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

In a previous letter (*1*), it was pointed out that the values quoted for the experimentally determined thermal-neutron disadvantage factors have not been corrected for the spatial dependence of neutron spectra, and this correction could substantially change the magnitude of these quantities. In reference to their experiments, Sher, Kouts, and Klein agree that such a correction should be applied to the measured disadvantage factors (*2*). However, their consideration of the magnitude of the correction deserves further discussion.

In ref. *1*, data are presented which show that the neutron temperature in a fuel rod in a light-water lattice is much larger than the physical temperature. For moderator-to-fuel ratios used in the light-water experiments, the moderator neutron temperature is also undoubtedly larger than its physical temperature but lower than the fuel temperature; therefore, a correction must be applied to the experimentally determined disadvantage factor. To illustrate that such a correction factor is indeed significant, an admittedly extreme example was chosen in which the fuel temperature was assumed to be 200°C and the moderator temperature 20°C, and the calculated correction factor is approximately 30%. This example is not meant to imply that a 30% correction factor should be applied to all measurements or that a 200°C difference in temperature exists between the fuel and moderator regions of the Sher, Kouts, and Klein experiments. In fact, the determination of a specific correction factor is dependent upon the physical properties of the lattice. Sher, Kouts, and Klein give evidence that for some of their lattices with ~1.3 wt.% U^{235} enrichment the neutron temperature difference between fuel and moderator is approximately 50°C, and about a 10% correction to the presently quoted experimental disadvantage factors is required (*2*). It would appear that for similar lattices with higher U^{235} enrichments, the neutron temperature difference between fuel and moderator would be greater, and could be 200°C for the appropriate fuel enrichment.

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A Note on the Measurement of Diffusion Parameters by the Pulsed-Neutron Source Technique

The pulsed-neutron technique has been used extensively to measure diffusion parameters in a variety of moderators. A description of the technique and an excellent summary of the present status of experiments may be found in a recent review by Beckurts (*1*). It may be seen from Beckurts' review that the values of the absorption cross section and of the diffusion constants obtained by the pulsed-neutron technique for the various moderators are quite consistent. However, some very puzzling discrepancies are observed between various measurements of the diffusion cooling constant, especially for crystalline moderators such as beryllium and graphite.

Beckurts proposes the following possible causes for these discrepancies: (1) the role of B^6 terms, (2) the effect of higher harmonics, and (3) the importance of the data-evaluation schemes. For the case of beryllium, at least, different laboratories measure different decay constants for the same value of the buckling (*2*, *3*). Such discrepancies cannot be blamed on B^6 terms. It is also difficult to see how the effect of harmonics may not be properly accounted for since, for a small cube of a moderator with low absorption, the first spatial harmonic decays almost twice as rapidly as the fundamental mode.

The purpose of this note is to propose another possible cause for the observed discrepancies in the measurements of decay constants. It appears that under certain conditions the decay of the neutron population out of a moderating assembly may never be strictly exponential. In this case, the "asymptotic decay constant" is not directly measurable and the diffusion cooling constant is not a well defined concept. The argument will be presented in some detail for the case of beryllium; however, the general conclusions should certainly be valid for crystalline moderators like graphite and beryllium oxide, and perhaps other materials as well.

The elastic transport cross section of beryllium has been computed by Bhandari (4) and is shown in Fig. 1. It changes very rapidly with energy and reaches its maximum value where the neutron wavelength is just equal to the distance between parallel planes of Miller's index (1, 0, 1). This corresponds to a neutron energy of 6.85 mev (millielectron volt) and to a velocity of 1.142×10^5 cm/sec. At this energy the transport cross section is about 18 barns and changes very little with moderator temperature. Below this energy the cross section drops stepwise to a value of 5.5 barns.

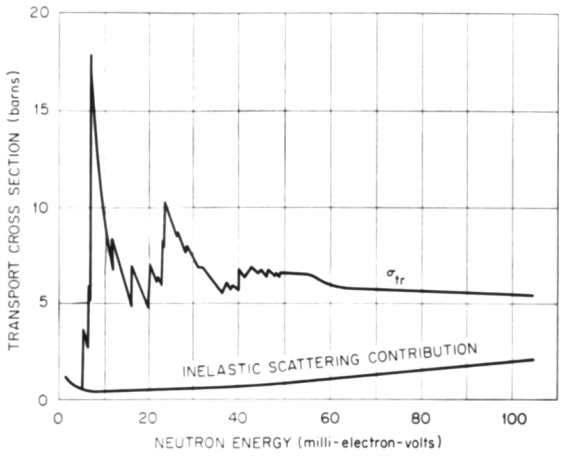

FIG. 1

Above 6.85 mev it is inversely proportional to the square of the neutron energy, up to about 11.6 mev.

In the energy region around 6.85 mev the inelastic scattering cross section of beryllium is a slowly varying function of energy, but it is strongly temperature dependent. At 300°K it has a value of 0.4 barns, and around this temperature it is proportional to the power 7/2 of the absolute temperature (*5*, *6*).

The neutrons with energies just above 6.85 mev have a very small diffusion coefficient. They have also a very small probability of changing energy, since the scattering is almost entirely elastic. Hence, in a small beryllium assembly neutrons of this energy are "trapped" and remain longer than "average" neutrons. For a neutron of energy 6.85 mev the probability per unit time to be transferred into another energy (to undergo inelastic scattering) is given by:

$$\alpha_{in} = vN\sigma_{in}$$
$$= 1.142 \times 10^2 \times 0.12 \times 0.4 = 5.5 \text{ msec}^{-1} \quad (\text{at } 300°\text{K})$$

where

v = velocity corresponding to 6.85 mev = 1.142×10^2 cm/msec

N = density of beryllium atoms = 0.12 atom/barn·cm,

σ_{in} = inelastic scattering cross section = 0.4 barn at 300°K.

The probability per unit time of being absorbed is independent of energy and equal to:

$$\alpha_a = v_0 N\sigma_a(v_0) = 2.2 \times 10^2 \times 0.12 \times 10^{-2} = 0.264 \text{ msec}^{-1}$$

where $\sigma_a(v_0) = 10^{-2}$ barns is the absorption cross section of beryllium for neutrons of velocity $v_0 = 2.2 \times 10^2$ cm/msec. Finally, the probability per unit time that a neutron of 6.85 mev will leak out of a beryllium assembly is given by:

$$\alpha_l = \frac{v}{3N\sigma_{tr}} B = \frac{1.142 \times 10^2}{3 \times 0.12 \times 18} B^2 = 17.6\, B^2 \text{ msec}^{-1}$$

where

σ_{tr} = 18 barns is the transport cross section,

B^2 = the buckling of the assembly in cm^{-2}.

So, if neutrons of all energies are introduced in a beryllium assembly of 0.072 cm^{-2} buckling, those neutrons that happen to have an energy of 6.85 mev would be removed (by leakage, absorption, or transfer to another energy) with a decay constant:

$$\alpha = \alpha_{in} + \alpha_a + \alpha_l = 5.5 + 0.264 + 1.267 = 7.0 \text{ msec}^{-1}.$$

Hence the asymptotic decay constant of such an assembly must be equal or smaller than 7 msec^{-1}. The decay constant of neutrons of higher energies will be larger because of increased leakage probability. The "asymptotic decay constant" experimentally obtained for this assembly is 8.5 ± 0.3 msec^{-1} (*2*). This clearly shows that these neutrons initially at an energy of 6.85 mev escape observation. This should be expected because of the limited precision of the measurements and since very few neutrons of the initial distribution have 6.85 mev. The peak of the cross section around 6.85 mev is very narrow. It can be easily computed that the transport cross section is larger than 15 barns only from 6.85 mev to 7.39 mev. If the initial distribution of the neutrons is Maxwellian (with a temperature of 300°K) about 0.5% of the neutrons are in the 0.54 mev wide trap above 6.85 mev.

It is difficult to determine after how long a time the decay will become exponential, if it ever does. This time is a sensitive function of the initial conditions and of the differential inelastic scattering cross sections. An approximate two-group computation suggests that the neutrons within the trap, 6.85 mev $< E <$ 7.39 mev will dominate the decay within less than one millisecond.[1] Since within this energy interval the transport cross section is inversely proportional to the square of the energy, the energy dependent decay constant of the trapped neutrons, out of the assembly considered, is given by:

$$\alpha(E) = \alpha_{in}(E_0) \frac{E^{1/2}}{E_0^{1/2}} + \alpha_a + \alpha_l(E_0) \frac{E^{5/2}}{E_0^{5/2}}$$

where E_0 = 6.85 mev, E is the trapped neutron energy in mev, and where the α's have been defined above. The decay constant of the neutrons of 7.39 mev is 7% larger than that of the neutrons of 6.85 mev, and, in between these two limits, the decay constant varies continuously with energy.

Under such conditions it is not possible to measure the asymptotic decay constant, since the lowest component of a composite decay curve can be isolated from the other components only if the smallest decay constant is appreciably smaller than the decay constants of the faster components.

Two remarks must now be made on the argument just presented: (1) The neutron trap above 6.85 mev is by far the most effective trap in beryllium, but it is by no means unique. In fact, the transport cross section exhibits a sharp peak every time the neutron wavelength is just equal to the distance between parallel planes of a Miller index. (2) The effectiveness of the traps is a very strong function of the moderator temperature because the inelastic cross section depends so sensitively on the moderator temperature. Above room temperature the trap effect in beryllium is probably negligible for all bucklings of practical interest. On the other hand, at a temperature of 273°K the probability for inelastic scattering out of the 6.85 mev trap becomes α_{in} = 4.0 msec^{-1} (instead of 5.5 msec^{-1} at 300°K). The decay constant out of the trap at 6.85 mev for an assembly of 0.054 cm^{-2} buckling becomes α = 4.0 + 0.264 + 0.950 = 5.21

[1] Pointed out by Dr. A. F. Henry.

msec^{-1}. The experimentally measured "asymptotic decay constant" for this assembly at 273°K is 6.4 ± 0.2 msec^{-1} (*7*). Even for this larger assembly, at the lower temperature, the contribution of the trapped neutrons to the decay is not observed.

In conclusion, it must be said that the trap effect does not appreciably affect the measurements of absorption cross sections and diffusion constants by the pulsed-neutron-source method, at least at room temperature and above, since these parameters can be obtained from measurements on large assemblies for which the over-all leakage probability is smaller than the energy transfer probability out of the trap. In these assemblies a true equilibrium energy distribution is established and an asymptotic decay constant may be measured.

However, the determination of the diffusion cooling constant and of terms proportional to B^6 or B^8 must rely on measurements of the asymptotic decay constants in small assemblies. For small assemblies the over-all leakage probability is comparable to the inelastic scattering probability out of the trap, and the instantaneous decay constant keeps decreasing almost until the last neutron has left the assembly. Under these conditions, the asymptotic decay constant is not a measurable quantity and the diffusion cooling constant is not a very useful concept.

The "measured" asymptotic decay constant of a small assembly is a function of the intensity of the pulsed-neutron source available, of the practical limitation imposed by the background conditions, and of the data evaluation scheme. It also depends to a lesser extent on the energy of the pulsed-neutron source, the duration of the pulses, and the geometry of the experiment, as such factors determine the ratio of the number of neutrons trapped to the total number of neutrons at the initial time. Therefore, it does not appear too surprising that as the precision of the measurements increases the discrepancy between the "asymptotic decay constants" obtained for the same assembly but under different experimental conditions also increases.

Finally, it must be noted that the trap effect is not taken properly into account by the usual diffusion cooling calculations. Indeed, because of mathematical difficulties, these calculations always assume a smooth variation of the transport cross section (E^α-law or incoherent approximation) and an equilibrium neutron spectrum that can be expressed as a Maxwellian, or a Maxwellian multiplied by a finite number of terms of a power expansion, or a Wigner-Wilkins spectrum (*8–10*), whereas the equilibrium spectrum of the trapped neutrons is better approximated by an energy δ-function.

The author would like to acknowledge some very helpful comments of Dr. A. F. Henry in evaluating some of the consequences of the trap effect described in this note.

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The Resonance Integral For Uranium Metal and Oxide

Smith, *et al.* (*1*) have made a careful study of the relative epicadmium absorption in uranium metal and oxide. Their measurements yield a larger ratio of metal to oxide resonance absorption than was calculated from the formulas obtained earlier at our laboratory (*2*). As our measuring technique has much improved since the old experiments, a minor set of new measurements has been performed.

The method of measurement was similar to the one used for the study of thorium metal (*3*). Thus the calibration in barns was obtained by means of the dilute resonance integral for gold and the thermal cross sections for gold and uranium (for the old measurements (*2*) the dilute resonance integral for uranium was used). The arrangement for the irradiations is illustrated in Fig. 1. The neutron flux was monitored with gold foils. They were placed about 12 cm above the samples to avoid screening effects from the uranium resonances. The diameters of the samples were 28 and 10 mm for the metal, and 17 and 12.5 mm, respectively, for the oxide.

The measurements for the metal yield values about 4.5% larger than those calculated from the formula given in (*2*). For the oxide rods, however, the resonance integrals obtained agreed to within 0.5% with the earlier results. The old formula for the oxide, $RI = 4.15 + 26.6\ \sqrt{S/M}$, may therefore still be used. For the metal the following new expression should be recommended

$$RI = 2.95 + 25.8\ \sqrt{S/M}$$

Taking into account the uncertainties in the spectrum corrections (compare (*3*)) and in the cross sections used for the absolute calibration, the standard deviations for the