

$$\epsilon_2 = E + \frac{1}{15} (7 d_{02} + 3 d_{03} + 2 d_{04} + 3 d_{01} - 4 d_{23} - 1 d_{34} \\ + 1 d_{41} + 4 d_{12})$$

$$\epsilon_3 = E + \frac{1}{15} (7 d_{03} + 3 d_{04} + 2 d_{01} + 3 d_{02} - 4 d_{34} - 1 d_{41} \\ + 1 d_{12} + 4 d_{23})$$

$$\epsilon_4 = E + \frac{1}{15} (7 d_{04} + 3 d_{01} + 2 d_{03} + 3 d_{02} - 4 d_{41} - 1 d_{12} \\ + 1 d_{23} + 4 d_{34}) \text{ etc.}$$

There are many other problems connected with an optimal procedure for determining the values of the derived etalons but their solutions are not within the framework of this paper. The aim of this paper was

only to draw the attention to problems mentioned in the title.

*Acknowledgement.* Finally I should like to express my thanks to Dr. J. Hajda, CSc. researcher of the Institute of Theory of Measurement who suggested the above problem.

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## Calculation of Thermal Neutron Absorption in Cylindrical and Spherical Neutron Sources

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#### Abstract

A calculation of the thermal neutron self-absorption for cylindrical or spherical neutron sources has been made. The calculations are confirmed by the experimentally-measured difference in manganous sulfate bath activity for bare and cadmium-covered Pu-Be and Am-Be neutron sources. The calculation is done in single interaction approximation and assumes that the incident thermal neutron flux is isotropic. The source material may be fissionable and be covered by up to three cladding materials. A computer program has been written for the numerical calculations.

#### 1. Introduction

This calculation has been carried out in connection with a program to reduce the uncertainties in the corrections applied to the manganous sulfate bath calibration of neutron sources [1, 2]. The correction considered here is to account for the reduction of the manganese activity due to the loss of thermalized neutrons absorbed in the neutron source itself.

Ryves [3] has calculated the thermal-neutron flux density within a spherical cavity surrounding a neutron source in a weak boron solution, for a wide range of cavity radii, boron concentrations and neutron source spectra. The results were applied to the manganous sulfate bath technique for several sources by multiplying the calculated flux density by the total neutron capture cross section of cavity and source. The results of his report are suitable for sources which can be regarded as "thin" for thermal neutron capture.

Cap *et al.* [4] have calculated the effect of the moderator on the source strength of a fissionable source. The source encapsulations were neglected and

absorption in the source itself was handled by a six group-diffusion equation.

The calculation presented here is applicable to sources of all ranges of absorption or fission cross section and encapsulations of any material. The calculation does require a measurement or knowledge of the thermal-neutron flux at the source location.

This calculation will take into account four different materials in the source. The materials are three cladding and one, possibly fissionable, core material. The calculation is carried out in a single interaction approximation, i.e., the effects of elastic and inelastic scattering of thermal neutrons are neglected. This approximation is adequate because the neutrons are in thermal equilibrium and because the correction which is applied to the calibration is small (typically 1% or less). Two cases are available. The first is for a spherically symmetric source and the second is for a cylindrically symmetric source. Each consists of a core and up to three cladding layers. The thicknesses of the ends and side of a cladding cylinder may all be different.

The thermal-neutron flux is assumed to be isotropic. This enables one to carry out the computation as a sum of monodirectional fluxes from different directions. All integrations are performed with Weddle's formula (5).

#### 2. Description of the Calculation

Part I. The probability of neutron loss for a given neutron direction and position.

If the source materials are labeled A, B, C, and D from the inside out, then the probability of a thermal

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neutron interacting in passing through the source is

$$P(t) = 1 - e^{-\Sigma_a a(t) - \Sigma_b b(t) - \Sigma_c c(t) - \Sigma_d d(t)} \quad (1)$$

where  $a(t)$  is the thickness of "A" material for this particular direction and location of passing through the source,  $b(t)$  is the total thickness of "B" material, etc., and " $t$ " denotes any particular path through the source. The  $\Sigma$ 's are the appropriate macroscopic cross sections,  $\Sigma_a$  being the sum of the absorption and fission cross sections for the "A" material,  $\Sigma_b$  being the absorption cross section for the "B" material, etc.

The probability of fission in the fissionable material is given by

$$P_f(t) = \frac{\Sigma_{af}}{\Sigma_a} [1 - e^{-\Sigma_a a(t)}] [e^{-(\Sigma_a d'(t) + \Sigma_{cf}(t) + \Sigma_b b'(t))}] \quad (2)$$

where  $d'(t)$  is the thickness of the "D" layer passed in this direction and location going from the outside into the "A" material,  $c'(t)$  the thickness of the "C" layer passed in going into the source, etc., and  $\Sigma_{af}$  is the macroscopic fission cross section of material "A".

Part II. Case 1. A spherically symmetric source.

In this case the probability of neutron disappearance for a neutron striking the source is given by

$$P = \frac{\int_0^R 2\pi y P(y) dy}{\pi R^2} \quad (3)$$

where  $R$  is the outer radius of the source and  $y$  is the perpendicular distance from the center of the source to the path through it. The probability of fission is similarly

$$P_f = \frac{\int_0^R 2\pi y P_f(y) dy}{\pi R^2} \quad (4)$$

The probability of loss of neutrons from the bath per neutron striking the source is therefore

$$P_L = P - kP_f \quad (5)$$

where  $k$  is the number of neutrons per fission interaction.

Part II. Case 2. A cylindrically symmetric source.

The notation for the dimensions of the source is given in Fig. 1. Since an isotropic flux is assumed it is necessary only to consider monodirectional fluxes from an appropriate number of different directions. By symmetry it is most convenient to use cylindrical coordinates. For convenience in normalization and avoidance of solid angle considerations an imaginary

sphere is placed about the source and all neutrons striking this sphere are included in the integrals and probabilities. This procedure does not affect the results when they are finally expressed in terms of neutrons lost as a percent of the source strength or in terms of a given thermal flux in the vicinity of the source.

Given that a neutron enters this imaginary sphere, let the probability of transmission without interaction be  $P_T$ , probability of interaction be  $P_I$ , probability of absorption be  $P_A$  and the probability of fission be  $P_f$ . The probability of neutron loss in the source  $P_L$  is

$$P_L = P_I - kP_f = 1 - P_T - kP_f \quad (6)$$

where  $k$  is the number of neutrons per fission. The program computes  $P_T$  and  $P_f$  by integration and  $P_I$  and  $P_A$  by  $P_I = 1 - P_T$  and  $P_A = P_I - P_f$ .

Part III. The integration and normalization.

Consider the cylindrical source surrounded with a sphere centered about the core material with radius  $R$ , just large enough to enclose the outer cylinder as shown in Fig. 1a. The thermal-neutron flux is taken to be isotropic. Neutron flux is the number of neutrons per second entering a sphere of 1 square centimetre cross section. The number of neutrons per second entering the sphere surrounding the cylindrical source is the cross sectional area of the surrounding sphere times the thermal flux.

Since the geometry is symmetric about the  $x$ -axis, the probability for various results for an isotropic flux will equal the probabilities evaluated assuming a flux uniformly distributed in  $z$ ,  $w$ , and  $\phi$  but all paths constrained to lie in planes parallel to the  $x-y$  plane. The neutron paths are perpendicular to all points along the  $w$ -axis within the sphere and  $\phi$  is the angle between the  $y$  and  $w$  axes. The probability of transmission of a neutron through the imaginary sphere averaged over all directions is thus

$$P_T = \frac{R}{2\pi} \int_{-R}^R \int_0^{2\pi} \int_{-r(z)}^{r(z)} dw P_T(z, w, \phi) [2\pi(\pi R^2)] \quad (7)$$

The probability of transmission,  $P_T(z, w, \phi)$  is

$$P_T(z, w, \phi) = e^{-(\Sigma_a a(z, w, \phi) + \Sigma_b b(z, w, \phi) + \Sigma_c c(z, w, \phi) + \Sigma_d d(z, w, \phi))} \quad (8)$$

where the  $\Sigma$ 's are the macroscopic cross sections. Parameters are as in the spherical case and the  $a$ ,  $b$ ,  $c$ , and  $d$  are the thicknesses of the A, B, C, and D materials as seen at the particular  $z$ ,  $w$ , and  $\phi$ . For a

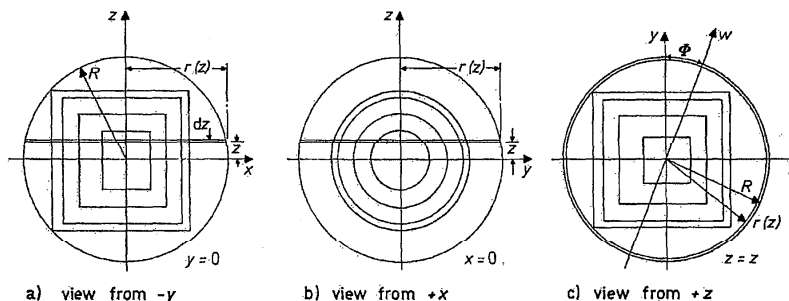


Fig. 1. Diagram for the cylindrical source program. The cylindrical source is surrounded by an imaginary sphere with center on the  $X$  axis in the middle of the core material. Fig. 1c shows the intersection of a plane parallel to the  $X-Y$  axis with the source at  $Z$ .

transparent source the macroscopic cross sections are all zero and Eq. (7) integrates to unity. The fission probability is similarly

$$P_F = \int_{-R}^R dz \int_0^{2\pi} d\varphi \int_{-\tau(z)}^{\tau(z)} dw P_f(z, w, \varphi) / 2\pi(\pi R^2) \quad (9)$$

where

$$P_f(z, w, \varphi) = (1 - e^{-\beta}) e^{-\gamma} (\sigma_{at}/\sigma_a) .$$

In  $P_f(z, w, \varphi)$ ;

$$\beta = \Sigma_a a(z, w, \varphi) ,$$

$1 - e^{-\beta}$  is the probability for interaction in the "A" material,

$$\gamma = \Sigma_b b_i(z, w, \varphi) + \Sigma_c c_i(z, w, \varphi) + \Sigma_d d_i(z, w, \varphi) ,$$

where  $b_i(z, w, \varphi)$  is the thickness of the "B" layer transversely passing into the "A" material and likewise for  $c_i$  and  $d_i$ ;  $e^{-\gamma}$  is the probability for passing through the cladding material without interaction  $\sigma_{at}$  is the fission cross section for the "A" material, and  $(\sigma_{at}/\sigma_a)$  is the probability for fission, given that an interaction occurs in the "A" material.

The cylindrical and spherical source absorption programs together with condensed versions of each program output are to be published as an NBS Technical Note [6]. The calculations were performed on a Univac 1108. The cylindrical program required 2992 words of memory with a run time of about 10 min and the spherical program required 1437 words of memory with a run time of 10 seconds.

### 3. Experimental Verification of the Calculation

#### a. Measurement of Manganous Sulfate Bath Activations by Bare and Cadmium-covered Sources

The validity of the source absorption program can be tested by its ability to predict the change in bath activity for irradiations by bare and cadmium-covered sources. A direct measurement of neutron self absorption cannot be made, but it can be changed by encapsulating the source in another material. This has been done for two Pu-Be and one Am-Be neutron sources using 2.5 mm thick cadmium covers.

The bath was 1.27 m in diameter and contained 441 g/l of manganous sulfate in distilled water. The sources were placed in a tetrafluoroethylene spherical shell, 10.16 cm outside diameter by 5 mm wall thickness, and then located at the center of the manganous sulfate bath.

A 1.3 l/s pump circulated and mixed the bath solution continuously. About 0.1 l/s were pumped to a remote and shielded sodium-iodide crystal and multi-scale counting system. The heat introduced by the pump was removed by a water-cooled heat exchanger so that the bath remained at room temperature.

The discrimination level of the counting system was set at about 20 keV and then checked daily with a precision pulser. The gain of the photomultiplier tube was checked for stability by measuring the pulse height spectrum of the 850 keV line of iron-56 of the bath activity each day. One percent shift in gain resulted in a count rate change of 0.03 %. This in turn corresponded to a 0.75 % change in high voltage, which was regulated to 0.03 %. The discriminator level shifts from day to day were at most responsible for shifts in count rate of 0.03 %. The drift of the bath count rate

was less than 0.01 % per degree C and the bath temperature was constant to within 2 degrees C.

The saturated bath count was recorded in 1000 sec intervals for total counts ranging from about 6 million to 100 million each day depending upon the emission rate of the source. Each day the recorded data were statistically analyzed within minutes via a data telephone line to a computer, so that drifts or troubles with the recording system could be quickly detected.

It was found that a single source could be placed anywhere within the tetrafluoroethylene cavity in a silk net or rest on the bottom without affecting the count rate. Most data, but not all, were taken with the source mounted near the center of the cavity in the silk net.

The dimensions and macroscopic cross-sections of the three sources used in this experiment are listed in the samples of the program output [6]. Pu-Be A contained 9.92 g of plutonium and 4.97 g of beryllium, and was encapsulated in nickel. Pu-Be M-621 contained 79.89 g of plutonium and 39.3 g of beryllium with an inner liner of tantalum and outer encapsulation of stainless steel. The Am-Be source contained 2 g of americium and 22.6 g of beryllium with an inner liner of tantalum and outer encapsulation of stainless steel.

#### b. Evaluation of Neutron Flux as Input for the Calculation

The computer program assumes that the reaction rate is proportional to the product of the thermal-neutron flux and thermal-neutron cross section for each of the cladding and core materials. This calculation used the neutron flux as measured with an indium or gold foil by the cadmium-difference method as the thermal-neutron flux in the program input for a bare source. The spectrum present in a small void at the center of a manganous sulfate bath due to a source located in that same void is not well-moderated and the neutron temperature is higher than the moderator temperature. Cadmium ratios were about 3.3 for gold foils and 5.8 for indium foils. Moreover, the presence of elements such as plutonium 239 in the source, tantalum in the liner, and cadmium in the outer capsules, with resonances in the thermal and epithermal region complicates the simple comparison intended between the experiment and calculation.

The cadmium-difference flux was measured with gold and indium foils for the plutonium-beryllium and americium-beryllium spectra in the manganous sulfate bath. The foils were counted with a sodium-iodide crystal and were calibrated in the standard thermal-neutron flux at this laboratory. The thermal flux in percent of source neutrons per centimeter squared determined by this method for a plutonium source was  $(0.122 \pm 0.003)$  with indium foils and for the americium source were  $(0.122 \pm 0.003)$  for gold foils and  $(0.119 \pm 0.003)$  for indium foils.

The thermal flux which interacts with the 0.3 eV resonance in plutonium 239 is included in the cadmium-difference measurement. The fact that the cross section is not  $1/v$  is unimportant because the absorber is infinitely thick. A larger fraction of neutrons may be absorbed in the resonance but the increased absorption there results in fewer fission absorptions because

the ratio of absorption to fission has a maximum under the resonance.

The epithermal flux, which interacts in any of the cladding or source materials, is not included in the cadmium-difference flux, which is used with the thermal neutron cross section of the respective material to determine absorption and fission. If the relative fraction of cadmium-difference neutrons, which interact in each of the core and cladding materials, is known, the additional epithermal neutron absorption in any particular material can be calculated as a small correction for an assumed epithermal spectrum.

In order to calculate the relative fraction of neutrons absorbed in each material the program for the simpler spherical case was modified to record the interactions in all materials separately. This was not done for the cylindrical case because the run time would become prohibitively long. It was also felt that sufficient information could be gained by performing the calculation on a spherical source of equivalent volume to the cylindrical source in question.

Accordingly, the calculation was performed for spherical sources with volumes of source material and cladding materials equal to those in the two Pu-Be and Am-Be cylindrical sources used in this experiment. The fact that the net absorption for the two Pu-Be sources increased by only 0.03% and 0.12% and the Am-Be source by -0.01% indicates that the equivalent spherical source is a good approximation to the cylindrical source.

is an exaggerated example of the correction for epithermal neutrons, assume 20% more interactions in the tantalum and 5% more absorption in the source material than would be predicted by the cadmium-difference flux measurement due to the presence of epithermal neutrons and significant epithermal cross section. This type of calculation has been done for thin foils in a well moderated neutron flux by Westcott *et al.* [7, 8]. Correcting the interaction probabilities for the above cases and multiplying by the measured cadmium-difference flux and area of the source would make the following changes in the source absorption: an increase of -0.02% for Pu-Be A, 0.13% for Pu-Be M-621, and 0.13% for the Am-Be source. This indicates that the correction for neglect of epithermal neutrons in the calculation for bare sources is small.

When the sources are encapsulated in 2.5 mm cadmium, no thermal neutrons penetrate into the source itself. This fact is verified by the computer program. Cadmium will also absorb neutrons in its various resonances in the epithermal region and all primary neutrons that are emitted from the source must now penetrate through the 2.5 mm layer of cadmium before entering the bath.

The fast, primary neutrons captured in cadmium were calculated using a modification of this same computer program. All fast neutrons were assumed to start at the center of the source material at the average source neutron energy. The attenuation through the outer cylindrical shell of cadmium was tallied for the absorption cross-section of the average energy source neutron, and amounted to about 0.1%. A cadmium cross-section of 0.079 b at 4.0 MeV was used for the plutonium spectrum and 0.075 b at 4.5 MeV for the americium spectrum [9].

The effective flux for thermal plus epithermal flux in the cavity was measured for cadmium by introducing a 2.5 cm diameter sphere of cadmium into the cavity with the source in question. The cadmium was suspended in a net at the top of the cavity and the source rested on the bottom. The percentage difference in bath activity with and without the cadmium sphere was divided by the cross-sectional area of the sphere to determine the percent flux in neutrons per  $\text{cm}^2$  per source neutron. In the case of the smaller plutonium-beryllium source the effective flux was found to be 33% higher than the cadmium-difference measurement. This value was used for the calculation of source absorption when it was encapsulated in cadmium.

In the case of the americium-beryllium source the effective flux for cadmium was found to be about 9% lower than the cadmium-difference flux, but the agreement was still within the error of both methods. The lower value is almost certainly due to flux depression, because of the very close proximity of the cadmium to the larger americium-beryllium source in the confined volume of the tetrafluoroethylene shell. The separation was much more favorable with the smaller plutonium-beryllium source. The flux which was used for the cadmium-encapsulated americium-beryllium source was the cadmium-difference measurement.

#### 4. Results

Table 1 lists the results of the comparison between the calculation and experiment for the three different neutron sources, Pu-Be A, Pu-Be M-621, and an Am-Be source. The interaction and fission for each source are listed in rows A and B with their differences in row C for the bare sources, using the cadmium-difference flux for each source. The slow neutron absorption when the sources are encapsulated in cadmium are listed in row D and the corresponding fast neutron absorption in row E. The calculated difference between bare and cadmium-covered source absorptions are listed in row F and the measured differences from bath activities are listed in row G.

The good agreement between the calculated and measured values of net absorption confirms the fact that the single interaction approximation, computer calculation of source absorption is quite reliable.

The results for Pu-Be sources also show that the absorption interactions are almost cancelled by the fission in the plutonium. A smaller fraction of fission neutrons are produced in the plutonium, when the source has an inner liner of tantalum, because of the large macroscopic absorption cross section of tantalum.

The calculation shows that corrections to managanous sulfate bath calibrations for self absorption of thermal neutrons in the source are very important for sources with large macroscopic absorption or fission cross sections. Plutonium-beryllium source calibrations are fortuitously less subject to error from this cause because of the cancelling effect of the fission neutrons from plutonium.

The calculation of self absorption in a cylindrical source can be quite accurately approximated by performing the calculation on a spherical source with equivalent volumes of source materials.

This calculation has been used to predict the difference in source strength in a water moderator for

the plutonium-beryllium source of Cap *et al.* [4] with and without a cadmium cover. They reported an experimentally measured difference of 3.62%. They calculated the percent of fission and absorbed neutrons in the core material to be 2.86% and 5.43%, respectively. From the difference of these numbers they infer that 1.05% of the neutrons are absorbed in the source encapsulation. The quantities calculated for rows A to F of our Table 1 for their source were: 2.068%, 1.761%, 0.306%, 3.89%, 0.04%, and 3.62%. Our calculated net difference is in very good agreement with their experimentally measured difference, but we do not agree with their calculated values of fission and absorbed neutrons in the core material.

Our calculation predicts 1.17% absorption in the encapsulating material, 0.63% fission interaction in the core, and 0.27% absorption in the core. Because the interaction in the core is 0.90% compared to 1.17% in the encapsulation, the simplification of Cap *et al.* to ignore the encapsulation in their calculation is not warranted. Moreover, using our spherical source absorption program for the bare equivalent spherical source of Cap *et al.*, we calculate a neutron absorption interaction of 0.44%, neutron fission interaction of 1.01%, and neutron fission emission of 2.82%. According to our calculation, therefore, the bare source has a net neutron gain of 1.37% compared to a net loss of 0.306% for the encapsulated source.

Table 1. Results of intercomparison between calculation and experiment

	Pu-Be A (%)	Pu-Be M-621 (%)	Am-Be (%)
A Calculated absorption plus fission interaction for the bare source irradiation (neutron disappearance)	0.652	2.068	0.966
B Calculated fission neutron emission for bare source irradiation	0.809	1.864	0.003
C Bare irradiation net source absorption (A - B)	-0.157	0.143	0.963
D Calculated slow-neutron absorption for cadmium covered source <sup>a</sup>	1.824	4.094	2.193
E Calculated fast-neutron absorption for cadmium covered source	0.10	0.11	0.11
F Calculated difference between bare and cadmium-covered source absorption (D + E - C)	2.08	4.06	1.34
G Experimentally measured difference from bath irradiations	1.93 ± 0.03	4.05 ± 0.1	1.44 ± 0.02

<sup>a</sup> The cadmium-difference flux is used for A and B, but an effective flux for cadmium, as explained in the text, is used for D.

In our calculation we have used macroscopic fission and absorption cross sections for the Pu-Be<sup>13</sup> core material of 4.345 per cm and 1.883 per cm. The isotopic content is usually 91% Pu 239, 8% Pu 240, and 1% Pu 241. The mass ratio of plutonium to beryllium of Cap *et al.* was 2.13 and the density of the compressed mixture 3.7 g/cm<sup>3</sup>. The ratio of fission interaction to total interaction in the core should be  $4.345/(4.345 + 1.883) = 0.698$  and, if the number of neutrons per fission is 2.8, the ratio of fission produced neutrons to absorbed neutrons is  $2.8 \times 0.698 = 1.95$ . The ratio reported by Cap *et al.* is  $2.86\%/5.43\% = 0.527$ .

We assumed that the cadmium cover used by Cap *et al.* was 1 mm thick. A value of 0.167% source neutrons per cm<sup>2</sup> was determined from their Fig. 1 of the cadmium-difference flux together with a neutron production rate of the source of  $1.15 \times 10^6$  neutrons per second per gramme of plutonium, which we have found to be the average for many calibrated sources. The exact source dimensions and construction were available from the manufacturer's catalog.

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