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SPATIAL DISTRIBUTION MEASUREMENTS OF FISSION NEUTRONS IN WATER AS AN OXYGEN DATA TEST

(AWBA DEVELOPMENT PROGRAM)

FEBRUARY 1978

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MASTER

BETTIS ATOMIC POWER LABORATORY

WEST MIFFLIN, PENNSYLVANIA

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FISSION NEUTRONS IN WATER AS AN OXYGEN DATA TEST
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FOREWORD

The Shippingport Atomic Power Station located in Shippingport, Pennsylvania was the first large-scale, central-station nuclear power plant in the United States and the first plant of such size in the world operated solely to produce electric power. This project was started in 1953 to confirm the practical application of nuclear power for large-scale electric power generation. It has provided much of the technology being used for design and operation of the commercial, central-station nuclear power plants now in use.

Subsequent to development and successful operation of the Pressurized Water Reactor in the AEC-owned reactor plant at the Shippingport Atomic Power Station, the Atomic Energy Commission in 1965 undertook a research and development program to design and build a Light Water Breeder Reactor core for operation in the Shippingport Station. In 1976, with fabrication of the Light Water Breeder Reactor (LWBR) nearing completion the Energy Research and Development Administration established the Advanced Water Breeder Applications program (AWBA) to develop and disseminate technical information which would assist U.S. industry in evaluating the LWBR concept. All three of these reactor development projects have been administered by the Division of Naval Reactors with the goal of developing practical improvements in the utilization of nuclear fuel resources for generation of electrical energy using water-cooled nuclear reactors.

The objective of the Light Water Breeder Reactor project has been to develop a technology that would significantly improve the utilization of the nation's nuclear fuel resources employing the well-established water reactor technology. To achieve this objective, work has been directed toward analysis, design, component tests, and fabrication of a water-cooled, thorium oxide fuel cycle breeder reactor to install and operate at the Shippingport Station. Operation of the LWBR core in the Shippingport Station started in the Fall of 1977 and is expected to be completed in about 3 to 4 years. Then the fissionable fuel inventory of the core will be measured. This effort, when completed in about 2 to 3 years after completion of LWBR core operation, is expected to confirm that breeding actually took place.

The Advanced Water Breeder Applications (AWBA) project was initiated to develop and disseminate technical information that will assist U.S. industry in evaluating the LWBR concept for commercial-scale applications. The project will explore some of the problems that would be faced by industry in adapting technology confirmed in the LWBR program. Information to be developed includes concepts for commercial-scale prebreeder cores which will produce uranium-233 for light water breeder cores while producing electric power, improvements for breeder cores based on the technology developed to fabricate and operate the Shippingport LWBR core, and other information and technology to aid in evaluating commercial-scale application of the LWBR concept.

Technical information developed under the Shippingport, LWBR, and AWBA projects has been and will continue to be published in technical memoranda, one of which is this present report.

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ABSTRACT

The spatial distribution of the total neutron density from a ^{252}Cf source in pure water was measured to high statistical precision at distances from 11 to 80 cm from the source. Assuming the adequacy of the ENDF/B-IV hydrogen, and reasonable constraints on the fission spectrum mean energy, good agreement between experiment and a one-dimensional transport calculation was obtained for both ENDF/B-III and IV oxygen, with Version III slightly better. However, small residual differences remained that could not be removed by adjustment of the spectrum mean energy alone.

I. Introduction

In a program to determine a suitable cross section set for use in criticality calculations for highly enriched ^{233}U systems, McNeany and Jenkins¹ at the Oak Ridge National Laboratory recently reported comparisons of experiment and calculation for a set of six simple homogeneous assemblies. The assemblies were characterized by Hydrogen/ ^{233}U (H/U) ratios varying from zero, for a sphere of bare unreflected ^{233}U metal, to nearly 2000. Two cross section sets were studied, ENDF/B-IV and the 16 group Hansen Roach Library² developed at Los Alamos in 1961. Except for the bare metal assembly, calculations with ENDF/B data showed an upward trend in eigenvalue with decreasing H/U. The discrepancy reached over 2% at an H/U value of 154. One critical, with an H/U of 39, was actually 7% high. However, the paraffin reflector of this assembly makes it difficult to interpret. Calculations with the Hansen Roach data set, on the other hand, showed good agreement with experiment throughout the range of H/U ratios. It was concluded that the ENDF/B-IV data file was not satisfactory for the calculation of multiplication factors in highly enriched ^{233}U systems having H/U ratios less than about 400.

McNeany and Jenkins also analyzed two highly enriched ^{235}U assemblies with the ENDF/B and Hansen Roach data sets. Agreement among the calculations and experiments was good. However, one of the assemblies was of bare metal, a core type which, for ^{233}U , gave an anomalously low eigenvalue result with ENDF/B. The second assembly had an H/U ratio of 1393, for which good agreement would be expected also for a ^{233}U fueled system. Therefore, no real conclusions could be drawn from this

work regarding a possible eigenvalue discrepancy trend for ^{235}U similar to that observed in ^{233}U systems.

Ullo and Hardy³ extended the eigenvalue study of the ENDF/B-IV evaluation to 17 highly enriched homogeneous ^{233}U and ^{235}U critical assemblies, including most of those calculated by McNeany and Jenkins. Agreement with the ORNL results was good, with the additional observation of the same upward trend in eigenvalue with decreasing H/U for the ^{235}U systems. It was also demonstrated that the eigenvalue error was strongly correlated with the leakage fraction for both fuel types.

A variety of modifications to the input nuclear data (ENDF/B-IV) were considered by Ullo and Hardy, including fuel, oxygen and hydrogen cross sections, and fission neutron spectra average energy and shape. Eigenvalue sensitivities to changes in each of the data types were evaluated to identify possible candidates contributing to the calculated eigenvalue error. This study revealed that, for high leakage assemblies, the calculated eigenvalues were very sensitive to the oxygen cross sections, as reflected by results for the various ENDF/B versions of oxygen (I, III, and IV, since I and II are identical). In particular, the use of ENDF/B-I oxygen reduced the calculated eigenvalue by approximately 0.6% compared to ENDF/B-IV, about 1/3 the maximum observed error. Version III showed an intermediate behavior.

The low leakage assemblies were insensitive to the oxygen data set. Thus, in general the calculated eigenvalue dependence on the oxygen data set was shown to be correlated with leakage. This means that, at least to first order, oxygen affects the eigenvalue by modifying core leakage in the direction required to reduce the eigenvalue error. This, coupled

with the fact that oxygen was a common element in both fuel systems studied, prompted a closer look at the oxygen cross sections.

The present effort was undertaken to provide a clean independent integral test of the ability of the various ENDF/B oxygen evaluations to describe correctly neutron transport in an oxygen containing medium. The work consists of three parts. In the first, a brief calculational survey of neutron transport in various oxygen containing media to identify an optimum one in which to perform the experiment is described. Available integral data for this medium is then discussed. In the second, the present experimental arrangement and its calculational model are then described. Finally, a comparison of experiment and calculation is presented along with the experimental data which can be used for testing any other evaluation.

II. Preliminary Calculations

Any experiment to be considered should be simple from the point of view of analysis, i.e., the geometry should be easily described, and the sensitivity to oxygen data should be reasonable and should not be confounded by uncertainties in the data for other constituents. It was decided that an experiment to measure the neutron distribution in a near infinite oxygen bearing medium from a near point fission source satisfied these conditions. The media chosen for the survey calculations were water, solid CO₂, and liquid oxygen.

The calculations were performed for a point source-point detector with P7MG, an extension of P3MG⁴, a one-dimensional P_L program. The calculations contained 57 fast groups (the MUFT 54 group scheme with three additional $\Delta u = 0.25$ groups above 10 MeV). One thermal group (0.0 - 0.625 eV) with cross sections averaged in a 25 group infinite medium calculation was

also used. The flux angular treatment was double P_1 thermally and P_7 fast. The region of solution extended over 100 cm, and the boundary conditions employed were current = 0 at the origin and flux = 0 at the right hand boundary. At the origin was a point fission source with a spectrum that was reported in Ref. 5.

The results of the calculations are shown in Figs. 1 through 3, where we have plotted the ratio of the thermal neutron flux using ENDF/B-I and III oxygen and an early modified Troubetzkoy⁷ version to that for ENDF/B-IV. The hydrogen and carbon data were taken from ENDF/B-IV for all calculations. Based on these results, water was chosen as the medium to perform the neutron spatial distribution measurements. The reasons for this choice were as follows:

- 1) Meaningful differences between different oxygen data sets in liquid oxygen do not appear until large distances (~ 100 cm) from the source. Therefore, a large system would have to be constructed, with the attendant problems of handling liquid oxygen.
- 2) The CO_2 system would also have to be large to detect a difference between oxygen data sets. In addition, the analysis would be complicated by the presence of the carbon scattering resonances, where characterization and uncertainties might confound the oxygen data comparisons.
- 3) The hydrogen cross section is well known. Water is a convenient medium from an experimental viewpoint, with the important parameters of purity and density (i.e., temperature) easily controlled. In addition, preliminary tests with an available ^{252}Cf spontaneous fission source indicated that the required statistical accuracy of $\pm 1\%$ beyond 50 cm could be achieved with a thermal detector.

Calculations similar to those above for each of the three media were carried out at indium and gold resonance energies. An enhanced sensitivity to oxygen was observed. However, this advantage was outweighed by the much lower flux levels.

The literature is replete with reports of neutron distribution measurements in pure water, and it is of interest to examine these for application to the problem at hand, i.e., determination of a best available data set for oxygen. The measurements reported are generally of two types, age and spectrum shape.

The age is proportional to the second spatial moment of the flux distribution (generally determined at indium resonance energy, 1.46 eV) from a point or plane source (with point or plane detector). The contribution to the second moment falls off very rapidly with distance beyond a few centimeters from the source, with over 90% of the moment integral accumulated within 20 cm. The emphasis in age measurements is therefore on relatively short distances. In this region, the flux distribution is not very sensitive to scattering from oxygen and, in fact, the fission age in water decreases only about 6% if oxygen slowing down is left out completely. Table I lists the ages calculated using P7MG for a ^{252}Cf fission source⁵ for three ENDF/B oxygen evaluations. Only the differences are significant here, and these are seen not to exceed about 1%. This is less than the experimental uncertainty for the best known flux age, that from a ^{235}U fission source. Therefore, neutron age results cannot be used to provide a conclusive test of the adequacy of different oxygen cross section sets.

While age results alone are not useful here, Spiegel⁶, in a recent measurement of the age of ^{252}Cf fission neutrons in water, took indium

activation data out to 65 cm. His paper lists the results of a smooth curve fit to the normalized data at 0.5 cm intervals. An attempt was made to analyze these data to determine if some information about oxygen could be extracted. Calculations were performed again with P7MG. The source encapsulation was described as an aluminum sphere of radius 0.435 cm, with the ^{252}Cf fission neutrons originating from a region at the center with a 1 mm radius. The P_0 flux in multigroup 54 (1.855 - 1.439 eV) was computed at 0.5 cm intervals along a radius from the source and compared with experiment. The fission spectrum in Ref. 5 was employed. Four sets of calculations were performed using different oxygen data sets: ENDF/B-I, III, IV and the previously mentioned evaluation by Troubetzkoy⁷. Employing the same normalized units for experiment and calculation, the ratios of experiment to calculation are shown plotted in Fig. 4. It can be seen from the figure that good agreement for the spatial distribution of indium resonance neutrons is not achieved for any of the oxygen data sets.

The sensitivity of these calculations to the choice of a ^{252}Cf fission neutron spectrum representation was next investigated. The calculation was repeated using the ENDF/B-I oxygen and a Maxwellian representation with the same average energy as that of the complex spectrum of Ref. 5, 2.10 MeV. As shown in Fig. 5, no significant difference was found. The average energy of the Maxwellian was then raised to 2.154 MeV, which was judged to be a harder spectrum than the upper limit of the error. The results are displayed in Fig. 5; significant differences still remain. It was concluded that no reasonable hardening or change in shape of the ^{252}Cf fission neutron spectrum would produce good experiment/calculation agreement over the spatial

range spanned by Spiegel's measurements. The reasons for this are not known.

Spectrum measurements in good geometry have proven useful in the testing of total cross section data^{9, 10, 11}. However, in evaluating the ability of an oxygen data set to describe leakage correctly, scattering angular distributions are important, and any experiment to be considered should be sensitive to them. Some of the spectrum measurements also required unfolding of a proton recoil distribution and resolution broadening of the calculation before a comparison with calculation could be made. These procedures introduce significant uncertainties.

Based on the above considerations, it was decided that a new experiment that would emphasize the deep penetration of neutrons in water to an accuracy of $\sim \pm 1\%$ at distances out to 70 - 75 cm was needed. In this way (assuming the adequacy of ENDF/B-IV hydrogen and reasonable constraints on the shape and mean energy of the ²⁵²Cf spectrum), experiment/calculation comparisons could be used to assess the adequacy of different oxygen cross section data sets.

III. Experimental Arrangement

(a) Detectors

Since the previously described calculations indicated significant sensitivity to oxygen scattering of the thermal neutron flux distribution in water, it was decided to use a thermally sensitive detector. The advantages of this choice compared to a resonance detector are high efficiency and the option of using a "live" system, i.e., a neutron counter rather than the activation of a foil. This, again, permits a higher rate of data accumulation. The requirements for the detector

are that it be small to minimize water displacement, have adequate efficiency, and very low γ -ray sensitivity. The associated electronics should also have a high count rate capability (which is, of course, closely related to γ -ray sensitivity). In addition, it should be possible to represent accurately the energy dependence of the detector efficiency in the calculation.

All of these requirements were met using miniature boron lined neutron detectors with a fast electronics system. The specifications for the detectors were determined from preliminary tests with available detectors and ^{252}Cf sources. Three detectors were fabricated, designated as the high efficiency, low efficiency, and zero efficiency detectors. Except for the boron linings, all were made of identical materials and had the same dimensions, 0.635 cm diameter with 0.635 cm active length. The high sensitivity detector, which covered the distance range of 22 to 80 cm from the source, had an enriched (92%) ^{10}B lining 0.2 mg/cm² in thickness. The low sensitivity detector, with a depleted (11%) ^{10}B lining approximately 0.1 mg/cm² thick, covered the distance range of 11 to 45 cm. The detector sensitivities, as provided by the manufacturer, were 7.2×10^{-3} and 3.3×10^{-4} c/s per unit incident moderated neutron flux. Both linings were optically thin to thermal neutrons so that the energy dependence of the detector efficiencies corresponds to the shape of the ^{10}B absorption cross section. This allowed a simple representation of the detector in the calculation, as discussed below.

The zero sensitivity detector had no boron lining and was used to determine the γ -ray response. This is of special importance

since even a low γ -ray sensitivity, caused primarily by pulse pileup, could seriously distort the measured neutron distribution.

It should be pointed out that the ^{10}B cross section is accurately $1/V$ in shape throughout the energy range of importance here. Therefore, this experiment measured the spatial distribution of the total neutron density, independent of the slowing down spectrum shape at each measurement location.

The detectors were placed at the bottom of thin walled (0.50 mm) zircaloy tubes 0.77 cm in diameter and 152 cm long. Low capacitance cable coupled the detectors to a fast rise time (16 ns), short clipping time (150 ns), charge sensitive preamplifier designed specifically to reduce γ -ray pulse pileup in high γ -fields. This preamplifier had the additional advantage of very low input impedance, thereby accomodating long cable lengths at the input. The output of the preamplifier went directly to a fast, fixed threshold discriminator.

(b) Water Tank

The experiment was performed in a cylindrical stainless steel water tank, 229 cm in diameter and 183 cm high. The tank was filled with deionized water, which was continuously circulated through a multibed resin column to maintain water purity. Water temperature, as measured by an alcohol thermometer, was $20.5 \pm 0.2^\circ\text{C}$ and was controlled to within 0.1°C by means of a proportional temperature controller which operated a 1500 watt heater.

(c) Neutron Source

The ^{252}Cf spontaneous fission isotope was contained in a standard Oak Ridge National Laboratory NSD encapsulation, as shown in Fig. 6.

Also included in the figure is all of the information on encapsulation dimensions and materials as provided by ORNL. The ^{252}Cf content at the time of those measurements was 1.3 mg. The largest contribution to the dimensional uncertainty in this experiment was the radial location of the ^{252}Cf within the resin. For purposes of experiment/calculation comparisons, it was assumed that the ^{252}Cf was located at the center with a radius of 1 mm. The source was threaded into a 1.90 cm high stainless steel plug pressed into the end of a thin walled zircaloy tube 2.0 cm in diameter and 200 cm long. Holes drilled into the tube allowed it to fill with water.

(d) Detector and Source Support Systems

The source and detector tubes were rigidly suspended from platforms placed on a pair of heavy aluminum rails 12.7 cm high and 3.8 cm wide resting on the 9 cm wide flange on the top of the tank. Machining precision was kept within ± 0.025 mm for all important surfaces to insure good parallelism and perpendicularity. These measures were necessary since the distance scale was set at only a single location.

The detector platform rode on 4 nylon wheels whose larger central diameter fit snugly into the rail grooves while the smaller diameter inner and outer edges rested on the rails. Holes 3 mm in diameter were precision drilled every centimeter along the top of both rails. Close fitting steel pins passing through the platform and into the holes provided positive locking for good positional reproducibility.

The distance scale was set by placing the source platform so that the source was about 76 cm from the tank wall ($1/3$ of the tank diameter) and 90 cm below the water surface. The detector platform

was then wheeled in close to the source and the detector height varied until the maximum count rate location was found. This was done separately for each detector, and insured that the source and detector were at the same effective heights. An aluminum rod with a hole drilled through its center, such that the detector tube fit through snugly, was then placed at the detector location. The rod length had been accurately machined so that the distance from the center of the source encapsulation to the detector center line, when the rod end made electrical contact with the surface of the source, was 17 cm. Contact was made by varying the source platform location with the detector platform pinned in place. The source platform was then clamped to the rails. The estimated uncertainty in this procedure was ± 0.5 mm.

IV. Results

(a) Experimental Data

Data were taken every centimeter, from 11 to 45 cm for the low efficiency detector and 22 to 80 cm for the high efficiency detector. Statistical precision was limited by drift in the electronics which required frequent normalizations at standard locations. Runs were repeated until the standard deviation at each point was 1% or better. The count rate data in the overlap region from 22 to 45 cm were ratioed point by point, and a mean and standard deviation for the high to low efficiency detector of 21.900 ± 0.215 were obtained. The standard deviation is consistent with the single measurement precision for the ratio and it was concluded that, within statistics, no shape difference was observed between the two detectors in the overlap region.

The mean is also in excellent agreement with the value of 21.8 obtained from the ratio of the detector sensitivities provided by the manufacturer.

The results for the combined set of data, normalized to the high efficiency detector, are listed in Table II and plotted in Fig. 7. No smoothing has been performed. The data are fully corrected, as indicated below, and may be directly compared with calculation.

(b) Corrections

(i) Background

By background, we mean counts due to electronic noise, cosmic rays, etc. A linear least squares fit to the $\ln(Ar^2)$ data of Table II from 71 to 80 cm was made to obtain a relaxation length in this region. This was found to be 8.84 cm. Data were also accumulated at 104 cm, the farthest location the detector platform could be accurately located. This location is still 50 cm from the tank wall and a factor of over 20 lower in signal than the 80 cm point. An excess of 0.0006 c/s over that predicted for 104 cm from the relaxation length was observed and attributed to "background". No attempt was made to assign an uncertainty to this value since, as can be seen from Table II, the maximum correction is only about 0.5%.

(ii) γ -Ray Sensitivity

The γ -ray response was found to be a strong function of detector bias, as illustrated in Fig. 8. The operating biases of the two neutron sensitive detectors, shown in Fig. 8,

were a compromise between low γ -ray response and the need to operate on the detector plateaus to minimize drift. The spatial dependence of the γ -ray response at 875 volts bias, corresponding to the low sensitivity detector, is shown in Fig. 9. The ratios of the zero sensitivity to low sensitivity detector responses at each of the measurement points are also shown in Figure 9. The statistical uncertainty for the three points at 30, 35 and 40 cm is about 20%. Within measurement precision, it can be seen that the ratios are constant at approximately 0.6%. This is a surprising result and is evidently caused by a balance between rapidly decreasing neutron and pulse pileup signals. This ratio would be a factor of over 50 lower for the high sensitivity detector. Therefore, no γ -ray response correction was applied to either of the neutron sensitive detectors.

(iii) Count Rate Losses

The maximum count rate encountered in this experiment was 2768 c/s for the high sensitivity detector at 22 cm. The expected count rate loss at this value, based upon measurements taken with a critical assembly on a stable period, was 0.05%, with an estimated uncertainty of 25%. This is negligible compared to the statistical uncertainty, so no corrections were made. A check on this procedure is provided by the excellent shape agreement in the overlap region of the high and low sensitivity detectors.

(iv) Photoneutron Reactions

At long distances from a fission source in water, photoneutrons from (γ, n) reactions on the deuterium in the water become significant¹². However, at 80 cm, the contribution is estimated to be only about 0.1%, so no correction for this effect was necessary.

(c) Comparison With Calculation

To compare with the experimental data in Table II, 57 group P7MG calculations were performed similar to the survey calculations described earlier. The major differences were in the source and detector descriptions. The source dimensions and compositions as represented in the calculation are shown in Fig. 10. The 1.38 cm outer radius was obtained by sphericizing the source body and plug into which it was threaded. Similarly, the pressed aluminum pellet was sphericized to form the intermediate radius. The resin was distributed homogeneously in the pressed aluminum, and fission neutrons originated within the 0.1 cm radius about the center.

The calculations included a trace (10^{-15} atoms/b-cm) of ^{10}B uniformly distributed throughout the water. The ^{10}B absorption fraction was then calculated every cm as a function of distance from the source. In the region of solution, no mesh spacing was greater than 0.385 cm. This spacing was found to be adequate based on a mesh sensitivity study. The quantity calculated is given by

$$A^{B10} = \int_E \int_V \Sigma_a^{B10}(E) \phi(r, E) dE d\vec{r}. \quad (1)$$

Due to the $1/V$ absorption cross section of the ^{10}B , Eq. (1) can be

rewritten as:

$$\begin{aligned}
 A^{B10} &= N^{B10} \sigma_{a_o}^{B10} \int_E \int_V n(E, r) dE d\vec{r} \\
 &= 4 \pi N^{B10} \sigma_{a_o}^{B10} \int_E \int_{R_i}^{R_{i+1}} n(E, r) dE r^2 dr. \quad (2)
 \end{aligned}$$

The energy integration is over the range from 0.0 to 21.0 MeV and the volume integration in each case is over the spherical shell between R_i and R_{i+1} , where $(R_{i+1} - R_i) = 0.77$ cm. This is just the diameter of the detector tube. Dividing Eq. (2) by the volume yields the volume normalized ^{10}B absorption rate

$$A^{B10} \left(\frac{R_i + R_{i+1}}{2} \right) = N^{B10} \sigma_{a_o}^{B10} \eta \left(\frac{R_i + R_{i+1}}{2} \right); \quad (3)$$

where

$$\eta \left(\frac{R_i + R_{i+1}}{2} \right) = \frac{3 \int_E \int_{R_i}^{R_{i+1}} n(E, r) r^2 dr dE}{(R_{i+1}^3 - R_i^3)} \quad (4)$$

is the spatially averaged total neutron density which is assigned to the midpoint of the spherical shell. Hence, the calculations yield a quantity which is directly proportional to the total neutron density as a function of radius and which is directly comparable to the measurements.

Before presenting comparisons with the various oxygen data sets, Fig. 11 illustrates the strong dependence of neutron density on oxygen scattering at increasing source-detector distances. For this calculation, oxygen scattering was deleted completely, and the

EXP/CALC ratio was normalized at 13 cm. As expected, the calculation increasingly over-predicts the density, with the error reaching over a factor of 6 at 80 cm.

In Fig. 12, we show the EXP/CALC comparison for the early modified Troubetzkoy⁷ evaluation. The data have been normalized in this, as well as all subsequent, comparison in the 13-20 cm region. The first two points were not included in the normalizations since the slowing down curve (Fig. 7) shows increased curvature here. This indicates that those points would be more sensitive to the source location and its details. It can be seen in Fig. 12 that the EXP/CALC ratio is poor beyond 40 cm with the discrepancy increasing to 20% near 70 cm. The inadequacy of this oxygen evaluation has been observed by others^{10,13}.

The striking improvement obtained for ENDF/B-I oxygen is shown in Fig. 13. The cross sections of importance here were taken by ENDF from the work of Slaggie and Reynolds¹⁴. These cross sections were maintained in ENDF/B-II. Agreement between experiment and calculations is seen to be excellent out to about 50 cm with the calculation over-predicting slightly the neutron density beyond this distance.

The results for ENDF/B-III and IV are shown in Figs. 14 and 15 respectively. Two observations may be made from these figures. First, except for the slightly faster rise near 25 cm and above 75 cm for ENDF/B-IV, they are very close in shape. Secondly, both evaluations systematically underpredict the neutron density by 1.5-2% beyond 30 cm. The relative behavior of all three evaluations is,

incidentally, consistent with their eigenvalue response, i.e., a higher neutron density further out which is indicative of increased leakage and lower eigenvalue.

Since we have standardized the use of ENDF/B-IV hydrogen cross sections in this work, the only other variable that can affect the EXP/CALC comparisons is the characterization of the ^{252}Cf fission neutron spectrum. The spectrum model used in all calculations above was that reported by Green et al⁵. It has a complex shape with a mean energy of 2.105 MeV. This is at the low end of the range of measurements and is significantly softer than the reference Maxwellian average energy of 2.13 MeV proposed in the recent NBS evaluation¹⁵. It was judged that the most likely range of mean energies for the ^{252}Cf spectrum is $2.10 < \bar{E} < 2.13$ MeV. The effect of increasing the mean energy is to worsen the ENDF/B-I comparison while improving those for Versions III and IV. For this reason we consider Versions III and IV to be in better agreement with the present experiment than Version I oxygen.

The effect of changes in mean energy and shape of the fission neutron spectrum were investigated for Version IV oxygen by calculations using Maxwellian distributions at the estimated limits of the mean energy values of 2.10 and 2.13 MeV. The lower value also corresponds to the same mean energy as the complex spectrum model and provides a measure of the shape sensitivity. The results are shown in Fig. 16. Comparison of Fig. 16 with Fig. 15 shows the shape change to have a small effect with a maximum difference in the EXP/CALC ratio of about 1% near 40 cm. Of far greater importance is the mean energy change. For

the 27 keV difference the ratio is seen to swing from below to above unity, reaching values of 0.97 and 1.02, for a difference of 5% beyond 70 cm. Therefore, for a reasonable choice of limits on the mean energy of the ^{252}Cf fission spectrum, Version IV oxygen brackets the experimental results.

In Fig. 17, we show the EXP/CALC ratio for an intermediate energy ($\bar{E} = 2.118$ MeV) Maxwellian. Agreement is seen to be within $\pm 1.5\%$ over the entire range from 11 to 80 cm. Comparison of Figs. 14 and 15 shows that Version III oxygen will behave similarly, although with a significantly smaller bulge between 25 and 40 cm, so that agreement will be about $\pm 1\%$. The agreement between experiment and calculation using both ENDF/B III and IV must be considered excellent in view of the change in magnitude of nearly 4 decades in the neutron density. Nonetheless, it is apparent from Fig. 17 that systematic differences between experiment and calculation do exist, and these differences may be a result of the inadequacy of the Maxwellian description for the ^{252}Cf fission neutron spectrum.

V. Conclusions

The spatial distribution of the total neutron density from a near point fission ^{252}Cf neutron source in pure water has been measured every cm from 11 to 80 cm to a statistical precision of better than 1%. The purpose of the measurements was to provide data for testing the ability of various oxygen evaluations to describe adequately neutron leakage in oxygen containing systems. The measurements were compared to spherical geometry multigroup transport calculations. Assuming the adequacy of the ENDF/B-IV hydrogen and a reasonable constraint on the ^{252}Cf fission

spectrum mean energy, good agreement between experiment and calculation was obtained for both ENDF/B-III and IV oxygen with Version III slightly better. A shift of ± 13 keV in mean energy produced strong disagreement at long penetration distances for both evaluations.

The remaining differences between experiment and calculations for both Versions III and IV oxygen, while small, are systematic and cannot be removed by adjustment of the fission neutron spectrum mean energy alone. This may be due to inadequacy of the Maxwellian description of the fission neutron energy distribution.

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List of Tables

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Table I

CALCULATED AGES FOR DIFFERENT ENDF/B OXYGEN EVALUATIONS^a

<u>Version</u>	<u>Ages (cm²)</u>
I	27.1
II	26.9
III	26.8

^a Fission spectrum from Ref. 5.

Table II

COUNT RATE DATA

<u>r (cm)^a</u>	<u>A^b (c/s)</u>	<u>r (cm)^a</u>	<u>A^b (c/s)</u>	<u>r (cm)^a</u>	<u>A^b (c/s)</u>
11	44,917	34	194.3	57	3.301
12	35,216	35	158.1	58	2.801
13	27,485	36	130.4	59	2.386
14	21,217	37	106.9	60	2.041
15	16,460	38	87.62	61	1.764
16	12,643	39	72.83	62	1.516
17	9855	40	60.17	63	1.305
18	7558	41	50.07	64	1.113
19	5880	42	41.62	65	0.9650
20	4583	43	34.67	66	0.8270
21	3565	44	28.87	67	0.7185
22	2796	45	24.15	68	0.6192
23	2200	46	20.33	69	0.5356
24	1735	47	17.20	70	0.4651
25	1370	48	14.34	71	0.4039
26	1084	49	12.17	72	0.3501
27	872.1	50	10.27	73	0.3050
28	695.3	51	8.649	74	0.2644
29	554.5	52	7.360	75	0.2289
30	449.9	53	6.245	76	0.2003
31	361.4	54	5.263	77	0.1733
32	291.5	55	4.493	78	0.1518
33	236.9	56	3.853	79	0.1312
				80	0.1145

^a Distance between centerline of detector and source.

^b Count rate normalized to the high efficiency detector.

Figure Captions

- Fig. 1 Calculated sensitivity of the spatial neutron distribution in water to different oxygen data sets.
- Fig. 2 Calculated sensitivity of the spatial neutron distribution in solid CO₂ to different oxygen data sets.
- Fig. 3 Calculated sensitivity of the spatial neutron distribution in liquid oxygen to different oxygen data sets.
- Fig. 4 Experiment/calculation comparisons for the measured, Ref. 6, indium activations out to 65 cm using different oxygen data sets. All comparisons are normalized to 1.0 at $r = 6.5$ cm.
- Fig. 5 Experiment/calculation comparisons for measured, Ref. 6, indium activations using different ²⁵²Cf fission spectrum representations. All comparisons are normalized to 1.0 at $r = 6.5$ cm.
- Fig. 6 ²⁵²Cf neutron source encapsulation used in present experiment.
- Fig. 7 Semi-log plot of experimental counting data. r is the distance between source and detector center lines and A the detector count rate (c/s) normalized to the high efficiency detector.
- Fig. 8 Response of the zero sensitivity detector as a function of applied voltage. The operating (detector bias) of the two neutron sensitive detectors are also shown.
- Fig. 9 Spatial response of the zero sensitivity detector at 875 volts bias. The ratios of the zero to low sensitivity detector count rates are shown in parentheses.
- Fig. 10 Source representation in PTMG calculation.

- Fig. 11 EXP/CALC comparison for oxygen deleted from the calculation.
Normalization to 1.0 at $r = 13$ cm.
- Fig. 12 EXP/CALC comparison for the modified Troubetzkoy⁷ evaluation
for oxygen. Normalization in 13-20 cm region.
- Fig. 13 EXP/CALC comparison for ENDF/B-I oxygen. Normalization in
13-20 cm region.
- Fig. 14 EXP/CALC comparison for ENDF/B-III oxygen. Normalization in
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- Fig. 16 EXP/CALC comparisons for ENDF/B-IV for two different Maxwellian
neutron spectra.
- Fig. 17 EXP/CALC comparison for ENDF/B-IV to best fit Maxwellian.

FIGURE I

FIGURE 2

FIGURE 3

FIGURE 4

FIGURE 5

MODEL NSD
DOUBLE ENCAPSULATED

a BODY WEIGHT 15.639 g, CAP WEIGHT 6.045 g

b BODY WEIGHT 8.007 g, CAP WEIGHT 4.135 g

c 0.1 g DRY RESIN WITH THE FOLLOWING CALCULATED
ANALYSES (w/o): C, 54.6%; H, 4.6%; O, 24.5%; S, 16.4%

FIGURE 6

FIGURE 7

FIGURE 9

FIGURE 10

FIGURE 11

FIGURE 12

FIGURE 13

FIGURE 14

FIGURE 15

FIGURE 16

FIGURE 17