FIG. 5. Effect of organic additives on CO_2 sintering of UO_2 powder.

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A Remark on the Measurement of the Diffusion Coefficient for Thermal Neutrons*

In a recent article in this Journal, Baumann (1) has reported a measurement of the diffusion coefficient (D) of thermal neutrons in heavy water as a function of temperature in the range 20°C to 220°C. The technique used was to measure the diffusion length (L) as a function of copper absorption cross section (Σ_{Cu}). The copper was added in the form of thin tubes. The reciprocal diffusion coefficient is the slope of curve of $1/L^2$ vs. $\bar{\Sigma}_a$ at $\bar{\Sigma}_a = 0$. Here $\bar{\Sigma}_a$ is the Maxwellian (at the moderator temperature) averaged total absorption (copper plus heavy water) cross section. Several corrections must be made to the raw experimental data and these have been made with great care. The correction for neutron spectrum, however, was made on an intuitive basis and overestimates the correction. In this note we show how the correction should be made, estimate the size of the

correction from a theoretical scattering model for heavy water, and recompute D from Baumann's data.

Baumann recognizes that the neutron spectrum is distorted from the equilibrium Maxwellian spectrum at the temperature of the medium, T_0, by absorption and diffusion. He then characterizes this distorted spectrum by a Maxwellian at a "neutron temperature" T_n and uses a formula given by Coveyou, Bate, and Osborn (2) to relate T_n to T_0 and $\bar{\Sigma}_a$. Using an empirical expression for the Maxwellian averaged D as a function of temperature, and the $\sqrt{T_0/T_n}$ variation for the absorption cross section, the diffusion length is corrected back to a neutron temperature equal to the moderator temperature. Our objections to this procedure are:

1. The formula of Coveyou was derived for slowing down spectra where most of the distortion is caused by the $1/E$ tail of the spectrum. In the present experiment there is no slowing-down source, the spectra have no $1/E$ tail, and the distortion for a given absorption is much smaller (3-6).

2. The actual spectrum distortion can only be approximated by a shifted Maxwellian. Thus, while it may be possible to describe the shift of one parameter (say $\bar{\Sigma}_a$) by an effective Maxwellian temperature shift, the shift of another parameter (D) may require a different Maxwellian temperature shift.

The first of these objections is the most important since it determines the size of the correction. The second objection merely states that the form of the correction used was intuitive and lacked a firm theoretical foundation. An appropriate treatment of this correction has been discussed by Honeck (7). Koppel and Starr (8) have treated a similar measurement in light water in the manner described below.

The reciprocal diffusion length in a poisoned moderator can be expressed by

$$\frac{1}{L^2} = \alpha_1 \Sigma_{a_0} - \alpha_2 \Sigma_{a_0}^2 + \cdots \qquad (1)$$

where Σ_{a_0} is the total absorption cross section at an energy equal to kT_0 and the α's are coefficients that depend upon the scattering properties of the moderator. We will ignore the higher order terms α_3, α_4, etc. since, in the present experiment, the maximum Σ_{a_0} is quite small and the second term in (1) is only a few percent of the first. In other experiments with larger Σ_{a_0} the higher order terms may be important. Rewrite Eq. (1) in the form:

$$\frac{1}{L^2} = \frac{\sqrt{\pi}}{2} \frac{\Sigma_{a_0}}{D} \left(1 - \frac{\alpha_2}{\alpha_1} \Sigma_{a_0}\right). \tag{1'}$$

The desired correction for changes in neutron spectrum (and also for transport effects) is the term $(\alpha_2/\alpha_1)\Sigma_{a_0}$. For the range of Σ_{a_0} under consideration here, this correction is equivalent in form to that used by Baumann but is only about $\frac{1}{3}$ as large.

If $\Sigma_a(E)$ varies like $1/v$ the coefficients α_1 and α_2 are simply related to the more familiar parameters D_0 and C for the pulsed neutron experiment. The time decay constant λ in a pulsed assembly with buckling B^2 is given by

$$\lambda = v_0 \Sigma_{a_0} + D_0 B^2 - C B^4 + \cdots \tag{2}$$

where v_0 is the neutron velocity at energy kT_0, C is the diffusion cooling coefficient, and $D_0 = 2v_0 D/\sqrt{\pi}$. Then the diffusion length parameters are related to the pulsed parameters by

$$\alpha_1 = \frac{v_0}{D_0} = \frac{\sqrt{\pi}}{2D} \tag{3a}$$

$$\alpha_2 = \frac{v_0^2 C}{D_0^3} = \frac{\pi^{3/2}}{8} \frac{C}{v_0 D^3} \tag{3b}$$

$$\frac{\alpha_2}{\alpha_1} = \frac{v_0 C}{D_0^2} = \frac{\pi}{4} \frac{C}{v_0 D^2} \tag{3c}$$

Numerical values of α_1 and α_2 were computed in the following way. Scattering kernels for heavy water at various temperatures were generated using an incoherent model of the Nelkin type which represents the internal motions of the deuterium atoms by four discrete harmonic oscillators and a gaslike translation of the D_2O molecule. This model has been previously reported (9). The results compared with D_0 and C measured by Ganguly and Waltner (11) indicates that this model correctly predicts his values of λ within the experimental uncertainties. We then believe our values of α_2/α_1 are correct to within about 10%. Since the correction term itself is of the order of $\frac{1}{2}\%$ or less, the error in D due to uncertainty in α_2/α_1 is negligible. Given the scattering kernels (P_0 and P_1 components), the diffusion length is calculated for a given $1/v$ absorption concentration by direct solution of the transport equation (7). The values of L^2 for a series of Σ_{a_0} are then fit to a series like (1) to obtain α_1, α_2, α_3, etc. Our results are listed in Table I. Values at the experimental temperatures were taken from a smooth curve through the computed values.

We next calculate the value of D at each temperature from the rearranged form of Eq. (1')

$$D^{-1} = \sum_{n=1} W_n$$

$$\cdot \left\{ (\bar{\Sigma}_{Cu_n} + \bar{\Sigma}_w) L_n^2 \left[1 - \frac{\alpha_2}{\alpha_1} \frac{2}{\sqrt{\pi}} (\bar{\Sigma}_{Cu_n} + \bar{\Sigma}_w) \right] \right\}^{-1} \tag{4}$$

TABLE I

COMPUTED DIFFUSION PARAMETERS OF D_2O

T (°C)	20	77	127	177
ρ (gm/cc)	1.106	1.080	1.039	0.987
α_1 (cm^{-1})	1.069	1.021	0.967	0.906
α_2	2.63	2.35	2.13	2.03
α_2/α_1 (cm)	2.46	2.30	2.20	2.24
D_0 (10^5 cm^2/sec)	2.057	2.354	2.658	3.011
C (10^5 cm^4/sec)	4.73	5.31	6.05	7.43

TABLE II

DIFFUSION COEFFICIENT (cm)

	T (°C)	D (99.3%)	D (100%)
Uncorrected for neutron spectrum (From (1))	20	0.827 ± 0.010	
	100	0.897 ± 0.011	
	165	0.974 ± 0.012	
	220	1.073 ± 0.013	
Corrected for neutron spectrum by Baumann (From (1))	20	0.816 ± 0.010	0.841 ± 0.010
	100	0.887 ± 0.011	0.912 ± 0.011
	165	0.963 ± 0.012	0.989 ± 0.012
	220	1.063 ± 0.013	1.091 ± 0.013
Corrected for neutron spectrum with theoretical values of α_2/α_1	20	0.823 ± 0.010	0.848 ± 0.010
	100	0.895 ± 0.011	0.920 ± 0.011
	165	0.969 ± 0.012	0.995 ± 0.012
	220	1.070 ± 0.013	1.098 ± 0.013
Kash and Woods (12), corrected for neutron spectrum with theoretical values of α_2/α_1 (D_2O here is 99.6%)	23	0.842 ± 0.013	0.855 ± 0.013
Dexter et al. (13), corrected for neutron spectrum with theoretical values of α_2/α_1	18		0.84 ± 0.03
			0.87 ± 0.02

where n refers to the nth poison concentration, L^2 is the measured diffusion area corrected for everything except neutron temperature and H_2O contamination, $\bar{\Sigma}_{Cu}$ and $\bar{\Sigma}_w$ are Maxwellian averaged (at the moderator temperature) absorption cross sections for copper and D_2O, and W is a normalized weight proportional to the reciprocal of the percentage experimental errors reported by Baumann. The value of $\bar{\Sigma}_w$ was selected such that the variance of the individual terms in (4) was minimized. The results of these calculations are given in Table II along with those of Baumann. We note that all corrections are small compared to the experimental error. However, neglect of the spectrum correction, or use of an incorrect one, leads to a systematic error. Baumann's correction for neutron temperature decreases D by about 0.010, while our correction reduces D by about 0.003, a factor of 3 lower.

In Table II we also give the result of applying our analysis to the data of Kash and Woods (12), and the data of Dexter et al. (13). Dexter's most highly poisoned measurement is omitted because an asymptotic distribution was not achieved, the reported L being less than the scattering mean free path.

Baumann has used the formula

$$\frac{1}{D} = \frac{1-f}{D(D_2O)} + \frac{f}{D(H_2O)} \qquad (5)$$

to correct for light water contamination. Here, f is the mole fraction of light water. A more accurate expression for small values of f can be written

$$\overline{\frac{1}{D}} = \frac{1-f}{D'(D_2O)} + \frac{f}{D(H_2O)}$$
$$\cdot \int_0^\infty dE\, M(E) \left[\frac{\lambda(E)}{\langle\lambda\rangle}\right]^2_{D_2O} \left[\frac{\langle\lambda\rangle}{\lambda(E)}\right]_{H_2O} \qquad (6)$$

where

$$\lambda(E) = 1/\Sigma_{tr}(E)$$

$$\langle\lambda\rangle = \int dE\, M(E)\lambda(E).$$

The integral in (6) was evaluated and found to be 1.027 and independent of temperature. Since the second term in (5) is only 3% of the first term for this experiment, the use of Eq. (5) to correct for light water will be accurate to better than 0.1%.

A final question arises as to whether a spectrum correction for an infinite homogeneous medium can be applied to a heterogeneous lattice measurement. We have attempted to answer this question by calculating the actual spatial and energy distribution of the flux in Baumann's 11.56 cm pitch lattice at room temperature (the most highly absorbing case). For this case we find that the non-Maxwellian correction reduces $1/L^2$ by 0.650% for a homogeneous medium and by 0.621% for the heterogeneous lattice. Thus the usual diffusion cooling (heating in this case) correction can be applied in this case. It should be noted, however, that the lattices considered here are very dilute and the copper tube is quite thin. The variation of the average velocity is only 3% across the cell.

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Re: A Remark on the Measurement of the Diffusion Coefficient for Thermal Neutrons*

Dr. Honeck's new calculations of the spectral hardening correction for my measurements of the thermal diffusion coefficient for D_2O make an interesting and valuable addition to the study. Since my original paper was primarily experimental, the analysis was deliberately kept at the simplest level consistent with the precision of the data, but the corrections were given separately to make possible later improvements such as those suggested. Like Dr. Honeck, I had found that use of more precise terms for the H_2O and heterogeneity corrections made essentially no difference in the numerical results. Likewise a test of the spectral hardening term against H_2O data had given excellent results as had the test against Dexter's D_2O data.

The rather large discrepancy between the term used and Dr. Honeck's calculations for D_2O is, in fact, rather surprising. Diffusion cooling terms calculated by my method and by Honeck for H_2O and graphite are in good agreement both with each other and with experiments. Calculations by my method also agree well with the average of the diffusion cooling measurements for beryllium. Further, in the case of D_2O my correction reconciles Dexter's data somewhat better than does Honeck's. On the other hand Honeck's corrections appear to rest on much more secure theoretical grounds and also give better agreement with the measured diffusion cooling term for D_2O in pulsed neutron experiments.

To help resolve the discrepancy we are planning experiments with deep poisoning cases. Monte Carlo calculations are also underway to determine the effect of epithermal neutron feed on the equilibrium thermal energy distribution in poisoned D_2O.

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