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CALCULATED ENHANCEMENT OF THERMAL NEUTRON FLUX FROM A ^{252}Cf SOURCE

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ABSTRACT

The thermal neutron flux obtainable from a ^{252}Cf neutron source in a moderated subcritical multiplying system was studied by means of the ANISN code. The dependence of the flux on k_{eff} of the system, the Σ_{aeff} of the system, the ratio of fissionable material to purely absorbing material, and the type of moderating material are discussed. Two-group analytic solutions are presented to show the approximate dependence of the flux on the various parameters. Multiplying media and nonmultiplying media are compared.

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INTRODUCTION

Californium-252 decays by spontaneous fission with an exceptionally high neutron output of 2.311×10^{12} neutrons/sec-g. Because of this high output, ^{252}Cf should be particularly well suited in applications with compact neutron sources that emit $>10^9$ neutrons/sec. One of the many applications suggested for ^{252}Cf is to use its emitted neutrons to provide a sustained neutron flux for neutron activation analysis and short-lived nuclide production.[1] Because the neutrons emitted by ^{252}Cf have an average energy of 2.3 Mev, the thermal neutrons for these applications are obtained by placing the source in a moderating medium. Media that multiply as well as thermalize the neutrons can be used under certain conditions to enhance the neutron flux; the neutron flux level would increase as the assembly approaches criticality. If such systems are constrained to operate in a subcritical mode, they can provide many of the advantages of a small reactor without incurring as many safety and administrative costs.

To aid evaluations of ^{252}Cf in such applications, the thermal neutron flux levels from a ^{252}Cf source in various media were calculated. Results are reported for homogeneous mixtures of ^{235}U-H_2O and ^{235}U-D_2O. For comparison the thermal neutron flux levels of three nonmultiplying moderators - H_2O, graphite, and D_2O - were also calculated.

SUMMARY

Of the nonmultiplying media considered, water, because of its excellent slowing-down properties, yields the highest thermal neutron flux in the vicinity of the source. This peak flux was computed to be 0.012 neutron/cm^2-sec for a source strength of 1 neutron/sec.

The thermal flux level in the moderator can be amplified by admixing fissile material. The addition of a mixture of fissile and absorbing materials to the moderator, however, might actually depress the flux at the source. Water, for example, can support a higher thermal flux at the source than can natural uranium-light water lattices which suffer from a large number of parasitic absorptions by ^{238}U. Amplified flux levels can be attained in systems which minimize the ^{238}U absorption, such as homogeneous mixtures of ^{235}U-H_2O or ^{235}U-D_2O, but here additional effort is required to ensure subcriticality. Fortunately, for the same degree of subcriticality, the thermal flux near the source is highest for the minimum mass of fissile material. Thus, the optimum flux condition is attained in systems which most easily remain subcritical. At the minimum ^{235}U mass, a ^{235}U-H_2O medium with an effective multiplication constant $k = 0.980$ can yield a thermal flux at the source which is approximately fives times that obtained in water. Still higher amplification can be achieved by drawing closer to criticality, because the thermal flux throughout a multiplying medium is approximately proportional to $1/(1-k)$.

DISCUSSION

CALCULATIONS

The neutron flux levels were calculated with the one-dimensional discrete ordinates code ANISN[2] in the S_{16} approximation. The sixteen group cross section sets developed by Hansen and Roach were employed.[3] These cross sections contain P_1 scattering for hydrogen and deuterium, but are limited to P_0 scattering in the transport approximation for all other nuclides. The source was taken to be a dispersion of ^{252}Cf in a 1-cm^3 aluminum sphere. The fission neutron spectrum of ^{252}Cf was assumed to be the same as for ^{235}U.[4] This spectrum and the Hansen and Roach sixteen group structure are shown in Table I.

TABLE I

Energy Spectrum of Neutrons from Source

Group	Energy Range	Relative Number of Neutrons Emitted by Source per Second
1	3.0 - ∞ Mev	0.204
2	1.4 - 3.0 Mev	0.344
3	0.9 - 1.4 Mev	0.168
4	0.4 - 0.9 Mev	0.180
5	0.1 - 0.4 Mev	0.090
6	17.0 -100.0 keV	0.014
7	3.0 - 17.0 keV	—
8	0.55 - 3.0 keV	—
9	100.0 -500.0 eV	—
10	30.0 -100.0 eV	—
11	10.0 - 30.0 eV	—
12	3.0 - 10.0 eV	—
13	1.0 - 3.0 eV	—
14	0.4 - 1.0 eV	—
15	0.1 - 0.4 eV	—
16	thermal	—

NONMULTIPLYING MEDIA

The calculated thermal (group 16) neutron flux distributions in water, graphite, and heavy water are shown in Figure 1. Results from similar calculations for these and other moderators have also been reported by Nichols.[5] In the present calculations, each moderator was assumed to have a radius of 68 cm. Due to the large neutron diffusion lengths for graphite and heavy water, only 11 and 3% of the source neutrons were absorbed by these systems, respectively. Nevertheless, on the basis of simple age-diffusion theory calculations, the thermal flux at the source is expected to be no more than a few percent higher for effectively infinite systems.

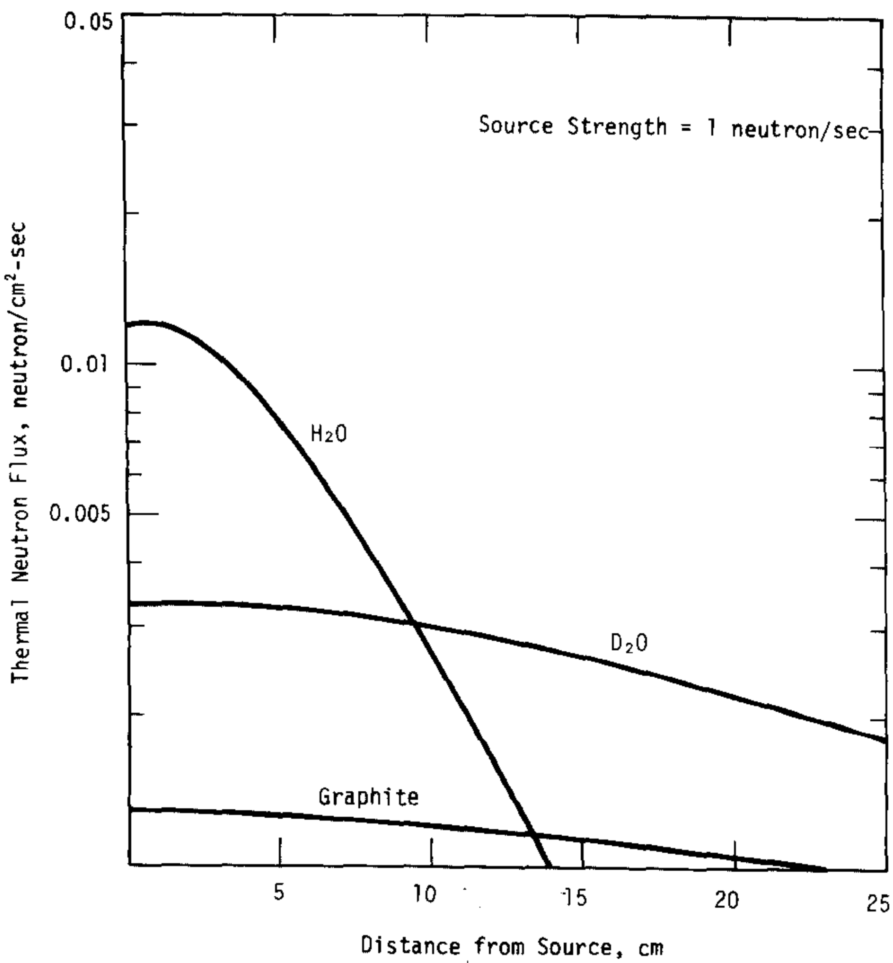

FIG. 1 THERMAL FLUX DISTRIBUTIONS FROM A ^{252}Cf SOURCE IN H_2O, D_2O, AND GRAPHITE

According to two-group diffusion theory, moderators that support higher fluxes at the source have a long thermal neutron diffusion length (small absorption cross section and large thermal diffusion coefficient) and small τ (Fermi age from fission to thermal energy, Figure 2). For H_2O, D_2O, and graphite, the implications of diffusion theory are in qualitative agreement with the results from ANISN, but are qualitative only, since the two-group model generally yields fluxes which are too high in the vicinity of the source. (The derivation of Figure 2 by two-group diffusion theory is given in Appendix A.[6])

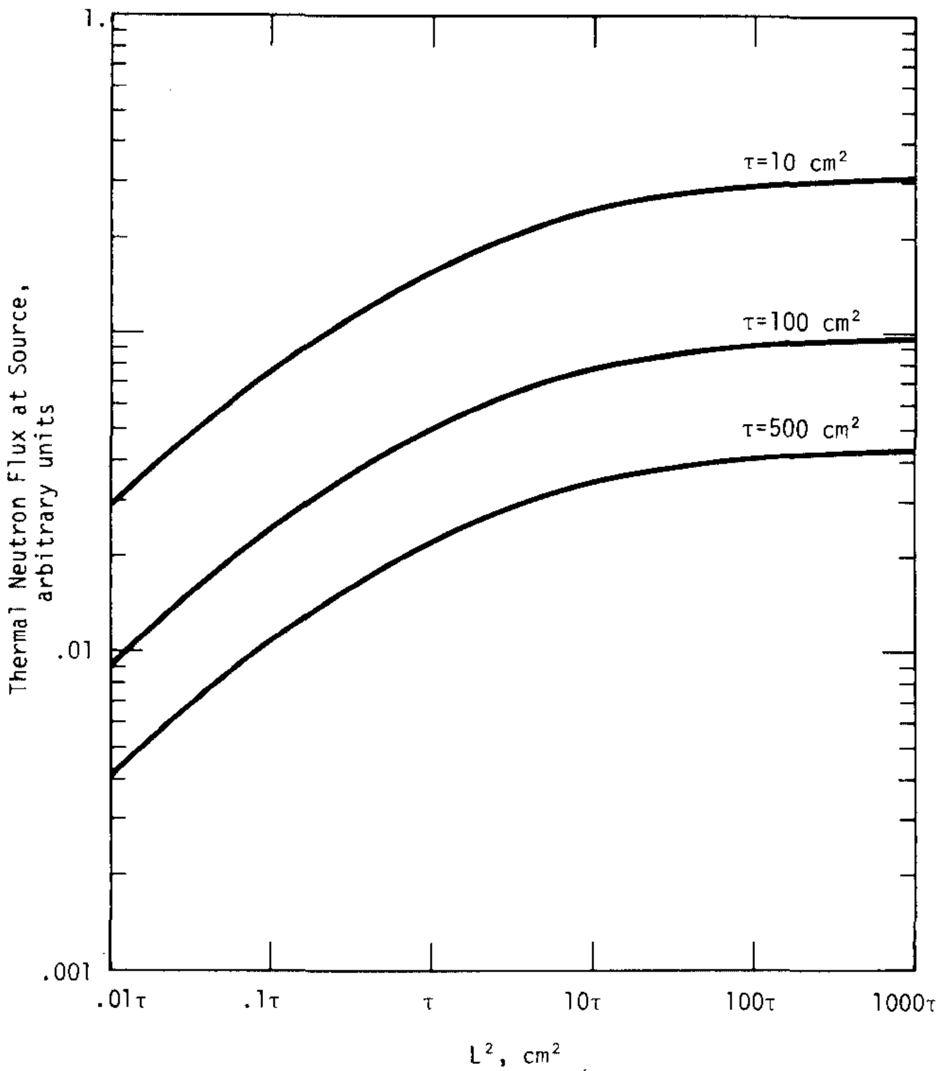

FIG. 2 THERMAL NEUTRON FLUX AT THE SOURCE AS A FUNCTION OF L^2 AND τ, ASSUMING A CONSTANT THERMAL DIFFUSION COEFFICIENT

The ratio of the thermal neutron flux to the total neutron flux above 0.10 eV is given as a function of distance from the source in Figure 3. These curves, obtained with ANISN, illustrate the large variation in the neutron spectrum near the source. This variation of the neutron spectrum could be significant in applications that require standards and unknown samples to be irradiated in identical neutron spectra.

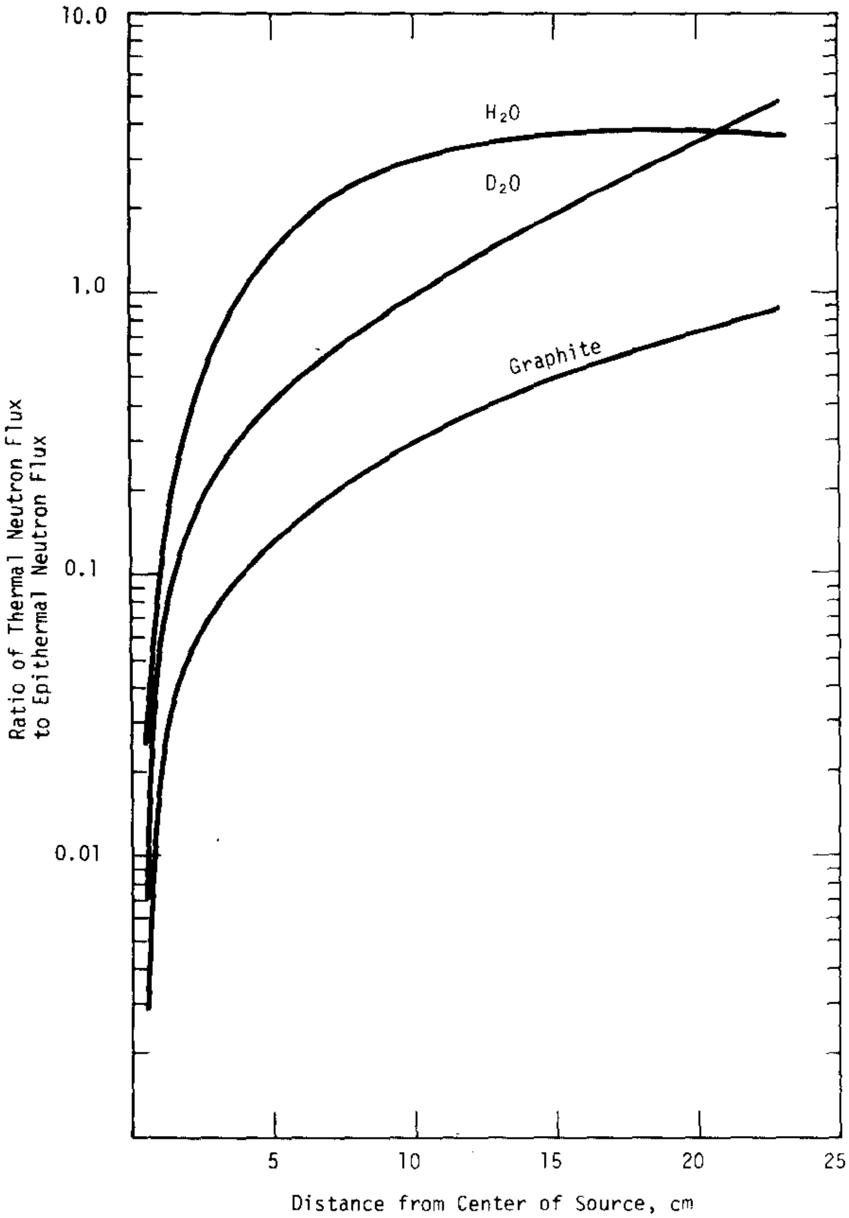

FIG. 3 SPECTRAL VARIATION IN H_2O, D_2O, AND GRAPHITE

To derive a nearly optimum case, ANISN calculations were performed to determine the neutron flux from a ^{252}Cf source in homogeneous mixtures of ^{235}U and light water.

By use of ANISN to compute k, the core volumes and ^{235}U masses for ^{235}U-H_2O spheres with 4-in. H_2O reflectors were calculated for various degrees of criticality. The results are shown in Figures 4 and 5. To provide a measure of the accuracy of the calculations, the figures also display the results, as reported in Reference 7, of criticality studies made at Oak Ridge National Laboratory. The Oak Ridge studies were based on homogeneous, water-moderated, water-reflected spheres of uranium with 93.2% ^{235}U.

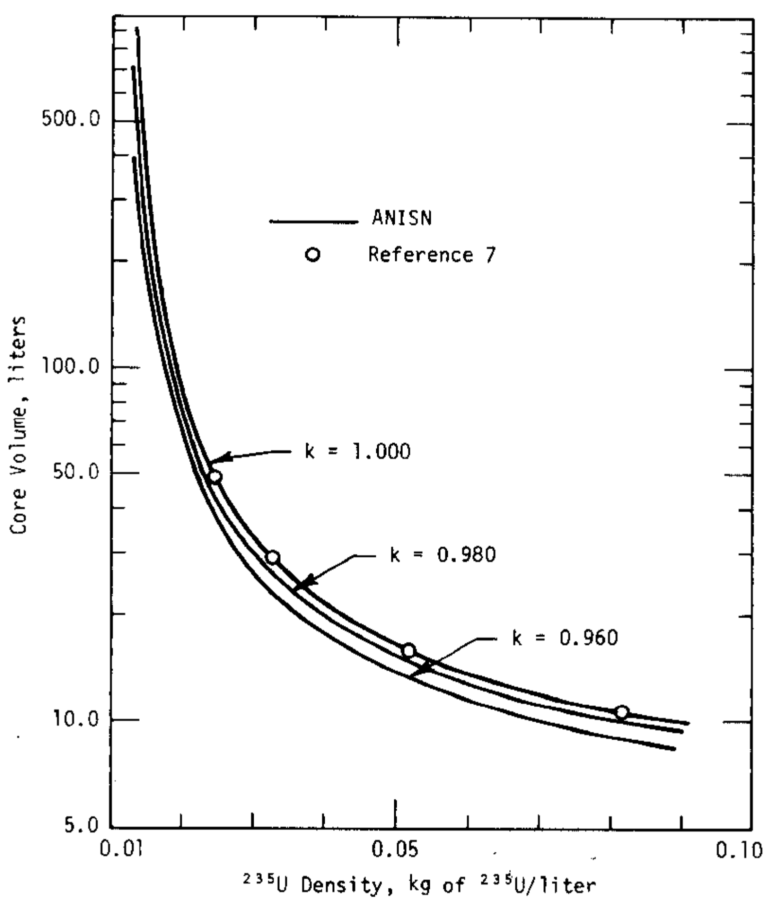

FIG. 4 CORE VOLUME AS A FUNCTION OF k AND ^{235}U DENSITY FOR WATER-REFLECTED ^{235}U-H_2O SPHERES

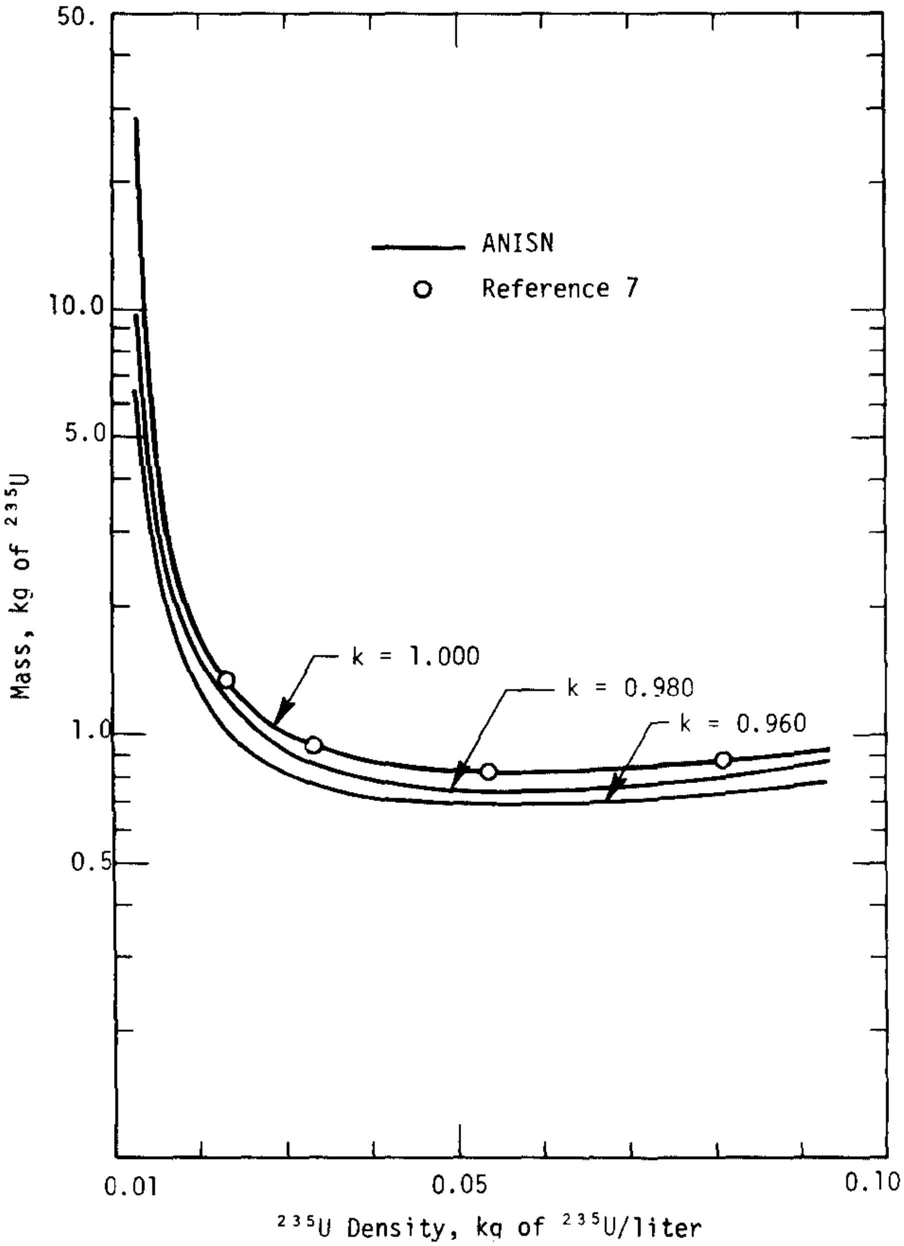

FIG. 5 DEPENDENCE OF ^{235}U MASS ON k AND ^{235}U DENSITY FOR WATER-REFLECTED ^{235}U-H_2O SPHERES

Figures 6 and 7 display the ANISN results for the thermal neutron flux at the source as a function of ^{235}U density and core radius. Comparison of Figures 5 and 6 shows that the thermal flux at the source is nearly maximum at the minimum ^{235}U mass, in accordance with the predictions of diffusion theory (Appendix B).

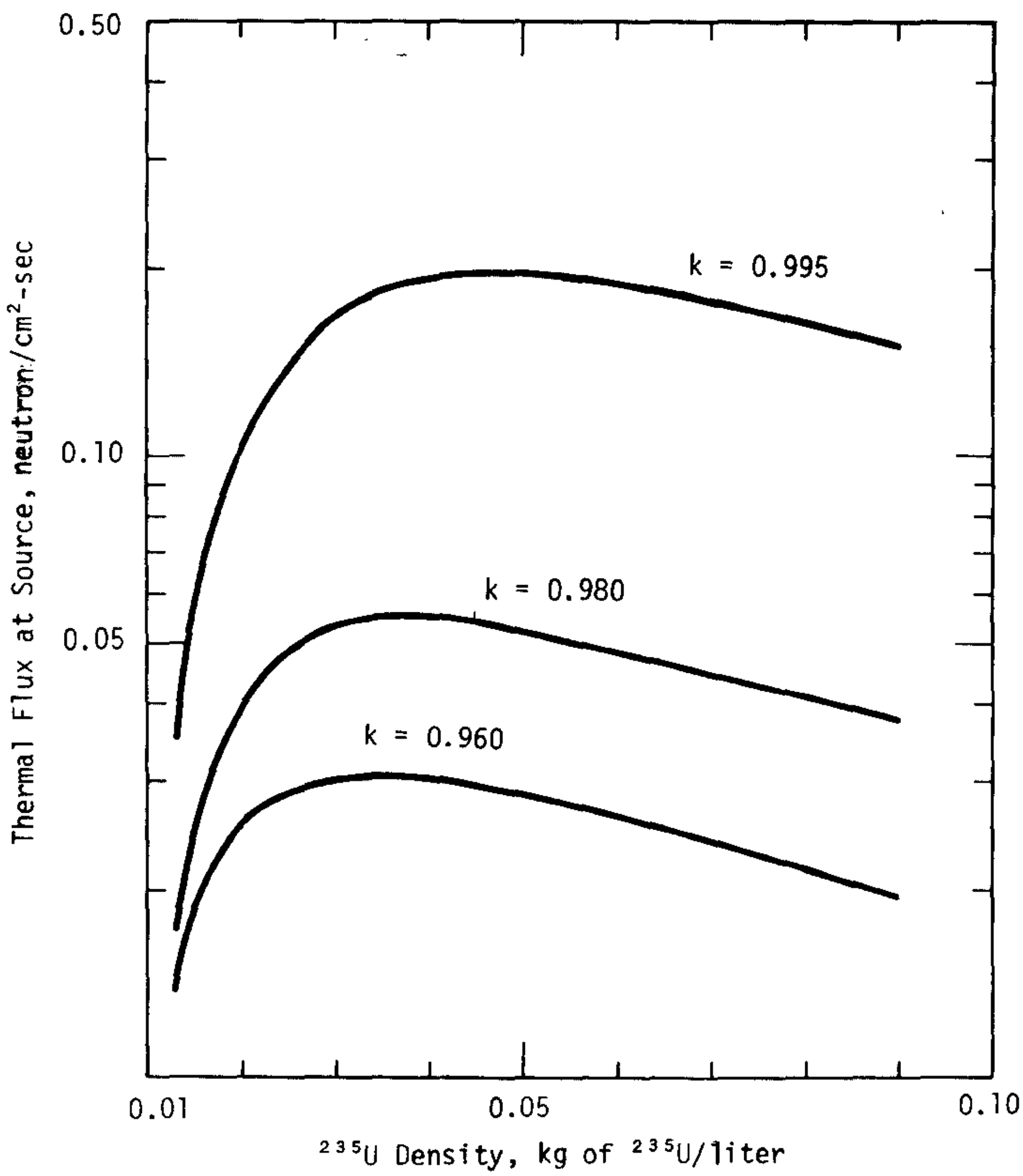

FIG. 6 PEAK THERMAL NEUTRON FLUXES IN WATER-REFLECTED ^{235}U-H_2O SPHERES AS A FUNCTION OF ^{235}U DENSITY AND k

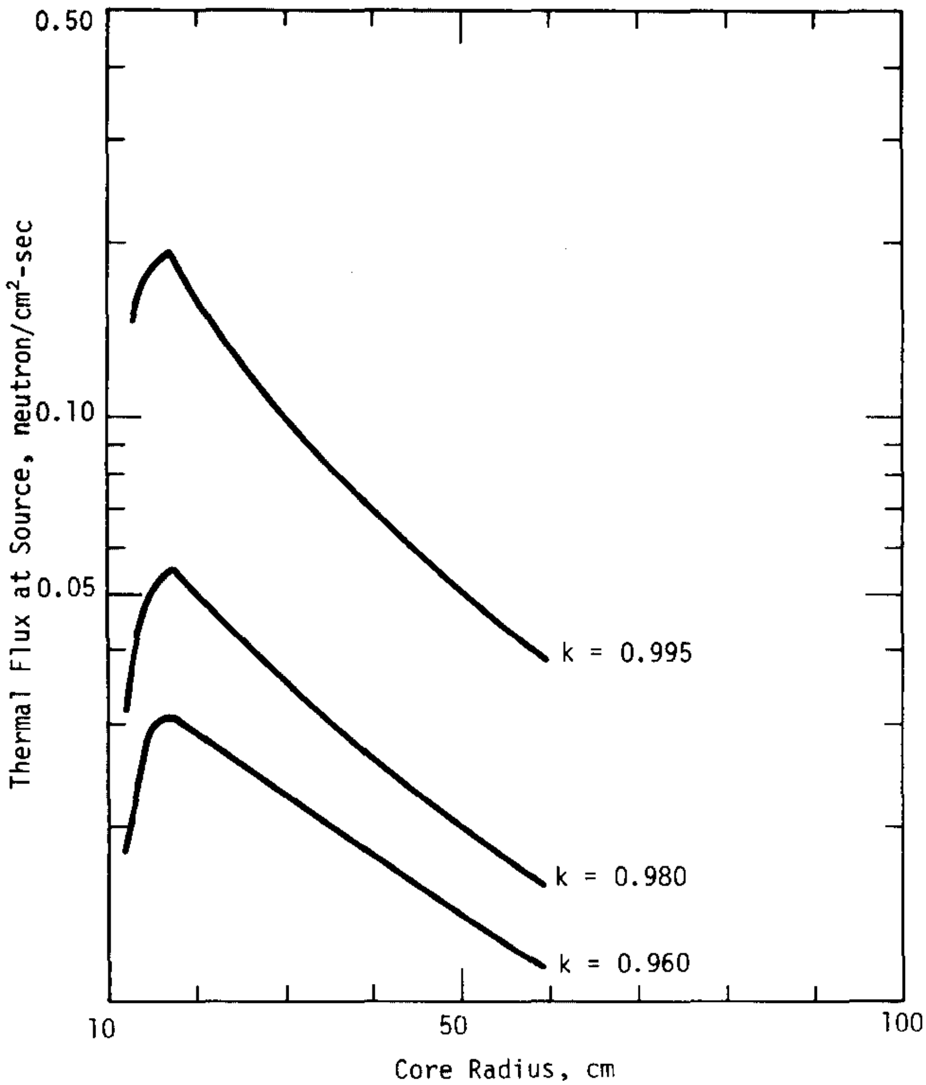

FIG. 7 PEAK THERMAL NEUTRON FLUXES IN WATER-REFLECTED $^{235}U-H_2O$ SPHERES AS A FUNCTION OF CORE RADIUS AND k

For $^{235}U-H_2O$ spheres, core radii of approximately 17 cm appear most advantageous, not only because they provide the highest possible thermal neutron flux near the source for a given value of k, but also because at the minimum critical mass they can not go critical due to accidental changes in geometry or dilution. The effect that the size of the core has on the thermal flux distribution can be seen in Figure 8, where the thermal flux versus distance from the source is plotted for systems with k = 0.980 and core radii of 17.8 and 55.4 cm.

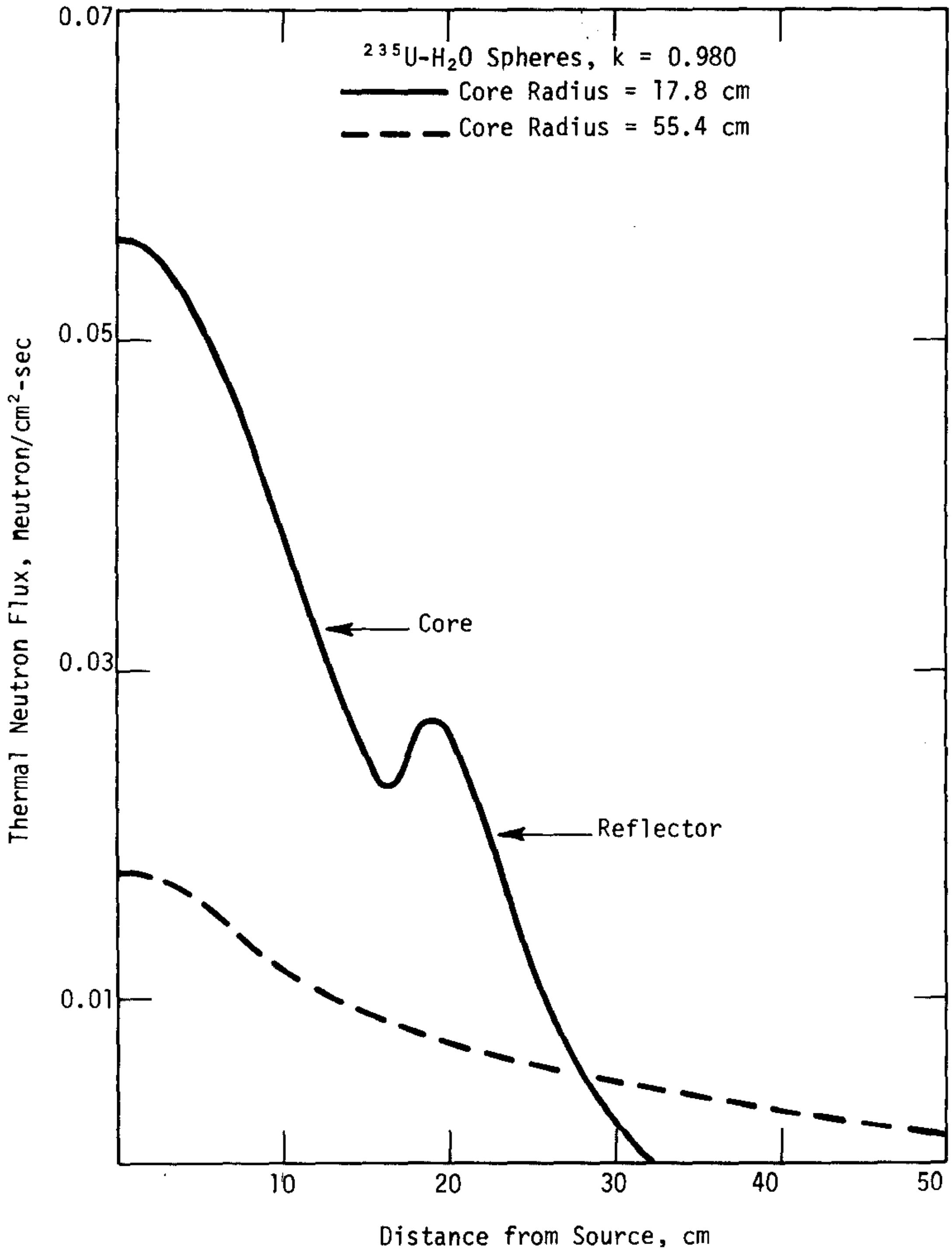

FIG. 8 THERMAL FLUX DISTRIBUTIONS IN $^{235}U-H_2O$ SPHERES WITH CORE RADII OF 17.8 cm AND 55.4 cm

Due to the dependence of the flux on the fuel mass, the desired size of the subcritical multiplying system largely influences the choice of moderator to be used. Figure 9 is a reproduction of the results of critical mass calculations by Safonov[8] for bare ^{235}U systems with various moderators. On the basis of these results and Equation 14 (Appendix B), ^{235}U-H_2O systems with core radii of 17 cm should yield higher thermal fluxes at the source than can be attained in systems with the same k using either D_2O or graphite. On the other hand, if systems with core radii of approximately 60 cm are desired, Figure 9 indicates that D_2O will yield a higher thermal flux near the source than H_2O. Table II compares the peak thermal fluxes in various ^{235}U-H_2O and ^{235}U-D_2O systems when k = 0.980. These results were computed using ANISN and assuming a 4-in. H_2O reflector in each case. The thermal flux at the source in a ^{235}U-D_2O medium with radius equal to 57 cm and k equal to 0.980 is fifteen times greater than that attained in D_2O alone.

TABLE II

COMPARISON OF THERMAL NEUTRON FLUXES NEAR ^{252}Cf SOURCE FOR ^{235}U-H_2O AND ^{235}U-D_2O SPHERES WITH k=0.980

Core Radius, cm	Thermal Flux at Source, neutron/cm^2-sec[a] ^{235}U-H_2O	^{235}U-D_2O
23	.045	.001
39	.027	.0362
57	.017	.0501
83	.009	.0421

a. For source strength of 1 neutron/sec.

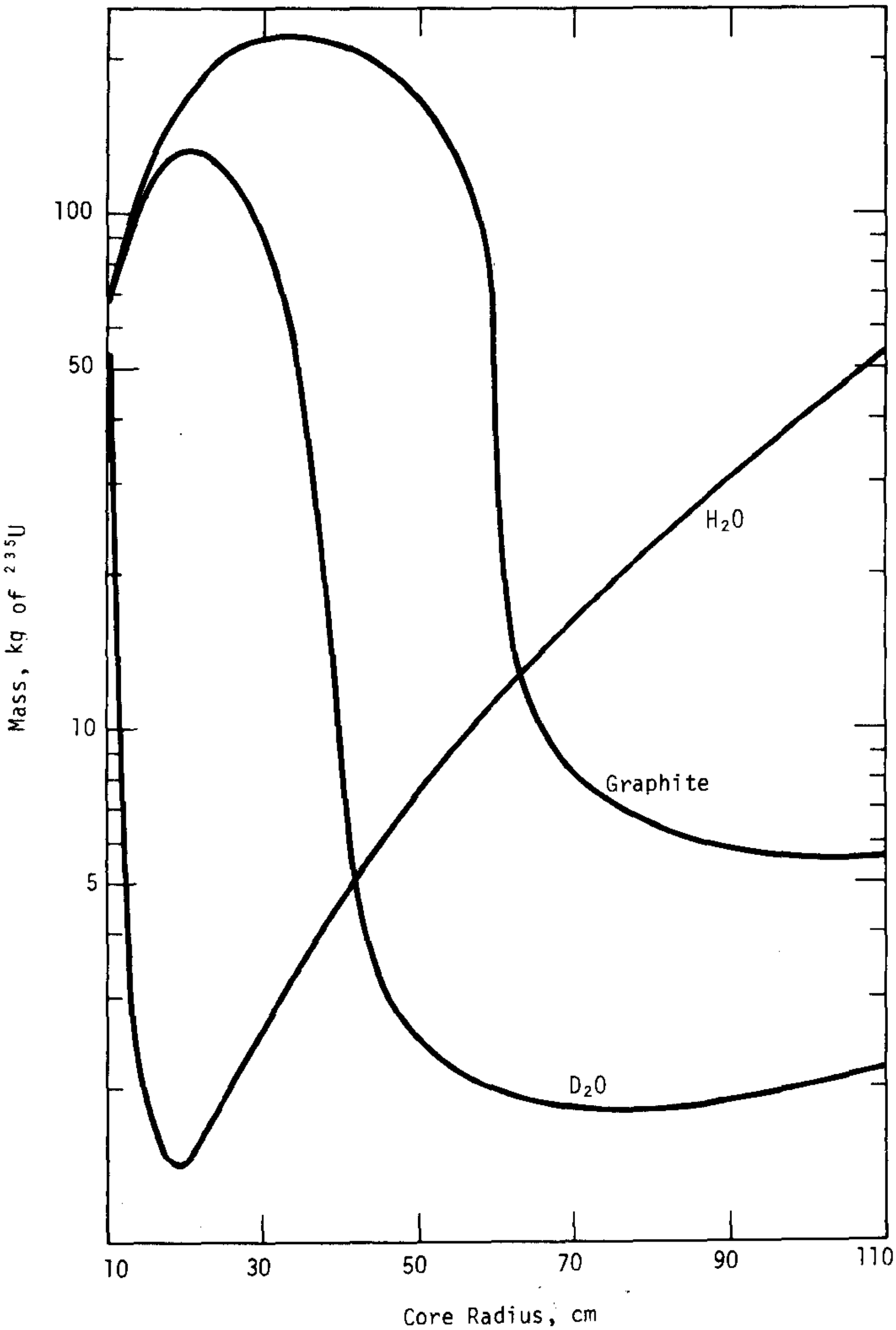

FIG. 9 CRITICAL MASSES OF ^{235}U IN BARE SPHERES AS A FUNCTION OF MODERATOR AND RADIUS (REF. 8)

The many well-documented results from critical mass studies may be applied in other ways to enhance the flux levels in subcritical systems. For example, it has been shown that ^{235}U-H_2O systems with H_2O reflectors require slightly more ^{235}U for criticality than those with thick D_2O or graphite reflectors.[9,10] Consequently, for subcritical systems having the same k, the magnitude of the thermal flux near the source will be slightly lower with H_2O reflectors than with D_2O or graphite. Beryllium reflected systems should yield the highest thermal flux at the source, because systems with thick beryllium reflectors (>30 cm) require the least fuel for criticality; approximately 250 g of ^{235}U for ^{235}U-H_2O cores 9 cm in radius.[9,10] For a beryllium reflected ^{235}U-H_2O system with a core radius of 9.4 cm and k = 0.980, the thermal flux at the source was calculated to be 0.073 n/cm^2-sec. This value is approximately 1.3 times higher than the peak thermal flux for H_2O reflected systems with the same k, and a factor of 6 higher than the thermal flux at the source in an effectively infinite sphere of water.

APPENDIX A

TWO-GROUP DIFFUSION THEORY FOR NONMULTIPLYING MEDIA

The fast (ϕ_1) and thermal (ϕ_2) group fluxes from a point source in an infinite, nonmultiplying medium have the form

$$\phi_1(r) = \frac{q}{4\pi r\tau\Sigma_R} - e^{-r/\sqrt{\tau}} \qquad (1)$$

and

$$\phi_2(r) = \frac{q}{4\pi r\Sigma_a} \; \frac{e^{-r/L}}{L^2 - \tau} - \frac{e^{-r/\sqrt{\tau}}}{L^2 - \tau} \qquad (2)$$

where r is the distance from the source and q denotes the source strength (neutron/sec). The other parameters in these equations are: Σ_a, the macroscopic thermal neutron absorption cross section; L, the thermal diffusion length; τ, the Fermi age from fission to thermal energy; and Σ_R, the cross section for the removal of neutrons from the fast group by down-scatter. The above expressions for the group fluxes ignore fast neutron absorption. The thermal neutron flux at the source is obtained by letting $r \rightarrow 0$ in Equation 2:

$$\phi_2(o) = \frac{q}{4\pi\Sigma_a L^2} \cdot \frac{1}{\tau(1 + \tau/L)} \qquad (3)$$

APPENDIX B

TWO-GROUP DIFFUSION THEORY APPLIED TO A ^{252}Cf SOURCE IN MULTIPLYING MEDIA

In multiplying media, it is again helpful to establish certain qualitative features of the neutron flux levels using two-group diffusion theory. In spherical geometry, the appropriate equations are:

$$-D_1 \nabla^2\phi_1(r) + (\Sigma_R + \Sigma_{a1})\phi_1(r) - \nu\Sigma_f\phi_2(r) = S(r) \qquad (4a)$$

and

$$-D_2 \nabla^2\phi_2(r) + \Sigma_{a2}\phi_2(r) - \Sigma_R\phi_1(r) = 0 \qquad (4b)$$

where D_i and Σ_{ai} denote the diffusion coefficient and macroscopic absorption cross section associated with group i (i = 1,2). Here fissions induced by fast neutrons are neglected. The function S(r) represents the spatial distribution of the ^{252}Cf source (neutrons emitted per cm^3 per second at r), and is assumed to have the shape depicted in Figure B-1. The group fluxes are required to vanish at R, the extrapolated radius of the sphere:

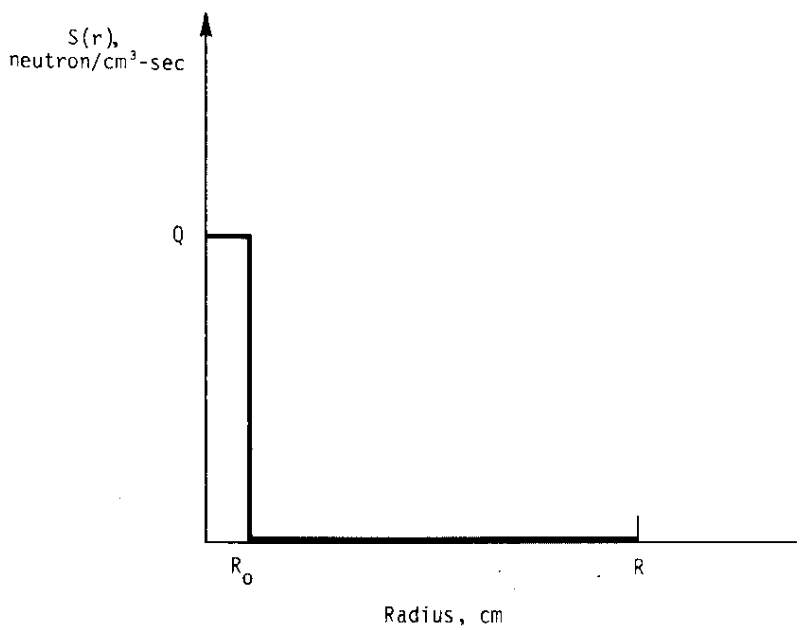

FIG. B-1 DISTRIBUTION OF NEUTRON SOURCE

$$\phi_i \; (R) = 0 \tag{5}$$

where

$$i = 1,2$$

The above problem may be conveniently solved in terms of the eigenfunctions of the corresponding critical problem. In this approach, the group fluxes and source distribution are represented by the following expansions:

$$\phi_1 \; (r) = \frac{1}{r} \sum_{n=1}^{\infty} a_n \sin B_n r \tag{6a}$$

$$\phi_2 \; (r) = \frac{1}{r} \sum_{n=1}^{\infty} b_n \sin B_n r \tag{6b}$$

and

$$S(r) = \frac{1}{r} \sum_{n=1}^{\infty} q_n \sin B_n r \tag{6c}$$

where

$$B_n = \frac{n\pi}{R} \tag{7}$$

The orthogonality of the sine terms can be utilized to show that

$$q_n = \frac{2}{R} \int_0^R rS(r) \sin B_n r dr$$

$$= \frac{2Q}{RB_n^2} (\sin B_n R_o - B_n R_o \cos B_n R_o) \tag{8}$$

The coefficients a_n and b_n in Equations 6a and 6b can be related to q_n by substituting the expansion for the group fluxes into Equations 4a and 4b. In this manner it can be shown that

$$\phi_1 (r) = \frac{1}{r} \sum_{n=1}^{\infty} q_n \frac{\sin B_n r}{(D_1 B_n^2 + \Sigma_{a_1} + \Sigma_R)(1-k_n)} \tag{9a}$$

and

$$\phi_2 (r) = \frac{1}{r} \sum_{n=1}^{\infty} q_n \frac{\Sigma_R \sin B_n r}{(D_2 B_n^2 + \Sigma_{a_2})(D_1 B_n^2 + \Sigma_{a_1} + \Sigma_R)(1-k_n)} \tag{9b}$$

where

$$k_n = \frac{\Sigma_R}{D_1 B_n^2 + \Sigma_{a1} + \Sigma_R} \cdot \frac{\nu\Sigma_f}{D_2 B_n^2 + \Sigma_{a1}} \qquad (10)$$

It will be noted that k_1 is the usual two-group multiplication constant if fast fission is neglected.

Of special interest is the behavior of the thermal flux in systems which are nearly critical, i.e., cases in which the fundamental mode in Equation 9b dominates. Substituting the explicit expression for q_1 into Equation 9b and dropping the subscript 1 from k_1 and B_1, we have for $k \approx 1$

$$\phi_2(r) = \frac{2QRk}{\pi^2(1-k)} \cdot \frac{\sin BR_o - BR_o \cos BR_o}{\nu\Sigma_f} \cdot \frac{\sin Br}{r} \qquad (11)$$

The above equation implies that near criticality the change in the neutron flux due to a change in k is characterized primarily by the variation in the factor 1/(1-k). This property is sometimes used for determining the multiplication constants for subcritical assemblies.

In the region of maximum flux near the source, Equation 11 has the form

$$\phi_2(o) = \frac{2Qk}{\pi(1-k)} \cdot \frac{(\sin BR_o - BR_o \cos BR_o)}{\nu\Sigma_f} \qquad (12)$$

This expression can be simplified by noting that for most systems under consideration, the product BR_o is sufficiently small to make the identification

$$\sin BR_o - BR_o \cos BR_o = \tfrac{1}{3}(BR_o)^3 = \frac{\pi^3 V_o}{3V} \qquad (13)$$

Here V and V_o are the volumes of the system and source, respectively. Equation 12 can then be rewritten to obtain

$$\phi_2(o) = \frac{2\pi^2 QV_o}{3\nu\sigma_f NV} \; \frac{k}{1-k} \qquad (14)$$

where σ_f and N denote the macroscopic fission cross section and atom density for the fissile material in the moderating medium. It follows that the relationship between the thermal fluxes in systems having multiplication constants k and k' is

$$\phi_2'(o) = \frac{k'(1-k)NV}{k\,(1-k')N'V'} \cdot \phi_2(o) \qquad (15)$$

One important conclusion that can be drawn from the above equations is that, for a given value of k, the thermal neutron flux at the source varies inversely with the mass of fissile material in the system. An analogous dependence of the flux on the fuel mass is experienced in thermal reactors held at constant power. To attain high thermal fluxes, highly multiplying systems with the minimum mass of fissile material are best.

Although the above treatment ignores the fast fission effect, Equation 14 can be used to estimate the thermal flux at the source when only a small percentage of the fissions are induced by fast neutrons. If Equation 14 is applied to natural uranium-light water systems, such systems would not be expected to yield high thermal fluxes near the source because they must contain large amounts of uranium (must have large volumes) for k to be near unity. Calculations using the ANISN code demonstrated that the addition of natural uranium to water in fact depresses the thermal flux at the source. The flux levels can be increased, however, by using fuels enriched in ^{235}U.

REFERENCES

1. J. L. Crandall. "Survey of Applications for ^{252}Cf." *Isotop. Radiat. Tech.* *7*(3), 306 (1970).

2. W. W. Engle, Jr. *A Users Manual for ANISN: A One Dimensional Discrete Ordinates Transport Code With Anisotropic Scattering.* USAEC Report K-1693, Oak Ridge Gaseous Diffusion Plant, Oak Ridge, Tenn. (1967).

3. G. E. Hansen and W. H. Roach. *Six and Sixteen Group Cross Sections for Fast and Intermediate Critical Assemblies.* USAEC Report LAMS-2543, Los Alamos Scientific Laboratory, Los Alamos, New Mexico (1961).

4. J. W. Meadows. "^{252}Cf Fission Neutron Spectrum from 0.003 to 15.0 Mev." *Phys. Rev. 157*, 1076 (1967).

5. J. P. Nichols. "Design Data for ^{252}Cf Neutron Source Experiments." *Nucl. Appl. 4*, 382 (1968).

6. K. H. Beckurts and K. Wirtz. *Neutron Physics.* Springer-Verlag, New York (1964).

7. H. C. Paxton, J. T. Thomas, D. Callihan and E. B. Johnson. *Critical Dimensions of Systems Containing* ^{235}U, ^{239}Pu, *and* ^{233}U. USAEC Report TID-7028 (1964).

8. G. Safonov. *A Brief Description of Some RAND Reactor Studies.* RM-847, Rand Corp., Santa Monica, Calif. (1952).

9. G. A. Jarvis and C. B. Mills. *Critical Mass Reduction.* USAEC Report LA-3651, Los Alamos Scientific Laboratory, Los Alamos, New Mexico (1966).

10. R. I. Brasier, L. D. P. King, and C. B. Mills. "Application of Low Critical Mass Studies to Reactor Design." *Nucl. Appl. 6*, 266 (1969).