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FISSION PRODUCT TRANSIENT MEASUREMENTS IN THE ADVANCED REACTIVITY MEASUREMENT FACILITIES

MASTER

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Fission Product Transient Measurements
in the Advanced Reactivity Measurement Facilities

R. G. Nisle, I. E. Stepan, D. A. Millsap, and E. Fast

Summary

The yields of ^{135}I in the fission of ^{233}U , ^{239}Pu , and ^{241}Pu relative to that for ^{235}U have been measured by an integral method using the Advanced Reactivity Measurement Facilities (ARMF I and II). The xenon reactivity transient was measured following irradiation of the fissile isotope in the Materials Testing Reactor (MTR) for periods from 2-1/2 to 110 days. The use of the two ARMF reactors permits the calculation of the absorption cross sections of both simple absorbers and fissile atoms. Calibration and data reduction techniques make extensive use of digital computer codes and take account of nonlinear reactivity responses of the reactors. The ^{135}I yield ratios, relative to ^{235}U , were found to be 0.825 ± 0.072 for ^{233}U , 1.006 ± 0.067 for ^{239}Pu , and 1.221 ± 0.089 for ^{241}Pu . The quoted uncertainties are confidence intervals at the 90% confidence level.

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Fission Product Transient Measurements in the Advanced Reactivity Measurement Facilities

R. G. Nisle, I. E. Stepan, D. A. Millsap and E. Fast

Introduction

Early measurements^(1,2,3) of reactivity transients arising from the decay of fission products were made on the Reactivity Measurement Facility (RMF). With the completion of the Advanced Reactivity Measurement Facilities⁽⁴⁾ (ARMF I and II) improved data handling techniques became necessary to take advantage of the increased precision of these reactors. This report will describe some of these improvements, notably, an extension of fuel and poison calibration procedures to account for the nonlinear response of these reactors to large samples. Other improvements are (a) data reduction procedures have been mechanized through the use of digital computer codes and (b) an improved method for measurements of flux level and spectrum in the irradiating facility (Materials Testing Reactor) has been used.

Tentative values of the fission yield of ^{135}I in the cases of ^{233}U and ^{239}Pu relative to that for ^{235}U have been reported previously⁽⁵⁾. This report presents a detailed description of the experimental and analytical techniques that were used and a revision of results based on additional data, as well as results subsequently obtained on the ^{135}I yield for ^{241}Pu relative to that for ^{235}U .

Description of the Experiment

The basic principles of the method used here have been reported elsewhere⁽⁶⁾. Variations in data handling techniques as used by

others have also been reported⁽¹⁾. Early reports of work at this laboratory assumed the reactivity response to be a linear function of fuel and/or poison content. The improved precision of the ARMF reactors, however, makes it necessary to take account of nonlinear effects⁽⁸⁾. The experiment may be divided into four phases:

(a) irradiation of the sample and measurement of its reactivity subsequent to irradiation, (b) fuel and poison calibration of the ARMF reactors, (c) reduction of the raw reactivity data, and (d) analysis of the reduced data and error analysis. This section will be confined to phases (a) and (c). The other two will be dealt with in separate sections.

The sample design is shown in Figure 1. The ARMF core configurations with the measuring locations indicated are shown in Figures 2a and 2b.

Sample irradiations were made in both an MTR rabbit facility (VH-4) and in a tank location (L-52). The various specimens were irradiated for periods varying from 2-1/2 to about 110 days at flux levels of about 1.6×10^{14} nv. Irradiations up to about 350 hours were made in the rabbit facility. About twenty minutes is an average transfer time when the rabbit facility is used, with an occasional transfer being accomplished in as short a time as ten minutes. Such rapid transfer is desirable to permit the taking of reactivity measurements during as much of the early part of the xenon and rhodium transients as possible. The long irradiation times were used to study the buildup of gross fission products.

Immediately upon removal of the samples from the MTR a sequence of measurements were begun in the ARMF I and ARMF II. Usually two

samples were irradiated simultaneously. They were then measured alternately in first one reactor (ARMF I or II) then in the other. Both reactors were in continuous operation and hence both samples could be measured at once. In this way reactivity transients on each of two samples were obtained in two reactors concurrently. About eight minutes were required to take one reading on one sample. This sequence of measurements continued for about 40 hours. Additional readings were taken at intervals during the following 500 to 1000 hours. Typical plots of reactivity ($\Delta k/k$) versus time are shown in Figure 3.

Similar measurements were taken on the empty reactor and on a variety of calibration samples containing different amounts of the various fuels and different amounts of lithium. The lithium samples constitute $1/v$ reference absorbers. Calibration samples containing both fuel and lithium were also used. Such measurements were taken at intervals of one to four hours during the transient measurement, and are used to detect drift or any possible malfunction of the reactor and to calibrate the reactor.

The reduction of the raw data has been mechanized through the use of digital computer codes. Regulating rod position is converted to net reactivity values by a program⁽⁹⁾ that also corrects for reactor drift. By means of a calibration equation the reactivity values are translated to equivalent $1/v$ cross section values with another computer code. Reactivity measurements, made in two reactors having different relative worths of thermal and fission neutrons provide a non-destructive means for finding both fuel and poison cross sections of the sample. A computer code⁽¹⁰⁾ that does a nonlinear simultaneous equation solution is used for this purpose.

One now has a list of cross sections, $\hat{\Sigma}_p(t_i)$ at a sequence of times, t_i . It has been shown⁽¹¹⁾ that the time dependent cross section, $\hat{\Sigma}_p(t)$, is representable by an equation containing an exponential term for each of the isotopes, ^{135}I , ^{135}Xe , ^{149}Pm , ^{105}Ru , and ^{105}Rh , plus a constant term. The coefficients of these exponentials are functions of the various yields, half-lives, and the fractional burnup of the fuel. A nonlinear least squares curve fitting code calculates these coefficients in the equation

$$\begin{aligned} \hat{\Sigma}_p(t_i) = & C_1 + C_2 e^{-\lambda_I t_i} + C_3 e^{-\lambda_{Xe} t_i} + C_4 e^{-\lambda_{Pm} t_i} \\ & + C_5 e^{-\lambda_{Ru} t_i} + C_6 e^{-\lambda_{Rh} t_i} \end{aligned} \quad (1)$$

where the λ 's are the decay constants for ^{135}I , ^{135}Xe , ^{149}Pm , ^{105}Ru , and ^{105}Rh . In the cases of ^{233}U and ^{235}U , however, it has been found that a better fit results when only the first four terms are used.

Calibration Methods

A fuel and a poison calibration of the measuring locations in the ARMF reactors are necessary to evaluate the reactivity worth of these quantities in the particular sample geometry used. When the reactivity worths are known it becomes possible to calculate equivalent fuel and poison contents of the samples from the measured reactivities. The basic principles of the method are described in Ref. 6. This section will describe the refinements that have been introduced to account for the nonlinear response of the reactors to large samples.

The reactivity response of a reactor to the introduction of a quantity of a simple neutron absorber (poison) may be written

$$\rho_p = \bar{w}_p \hat{\Sigma}_p G(\hat{\Sigma}_p) \quad (2)$$

where

ρ_p = reactivity change as indicated by a change in position of the calibrated regulating rod

$\bar{w}_p$ = reactivity worth of poison averaged over the flux spectrum in the measuring location, reactivity per unit effective cross section

$\hat{\Sigma}_p = VN\hat{\sigma}_p$ = effective absorption cross section of the sample producing the reactivity change, cm^2

$G(\hat{\Sigma}_p)$ = nonlinear factor (dimensionless)

V = volume of sample

N = atom density of absorber

$\hat{\sigma}_p$ = effective microscopic cross section

In addition to the requirement that Eq. (2) shall give a good fit to the experimental data, the factor $G(\hat{\Sigma}_p)$ must be of such form that

$$\lim_{\hat{\Sigma}_p \rightarrow 0} G(\hat{\Sigma}_p) = 1 \quad (3)$$

The proportionality factor, $\bar{w}_p$, therefore, may be interpreted as the slope of the $(\rho, \hat{\Sigma}_p)$ curve at the origin. It has been found that an expression⁽⁹⁾ for $G(\hat{\Sigma}_p)$ that fits the data is

$$G(\hat{\Sigma}_p) = [1 - \exp(-K\hat{\Sigma}_p)][K\hat{\Sigma}_p]^{-1} \quad (4)$$

where K is a constant. Equation (4) is also seen to satisfy the requirement defined by Eq. (3). When Eq. (4) is substituted into Eq. (2), one has

$$\rho_p = \bar{w}_p [1 - \exp(-K\hat{\Sigma}_p)] \quad (5)$$

where

$$W_p = \bar{w}_p / K.$$

The process of calibration for poison response, therefore, consists of evaluating W_p and K in Eq. (5) by the use of poison calibration standards.

The reactivity response of a reactor to the introduction of a quantity of fuel is complicated by the fact that it is a composite of two events -- the absorption of neutrons and the subsequent production of neutrons by fission. If fission did not occur, then the reactivity response would be no different than for any other absorber. Furthermore, the component of the reactivity response due to the fission event is due solely to the response of the reactor to neutrons produced in fission. If it can be assumed that the absorption of fission neutrons in the sample is negligible, then the nonlinear factor should have the same form as for a simple neutron absorber. Therefore, one may write

$$\rho_f = (\bar{w}_p + \bar{\eta}\bar{w}_f) \hat{\Sigma}_a G(\hat{\Sigma}_a) \quad (6)$$

where

ρ_f = reactivity change resulting from the introduction of a fuel sample

$\bar{w}_f$ = reactivity worth of fission neutrons in the measuring location averaged over the fission spectrum, reactivity per fission neutron

$\hat{\Sigma}_a = VN\sigma_a$ = effective absorption cross section (fission + capture) of the fuel sample producing the reactivity change

$\bar{\eta}$ = neutron multiplication constant averaged over the fuel sample content and over energy.

Since $G(\hat{\Sigma}_a)$ is assumed to be of the same form as $G(\hat{\Sigma}_p)$ one may substitute an expression similar to Eq. (4) in Eq. (6). The result is

$$\rho_f = [W_p + \bar{\eta}W_f][1 - \exp(-K\hat{\Sigma}_a)] \quad (7)$$

where

$$W_f = \bar{w}_f/K$$

When fuel samples only are available it is impossible to evaluate both W_p and W_f . Equation (7) can only be used to find K and $[W_p + \eta W_f]$. However, Eq. (7) may be generalized by writing

$$\rho_i = [W_p + \bar{\eta}_i W_f][1 - \exp(-K\hat{\Sigma}_i)] \quad (8)$$

where ρ_i = reactivity response of any isotope, i

$\hat{\Sigma}_i$ = the corresponding effective absorption cross section as previously defined

Then $\bar{\eta}_i = 0$ for a simple absorber and Eq. (8) reduces to Eq. (5). Thus both pure fuel and pure poison samples may be used in Eq. (8) to evaluate W_p , W_f (or $\bar{\eta}W_f$), and K by a least squares fitting process.

Because of the fact that $\bar{\eta}$ is also an effective value that varies with the flux spectrum it is convenient to calculate the product $\bar{\eta}W_f$ when only one fissile isotope is present. For plutonium a better calibration results when an effective sample- η is used. By this process both ^{239}Pu and ^{241}Pu can be accounted for as well as the variation of effective η with flux spectrum.

It is not enough to have a calibration for fuel alone, or for poison alone, because the irradiated samples contain both, and one

would like to be able to measure how much of each is present. Since the form of the nonlinear factor, $G(\Sigma)$, has been shown⁽⁹⁾ to depend on absorption only, the reactivity of a composite sample may be found by summing a series of expressions of the form of Eq. (8). The result is

$$\rho_{p+f} = [W_p \hat{\Sigma}_p + (\bar{n}W_f) \hat{\Sigma}_a] G(\hat{\Sigma}_p + \hat{\Sigma}_a) \quad (9)$$

Let $\hat{\Sigma}_p + \hat{\Sigma}_a = \hat{\Sigma}_T$ and substitute Eq. (4). The result is

$$\rho(\hat{\Sigma}_T) = [W_p + \bar{n}W_f \frac{\hat{\Sigma}_a}{\hat{\Sigma}_T}] [1 - \exp(-K\hat{\Sigma}_T)] \quad (10)$$

Equation (10) describes the reactivity response of the reactor to a sample containing both fuel and poison in terms of the proportionality constants W_p and W_f , the sample content parameters $\bar{n}$, $\hat{\Sigma}_p$, and $\hat{\Sigma}_a$, and the nonlinear parameter K . The calibration process consists in evaluating W_p , W_f , and K by use of samples of known values of $\hat{\Sigma}_p$, $\hat{\Sigma}_a$, and $\bar{n}$.

Reactivity effects due to scattering cross sections are negligible compared with those due to absorption or fission cross sections.

Furthermore, the calibration samples used to measure W_p , W_f , and K contained the same amounts of the fissile isotopes as the irradiation sample. The low concentration of the fissile isotope in the sample further tends to reduce the contribution of scattering to the measured reactivity.

Data Analysis

The problem of data analysis is the inverse of the calibration problem discussed above. Since the same sample cannot be measured in

two different reactors at the same time, it is necessary to calculate simultaneous reactivity values from smoothed data. This section describes the smoothing method as well as the yield and cross section calculations.

The operation of the two-location method depends on the fact that in different measurement locations the relative values of W_p and W_f are different. Two different locations in one reactor could be used. However, in this experiment two reactors, the ARMF I and ARMF II, were used. Aside from the fact that these reactors were available, there are two reasons for using the ARMF II rather than a second location in the ARMF I: (a) The ARMF II is much more sensitive to poison than any location in the ARMF I. (b) The measuring location in the ARMF II is more symmetrically located than any second location in the ARMF I, and is, therefore, less sensitive to positioning errors.

One has, therefore, two sets of reactivity values as a function of time and two sets of the parameters W_p , W_f , and K . The two sets of reactivity data cannot be used as they are, however, in the two resulting simultaneous equations because they do not contain simultaneous pairs of reactivity values. It is first necessary to smooth the raw data by means of a curve fit and then calculate simultaneous pairs from the smooth fit. The nonlinear least squares computer code previously mentioned can be used for this purpose. The equation fitted by that code is

$$\rho(t) = A_1 + A_2 e^{-\lambda_I t} + A_3 e^{-\lambda_{Xe} t} + A_4 e^{-\lambda_{Pm} t} + A_5 e^{-\lambda_{Ru} t} + A_6 e^{-\lambda_{Rh} t} \quad (11)$$

Because of the nonlinear relationship between ρ and $\hat{\Sigma}_p$ there is a

question as to the propriety of writing Eq. (11) in the same form as Eq. (1). This question will be discussed in greater detail in the section on error sources.

Simultaneous reactivity pairs, $\rho_1(t_n)$ and $\rho_2(t_n)$ are then used in a pair of equations of the form of Eq. (10) with appropriate subscripts to designate the measuring location. The mixed nature of these equations necessitates a double iteration -- one for $\hat{\Sigma}_a$ and one for $\hat{\Sigma}_p$.

Since Eq. (10) is expressed in terms of effective cross sections, these variables will be different in the two reactors. Therefore, it is necessary to introduce effective cross section factors to have a variable that has the same value in the two corresponding simultaneous equations. Nisle's formulation^(12,13) was used for this purpose. Thus, one has

$$\begin{aligned}\hat{\Sigma}_p &= \Sigma_p P_y(F_1, T) \\ \hat{\Sigma}_a &= \Sigma_a P_x(F_1, T)\end{aligned}$$

where

$$\Sigma_a = NV\sigma_a, \Sigma_p = NV\sigma_p \quad (2200 \text{ m/s values})$$

$$P_x(F_1, T), P_y(F_1, T) = \text{effective cross section factor for isotopes } x, y.$$

F_1 is a parameter characterizing the resonance fraction of the flux spectrum.

T is the neutron temperature.

Σ_p, Σ_a are the required variables in Eq. (10).

Thus, the two-location method (two reactors in the present experiment) yields 2200 m/s absorption cross sections for the poisons and fuels in a sample.

One now has a value for Σ_a which is a constant since the burn up of fuel during the reactivity measurement is negligible, and a sequence

of values for $\Sigma_p(t)$. One also has values for the coefficients in Eq. (1) as previously described. It is now possible, in principle at least, to find the yield for ^{135}I and $^{135}\text{I} + ^{135}\text{Xe}$, the effective poison cross section at time zero (time of removal from the irradiating flux), and the effective poison cross section of the long half-life fission products. Relative magnitudes of the calculated quantities and experimental errors make it impractical to find all these quantities with sufficient accuracy to be meaningful. Nevertheless, the principles will be discussed.

Expressions for the coefficients in Eq. (1) are derived in reference (6). The equation pertinent to the calculation of the fission yield of ^{135}I is, after dropping negligible terms

$$C_2 = - \hat{\sigma}_{\text{Xe}} \hat{\Sigma}_{\text{fo}} \phi \left[\frac{\gamma_{\text{I}}}{\lambda_{\text{I}} - \lambda_{\text{Xe}}} \right] \exp(-\hat{\sigma}_{\text{a}} \phi T_{\text{r}}) \quad (12)$$

where the

λ 's = decay constants with appropriate subscripts,

γ 's = fission yield with appropriate subscript,

T_{r} = duration of irradiation,

$\hat{\sigma}_{\text{Xe}}$ = effective ^{135}Xe cross section in the measuring reactor,

ϕ = flux appropriate to the effective cross section formalism used,

$\hat{\sigma}_{\text{a}}$ = effective absorption cross section of the fuel in the irradiating reactor,

$\hat{\Sigma}_{\text{fo}} = VN_0 \hat{\sigma}_{\text{f}}$ = effective fission cross section in the fuel at the beginning of irradiation.

Thus, Eq. (12) may be used to calculate the yield when an experimental value for C_2 has been found and the remaining factors are known from

other sources. All the remaining factors are not as accurately known as one would desire, however. For example, the decay constants λ_I and λ_{Xe} are known to only about two significant figures and their difference is small. Therefore, the accuracy of this difference suffers. The effective cross section of ^{135}Xe is another quantity of limited accuracy.

For these reasons it is the yield ratios rather than the absolute yields that are calculated from reactivity transient data. For the case of ^{233}U and ^{235}U one has

$$\frac{\left[Y^{135}\text{I} \right]_{233\text{U}}}{\left[Y^{135}\text{I} \right]_{235\text{U}}} = \frac{\left[C_2 \right]_{233\text{U}}}{\left[C_2 \right]_{235\text{U}}} \frac{\left[\hat{\Sigma}_{f0} \phi \exp(-\hat{\sigma}_a \phi_r^{\text{U}}) \right]_{235\text{U}}}{\left[\hat{\Sigma}_{f0} \phi \exp(-\hat{\sigma}_a \phi_r^{\text{T}}) \right]_{233\text{U}}} \quad (13)$$

Similar equations apply when the yield of ^{135}I for ^{239}Pu and ^{241}Pu fission relative to that for ^{235}U is to be found.

In Eq. (13) the exponential term describes the fractional burn up of the fuel. To derive the exponential term it is necessary to use an effective cross section factor appropriate to the irradiation location in the MTR. This situation leads to complications, since the flux wires are too far from the fuel to give an accurate value for either flux level or spectrum.

The reactivity content calibration and the two-reactor method, however, provide means for using the sample as its own flux detector. To this end the burn-up term is rewritten

$$\hat{\Sigma}_{f0} \phi \exp(-\hat{\sigma}_a \phi_r^{\text{T}}) = N_0 V (\hat{\sigma}_f \phi) \exp(-\hat{\sigma}_a \phi_r^{\text{T}}) \quad (14)$$

The exponential term in Eq. (14) is evaluated by using the initial fuel content and the post-irradiation fuel content, or by using the

^{137}Cs method of measuring number of fissions. These quantities are obtained from chemical analyses. Then one has

$$\frac{M}{M_0} = \exp(-\hat{\sigma}_a \phi T_r) \quad (15)$$

where the M 's are grams of fuel from which the term $(\hat{\sigma}_a \phi)$ is easily obtained, since T is accurately known.

The term $\sigma_f \phi$ in Eq. (14) can now be derived as follows:

Note that $\hat{\sigma}_a = \sigma_a P_a(F_1, T)$ and that $\hat{\sigma}_f = \sigma_f P_f(F_1, T)$. Furthermore, σ_f/σ_a is the ratio of the 2200 m/s cross sections. Hence

$$\hat{\sigma}_f \phi = \hat{\sigma}_a \phi (\sigma_f/\sigma_a) [P_f(F_1, T)/P_a(F_1, T)]. \quad (16)$$

The ratio of P -factors is insensitive to flux spectrum in the range of temperature and resonances fluxes encountered in the MTR. The latter ratio has been calculated from tabulations in reference 14. They are

$$^{233}\text{U}, \quad \frac{P_f(F_1, T)}{P_a(F_1, T)} = 1.030 \pm .002$$

$$^{235}\text{U}, \quad \frac{P_f(F_1, T)}{P_a(F_1, T)} = 0.985 \pm .003$$

$$^{239}\text{Pu}, \quad \frac{P_f(F_1, T)}{P_a(F_1, T)} = 0.985 \pm .005$$

$$^{241}\text{Pu}, \quad \frac{P_f(F_1, T)}{P_a(F_1, T)} = 1.002 \pm 0.001$$

The effective fission product cross sections may be obtained directly from the coefficients of Eq. (1). When $T = 0$, each of the exponentials equals one and the result is

$$\Sigma_p(0) = C_1 + C_2 + C_3 + C_4 + C_5 + C_6 \quad (17)$$

When t is very large the exponentials approach zero, and the result is

$$\Sigma_p(t \text{ is large}) = C_1 \quad (18)$$

If the cross section per fission is required, it is simply necessary to divide Eq. (17) or (18) by the number of fissions derived from the burn-up calculation, which may use either chemical analysis or the Σ_a of Eq. (10).

If one assumes the fission yield of ^{135}I to be known, Eq. (12) could be used to find the effective cross section of ^{135}Xe , $\hat{\sigma}_{\text{Xe}}$, but the result is expected to be of limited accuracy because of the term $(\lambda_{\text{I}} - \lambda_{\text{Xe}})$.

The equations for the mass-105 chain are the same as those for the mass-135 chain with the appropriate constants for ^{105}Ru and ^{105}Rh replacing those for ^{135}I and ^{135}Xe , respectively. Since, however, the ^{105}Ru yields for ^{235}U and ^{233}U are very small (about 0.8% and 0.5% respectively) the corresponding coefficients in Eq. (1) are of limited accuracy. In the case of the plutonium isotopes the ^{105}Ru yields are comparable to those of ^{135}I . Furthermore, the $(\lambda_{\text{Ru}} - \lambda_{\text{Rh}})$ term in Eq. (12) is of better accuracy than the corresponding term for the $^{135}\text{I} - ^{135}\text{Xe}$ case. The ratio approach is not suitable in the case of the plutonium isotopes (^{239}Pu and ^{241}Pu), since the accuracy of the C_5 coefficient in Eq. (1) for ^{235}U is very poor. It might appear that Eq. (12) might be used directly to derive an ^{135}I yield value. Here again the poor accuracy of the $\hat{\sigma}_{\text{Xe}}$ term impairs the accuracy of the result.

Sources of Error

The various sources of error may be classified as follows:

- (a) reactivity measurement uncertainties,
- (b) sample composition uncertainties,
- (c) uncertainties due to assumptions made in the data analysis methods,
- (d) uncertainties in the values of various factors derived from other sources.

Reactivity measurement uncertainties are considered to be the least important. Although the absolute reactivities may be in error the relative values and precision of measurement is very good. The standard deviation calculated from several hundred readings on both stable and radioactive samples is less than $0.2 \times 10^{-6} \Delta k/k$. These measurements were spread over a period in excess of two years and total sample worth varied from -1.0 to $+300 \times 10^{-6} \Delta k/k$. Since C_2 depends on curve shape and not on absolute reactivity values, the uncertainty introduced from this source is, therefore, considered negligible.

Uncertainties in sample composition effect the result through the calibration of the reactivity worth of fuel and poison sample in the applicable measuring locations. Measurements on the calibration standards showed deviations between 3.0 and $4.0 \times 10^{-6} \Delta k/k$. This is much larger than can be attributed to uncertainties in the reactivity measurements. Such deviations were attributed to the fact that sample content could possibly be in error since the value used was not the direct result of analysis but was inferred from analysis of lathe turnings taken next to the alloy specimen actually used to fabricate

the sample. To solve the problem of sample content several calibration samples were sacrificed to get destructive analyses of their content after making extensive reactivity measurements on them. The resulting calibration was then used as a non-destructive analysis method on the remaining samples. This process largely eliminated the previously unexplained deviations.

The assumption that Eq. (1) is sufficient to represent the change in poison cross section over the period of the transient reactivity measurements requires investigation. In order to test this assumption for short half-life products, a curve fit was made to the same set of cross-section data with successively larger portions of the early data deleted. For each of these cases the cross section extrapolated to $t = 0$ was calculated. It was found that the variation in the extrapolated value was less than 0.5% for deletions up to 6 hours. Furthermore, it has been found that the standard deviation of the curve fit is about the same as that for repeated measurements on a stable sample. It is concluded, therefore, that Eq. (1) is sufficient to represent the change in poison cross section within the accuracy of the measurements; and that there are no short half-life products of high cross section present in sufficient quantities to be observed. The sufficiency of Eq. (1) requires qualification, however. It is derived on the assumption that the irradiating flux is constant. In a reactor such as the MTR many experiments are being conducted simultaneously and oftentimes there will be a power reduction or an inadvertant scram attributable to one of these other experiments. The balance of fission products in the sample is, therefore, upset to a degree depending on the number and duration of such power reductions. This situation

existed for one value of the ^{135}I yield ratio for ^{233}U in particular. The limited number of data points (3), however, made it uncertain whether the rejection of the one value was justified, even though there were 15 power reductions during the irradiation. A fourth irradiation was made in which the irradiation flux level was constant. By the use of standard techniques⁽¹⁴⁾ for the rejection of Outliers a rejection factor for the suspected value was found to be 0.656 whereas the value given for 90% confidence was 0.679. Rejection, therefore, is not justified on strictly mathematical grounds, although the fact that the calculated factor is so close to the cut-off value renders the test inconclusive. It is felt, therefore, that a judgement based on a knowledge of the experimental conditions, namely, the departure of those conditions from the conditions specified by the equations, is sufficient to justify rejection. If the suspected value is retained, the yield is 0.778 ± 0.117 ; if not, the yield is as quoted in Table I.

Uncertainties associated with assumptions implicit in Eq. (11) may be described as follows: for the case of very dilute samples in which the function $G(\Sigma_T)$ approaches unity it is easy to derive Eq. (11) from Eq. (1). For the nonlinear case each of the coefficients in Eq. (11) would be of the form

$$A_i = \frac{W_p C_i [1 - \exp[-K(\hat{\Sigma}_p(t) + \hat{\Sigma}_a)]]}{K(\hat{\Sigma}_p(t) + \hat{\Sigma}_a)} \quad (19)$$

A rigorous treatment shows, therefore, that the coefficients in Eq. (11) are not constants, but are themselves functions of time. The problem is determinant and hence solvable in principle, but probably not worth

the effort required. An error study has indicated that the uncertainty introduced by this assumption is no greater than about 1%.

The burn-up terms in brackets on the right side of Eq. (13) constitute data that must, ultimately, be derived from non-reactivity measurements, namely chemical and spectrochemical analyses, either directly or through the medium of a calibration. Pre- and post-irradiation analyses of the samples can be used to evaluate the exponential, $e^{-\sigma_a \phi T_r}$. Alternatively a method based on reactivities measured in two reactors as described in the section on Data Analysis can be used. This method uses calculations of pre- and post-irradiation Σ_a values, which when converted to fuel content gives $e^{-\sigma_a \phi T_r}$ by Eq. (15). Since both methods depend ultimately on a chemical analysis, the uncertainties are about the same.

By restricting results to the yield ratio given by Eq. (3), the need for data from other sources is minimized. The only additional data, other than sample content, are the decay constants used in Eq. (1). By varying the λ values used in Eq. (1) it was found that a 1% variation in λ produced about a 3% variation in the calculated coefficients. This same imposed variation in the λ 's produced a larger standard deviation of the curve fit. Hence the values of λ that were used were those that produced the smallest standard deviation of the fit, which λ 's incidently were the same as the literature values.

The process used to evaluate $\sigma_f \phi$ and $\sigma_a \phi$, based on burn-up measurements further reduces the reliance on data from other sources. By this method the sample is made to be its own flux detector and the problem of converting flux wire data to sample flux level and flux spectrum is avoided.

In summary, therefore, one may say that reactivity measurements introduce negligible uncertainties; sample composition uncertainties are minimized by a combination of reactivity calibrations and direct chemical analysis; the chemical analysis uncertainties (confidence interval) amount to about 2% at the 95% confidence level; and uncertainties in data from other sources have been reduced to those associated with the decay constants, λ in Eq. (1).

Results and Discussion

Tentative values based on limited data were reported in a Technical Note⁽⁵⁾. The uncertainties quoted there were best estimates of the standard deviation, which admittedly are not very significant for such limited data. A better method that accounts for the smallness of the sample is to quote a confidence interval at a given level of confidence⁽¹⁴⁾. The uncertainties quoted here are confidence intervals at the 90% confidence level.

The case of ^{241}Pu requires special consideration, since the 14-year half-life is sufficiently short that the resulting reactivity change is significant over the period of time that transient measurements are being made. To evaluate this effect one unirradiated ^{241}Pu sample was measured about once a week over a period of about 18 months. Measurements in two reactors were then used to measure the decay of ^{241}Pu and build up of ^{241}Am .

In the case of the fission product transient the effect just discussed is superposed upon the effect arising from fission product decay. Thus, instead of the fuel content remaining constant after irradiation it will decrease, and the measured poison cross section

will be affected by the ^{241}Am build up. During the period that the xenon cross section dominates, however, these effects are relatively constant. Hence, the process of fitting Eq. (1) to the data is still valid and no appreciable additional error in the yield ratios result from this source.

However, corrections that account for the ^{241}Pu decay must be made to $\hat{\Sigma}_{f0}$ in Eq. (13) and to the quantity, $\hat{\sigma}_a \phi T_r$, the argument of the exponential in Eq. (13); that is, $\hat{\sigma}_a \phi T_r$ must be replaced by $(\hat{\sigma}_a \phi + \lambda) T_r$, and $\hat{\Sigma}_{f0}$ must be corrected for the decay of ^{241}Pu to account for the time between the determination of sample content and the beginning of irradiation.

The relative fission yields for the fissile isotopes measured are shown in Table I together with results published by others.⁽⁷⁾ The results quoted here are a revision of previously published results⁽⁵⁾ as are those of Okazaki and Walker⁽⁷⁾. In the latter publication the values for ^{233}U and ^{239}Pu have been revised upward slightly from previously quoted values⁽¹⁵⁾ and a value for ^{241}Pu has been added. The uncertainties quoted by them are the r.m.s. deviations based on all experimental values. Hence they are not strictly comparable to the uncertainties quoted in Table I. No attempt has been made to make a conversion to comparable uncertainties since all pertinent data were not available. The uncertainties quoted in reference 5 were also r.m.s. deviations and hence, they were recalculated using the confidence interval method.

Although measurements are continuing on the fission product cross sections, interim results appear to be worth quoting.

Figures 4 and 5 show reactivity measurements on ^{233}U and ^{235}U

samples irradiated to about 80% burn up. The reactivity change is seen to be very small—even for decay times as large as 12,000 hours ($\approx 1\frac{1}{2}$ years). Although these figures appear to show a small negative slope in the fitted curve, the 95% confidence band is about $0.2 \times 10^{-6} \Delta k/k$ wide, which is greater than the observed reactivity decrease. The results to date, therefore, are inconclusive. Measurements are continuing in an attempt to resolve this question.

Conclusions

Final results on the yield ratio for ^{135}I have been given for ^{233}U , ^{239}Pu and ^{241}Pu relative to ^{235}U . These results differ somewhat from preliminary results previously published. Unscheduled power reductions occurring during the irradiation period, even though of short duration, were found to produce erratic results; hence, only those transients, for which the irradiating flux level was constant, were used in calculating the relative yields reported. Preliminary results reported earlier used some of these transients that were later discovered to be unacceptable on the grounds of constancy of the irradiating flux. These were rejected on such grounds even though strict interpretation of statistical tests might indicate otherwise.

Results on the decay of long half-life fission products are also presented. A small decay of about the same magnitude as the uncertainty in the reactivity measurements was observed. The results are, therefore, inconclusive as to the value of the decay rate, if any. If one assumes the decay rate to be negligible, then one could conclude that, on the average, the cross sections of the fission products are about the same as those of the decay products.

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Table I

Yield Ratios of ^{135}I in the Fission of Various IsotopesRelative to ^{235}U

	<u>This Paper</u>	Nisle & ⁽⁵⁾ <u>Stepan</u>	Okazaki & ⁽⁷⁾ <u>Walker</u>	Weaver, et al ⁽¹⁶⁾ <u></u>
^{233}U	0.825 ± 0.072^a	0.700 ± 0.156^b	0.761 ± 0.011^c	0.749
^{239}Pu	1.006 ± 0.067^b	1.204 ± 0.214^b	0.955 ± 0.014^c	1.024
^{241}Pu	1.221 ± 0.089^b	_____	1.205 ± 0.030^c	_____

NOTES: a - average of three values - confidence interval at the 90% level (see text)

b - average of two values - confidence interval at the 90% level (see text)

c - uncertainties quoted as r.m.s. deviations (see text)

Fig. 1 Sample design

Fig. 2a ARMF I core diagram showing measurement location

Fig. 2b ARMF II core loading showing measurement location

Fig. 3 Typical xenon reactivity transients in the ARMF II

Fig. 4 Reactivity measurements on irradiated ^{233}U in the ARMF I

Fig. 5 Reactivity measurements on irradiated ^{235}U in the ARMF I