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QUARTERLY PROGRESS REPORT
JANUARY – MARCH, 1957

ATOMICS INTERNATIONAL

A DIVISION OF NORTH AMERICAN AVIATION, INC.

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ABSTRACT

NUCLEAR MEASUREMENTS

A series of exponential experiments with one-inch diameter uranium rods in diphenyl has been started. Initial measurements have been made to determine the height of thermal column and amount of external shielding needed. Fuel enrichments of 0.5, 0.7, and 0.9 per cent U^{235} will be used with one-inch diameter rods at lattice spacings varying from 1.5 to 2.5 inches. New techniques for making flux distribution measurements are being developed in order to overcome the problems which have arisen because of the use of diphenyl as a moderating material for the experiments.

By application of the age-diffusion theory approximation, the non-leakage probability of neutrons from a spherical medium containing a centrally-located fast source is shown, for large sphere radius, to be proportional to exp $(-\pi^2M^2/3R^2)$, where R is the radius of the sphere and M^2 the migration area of the neutrons in the medium. This functional dependence was verified by experimental measurement of the non-leakage probabilities of neutrons from fast sources contained in spheres of water, titanium hydride, and calcium hydride. For a mock-fission source, analysis of the data leads to values of 43 ± 2, 24 ± 2, and 100 ± 10 cm^2 for the migration areas in these materials, respectively.

NEUTRON THERMALIZATION

An effort was made to relax two approximations upon which the Wilkins equation rests. The one condition, i.e., no chemical binding, has been relaxed in an approximate way involving the use of a judiciously chosen three-parameter approximation for the center-of-mass scattering cross section, which was introduced by Brown and St. John. A differential equation analogous to the Wilkins equation but involving this approximation was obtained and is now being coded for IBM 704 solution.

An attempt was made to relax the second condition, i.e., heavy moderator mass, and it became clear that more refined approximations to the Wigner-Wilkins kernel would not lead to further simplification.

I. NUCLEAR MEASUREMENTS

A. ORGANIC MODERATED LATTICES (W. W. Brown, M. J. Glaubman)

The development of reactors using organic moderators has created a need to extend out knowledge of the basic properties of lattices to include systems of this type. As a first step, a series of exponential experiments have been planned on lattices of one-inch-diameter uranium rods in diphenyl. Fuel loadings of three different enrichments, 0.5, 0.7, and 0.9 per cent U^{235}, will be used and lattices at three or four different spacings will be investigated. The fuel and aluminum cladding is identical with that used in previous lattice studies made in heavy water and graphite moderators. The results of all three studies may thus be directly correlated from the standpoint of determining whether the calculational procedures that worked well for the interpretation of the heavy water data can be more generally applied or not. The work will also provide additional information for the study of the empirical relation between the measured excess absorption of a lattice and that calculated on the basis of diffusion theory.

A 2-1/4-foot-diameter aluminum tank to contain the lattices has been mounted on top of the thermal column of a water boiler reactor. The lid and sides of the tank are cadmium clad to restrict entry of thermal neutrons into the system except through the tank bottom. The last 17 inches of the thermal column have been reduced from 5 feet to 2-1/4 feet in diameter to match the geometry of the tank. External heaters maintain the lattice temperature at 200° F -- just above the diphenyl melting point.

Certain nuclear parameters of each lattice set-up can be calculated from measured thermal neutron distributions along the vertical axis of the lattice and along the horizontal radius of an individual cell. The horizontal distribution of thermal neutrons in the moderator, without fuel present, is also required so that the extrapolated radius of the system can be determined directly rather than be obtained by calculation from the geometry of the system. In a tank with a small radius, the error in a calculated value would contribute considerably to the error in the lattice buckling.

Measurements by foil activation have already been started in one of the lattices, viz., 0.9 per cent enriched uranium rods in a square array with a two-inch spacing between centers. Vertical and horizontal distributions have also been obtained in the moderator without the fuel present. At first, these measurements were made with the system situated on a thermal column with a total height of 36 inches. The horizontal distribution of neutrons in the pure diphenyl measured under these conditions resulted in an excessively large extrapolated radius. Measurement of the indium resonance neutron distribution showed that this was because of epithermal neutrons entering through the side of the tank. To correct for this, the contribution to the thermal neutrons in the tank resulting from the leaking in of epithermals was measured by placing a 1/16-inch-thick cadmium sheet under the tank, so that only epithermal neutrons could enter, and then measuring the thermal neutron distribution. The resulting thermal neutron flux contribution was about 20 or 30 per cent of the total observed without the cadmium sheet. This is considered too large a correction to make and therefore it was decided first to reduce it by increasing the height of the thermal column by 17 inches. The increase will not only reduce the epithermals entering through the tank bottom, but will permit reduction of their entry through the sides of the tank by the stacking of more paraffin over open regions across the top of the reactor.

The use of diphenyl as a moderating material has created new problems in the exposure and handling of foils in the exponential assembly. New types of foil holders are under development in order to solve these problems.

B. ABSORPTION OF NEUTRONS IN HYDROGENOUS SPHERES (W. W. Brown)

In general it has been found useful to apply the concept of Fermi age to hydrogenous media. Measurement of either neutron age or migration area for various fast neutron sources in such media usually involves the use of sufficiently large quantities of material so that corrections for leakage can be neglected. In the following analysis, a method is suggested for measuring migration area in small systems -- a method that can be applied when only small amounts of material are available.

In a spherical medium of extrapolated radius R containing a centrally-located fast-neutron point source, the thermal flux obtained from the age-diffusion theory equations, at any distance r from the source, is proportional to

$$\sum_{n=1}^{n=\infty} \frac{n}{R^2 + N^2\pi^2L^2} \exp\left\{-\frac{n^2\pi^2}{R^2}t\right\} \sin\frac{n\pi r}{R} \quad ,$$

where L is the diffusion length and t the age to thermal energy. The integral of this over the volume of the sphere is

$$\sum_{n=1}^{n=\infty} \frac{R^2\, e^{-\frac{(2n-1)^2\pi^2 t}{R^2}}}{R^2 + (2n-1)^2L^2\pi^2} - \frac{R^2 e^{-\frac{(2n)^2\pi^2 t}{R^2}}}{R^2 + (2n)^2L^2\pi^2}$$

which is proportional to the non-leakage probability. For large R, the sum may be replaced by an integral by making the following substitutions:

$$w = \frac{(2n-1)\pi}{R} \qquad \frac{R}{2\pi}\, dw = dn = 1 \quad .$$

This leads to an integral, I, given by

$$I = \frac{R}{2\pi} \int_o^\infty \left[\frac{e^{-w^2t}}{1 + w^2L^2} - \frac{e^{-\left(w + \frac{\pi}{R}\right)^2 t}}{1 + \left(w + \frac{\pi}{R}\right)^2 L^2} \right] dw$$

$$= \frac{R}{2\pi} \int_o^\infty \frac{e^{-w^2t}}{1 + w^2L^2}\, dw - \frac{R}{2\pi} \int_{\pi/R}^\infty \frac{e^{-f^2t}}{1 + f^2L^2}\, df$$

(where f = w + π/R)

$$= \frac{R}{2\pi} \int_{o}^{\pi/R} \frac{e^{-f^{2}t}}{1 + f^{2}L^{2}} \, df$$

For large R, f is small and the integrand may be approximated by

$$1 - f^{2}M^{2}, \quad \text{where } M^{2} = t + L^{2} \quad .$$

The integral then becomes

$$I = \pi \left(1 - \frac{\pi^{2}M^{2}}{3R^{2}} \right)$$

$$\approx exp \left\{ - \frac{\pi^{2}M^{2}}{3R^{2}} \right\}$$

If the non-leakage were directly measured for several values of R, then, from a semilogarithmic plot of the non-leakage probability as a function of R^{-2}, the migration area M^{2} can be calculated.

The thermal neutron distributions have been measured, by foil-activation techniques, in spheres (or assemblies of pressed one-inch cubes approximating spheres) of light water, titanium hydride, and calcium hydride. In the water both mock fission and polonium-beryllium neutron sources were used at the sphere center. In the other materials, only the mock fission source was used. The spheres ranged in size from 8 to 15 inches in diameter. Graphical integration of the measured thermal fluxes over the sphere volumes gave, for a given material, numbers proportional to the non-leakage probability in each case. In particular, for the light-water case, measurements were also made in an effectively infinite volume. The ratio of the flux integrations in the spheres to that for the infinite case gives the absolute value of the non-leakage probability for the water spheres, and should provide a check on the linearity, for large R, of a semilogarithmic plot of the non-leakage against R^{-2}. Linear extrapolation to large R of the line connecting the measured points in the region of finite R should intercept the non-leakage axis at 1.0 if the above age-diffusion theory analysis holds. The results, given in Fig. 1, show this indeed to be the case. There was not enough material to make infinite medium measurements in the calcium and titanium hydrides. However, the values of the infinte integrals for these materials were obtained from that of water by multiplying by the respective ratios of the thermal macroscopic absorption cross sections of these materials to that of water. As shown in the figure, the non-leakage probabilities so calculated for these materials also extrapolate to large R in a manner consistent with the analysis. The radii used in plotting these data are the radii of the physical systems -- not the extrapolated radii. The integrations were carried only to the physical boundary.

If it is assumed that the analysis applies to the above experimental situation, an effective migration area can be calculated from the slopes of the curves for each of the materials in which measurements were made. The results are collected in Table I. The results in light water are in agreement with measurements made in extensive media at other laboratories[1, 2] with similar, although not identical, sources.

TABLE I

EFFECTIVE MIGRATION AREAS

Material	Density ($gm\text{-}cm^{-3}$)	Neutron Source	Migration Area, cm^2	
			From Measurements in Large Systems[1, 2]	Present Measurements in Small Systems
H_2O	1.00	Fission	40	
		Mock fission		43 ± 2*
		Po-Be	66	71 ± 4
$TiH_{1.74}$	3.21	Mock fission		24 ± 2
$CaH_{1.56}$	1.41	Mock fission		100 ± 10

*Errors are estimates based on statistical fluctuation only

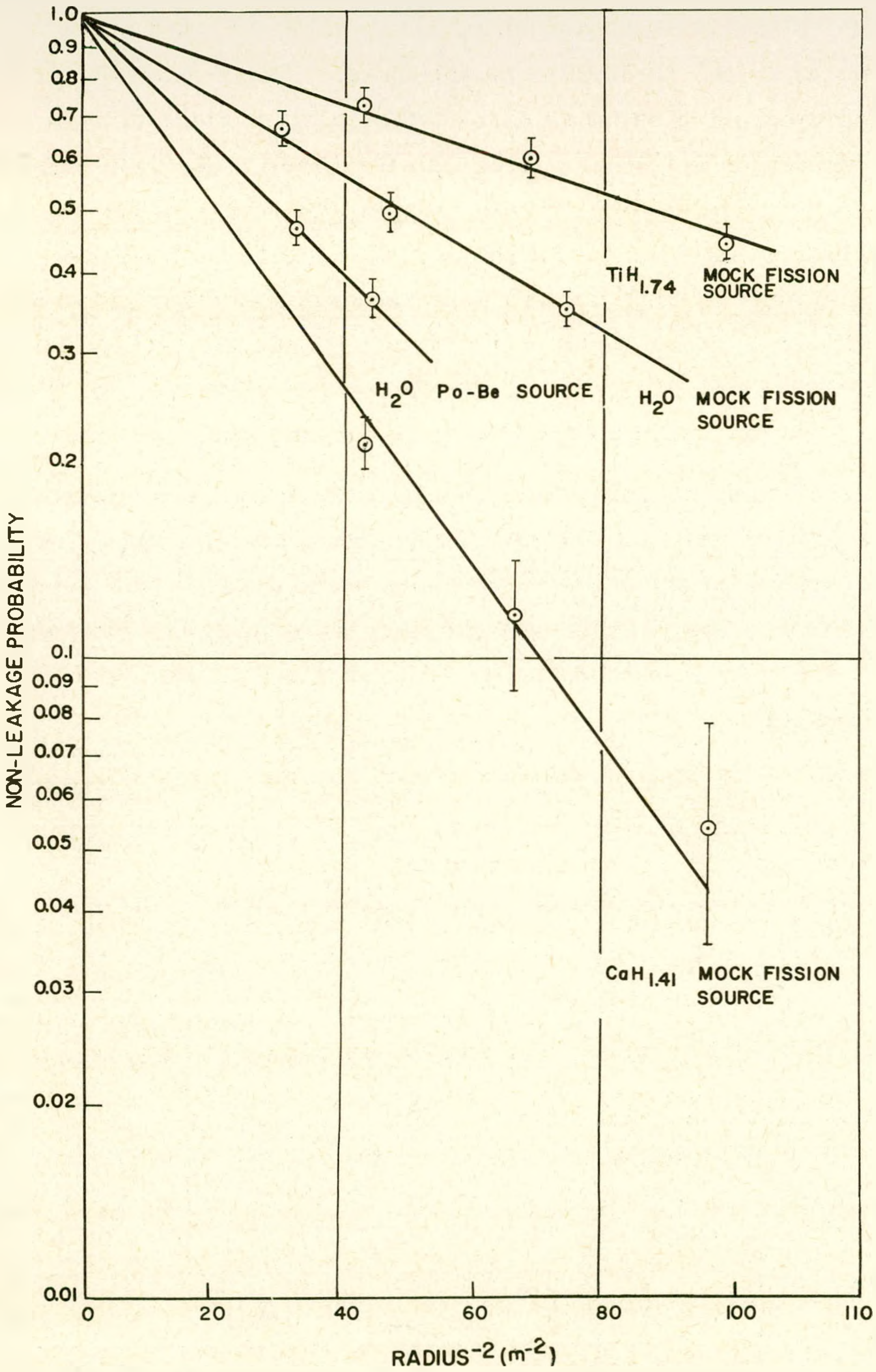

Fig. 1. Non-Leakage Probabilities of Neutrons From Fast Sources at the Centers of Hydrogenous Spheres

II. NEUTRON THERMALIZATION

A. THERMALIZATION IN CHEMICALLY BOUND MODERATORS OF INTERMEDIATE MASS (M. N. Moore)

The Wilkins equation for the neutron energy spectrum in equilibrium with a Maxwellian gas entails, among others, the following two approximations on the exact (Boltzmann) equation:

1) The mass of the gas particle is large with respect to that of the neutron.

2) The scattering cross section in the center of mass system is independent of the relative velocity.

In connection with each of these approximations, there arises two questions of interest:

1) In view of the Wigner-Wilkins solution for thermalization in hydrogen on the one hand and the Wilkins solution suitable for heavy masses on the other, what is the neutron spectrum in equilibrium with moderators of intermediate mass?

2) What is the neutron spectrum in equilibrium with a chemically-bound system for which the scattering cross section in the center-of-mass system is not independent of relative velocity?

To answer the first question, an attempt was made to simplify the kernel of the Boltzmann equation by the use of approximations to the kernel. However, it was found that since the kernel is a small linear combination of several large quantities, an approximation able to give the moments up to second order was more complicated than these moments themselves. Furthermore, there is no reason to expect this situation to improve at higher moments.

A complete answer to the second question is extremely complicated. H. Hurwitz[3] has shown, from conservation of neutrons and the principle of detailed balancing, that in general, $\Psi(E)$, the deviation from a Maxwellian flux $\phi_o(E) = Ee^{-E/RT}$, obeys the following integral equation:

$$[\sigma_s(E) + \sigma_a(E)]\,\Psi(E) = \int \sigma_s(E \rightarrow E')\,\Psi(E')\,\phi E \qquad \ldots(1)$$

where

E, E' = neutron energies in the laboratory system,

$\sigma_s(E)$ = the neutron scattering cross section of the moderator,

$\sigma_a(E)$ = the neutron absorption cross section of the moderator,

$\sigma_s(E \rightarrow E')$ = the energy transfer cross section of the moderator, for neutrons initially of energy E and after collision of energy E',

$\phi(E)$ = the neutron flux,

and where

$$\phi(E) = \phi_o(E)\Psi(E) \quad .$$

When Eq. (1) is used to describe the neutron spectrum in a chemically-bound moderator, the effects of chemical binding are, of course, contained in the energy transfer cross section. In calculating this cross section, such difficulties as anisotropic scattering in the center-of-mass system, distribution of excitation energy among various types of excited states, correlations between these states, and interference between the scattering center and its bound neighbors are typical complications. In an effort to obtain some reasonable results for the spectrum in chemically-bound systems, notably liquid D_2O and H_2O, Brown and St. John[4] developed a formalism whereby the above complications could be avoided by the use of the empirical $\sigma_s(E)$ in connection with Eq. (1). This formalism involved approximation of the scattering cross section, as a function of the relative velocity v_r, by a three-parameter function of the form

$$\sigma_s(v_r) = C + Be^{-kv_r^2} \quad . \qquad \text{...(2)}$$

The constants B, C, and K are determined from the empirical curve of $\sigma_s(E)$. The approximation was found to be quite satisfactory in fitting the experimental $\sigma_s(E)$ for both liquid D_2O and H_2O and hence presumably for more complicated systems. The energy transfer cross section was inferred from the total scattering cross section by replacing the moderator by a monatomic gas with a suitably chosen "effective" mass. This mass was taken to be the reciprocal of the trace of the "effective reciprocal mass tensor" used by Sachs and Teller[5] to represent the dynamical properties of rigid-rotator molecules. Using the resulting energy-transfer cross section as the kernel, Brown and St. John could then solve Eq. (1) numerically.

An alternative solution to (1) may be found if one exploits the insensitivity of $\psi(E)$ to changes in E at low energies. Following the method of Hurwitz, $\Psi(E')$ is expanded in a Taylor's series about E, so that (1) then takes the form

$$\sigma_a(E)\Psi(E) = \sum_{k=1}^{\infty} \frac{1}{k!} M_k(E)\,\Psi^{(k)}(E) \quad , \qquad \text{...(3)}$$

where

$$M_k(E) = \int_0^{\infty} \sigma_s(E \to E')(E' - E)^k \phi E'$$

and

$$\Psi^{(k)}(E) = \frac{d^k\Psi}{dE^k} \quad .$$

At low energies ($E << kT$), σ (E) varies slowly with E and hence terms having $k > 2$ are neglected. For higher energies, if the mass of the moderator is large, Hurwitz has shown that the M_k (E) are so small for $k > 2$ that the M_k (E) $\Psi^{(k)}$(E) in this region are negligible compared with the M_k (E) $\Psi^{(k)}$(E) for $k \leq 2$. This condition holds in the case of $\sigma_s(v_r)$ constant and $E << \frac{MkT}{4}$.

Just how stringent this heavy mass requirement is, from a practical standpoint, insofar as the neutron spectrum is concerned, is not clearly understood, nor is it clear what the appropriate energy range of validity of such an approximation by a second-order differential equation, for $\sigma_s(v_r)$ not constant, should be.

It has been felt worthwhile, therefore, to develop an IBM 704 code to solve (3) for $k \leq 2$, where the M_k(E) are computed from (2) using the observed σ_s(E) to determine the constants B, C, and K. Such a code has a two-fold value:

1) Using the Brown-St. John numbers, a comparison of spectra calculated by the code with those calculated by Brown and St. John from the exact expression (1) would yield information concerning the stringency of the heavy mass requirement, and,

2) the solutions calculated by the code, which are valid at least for heavy masses, could then be used to calculate thermal group constants for chemically-bound, non-hydrogenous moderators.

The analytical problem has been solved and coding has been initiated.

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