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REVIEW OF EXPERIMENTAL AND THEORETICAL STUDIES CONCERNING THE AGE OF FISSION NEUTRONS IN HEAVY WATER

M. Tobias

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M. Tobias

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ABSTRACT

A survey of the available literature reveals that four separate measurements of the age of fission neutrons to indium resonance in heavy water have been made between 1944 and 1956. The results range between 104 and 111 cm^2 and are in close agreement considering the experimental errors involved. The most recent measurements (by J. W. Wade) appear to be the most elaborate and reliable yet made and are therefore recommended for use. Wade reports a value of 109 ± 3 cm^2 for the age in 99.8% D_2O.

Theoretical evaluations of the age show much closer agreement with the experimental results than is the case for light water. Although experimental measurements have shown that a Gaussian slowing down model is not applicable to heavy water, it is found that numerical integration of $D/\xi \Sigma_s$ over the lethargy range from fission energy to indium resonance is in good agreement with experiment. Monte-Carlo calculations give values of the order of 110 cm^2 but are not yet fully corrected for the effect of anisotropic scattering.

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INTRODUCTION

The utility of the value of neutron age in reactor calculations is well known and will not be discussed here. The scope of this report is restricted primarily to values which have been obtained for fission neutrons in heavy water slowed down to indium-resonance energy (1.44 e.v.); the "age-to-thermal" is not discussed here in any detail as such a quantity is as yet not clearly defined and is subject to dispute.[2]

The definition itself of age is handled with some ambiguity in published literature. For present purposes, it is defined in terms of the second moment of the slowing down distribution from a source of fission neutrons to the point of detection. In the case of an isotropic point source, the age is taken as $\frac{\overline{r^2}}{6}$, and for a uniform plane source, $\frac{\overline{z^2}}{2}$. These are the principal definitions employed in experimental work.[6,9,11,14] However, it is sometimes considered to be the appropriate value to be employed in the fast nonleakage probability $\left(\text{expressed in the Gaussian form as, } e^{-B^2\tau}\text{, or in the group-diffusion form, as } (1 + \tau B^2)^{-1}\right)$.[7,8] It is clear, however, that the second moment definition of age given above is unequivocal and is the meaning chosen for the purposes of this report.

EXPERIMENTAL MEASUREMENTS

The earliest experimental measurement of the age of fission neutrons in heavy water appears to be one made by Fermi and his co-workers[11] in March 1944 using the Argonne CP-3 lattice. As a source of fission neutrons, 33 g of enriched uranium was used. Indium foils, 4 cm x 6.5 cm x 93 mg/cm^2, were placed in cadmium cylinders having walls two mm thick. The activation of each foil was measured with three Geiger counters. The foils were irradiated with and without the fission source. The results obtained with the latter procedure gave the background activity (which was not due to the slowed down fission neutrons).

Figure 1 gives the curve of activity vs the square of the distance from the source for the data of this experiment plotted on semi-log paper. Such a plot would be a straight line if the neutron slowing down process were continuous as supposed in the Fermi-age model since this theory predicts a distribution of the form $e^{-r^2/4\tau}$.

Nordheim and Cashwell[3] fitted the data with a function of the form

$$\text{Activity} = \frac{(\text{Const.})\, e^{\tau_2/\tau_1}}{8\,\tau_1\,\pi\, r} \left\{ e^{-\frac{r}{\sqrt{\tau_1}}} \left[1 + \operatorname{erf}\left(\frac{r}{2\sqrt{\tau_2}} - \sqrt{\frac{\tau_2}{\tau_1}} \right) \right] - e^{\frac{r}{\sqrt{\tau_1}}} \left[1 - \operatorname{erf}\left(\frac{r}{2\sqrt{\tau_2}} + \sqrt{\frac{\tau_2}{\tau_1}} \right) \right] \right\} \qquad (1)$$

where r is the distance of the detector from the source.

In the mathematical model used to obtain this function it is supposed that neutrons "diffuse" from a point source with a reciprocal "diffusion length"

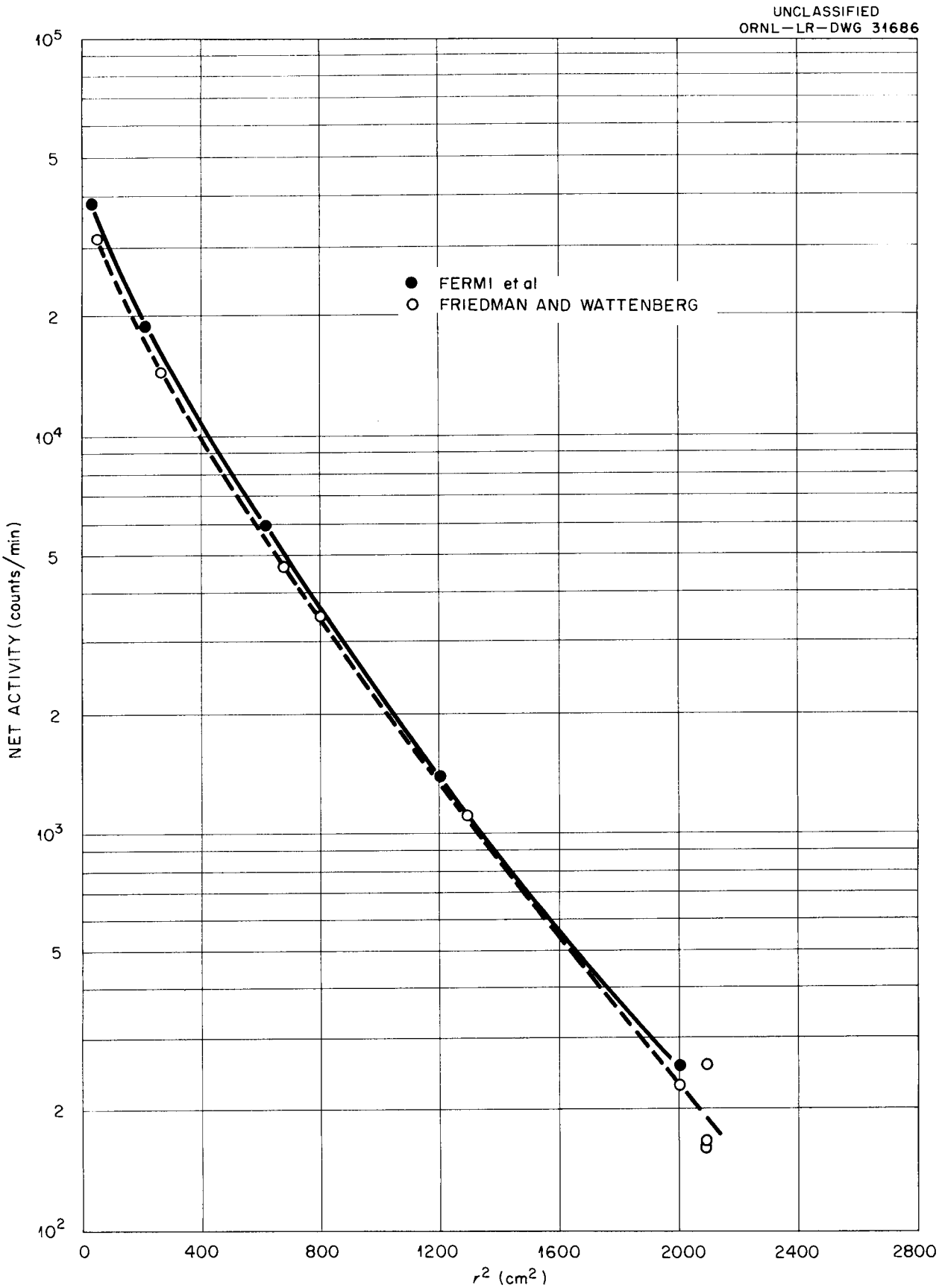

Fig. 1. Solid Curve: Indium Resonance Activity vs r^2 in 99.85% D_2O as Determined by Fermi et al., Ref. 11. Dashed Curve: Same for 99.8% D_2O as Determined by Friedman and Wattenberg, Ref. 6.

$\frac{1}{\sqrt{\tau_1}}$ to a point r_1; from r_1 they are slowed to a point r_2 with a Gaussian age distribution.* It can be shown that the total age is simply expressed as follows:

$$\tau = \frac{\overline{r^2}}{6} = \tau_1 + \tau_2 \qquad (2)$$

Using this analysis the total age, τ, was found to be 104 cm^2 for D_2O of 99.85% isotopic purity. Figure 2 compares the experimental results with equation (1) as well as with two-group, three-group, and age correlations.[3]

A similar experiment, performed in May 1945 and reported by Friedman and Wattenberg,[6] yielded an age to indium resonance of 106 cm^2 in 99.8% D_2O** when the data were correlated with equation (1) above. The best values of τ_1 and τ_2 were found to be 58 cm^2 and 48 cm^2, respectively; however, these numbers are not as accurately defined as the sum, τ. These measurements were conducted in 2700 liters of D_2O contained in a cylindrical tank atop the thermal column of the CP-2. An aluminum capsule suspended 25 cm above the bottom of the tank and containing 15.018 g of U_3O_8 enriched to 10% of U^{235} was employed as the source of fission neutrons. Indium foils were mounted in cadmium cylinders spaced at 10 cm intervals along a piece of 1/4-in. aluminum tubing. The cadmium was 2.5 mm thick, while the indium foils were 4 cm x 6.5 cm x 93 mg/cm^2. Each foil was measured on three inter-calibrated Geiger counters. Corrections were made for background activity

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* The same expression results if the two assumed slowing down processes are reversed.

** The value of 100 ± 5 cm^2 cited in BNL-170 and in the Reactor Handbook, Vol. 1, p 485 (AECD-3465, Feb. 1955) appears to be based in part upon preliminary reports published prior to CP-3406 and CP-3453 which cite some estimates for τ less than 100 cm^2 to In resonance. There appears to be no <u>final</u> report of experimental or theoretical work which would justify a value as low as this.

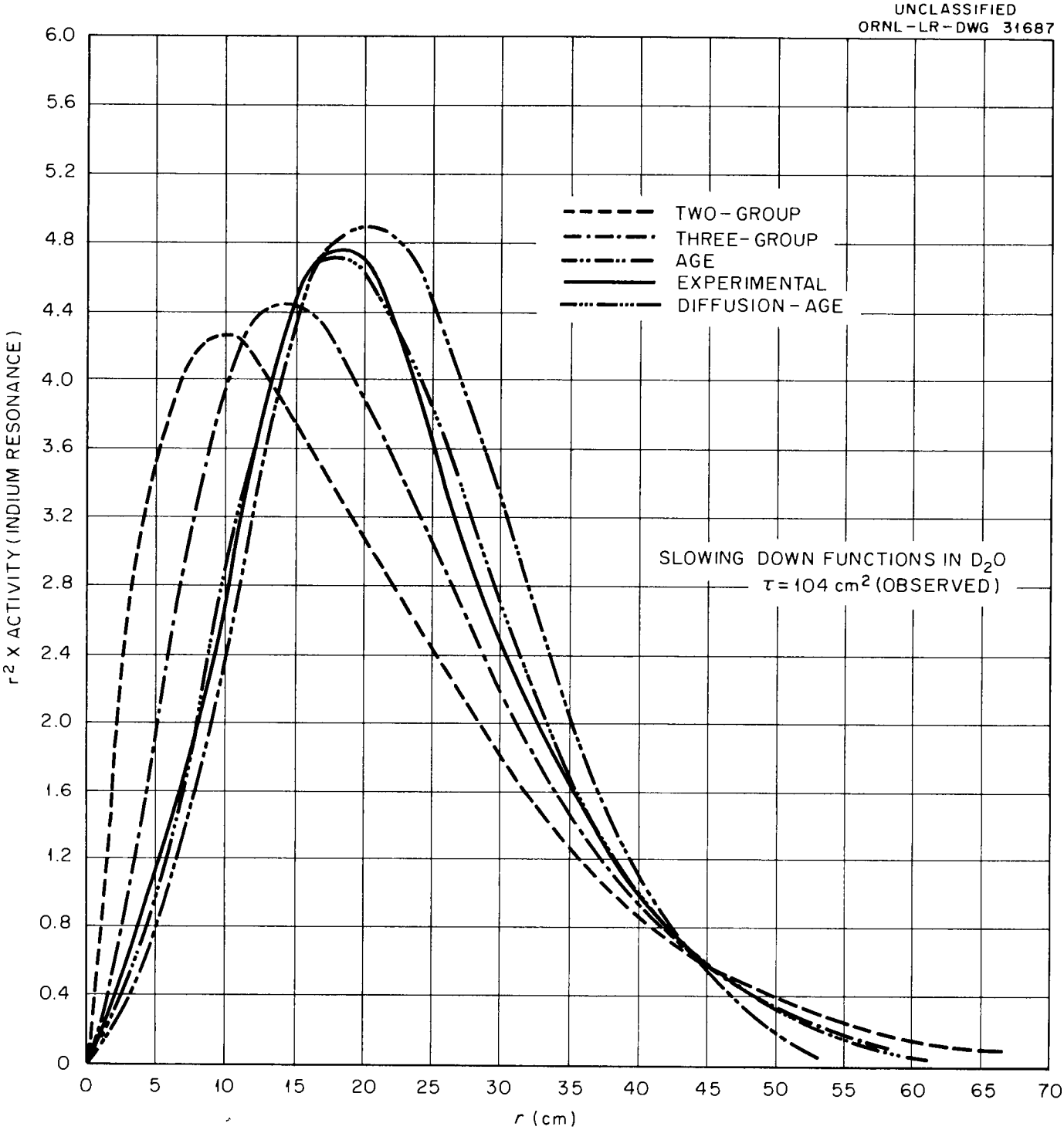

Fig. 2. Comparison of Results of Ref 11, Fermi et al, Plotted as r^2 X Indium Resonance Activity vs. r with Various Slowing Down Models, Ref 3.

and for the effective position of the foils. (The latter correction was associated with the nonlinear flux distribution along the 6.5 cm long foils.) Figure 1 shows the indium resonance neutron distribution obtained; the distribution and curvature are quite similar to those reported in CP-3406, confirming the conclusion that the slowing down kernel is not a simple Gaussian.

Olcott[9] has obtained a value of 111 $\pm$ 1 cm^2 for the age in 99.8% heavy water. In his work, an enriched uranium sphere was placed at the center of a 35-in. diameter sphere of heavy water. In contrast to the previously mentioned measurements, the plot of logarithmic activity vs r^2 is quite linear (Fig. 3) enabling correlation by $e^{-r^2/4\tau}$. Since the experimental procedure for the age measurement is not given in detail, it is not possible to judge whether the claimed accuracy is justified. The graph given by Olcott (of which Fig. 3 is an approximate copy) indicates that a much lower counting rate appears to have been employed than in any of the other measurements cited here.

The most recent measurements (1956) using the technique of activity measurement at various distances from a fission neutron source are those of Wade.[14] These experiments are reported in considerably greater detail than any of the previous measurements; in addition, the technique employed appears superior. For example, the instrumentation used is obviously more advanced than that available to the Chicago groups. Further, higher counting rates and smaller foils were used resulting in improved statistics and smaller geometry corrections. Minor corrections for activity induced by high energy neutrons were also made which appear to have been neglected by other workers. Wade obtains 109 $\pm$ 3 cm^2

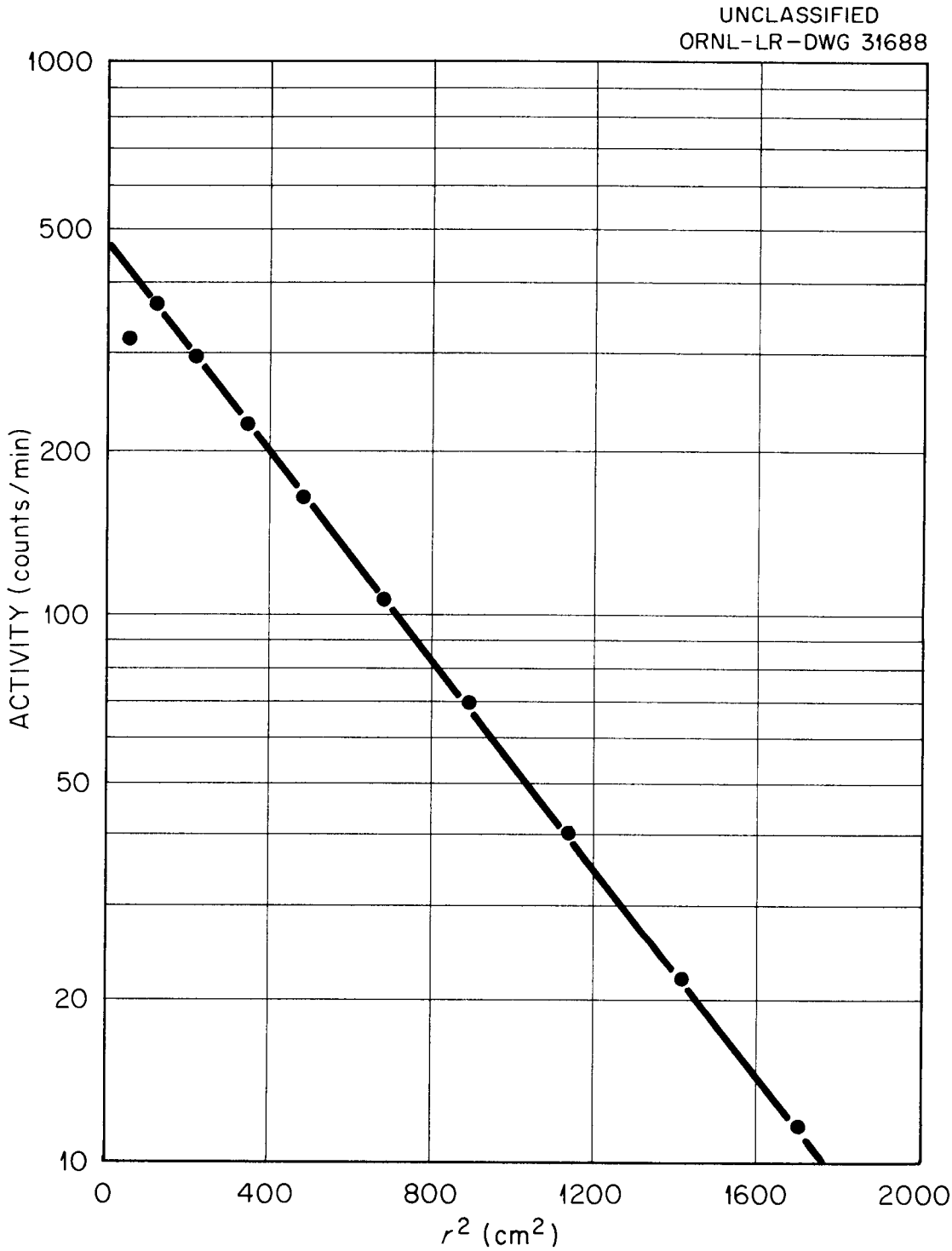

Fig. 3. Indium Resonance Activity vs r^2 in 99.8 % D_2O as Determined by Olcott, Ref. 9.

in 99.8% D_2O. Figure 4 shows the activity distribution obtained for this composition. Figure 5 shows his results for other percentages of D_2O.

It is of interest to compare these direct measurements with one in which an attempt was made to determine the appropriate value of τ to be incorporated into an estimate of fast nonleakage probability in a reactor. Such an experiment is reported by Kash[7] in which the age to "thermal" energy was computed by comparing the properties of an exponential lattice moderated with heavy water with those of the same lattice when the water contained 150 mg of B_2O_3 per liter. Cohen[2] states that the assumption is made that only the leakage and thermal absorption properties of the lattice are changed by the addition of poison. Hence, η, ϵ (the fast fission factor), p (the resonance escape probability) and τ will remain unchanged while L^2, f, and B^2 (the diffusion length squared, the thermal utilization, and the buckling, respectively) are changed. This assumption is valid if the additional poison is insufficient to alter the neutron spectrum. With these conditions one can write

$$\left(\bar{B}^2 - \bar{B}^2\right)\tau = \ln\left[\frac{1 + \bar{B}^2\,\bar{L}^2}{1 + B^2\,L^2}\,\frac{f}{\bar{f}}\right]$$

where the barred values refer to the poisoned lattice. The result obtained using this formula is $107 \pm 5\ cm^2$. The major contribution to the uncertainty in this value is said by the author to be the uncertainty in the cross sections involved.

All the reports on experimental measurements seem to have one common failing, namely, that little is said concerning the accuracy of the cited purity of the heavy water used. Since the age is quite sensitive to the percent of light water present for high concentration D_2O mixtures, the analysis of the mixture clearly deserves more discussion than is generally given.

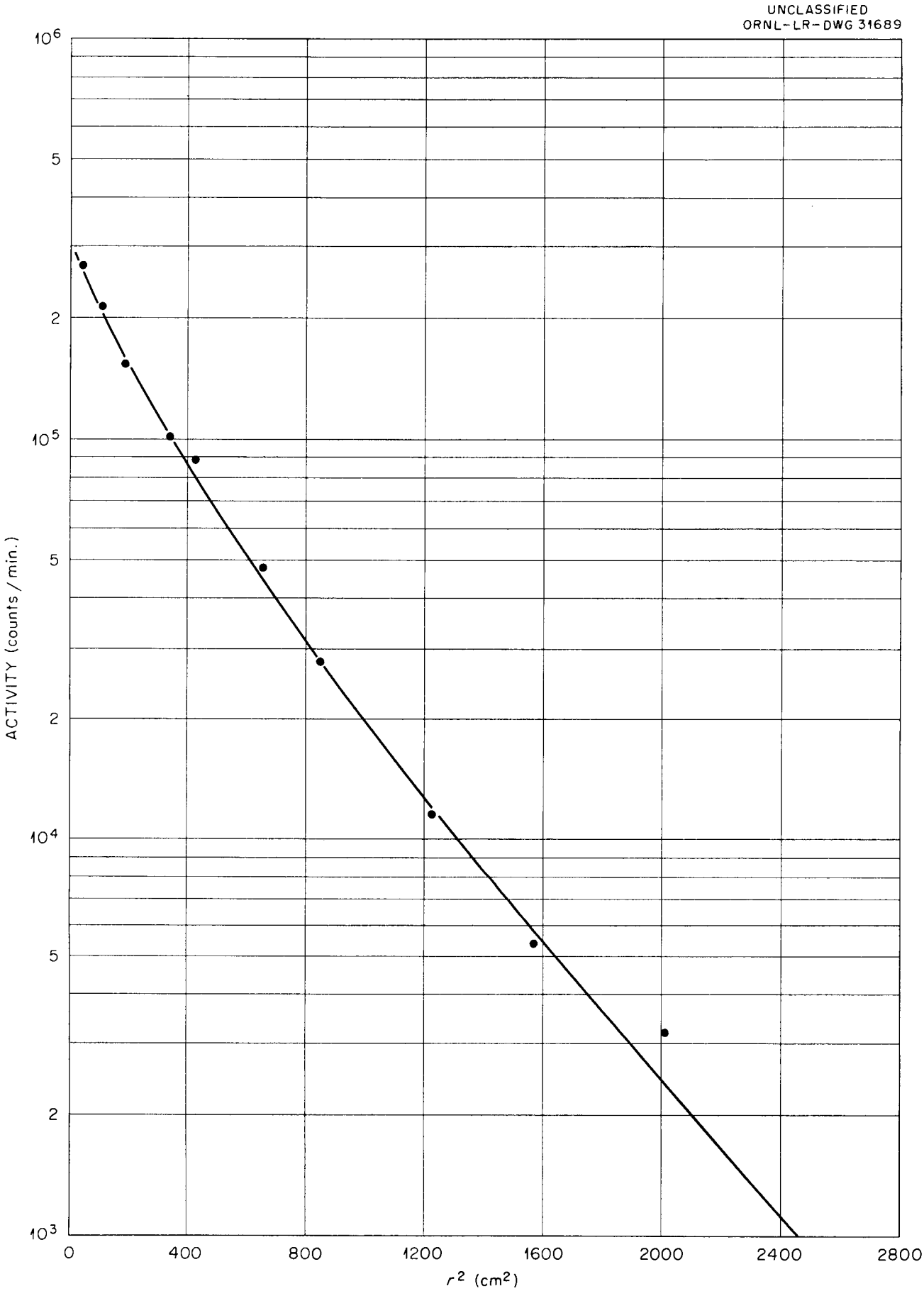

Fig. 4. Indium Resonance Activity vs r^2 in 99.8 % D_2O as Determined by Wade, Ref. 14.

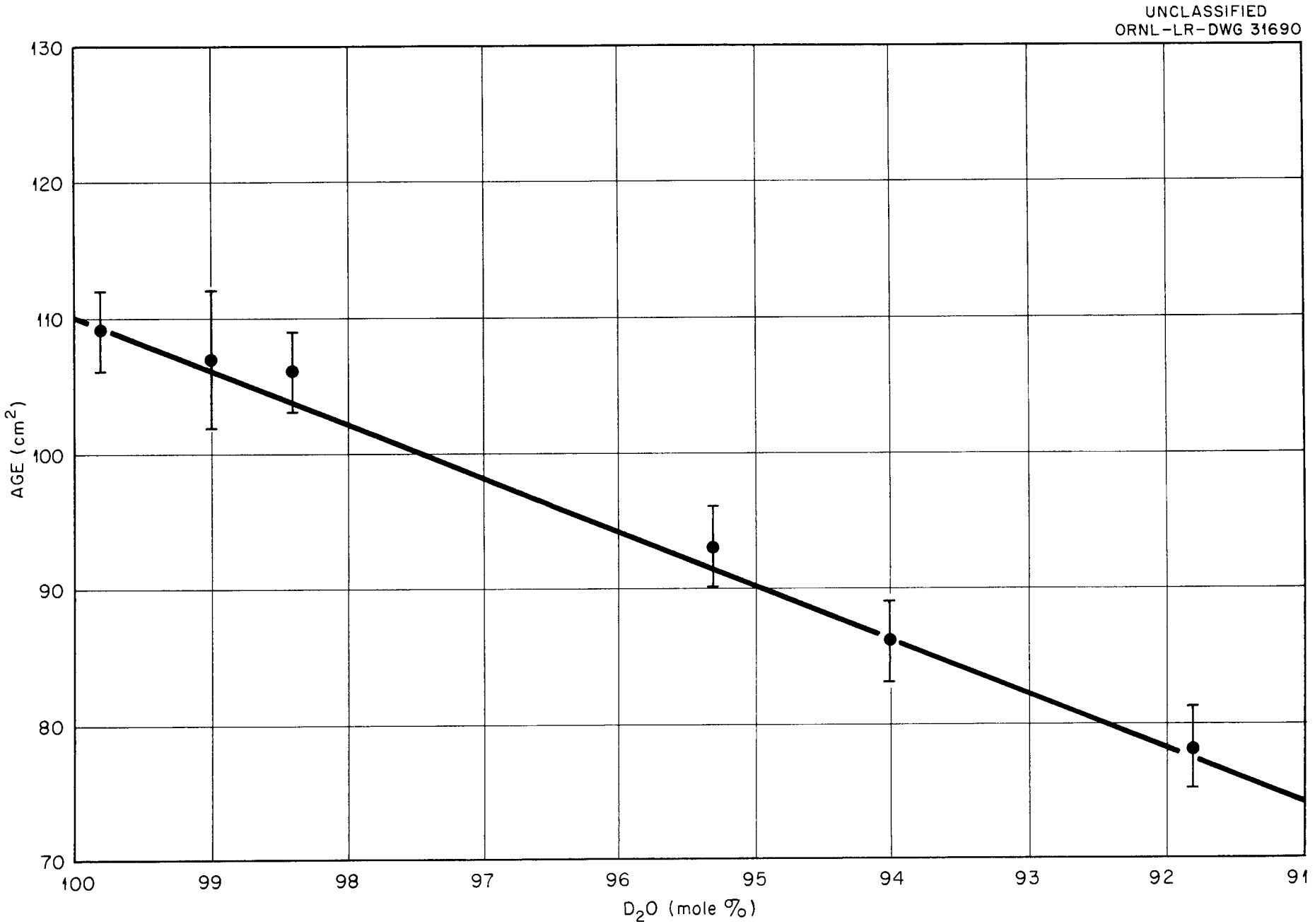

Fig. 5. Age to Indium Resonance for Fission Neutrons vs Moderator Purity as Determined by Wade, Ref. 14. (Error limits shown are those cited by Wade)

THEORETICAL ESTIMATES OF AGE

Fermi[5] appears to have been the first to estimate the age of heavy water on the basis of cross section measurements. The procedure he followed was evaluation of the integral

$$\tau = \int_{E_1}^{E_o} \frac{\lambda^2(E)}{3(1 - \bar{\mu})\xi} \frac{dE}{E} \qquad (2)$$

Taking E_o and E_1 as 2×10^6 e.v. and 0.1 e.v. respectively, a value of 121.7 was obtained. Using the cross section data cited by Fermi, the age is 103 cm^2 from 2×10^6 e.v. to 1.44 e.v. Addition of a first and last flight correction would give about 110 cm^2. It is surprising how good an agreement with experiment may be obtained by this simple integration procedure. Plass,[10] who included the fission spectrum measured by Richards,[12] obtained a value of 106 cm^2. Unfortunately, few details are given, but it is probable that the value was obtained by integration using a formula like equation 2 above. Inclusion of first and last flight corrections is not mentioned, nor are the cross sections employed cited.

A far more elaborate treatment is that employed by H. D. Brown[1] and by Coveyou and Sullivan.[4] Here the age was computed by Monte Carlo techniques applied to the momemts method.[15] Since this is equivalent to a numerical solution of the Boltzmann equation, it is possible, in theory at least, to achieve any desired degree of accuracy, provided sufficient information concerning cross sections and scattering angles is available. In the case of Brown's work, however, both deuterium and oxygen are assumed to scatter isotropically; Coveyou and Sullivan, on the

other hand, include the anisotropy of oxygen scattering (predominantly forward) but not that in deuterium (predominantly backward below 2 mev). Figure 6 compares the results obtained by these workers with the data of Wade for fission neutrons in various D_2O-H_2O mixtures. Agreement between calculation and experiment is seen to be improved by the inclusion of the anisotropic scattering in oxygen. (It is of interest to point out here that in 1947, Rose[13] computed a decrease of 2.1 cm^2 due to anisotropic scattering in deuterium on the basis of the limited information available to him at that time.)

SUMMARY

The most reliable experimental value for the age of fission neutrons to indium resonance in 99.8% D_2O appears to be 109 $\pm$ 3 cm^2, (Wade, 1956). Other work is in close agreement, ranging from 104 (Fermi) to 111 cm^2 (Olcott). Simple numerical integration calculations fall in this range; Monte Carlo estimates are likewise in good agreement but the full effects of anisotropic scattering have not yet been included in these calculations.

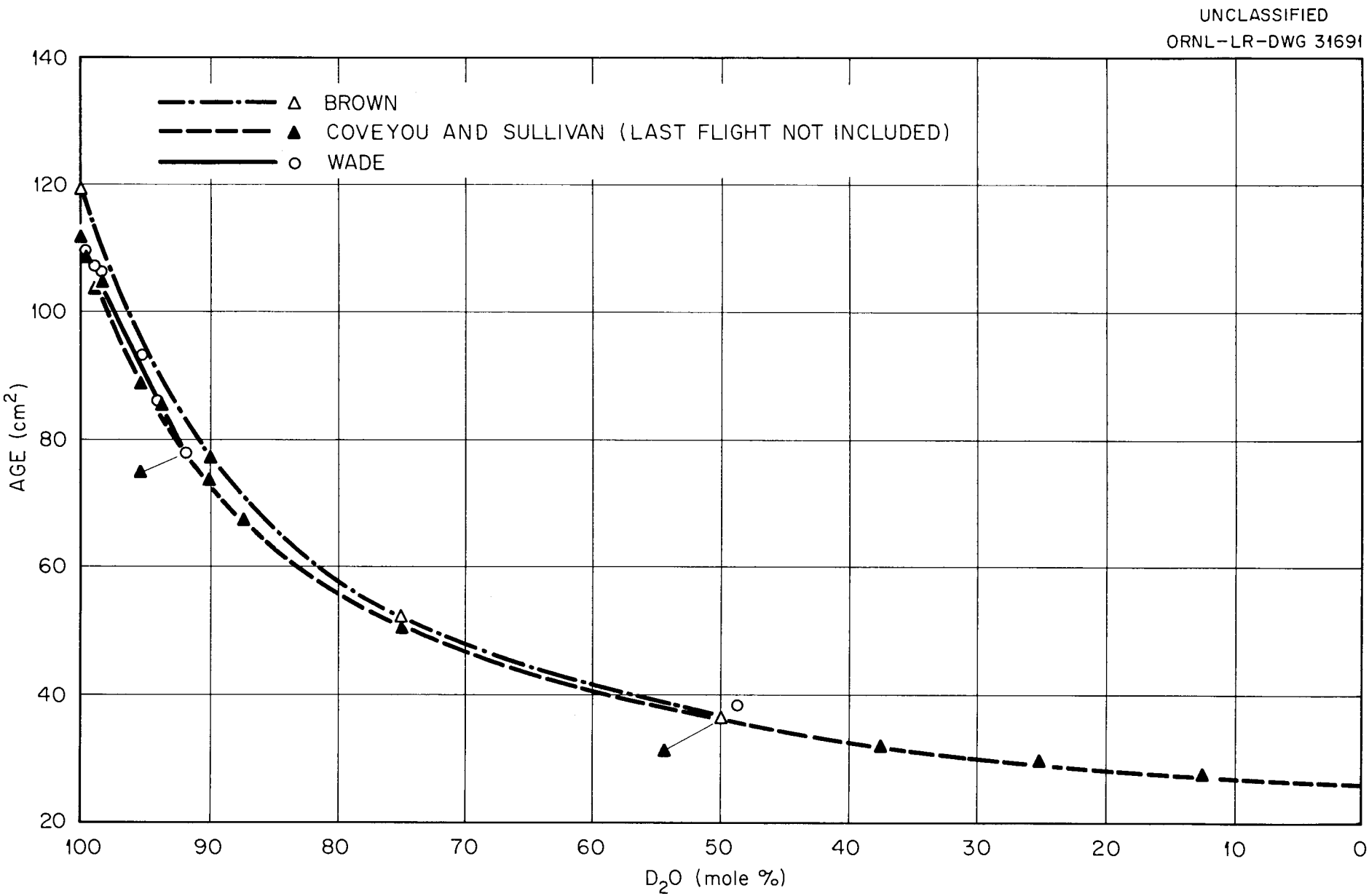

Fig. 6. Comparison of Monte Carlo Calculation of Age of Fission Neutrons in Various Mixtures of Light and Heavy Water with Experimental Data of Wade, Ref. 4.

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