and Kaplan have presented a method for performing such an analysis together with the experimental comparison (*6*).

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Measurements of Relative Pu Fission Rates in Slightly Enriched Uranium–Water Lattices

The fission activation of Pu^{240} relative to Pu^{239} and Pu^{241} has been measured for the TRX facility, a slightly enriched uranium–light water moderated reactor. The lattices in which the experiments were performed were composed of 1.3 wt % enriched uranium metal fuel rods, 0.387 in. in diameter, with a water to uranium volume ratio of either 2.35:1 or 1:1. The experiments described here are an extension of previously reported measurements for these lattices (*1*, *2*).

The Pu was in the form of 1 mg/cm² deposits on highly pure nickel foils 0.010 in. thick. The isotopic concentrations of the deposits are given in Table I.

The presence of approximately 19% Pu^{239} in the "Pu^{240}" deposit introduced the problem that the Pu^{240} isotope contributes only of the order of 3–4% to the total fission activity of this deposit irradiated in these lattices. This relative contribution was increased to roughly 30% by irradiating this deposit Cd covered, thereby suppressing the thermal activation contribution of the Pu^{239}.

The ratio of the measured total fission product activity of a Cd covered "Pu^{240}" deposit to a bare "Pu^{239}" deposit is given by:

$$\gamma(t)_{49}^{40} = \frac{u(t)^{40}N_{40}^{40}I^{40} + u(t)^{41}N_{40}^{41}I_\epsilon^{41} + u(t)^{49}N_{40}^{49}I_\epsilon^{49}}{u(t)^{49}N_{49}^{49}I^{49}}$$

where $\gamma(t)_{49}^{40}$ is the relative activity at time t after irradiation, N_k^i is the number of atoms of isotope i in deposit k and $u(t)^i$ is a time function which reflects the gross fission product decay rate of isotope i. The $u(t)^i$ and hence the $\gamma(t)$ are functions of the time of irradiation and the time t at which the deposits are counted. It is implicitly assumed that $u(t)^{49}$ and $u(t)^{41}$ are independent of the incident neutron energy initiating fission. No time dependence was detected in the Cd ratio measurements. The various fission activation integrals are denoted by the symbol I; i.e.,

TABLE I

Plutonium Deposit Enrichments

Deposit	Pu^{239}(%)	Pu^{240}(%)	Pu^{241}(%)	Pu^{242}(%)
"Pu^{239}"	99.36	0.63	0.01	
"Pu^{240}"	18.64	80.08	1.14	0.14
"Pu^{241}"	9.02	31.01	57.96	2.01

$$I^i = \int_0^\infty \sigma_f{}^i(E)\phi(E)\,dE \quad \text{and} \quad I_\epsilon{}^i = \int_\epsilon^\infty \sigma_f{}^i(E)\phi(E)\,dE$$

where ϵ is the Cd cutoff energy (~0.45 ev). The Pu^{242} contribution was neglected.

If the above expression is solved for the ratio of the Pu^{240} to Pu^{239} fission integrals, the result is the following:

$$\frac{\int_0^\infty \sigma_f^{40}(E)\phi(E)\,dE}{\int_0^\infty \sigma_f^{49}(E)\phi(E)\,dE} = P(t)_{40}^{49}\left\{\left[\frac{N_{49}^{49}}{N_{40}^{40}}\gamma(t)_{49}^{40} - \frac{N_{40}^{49}}{N_{40}^{40}R^{49}}\right] - P(t)_{49}^{41}\frac{N_{40}^{41}}{N_{40}^{40}}\frac{I^{41}}{I^{49}R^{41}}\right\} \tag{1}$$

where R^i is the measured Cd ratio of the ith isotope and $P(t)_m{}^i$ is the ratio $u(t)^i/u(t)^m$ and accounts for the relative gross fission product decay rates of isotopes i and m.

In a similar manner, the following is obtained:

$$\gamma(t)_{41}^{40} = \frac{u(t)^{40}N_{40}^{40}I^{40} + u(t)^{41}N_{40}^{41}I_\epsilon^{41} + u(t)^{49}N_{40}^{49}I_\epsilon^{49}}{u(t)^{40}N_{41}^{40}I^{40} + u(t)^{41}N_{41}^{41}I_\epsilon^{41} + u(t)^{49}N_{41}^{49}I_\epsilon^{49}}$$

$$\frac{\int_0^\infty \sigma_f^{40}(E)\phi(E)\,dE}{\int_0^\infty \sigma_f^{41}(E)\phi(E)\,dE} = P(t)_{40}^{41} \times \frac{\left[\frac{N_{40}^{41}}{N_{41}^{41}R^{41}} - \gamma(t)_{41}^{40}\right] + P(t)_{41}^{49}\frac{N_{41}^{49}}{N_{41}^{41}}\frac{I^{49}}{I^{41}}\left[\frac{N_{40}^{49}}{N_{41}^{49}R^{49}} - \gamma(t)_{41}^{40}\right]}{\frac{N_{41}^{40}}{N_{41}^{41}}\left[\gamma(t)_{41}^{40} - \frac{N_{40}^{40}}{N_{41}^{40}}\right]} \tag{2}$$

The experimental procedure is similar to the previously reported measurements (*1*, *2*). The quantity $\gamma(t)$ was obtained by irradiating the deposits in a split fuel rod and counting the resultant fission product activity with a scintillation counter biased to reject pulses having equivalent energies less than 400 kev. The quantities designated as $P(t)$ were obtained from a separate experiment utilizing a double fission chamber technique (*2*).

A significant effect in the measurements with plutonium deposits that was not important in the previous uranium work was the large α particle emission rate which tends to produce poor plateaus in the fission chamber. In order to decrease the contribution of the α background relative to the fission product ionization, the anode to cathode spacing was varied to give the maximum fission to α ratio consistent with a reasonable gain in the system. The rise time of the system was also improved by using a transistorized pre-amp feeding a nonoverloading amplifier. The fission chamber

TABLE II

RELATIVE FISSION ACTIVATION INTEGRALS

	Experimental	Calculated
(A) 2.35:1 Lattice		
$\frac{\int_0^\infty \sigma_f^{40}(E)\phi(E)\,dE}{\int_0^\infty \sigma_f^{49}(E)\phi(E)\,dE}$	0.0053 ± 0.0007	0.0032
$\frac{\int_0^\infty \sigma_f^{40}(E)\phi(E)\,dE}{\int_0^\infty \sigma_f^{41}(E)\phi(E)\,dE}$	0.0038 ± 0.0007	
$\frac{\int_0^\infty \sigma_f^{28}(E)\phi(E)\,dE}{\int_0^\infty \sigma_f^{25}(E)\phi(E)\,dE}$	0.00131 ± 0.00006	
(B) 1:1 Lattice		
$\frac{\int_0^\infty \sigma_f^{40}(E)\phi(E)\,dE}{\int_0^\infty \sigma_f^{49}(E)\phi(E)\,dE}$	0.0096 ± 0.0010	0.0052
$\frac{\int_0^\infty \sigma_f^{40}(E)\phi(E)\,dE}{\int_0^\infty \sigma_f^{41}(E)\phi(E)\,dE}$	0.0096 ± 0.0013	
$\frac{\int_0^\infty \sigma_f^{28}(E)\phi(E)\,dE}{\int_0^\infty \sigma_f^{25}(E)\phi(E)\,dE}$	0.00238 ± 0.00012	
(C) $P(t)_{49}^{40} = 0.98 \pm 0.05$	$P(t)_{49}^{41} = 1.11 \pm 0.04$	$P(t)_{40}^{41} = 1.13 \pm 0.07$

was Cd covered to suppress the Pu^{239} activation in the "Pu^{240}" deposit.

The relative weights of the Pu on the deposits were obtained by calibration in a highly thermalized spectrum with a measured Cd ratio of the order of 60.

A correction was applied to the measured relative fission integrals given by Eqs. (1) and (2) to account for the self-shielding of the 1 ev Pu^{240} fission resonance by the high ($\sim 10^5$ barns) capture resonance also present at this energy. Using the cross sections given in BNL-325, 2nd ed., (*3*) and the energy dependent flux as given by the Muft code (*4*), the contribution of this fission resonance to the total Pu^{240} fission activity in a "Pu^{240}" deposit was estimated to be 11% for our spectrum. Assuming an isotropic flux and slab geometry, the self-shielding of the 1 ev fission resonance was estimated to be 13%. Thus the total Pu^{240} fission activity was increased by $11\% \times 13\% = 1.4\%$.

The results obtained are listed in Table II. The deposits were activated and counted over a range of time intervals; the irradiation times varied from 10 min to 60 min and the counting intervals from 10 min to 90 min after irradiation. Within experimental error, no time dependence was detected in either of the measured quantities $\gamma(t)$ or $P(t)$ for any combination of irradiation and counting times within the above interval. However, it should be noted that, in principle, the $P(t)$ values listed in Table II are dependent upon the neutron energy spectrum, counting bias (400 kev), scintillation crystal efficiency, etc. utilized in these measurements.

The listed uncertainties are a result of combining the following individual uncertainties:

1. 3% in the weight calibrations.
2. 1% in the measurements of $\gamma(t)$.
3. 3% in the measured Cd ratios.
4. 5–6% in the measurements of $P(t)$.

The final listed errors of 10–20% in the measured relative fission integrals are an indication of the difficulty encountered in detecting the Pu^{240} activity with these particular "Pu^{240}" deposits. The results for the Pu^{241} fission integral relative to the Pu^{239} fission integral agree with the previously measured values, within experimental error, although the experimental uncertainties are large in the present measurements.

A comparison with calculated results is shown in Table II in the case of Pu^{240} relative to Pu^{239}. The fission cross sections utilized were obtained from BNL-325, 2nd ed., and the energy dependent flux constructed for these lattices in refs. *1* and *5*. It is seen that the calculated values are at least 30–40% lower than the measured results. A large portion of this difference is probably due to the lack of adequate cross section data for Pu^{240}, particularly in view of the relatively good agreement between calculated and measured results given in ref. *1*. Calculations were also performed using the Pu^{240} cross section data of Nesterov and Smirenkin (*6*) in which the threshold was determined experimentally to extend to 0.15 Mev in contrast to the value of approximately 0.26 Mev shown in BNL-325. However, only a 1% increase results in the calculated values using this data. Lack of detailed Pu^{241} cross section data prevented any calculations of the corresponding Pu^{240} to Pu^{241} relative fission integrals.

An important conclusion can be drawn by noting the difference between the measured Pu results of this work and those of previous experiments for U, as listed in Table II for these lattices. It is seen that, for the neutron energy spectrum generated in the slightly enriched uranium–water moderated systems investigated, the Pu^{240} fission activation integral contribution relative to Pu^{239} appears to be of the order of 3–4 times as important as the corresponding U^{238} fission activation integral contribution relative to U^{235}.

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