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in a Monoatomic Heavy Gas

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FROM: S. N. Purohit

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ABSTRACT

The time-dependent energy spectra, for times greater than the slowing-down time, were generated in a monoatomic heavy gas with the help of a multigroup formalism. These spectra were obtained for the infinite as well as finite media of beryllium and graphite. The behavior of asymptotic energy spectra during the last stage of neutron thermalization and diffusion periods was studied. The thermalization time constant for the establishment of the final Maxwellian velocity distribution of neutrons, in a monoatomic heavy gas, was estimated to be equal to $(1.174 \xi \sum_{g0} v_0)^{-1}$. Total thermalization times for neutrons in beryllium and graphite were found to be equal to 114 and 238 μ sec, respectively. Using the energy-dependent transport mean free path, the diffusion cooling coefficient for beryllium was calculated to be equal to 0.890 cm^2 . For graphite, under the constant diffusion coefficient assumption, the diffusion cooling coefficient was determined to be equal to 1.922 cm^2 .

1. INTRODUCTION

Time-dependent studies of neutron moderation were undertaken by Ornstein and Uhlenbeck (1), Marshak (2), von Dardel (3), Kazarnovsky (4), Waller (5), Eriksson (6), and others. Amaldi (7) has reviewed most of these studies in an excellent article. In all these studies the atoms of the moderator were assumed to be at rest and free. This paper gives the results of studies of the time behavior of the neutron energy distribution in a heavy gas for times greater than the slowing-down time, with the objective of obtaining the characteristics of the energy distribution during the last stage of neutron thermalization and diffusion. The behavior of the energy distribution of neutrons during these periods is required for analyzing and interpreting the results of pulsed-neutron experiments. Moreover, knowledge of time-dependent energy spectra below 1 ev in a heavy gas is in itself a sufficient motivation.

Using the Wilkins time-dependent differential equation for a heavy gas, time-dependent energy spectra have been generated for several interesting physical cases for beryllium and graphite. With the help of these energy spectra, the total thermalization time, the thermalization time constant for the establishment of Maxwellian distribution, and the characteristics of the asymptotic energy spectra in finite media--i.e., the average speed, the decay constant, and the diffusion cooling coefficient--have been determined.

A multigroup formalism based upon the division of the thermal energy interval between zero and E_T into a finite number of energy groups has been developed. The thermal cutoff energy is represented by E_T and is taken to

be equal to 10 KT. The multigroup method reduces the problem to the solution of a set of coupled, linear, first-order differential equations, which are solved by means of an electrical analog computer, with time as a continuous variable. The analog computer simulates the time behavior of a pulse of neutrons during thermalization and diffusion periods and gives the energy spectrum at any desired time. The studies reported in this paper have also been treated in detail in another publication (8). Purohit and Zweifel (9) reported some of the earlier results.

2. THEORY

In the diffusion approximation the distribution of neutrons in the thermal energy range is given as follows:

$$\begin{aligned} \frac{1}{v} \frac{\partial \phi(E, \underline{r}, t)}{\partial t} = & - (\Sigma_a - D \nabla^2) \phi(E, \underline{r}, t) - \Sigma_s \phi(E, \underline{r}, t) \\ & + \int_0^{E_T} \Sigma_s(E') \phi(E', \underline{r}, t) F(E' \rightarrow E) dE' \\ & + \int_{E_T}^{E/\alpha} \Sigma_s(E') \phi(E', \underline{r}, t) F(E' \rightarrow E) dE' + S(E, \underline{r}, t) \end{aligned} \quad (1)$$

In Eq. (1) the various symbols have their conventional meaning. The slowing-down integral is broken up into two parts, as a matter of convenience. During the energy interval ($0 \leq E \leq E_T$) the scattering frequency $F(E' \rightarrow E)$ is given by the thermalization model, and in the energy range ($E_T \leq E \leq E/\alpha$) by the slowing-down model. Expanding $\phi(E, \underline{r}, t)$ into a

series of orthonormal space functions, we get

$$\phi(E, \underline{r}, t) = \sum_n \phi_n(E, t) \phi_n(\underline{r}) \quad (2)$$

$$\nabla^2 \phi_n(\underline{r}) = -B_p^2 \phi_n(\underline{r}) \quad (3)$$

$$\phi_n(\underline{r}) = 0 \text{ at the extrapolated boundary}$$

We assume the extrapolated distance to be constant with energy. The external source is assumed to be monochromatic and pulsed, and can be expressed mathematically as follows:

$$\begin{aligned} S(E, \underline{r}, t) &= S_0 \delta(E - E_0) \delta(t) \phi(\underline{r}) \\ &= 0 \quad \text{for } 0 \leq E \leq E_T \end{aligned} \quad (4)$$

Using the above equations, we get the following equation for any spatial mode having the geometrical buckling B^2 :

$$\begin{aligned} \frac{1}{v} \frac{\partial \phi(E, t)}{\partial t} &= - \left(\sum_a + DB^2 \right) \phi(E, t) - \sum_s \phi(E, t) \\ &+ \int_0^{E_T} \sum_s(E') \phi(E', t) F(E' \rightarrow E) dE' \\ &+ \int_{E_T}^{E/\alpha} \sum_s(E') \phi(E', t) F(E' \rightarrow E) dE' \end{aligned} \quad (5)$$

Source for Thermalization

The neutrons which serve as a source for the thermalization problem are the last collision neutrons arriving at an energy E less than E_T . These neutrons will come from the elastic scattering collisions at some energy between E_T and E_T/α . The neutrons which are scattered between this interval become the source for the neutrons of energy less than E_T . This source is given by the slowing-down scattering integral, and is represented by

$$S(E, t) = \int_{E_T}^{E/\alpha} \Sigma_s(E') \phi(E', t) P(E' \rightarrow E) dE' \quad (6)$$

$S(E, t)$ is represented by the Dirac delta function in the following manner:

$$S(E, t) = \delta(t - \bar{t}_s) \int_{E_T}^{E/\alpha} \Sigma_s(E') \phi(E') P(E' \rightarrow E) dE' \quad (7)$$

where $\bar{t}_s$ is the mean slowing-down time for the source neutrons to have their final energy between E_T and E_T/α . The above representation [Eq. (7)] for the source is essentially given by the continuous slowing-down theory. Using the expression for $\phi(E')$, as given by the age theory, we obtain for $S(E, t)$:

$$S(E, t) = \frac{\delta(t - \bar{t}_s)}{\xi(1 - \alpha)} \left(1 - \frac{\alpha E_T}{E} \right) pe^{-B^2 \tau} \quad \text{for } \alpha E_T \leq E \leq E_T \quad (8)$$

= 0 otherwise

In deriving the above result, we have made the following assumptions:

1. The slowing-down time for the neutrons to a final energy in the energy interval between E_T and E_T/α can be given by $\bar{t}_s$.
2. The resonance escape probability p and the nonleakage probability $e^{-B^2\tau}$ remain constant in the energy interval between E_T and E_T/α .

It must, however, be pointed out that the details of the initial source are not important for the study of the asymptotic behavior of neutrons. Using the above source we shall generate the time-dependent energy spectra for times greater than $\bar{t}_s$ for beryllium and graphite. $\bar{t}_s$ is equal to 14 and 38 μsec for beryllium and graphite, respectively.

Heavy-Gas Differential Equation

The energy exchange scattering kernel for a monoatomic gas was obtained by Wigner and Wilkins (10), and they assumed the atoms of the gas to have a Maxwellian velocity distribution. For a heavy mass, the integral equation (5) can be transformed into the Wilkins differential equation (11):

$$E \frac{\partial^2 \phi(E, t)}{\partial E^2} + E \frac{\partial \phi(E, t)}{\partial E} + \phi(E, t) \left[1 - \frac{\sum_a(E) + D(E) B^2}{\xi \sum_{s0}} \right] = \frac{1}{\xi \sum_{s0} v} \frac{\partial \phi(E, t)}{\partial t} \quad (9)$$

In Eq. (9) the energy variable is expressed in units of KT , with T as the moderator temperature. The functions $\partial \phi(E, t)/\partial E$ and $\partial^2 \phi(E, t)/\partial E^2$ are expressed in the following manner: let ΔE be the energy interval between

the $(n+1)$ th and the n th energy groups; let $\Delta E'$ be the energy interval between the n th and $(n-1)$ th energy groups. If ϕ_{n+1} , ϕ_n , and ϕ_{n-1} represent the neutron distributions for $(n+1)$, (n) , and $(n-1)$ th energy groups, then we have:

$$\left(\frac{\partial \phi}{\partial E}\right)_n = \frac{1}{2} \left(\frac{\phi_{n+1} - \phi_n}{\Delta E} + \frac{\phi_n - \phi_{n-1}}{\Delta E'} \right) \quad (10)$$

$$\left(\frac{\partial^2 \phi}{\partial E^2}\right)_n = \frac{2}{\Delta E + \Delta E'} \left(\frac{\phi_{n+1} - \phi_n}{\Delta E} - \frac{\phi_n - \phi_{n-1}}{\Delta E'} \right) \quad (11)$$

Using Eqs. (10) and (11), we get the following equation representing the n th energy group which is coupled to the $(n+1)$ th higher group and $(n-1)$ th lower group:

$$\begin{aligned} \phi_{n+1} \left[\frac{2 E_n^{3/2}}{\Delta E (\Delta E + \Delta E')} + \frac{E_n^{3/2}}{2 \Delta E} \right] \\ - \phi_n \left\{ E_n^{3/2} \left[\frac{2 (\Delta E + \Delta E')}{\Delta E' \Delta E (\Delta E + \Delta E')} + \frac{1}{2} \left(\frac{1}{\Delta E} - \frac{1}{\Delta E'} \right) \right] + E_n^{1/2} [1 + \beta(E_n)] \right\} \\ + \phi_{n-1} \left[\frac{2 E_n^{3/2}}{\Delta E' (\Delta E + \Delta E')} - \frac{E_n^{3/2}}{2 \Delta E'} \right] = \alpha \frac{\partial \phi_n}{\partial t} \end{aligned} \quad (12)$$

where

$$\alpha = \frac{1}{\sum_{s=0}^S v_s \sigma_s}$$

$$v = E^{1/2} v_0$$

$$\beta(E_n) = \frac{\sum_a(E_n) + D(E_n) B^2}{\sum_{s0}}$$

Equation (12) was employed to generate the time-dependent energy spectra using the source given by the slowing-down theory according to Eq. (8). For the set of problems dealing with beryllium, 20 energy groups were employed between the zero- and 10-KT energy interval. Ten groups of 0.1-KT width were chosen between zero and 1 KT, and the other ten groups between 1 KT and 10 KT of 1-KT width. For the set of problems dealing with graphite, the energy interval between zero and 10 KT was divided into 14 energy groups.

3. RESULTS

The results obtained for the infinite and finite media of beryllium and graphite are discussed below and are compared with the analytical and experimental results.

Infinite Medium, Zero Absorption

The energy spectra for beryllium are plotted in Fig. 1 at different times in the interval of $t = \bar{t}_s$ to $t = 350 + \bar{t}_s$ μ sec, and are plotted in Fig. 2 for graphite in the interval of $t = \bar{t}_s$ to $t = 510 + \bar{t}_s$ μ sec. These energy spectra converge finally into the Maxwellian distribution $M(E) = Be^{-E}$. The degree of convergence is very good. After attaining the Maxwellian distribution the neutrons retain this spectrum at all further times. In the nonabsorbing infinite medium the final asymptotic energy distribution is Maxwellian. This is a basic result of the H theorem of statistical mechanics and holds good for any scattering model.

The time behavior of a few energy groups is given in Figs. 3 and 4

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Fig. 1. Time Behavior of Neutron Energy Spectrum in Beryllium for Infinite Medium and Zero Absorption.

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Fig. 2. Time Behavior of Neutron Energy Spectrum in Graphite for Infinite Medium and Zero Absorption.

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Fig. 3. Time Behavior of Energy Groups in Beryllium for Infinite Medium and Zero Absorption.

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Fig. 4. Time Behavior of Energy Groups in Graphite for Infinite Medium and Zero Absorption.

to demonstrate the existence of the asymptotic distribution. Parallel curves indicate the existence of the asymptotic energy spectrum after the thermalization time, defined as the total time required by the source neutrons to have slowed down and to have been completely thermalized so as to attain the asymptotic energy distribution and to retain it for all times thereafter. It may be noted that the precise determination of the thermalization time is not possible, as it is impossible to determine the state of complete thermalization. However, an estimate with an uncertainty of the order of 5 to 10% can be given with the help of the curves in Figs. 3 and 4.

The thermalization time for beryllium is estimated to be equal to 100 μ sec plus the mean slowing-down time of 14 μ sec. Kothari and Singwi (12) report the thermalization time in crystalline beryllium to be about 145 μ sec, and calculated that the time required by the neutrons to reach the moderator temperature 310°K from 2000°K to be about 130 μ sec.

For graphite, we estimate the thermalization time to be 200 μ sec plus the mean slowing-down time of 38 μ sec. In their experiments, Antonov et al. (13) report the thermalization time in graphite to be 200 μ sec. Beckurts (14) determined the thermalization time constant for graphite from his experiments to be 185 ± 40 μ sec, and Kothari and Khubchandani (15) reported it to be 170 μ sec. The thermalization time constant in graphite might be of the same order as the thermalization time, as indicated by the results of Beckurts and Antonov. It is interesting to note that the estimates of the thermalization time for beryllium and graphite given by the neutron distribution $\phi(E, t)$ on a heavy-gas model are of the same order as those given by other authors for the crystalline medium.

The curves given in Figs. 3 and 4 provide excellent data for determining the thermalization time constant. We use the beryllium data for $\phi(E, t)$ in the time interval between 60 and 120 μsec , and fit in the following manner, using the Cornell method (16):

$$\phi(E, t) = \phi_0(E) + \phi_1(E) e^{-\lambda_1(t-60)}, \quad \text{for } t \geq 60 \mu\text{sec} \quad (13)$$

In the above equation t equal to zero corresponds to t equal to $\bar{t}_g$. $\phi_0(E)$ and $\phi_1(E)$ are plotted in Fig. 5. λ_1 is calculated to be equal to $4.75 \times 10^4 \text{ sec}^{-1}$ for beryllium; when converted, λ_1 is found to be equal to $1.176 \frac{E}{\sum_{s0} v_0}$ units. Figure 6 shows plots of $\phi_1(E)$ obtained numerically and analytically, with the curves normalized at 0.4 KT. The analytical curve was obtained by using the first four associated Laguerre polynomials of order one (8). The agreement is excellent on the low-energy side. Both curves have a minimum at 0.4 KT and a maximum at 2.5 KT, but the numerical curve has a zero at 1.23 KT and the analytical curve a zero at 1.27 KT. The analytical result for λ_1 using the first four associated Laguerre polynomials of order one is equal to $1.274 \frac{E}{\sum_{s0} v_0}$ (8). The thermalization time constant is the reciprocal of λ_1 . For beryllium the thermalization time constant according to the numerical and analytical values of λ_1 is equal to 21.05 and 19.5 μsec , respectively. Kothari and Singwi (12) and von Dardel and Sjöstrand (17) report 21 and 22 μsec , respectively. The analytical value of λ_1 would decrease if more than four polynomials were used.

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Fig. 5. Exponential Fit of the Infinite-Medium Data for Beryllium.

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Fig. 6. Comparison of Numerical and Analytical Eigenfunction $\phi_1(E)$.

Finite Medium, Zero Absorption

Time-dependent energy spectra for B^2 equal to 2.96×10^{-2} , 3.96×10^{-2} , 5.36×10^{-2} , and $7.18 \times 10^{-2} \text{ cm}^{-2}$ for beryllium assemblies are plotted in Figs. 7 through 10. These assemblies were used by deSaussure and Silver (18) in their experiments. The transport mean free path was taken to be energy dependent, as given by Bhandari (19). In Fig. 11 the energy spectra for graphite at 300 μsec are plotted for several values of B^2 . These spectra were generated under the constant diffusion coefficient assumption, using Beckurts' (14) experimental value of the diffusion coefficient of 0.968 cm. The asymptotic energy spectra for B^2 equal to zero and $7.18 \times 10^{-2} \text{ cm}^{-2}$ for beryllium are plotted in Fig. 12. The asymptotic energy spectra for B^2 equal to zero and $18.5 \times 10^{-2} \text{ cm}^{-2}$ for graphite are given in Fig. 13. These curves demonstrate the shift of the energy spectrum toward the low-energy side with increase in the geometrical buckling. The softening of the energy spectrum in a finite medium is due to the preferential leakage of neutrons of higher energy. The above results furnish the direct evidence of the diffusion cooling.

The time behavior of the energy distribution after the thermalization time was found to be given by a decaying exponential. In all the previous studies it was assumed that at sufficiently long times the neutron distribution $\phi(E, t)$ can be characterized by a single decay constant. The existence of an asymptotic distribution in a finite medium having $B^2 \leq 7.18 \times 10^{-2} \text{ cm}^{-2}$ for beryllium and $B^2 \leq 18.5 \times 10^{-3} \text{ cm}^{-2}$ for graphite has been established in this study. Von Dardel and Sjöstrand (20) obtained the diffusion cooling coefficient from the measurements of the decay constants

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Fig. 7. Time Behavior of Neutron Energy Spectrum in Finite Medium of Beryllium with Geometrical Buckling $B^2 = 2.96 \times 10^{-2} \text{ cm}^{-2}$.

Fig. 8. Time Behavior of Neutron Energy Spectrum in Finite Medium of Beryllium with Geometrical Buckling $B^2 = 3.96 \times 10^{-2} \text{ cm}^{-2}$.

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Fig. 9. Time Behavior of Neutron Energy Spectrum in Finite Medium of Beryllium with Geometrical Buckling $B^2 = 5.36 \times 10^{-2} \text{ cm}^{-2}$.

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Fig. 10. Time Behavior of Neutron Energy Spectrum in Finite Medium of Beryllium with Geometrical Buckling $B^2 = 7.18 \times 10^{-2} \text{ cm}^{-2}$.

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Fig. 11. Neutron Energy Spectrum at 300 μsec in Graphite for Various Geometrical Bucklings, B^2 .

Fig. 12. Equilibrium Neutron Energy Spectra in Beryllium for Geometrical Buckling $B^2 = 0$ and $7.18 \times 10^{-2} \text{ cm}^{-2}$.

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Fig. 13. Equilibrium Neutron Energy Spectra in Graphite for Geometrical Buckling $B^2 = 0$ and $18.5 \times 10^{-3} \text{ cm}^{-2}$.

in finite media. The decay constant is given by the following equation:

$$\lambda_0 = (\overline{Dv})_0 (1 - CB^2)B^2 \quad (\text{zero absorption}) \quad (14)$$

$(\overline{Dv})_0$ is averaged over the Maxwellian distribution; C is the diffusion cooling coefficient. By fitting the decay constants against B^2 , we get the following result for beryllium:

$$\lambda_0 = 1.267 \times 10^5 (1 - 0.890 B^2)B^2 \quad (15)$$

The value of $(\overline{Dv})_0$ averaged over the Maxwellian distribution is equal to $1.267 \times 10^5 \text{ cm}^2/\text{sec}$, which agrees with the experimental values of $1.25 \times 10^5 \text{ cm}^2/\text{sec}$ and $1.25 (+0.06) \times 10^5 \text{ cm}^2/\text{sec}$ as given by Komoto and Kloverstrom (21) and deSaussure and Silver (18), respectively. The diffusion cooling coefficient given by Eq. (15) is equal to 0.890 cm^2 .

Hurwitz and Nelkin (22) and Häfele and Dresner (23) have given the theoretical expressions for the diffusion cooling coefficient for a heavy gas using the constant diffusion coefficient assumption. The value of diffusion cooling coefficient according to the formulas given by these authors using $(\overline{Dv})_0$ equal to $1.25 \times 10^5 \text{ cm}^2/\text{sec}$ is found to be 0.46 cm^2 . Nelkin (24) and Singwi (25) gave the diffusion cooling coefficient for the crystalline model. Singwi's (25) value using the constant diffusion coefficient for beryllium is 0.9 cm^2 for $(\overline{Dv})_0$ equal to $1.25 \times 10^5 \text{ cm}^2/\text{sec}$; his estimate of the diffusion cooling coefficient is 50% higher than the value obtained for the heavy-gas model under the constant diffusion coefficient. Using the energy-dependent transport mean free path given by

Bhandari, we have found the diffusion cooling coefficient to be 50% higher than for the constant diffusion coefficient for the heavy-gas model. It should, therefore, be concluded that the variation of transport mean free path with energy is as important as the crystalline effects in the calculation of the diffusion cooling coefficient for beryllium. The experimental values of the diffusion cooling coefficient as reported by deSaussure and Silver (18), Antonov et al. (13), and Komoto and Kloverstrom (21) are 1.1 ± 0.9 , 2.5 ± 0.9 , and $2.0 \pm 0.9 \text{ cm}^2$, respectively. The result of deSaussure and Silver (18) is of the same order as the theoretical value of Singwi (25) neglecting the energy dependence of transport mean free path. It is speculated that the exact theoretical value would be about 50% higher than the experimental result of deSaussure and Silver, which appears to be low.

For graphite the diffusion cooling coefficient using the constant diffusion coefficient assumption is estimated to be 1.922 cm^2 . The experimental value of Beckurts (14) is $7 \pm 1.0 \text{ cm}^2$. For the constant diffusion coefficient equal to 0.9 cm, Singwi (25) has estimated the diffusion cooling coefficient to be equal to 7.0 cm^2 for the crystalline medium. Parks et al. (26) have given 6.80 cm^2 for graphite, using the crystalline model. The diffusion cooling coefficients obtained experimentally as well as theoretically for the crystalline model are larger than for the heavy-gas model. It may, therefore, be concluded that the heavy-gas model underestimates the diffusion cooling coefficient for graphite. A similar observation was previously made by Hurwitz and Nelkin (22).

The average speeds, $\bar{v}$, corresponding to the asymptotic energy spectra for various values of B^2 are presented in Table I for beryllium and graphite. If the average speed is proportional to the square root of neutron temperature, which is true if the neutrons have a Maxwellian distribution, then the change in average speed is a measure of the change in neutron temperature with B^2 .

By fitting the values given in Table I against B^2 we get the following results:

$$\bar{v} = 1.088 v_0 (1 - 1.852 B^2) \quad (\text{graphite}) \quad (16)$$

$$\bar{v} = 1.110 v_0 (1 - 0.518 B^2) \quad (\text{beryllium}) \quad (17)$$

For the Maxwellian distribution corresponding to B^2 equal to zero the exact theoretical value is equal to $1.129 v_0$. Equations (16) and (17) should give the change in $\bar{v}$ with the change in B^2 correctly.

4. CONCLUSION

The multigroup method developed in this study is unique, since it simulates the physical situation and follows a pulse of neutrons during the thermalization and diffusion periods. The advantage of the method is that it describes the neutron distribution function $\phi(E, t)$ completely, provided that the initial source is given exactly. It gives the correct asymptotic behavior which describes the last stage of neutron thermalization and diffusion. It might be possible to generate the time-dependent energy spectra for the crystalline model by a similar multigroup technique. The

Table I

Average Speed, $\bar{v}$, in Beryllium and Graphite

B^2 (cm ⁻²)	$\bar{v}$ (v_0 units)
Beryllium	
0	1.106
2.94×10^{-2}	1.0941
5.36×10^{-2}	1.0919
7.18×10^{-2}	1.0586
Graphite	
0	1.0914
2.96×10^{-3}	1.0820
4.62×10^{-3}	1.0789
8.21×10^{-3}	1.0655
18.50×10^{-3}	1.0513

limitations of the method are governed by the machine used to generate the distributions. The accuracy of the results obtained increases with the number of groups, which is limited by the machine itself. The uncertainty in the beryllium results is less than 5% and for graphite is of the order of 5%.

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Distribution

1. F. S. Alsmiller	22. J. T. Mihalcz
2-5. E. P. Blizard	23. F. J. Muckenthaler
6. A. D. Callihan	24. R. W. Peelle
7. E. C. Campbell	25. S. K. Penny
8. G. T. Chapman	26. R. Perez-Belles
9. R. R. Coveyou	27-52. S. N. Purohit
10. G. deSaussure	53. A. Simon
11. L. Dresner	54. M. J. Skinner
12. R. Gwin	55. E. G. Silver
13. K. M. Henry	56. K. S. Singwi
14. L. B. Holland	57. J. T. Thomas
15. E. Inonu	58. A. M. Weinberg
16. E. B. Johnson	59. W. Zobel
17. W. H. Jordan	60-61. Central Research Library
18. P. Kasten	62-73. Laboratory Records Department
19. T. A. Love	74. Laboratory Records, ORNL R.C.
20. R. E. Maerker	75-89. Technical Information Service,
21. F. C. Maienschein	AEC

External

90. N. Corngold, Brookhaven National Laboratory, Upton, Long Island, N. Y.
91. J. Chernick, Brookhaven National Laboratory, Upton, Long Island, N. Y.
92. H. J. Comberg, Nuclear Engineering Dept., University of Michigan, Ann Arbor, Michigan
93. R. V. Meghreblan, Jet Propulsion Laboratory, Pasadena, California
94. M. Melkin, General Atomics, P.O. Box 608, San Diego 12, California
95. R. K. Osborn, University of Michigan, Ann Arbor, Michigan
96. D. E. Parks, General Atomics, P.O. Box 608, San Diego 12, California
97. P. P. Zweifel, Nuclear Engineering Dept., University of Michigan, Ann Arbor, Michigan