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**AEC Research and
Development Report**

**Knolls
Atomic Power
Laboratory**

**Neutron
Spectra
Measurements**

R. S. Stone

R. E. Slovacek

September 15, 1957

Operated for the
United States Atomic
Energy Commission by

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NEUTRON SPECTRA MEASUREMENTS

R. S. Stone, R. E. Slovacek

September 15, 1957

R. S. Stone

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KNOLLS ATOMIC POWER LABORATORY
Schenectady, New York
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ABSTRACT

Thermal neutron spectra have been measured with time-of-flight techniques. Spectra were obtained for pure water and for a nearly homogeneous subcritical assembly where the ratio of thermal absorption to high energy scattering cross section was ~ 0.3 . For each medium, spectra were measured at 298 and 586°K.

The experimental results are presented and compared with calculated theoretical spectra. The agreement between theory and experiment is excellent for the multiplying media. On the basis of this agreement, one concludes that chemical binding effects in light water play a negligible role in determining the equilibrium neutron spectrum in water assemblies.

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NEUTRON SPECTRA MEASUREMENTS

R. S. Stone, R. E. Slovacek

INTRODUCTION

The thermal neutron spectrum in a finite medium with absorption present is of considerable interest both practically in reactor design; and theoretically in understanding neutron thermalization. A rather complete discussion of the present status of the theory and measurement of reactor spectra can be found in the literature.¹

Wigner and Wilkins² calculated the steady-state spectrum in a partially absorbing infinite homogeneous medium with a monatomic gaseous moderator whose scattering nuclei have a Maxwellian energy distribution. The scattering nuclei were assumed to have a constant scattering cross section and a mass equal to that of the neutron while the absorption cross section was assumed to vary as $1/v$. The effects of molecular binding and intermolecular forces in the liquid state were not considered.

Recently Amster³ has extended the previous work so that the neutron spectrum can be calculated for a finite homogeneous medium with the absorption cross section varying arbitrarily as a function of neutron energy. The conditions on the scattering nuclei remained the same as in the Wigner-Wilkins calculation.

These calculations are usually applied to water-moderated assemblies. However, hydrogen, in water, does not completely fulfill the assumptions made on the scatterer in the calculations. Measurements are thus needed to observe the magnitude of the molecular and intermolecular binding effects on spectra.

The measurement of neutron spectra in partially absorbing media has been undertaken recently with two techniques, both of which use time of flight. By using a subcritical assembly in a thermal column with a slow neutron chopper, the authors⁴ obtained preliminary results which indicated good agreement with the Wigner-Wilkins calculations. A pulsed neutron technique was used by Poole⁵ who obtained the spectra in homogeneous light water solutions with various boron concentrations. Amster⁶ has compared Poole's measurements with the calculated spectra and also finds good agreement over most of the energy range except for a consistent deviation in the joining region between the hardened thermal spectrum and the dE/E slowing region.

In this series of measurements a non- $1/v$ absorber was used over a wider temperature range (298 to 586°K) than used previously. In addition, the spectra in the joining energy region was investigated with greater resolution than that used in the previous measurements.

EXPERIMENTAL ARRANGEMENT

The slow chopper technique for measuring spectra was described in detail in KAPL-1499.⁷ Equipment descriptions not contained in that report are included here for completeness.

The arrangement of experimental equipment is shown schematically in Figure 1. The Thermal Test Reactor serves as a source of thermal neutrons to excite an enriched uranium fission plate located in the reactor thermal column. The thermal neutron flux at operating power is about 10^{10} neutrons/cm²-sec incident on the plate. The high-energy neutrons which result from fission in this plate are transmitted through the pressure vessel wall more easily than the thermal neutrons. These neutrons excite the 16-in.-long and 10-in.-diam subcritical assembly which is located in the rear of the vessel. A 2-in.-diam reentrant hole penetrates 8 in. deep into the forward end of the assembly. This hole is filled with a thin-walled helium tank to provide a method of extracting a neutron beam from the bottom of the hole with small perturbation to the medium. The beam leaves the high-pressure region through a 0.030-in.-thick stainless steel high-pressure rupture disk and is collimated by a 1-meter-long boron carbide collimator assembly.

Time-of-Flight Equipment

The emergent beam is chopped by a rotating flat-plate shutter. The 2-in.-high rotor plate stack consists of 1/16-in.-thick borated phenolic resin plates and 1/16-in.-thick aluminum "window frames" serve as plate spacers. This plate stack was capped with borated phenolic resin to form the 6-in.-diam, 6-in.-long rotor cylinder. A shrunk-fit, thin-walled, stainless steel shell provided mechanical support for the rotor.

Since the spectra under investigation contain a relatively large number of high-energy neutrons, boron or cadmium cannot be used as shutter materials. The shutter material must have a high strength-to-weight ratio and must have a large removal cross section at high energies. It was for this reason that the hydrogenous phenolic compound was used.

After a flight path length of three meters, the chopped neutron beam was detected by an array of 15 proportional counters. These counters (2 in. diam) were filled with $B^{10}F_3$ to 1 atmosphere. The active length

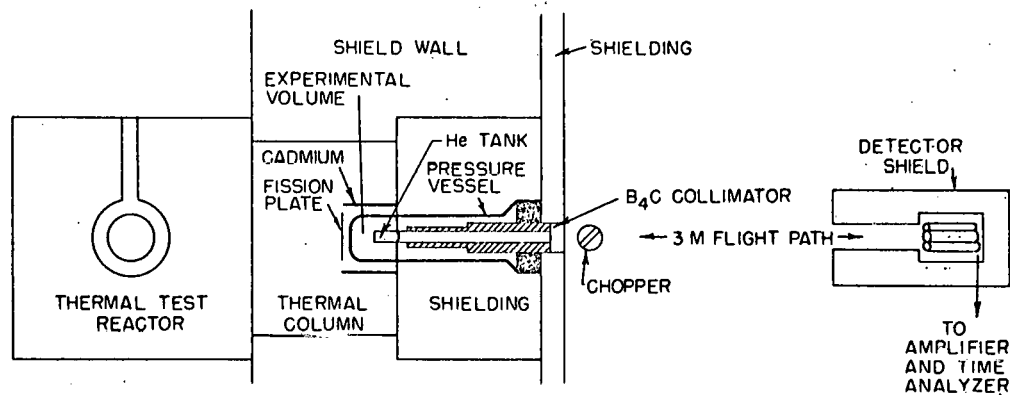


FIGURE 1 EXPERIMENTAL ARRANGEMENT

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of the counters was 7 in. in the beam direction. The ends of the counters were made of aluminum oxide ceramic, whose measured transmission was nearly energy independent over the range of interest. For higher resolution work, it was possible to use only the central row of five counters as a detector. The counter assembly was housed in a large beam catcher whose walls were composed of 6-in.-thick borated paraffin.

The output pulses from the counter were amplified and recorded as to time of arrival in a 256-channel time analyzer.⁸ Channel widths of from 2.5 to 80 μsec were available. For this series of experiments, 40 μsec channels were used. The channelizer was gated on by a pulse from the rotating shutter. The arrival of this pulse was adjusted so that a counter signal, corresponding to an infinite velocity neutron, would be recorded at the beginning of time channel 129. The system parameters were adjusted so that the flight time of a cutoff neutron was less than one-half the time between bursts. Since the rotor construction is symmetric about a plane passed through the center of its plate stack, the time dependent background as a result of transmission through the shutter will also be symmetric about flight time equal to zero. One can

obtain, therefore, signal counting rates corrected for background by reflecting the data about channel 129 and subtracting.

Information stored in each of the 256 channels is printed out on IBM cards and forms an input deck for the IBM-704 computer. This computer has been programmed to reduce the data and yields, as an output, the flux per electron volt as a function of energy.⁹ This program corrects for background, energy dependent system transmission, and system resolution as discussed in the Experimental Corrections section.

Sources

Two types of media were investigated in the beam source region. In one arrangement, the 16-in.-long, 10-in.-diam experimental volume in the rear of the pressure vessel was filled with pure water. For the second type of source region, a multiplying medium was used. Since chemical binding effects on spectra are not well understood even at room temperature, it was of interest to observe their variation, if any, with temperature. Facilities were available to raise the temperature of the source region up to 600°F.

The multiplying medium or matrix consisted of an array of alternate planes of light water, zirconium, and enriched uranium. The spacing between fuel planes was 0.2 in. The fuel was 0.0013 in. thick and was clad with Zircaloy to enable operation of this subcritical assembly at elevated temperatures. The composition of the assembly was chosen so that the ratio

$$\frac{\Sigma_a(0.025 \text{ ev})}{\xi \Sigma_s(1 \text{ ev})} = 0.29$$

Under these conditions, with $\Sigma_a \gg DB^2$, the measured spectrum should well represent the spectrum in an infinite medium of this absorption with a small leakage correction.

The lateral surface of the cylindrical source region was bounded by a 1-1/4-in.-thick carbon steel pressure vessel wall which was covered by a 2-in. layer of thermal lagging and cadmium sheet. Since the buckling would be different to calculate under the existing source and boundary conditions, activation experiments were conducted with manganese or fuel strips to measure the axial and radial flux traverses for each source.

The results of these activations yielded several bits of information. The perturbation of the reentrant hole on the axial gradient was found to be < 3%. In the case of the room temperature matrix, bare and cadmium-covered strip measurements of the flux distribution in both the radial and

axial directions were obtained. The thermal and epithermal flux distributions in the beam source region differed by < 6% and indicated that the spectrum was a rapidly varying function of position only near the boundaries of the matrix. In the beam source region, the flux was found to be represented, to a good approximation, by the function,

$$\phi(E, r, z) = \phi_0(E)e^{-\gamma z} J_0(\alpha r)$$

The buckling will then be given by $B^2 = \alpha^2 - \gamma^2$.

The measured values of α , γ , and B^2 with their estimated errors are shown in Table 1. The values of $D(E_0)B^2$ and $\Sigma_a(E_0)$ are also included for each medium where E_0 is the neutron energy corresponding to the ambient moderator temperature. The values of α and γ were obtained from a least squares fit to the activation data for a distance of three thermal diffusion lengths about the source center.

The magnitude of the measured buckling was used to determine the leakage correction, DB^2 , which was added to the absorption cross section in calculating the spectrum. As a consequence of the bare and cadmium-covered activations in the matrix, the variation of the leakage correction was taken to be the same as the variation of D with energy. The axial flux measurement was also used in relating the measured beam current to the flux in the source region.

In the case of pure water, Σ_a is not large compared to DB^2 for all energies of interest. Therefore, the calculated spectrum will be very sensitive to the variation of both Σ_a and DB^2 with energy. Since the buckling as a function of energy could not be obtained, no leakage correction was applied to the absorption cross sections in calculating the spectrum in water.

EXPERIMENTAL CORRECTIONS

Since several energy dependent corrections must be applied to the experimental data, they will be considered here for completeness although some have been discussed previously.⁷

TABLE I. Measured Values of α , γ , and B^2

<u>Medium</u>	<u>T,</u> <u>°K</u>	<u>E₀,</u> <u>eV</u>	<u>α,</u> <u>cm⁻¹</u>	<u>γ,</u> <u>cm⁻¹</u>	<u>B²,</u> <u>cm⁻²</u>	<u>D(E₀)B²,</u> <u>cm⁻¹</u>	<u>$\Sigma_a(E_0)$,</u> <u>cm⁻¹</u>
H ₂ O	298	0.0257	0.172 ± 0.020	0.166 ± 0.001	-0.057 ± 0.010	-0.0064	0.0220
H ₂ O	586	0.0505	0.112 ± 0.010	0.098 ± 0.001	-0.003 ± 0.002	0.0007	0.0115
Matrix	298	0.0257	0.093 ± 0.006	0.056 ± 0.001	0.0055 ± 0.0011	0.0011	0.193
Matrix	586	0.0505	0.056 ± 0.004	0.036 ± 0.001	0.0018 ± 0.0014	0.0007	0.138

The experimental data can be considered to be a function of the flight time, t , of the form $C(t)$. Since the time of flight, t , uniquely specifies the neutron energy, E , through the relation

$$E = 1/2 \frac{mL^2}{t^2}, \quad (1)$$

where

E = neutron energy,
 m = neutron mass,
 L = flight path length, and
 t = flight time,

the function $C(t)$ specifies the related function $C(E)$ uniquely. However, the change of metric from unit time to unit energy implies that

$$C(E) = C(t) \frac{dt}{dE} \quad (2)$$

or

$$C(E) = C(t) \left[-\frac{t^3}{mL^2} \right] \propto t^3 C(t) \quad (3)$$

That is, the count per unit energy is proportional to the count per unit flight time times the neutron flight time cubed.

Beam Attenuation

A correction to the experimental data must be made for the attenuation effect of the materials in the beam. If the neutron beam current density in the region of the source is $J_S(E)$ and the observed neutron beam current density at the detector is $J_D(E)$, these two current densities will be related by the removal cross section of the materials in the beam. That is,

$$J_S(E) = J_D(E) e^{(\Sigma_{t_1}(E)x_1 + \Sigma_{t_2}(E)x_2 + \dots)}$$

where $\Sigma_{t_i}(E)$ is the total cross section and x_i is the thickness of the i^{th} material in the beam.

Shutter Transmission

The transmission of a rotating flat-plate neutron shutter is a function of neutron energy. This energy dependence has previously been reported in detail.⁷ If one defines the quantity $\beta = \frac{v_{co}}{v}$ as being equal to the ratio of the shutter cutoff neutron velocity to the neutron velocity under consideration, then the relative shutter transmission is given by

$$\tau(\beta) = 1 - \frac{8}{3}\beta^2 \quad ; \quad 0 \leq \beta \leq 1/4 \quad (5a)$$

$$\tau(\beta) = \frac{8}{3}\beta^2 - 8\beta + \frac{16}{3}\sqrt{\beta} \quad ; \quad 1/4 \leq \beta \leq 1 \quad (5b)$$

Counter Efficiency and Effective Flight Path Length

Two additional corrections must be made to the observed data for the variation with energy of counter efficiency and mean stopping position within the counter.

The counter efficiency as a function of energy is given by

$$\epsilon(E) = 1 - e^{-\Sigma_a(E)l} \quad (6)$$

where $\Sigma_a(E)$ is the macroscopic absorption cross section of the counter filling gas and l the length of the counter.

The mean stopping position of a neutron of energy E as measured from the front of the counter is given by

$$\bar{\lambda}(E) = \frac{\int_0^l \lambda \Sigma_a(E) e^{-\Sigma_a(E)\lambda} d\lambda}{\int_0^l \Sigma_a(E) e^{-\Sigma_a(E)\lambda} d\lambda} \quad (7)$$

The effective flight path length is then given by

$$L(E) = L' + \bar{\lambda}(E) \quad (8)$$

where L' is the flight path from the center of the chopper to the front of the active counting volume.

Flux Gradient Correction

Since it is the spectrum of the neutron beam current rather than the spectrum of the neutron flux at the origin of the beam that is measured, an additional correction must be made. The energy dependence of the flux and the beam current are related in the diffusion theory approximation by the following relationship,

$$J_+(E) = \Phi_0(E) - \lambda_T(E) \left. \frac{d\Phi}{dz} \right|_{z=0} \quad (9)$$

The transport mean free, path, $\lambda_T(E)$, in the medium can be obtained from a knowledge of the material cross sections in the assembly and the flux gradients in the beam direction can be obtained from the activation experiments. One can then calculate an energy dependent correction term to the observed current spectrum of the following form

$$\Phi_0(E) = \frac{J_+(E)}{1 - \lambda_T(E) \left. \frac{d}{dz} \ln \Phi(E) \right|_{z=0}} \quad (10)$$

Resolution Corrections

A detailed discussion of the resolution of neutron time-of-flight spectrometers has been presented.⁷ In this earlier discussion, it was assumed that the time-of-flight spectrum of the beam was slowly varying over the instrumental time resolution range.

Frost¹⁰ has shown that an additional correction can be made for the effect of finite resolution in the range where the slowly varying condition does not apply. If one defines the input time-of-flight spectra to the spectrometer as $I(t)$, the observed spectra as $O(t)$, and the spectrometer resolution function as $R(t)$, then

$$O(t) = \int_{-\infty}^{\infty} R(\lambda) I(t + \lambda) d\lambda \quad (11)$$

If $I(t)$ is a continuous function with continuous derivatives, it can then be expanded in terms of a Taylor's expansion

$$O(t) = \int_{-\infty}^{\infty} R(\lambda) \sum_{n=0}^{\infty} \frac{\lambda^n}{n!} \frac{d^n I(t)}{dt^n} d\lambda \quad (12)$$

If the series is uniformly convergent, then

$$O(t) = \sum_{-\infty}^{\infty} \frac{1}{n!} \frac{d^n I(t)}{dt^n} \int_{-\infty}^{\infty} \lambda^n R(\lambda) d\lambda \quad (13)$$

The function $R(\lambda)$ has previously been shown to be even in its argument, and the integral of the function can be normalized to unity. Under these conditions, we may rewrite, neglecting fifth and higher order derivatives,

$$O(t) = I(t) + \frac{1}{2} \frac{d^2 I(t)}{dt^2} \int_{-\infty}^{\infty} \lambda^2 R(\lambda) d\lambda + \frac{1}{24} \frac{d^4 I(t)}{dt^4} \int_{-\infty}^{\infty} \lambda^4 R(\lambda) d\lambda \quad (14)$$

By differentiating with respect to time and again neglecting fifth and higher order derivatives we obtain

$$\frac{d^2 O(t)}{dt^2} = \frac{d^2 I(t)}{dt^2} + \frac{1}{2} \frac{d^4 I(t)}{dt^4} \int_{-\infty}^{\infty} \lambda^2 R(\lambda) d\lambda \quad (15)$$

and

$$\frac{d^4 O(t)}{dt^4} = \frac{d^4 I(t)}{dt^4} \quad (16)$$

By substituting these two results in Equation (14) and by rearranging terms, one obtains

$$I(t) = O(t) \left[1 - \frac{1}{2} \frac{1}{O(t)} \frac{d^2 O(t)}{dt^2} \int_{-\infty}^{\infty} \lambda^2 R(\lambda) d\lambda + \left\{ \frac{\left(\int_{-\infty}^{\infty} \lambda^2 R(\lambda) d\lambda \right)^2}{4} - \frac{\int_{-\infty}^{\infty} \lambda^4 R(\lambda) d\lambda}{4!} \right\} \frac{d^4 O(t)}{O(t) dt^4} \right] \quad (17)$$

The even moments of the resolution function can be obtained from the analytic expression derived in KAPL-1499.⁷ The output function and its derivatives are obtained directly from the experimental data. Therefore, a numerical correction can be calculated with Equation (17) that will correct for variation of the input spectrum over the range of the resolution function. In the case of the pure water spectra where the counting rate increases sharply between the dE/E region and the thermal region of the spectrum, the resolution correction was as large as 20%.

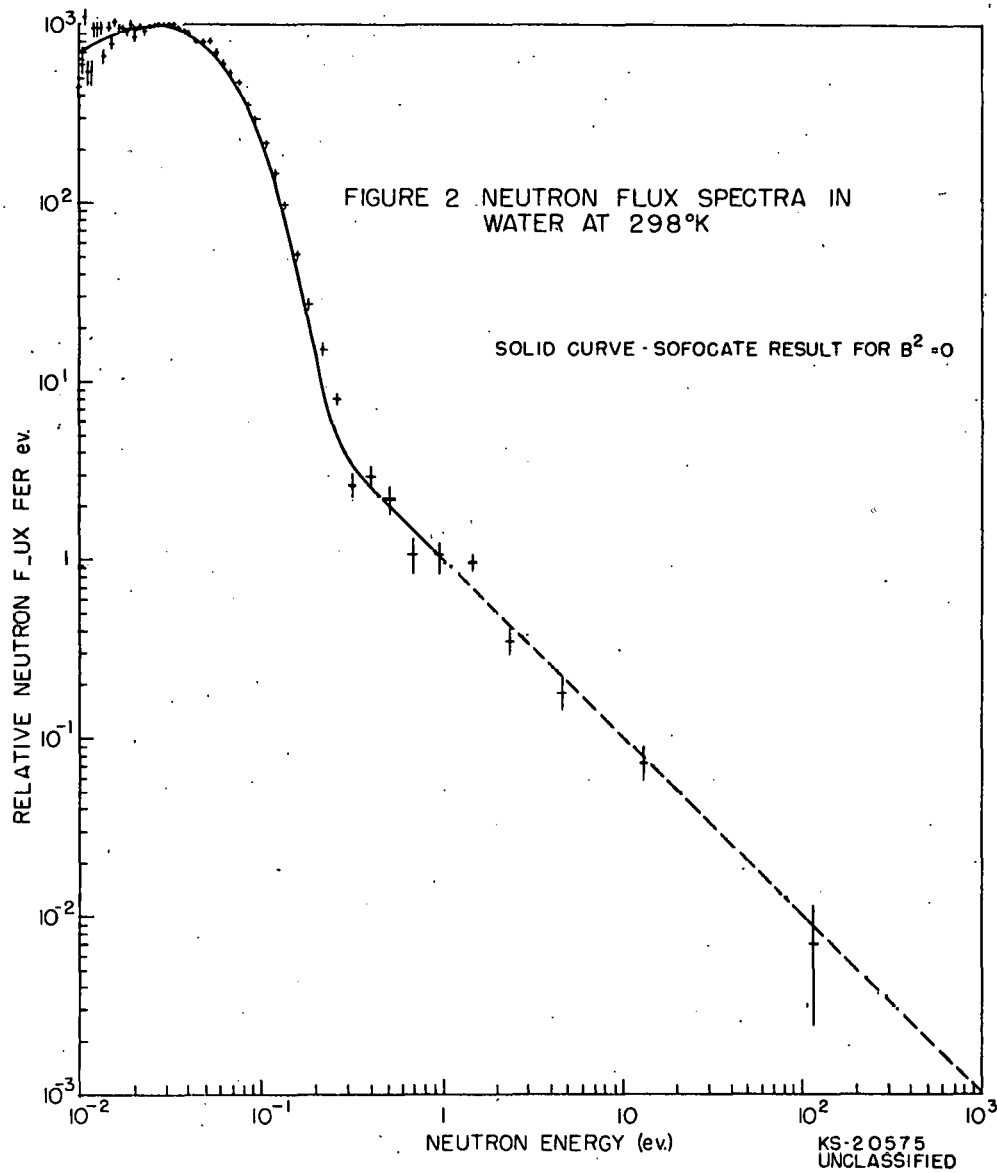
COMPARISON OF THEORY WITH EXPERIMENT

Spectra were calculated for each source-medium with Amster's SOFOCATE code.³ For the two types of media, pure water and matrix, calculations were performed for moderator temperatures which corresponded to the measured temperature used in the spectrum measurements, namely 298 and 586°K.

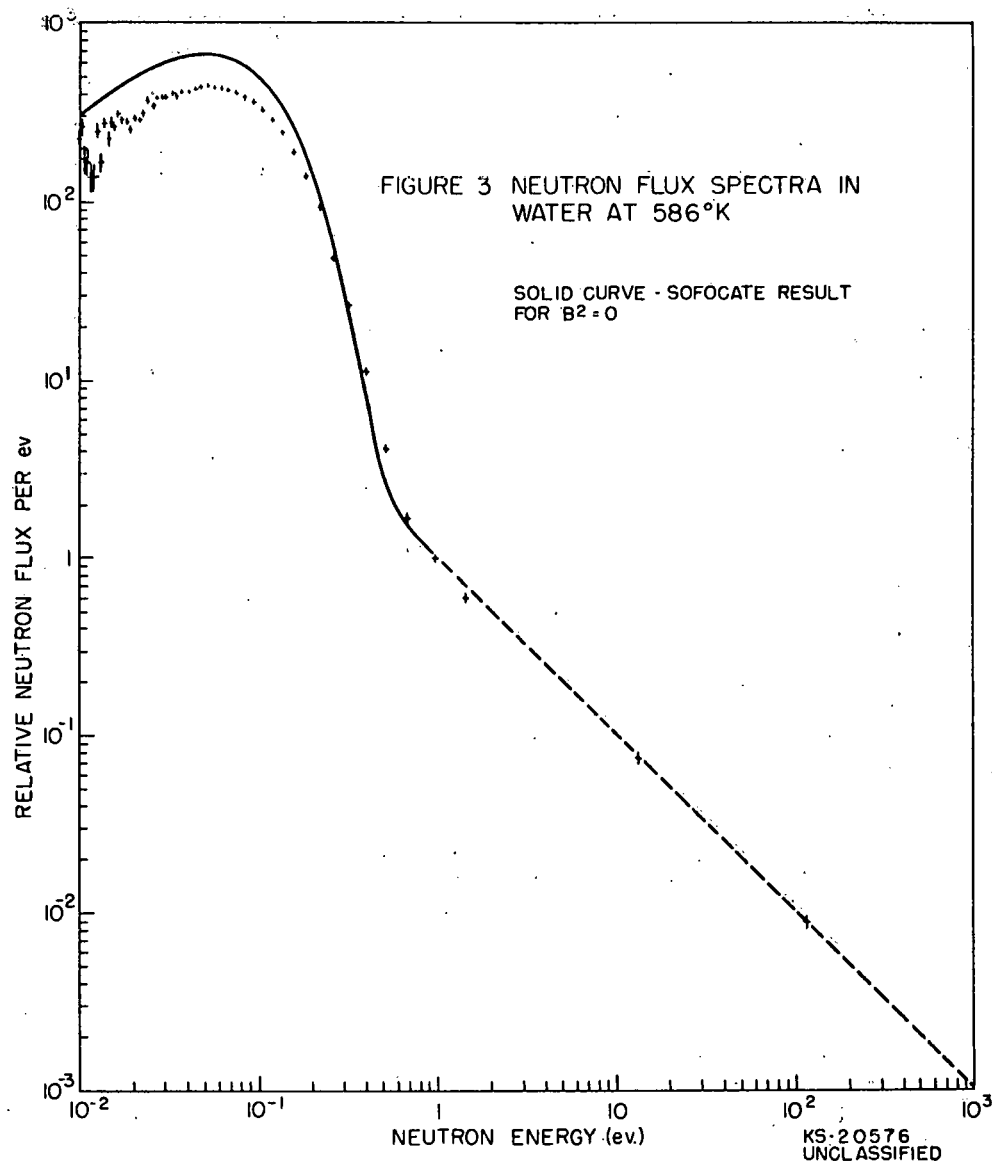
Included in the code is the leakage term DB^2 to correct Σ_a in a finite medium. For the hot and cold matrix cases, the appropriate measured value of B^2 was used in the calculations. Since $\Sigma_a(E_0) \gg DB^2$ for the multiplying media, this corresponds to a small correction (~1%) to the spectra and the calculated spectra will be insensitive to errors in the measurement of $B^2(E)$. In addition, the calculation treats only an equivalent homogeneous assembly with the same average composition as the heterogeneous experimental assembly. This approximation is good over most of the energy range except below 0.2 ev where self-shielding effects would tend to reduce the effective homogeneous Σ_a value obtained from the material composition. A self-shielding factor was determined for the mean fuel Σ_a and corresponds to the spectrum by using Bohl's results for a repeating slab array.¹¹ The self-shielding factors, which were then applied throughout the thermal range in the calculations, were 0.898 for the case of the 298°K matrix and 0.924 for the case of the 586°K matrix.

In the case of pure water, Σ_a is not large compared to DB^2 over the entire energy range of interest. The calculated spectrum will then be very sensitive to $B^2(E)$ as well as $D(E)$. For the particular pure water spectrum source used, it is quite likely that the fast leakage is different from the thermal leakage. Since the measurement of $B^2(E)$ was not available, the thermal leakage correction was not made to Σ_a in the case of pure water.

The results appear in Figures 2 through 5 where the calculated spectra appear as solid curves. The dashed line is the dE/E spectrum above 1 ev that is assumed in the calculations. In these figures, the crosses are measured values of the flux with the vertical extent of the cross indicating the probable error in the relative flux as determined from the counting statistics for each channel. Both the calculated spectra and measured spectra were normalized to give a slowing down flux of 1 neutron/cm²-sec-ev at an energy of 1 ev. To obtain this, the experimental data above 1 ev were fitted to a dE/E spectrum in each case.



The room temperature water spectra are shown in Figure 2. The good agreement between the measured spectrum in a finite medium and the infinite medium calculated spectrum is probably fortuitous. From Table 1 it is seen that the leakage effect at ~ 0.025 e.v. could be appreciable ($\sim 30\%$).



The leakage effect at 1 eV is even greater. The water spectra at 586°K is shown in Figure 3. Here the measured thermal flux seems to be low by ~50%. This is probably accounted for by the difference between the thermal leakage and fast leakage effects. The shape of the measured hot- and cold-water spectra in the energy range between thermal energy and 1 eV exhibit no large discrepancies from the calculated infinite medium spectra.

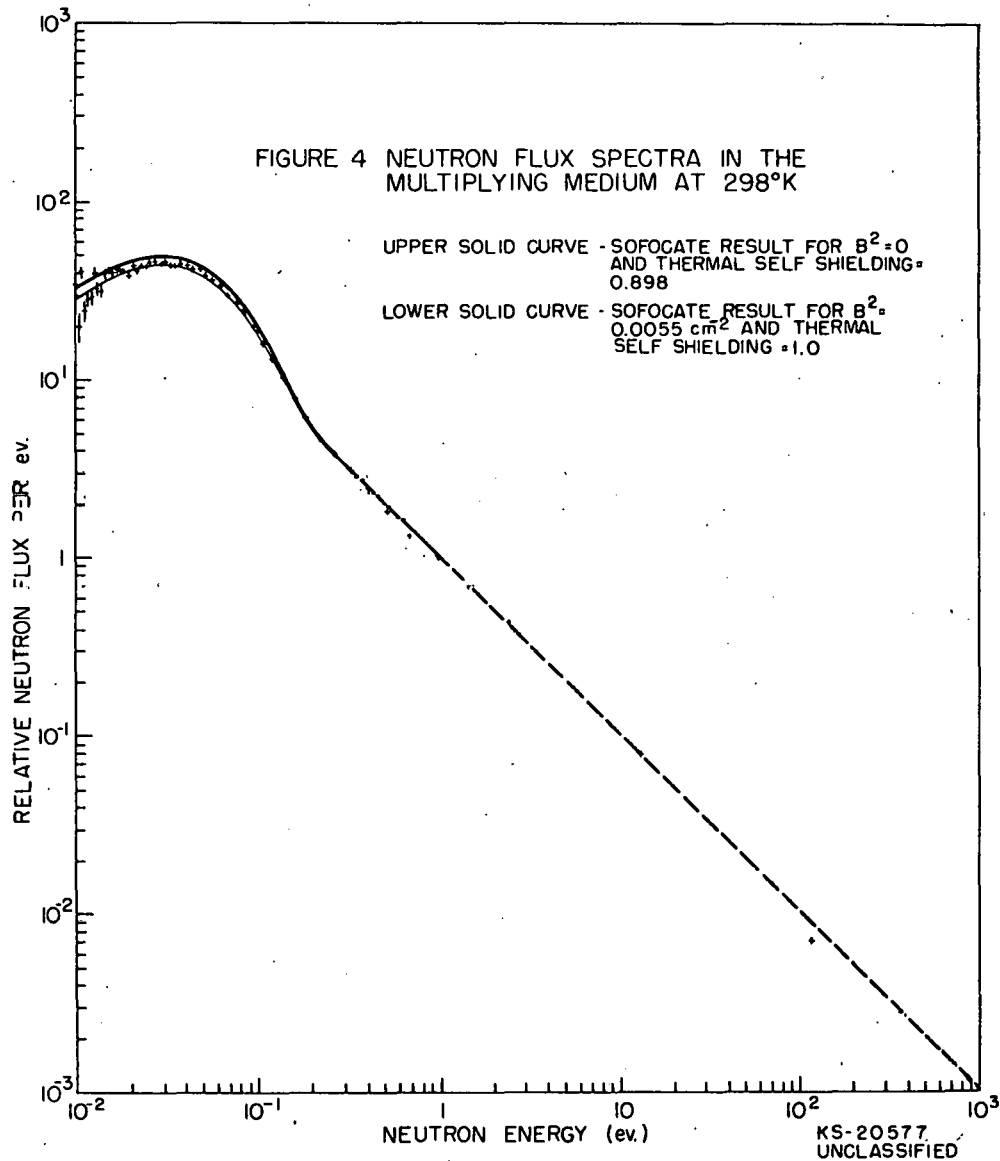


Figure 4 shows the room temperature spectra in the multiplying medium. The lower solid curve is the calculated spectrum when one assumes a self-shielding factor of 1.0. The upper curve is for a self-shielding factor of 0.898. Above the thermal energy region, the agreement is excellent. In the thermal region, the measured points lie between the two calculated curves. Apparently an average self-shielding factor is not adequate to describe the effect and perhaps one should use an energy dependent correction here. No significant deviations caused by chemical binding effects appear in the measured spectrum.

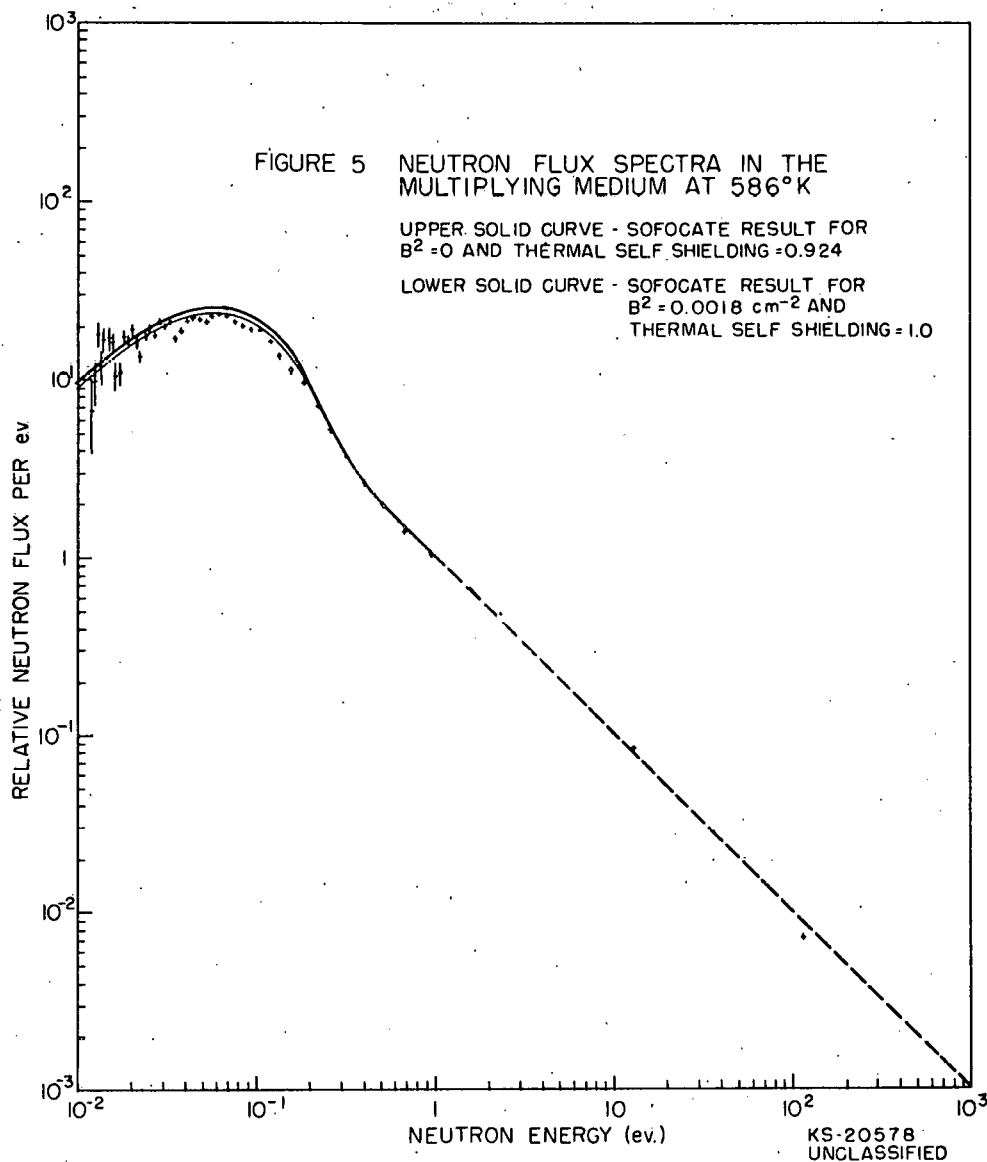


Figure 5 shows the spectra in the multiplying medium at 586°K. Again the lower solid curve is for a self-shielding factor of 1.0, while the upper curve is for a self-shielding factor of 0.924. The agreement is not as good as in the room temperature case, but it is still good. Again the shape of the curve above the thermal energy range is in excellent agreement with calculations.

The general agreement over the entire energy range of interest is consistent with Poole's results. However, Poole's results indicated a consistent deviation in the joining region between the dE/E and "hardened" thermal spectra where chemical binding effects may be noticeable. The present results indicate no deviations in this region. It is to be noted that the count rate increases sharply in this region and that the resolution correction can be as large as 20% in the case of pure water and 6% for the matrix. In the case of pure water, the ratio of thermal to dE/E flux is larger than it is for the matrix. Higher resolution would tend to reduce the correction and thus the uncertainty associated with the measured point. The data presented here were obtained with $\Delta E/E = 1/1$ at 3.8 ev.

A comparison between the measured and calculated spectra can be made by using each spectrum to calculate the average cross section, $\bar{\sigma}$, of a $1/v$ absorber for the overlapping energy range, 0.01 to 1.0 ev. One can form the ratio

$$\left(\frac{\bar{\sigma}}{\sigma_0} \right) = \frac{E_0^{1/2} \int_{0.01}^1 E^{-1/2} \Phi(E) dE}{\int_{0.01}^1 \Phi(E) dE} \quad (18)$$

where σ_0 is the cross section at $E_0 = 0.0252$ ev.

Table 2 contains the results of these calculations and the estimated errors of this ratio. The error in the ratio using the measured spectrum is based on counting statistics, the error in the energy calibration, and the error in the transmission correction for materials in the beam. The error in the ratio using the calculated spectrum is based on the uncertainties in the composition cross sections and on the errors in the measured temperatures of the media.

As indicated from the plotted results, the agreement between the experimental value and the calculated value obtained with no self-shielding correction in calculating the spectrum is good. The self-shielding corrections seem to be too large since the experimental and calculated results do not agree with their probable error ranges. In any case, the agreement is within about 4%.

TABLE 2. Average $1/v$ Cross Sections

<u>Source</u>	$\bar{\sigma}/\sigma_0$	
	<u>Experimental</u>	<u>Calculated</u>
298°K Matrix	0.565 ± 0.004	
Self-Shielding Factor = 1.0		0.574 ± 0.009
Self-Shielding Factor = 0.898		0.587 ± 0.009
586°K Matrix	0.458 ± 0.006	
Self-Shielding Factor = 1.0		0.469 ± 0.006
Self-Shielding Factor = 0.924		0.479 ± 0.006

CONCLUSION

Chemical binding effects on the spectra in water-moderated reactors appear to be extremely small. Consequently, spectra as calculated by the SOFOCATE code should be quite accurate for media with $\Sigma_a/\xi\Sigma_S \leq 0.3$. The average cross sections that are used in reactor calculations obtained with the code should be good to within 4%.

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