the activity ratios. The inspector cannot be expected

to do much more than operate the equipment and recognize malfunctioning, if and when they occur.

Initially, it is recommended that the digital computer portion of a safeguards system be deleted and that the information obtained from the detector system be channeled via teletype to a computer system analysis center. For instance, the data could be obtained by the inspector via the experimental equipment and the data transmitted via teletype to an analysis team at some distant point where the required software computer programs exist. The analysis center would then process the data, determine the resultant conclusion and transmit these via teletype back to the inspector. This entire process could be performed quite quickly via a teletype system.

While the hardware system is being constructed, development work on the technique could continue by analyses of data which already exist. There is a significant inventory of nondestructive and destructive data on various types of reactor fuel in existance and this data could be used to further develop the technique or techniques. The idea is to determine the degree of information which is required to determine fuel burnup to the optimum accuracy. Such questions as whether or not one needs to know the discharge date of the fuel or certain information about reactor types is yet to be ascertained. After the prototype system is fully constructed one could then begin to analyze fuel at a reprocessing plant and continue the sensitivity analyses to determine the extent of information required about the reactor type in order to obtain the 5% accuracy on fuel burnup. It is envisioned that approximately two years would be required to develop and prove the feasibility of the gamma ray technique.