Letters to the Editors

Spatial Dependence of Thermal-Neutron Spectra and the Interpretation of Thermal Utilization Measurements

The thermal utilization, f, is usually defined as the number of thermal neutrons absorbed in the fuel per thermal neutron absorbed in the lattice. For a low U²³⁵ enrichment lattice consisting of uranium fuel rods

$$f = \left\{ 1 + \frac{\phi_{\rm m} \, V_{\rm m} \, N_{\rm m} \, \bar{\sigma}_{\rm m}(T_{\rm m}) + \phi_{\rm el} \, V_{\rm el} \, N_{\rm el} \, \bar{\sigma}_{\rm cl}(T_{\rm cl})}{\phi_{\rm u} \, V_{\rm u} [N_{25} \, \bar{\sigma}_{25}(T_{\rm u}) + N_{28} \, \bar{\sigma}_{28}(T_{\rm u})]} \right\}^{-1} \quad (1)$$

where V is the volume of the material; N is the atomic density; $\overline{\sigma}$ (T) is the microscopic absorption cross section averaged over an appropriate thermal-neutron spectrum characterized by a neutron temperature, T; and ϕ denotes the neutron flux. The subscripts m, cl, u, 25, and 28 refer to moderator, clad, uranium, U²³⁵, and U²³⁶, respectively.

The experimental determination of the thermal utilization involves the introduction of foils, usually U²³⁵ or Dv, in the different regions within the lattice. The resulting activation of these foils determines the neutron density in each region. Since the activation in each region is proportional to the product of the neutron flux and cross section. it is not necessary to make any experimental correction for different effective neutron temperatures in each lattice region provided the foils have the identical neutron absorption energy dependence as the other materials in the lattice. The fact that the foils do have a slightly different energy dependence introduces an extremely small correction (1, 2). Besides the f value, additional quantities that are often quoted as being measured are the neutron-flux ratios or disadvantage factors, ϕ_m/ϕ_u and ϕ_{cl}/ϕ_u , which have been assumed to be proportional to the activation ratio (1, 2). These latter quantities are often used to verify the accuracy of theoretical methods that are used to calculate lattice neutron-flux distributions.

It may be worth while to point out that there are certain analytical simplifications in the experimental interpretation of these activation measurements that may not be entirely justified. First, in these measurements, foils may not have been placed in all regions in the lattice but only in the fuel and moderator regions. It is then necessary to make assumptions regarding the neutron spectrum existing in the cladding and structural materials. If the cladding and structural materials are weak neutron absorbers, e.g., aluminum, then only a small uncertainty in the measurement of thermal utilization will occur due to the assumptions made concerning the effective neutron temperature in these materials (2). However, if the cladding and structural materials are strong absorbers, e.g., stainless steel, then any uncertainties involved in the assumption of an effective neutron temperature for these regions can significantly alter the experimental determination of the thermal utilization.

Second, the pronounced spatial dependence of the thermal-neutron spectra will not usually permit equating the activation ratio to the flux ratio. Bigham has measured the temperature dependence of the ratio of the Pu²³⁹ to U²³⁵ average fission cross sections in a Maxwellian spectrum (3). In light water, at 50°C, a value of 1.44 was measured, whereas the value at 95°C was 1.51. Klein has made room temperature measurements of Pu²³⁹ fission reaction rates relative to U²³⁵ in light-water uranium-metal lattices containing 1.3% U²³⁵ enrichment (4). The measured ratios of the Pu²³⁹ to U²³⁵ fission reaction rates below the cadmium cutoff is 2.00 for a 2.35:1 water-to-metal lattice, and 2.37 for a 1:1 water-to-metal lattice. These latter ratios are approximately proportional to the ratio of the average Pu²³⁹ to U²³⁵ thermal-fission cross sections. The results of Bigham's measurements have been extended analytically by averaging the measured microscopic fission cross sections of Pu²³⁹ and U²³⁵ at higher temperatures and comparing with Klein's lattice measurements. In terms of an effective neutron temperature, these analyses show that the fuel regions in the lattices have a neutron temperature more than 250°C greater than a room temperature spectrum would predict. On the other hand, the neutron spectrum in the moderator can deviate considerably from the spectrum in the fuel rod as is shown in a calculation by Volpe and Klein (5). Assuming a Maxwellian-type spectrum in both moderator and fuel regions but with each Maxwellian at a different neutron temperature, the average neutron velocity is different in the two regions and, consequently, the average cross section of foils placed in the two regions is different. Therefore, a significant correction factor should be applied to the activation measurements before they can be considered proportional to the neutron fluxes in each region.

To illustrate by means of an extreme example: if the effective neutron temperature is 200°C in the fuel and 20°C in the moderator, the cross sections of U^{235} and Dy foils are approximately 30% greater in the moderator than in the fuel. A correction of this magnitude should be applied to the activation ratio to obtain the true disadvantage factor. The large difference in neutron temperature between the fuel and moderator used in this example is probably not typical of most lattices. However, this example does indicate that the spatial dependence of neutron spectra may not be slight and, furthermore, that the determination of the disadvantage factor requires a substantial correction to the measured activation ratio. This correction should be applied to presently quoted disadvantage factors.

It would seem that any analytical method requiring that the predicted values of both the thermal utilization and activation ratios agree with experiment should include the spatial energy dependence of neutrons. Recently, Honeck 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 Pu²⁴⁰ relative to Pu²³⁹ and Pu²⁴¹ 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/cm^2 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 $\gamma(t)_{49}^{49}$ 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

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

activation integrals are denoted by the symbol I; i.e.,

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

where ϵ is the Cd cutoff energy (~0.45 ev). The Pu²⁴² contribution was neglected.

If the above expression is solved for the ratio of the Pu²⁴⁰ to Pu²³⁹ fission integrals, the result is the following:

$$\frac{\int_{0}^{\infty} \sigma_{f}^{40}(E)\phi(E) \, dE}{\int_{0}^{\infty} \sigma_{f}^{49}(E)\phi(E) \, dE} = P(t)_{40}^{49} \left\{ \left[\frac{N_{49}^{49}}{N_{40}^{40}} \gamma(t)_{49}^{40} - \frac{N_{40}^{49}}{N_{40}^{40}} R^{49} \right] - P(t)_{49}^{41} \frac{N_{40}^{41}}{N_{40}^{40}} \frac{I^{41}}{I^{49}R^{41}} \right\}$$
(1)

where R^i is the measured Cd ratio of the *i*th 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:

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

$$\int_{0}^{\infty} \sigma_{f}^{40}(E)\phi(E) dE = P(t)_{40}^{41}$$

$$\times \frac{\left[\frac{N_{40}^{41}}{N_{41}^{41}R^{41}} - \gamma(t)_{41}^{40}\right] + P(t)_{41}^{49}\frac{N_{41}^{49}I^{49}}{N_{41}^{41}I^{41}}\left[\frac{N_{40}^{49}}{N_{41}^{49}R^{49}} - \gamma(t)_{41}^{40}\right]}{\frac{N_{41}^{40}}{N_{41}^{41}}\left[\frac{N_{40}^{40}}{N_{41}^{40}} - \gamma(t)_{41}^{40}\right]}$$

$$(2)$$

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