case the activity of foil 2 (induced by epicadmium neutrons passing through the detector in both directions) is subtracted from the activities of both foil 1 and foil 3, the resultant activity differences being caused entirely by capture of thermal neutrons. That is $\beta(\text{thermal}) = (a_1-a_2)/(a_3-a_2)$. This detector has the advantage of great compactness compared with the indium detector.

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An Improved Free-Surface Boundary Condition for the P-3 Approximation

One of the most common methods of solution of the monoenergetic neutron transport equation is to expand the angular dependence of the directional flux in a truncated spherical-harmonics series[1]. The resulting finite number of differential equations for the spatially dependent expansion coefficients are generally referred to as the P_N equations. N here refers to the upper limit on the summation sign of the flux expansion and thus designates the order of the expansion. Setting N equal to one yields the widely used diffusion approximation. If greater accuracy is required, the P_3 equations are often used. Only in very special instances are higher order expansions ($N > 3$) used in reactor design.

As is well known[1], in a finite-order expansion (N finite) one cannot satisfy exactly the boundary condition that, at a free surface, no neutrons return to the system, i.e., the directional flux vanishes over a hemisphere (2π) of solid angle. Thus an approximate boundary condition is required. In this letter we use the variational calculus to obtain this approximate free-surface boundary condition for the P_3 approximation. The P_1 free-surface boundary condition according to the variational method has been treated elsewhere[2] and leads to a linear extrapolation distance of 0.7071 mean free paths.

For definiteness, we consider the left-hand boundary of a slab system to be a free surface at $z = a$. The right-hand boundary is treated analogously. Slab geometry is considered for simplicity, but presumably the variational analysis could be carried through in other geometries. The P_3 directional flux expansion is

$$\phi(z,\mu) = \sum_{n=0}^{3} \left(\frac{2n+1}{2}\right) \phi_n(z) P_n(\mu), \qquad (1)$$

where $\phi(z,\mu)$ is the directional flux, z is the slab coordinate, μ is the cosine of the angle between the z axis and the velocity vector of the neutron, $P_n(\mu)$ is the n^{th} Legendre polynomial, and $\phi_n(z)$ is the n^{th} expansion coefficient, given by

$$\phi_n(z) = \int_{-1}^{1} d\mu P_n(\mu)\phi(z,\mu). \qquad (2)$$

Now, at the free surface, $z = a$, the rigourous transport-theory boundary condition is

$$\phi(a,\mu) = 0, \ (0 < \mu \leq 1). \qquad (3)$$

It is evident from the flux expansion, Eq. (1), that Eq. (3) cannot be satisfied exactly except by the trivial solution $\phi_i(a) = 0$, $0 \leq i \leq 3$. In particular, the structure of the P_3 equations demands, for a non-trivial solution, that there exist two linear relationships between the moments at the free surface. These two relationships can be written quite generally as

$$\phi_3(a) + A\phi_0(a) + B\phi_1(a) = 0, \qquad (4)$$

$$\phi_2(a) + C\phi_0(a) + D\phi_1(a) = 0. \qquad (5)$$

Mark and Marshak have each suggested methods for calculating A through D so as to approximate Eq. (3). (See reference 1 for a general discussion). Mark sets

$$\phi(a,\mu_i) = 0, \ (i = 1, 2), \qquad (6)$$

where μ_i are the two positive roots of $P_4(\mu_i) = 0$, and Marshak uses

$$\int_0^1 d\mu P_i(\mu)\phi(a,\mu) = 0, \ (i = 1, 3) \qquad (7)$$

as the boundary conditions. As argued by Davison[1] and shown by experience, Marshak's boundary conditions for the P_3 approximation, Eq. (7), are generally superior to those of Mark. (For high-order expansions, Mark's conditions may be better.) Thus, the Marshak P_3 free-surface boundary conditions are currently in general use. Using Eq. (1) in Eq. (7) and carrying out the angular integrations yields the Marshak conditions explicitly, which are, in the format of Eqs. (4) and (5),

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[1] B. DAVISON and J. B. SYKES, *Neutron Transport Theory*. Clarendon Press, Oxford, (1957).

[2] G. C. POMRANING and M. CLARK, JR., "The Variational Method Applied to the Monoenergetic Boltzmann Equation, Part I." *Nucl. Sci. Eng.*, 16, 147-154 (1963).

$$\begin{aligned} A &= -5/8, \\ B &= -1, \\ C &= +4/5, \\ D &= +8/5. \end{aligned} \tag{8}$$

We now determine A through D using the variational calculus and show that the resulting values lead to a significantly more accurate result for the linear extrapolation distance (Milne problem) than do the Marshak values given by Eq. (8). Including the boundary terms in the Lagrangian of the transport equation, as in reference 2, and restricting the adjoint trial function to be related to the trial function for the directional flux according to

$$\phi^*(z,\mu) = \phi(z,-\mu), \tag{9}$$

yields the following relationship at the free surface:

$$\int_0^1 d\mu\mu\,\phi(a,\mu)\,\delta\phi(a,-\mu) = 0, \tag{10}$$

where $\delta\phi(a,-\mu)$ is the first variation of the trial function evaluated at $z = a$ and $-\mu$. Equation (9) is a natural restriction to place on the adjoint trial function since the exact solutions for $\phi(z,\mu)$ and $\phi^*(z,\mu)$ obey this relationship. Using Eq. (1) as the trial function in Eq. (10) and carrying out all angular integration yields

$$\begin{aligned} &\left(\frac{1}{8}\phi_0 + \frac{1}{4}\phi_1 + \frac{5}{32}\phi_2\right)\delta\phi_0 \\ &-\left(\frac{1}{4}\phi_0 + \frac{9}{16}\phi_1 + \frac{1}{2}\phi_2 + \frac{7}{32}\phi_3\right)\delta\phi_1 \\ &+\left(\frac{5}{32}\phi_0 + \frac{1}{2}\phi_1 + \frac{25}{32}\phi_2 + \frac{3}{4}\phi_3\right)\delta\phi_2 \\ &-\left(\frac{7}{32}\phi_1 + \frac{3}{4}\phi_2 + \frac{147}{128}\phi_3\right)\delta\phi_3 = 0, \end{aligned} \tag{11}$$

where the argument of the moments, $z = a$, has been omitted for simplicity. Using Eqs. (4) and (5) in Eq. (11) to eliminate ϕ_2, ϕ_3, $\delta\phi_2$ and $\delta\phi_3$ and setting to zero the coefficients of $\phi_0\delta\phi_0$, $\phi_0\delta\phi_1$, $\phi_1\delta\phi_0$, and $\phi_1\delta\phi_1$ yields four nonlinear equations for the four unknowns, A through D:

$$\frac{1}{8} - \frac{5}{16}C + \frac{25}{32}C^2 - \frac{147}{128}A^2 = 0, \tag{12}$$

$$\frac{1}{4} - \frac{5}{32}D - \frac{1}{2}C + \frac{25}{32}CD + \frac{3}{4}BC + \frac{7}{32}A - \frac{3}{4}AD - \frac{147}{128}AB = 0, \tag{13}$$

$$-\frac{1}{4} + \frac{1}{2}C + \frac{7}{32}A - \frac{5}{32}D + \frac{25}{32}CD + \frac{3}{4}AD - \frac{3}{4}BC - \frac{147}{128}AB = 0, \tag{14}$$

$$-\frac{9}{16} + \frac{7}{16}B + \frac{25}{32}D^2 - \frac{147}{128}B^2 = 0. \tag{15}$$

Solving Eqs. (12) through (15) numerically, we find

$$\begin{aligned} A &= -0.532591, \\ B &= -0.868925, \\ C &= +0.744949, \\ D &= +1.522003. \end{aligned} \tag{16}$$

We note that these values follow the same trend as do the Marshak values, but are consistently smaller (in absolute value).

To compare the accuracy of the Marshak and variational P_3 free-surface boundary conditions, we consider the Milne problem, i.e., a pure isotropic scatterer occupying the halfspace $z > 0$ with a source at infinity. The P_3 equations for this problem are

$$\frac{d\phi_1(z)}{dz} = 0, \tag{17}$$

$$\frac{1}{3}\frac{d\phi_0(z)}{dz} + \Sigma\phi_1(z) + \frac{2}{3}\frac{d\phi_2(z)}{dz} = 0, \tag{18}$$

$$\frac{2}{5}\frac{d\phi_1(z)}{dz} + \Sigma\phi_2(z) + \frac{3}{5}\frac{d\phi_3(z)}{dz} = 0, \tag{19}$$

$$\frac{3}{7}\frac{d\phi_2(z)}{dz} + \Sigma\phi_3(z) = 0, \tag{20}$$

where Σ is the macroscopic collision cross section. The solution of Eqs. (17) through (20) which has the proper behavior at infinity is

$$\phi_0(z) = \alpha\Sigma z + \beta + \gamma e^{-\nu\Sigma z}, \tag{21}$$

$$\phi_1(z) = -\frac{1}{3}\alpha, \tag{22}$$

$$\phi_2(z) = -\frac{1}{2}\gamma e^{-\nu\Sigma z}, \tag{23}$$

$$\phi_3(z) = -\frac{3\nu}{14}\gamma e^{-\nu\Sigma z}, \tag{24}$$

where $\nu = \sqrt{35}/3$. Using Eqs. (4) and (5) as the boundary conditions at $z = 0$ gives

$$\alpha = \frac{3}{14}\left(\frac{3\nu C - 7A}{AD - BC}\right)\gamma, \tag{25}$$

$$\beta = \left(\frac{3\nu CD - 7BC - 14C(DA - BC)}{14C(AD - BC)}\right)\gamma. \tag{26}$$

Now, the linear extrapolation distance d, is defined as the reciprocal of the logarithmic derivative of the asymptotic scalar flux, ϕ_0, at the free surface. Thus we have

$$d = \frac{\beta}{\alpha}\lambda_{tr} = \frac{1}{3C}\left[\frac{3\nu CD - 7BC - 14C(DA - BC)}{3\nu C - 7A}\right]\lambda_{tr}, \tag{27}$$

where we have defined the transport mean free path, λ_{tr}, as $1/\Sigma$ for an isotropic scatterer. Using the Marshak values for A through D, Eq. (8), in Eq. (27) yields the familiar result $d = 0.7051\,\lambda_{tr}$. This value, when compared to the exact Wiener-Hopf result of $0.7104\,\lambda_{tr}$, is seen to be 0.75 percent

in error. Using the variational values for A through D, Eq. (16), in Eq. (27) yields $d = 0.7118\ \lambda_{tr}$, which is 0.20 percent in error. Thus the use of the variational boundary conditions has reduced the error by almost a factor of four. For completeness we mention the P_3 Mark value for d which is $0.6940\ \lambda_{tr}$, 2.31 percent in error. Although the Milne problem is primarily of academic interest, it is suggested that the variational boundary conditions derived here will yield better results than the Marshak conditions for practical problems involving free surfaces, such as criticality calculations, thermal-control-worth calculations, blackness-coefficient calculations, etc.

At a right-hand free surface, assumed to be at $z = b$, the variational boundary conditions, in the format of Eqs. (4) and (5) (with the argument $z = a$ replaced by $z = b$) are easily found from symmetry considerations to be

$$\begin{aligned} A &= +\,0.532591\ , \\ B &= -\,0.868925\ , \\ C &= +\,0.744949\ , \\ D &= -\,1.522003\ . \end{aligned} \qquad (28)$$

A more detailed account of this work, including generalization to even-order and higher odd-order expansions, and treatment of entrant boundary conditions, will be published in the future.

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Differences in the Removal of Activity from an Irradiated Thoria/Urania Slurry and from a Slurry Containing Adsorbed Activity

The method suggested for chemical reprocessing and purification of Th/U/O slurry type reactor fuels by Gardner[1] was tested with thoria/urania slurry used in an in-pile loop experiment. Less than 1% of the gross gamma activity due to fission products was removed from the slurry by leaching with 6 M HNO_3 (100 C) for 6 hours. A comparison of the gamma activity in the slurry before and after treatment showed no detectable selective removal of activity. Under his conditions, Gardner reports removal of 60 to 74.3% of the activity from an unirradiated slurry containing Ce^{144} tracer as a stand-in for fission products. Our study demonstrates that simple acid leaching can not be used to decontaminate irradiated thoria slurry for recycle.

The slurry used in our study was from an experiment of the Thorium Utilization Program of the Reactor Chemistry Division, Oak Ridge National Laboratory.[2] The slurry contained 1350g of Th/0.4% U^{235} oxide per kg of D_2O, to which was added a sufficient amount of palladium-treated thoria to bring the palladium concentration to 0.019 M. (The palladium was used as a catalyst for hydrogen/oxygen recombination.) The slurry had been prepumped for 900h prior to irradiation, and then irradiated to a total of 7×10^{18} fissions per gram of ThO_2. The operating temperature of the in-pile loop was 280 C, and pressurized oxygen was kept over the slurry. Initially the slurry particles averaged 2 μm in size and had a surface area of 2.7 m^2/g. Due to the combined effect of pumping and irradiation, the particles averaged about 0.75 μm in size and had a surface area of 32.9 m^2/g when discharged from the irradiation loop. At the time of the decontamination study, the slurry had been out of the reactor about 18 months and typical fission-product analyses were 1.5×10^7 dis min^{-1} g^{-1} of cesium, 1.3×10^9 of cerium, and 4.1×10^9 of zirconium.

The slurry used by Gardner as the basis for the originally suggested procedure was prepared by fixing $Ce(NO_3)_3$ containing Ce^{144} tracer on prepumped thoria/urania by autoclaving at about 600 F. The difference between the results obtained with this material and the irradiated slurry may be attributed to the following: 1) The activity present in our study came from in situ fission of uranium rather than adsorbed tracer 2) Although fission recoil will result in the removal of some of the fission fragments from the particle in which the fission takes place, the stopping point of many of these fragments will be in another thoria particle[3] 3) Fission within the thoria particles results in the ejection of some of the thoria into

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