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A MONTE CARLO STUDY OF NEUTRON THERMALIZATION*

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ABSTRACT

The slowing down of neutrons from fission to absorption energies in a homogeneous medium has been treated by a Monte Carlo method. Thermalization by elastic collisions in both light and heavy water was computed on an IBM 650. One-sixth the mean-square distance travelled by the neutrons was interpreted as the migration area. This quantity was analyzed to give the age, τ, both above and below the indium resonance, and the thermal diffusion constant, D, as functions of the moderator temperature, the effective moderator mass, the moderator composition, the scattering and absorption laws, and the energy distribution of the neutron source.

The migration area from the fission energy source to below the indium resonance energy (1.46 ev) was found to be 27.4 cm^2 for H_2O and 120 cm^2 for D_2O. By varying the absorption cross section, the migration area from the indium resonance energy to absorption of the neutrons could be decomposed into components D/L^2 and τ_s. D and τ_s were found to be 0.19 cm and 0.77 cm^2 respectively for H_2O, and 0.87 cm and 20 cm^2 for D_2O. D was found to increase with the thermal agitation of the moderator at the rate of 0.15%/°C for H_2O and 0.14%/°C for D_2O. Neutron energy distributions found from the scattering frequency agree with those derived by analytical methods. (auth)

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1. INTRODUCTION

A comprehensive study of neutron thermalization involves the diffusion of neutrons in space and energy as they are slowed from fission energies to thermal absorption. Such a study must be concerned with the energy of the source of neutrons, the scattering and absorption cross sections over the entire slowing-down range, the molecular mass of the moderator, its temperature, and the molecular binding.

The solution of the thermalization problem is needed to compute moderator properties such as migration areas and group constants. These in turn are used to estimate k_{eff} for reactors, for subcritical assemblies, and for containers of fissile materials. Further objectives of the calculation were to study the decomposition of the migration area into fast and thermal components, and to find the variation of the migration area with the temperature of the moderator.

Other slowing-down studies have been based on analytical evaluation of the moments of the slowing-down distribution (Wilkins, Hellens, and Zweifel (1956)). However, variation of parameters by analytical methods is difficult because of the complexity of the solutions. A question unresolved by the earlier work was the effect of temperature on the slowing-down phenomenon in the transition region between fast and slow neutrons. Parametric variation is particularly easy in Monte Carlo methods where the input data can have arbitrary form.

2. THE MONTE CARLO CODE

A Monte Carlo code for the IBM 650 computer is used at the Savannah River Laboratory to compute slowing down in an infinite homogeneous medium. Neutron histories are computed, collision by collision, as the neutrons are thermalized from the fission source. In each collision either a scattering or an absorption takes place according to the relative cross section at the neutron's energy. If the neutron is absorbed a new history is begun. If it is scattered, the energy loss in scattering is determined from the equations of elastic collision. It is assumed that the scattering is isotropic in the center-of-mass frame of reference. After scattering, a new event occurs, again as determined by the relative cross sections for scattering and absorption. This process is continued until the neutron is absorbed or until it is slowed down past the range of interest. The calculation was performed in two parts; slowing down from fission to indium resonance (1.46 ev) and slowing down from indium resonance to absorption. A 1/v absorption rate was assumed in the energy range below the indium resonance, but the code is not restricted to such an assumption.

Precision is improved and the computing time speeded by a statistical estimation of the migration area and through the extensive use of tables. Cross sections, source energy distributions, and moderator energy distributions are obtained, as needed, from tables instead of being computed from formulas.

Eight digit random numbers, R, were generated by a process suggested by Lehmer (1951) which is well suited for application to digital computers:

$$R_{n+1} = 23R_n \bmod (10^8 + 1)$$

The random numbers are used to choose scattering and absorption probabilities, angles of scattering, and target velocities.

A neutron history is acquired in the following sequence: a) select the initial neutron energy, b) determine the type of event, c) assign the scattering mass, d) determine the target velocity, e) calculate the energy loss, f) accumulate the addition to the migration area, then begin the next event. When a neutron is absorbed or scattered out of the energy range of interest, the history is terminated. These steps are discussed in the following paragraphs.

Initial Energy

The initial energy may be the same for all neutrons as it is for slowing down from indium resonance to absorption, or it may be taken in tabulated order from a properly biased table as it is for high energy calculations. The source for high energy scattering was taken to be a fission energy distribution in the form of a least-squares maxwellian fit to data reported by Rosen (1955). Sequential sampling of the entire table assures an accurate representation.

Type of Event

The probability, p_i, of an event of type i occurring among competing events is $p_i = \frac{\sigma_i}{\Sigma_j \sigma_j}$, where the σ_j are macroscopic cross sections in the laboratory frame of reference. Tables of $P_i = \sum_{k=1}^{i} p_k$ are stored in the IBM 650 as functions of energy. An event is of type i if a number R, selected at random from the range 0 to 1, is in the interval $P_{i-1} \leqslant R < P_i$. If the event is an absorption, the migration area at which absorption occurs, the

energy at which absorption occurs, the nuclear identity of the absorbing atom, the number of the neutron, and the number of the collision are recorded. A new neutron is then selected from the source.

Scattering Mass

Chemical binding of the moderator (target) atoms, such as that encountered in water moderators, increases the inertia of the target in collision. The influence of this effect is approximated in the calculation by using a larger mass for the target atom.

When the incident neutron has energy in excess of the vibrational binding energy of the molecule, E*, it is assumed that the target is unbound and free atom masses are used. When the energy of the incident neutron is less than E*, the effective masses derived by Brown and St. John (1954) are employed. Thus, in D_2O, the effective mass of oxygen, A(O), is 16 and that of deuterium, A(D) is 2 for events occurring above E* = 0.143 ev. A(O) is 20 and A(D) is 3.6 for events occurring below E*. Similarly for H_2O, A(O) is 18 and A(H) is 1.9 for events occurring below E* = 0.226 ev.

Target Velocity

The probability, p, of collision between a neutron of velocity $\vec{u}$ and a target of velocity $\vec{U}$ depends upon the relative velocity and the thermal distribution of the target atom velocities. It is customary to assume that the target atoms possess a maxwellian distribution of velocities, $M(\vec{U})$, in the laboratory system and that the scattering cross section in the center-of-mass coordinates is independent of the target velocity. The probability of collision

with a moderator atom is obtained by integrating over all possible velocities.

$$\int p \, d\vec{U} = \int_0^\infty |\vec{u} - \vec{U}| \, M(\vec{U}) \, d\vec{U}$$

$$= \frac{1}{\beta \sqrt{\pi}} e^{-\beta^2 u^2} + \left(\frac{1}{2\beta^2 u} + u\right) \mathrm{Erf}(\beta u)$$

where $\beta^2 = \frac{A}{2kT}$, kT represents the moderator temperature, and $\mathrm{Erf}(x) = \frac{2}{\sqrt{\pi}} \int_0^x e^{-y^2} dy$ is the error function. The target velocity must be selected from equally weighted samples of this integral probability.

Since the integral probability is complex and difficult to decompose into equally weighted samples, we deal instead with the differential probability. The selection of U is performed by a rejection technique. An upper bound of the integrand is u+U. Thus, an upper bound of the probability is

$$\int p_{max} \, d\vec{U} = u \int_0^\infty M(U) dU + \int_0^\infty U \, M(U) dU$$

$$\equiv u+I$$

A random number R_1 is drawn to decide the selection of U. If $R_1 \leqslant \frac{u}{u+I}$, a value of U is taken at random from a table biased with a maxwellian distribution. If $R_1 > \frac{u}{u+I}$, U is selected from a table biased with a U-weighted maxwellian distribution. Although the U so obtained is representative of the upper bound probability, p_{max}, the actual probability is more restrictive. Every velocity U is chosen too frequently by the p_{max} sampling. The frequency should be reduced by the fraction by which p_{max} exceeds the actual p. Thus a second random number, R_2, is drawn to test the choice of U. If $R_2 \leqslant \frac{\vec{u}-\vec{U}}{u+U}$, U is accepted as the target velocity. Otherwise the

sample is rejected and another is chosen in the same manner.

Scattering Mechanics

The equations of neutron moderation are taken from simple "billiard-ball" mechanics. Details of the j^{th} neutron collision with the moderator atoms are derived from the conservation of energy and momentum. Upon collision, the following scattering data are calculated sequentially to obtain the energy loss and the scattering angle.

$$x_j^2 = \frac{1}{(A_j+1)^2}\left[u_j^2 + A_j^2\, U_j^2 + 2A_j u_j U_j \cos a_j\right]$$

$$v_j^2 = \frac{A_j^2}{(A_j+1)^2}\left[u_j^2 + U_j^2 - 2u_j U_j \cos a_j\right]$$

$$E_{j+1} = x_j^2 + v_j^2 + 2x_j v_j \cos b_j$$

$$\cos c_j = \frac{1}{x_j(A_j+1)}\left[u_j + A_j\, U_j \cos a_j\right]$$

$$\cos \Psi_j = \cos b_j \cos c_j + \sin b_j \sin c_j \cos \phi_j$$

$$\mu_j = \frac{1}{u_j+1}\left[v_j \cos \Psi_j + x_j \cos c_j\right]$$

The flight parameters are defined as follows:

$\vec{x}$ - velocity of the center gravity

$\vec{v}$ - velocity of the neutron in the barycentric system before collision

A - effective mass of the target

a - angle between $\vec{u}$ and $\vec{U}$

b - angle between $\vec{x}$ and $\vec{v}$

c - angle between $\vec{x}$ and $\vec{u}$

Ψ - angle between $\vec{v}$ and $\vec{u}$

μ - angle between $\vec{u}_j$ and $\vec{u}_{j+1}$

ϕ - azimuth of $\vec{v}$ around $\vec{x}$

E - energy of the neutron

The mass of the neutron is taken as unity and the units of velocity are such that $u = \sqrt{E}$. The index j is the number of the collision. The solid angle of scattering in the barycentric system (i.e., Φ and cos b) is selected at random.

Accumulation of M^2

The migration area, M^2, is defined as one-sixth the mean-square distance, $\overline{r^2}$, travelled by the neutrons in slowing down.

When the flight parameters have been obtained the contribution to the migration area is computed and the contents of the storage for $\overline{r^2}$ are brought up to date. A general expression for the second moment of the slowing-down distribution derived by Coveyou (1956) was adapted for the code. The mean-square distance travelled in the course of j collisions is

$$\overline{r^2} = 2 \sum_{\alpha=1}^{j} \sum_{\beta=1}^{\alpha} \lambda_\alpha \lambda_\beta \, (\mu_\beta \mu_{\beta+1} \cdots \mu_{\alpha+1})$$

The mean-free-paths, λ, are picked from a table where they are stored as functions of energy. The quantity μ_β is the computed cosine of the scattering angle at collision β.

The use of the general expression for $\overline{r^2}$ is a major factor in the speed and precision of the computation. This expression gives the proper average of all possible path lengths between successive collisions, and all possible azimuthal angles of scattering. Thus, every neutron history is a statistical representation of all neutrons that have experienced the same succession of energies of degradation. Only those spatial details of the scattering event necessary to obtain the cosine of the scattering angle, μ, were required in the computation.

The flight between collisions has now been completed and pertinent data are punched. The next event (scattering or absorption) is determined from the energy as before, and the process is repeated. A neutron history is terminated when the neutron is absorbed or when its energy falls below some cutoff level.

The speed of computing neutron histories depends upon the number of collisions required per history. A large fraction of the machine time is spent in the accumulation of the $\overline{r^2}$ function. The contributions of flights with large j involve many more arithmetic operations than those for small j. The computing rate R in terms of the average number of collisions per neutron, J, is given by the empirical formula

$$R(\text{neutrons/hr}) = \frac{3900}{J^2} + \frac{550}{J}$$

Useful results can be obtained in a double shift of IBM 650 operation. For example the migration area from fission to 1.46 ev in H_2O was computed from 750 neutron histories in sixteen hours with a standard deviation of 0.6%.

3. RESULTS

A. Migration Areas

The method is primarily one for computing migration areas in infinite homogeneous moderators. Diffusion constants and temperature coefficients were obtained by variation of input parameters in the migration area calculation. Components of the migration area above and below the indium resonance (1.46 ev) were obtained separately. The familiar discrepancy between computed and

measured values of the migration area for H_2O from fission to 1.46 ev remained; the computed value was 3 cm^2 below the measured value. For D_2O, however, the computed value was 8 cm^2 above the measured value. The results are listed in Table I.

TABLE I

CALCULATED MIGRATION AREAS

Fission to the First Scattering Below 1.46 ev

	H_2O	50% H_2O 50% D_2O	25% H_2O 75% D_2O	10% H_2O 90% D_2O	D_2O	HNO_3 3.5 N	HNO_3 7.9 N
M_f^2, cm^2	27.4	37.3	52.6	77.6	120.3	29.3	31.1

1.46 ev to Absorption

H_2O			D_2O		
kT	Σ_a	M_s^2	kT	Σ_a	M_s^2
0.0257 ev	0.13 cm^{-1}	2.44 cm^2	0.0257 ev	0.011 cm^{-1}	108 cm^2
	.17	2.01		.022	64.5
	.26	1.60		.033	49.1
	.52	1.12		.033(E*=0)	50.3
	.78	0.90		.260	14.3
0.0350	.13	2.72	0.0400	.011	129
	.26	1.74		.022	73.9

NOTES: The migration area from fission to 1.46 ev is calculated to be 26.7 cm^2 for H_2O and 119.5 cm^2 for D_2O.

Σ_a is defined as the maxwellian average of the 1/v cross section at 25°C.

The case with E*=0 shows the effect of neglecting chemical binding.

The energy range for slowing down is divided at 1.46 ev, the energy of the In-115 resonance, to permit the results to be compared with experiments. In the high energy range the source of neutrons was taken to be the fission energy distribution. The system was assumed to be nonabsorbing, and the moderator atoms were assumed to be unbound and at rest. When a neutron emerged from collision with energy less than 1.46 ev, its contribution to M^2 was computed and the history terminated. In the low energy range the source energy was assumed constant at 1.46 ev. The absorption rate was taken to behave as 1/v and was varied in strength to study the validity of the relation $M^2 = \frac{D}{\Sigma_a} + \tau$. The moderator atoms were in a maxwellian distribution and were assumed bound for collisions occurring below the threshold of vibration, E*. Neutron histories were terminated by absorption.

The fast and slow components so obtained cannot simply be added to yield the total migration area because slowing down in the energy "overlap" of the first collision below 1.46 ev is computed for both components. Migration areas through the energy overlap were obtained from short runs using a $\frac{1}{E}$ distribution above 1.46 ev as a source. Both M^2 for scatterings to energies below 1.46 ev and M^2 for scatterings to energies within the interval 1.53 to 1.39 ev were accumulated. The difference between these two concepts of M^2, 0.7 cm^2 for H_2O and 0.8 cm^2 for D_2O, must be subtracted from the migration area to below 1.46 ev in order to obtain the migration area to 1.46 ev.

Olcott (1956) measured 111 ± 1 cm^2 and Wade (1957) measured 109 ± 3 cm^2 for the migration area from fission to indium in D_2O contaminated with 0.2% H_2O. The value interpolated for this

D_2O purity from the Monte Carlo calculations is 118 cm^2. Barkov and Mukhin (1956) measured 29.4 ± 1.5 cm^2 and Hill, Roberts, and Fitch (1955) measured 30.8 cm^2 for H_2O. The calculation gives 26.7 cm^2. Hurwitz and Zweifel (1955) computed the migration area of H_2O to be 23.6 by a moments treatment of a similar model.

Including the effects of anisotropic scattering of oxygen in the calculation would bring the value of H_2O into better agreement with experiments but would worsen the agreement for D_2O. Goldstein and Certaine (1957) have found the effect of anisotropic scattering to be small. They also show that the low value computed for H_2O does not arise from possible errors in the fission spectrum.

Experiments that determine migration area by indium foil activations require correction for the width of the activation resonance and the thickness of the foil. The observed activation is the average value over the width of the resonance. Corrections to convert these results to the values that would be obtained with resonances of zero width were estimated. The migration area in H_2O, as computed to the wide energy band 1.10 to 1.94 ev which is embraced by the indium resonance, was 0.8 cm^2 larger than that to the smaller interval 1.39 to 1.53 ev. For D_2O the migration area to the wide band was found to be 2.8 cm^2 greater than to the narrow band. Presumably, the experimental data in the literature were not corrected for this effect.

B. Diffusion Constants

The migration area of neutrons through the thermal range of energies may be related to the absorption rate by the expression $M^2 = \frac{D}{\Sigma_a} + \tau_s$ where D, the diffusion constant, depends upon the temperature and scattering properties of the moderator, Σ_a is the

effective absorption cross section, and τ_s is the neutron age from 1.46 ev to thermal energy. Migration areas were computed for different absorption strengths to check the validity of the linear formula and to evaluate the magnitudes of τ_s and D. An absorption cross section varying as 1/v was assumed throughout.

For high values of Σ_a the neutron path lengths must approach zero and a linear relation cannot be valid. However, a linear relation does hold over a large range of $L^2 = \frac{D}{\Sigma_a}$ for both H_2O and D_2O as shown in Figures 1 and 2. The linear portions extrapolate to the component of M^2 that does not vary with changes in Σ_a. This component is interpreted as the epithermal age; $\tau_s = 0.77$ cm^2 for H_2O and $\tau_s = 20$ cm^2 for D_2O. The energy at which neutrons may be considered thermal and to which τ_s should be computed has been debated by Cohen (1955). Our value of τ_s for D_2O agrees with the usual 20 cm^2 derived from the assumption that the neutron becomes thermal at kT rather than the 6 cm^2 resulting from the assumption that the neutron becomes thermal at about 15 kT. Barkov, Makarin, and Mukhin (1956) have measured τ_s to be 1.0 ± 0.5 for H_2O.

In order to obtain D from L^2, we arbitrarily define Σ_a to be the average cross section of the 1/v absorber in a maxwellian spectrum with temperature corresponding to kT = 0.025 ev, or $\sqrt{\pi}/2$ times the 2200 m/sec cross section. The diffusion constants are then found to be 0.19 cm for H_2O and 0.87 cm for D_2O. From values of the transport mean-free-path given in the Reactor Handbook (1955), one finds $D = \frac{\lambda tr}{3} = 0.16$ cm for H_2O and 0.88 cm for D_2O. The diffusion constant increases with higher moderator temperatures at constant moderator density through the influence of the increased

agitation of moderator atoms. The effect is reflected in the higher slopes of the curves in Figures 1 and 2. The coefficient for this effect alone was found to be 0.15%/°C for H_2O and 0.14%/°C for D_2O. In addition, the usual effects of density changes and spectral hardening must be included in the over-all temperature coefficient of M^2.

C. Neutron Energy Distributions

Neutron energy spectra are obtained from the Monte Carlo code by merely accumulating the number of scatterings that occur within each energy increment. An example is graphed in Figure 3. The distribution obtained by Monte Carlo coincides with that obtained from an analytical solution (Brown 1956). The comparison with the spectrum computed by a formula of Cohen (1955) implies that the disagreement about the epithermal age is in the definition of τ_s and does not extend to the spectrum.

Although the direct calculation of simple spectra is faster than the Monte Carlo calculation, it is still practical to obtain spectra from the code when it is also used for other purposes. The accumulation of a spectrum is incidental to the computation of M^2 and involves no additional computing time. For instance, at the same time that temperature coefficients of M^2 are computed, the temperature coefficients of the spectra can be obtained. In some cases parametric changes are more easily encompassed by the Monte Carlo method. Unusual situations, such as the effect of thermal resonances upon the spectrum, can be surveyed without additional coding.

4. CONCLUSION

The code is fast and accurate enough to find frequent application at the Savannah River Laboratory. Migration areas calculated by the Monte Carlo method are based on the same physical model that is used for the analytical methods (Hurwitz and Zweifel, 1955). The results are thus subject to similar errors such as scattering isotropy and fission source error, but useful information is obtained by normalizing to the available experimental points. The code is well suited to the study of the temperature variations, the effects of mixtures, arbitrary absorption and scattering functions, and arbitrary source distributions. The treatment could be extended to finite moderators by the usual artifice of adding the leakage to the absorption rate: $\Sigma_a^{eff} = \Sigma_a + DB^2$ where B^2 is the buckling.

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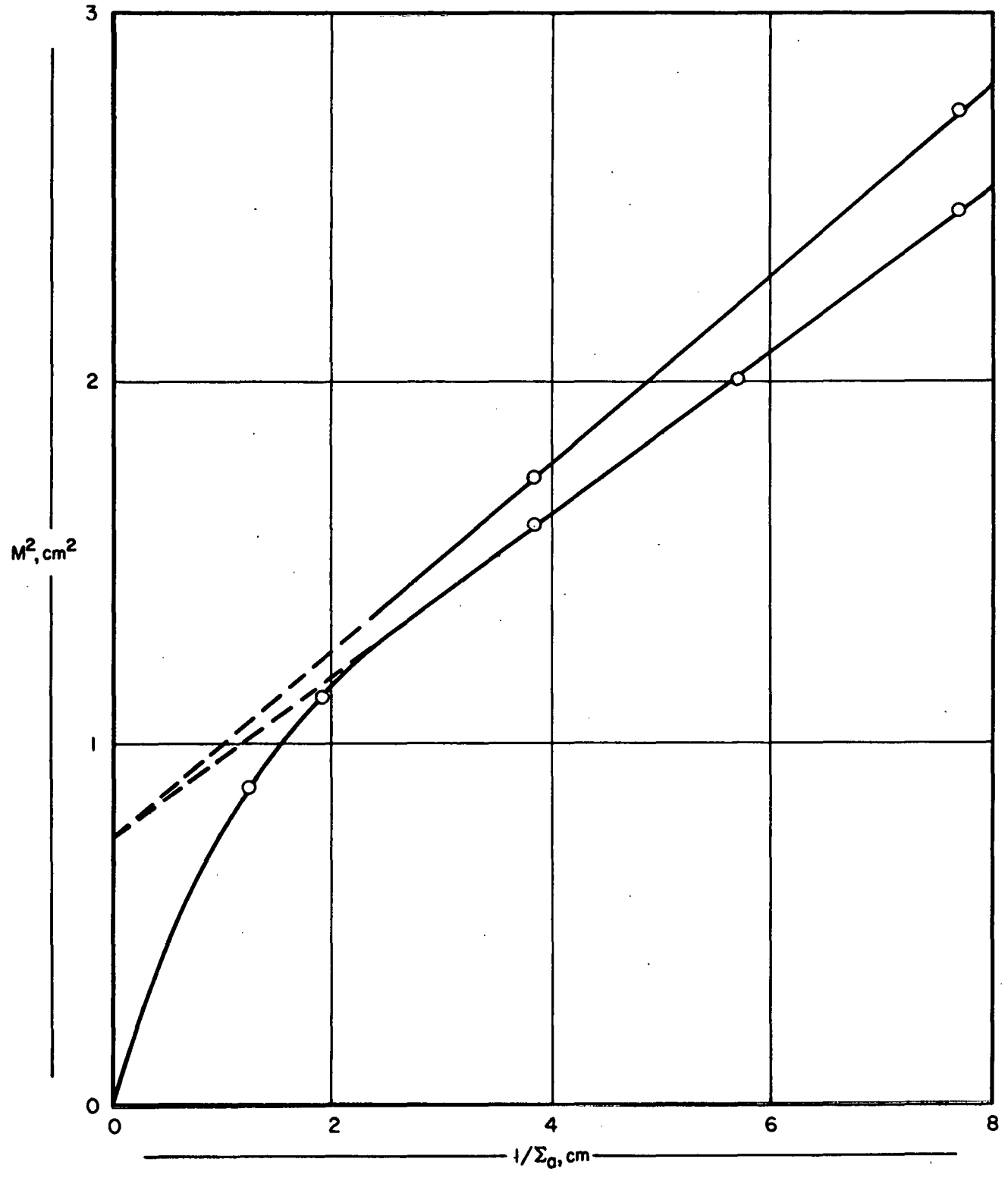
Note to Figures 1 and 2

The plotted migration areas were obtained by Monte Carlo calculations. The straight lines are least-square fits to the linear portion of the data and show the range of validity of the relation $M^2 = \frac{D}{\Sigma_a} + \tau$. If Σ_a is defined as the average of the $\frac{1}{v}$ cross section in a maxwellian flux at 25°C, D is found to be 0.19 cm for H_2O and 0.87 cm for D_2O. The values of kT refer to moderator temperatures.

Note to Figure 3

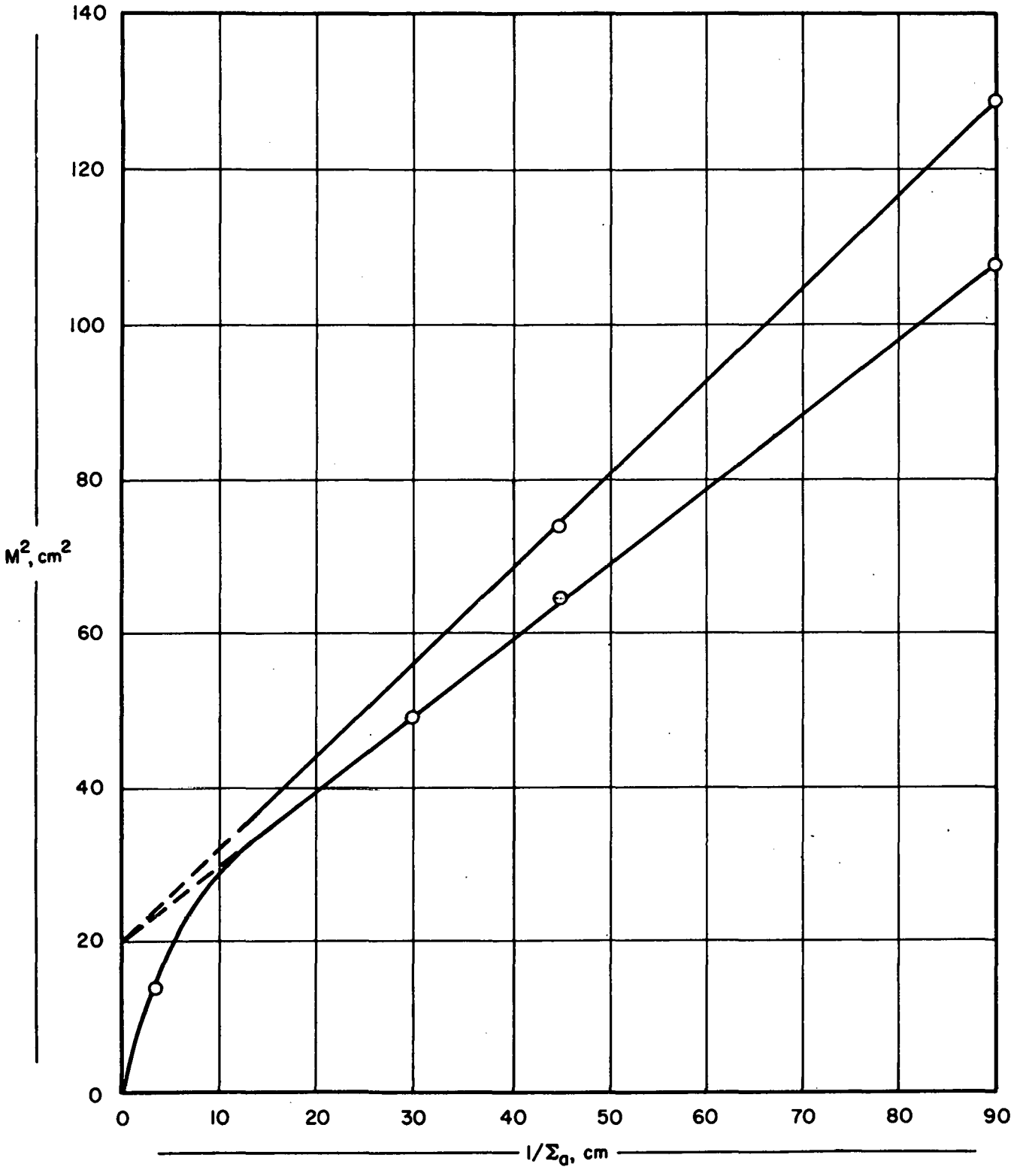
The distribution derived by Monte Carlo coincides with the analytic solution. The dashed curve is obtained from a formula of Cohen (1955). $\Sigma_a = 0.011\ cm^{-1}$ $kT = 0.0257$ ev

FIGURE 1



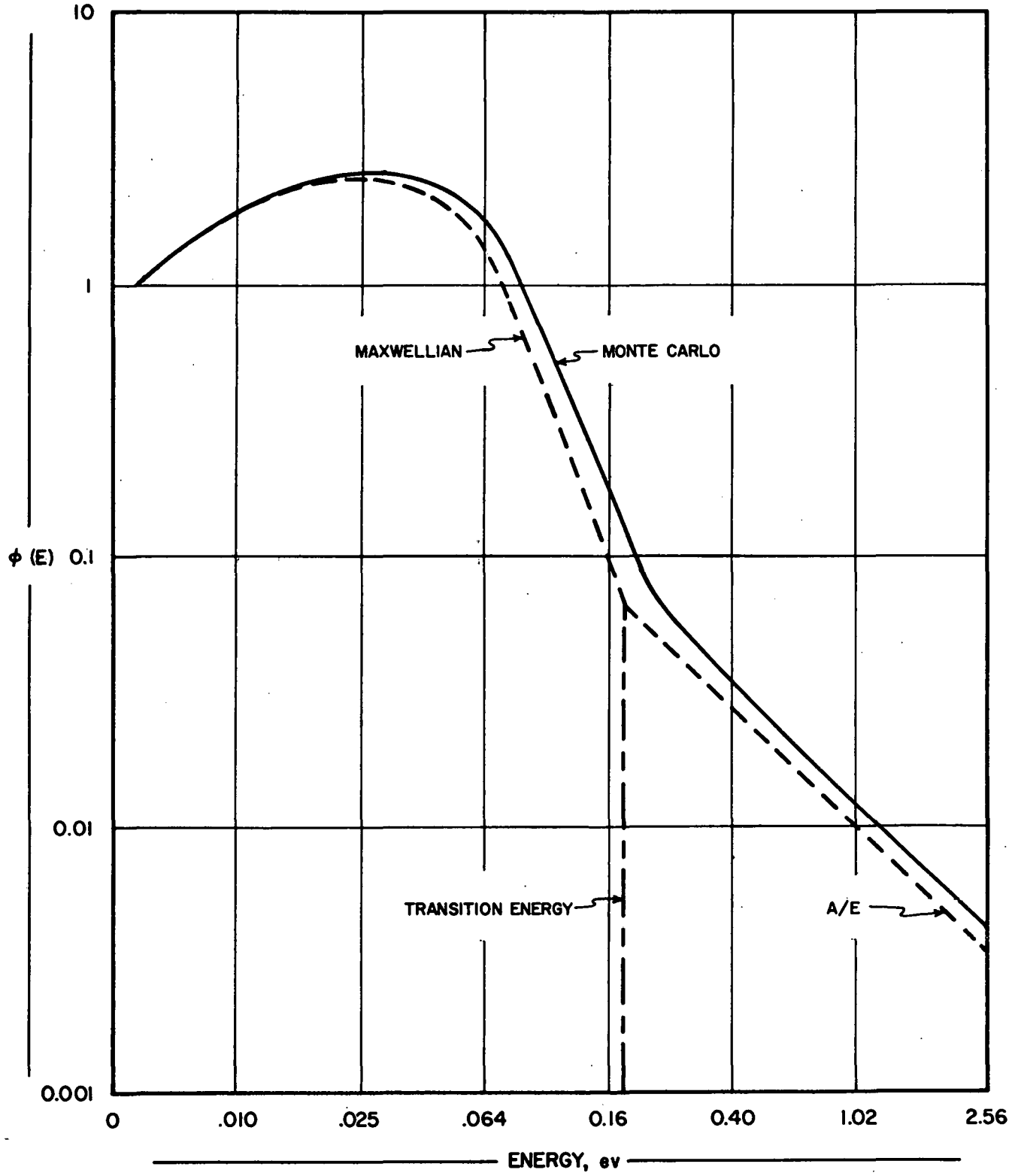
MIGRATION AREAS IN H_2O FROM 1.46 ev TO ABSORPTION

FIGURE 2



MIGRATION AREAS IN D_2O FROM 1.46 ev TO ABSORPTION

FIGURE 3



NEUTRON ENERGY DISTRIBUTION IN D_2O