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AN ESTIMATION OF PHOTONEUTRONS FROM CARBON-13 IN AN OIL SHIELD

D. K. Trubey

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NEUTRON PHYSICS DIVISION

AN ESTIMATION OF PHOTONEUTRONS FROM CARBON-13 IN AN OIL SHIELD*

D. K. Trubey

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OAK RIDGE NATIONAL LABORATORY
Oak Ridge, Tennessee
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ABSTRACT

Measurements of thermal-neutron flux in the presence of high-energy gamma rays in oil indicate that photoneutrons comprise a large fraction of the total thermal-neutron flux at large distances from the neutron source. This is attributed chiefly to the (γ,n) reaction in C^{13}. Measurements at the Lid Tank Shielding Facility in oil with and without a bismuth slab to reduce the gamma rays, and consequently the photoneutrons which they produced, agreed with calculated fluxes.

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INTRODUCTION*

Measurements of thermal-neutron fluxes in the presence of high-energy gamma rays in liquid media containing hydrogen or carbon and hydrogen indicate that photoneutrons comprise a large fraction of the total thermal-neutron flux at large distances from the neutron source. The evidence for this is a change in slope of the flux plotted as a function of distance from the neutron source. At large distances, the slope is characteristic of gamma-ray attenuation in the medium rather than of neutron attenuation. For example, the estimated photoneutron flux produced by the $D(\gamma,n)H$ reaction has been shown to be nearly equal to the total flux measured in the water around the Bulk Shielding Reactor at distances of about 2 m or more.[1] More recently at the Lid Tank Shielding Facility (LTSF) the photoneutron process has been observed to be even more important at distances closer to a fission source in media containing carbon (GE 10-C insulating oil).[2] It has been assumed that this could be attributed chiefly to the (γ,n) reaction in C^{13} induced by the high-energy capture gamma rays from an Inconel plate adjacent to the source plate.[3] In order to verify this assumption a calculation has been performed using recently determined values[4] of the cross section for this reaction and the results have been compared with the results of the LTSF experiment.

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* An abbreviated version of this report was previously published in ORNL-2081, "Appl. Nuclear Phys. Div. Ann. Prog. Rep. for Period Ending Sept. 10, 1956," ORNL-2081, p. 168.

1. J. M. LaRue, G. P. Letz, T. J. Morley, and J. N. Renaker, "An Estimate of the Photoneutron Flux in the Water Surrounding the Bulk Shielding Facility Reactor," ORNL-CF-51-8-290 (Aug. 24, 1951) (Confidential, Undocumented); see also G. deSaussure, "Calculation of the Photoneutron Flux in Water Near the Bulk Shielding Reactor," ORNL-2545 (May, 1958); also published in ORNL-2389, "Appl. Nuclear Phys. Div. Ann. Prog. Rep. for Period Ending Sept. 1, 1957," p. 22.

2. G. T. Chapman, J. B. Dee, J. M. Miller, and W. J. McCool, "Measurements of an Effective Neutron Cross Section of Lithium at the Lid Tank Shielding Facility," ORNL-CF-54-11-3 (Nov. 2, 1954) (Declassified).

3. That this reaction is responsible was first suggested by H. Goldstein of NDA.

4. B. C. Cook, and V. L. Telegdi, private communication to E. Guth, Jan. 1956; the $C^{13}(\gamma,n)C^{12}$ cross section has been calculated by G. deSaussure and E. Guth of ORNL, see ORNL-2081, "Appl. Nuclear Phys. Div. Ann. Prog. Rep. for Period Ending Sept. 10, 1956," p. 167.

I. ESTIMATION OF THE PHOTONEUTRON FLUX IN OIL

Most of the gamma rays which produce neutrons in C^{13} (the dominant process in the experiment) at large distances are very energetic (over 5 Mev); therefore, in the LTSF experiment[2] the gamma rays from the fission process in the source plate did not contribute to the thermal-neutron flux in the oil and were neglected in the calculation. However, the oil was contained in a steel tank which had an Inconel "window" (1/8 in. thick, 28 in. in diameter) in the section adjacent to the source plate. This Inconel produced capture gamma rays that were considered as the source gamma rays for the calculation of the photoneutron production. A simple spectrum of source energies (five groups) was computed from the known thermal-neutron flux at the location of the Inconel plate, from the absorption cross section of the elements in the Inconel, and from the estimations[5] of the capture gamma-ray distribution (see Table 1).

Table 1. Capture Gamma-Ray Spectrum Resulting from a Thermal-Neutron Flux of 6.8×10^7 Neutrons/cm^2·sec Incident on the Inconel Window

Composition of Inconel				Source Strength at Center of Source* (photons/cm^3·sec)				
Material	Percent	σ_a (barns)	Captures cm^3-sec	6 Mev	7.6 Mev	8.5 Mev	9.0 Mev	9.72 Mev
Cr	14	2.9	2.38×10^6	4.28×10^5				1.64×10^6
Fe	6	2.43	7.96×10^5	1.75×10^5	3.98×10^5			
Ni	72	4.5	1.69×10^7	5.07×10^6		2.28×10^6	7.12×10^6	
		Total γ's/cm^3·sec		5.67×10^6	3.98×10^5	2.28×10^6	7.12×10^6	1.64×10^6
		Total γ's/cm^2·sec		1.80×10^6	1.20×10^5	7.24×10^5	3.14×10^6	5.21×10^5

*In the calculations the average source strength, which was taken as 66% of these values, was used.

For energies above 7 Mev, the binding energy of the nucleus was the energy assumed. It was also assumed that the attenuation of both the neutron and gamma-ray fluxes in the Inconel was negligible. The gamma-ray flux for each group was then computed at various distances by multiplying the source strength by the attenuation factor for an isotropic plane circular source in an infinite medium and adding a built-up flux obtained from higher groups. This built-up flux was computed from published differential energy spectra in water[6] by the method presented in Appendix A.

5. P. Mittleman, "Gamma Rays Resulting from Thermal-Neutron Capture," NDA-10-99 (Oct. 6, 1953).

6. H. Goldstein and J. E. Wilkins, "Calculations of the Penetration of Gamma Rays," NYO-3075 (June 30, 1954).

The photoneutron thermal flux resulting from the gamma-ray flux was computed from the following relation:

$$\phi_{th}(z)\Sigma_a = \sum_{E=0}^{\infty} \Gamma(z,E)\Sigma_p(E)$$

where

ϕ_{th} = photoneutron thermal flux,

Σ_a = macroscopic thermal-neutron absorption cross section in the medium,

$\Gamma(z,E)$ = photon flux of energy E at z distance from the source

$$= \Gamma(0,E)\left\{\frac{E_1(\mu z) - E_1\left[\mu z\sqrt{1 + (a^2/z^2)}\right]}{2}\right\} + \text{built-up group},$$

$$E_1(\mu z) = \int_{\mu z}^{\infty} \frac{e^{-y}}{y}\, dy,$$

μ = linear total absorption coefficient for gamma rays of energy E,

a = radius of the source (both LTSF source plate and Inconel plate),

z = distance from the source,

$\Sigma_p(E)$ = photoneutron production macroscopic cross section for gamma rays of energy E.

The justification for the equation used is presented in Appendix B. It is apparent that it is correct if the migration of neutrons can be neglected. This is permissible in this case because neutrons are attenuated very rapidly compared to gamma rays.

The cross sections used are given in Table 2 and the results are shown in Table 3.

Table 2. Cross Sections Used for Oil
[Density = 0.876 g/cm^3; Composition = $(CH_2)_n$]

	Cross Sections (cm^{-1})				
	6 Mev	7.6 Mev	8.5 Mev	9 Mev	9.72 Mev
μ	2.58×10^{-2}	2.28×10^{-2}	2.16×10^{-2}	2.08×10^{-2}	2.00×10^{-2}
$\Sigma_p^{C^{13}}$	2.09×10^{-8}	1.88×10^{-7}	2.93×10^{-7}	3.55×10^{-7}	4.81×10^{-7}
Σ_p^{D}	2.56×10^{-8}	2.14×10^{-8}	2.02×10^{-8}	1.90×10^{-8}	1.85×10^{-8}

Table 3. Computed Photoneutron Flux Values

E(Mev)	Thermal-Neutron Flux (nv_{th}) z = 130 cm	z = 140 cm	z = 150 cm	z = 160 cm
	From Uncollided Photons			
6	1.18×10^{-3}	8.1×10^{-4}	5.4×10^{-4}	3.8×10^{-4}
7.6	5.2×10^{-4}	3.8×10^{-4}	2.6×10^{-4}	1.9×10^{-4}
8.5	5.18×10^{-3}	3.71×10^{-3}	2.60×10^{-3}	1.90×10^{-3}
9	2.962×10^{-2}	2.126×10^{-2}	1.49×10^{-2}	1.098×10^{-2}
9.7	7.18×10^{-3}	5.19×10^{-3}	3.66×10^{-3}	2.70×10^{-3}
Total	4.368×10^{-2}	3.135×10^{-2}	2.197×10^{-2}	1.615×10^{-2}
	From Built-up Photons			
6	1.39×10^{-3}	1.07×10^{-3}	8.1×10^{-4}	6.3×10^{-4}
7.6	3.52×10^{-3}	2.70×10^{-3}	2.03×10^{-3}	1.60×10^{-3}
8.5	1.98×10^{-3}	1.54×10^{-3}	1.16×10^{-3}	8.5×10^{-4}
9	1.48×10^{-3}	1.15×10^{-3}	8.8×10^{-4}	6.8×10^{-4}
9.7	2.4×10^{-4}	1.8×10^{-4}	1.4×10^{-4}	1.1×10^{-4}
Total	8.61×10^{-3}	6.64×10^{-3}	5.02×10^{-3}	3.87×10^{-3}
Grand Total	5.23×10^{-2}	3.80×10^{-2}	2.70×10^{-2}	2.00×10^{-2}

II. COMPARISON OF EXPERIMENTAL AND ESTIMATED NEUTRON FLUXES

The thermal-neutron fluxes measured in oil at various distances from the LTSF source plate are shown in Fig. 1 and Table 4. The insertion of a bismuth slab in the oil reduced the gamma rays sufficiently so as to effectively eliminate the photoneutrons without any other observable effect. In order to verify that the (γ,n) reaction in C^{13} was indeed responsible for the increased thermal-neutron flux, the estimations of the expected flux due to photoneutrons were added to the measured flux values obtained while the gamma rays were suppressed. The result was equal to the measured total flux within the experimental uncertainty. Plots of the estimated and measured total fluxes are compared in Fig. 2. The estimated photoneutron flux at various distances from the source plate is also presented in Fig. 2.

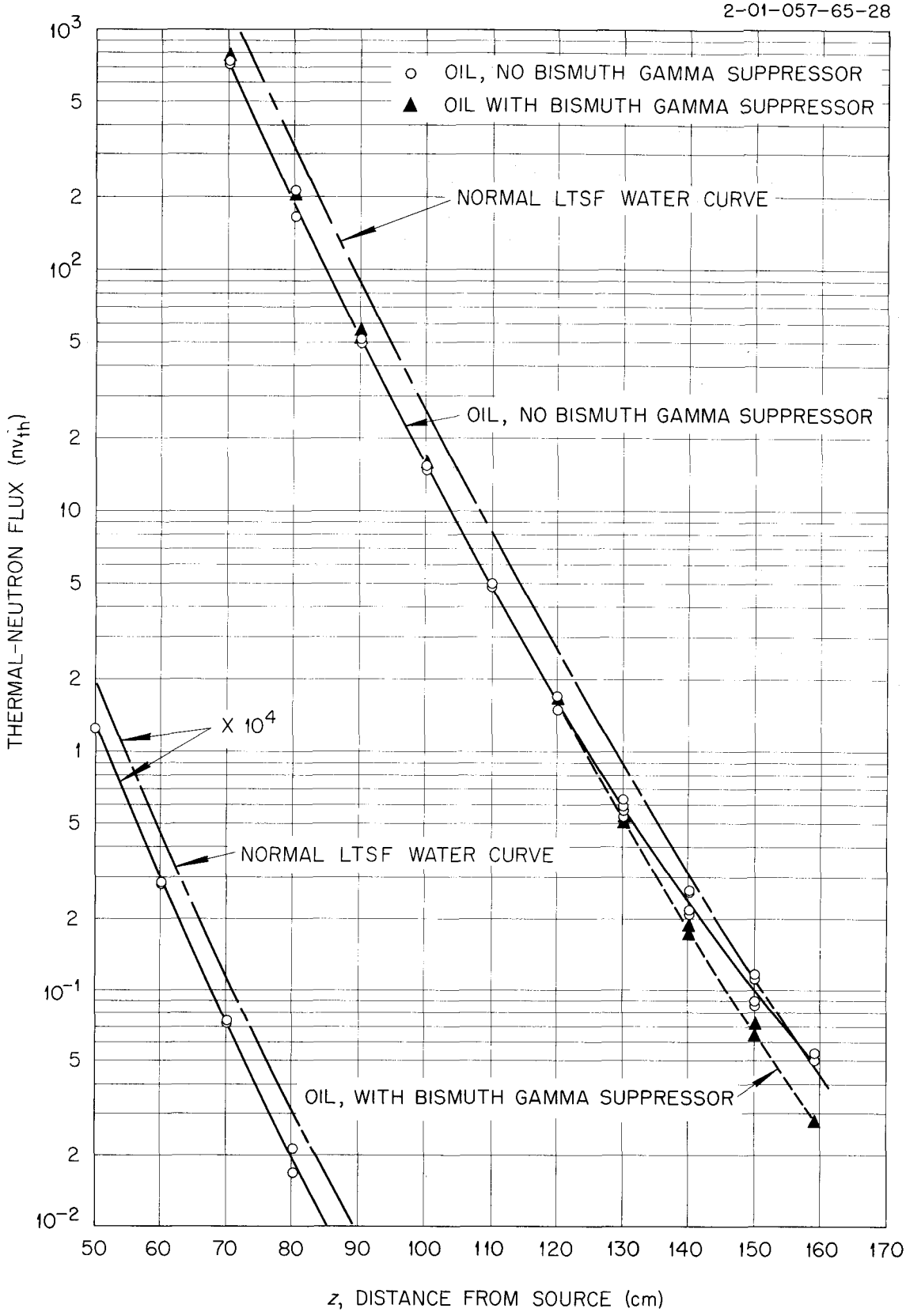


Fig. 1. Thermal-Neutron Flux in Oil With and Without Bismuth Slab to Suppress Photo-Neutron Production.

Table 4. Measured Thermal-Neutron Fluxes in Oil[a]

Detector[b]	Thermal-Neutron Flux (neutrons/cm^2-sec)			
	z = 130 cm	z = 140 cm	z = 150 cm	z = 159 cm
	With Bismuth Slab			
SB	4.99×10^{-1}	1.69×10^{-1}	6.39×10^{-2}	2.73×10^{-2}
DB	5.19×10^{-1}	1.85×10^{-1}	7.08×10^{-2}	
	Without Bismuth Slab			
SB	5.60×10^{-1}	2.15×10^{-1}	8.54×10^{-2}	5.35×10^{-2}
SB	5.35×10^{-1}	2.11×10^{-1}	8.93×10^{-2}	4.97×10^{-2}
DB	5.93×10^{-1}	2.57×10^{-1}	1.15×10^{-1}	
DB	6.27×10^{-1}	2.62×10^{-1}	1.11×10^{-1}	

a. This data was taken from Ref. 2.

b. SB indicates single-barrel 12-1/2-in. BF_3 Counter; DB indicates double-barrel 12-1/2-in. BF_3 counter.

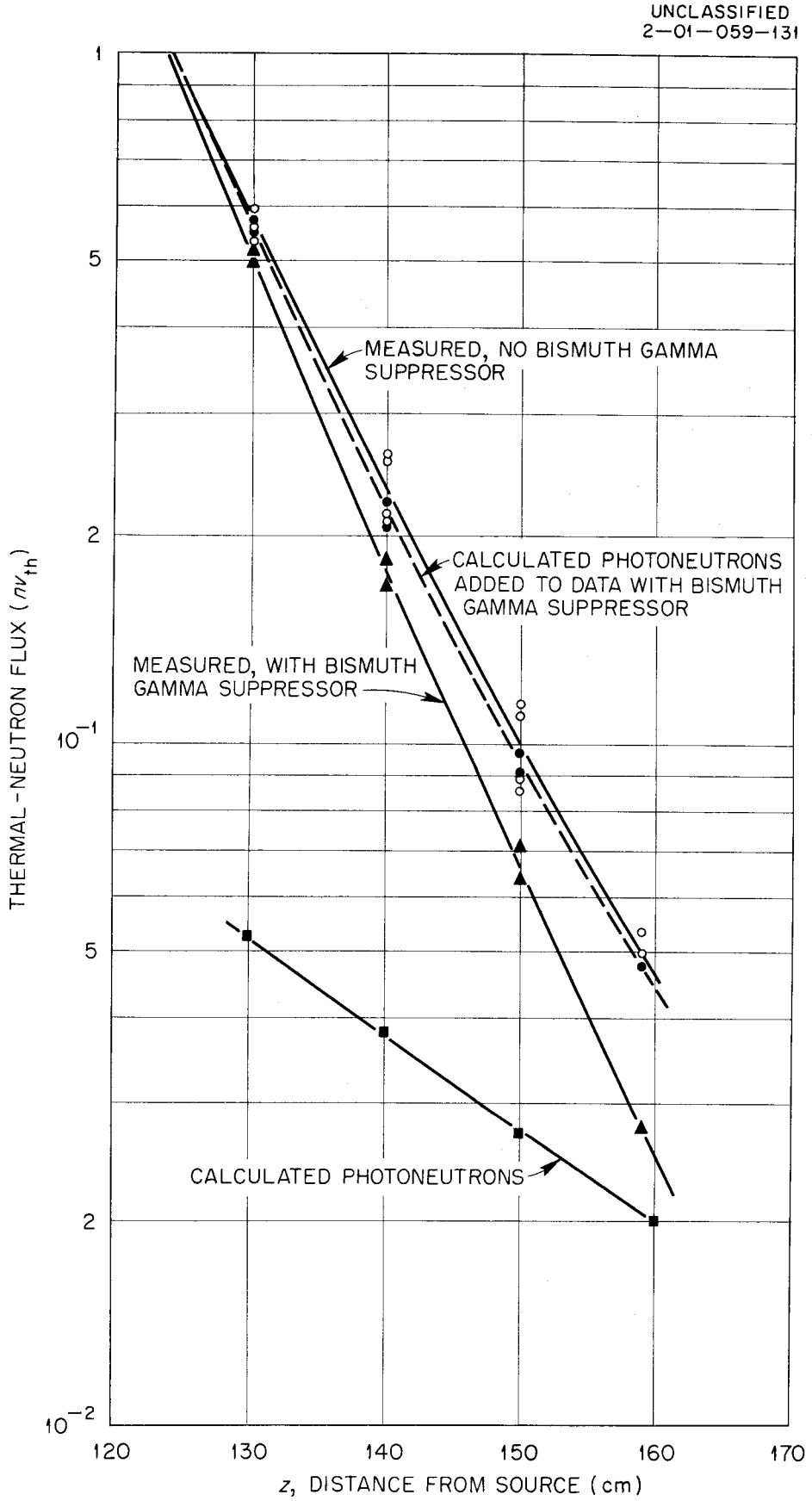


Fig. 2. Measured and Calculated Thermal-Neutron Flux in Oil, Showing Measured and Calculated Components.

Appendix A

METHOD FOR COMPUTING BUILT-UP GAMMA-RAY FLUX IN OIL

The built-up gamma-ray flux at various points in the oil medium at the LTSF was calculated by use of a quantity I_o which has been tabulated for several source energies by Goldstein and Wilkins.[6] This quantity is called the differential energy spectrum and it represents the energy flux of photons of energy E per unit energy range which a nondirectional detector would measure. It is tabulated as $4\pi r^2 e^{\mu_o r} I_o$, where r is the distance between a point source and the detector. The source gives 1 photon/sec of energy E_o which corresponds to a total gamma-ray absorption coefficient μ_o.

In order to remove the point source geometry effect, the attenuation function for the circular plane source of radius a may be used as follows:

$$\text{(A.1)} \qquad I(E)\Delta E = \int_E^{E+\Delta E} \Gamma_o(E_o)(I_o 4\pi r^2 e^{\mu_o r})\left\{\frac{E_1(\mu_o r) - E_1\left[\mu_o r\sqrt{1 + (a^2/r^2)}\right]}{2}\right\} dE$$

where

$$\Gamma_o = \text{source strength,}$$

$$= \Gamma_o(E_o)\left\{\frac{E_1(\mu_o r) - E_1\left[\mu_o r\sqrt{1 + (a^2/r^2)}\right]}{2}\right\} \overline{(I_o 4\pi r^2 e^{\mu_o r})}\Delta E,$$

$\overline{(I_o 4\pi r^2 e^{\mu_o r})}$ = average value over the range E to E $+\Delta E$.

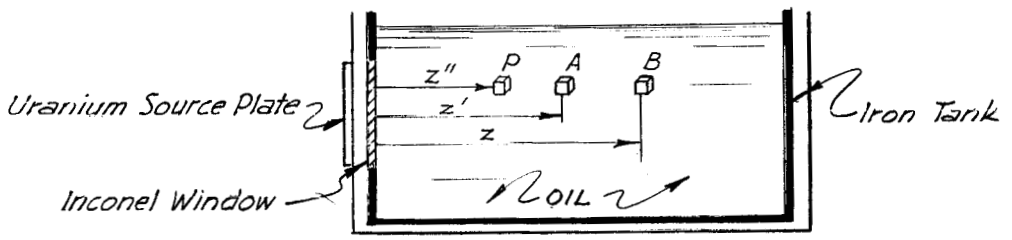
A number flux is obtained by dividing the above equation by E. Thus the built-up flux in a group is:

$$\text{(A.2)} \qquad \phi(E) = \frac{\Gamma_o(E_o)}{E}\left\{\frac{E_1(\mu r) - E_1\left[\mu r\sqrt{1 + (a^2/r^2)}\right]}{2}\right\} \overline{(I_o 4\pi r^2 e^{\mu_o r})}\Delta E$$

Appendix B

METHOD FOR COMPUTING PHOTONEUTRON FLUX IN OIL

As was mentioned previously, the source gamma rays were considered to be only those gamma rays which resulted from thermal-neutron capture in the Inconel window. Capture gamma rays which are born in the window make a collision with a C^{13} nucleus at any point P (see sketch), and a fast-neutron



Lid Tank Geometry

born at that point P arrives at thermal energy at point A and diffuses to point B where it is observed as a thermal flux. The number of neutrons arriving at thermal energy at point A per cubic centimeter per second is given by the following relation (only one gamma-ray energy is considered):

$$N_A = \int_{z''=0}^{\infty} \Gamma(z'')\Sigma_p q(z' - z'')dz'' \tag{B.1}$$

where

$\Gamma(z'')$ = gamma-ray flux at z",

Σ_p = (γ,n) cross section for C^{13},

$q(z' - z'')$ = probability of the neutron from a plane source slowing down to thermal energy at distance $(z' - z'')$.

For an infinite plane in an infinite medium, Fermi age theory gives:

$$q(z' - z'') = \frac{e^{-(z' - z'')^2/4\tau}}{\sqrt{4\pi\tau}} \tag{B.2}$$

(B.3) $$\Gamma(z'') = \Gamma_o e^{-\mu z''} \quad \text{(see footnote *)}$$

where $\Gamma_o = \Gamma(o)$. μ is defined by this equation and does not vary at large z". Substituting Eqs. B.2 and B.3 in B.1 gives

(B.4) $$N_A = \int_{z''=0}^{\infty} \frac{\Gamma_o e^{-\mu z''} \sum_p e^{-(z' - z'')^2/4\tau} \, dz''}{\sqrt{4\pi\tau}}$$

$$= \frac{\Gamma_o \sum_p}{\sqrt{4\pi\tau}} \int_{z''=0}^{\infty} e^{-\mu z''} e^{-(z' - z'')^2/4\tau} \, dz''$$

$$= \frac{\Gamma_o \sum_p}{\sqrt{4\pi\tau}} \int_{z''=0}^{\infty} e^{-\frac{1}{4\tau}\left[z''^2 + 2(2\tau\mu - z')z'' + z'^2\right]} \, dz''$$

Letting $2\tau\mu - z' = c$ gives

(B.5) $$N_A \doteq \frac{\Gamma_o \sum_p}{\sqrt{4\pi\tau}} e^{-\frac{1}{4\tau}(z'^2 - c^2)} \int_{z''=0}^{\infty} e^{-\frac{1}{4\tau}(z'' + c)^2} \, dz''$$

Now letting

(B.6) $$x = \frac{z'' + c}{\sqrt{4\tau}}$$

and

(B.7) $$dx = \frac{dz''}{\sqrt{4\tau}}$$

---

*The more accurate form, $\dfrac{E_1(\mu z'') - E_1\left[\mu z'' \sqrt{1 + (a^2/z''^2)}\right]}{2}$, will be substituted later.

and substituting these terms in Eq. B.5 gives

(B.8) $$N_A = \Gamma_o \Sigma_p e^{-\frac{1}{4\tau}(z^2 - c^2)} \frac{1}{\sqrt{\pi}} \int_{c/\sqrt{4\tau}}^{\infty} e^{-x^2}\, dx$$

$$= \Gamma_o \Sigma_p e^{-\frac{1}{4\tau}(z'^2 - c^2)} \left[\frac{\mathrm{erf}(\infty) - \mathrm{erf}\, c/\sqrt{4\tau}}{2} \right]$$

where

(B.9) $$\mathrm{erf}(x) = \frac{2}{\sqrt{\pi}} \int_o^x e^{-y^2}\, dy$$

but

(B.10) $$\mathrm{erf}(-x) = -\mathrm{erf}(x)$$

For $\mu \sim 0.03\ \mathrm{cm}^{-1}$, $\tau \sim 30\ \mathrm{cm}^2$, and $z' \sim 160$ cm, $c \sim -158$, and

(B.11) $$\mathrm{erf}(\infty) = 1$$

and

(B.12) $$\mathrm{erf}\, \frac{c}{\sqrt{4\tau}} \sim -\mathrm{erf}(14) = -1$$

and

(B.13) $$e^{-\frac{1}{4\tau}(z'^2 - c^2)} = e^{-\frac{1}{4\tau}(z'^2 - 4\tau^2\mu^2 + 4\tau\mu z' - z'^2)}$$

$$= e^{-(\mu z' - \tau\mu^2)}$$

$$\cong e^{-\mu z'} \qquad \text{if } z' \gg \mu\tau$$

Then

(B.14) $$N_A = \Gamma_o e^{-\mu z'} \Sigma_p$$

According to diffusion theory, the thermal-neutron flux at point B due to neutrons arriving at thermal at all points A is given by:

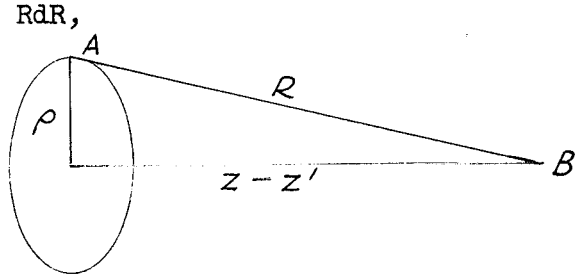
(B.15) $$\phi_B(z) = \Sigma_p \Gamma_o \int_{z'=0}^{\infty} \int_{\rho=0}^{\infty} e^{-\mu z'} \, 2\pi\rho d\rho dz' \left(\frac{e^{-KR}}{4\pi DR} \right)$$

$$= \frac{\Sigma_p \Gamma_o}{2D} \int_{z'=0}^{\infty} \int_{(z - z')}^{\infty} e^{-\mu z'} \, e^{-KR} \, dR$$

where

ρ = distance away from centerline (see sketch),

$\rho d\rho$ = RdR,



$\frac{e^{-KR}}{4\pi DR}$ = the diffusion kernel for a point source at distance R,

$K^2 = \Sigma_a / D$,

$$\phi_B(z) = \frac{\Sigma_p \Gamma_o}{2DK} \int_{z'=0}^{\infty} e^{-\mu z'} \, e^{-K(z - z')} dz'$$

$$= \frac{\Sigma_p \Gamma_o}{2DK} \left[e^{-Kz} \int_0^z e^{z'(K - \mu)} dz' + e^{Kz} \int_z^{\infty} e^{-z'(\mu + K)} dz' \right]$$

$$= \frac{\Sigma_p \Gamma_o}{2DK} \left[e^{-Kz} \frac{e^{z(K - \mu)} - 1}{K - \mu} + e^{Kz} \frac{e^{-z(\mu + K)}}{K + \mu} \right]$$

$$= \frac{\Sigma_p \Gamma_o}{2DK} \left[\frac{e^{-\mu z} - e^{-Kz}}{K - \mu} + \frac{e^{-\mu z}}{\mu + K} \right]$$

$$= \frac{\Sigma_p \Gamma_o}{2DK} e^{-\mu z} \left[\frac{1 - e^{-z(K - \mu)}}{K - \mu} + \frac{1}{K + \mu} \right]$$

but for $K \gg \mu$,

$$\phi_B(z) \cong \frac{\Sigma_p \Gamma_o e^{-\mu z}}{2DK} \left(\frac{2}{K}\right) = \frac{\Sigma_p \Gamma_o e^{-\mu z}}{\Sigma_a}$$

But the gamma-ray attenuation function from a finite plane is a difference of exponential integrals:

$$\phi_B(z) = \frac{\Sigma_p \Gamma_o}{\Sigma_a} \left\{ \frac{E_1(\mu z) - E_1\left[\mu z \sqrt{1 + (a^2/z^2)}\right]}{2} \right\}$$

where

$$E_1(x) \equiv \int_x^\infty \frac{e^{-y}\, dy}{y} ,$$

a = radius of source plate.

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