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DETERMINING INITIAL COMPOSITION OF REGENERATING IN-CORE NEUTRON FLUX DETECTORS

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DETERMINING INITIAL COMPOSITION OF
REGENERATING IN-CORE NEUTRON FLUX DETECTORS

By

W. L. BUNCH

Nucleonic Instrumentation
Instrument Research and Development
Physics and Instrument Laboratories
HANFORD LABORATORIES

September 11, 1963

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DETERMINING INITIAL COMPOSITION OF
REGENERATING IN-CORE NEUTRON FLUX DETECTORS

By W. L. Bunch

INTRODUCTION

In large reactors there is usually an incentive to operate each portion of the reactor at the maximum tolerable specific heat generation rate in order to efficiently utilize the fuel and plant capabilities to minimize the cost of producing electricity, isotopes, desalinized water, heat, and so on. Measurement of reactor power distribution to provide control information has been attempted in a number of ways; none has been entirely satisfactory. Wire activation techniques can be used to measure neutron flux distribution; however, such methods do not provide continuous information as is desirable for control purposes. Gamma sensitive chambers are vitiated by the residual activity which exists. Continuous monitoring of the neutron flux appears to be the most appropriate method of obtaining the desired distribution; however, an inherent disadvantage is the decrease in sensitivity due to transmutation of the detecting isotope by neutron absorption. The use of regenerating (Phoenix) detectors is one feasible method of extending the useful lifetime of sensitive in-core detectors located in relatively high flux environments. Regenerating detectors incorporate a fertile isotope in the sensing element; the fertile isotope is converted into a fissile isotope at approximately the same rate as the original fissile isotope is destroyed during continued exposure. The initial ratio of isotopes used in a given detector is dependent on the neutron flux environment in which it will reside. An experimental-calculational technique has been developed for determining the initial composition of the detectors to obtain a maximum useful lifetime in the reactor core. An experiment to determine the precision of the described technique is in progress.

SUMMARY

A computer program was written to calculate the initial ratio of isotopes which should be used in an in-core neutron flux detector to obtain the maximum useful lifetime where the useful lifetime is defined as that period of time during which the sensitivity of the detector remains within specified limits. Input data for the program include cross sections of the isotopes to be used. In order that various reactor environments might be considered, Westcott cross sections² have been employed. Westcott cross sections assume that the neutron spectrum in a thermal reactor can be characterized by the temperature, T , and by a spectral parameter, r . An experimental technique was developed for measuring these parameters. The evaluation of the parameter, r , is based on a standard technique comparing the relative radioactivity induced in bare and cadmium-covered cobalt samples. The determination of the neutron temperature, T , was made using a mass spectrometer to determine the isotopic changes in uranium and plutonium samples as a function of exposure in the neutron flux. This is a new technique which appears to be both simple and accurate. The irradiations were made in a water-cooled irradiation facility located in the KE Hanford production reactor. Based on the results of the experimental data obtained, twelve regenerating detector elements were encapsulated and are currently undergoing irradiation in the facility to determine how precisely the calculated behavior pattern can be achieved.

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DISCUSSION

A. Mathematical Basis

A regenerating detector could incorporate any number of fertile and fissile isotopes to achieve the desired characteristics. For mathematical simplicity, it was assumed that a regenerating detector would be composed of no more than three isotopes in a series, the first and third being fissionable and the middle isotope being the fertile material. The buildup and burnout of these isotopes is given by a set of differential equations

$$\frac{dN_i(t)}{dt} = \sum_{j=1}^3 C_{ij} N_j(t) \quad i = 1, 2, 3$$

where the N_i 's refer to the number of nuclei of that isotope in the detector and the C_{ij} 's are appropriate combinations of the neutron flux, ϕ , cross sections, σ , and decay constants, λ . In matrix notation

$$(C) = \begin{pmatrix} -\phi(\sigma_{12} + \sigma_{1f}) - \lambda_1 & 0 & 0 \\ \phi\sigma_{12} & -\phi\sigma_{23} - \lambda_2 & 0 \\ 0 & \phi\sigma_{23} & -\phi(\sigma_{34} + \sigma_{3f}) - \lambda_3 \end{pmatrix}$$

The first subscript refers to the isotope; for the cross sections, the second subscript indicates either conversion to the next isotope or fission. It is necessary that the decay constants be in compatible reciprocal time units. The solution of these differential equations can be written as

$$N_i(t) = \sum_{j=1}^3 \left[M_{ij} \sum_{k=1}^3 M_{jk}^{-1} N_k(t) \right] e^{v_j t} \quad i = 1, 2, 3$$

where the elements of the matrix, M , its inverse, M^{-1} , and the v_j 's are appropriate combinations of the elements of the C matrix. Symbolically, the relations are

$$\begin{aligned} N_1(t) &= N_1(0) e^{C_{11}t} \\ N_2(t) &= \left[\frac{C_{21}}{C_{11} - C_{22}} N_1(0) \right] e^{C_{11}t} + \left[N_2(0) - \frac{C_{21}}{C_{11} - C_{22}} N_1(0) \right] e^{C_{22}t} \\ N_3(t) &= \left[\frac{-C_{32}C_{21}}{(C_{33} - C_{11})(C_{11} - C_{22})} N_1(0) \right] e^{C_{11}t} + \left[\frac{-C_{32}C_{21}}{(C_{11} - C_{22})(C_{22} - C_{33})} N_1(0) + \frac{C_{32}}{C_{22} - C_{33}} N_2(0) \right] e^{C_{22}t} \\ &\quad + \left[-\frac{C_{32}C_{21}}{(C_{22} - C_{33})(C_{33} - C_{11})} N_1(0) - \frac{C_{32}}{C_{22} - C_{33}} N_2(0) + N_3(0) \right] e^{C_{33}t} \end{aligned}$$

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The fission rate of the detector per unit flux at any time depends on the number of fissile atoms which are present. As the equations show, the number of atoms of fissile material present in the detector as a function of time in a given environment is dependent only on the initial concentrations of the isotopes. The fission rate per unit flux or sensitivity is

$$S(N_1(0), t) = \sigma_{1f} N_1(t) + \sigma_{3f} N_3(t)$$

In a regenerating detector, it is desired that this fission rate be as nearly constant with time as possible. One method of achieving this result is to require that the square of the area between a constant median fission rate and the actual fission rate per unit flux be a minimum. Let F be the median fission rate. The area squared to an arbitrary fixed time, τ , is

$$Q = \int_0^{\tau} [S(N_1(0), t) - F]^2 dt$$

The partial derivatives of Q with respect to the $N_1(0)$'s are set equal to zero and solved simultaneously to determine the initial quantity of each isotope which should be utilized. The details of this solution are given in Reference 3. In matrix notation the solutions can be written

$$N_i(0) = F \sum_{j=1}^3 X_{ij} u_j \quad i = 1, 2, 3$$

where the elements of the matrix X_{ij} are complicated relations involving combinations of the elements of the C matrix and the u_j 's are exponential time functions involving certain of the elements of the C matrix evaluated at the specific time, τ . These solutions are too cumbersome to present symbolically here.

A computer program was written for the IBM-7090 to do the matrix manipulations required to solve for the initial isotopic composition of a regenerating detector as a function of the various input parameters. Required input parameters are the neutron flux level, the absorption and fission cross sections for each of the isotopes, the decay constants, an incremental time unit for τ , the number of incremental time units which are to be evaluated, and the maximum deviation of the fission rate per unit flux from the median value which is to be tolerated. The program first calculates the optimum initial composition for a single incremental time unit and determines the length of time a detector with this composition would remain within the specified sensitivity limits. These calculations are repeated for τ equal to two incremental time units, etc. up to the number specified. The optimum initial composition is that in which the fission rate per unit flux remains within the specified sensitivity limits for the longest period of time. For this optimum composition, the program

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prints out the fission rate history of the detector as a function of time. Table I and Figure 1 illustrate the results of the program for a postulated environment and compare regenerating detectors with simple U-235 or B-10 detectors.

In order for burnout of the detecting element to be important, the flux level must be high enough to make the product $\phi \sigma t \geq 0.1$ in a period of time equal to or less than the desired lifetime of the detector. When ϕ is of this order of magnitude, the product $\phi \sigma_1$ is significantly greater than the corresponding decay constant, λ_1 , for the uranium and plutonium isotopes considered. Thus, the detector sensitivity or fission rate can be plotted as a function of exposure rather than time without introducing significant error (see Figure 1).

B. Cross Section Notation

In the mathematical model of the regenerating concept, it was assumed that the interaction rate could be calculated as the simple product of the neutron flux, ϕ , and the macroscopic cross section, $N\sigma$. This common practice is possible only if a compatible definition of flux and cross section is used. In general, the reaction rate or saturated activity of a sample is

$$R = N \int_0^{\infty} n(v) v \sigma(v) dv$$

where N is the number of target nuclei present in the sample, $n(v)$ is the neutron velocity distribution, $\sigma(v)$ is the cross section for the event to take place as a function of incident neutron velocity, and v is neutron velocity. The neutron flux is

$$\phi = \int_0^{\infty} n(v) v dv$$

However, the cross section which should be used with this value of the flux in order to obtain the reaction rate cannot be defined. It has been noted that in many isotopes the activation cross section is inversely proportional to the incident neutron velocity or

$$\sigma(v) = \sigma_0 v_0 / v$$

where σ_0 is the cross section at an arbitrary velocity, v_0 . For isotopes with this type of cross section, the reaction rate is

$$R = N n v_0 \sigma_0$$

where

$$n = \int_0^{\infty} n(v) dv$$

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FIGURE 1

Comparison of Detector Burnout Behavior

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TABLE I

Comparison of Various Types of Detectors

Environment: $T = 100^{\circ}\text{C}$ $r = 0.04$

Type	Isotope	Cross Section (Barns) (f or α) (v)		Original No. of Atoms $\times 10^{-17}$	Detector Weight (mg)	"Useful Lifetime" ($\pm 10\%$ Sensitivity) $\times 10^{-20}$ nvt	Maximum Sensitivity at 10^{14} nv
Boron-Alpha	B-10	3840	-	0.26	0.00043	0.52	10^{10} α /sec.
Uranium-fission	U-235	557	110	1.80	0.07	3.0	10^{10} f/sec.
Regenerating Pu-fission	Pu-239	888	409	0.93	0.127	12.04	10^{10} f/sec.
	Pu-240	-	762	2.25			
	Pu-241	1145	431	0			
Regenerating U-fission	U-233	554	58	0	0.59	50.3	10^{10} f/sec.
	U-234	-	105	13.7			
	U-235	557	110	1.48			

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is the total neutron density. The conventional thermal neutron flux is defined as

$$\phi = nv_0 \text{ where } v_0 = 2200 \text{ meters per second}$$

Westcott, Walker, and Alexander have devised a system for determining the reaction rate of isotopes whose cross sections are not inversely proportional to velocity. This system retains the use and definition of conventional thermal neutron flux. They assumed that within a well moderated, thermal reactor, the neutron velocity distribution is composed of a thermal component which can be described by a Maxwellian distribution plus an epi-thermal component which is inversely proportional to the square of the velocity.

$$n(v) = \frac{4n}{\sqrt{\pi}} \left[(1-f) \frac{v^2}{v_T^3} e^{-\left(\frac{v}{v_T}\right)^2} + f \frac{\sqrt{\pi\mu}}{4} v_T \frac{\Delta}{v^2} \right]$$

where n is the total neutron density as before, f is the fraction of the neutron density in the epi-thermal component, v_T is the most probable velocity of the neutrons in the Maxwellian component, μ is a somewhat arbitrary constant which relates the cutoff velocity of the epi-thermal component, v_{co} , to the most probable velocity of the thermal component,

$$\mu = \left(\frac{v_{co}}{v_T} \right)^2$$

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and Δ is a step function which is zero for $v < \sqrt{\mu} v_T$ and is unity for $v > \sqrt{\mu} v_T$. The distribution function has been normalized so the integral of $n(v)$ over all velocities is equal to n , the total neutron density.

Using this assumed neutron velocity distribution for $n(v)$, the reaction rate can be written:

$$R = N n v_0 \sigma_0 \left[\frac{1}{v_0 \sigma_0} \int_0^{\infty} \frac{4}{\sqrt{\pi}} \left(\frac{v}{v_T} \right)^3 e^{-\left(\frac{v}{v_T}\right)^2} \sigma(v) dv + f \frac{\sqrt{\mu} v_T}{v_0 \sigma_0} \int_{\sqrt{\mu} v_T}^{\infty} \frac{\Delta \sigma(v)}{v} dv - \frac{f}{v_0 \sigma_0} \int_0^{\infty} \frac{4}{\sqrt{\pi}} \left(\frac{v}{v_T} \right)^3 e^{-\left(\frac{v}{v_T}\right)^2} \sigma(v) dv \right]$$

where σ_0 is the 2200 meter per second cross section. If sufficient experimental cross section data have been obtained to describe $\sigma(v)$, the integrals within the brackets can be evaluated for specific values of v_T . Let

$$g(T) = \frac{1}{v_0 \sigma_0} \int_0^{\infty} \frac{4}{\sqrt{\pi}} \left(\frac{v}{v_T} \right)^3 e^{-\left(\frac{v}{v_T}\right)^2} \sigma(v) dv$$

be a weighting factor which accounts for the non- $1/v$ behavior of the cross section for neutrons with a Maxwellian velocity distribution. Additionally, let

$$r = f \sqrt{\frac{\pi \mu}{4}}$$

$$s(T) = \frac{4}{\sqrt{\pi \mu}} \left[\frac{\mu v_T}{v_0 \sigma_0} \int_0^{\infty} \frac{\Delta \sigma(v)}{v} dv - g(T) \right]$$

The product $r s(T)$ becomes a weighting factor which accounts for the non- $1/v$ behavior of the cross section for a neutron distribution inversely proportional to the square of the velocity and in addition corrects for the fact all of the neutrons are not in the Maxwellian portion of the spectrum unless $r = 0$. The cross section for use with the conventional thermal neutron flux, $nv_0 = \phi$ becomes

$$\sigma = \sigma_0 \left[g(T) + r s(T) \right]$$

In order that calculated values of $g(T)$ and $s(T)$ might be utilized, it is necessary to establish the neutron temperature, T , and the spectral parameter, r , in the region where the sample is to be irradiated. The calculated reaction rates will be accurate only to the extent the actual neutron velocity distribution can be described by the relation assumed in the Westcott derivation.

C. Bases for Experimental Determinations

The evaluation of r and T requires at least two independent experiments. In the method employed, it was necessary to also determine the integrated neutron exposure, ϕt ; thus, three experiments were required. Experimental evaluation of these parameters is usually done with radioactinants,^{4,5,6} but the irradiation conditions encountered in the present experiment dictated that other techniques be devised in some cases. The evaluation of the parameters ϕt and T is based on the use of the mass spectrometer to determine the change in the isotopic composition which took place during an irradiation in the unknown environment. The mathematical basis for each of the three experiments will be discussed briefly.

1. Spectral Parameter, r

The spectral parameter, r , which relates the neutron density in the thermal and epi-thermal components of the spectrum, can be determined experimentally from the relative radioactivity of appropriate samples irradiated bare and with cadmium covers. Cadmium has a large cross section for thermal and low velocity neutrons and a relatively small absorption cross section for higher velocity neutrons. The transition from the large to the small cross section occurs rather abruptly at about 0.4 ev; therefore, a cadmium cover of appropriate thickness will absorb virtually all neutrons incident upon it with less than this energy and will transmit virtually all neutrons with more than this energy. Even though there is a transition energy region, a cadmium cutoff velocity can be defined such that transmission above

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that velocity is compensated by attenuation below that velocity. The effective cadmium cutoff velocity is a function of the thickness of the cadmium cover.

As defined previously, the reaction rate in a bare sample, which is proportional to the activity induced in the sample, is given as

$$R = N n v_0 \sigma_0 (g + r s)$$

If an appropriate detector is used such as Co-59, whose cross section to thermal and low energy neutrons is inversely proportional to velocity so $g(T) = 1$, the relation becomes

$$R = N n v_0 \sigma_0 (1 + r s)$$

If the Co-59 is covered with cadmium, the reaction rate becomes

$$R_{cd} = N n v_0 \sigma_0 + \frac{4}{\sqrt{\pi}} \frac{N_T}{N_0 \sigma_0} \int_{v_{cd}}^{\infty} \frac{\sigma(v)}{v} dv$$

where Δ has been replaced by unity since the integral is evaluated only at velocities greater than v_{cd} . In this expression the terms involving the Maxwellian distribution have been dropped since it is assumed that the integral of the Maxwellian from zero to v_{cd} is zero because of the cadmium cover and that the integral from v_{cd} to infinity is small enough to be insignificant. The thickness of the cadmium cover is such that the cadmium cutoff velocity, v_{cd} , is greater than the epi-thermal spectrum cutoff velocity, v_{co} ; therefore, the integral in the equation for R_{cd} can be written as the difference of the two integrals

$$\int_{v_{cd}}^{\infty} \frac{\sigma(v)}{v} dv = \int_{v_{co}}^{\infty} \frac{\sigma(v)}{v} dv - \int_{v_{co}}^{v_{cd}} \frac{\sigma(v)}{v} dv$$

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In addition, the same integral can be added to and subtracted from an equation without affecting the equality; thus, the reaction rate for a cadmium covered sample can be written as

$$R_{cd} = N n v_0 \sigma_0 + \left[\frac{4}{\sqrt{\pi}} \frac{N_T}{N_0 \sigma_0} \int_{v_{co}}^{\infty} \frac{\sigma(v)}{v} dv - \frac{4}{\sqrt{\pi}} \frac{N_T}{N_0 \sigma_0} \int_{v_{co}}^{v_{cd}} \frac{\sigma(v)}{v} dv + \frac{4}{\sqrt{\pi}} \frac{N_T}{N_0 \sigma_0} \int_0^{\infty} \left(\frac{N}{N_T} \right)^3 e^{-\left(\frac{N}{N_T} \right)^2} \sigma(v) dv - \frac{4}{\sqrt{\pi}} \frac{N_T}{N_0 \sigma_0} \int_{v_{co}}^{v_{cd}} \frac{\sigma(v)}{v} dv \right]$$

The first two terms of this relation are equal to $s(T)$ as defined previously. Under the assumption that the cross section of cobalt is inversely proportional to velocity at low energies, the third term in the brackets becomes

$$\frac{4}{\sqrt{\pi} \mu} = \frac{4 N_T}{\sqrt{\pi} N_{Co}}$$

Likewise, the fourth integral can be evaluated readily using this same cross section assumption

$$\frac{4}{\sqrt{\pi} N_0 \sigma_0} \int_{N_{Co}}^{N_{Cd}} \frac{\sigma(v)}{v} dv = \frac{4 N_T}{\sqrt{\pi}} \int_{N_{Co}}^{N_{Cd}} \frac{dv}{v^2} = \frac{4 N_T}{\sqrt{\pi} N_{Co}} - \frac{4 N_T}{\sqrt{\pi} N_{Cd}}$$

The reaction rate for a cadmium covered cobalt sample becomes

$$\begin{aligned} R_{Cd} &= N m v_0 \sigma_0 r \left[s(T) + \frac{4 N_T}{\sqrt{\pi} N_{Co}} - \frac{4 N_T}{\sqrt{\pi} N_{Co}} + \frac{4 N_T}{\sqrt{\pi} N_{Cd}} \right] \\ &= N m v_0 \sigma_0 r \left[s(T) + \frac{1}{\frac{\sqrt{\pi}}{4} \frac{N_{Cd}}{N_0}} \frac{N_T}{N_0} \right] \\ &= N m v_0 \sigma_0 r \left[s(T) + \frac{1}{K} \sqrt{\frac{T}{T_0}} \right] \end{aligned}$$

The factor K , which is proportional to the ratio v_{Cd}/v_0 , is a function of the thickness of the cadmium cover and relates the cadmium cutoff velocity to the base velocity of 2200 meters per second. Values of K for various thicknesses of cadmium have been calculated by Westcott, et al. The cadmium ratio is defined as the ratio of the activity or reaction rate of a bare to a cadmium-covered sample. For a cobalt sample,

$$C_R = \frac{R}{R_{Cd}} = \frac{1 + r s(T)}{r \left[s(T) + \frac{1}{K} \sqrt{\frac{T}{T_0}} \right]}$$

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Therefore, the spectral parameter, r , can be obtained from the cadmium ratio if the temperature, T , is known.

$$r = \frac{1}{C_R \left[s(T) + \frac{1}{K} \sqrt{\frac{T}{T_0}} \right] - s(T)}$$

The uncertainty in the value of r will be dependent both on the accuracy of the determination of the cadmium ratio and the neutron temperature.

2. Integrated Exposure, ϕt

A common method of determining the conventional thermal neutron flux or the integrated neutron exposure is to measure the radioactivity of an isotope induced in the sample. The accuracy of such a determination is limited by the absolute calibration of the detection system, by the uncertainty in the size (weight) of the sample, and by perturbations in the buildup-decay relations if $\phi \sigma \gg \lambda$ during the irradiation. The use of the mass spectrometer to analyze the change in isotopic ratio caused by the irradiation circumvents these limitations if

$$\int \phi \sigma dt \gg \int \lambda dt$$

for the sample. U-235 was found to be a good isotope for these irradiations because it met this requirement. In addition, its cross section is relatively independent of the temperature and hardness of the neutron spectrum (i.e., $g \approx 1.0$ and $s \approx 0$). The amount of U-235 in a sample as a function of exposure to conventional thermal neutron flux is

$$U_1(t) = U_1(0) e^{-\phi \sigma_1 t}$$

where $U_1(0)$ is the number of U-235 atoms present in the sample prior to the irradiation, σ_1 is the total neutron absorption cross section of U-235, and t is the exposure time in seconds. This equation assumes that the sample contains no U-234 and that $\phi \sigma_1 \gg \lambda_1$. The amount of U-236 in the sample as a function of exposure to conventional thermal neutron flux is

$$U_2(t) = \frac{\sigma_{12}}{\sigma_2 - \sigma_1} U_1(0) e^{-\phi \sigma_1 t} + \left[U_2(0) - \frac{\sigma_{12}}{\sigma_2 - \sigma_1} U_1(0) \right] e^{-\phi \sigma_2 t}$$

where σ_{12} is the capture cross section for the conversion of U-235 to U-236, σ_2 is the total absorption cross section of U-236, and $U_2(0)$ is the number of atoms of U-236 present in the sample prior to the irradiation. The ratio of U-236 to U-235 in the sample as a function of exposure can be obtained by dividing the second equation by the first.

When the ratio is simplified, the resultant equation becomes:

$$\frac{U_2(t)}{U_1(t)} = \frac{\sigma_{12}}{\sigma_1 - \sigma_2} \left[e^{(\sigma_1 - \sigma_2) \phi t} - 1 \right] + \frac{U_2(0)}{U_1(0)} e^{(\sigma_1 - \sigma_2) \phi t}$$

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If possible, a sample of uranium should be used which contains no U-236 initially; this would eliminate the second term in the expression. The cross section parameters for U-236 have not been investigated thoroughly; however, the U-236 cross section is small compared to that of U-235 so little error is introduced by the uncertainty in σ_2 .

Mass spectrometers are sensitive devices for measuring the relative isotopic abundance in samples; it is relatively easy to measure one part in a thousand. Since the ratio can be measured either way, a uranium sample could be evaluated from one part U-236 in a thousand parts of U-235 at the one extreme to a thousand parts of U-236 to one part U-235 at the other extreme. This provides a wide range in exposure which can be measured quite accurately by this method. Figure 2 is a curve showing the ratio of U-236 to U-235 as a function of conventional thermal neutron exposure under the assumption the sample contained no U-236 prior to the irradiation. The thickness of the line encompasses a neutron temperature range from 60°C to 100°C and a spectral parameter, r , from 0.030 to 0.034. For greater precision the variation in cross section with these parameters would need to be considered.

3. Neutron Temperature, T

The effective temperature of the Maxwellian portion of the neutron spectrum can be obtained by irradiating an isotope whose cross section is quite dependent on temperature. For example, the relative fission activity of Pu-239 and U-235 has been utilized as a measure of the temperature, since the cross section of Pu-239 is quite temperature dependent.⁷ Similarly, the relative radioactivity of Lu-177 and Lu-176mm has been used for this purpose.⁸ The use of the relative radioactivity of paired samples requires careful counting techniques and normalizing processes to account for differences in the samples, the decay spectra, and the half lives. In the present experiment, the mass spectrometer was used to measure the change in isotopic ratio which occurred during the irradiation. This technique circumvents many of the problems associated with the radioactive method.

As in the case of the uranium ratio, it can be shown that the ratio of Pu-240 to Pu-239 as a function of exposure is given by:

$$\frac{P_2(t)}{P_1(t)} = \frac{\sigma_{12}}{\sigma_1 - \sigma_2} \left[e^{(\sigma_1 - \sigma_2)\phi t} - 1 \right] + \frac{P_2(0)}{P_1(0)} e^{(\sigma_1 - \sigma_2)\phi t}$$

where P_2 refers to the number of atoms of Pu-240, P_1 refers to the number of atoms of Pu-239, and the cross section subscripts have similar meanings as in the preceding section but for plutonium rather than uranium isotopes. The plutonium cross sections in this relation are all temperature dependent and in addition are somewhat dependent on the spectral parameter, r . A preliminary estimate of the neutron temperature can be made using the following technique.

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FIGURE 2

Ratio U^{236}/U^{235} as a Function of Integrated Exposure to Conventional Thermal Neutron Flux

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Define

$$U = \frac{U_2(t)}{U_1(t)} - \frac{U_2(o)}{U_1(o)} e^{(\sigma_1 - \sigma_2)\phi t} = \frac{\sigma_{12}}{\sigma_1 - \sigma_2} \left[e^{(\sigma_1 - \sigma_2)\phi t} - 1 \right]$$

and

$$P = \frac{P_2(t)}{P_1(t)} - \frac{P_2(o)}{P_1(o)} e^{(\sigma_1 - \sigma_2)\phi t} = \frac{\sigma_{12}}{\sigma_1 - \sigma_2} \left[e^{(\sigma_1 - \sigma_2)\phi t} - 1 \right]$$

where the cross sections in the first relation refer to the uranium isotopes and those in the second relation refer to plutonium isotopes. For integrated neutron exposures which are small, the exponential in each case is approximately unity and the exponential minus one is approximately equal to the exponent. Thus, U is approximately the change in the uranium ratio effected by the irradiation and P is approximately the change in the plutonium ratio. In addition,

$$\frac{U}{P} \approx \frac{\sigma_{12}(U-235)}{\sigma_{12}(Pu-239)}$$

The ratio of these cross sections is plotted in Figure 3 as a function of temperature for three values of the spectral parameter, r. Given the mass spectrometer evaluations of the uranium and plutonium isotopic ratios before and after the irradiation, these curves can be used to quickly estimate the temperature of the neutron flux in which the samples were irradiated.

The final experimental values of r, ϕt , and T can be obtained by an iterative process using the data from each of the three irradiations. The neutron temperature is obtained from the relation for $P_2(t)/P_1(t)$ which is a function of the cross sections and the integrated exposure. Figures 4 and 5 show working curves which relate the cross sections and cross section ratios to the neutron temperature and the spectral parameter, r. These curves were generated for this specific experiment and encompass only the region of interest in this irradiation. The accuracy which can be achieved is limited by the uncertainty in the mass spectrometer determinations. Uncertainties in the evaluation of r and ϕt will also be reflected into the value of T.

D. Test Procedure

The irradiations were made in a water-cooled facility located in Test Opening 2B in the KE Hanford production reactor. A cross sectional view of this facility, known as a "magazine facility", is shown in Figure 6. The thimble, which is an aluminum tube 3.5 inches in outside diameter with a 0.125 inch thick wall, penetrates the reactor from the experimental level side to the control rod side and contains the single pass cooling water. Within this thimble are seven aluminum tubes each 0.76 inch outside diameter with 0.0625

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$\sigma_{12}(U^{235})/\sigma_{12}(Pu^{239})$

FIGURE 3

Neutron Temperature vs $\sigma_{12}(U^{235})/\sigma_{12}(Pu^{239})$

FIGURE 4

$\left(\frac{\sigma_{12}}{\sigma_1 - \sigma_2} \right)$ for Plutonium Isotopes vs r for Various Values of T

FIGURE 5
 $(\sigma_1 - \sigma_2)$ for Plutonium Isotopes vs r for Various Values of T

FIGURE 6

Cross Section of Magazine Facility

inch walls which contain the samples to be irradiated. The sample tubes penetrate entirely through the reactor and can be entered when the reactor is shut down without disrupting the coolant flow to the facility. For this irradiation the central sample tube was used and was filled with water to improve the thermal contact between the samples and the coolant. The samples were placed in 0.5 inch outside diameter aluminum containers which were sealed by a cold welding process. Samples were positioned within the containers with graphite spacers to obtain specific locations with respect to each other. All of the samples for this experiment were irradiated at the same time; it was assumed the flux level at each sample location would be the same because of the flattened distribution in that region of the reactor. These and other experimental data tend to justify this assumption.

1. Cobalt Samples

The cobalt samples were made by evaporating 99.9% cobalt onto pure (99.999%) aluminum backing plates in an evacuated system. The backing plates were 0.32 inches in diameter, approximately twenty mils thick, and weighed about sixty six milligrams each. A measured quantity of cobalt was evaporated and the thickness of the deposit was controlled by the separation distance to the backing plates. Attempts to determine the actual weight of cobalt deposited on a given backing plate by weighing the backing plate a number of times before and after the deposition process were apparently unsuccessful. The weighings indicated the weights of the planned 50 microgram deposits actually varied from 11 to 44 micrograms. The relative radioactivity of these foils did not indicate such large variations. Based on the radioactivity, it is estimated the weight of the deposit to be 24.6 ± 0.5 micrograms for the planned deposit of 50 micrograms, 42.9 micrograms for the planned deposit of 100 micrograms, and 55.8 micrograms for the planned deposit of 150 micrograms. For the cadmium ratio determinations, the actual weights are unimportant as long as each pair of foils is identical. The cadmium covers were 0.020 inch thick.

2. Uranium Samples

About 50 micrograms of U-235, free of U-236, were obtained. This was a material used for standardization of the mass spectrometer. The material was put into solution and the solution poured into a small aluminum capsule. Following drying, the small capsule was sealed. After the irradiation, the small capsule was opened and the uranium once again put into solution. A chemical separation was made to eliminate a major portion of the fission products, and the sample prepared for the mass spectrometer determination of the uranium isotopic ratios.

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3. Plutonium Sample

The most pure Pu-239 which could be obtained for these experiments contained about two parts per thousand of the isotope Pu-240. The plutonium was encapsulated and processed in a manner similar to that used for the uranium.

E. Results

1. Spectral Parameter, r

Essentially, two experiments were carried out simultaneously in determining the spectral parameter, r . In the first, the cadmium ratio, as a function of the thickness of the cobalt deposit, was measured to eliminate the effects of flux depression and self-absorption. In the second, the cadmium ratio, as a function of position within the reactor lattice, was measured to determine if any fine structure existed within the facility which would affect the behavior of a regenerating detector.

Table II summarizes the data obtained to determine the effect of the thickness of the cobalt deposit on the measured value of the cadmium ratio. On the basis of these data, it is concluded that the cadmium ratio is not dependent on the thickness of the cobalt deposit within the range of the experimental data. Calculations indicate there should be no dependence.

TABLE II

Cadmium Ratio As A Function Of Weight Of Cobalt Deposit

<u>Foil Number</u>	<u>Cover Material</u>	<u>Cover Thickness Inches</u>	<u>Cobalt Deposit Micrograms</u>	<u>Estimated Activity Micrograms</u>	<u>Dis/Sec</u>	<u>Cadmium Ratio</u>
1	Bare	-----	50	24.6	4.52×10^6	19.90
2	Cadmium	0.020	50	24.6	2.27×10^5	
6	Bare	-----	100	42.9	6.85×10^6	23.78
F	Cadmium	0.020	100	42.9	3.30×10^5	
7	Bare	-----	150	55.8	1.02×10^7	20.00
G	Cadmium	0.020	150	55.8	5.10×10^5	

Process tubes, containing natural uranium production fuel elements on a 7.5 inch square lattice array, pass by the magazine facility perpendicular to it and both above and below it. It is conceivable that the presence of these fuel elements, which are sources of fast neutrons and absorbers of thermal neutrons, could create a variation in the spectral parameter, r , within the irradiation facility. To investigate the magnitude of this variation, a set of four bare and four cadmium covered foils were irradiated to obtain ratios at one-quarter lattice unit intervals. The data from these irradiations are summarized in Table III. There is no apparent correlation between the cadmium ratio data and the position within the lattice. The data for foil number 3 seemed

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to be completely inconsistent; therefore, it was assumed that cobalt had been lost from the foil and this datum point was omitted in the analysis.

TABLE III

Cadmium Ratio As A Function Of Position Within Lattice

<u>Foil Number</u>	<u>Material</u>	<u>Cover Thickness - Inch</u>	<u>Goal Deposit Micrograms</u>	<u>Activity Dis/Sec</u>	<u>Cadmium Ratio</u>
1	Bare	-----	50	4.52×10^6	19.90
2	Cadmium	0.020	50	2.27×10^5	
3	Bare	-----	50	2.54×10^6	11.87
4	Cadmium	0.020	50	2.14×10^5	
5	Bare	-----	50	4.49×10^6	20.50
E	Cadmium	0.020	50	2.19×10^5	
C	Bare	-----	50	4.44×10^6	17.55
D	Cadmium	0.020	50	2.53×10^5	

The average value of the cadmium ratio, based on all of the data exclusive of foil number 3, is 20.35. The standard deviation of the mean was calculated to be 0.99. For this value of the cadmium ratio, the spectral parameter r can be calculated as a function of neutron temperature. For a cadmium cover thickness of 0.020 inch in an isotropic flux, the parameter K , which is related to the cover cutoff velocity, is 2.07. If it is assumed that the neutron temperature is 80°C , " s " for cobalt is 1.046 and the spectral parameter is calculated to be

$$r = 0.0322 \pm 0.0015$$

where the uncertainty is an estimated standard deviation based on the experimental uncertainty in the value of the cadmium ratio.

2. Integrated Exposure, ϕt

The uranium sample used in this irradiation contained no U-236 prior to the irradiation; hence, $U_2(0)/U_1(0) = 0$. The mass spectrometer evaluation of the ratio of U-236 to U-235 after the irradiation was 0.01276 ± 0.00008 , where the uncertainty is the estimated standard deviation in the analysis. An estimate of the thermal neutron exposure can be obtained from the curve in Figure 2. To make a more accurate evaluation it is necessary to make a calculation for specific values of r and T . On the basis of the preceding section, $r = 0.032$; assuming $T = 80^\circ\text{C}$, the relation becomes

$$0.01276 = 0.165 \left[e^{651 \phi t \cdot 10^{-24}} - 1 \right]$$

$$\phi t = 1.143 \times 10^{20} (\pm 0.007 \times 10^{20})$$

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where the uncertainty is the estimated standard deviation based on the accuracy of the mass spectrometer evaluation of the isotopic ratio.

The absolute value of the neutron flux, ϕ , in the facility can be obtained from the duration of the irradiation. From the time the reactor was half up until the time it was half down was 8.2 days or 7.08×10^5 seconds. The average value of the conventional thermal neutron flux in the magazine facility throughout the irradiation was thus $1.143 \times 10^{20} / 7.08 \times 10^5$, or

$$\phi = 1.61 \times 10^{14} \text{ neutrons/cm}^2 \text{ sec}$$

3. Neutron Temperature, T

The ratio of Pu-240 to Pu-239 in the sample used to determine the neutron temperature was measured to be 0.00163 with an estimated standard deviation of 0.00001 prior to the irradiation. Following the irradiation the ratio of the same isotopes was measured to be 0.0478 with a standard deviation of about 0.00025. A preliminary estimate of the neutron temperature can be made from the ratio of the uranium ratio to the plutonium ratio as

$$\frac{U}{P} = \frac{0.01276}{0.0478 - 0.00163} = 0.276$$

From Figure 3, this ratio of the cross sections indicates a temperature of about $85^\circ\text{C} \pm 50^\circ\text{C}$ depending on the value of r . Using the relation for $P_2(t)/P_1(t)$ and the specific values of r and ϕt quoted in the previous sections, the value of T is calculated with the aid of Figures 4 and 5 to be

$$T = 80^\circ\text{C} \pm 3^\circ\text{C}$$

where the quoted uncertainty is the standard deviation based on the mass spectrometer results for the isotopic ratio of plutonium. The estimated uncertainties in r and ϕ increase the uncertainty in T to about $\pm 5^\circ\text{C}$. This amount of uncertainty in the value of T would not increase significantly the quoted uncertainties in r and ϕt .

F. Computer Calculations

The experimental values of r and T quoted above establish the values for the plutonium cross sections for use in the computer program to calculate the optimum initial composition of regenerating detectors for irradiation in the facility. Input values for the calculation are given in Table IV along with the results.

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TABLE IV

Parameters for Optimum Regenerating Detector
for Magazine Facility

<u>Isotope</u>	<u>Cross Section, Barns</u>		<u>Decay Constant</u> <u>Seconds¹</u>	<u>Composition</u> <u>Percent</u>
	<u>Fission</u>	<u>Gamma</u>		
Pu-239	862	389	8.98×10^{-13}	27.3
Pu-240	0	661	3.24×10^{-12}	72.7
Pu-241	1124	423	1.68×10^{-9}	0

In this optimization calculation, the sensitivity of the detector was permitted to deviate from the median value by ten per cent. A time increment of one month was employed; therefore, an arbitrary neutron flux level of 3.8×10^{13} neutrons/cm² 3.8×10^{13} neutron/cm² sec was assumed in order to obtain an integrated exposure of 10^{20} neutrons/cm² per calculational month for ease in plotting. The calculated normalized fission rate of the optimum detector is shown in Figure 7. As shown, the useful lifetime of the detector within the ten per cent limits was calculated to be 1.2946×10^{21} neutrons/cm².

Plutonium with the optimum composition indicated in Table IV was not readily available. A simple mixture of available material yielded the following composition:

Pu-239	28.41%
Pu-240	71.43%
Pu-241	0.16%

Using the same cross sections, decay constants, and fluxes as before, the computer program was run with this input composition to determine the behavior which could be expected in the magazine facility environment. These results are also shown in Figure 7. The lifetime of detectors using this material is calculated to be 1.2612×10^{21} neutrons/cm² or just slightly less than the optimum. Therefore, a dozen elements with this composition were encapsulated in a manner similar to that used for the determination of T and these have been placed in the magazine facility and are currently undergoing irradiation. An element is to be discharged after each operating period and its isotopic composition determined. Integrated neutron flux exposures are to be determined from isotopic changes in samples of U-235 which are undergoing irradiation with the regenerating samples. The actual irradiation history of the regenerating plutonium detectors is to be compared with the calculated behavior shown in Figure 7. If good experimental agreement is obtained, it will be concluded the experimental-calculational technique described herein provides a valid method of determining the optimum composition for regenerating detectors for use in various reactors and reactor environments.

FIGURE 7

Calculated Relative Sensitivity of "Optimum" and "As-Built" Detectors as a Function of Exposure in Magazine Facility

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