Thermal Neutron Spectra in H₂O for Plane Geometry

An attempt was made to calculate thermal-neutron spectra in H_2O occurring in pulsed experiments. At first the energy-dependent Milne problem was treated, using Nelkin's scattering kernel for water.¹ The calculated energy distribution of neutrons at the interface was compared with a measured leakage spectrum from a large water geometry.² Because of the good agreement between theory and experiment, we then computed the leakage spectra of thermalized-neutron bursts for slabs of finite thickness and again obtained good agreement with corresponding experiments.^{2,3} The leakage spectra as well as the spectra in the center of the slabs show direct evidence for the diffusion cooling effect.

The energy-dependent Milne problem

To get the energy-dependent neutron flux at the boundary of a half-space, we solved the stationary Boltzmann equation in a P_1 -approximation, assuming a Maxwellian source at infinity. Our formalism is quite similar to that one of Conkie⁴ with two exceptions:

1. Instead of the Wigner-Wilkins kernel⁵ for monoatomic hydrogen we used Nelkin's¹ scattering kernel for water in our calculations.

2. We tried to describe the energy dependence with the aid of the functions $\omega_n(x) = x \cdot e^{-x} L_n^{(1)}(x)$ which are orthonormal in $0 \le x < \infty$ with the adjoint functions $\omega_n^+(x) = L_n^{(1)}(x)$. The $L_n^{(1)}(x)$ are normalized Laguerre polynomials of order one (see e.g. Reference 6). Conkie transformed the energy scale $0 \le E \le \infty$ into the finite range $0 \le y \le 1$ and then used Tchebycheff polynomials $T_m(y)$ instead of our $L_n^{(1)}(x)$. It seems to us that the second

difference is not a serious one but the use of Laguerre polynomials probably simplifies the calculations.

Our final result for the angular-dependent neutron flux after solving the matrix and eigenvalue equations, satisfying the P_1 - boundary conditions is the following one, written in the dimensionless variable $\epsilon = E/kT$: $\phi(z,\epsilon,\theta)$

$$= \left\{ \left[(q_o + z)\omega_o(\epsilon) + 3\cos\theta \sum_{p=0}^{p} C_{po} \omega_p(\epsilon) \right] + \sum_{j=1}^{p} e^{-g_j z} a_j \left[\sum_{p=0}^{p} (F_{pj}\omega_p(\epsilon) + 3\cos\theta C_{pj}\omega_p(\epsilon)) \right] \right\} .$$

Figure 1 shows the spectrum at the interface z = 0 compared with the measured leakage spectrum from a large water geometry $(15 \times 15 \times 15 \text{ cm} \text{ cube})$ taken from the work of Beckurts² together with the Maxwellian distribution at a moderator temperature that exists far away from the surface. Due to the preferential leakage of neutrons with high energies, the spectrum at the boundary is shifted towards higher energies.

Pulsed experiments

In the next step we tried to solve the timedependent source-free Boltzmann equation with a similar formalism. We only were interested in the asymptotic part of the time-dependent neutron flux in a pulsed experiment decaying with $e^{-\alpha t}$. The part of the Boltzmann equation proportional to $\frac{\alpha}{v} \sim \frac{\alpha}{\sqrt{\epsilon}}$ arising from the time dependence is easily taken into account since one is able to calculate the matrix elements $\int_0^\infty L_m^{(1)}$ (ϵ) $\frac{1}{\sqrt{\epsilon}}$ ϵ . $e^{-\epsilon}L_n^{(1)}(\epsilon)d\epsilon$ analytically (see Reference 6). All other matrices are just the same as in the Milne problem. If the values of the absorption cross section and the decay constant α are given, the computation of the problem is quite direct. At the end of the calculations the boundary condition will give us the thickness of the slab.

The energy-dependent asymptotic flux has the following form:

$$\phi(z,\epsilon) = \cos(Bz) \sum_{p=0}^{P} F_{po} \omega_{p}(\epsilon)$$

+
$$\sum_{j=1}^{r} a_j \cosh(g_j z) \sum_{p=0}^{r} F_{pj} \omega_p(\epsilon)$$
.

The measured spectra in Figure 2 belong to a $5 \times 5 \times 5$ cm cube² with a decay constant = 34 700 sec⁻¹ (buckling $B^2 = 0.94$ cm⁻²). In our calculations for an "essentially equivalent" slab we have used a value $\alpha = 34\ 900\ \text{sec}^{-1}$ ($B^2 = 0.935\ \text{cm}^{-2}$). One cannot fit exactly the experimental and theoretical values of the pair (α, B^2) because there exists a discrepancy in the diffusion constant

¹ M. S. NELKIN, *Phys. Rev.*, 119, 741 (1960).

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³W. REICHARDT, Private communication.

⁴W. R. CONKIE, Nucl. Sci. and Eng., 7, 295 (1960).

⁵E. P. WIGNER and J. E. WILKINS, Jr. A.E.C.D. - 2275 948).

⁶W. HÄFELE and L. DRESNER, Nucl. Sci. and Eng., 7, 304 (1959).



Fig. 1. Velocity-dependent neutron flux at the boundary of the halfspace compared with the thermal leakage spectrum in a large water geometry.



Fig. 3. Energy-dependent flux in the center and at the boundary of a 3-cm slab compared with experimental results.



Fig. 2. Calculated velocity-dependent neutron fluxes of a pulsed experiment in a 2.7-cm slab compared with the results of an essentially equivalent experiment.



Fig. 4. Calculated and measured leakage spectra of a 2cm slab compared with the leakage spectrum of a halfspace.

between theory and experiment. The theoretical curves for the velocity-dependent neutron flux in a 2.7-cm slab are in good agreement with the measurements in a $5 \times 5 \times 5$ cm cube in the center as well as at the boundary of the media (Fig. 2).

In another experiment³ the assumption of slab geometry has been fulfilled reasonably well $(2 \times 20 \times 20 \text{ cm} \text{ and } 3 \times 20 \times 20 \text{ cm} \text{ vessels}).$

In Figure 3 is shown the flux in the center and at the boundary of a 3-cm slab together with the experimental results. The leakage spectrum is shifted markedly towards higher energies compared with the spectrum in the center. In Figure 4 the calculated and measured spectra at the boundary of a 2-cm slab are compared. The solution at the boundary of the halfspace, which is also drawn in Figure 4, shows that the leakage spectrum of a small geometry is cooler than that of a large one. Both the spectra in the center and at the interface show this influence of the diffusion cooling effect (see e.g. Reference 2). Our calculations indicate that the energy dependence of thermal-neutron spectra belonging to the asymptotic flux in pulsed water geometries may be calculated very well with our formalism. A comparison of our solution of the Milne problem with Conkie's result shows that it is necessary to use an appropriate scattering kernel for water that takes into account the effects of chemical binding.

A detailed description will be published in Nukleonik.

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Received July 22, 1963

Decomposition of Defects by Neutrons in Reirradiated Graphite*

Identification of reactions causing defect decomposition during neutron irradiation still remains an important problem in radiation studies of graphite. "Radiation annealing" experiments in which a sample originally irradiated at one temperature is reirradiated at a higher temperature show that more damage is removed by this technique than by thermal annealing alone¹. Since these experiments involve simultaneous heating and reirradiation, it is not clear whether the effect represents defect decomposition, radiation enhancement of the thermal annealing reactions, or a combination of the two. The effects can be separated by performing the reirradiations and anneals alternately. When graphite is thermally annealed after each of a series of exposures at a fixed temperature, damage produced by the first irradiation can be removed, maintained at a constant value, or increased, by varying the exposure between anneals.

Studies were made on AGOT graphite samples subjected to more than twenty cycles of 30 C reirradiations followed by 350 C out-of-pile anneals. When the exposure between anneals was smaller than some critical value (determined by the temperature of the anneal and the total damage) each reirradiation and annealing cycle removed more net damage than the reirradiation produced. For exposures at the critical values, the reirradiation and annealing cycle removed a quantity of damage equal to that produced by the reirradiation. When the exposure between anneals exceeded the critical value, only a fraction of the damage produced by the reirradiation was removed during the following anneal. The observed effects are conclusive evidence that the reirradiations involve competitive reactions between production and decomposition of interstitial complexes. Critical exposure values and details on how the damage changes with exposure between anneals are given in reference².

Net changes in damage were determined from radiation-induced property changes, which were assumed to vary monotonically with the total defect concentration. This assumption is valid for c-axis changes and stored-energy changes when simple defects are formed^{3,4,5}. If the damage involves several different clusters of interstitials, variations in the proportionality constant per interstitial carbon atom must be considered. It is unlikely, however, that contributions to the c-axis due to changing proportionality constants in different species will parallel proportionality constant changes in the stored energy. Simultaneous measurements of c-axis, stored-energy and dimensional

^{*}This work was performed under the auspices of the U.S. Atomic Energy Commission.

¹A review of "radiation annealing" phenomena is given in *Nuclear Graphite* (Academic Press, New York (1962) 376-382), edited by R. E. NIGHTINGALE.

²D. G. SCHWEITZER and R. M. SINGER, "Effect of Irradiation Temperature and Annealing Temperature on Expansions and Contractions in Alternately Irradiated and Annealed Graphite". *ANS Transactions*, Vol 6, No 2, 383 (Nov. 1963).

³G. J. DIENES and G. H. VINEYARD, *Radiation Effects* in Solids Interscience Publishers Inc., New York (1957).

⁴D. G. SCHWELTZER, *Phys. Rev.* **128** 556 (1962).

⁵D. G. SCHWEITZER, "Determination of the Single Interstitial Migration Energy from Stored Energy and Thermal Resistivity Changes in Irradiated Graphite". Submitted for publication in *Carbon*.