The c-axis and dimensional points plotted from 0 to ~ 1000 MWd (1 MWd = 7×10^{16} nvt for neutrons with energies above 0.6 MeV) denote changes prior to the first anneal. Data between 1000 MWd and 2000 MWd were accumulated over a period of several years. At the end of the program, unirradiated stock material was continuously irradiated to check that irradiation and annealing conditions remained the same. The enclosed points show the data obtained. All values shown for exposures exceeding ~ 1000 MWd represent measurements after the 350 C anneal. Stored energy was determined from heats of combustion measurements made by the National Bureau of Standards. These measurements consumed appreciable portions of the sample and were made less frequently than those of the other properties. The variation in magnitude of exposures between anneals is shown by the position of the dimensional data points along the abscissa. Length changes were measured before and after most anneals and are shown in Figure 2. The dashed lines are estimates of the values not measured before annealing. The sample and aliquots were annealed at 350 C for 4 hours. No changes outside of experimental error occurred when the time of annealing was varied from 1 hour to 16 hours. C-axis values after irradiation at 30 C and prior to annealing did not change in aliquots that stood at room temperature for six years.

The decomposition during reirradiations demonstrated by these studies involves species formed during the anneals. Stored-energy and c-axis changes during the first irradiation can be quantitatively accounted for by a model which requires that the data reflect properties of one interstitial entity[4,5]. This model does not appear to account for experimental data if radiation decomposition reactions for the interstitial are included. It is therefore concluded that the interstitial formed during the first irradiation is more stable towards irradiation than are the clusters formed during anneals.

The mechanism for cluster break-up during reirradiations cannot be determined from these studies. It is possible that decomposition results from collision effects of energetic neutrons or from "hot atom"[1] reactions.

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Decoupling the Energy-Dependent Spherical Harmonics Equations*

We show how to transform the discrete energy approximation of the spherical harmonics equations for the slowing-down of both gamma rays and elastically scattered neutrons to a system of spatially decoupled simultaneous differential equations. The solution of the spherical harmonics moments problem in very high orders of approximation is made feasible by decoupling, since the solution of a large number of decoupled equations requires no more computational effort than the simultaneous solution of a modest number of coupled equations.

This method could be applied advantageously to the study of the deep penetration of gamma rays where the strong forward peaking of the photon distribution necessitates the use of high-order angular expansions. It could also be used to calculate neutron distributions in highly absorbing media and in light moderators in which the mathematically simple age approximation is inadequate.

In the spherical harmonics approximation of order L, the space-angle-energy particle distribution, $\Psi(z, \omega, \phi, u)$, which we restrict to an even function of the azimuthal angle, ϕ, is represented as

$$\Psi(z, \omega, \phi, u) = \sum_{\ell=0}^{L} \sum_{m=0}^{\ell} \sqrt{\frac{2\ell + 1}{4\pi} \frac{(\ell - m)!}{(\ell + m)!}}$$

$$\cdot \cos m\phi \, (1 - \omega^2)^{\frac{m}{2}} \, P_{\ell m}(\omega) \Psi_{\ell m}(z, u), \qquad (1)$$

where

z is the depth coordinate;

$\hat{\Omega} = \hat{i}(1 - \omega^2)^{\frac{1}{2}} \cos \phi + \hat{j}(1 - \omega^2)^{\frac{1}{2}} \sin \phi + \hat{k}\omega$ is a unit vector in the direction of particle motion;

$\hat{i}$, $\hat{j}$, and $\hat{k}$ are the unit vectors parallel to the x, y, and z Cartesian axes;

$\cos^{-1}\omega$ and ϕ are the polar and azimuthal angles on the surface of a unit sphere with its axis along the z-direction;

$P_{\ell m}(\omega)$ is the associated Legendre polynomial defined by

$$P_{\ell m}(\omega) = (1 - \omega^2)^{\frac{1}{2}} \frac{d^m P_{\ell}(\omega)}{d\omega^m},$$

where $P_\ell(\omega)$ denotes an ℓ-th degree Legendre polynomial,[1] u is the energy variable (lethargy for neutrons, wavelength for gamma rays);

$$\Psi_{\ell m}(z, u) = \sqrt{\frac{2\ell + 1}{4\pi} \frac{(\ell - m)!}{(\ell + m)!}}$$

$$\cdot \int_0^{2\pi} d\phi \cos m\phi \int_{-1}^{+1} d\omega P_{\ell m}(\omega) \Psi(z, \omega, \phi, u) \ . \tag{2}$$

The spherical harmonics moments, $\Psi_{\ell m}(z, u)$, satisfy the system

$$\sum_\lambda \left[\gamma_{m\ell\lambda} \frac{\partial}{\partial z} + \sigma_T(u) \delta_{\ell\lambda} \right] \Psi_{\lambda m}(z, u)$$

$$= \delta_{mo} \int_{u_o}^u f_\ell(u, u') \Psi_{\ell_0}(z, u') du'$$

$$+ S_{\ell m}(z, u) \begin{cases} 0 \leqq \ell \leqq L \\ 0 \leqq m \leqq \ell \end{cases} \tag{3}$$

where

$$\gamma_{m\ell\lambda} = \sqrt{\frac{(\ell - m)(\ell + m)}{(2\ell - 1)(2\ell + 1)}} \ \delta_{\lambda, \ell - 1}$$

$$+ \sqrt{\frac{(\lambda - m)(\lambda + m)}{(2\lambda - 1)(2\lambda + 1)}} \ \delta_{\lambda, \ell + 1} \ ; \tag{4}$$

$\sigma_T(u)$ is the macroscopic cross-section for all interactions;

$f(\mu | u, u')$ is angle-energy cross section for scattering a particle with incident energy u' and emergent energy u through an angle $\cos^{-1}\mu$;

$$f_\ell(u, u') = 2\pi \int_{-1}^{+1} P_\ell(\mu) f(\mu | u, u') d\mu .$$

$S_{\ell m}(z, u)$ is an expansion coefficient of the source distribution, $S(z, \omega, \phi, u)$, calculated in the same way as $\Psi_{\ell m}(z, u)$ in Equation (2).

Introducing a uniform mesh quadrature for the energy integral in Equation (3) and an obvious matrix notation, we obtain

$$\left\{ \gamma_m \frac{\partial}{\partial z} + \left[\sigma_T(u_i) - \delta_{mo} \alpha_i C \right] I \right\} \vec{\Psi}_m(z, u_i)$$

$$= \delta_{mo} \sum_{j=0}^{i-1} \alpha_j f(u_i, u_j) \vec{\Psi}_0(z, u_j) + \vec{S}_m(z, u_i) \ , \tag{5}$$

where $f_\ell(u_i, u_i) = C(u_i)$, a function independent of ℓ. (This is seen to be true by examining the forms of $f(\mu | u, u')$ for neutrons and gamma rays in References 2 and 3.)

Note from Equation (4) that γ_m is a nonsingular, symmetric, $(L - m + 1)^2$ matrix. Thus it is diagonalized by the unitary matrix, ρ_m, whose columns are its eigenvectors normalized to unit length. Thus

$$\gamma_m \rho_m = \rho_m \Gamma_m \ , \qquad \rho_m^T \rho_m = I, \tag{6}$$

where Γ_m is a diagonal matrix whose elements are the eigenvalues of γ_m .

Now, letting $\vec{\phi}_m = \rho_m^T \vec{\psi}_m$ and $\vec{\xi}_m = \rho_m^T \vec{S}_m$, we multiply Equation (5) from the left by ρ_m^T, and make use of Equation (6), to obtain

$$\left\{ \Gamma_m \frac{\partial}{\partial z} + \left[\sigma_T(u_i) - \delta_{mo} \alpha_i C \right] I \right\} \vec{\phi}_m(z, u_i)$$

$$= \delta_{mo} \sum_{j=0}^{i-1} \alpha_j \rho_m^T f(u_i, u_j) \rho_m \vec{\phi}_0(z, u_j)$$

$$+ \vec{\xi}_m(z, u_i) . \tag{7}$$

Since the slowing-down source which contributes to $\vec{\phi}_m(z, u_i)$ is determined by the $\vec{\phi}_0(z, u_j)$, $j < i$, this source is explicitly known when it is required to solve for the $\vec{\phi}_m(z, u_i)$. Thus the right hand member of Equation (7) is explicitly known then. Also the matrix operating on $\vec{\phi}_m(z, u_i)$ is diagonal. Hence the equations which determine the elements of a given $\vec{\phi}_m(z, u_i)$ are completely decoupled from each other and can be solved independently.

To determine the elements of ρ_m, note that a representative equation in system (6) is

$$\sum_\lambda \gamma_{m\ell\lambda} \rho_{m\lambda k} = \rho_{m\ell k} \Gamma_{mk} \ ,$$

$$k = 1, 2, \ldots, L - m + 1 . \tag{8}$$

Equation (8) is identical in form with the recursion formula for normalized associated Legendre polynomials.[1] Hence

[1] E. T. WHITTAKER and G. N. WATSON, *Modern Analysis*, 4th ed, pp. 308-309, Cambridge (1927).

[2] R. E. MARSHAK, *Reviews of Modern Physics*, 19, 3, p. 188 (July, 1947).

[3] W. HEITLER, *The Quantum Theory of Radiation*, Chapter V, 3rd ed., Oxford (1954).

[4] B. DAVISON, *Neutron Transport Theory*, p. 130, Oxford (1957).

$$\rho_{m\lambda k} = C_{mk} \sqrt{\frac{2\lambda + 1}{4\pi} \frac{(\lambda - m)!}{(\lambda + m)!}} \, P_{\lambda m}(\Gamma_{mk}). \quad (9)$$

The C_{mk} are determined by the normalization of the columns of ρ_m to unit length. Thus

$$C_{mk}^{-2} = \sum_{\lambda=0}^{L} \frac{2\lambda + 1}{4\pi} \frac{(\lambda - m)!}{(\lambda + m)!} \, P_{\lambda m}^{2}(\Gamma_{mk}). \quad (10)$$

The eigenvalues, Γ_{mk}, are determined by the requirement that, in an L-th order truncation,

$$\rho_{m, L+1, k} = 0 \quad \text{or} \quad P_{L+1, m}(\Gamma_{mk}) = 0.$$

The series for C_{ok}^{-2} can be summed by noting that[4]

$$\sum_{\lambda=0}^{L} (2\lambda + 1) \, P_{\lambda}(\omega_1) P_{\lambda}(\omega_2) \quad (11)$$

$$= \frac{L + 1}{\omega_1 - \omega_2} [P_{L+1}(\omega_1) P_{L}(\omega_2) - P_{L+1}(\omega_2) P_{L}(\omega_1)].$$

Taking limits as $\omega_2 \to \omega_1 = \Gamma_{ok}$ in Equation (11), and using Equation (10), we find

$$\sum_{\lambda=0}^{L} (2\lambda + 1) \, P_{\lambda}^{2}(\Gamma_{ok})$$

$$= (L + 1) \, P'_{L+1}(\Gamma_{ok}) \, P_{L}(\Gamma_{ok}). \quad (12)$$

But

$$P'_{L+1}(\Gamma_{ok}) = (1 - \Gamma_{ok}^{2})^{-1} (L+1) [P_{L}(\Gamma_{ok})$$

$$- \Gamma_{ok} P_{L+1}(\Gamma_{ok})]. \quad (13)$$

Substituting Equation (13) in Equation (12) gives

$$\sum_{\lambda=0}^{L} (2\lambda + 1) P_{\lambda}^{2}(\Gamma_{ok})$$

$$= (1 - \Gamma_{ok}^{2})^{-1} (L+1)^{2} P_{L}^{2}(\Gamma_{ok}) = 4\pi C_{ok}^{-2}. \quad (14)$$

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A Note on the Measurement of the Transport Mean Free Path of Thermal Neutrons in Graphite by a Poison Method*

J. M. Hendrie *et al.*,[1] have reported a measurement of the transport mean free path of thermal neutrons, λ_t, in graphite by a copper-poison technique which is analogous to the heavy-water/boron technique described earlier by S. W. Kash and D. C. Woods.[2] The value of $\lambda_t = 2.77 \pm 0.05$ cm reported by Hendrie is somewhat larger than the value of 2.65 ± 0.03 cm inferred from measurements by E. Starr and G. Price[3] for the same AA Graphite, using pulsed-neutron techniques. In addition, other measurements by the pulsed-neutron method (all referred to a graphite density of 1.60 g/cm^3) are tabulated in Table I and are seen to have an average value of approximately 2.59 cm. Measurements of λ_t by extrapolation distance, complex diffusion length, and averaged cross sections are also indicated in Table I, but are not considered to be as reliable as either the pulsed-neutron method or the poison method.

Recent measurements of the diffusion-cooling constant, c, as shown in Table II, indicate that the value is probably in the neighborhood of 38×10^5 cm^4/sec, rather than that of 12 or 16×10^5 cm^4/sec, which were reported earlier.[4,5] The transport mean free path reported by Hendrie *et al.*,[1] was not corrected for the effect of diffusion cooling because that effect was considered to be negligible. However, in view of the large values of c recently reported, the value of λ_t as measured by the copper-poison method has been recomputed and is reported herein.

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[1] J. M. HENDRIE, J. P. PHELPS, G. A. PRICE, E. V. WEINSTOCK, "Slowing Down and Diffusion Lengths of Neutrons in Graphite-Bismuth Systems", *Proc. of 2nd United Nations Int. Conf. on the Peaceful Uses of Atomic Energy, Geneva*, P/601 (1958).

[2] S. W. KASH and D. C. WOODS, "Measurements of the Transport Mean Free Path of Thermal Neutrons in D$_2$O by a Boron Poisoning Method", *Phys. Rev.* 90, 564 (1953).

[3] E. STARR and G. A. PRICE, "Measurement of the Diffusion Parameters of Graphite and Graphite-Bismuth by Pulsed Neutron Methods", *Proc. of the Brookhaven Conf. on Neutron Thermalization*, Vol. 3, 1034, BNL-719 (C-32) (1962).

[4] A. V. ANTONOV *et al.*, "A Study of Neutron Diffusion in Beryllium, Graphite, and Water by Impulse Method", *Proc. of the Int. Conf. on the Peaceful Uses of Atomic Energy, Geneva*, P/661 (1956).

[5] K. H. BECKURTS, "Measurements with a Pulsed Neutron Source", *Nuc. Sci. & Eng.*, 2, 516 (1957).