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**RESEARCH AND
DEVELOPMENT
REPORT**

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Research and Development Report

DETERMINATION OF FAST NEUTRON
SPECTRA IN A HEAVY WATER REACTOR
USING NUCLEAR EMULSIONS

by

Jimmie R. Youngblood

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DETERMINATION OF FAST NEUTRON SPECTRA IN A
HEAVY WATER REACTOR USING NUCLEAR EMULSIONS.

Jimmie R. Youngblood*

ABSTRACT

Nuclear emulsions have been exposed in the core of the Ames Laboratory Research Reactor to obtain the fast neutron flux spectrum (.8 to 5.0 MeV). It is found that the spectrum is generally similar to the Watt fission spectrum but with the peak shifted upward in energy to as high as 2.3 MeV. Superimposed on this are small depressions and peaks corresponding to the resonances and anti-resonances in the cross section of oxygen. Also, a theoretical approach to obtaining the neutron spectrum independent of experiment has been developed.

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I. INTRODUCTION

In recent years several theoretical neutron spectrum calculations for heavy water moderated reactors have been advanced. The need has been expressed for an experimental neutron spectrum in the MeV range which could be compared to the theoretical calculations.¹

An experiment was developed to obtain the neutron spectrum of the Ames Laboratory Research Reactor core which is heavy water moderated. Nuclear emulsions were chosen as the neutron detectors. The range of neutron energies analyzed is from .8 to 5.0 MeV. Poison strips of borated aluminum were inserted into the fuel elements in order to approximate an "end of life" core. This also allowed the use of a 24 element symmetrical core with the control rods almost completely withdrawn.

The center thimble and a point near the edge of the core near a fuel element were investigated to take advantage of the known fast neutron fluxes in these regions.

II. THEORY OF NUCLEAR EMULSION ANALYSIS.

A. Mechanics of Proton Recoil

Although the energy spectrum of the neutrons is not measured directly, it is inferred by measuring the tracks formed by the recoil protons in a nuclear emulsion. The protons form ionization trails after being bombarded by the neutrons and may be readily seen through an ordinary microscope.

The method of analysis used for the nuclear emulsions exposed in the core of the reactor takes into account the fact that the neutron source is not a beam or a point source but rather an extended volume yielding an anisotropic neutron flux.

Since for n-p scattering below 14 MeV the cross section in the center of mass system is invariant with angle, the fraction of protons scattered into a solid angle bounded by the cones of half angle β and $\beta+d\beta$ is,

$$dN(E, \beta) / N(E, \beta) = 2\pi \sin \beta d\beta \quad (1)$$

where $N(E, \beta)$ is the number of protons corresponding to neutron energy E scattered per unit solid angle at angle β .² Designating the laboratory angle by ψ , the kinematics of elastic scattering yield $\psi = \frac{1}{2} \beta$, and the ratio of solid angles in the laboratory and C.M. systems is $(4 \cos \psi)^{-1}$. We thus have in the laboratory system,

$$dN_L(E, \psi) = N(E, \beta) 4 \cos \psi 2\pi \sin \psi d\psi \quad (2)$$

where $dN_L(E, \psi)$ signifies the number of protons scattered into the solid angle $2\pi \sin \psi d\psi$. From $E_p = E_n \cos^2 \psi$, $dE_p = -2 E_n \cos \psi \sin \psi d\psi$ and, dividing Eq. (2) by this quantity, we have the relation,

$$dN_L / dE_p = \text{constant} \quad (3)$$

where E_p is the energy of the recoil proton. This implies that if one measures all proton recoils, each neutron interval will yield a proton energy distribution which is constant in magnitude from zero to the neutron energy.

If $F(E)$ is the neutron flux in neutrons/cm²-sec/MeV as a function of energy E , the number of proton recoils in a cm³ of a given substance due to neutrons of energy E is given by

$$dN_p = n_p F(E) \sigma(E) dE \quad (4)$$

where n_p is the number of hydrogen atoms/cm³ in the substance and $\sigma(E)$ is the neutron-proton scattering cross section in cm².³ The total number

of recoil protons N_p due to neutrons of all energies up to $E_{\max}$ can thus be obtained by integrating Eq. (4) as indicated below.

$$N_p = \int_0^{E_{\max}} n_p F(E) \sigma(E) dE. \quad (5)$$

Some pertinent neutron cross section data are supplied graphically in Appendix A.

Another expression for N_p can be obtained by observing a typical recoil proton spectrum as shown in Fig. 1.

Fig. 1. Recoil Proton Spectrum

Let $P(E_p)$ be the recoil proton spectrum in protons per MeV per cm^3 of detecting substance as a function of proton energy E_p . If we have a group of neutrons of energy E , it will contribute the following number of proton recoils:

$$dN_p = \frac{dP(E)}{dE} E dE \quad (6)$$

It should be noted that E_p and E are interchangeable in Eq. (6).

Integrating over-all neutron energies we have another expression for the total number of proton recoils N_p ;

$$N_p = \int_0^{E_{\max}} \frac{dP(E)}{dE} E dE. \quad (7)$$

Equating Eqs. (5) and (7) and differentiating both sides with respect to E we have

$$\begin{aligned} n_p F(E) \sigma(E) &= \frac{dP(E)}{dE} E \\ \text{or} \quad F(E) &= \frac{E}{n_p \sigma(E)} \frac{dP(E)}{dE}. \end{aligned} \quad (8)$$

$F(E)$ is the total integrated neutron flux (nvt) and $P(E)$ is the proton recoil spectrum.

This formula applies to all cases where all proton recoils in a given energy interval are measured. In the above derivation no restriction has been placed on the directions of the incident neutrons.

B. Data Analysis and Correction Terms

Equation (8) is valid if we have a volume detector and neglect escape from the emulsion surface. If tracks are accepted sufficiently close to the surface to permit escape, correction factors must be added.

A method proposed by Evans³ is used which consists of exposing two emulsions perpendicular to each other so that tracks in the 4π solid angle are obtained without using steeply diving tracks. The solid angles not used in the emulsions are indicated by the shaded regions of the cones in Fig. 2.

The probability of tracks beginning in the central zone of the emulsion is determined and the track is given this probability weight, W_a .

Fig. 2. Track Orientation in Emulsions

If a cross section of the emulsion could be seen divided into three zones as shown in Fig. 3, then the probability weight of a given proton track in the emulsion would be

$$W_a = \frac{2 - |(K_1 - 2)| - |(K_2 - 2)|}{2} \quad (9)$$

with $K_1 = 1 + \frac{Z_1 - A}{DS}$

and $K_2 = 1 + \frac{Z_2 - A}{DS}$

where

S = shrinkage factor

D = thickness of zone = $66.67 \mu m$

A = distance of bottom of first zone from the origin of the Z -coordinate system.

Fig. 3. Emulsion Zones

Tracks having endpoints in various zones will, according to Eq. (9), have the corresponding weights indicated in Fig. 4.

ZONE	1	2	3
1	0	.5	0
2	.5	1	.5
3	0	.5	0

Fig. 4. Track Probability Weights

Some protons escape from the emulsion, and tracks which reach the emulsion surface are, therefore, of indeterminate length. Allowance is made for this by rejecting all tracks of this type and applying to each track a weighting factor, W_b , to compensate for the number of similar tracks rejected.⁴ This factor is the inverse probability of a track of that length and orientation not reaching the surface of the emulsion, i. e.

$$W_b = \frac{t}{t - R \sin \theta} \quad (10)$$

where R = track length, θ = angle to emulsion surface and t = emulsion thickness.

The proton spectrum $P(E)$ is obtained from the track-length distribution. The relation which infers the proton energy from the track length has been obtained from Powell's range-energy curves,⁵ which analytically can be represented by the expression

$$E = 1.056 - 2.7944 \log R + 3.8695 (\log R)^2 - 1.8021 (\log R)^3 + 0.4348 (\log R)^4 \quad (11)$$

where E = proton energy in MeV and R = track length in μm .⁶

A computer program was prepared which first orients the tracks and selects those tracks for analysis which fall within certain specified incident angles. Next the program calculates the range, energy, and weight of each track. The weights are then added to various energy boxes to yield a proton track distribution as a function of energy. See Appendices B-1, B-2, and B-3.

In order to determine $\frac{dP(E)}{dE}$, one must calculate the derivative of the proton track distribution. This derivative is used in Eq. (8) to yield the neutron flux as a function of energy. (See Appendix C).

III. DETERMINATION OF THEORETICAL NEUTRON SPECTRA

A. Assumptions

A model for determining neutron spectra in a moderating medium has been developed which is essentially a one scattering approach. It is assumed for shallow penetrations that on an average there will not be many more than one scattering such that the scattered neutron will still be going in an essentially forward direction. This is especially true if one considers neutrons only above 1 MeV and for penetrations of less than 5 cm in heavy water.

Other assumptions are:

- (1) Elastic collisions are solely responsible for slowing down of neutrons.
- (2) Nuclei of moderator are at rest in the laboratory system before collision.
- (3) Scattering is spherically symmetric in the center of mass coordinate systems.
- (4) Total cross sections as a function of energy are used in the proper proportion for deuterium and oxygen.
- (5) Effective interaction probability is proportional to distance from source to detector (i. e. ∞ plane source).

The fission neutron source is assumed to have a spectrum predicted by the equation,

$$\Phi_{fiss}(E) = .484 e^{-E} \sinh \sqrt{2E} \quad (12)$$

where E is neutron energy in MeV. The removed neutron flux as a function of energy and distance from the source is taken to be

$$\Phi_{\text{removed}}(E, r) = r N_o \Phi_{\text{fiss}}(E) \sum_1 \frac{\rho_i}{A_i} \sigma_i(E) \quad (13)$$

where r = source to detector distance in cm, $N_o = 6.02 \times 10^{23}$, ρ = density of moderator, A = gram atomic weight, and $\sigma(E)$ = total cross section as a function of energy.

Another flux is considered which is the contribution from the scattered neutrons, referred to as the buildup flux. This expression is given by

$$\Phi_{\text{buildup}}(E, r) = \int_E^{10 \text{ MeV}} \Phi_{\text{removed}}(E', r) dE' / \int_E^{10 \text{ MeV}} dE' \quad (14)$$

Thus, the total flux at a distance r from the source as a function of energy is given by the fission flux minus the removed flux plus the buildup flux, i. e.

$$\Phi(E, r) = \Phi_{\text{fiss}}(E) - \Phi_{\text{removed}}(E, r) + \Phi_{\text{buildup}}(E, r) \quad (15)$$

B. Results

The results of the theoretical calculations are shown in Appendix D of this report. In general, there is a shift of the maximum in the flux spectrum from about .8 MeV upward to about 2.4 MeV as one proceeds a few centimeters from the source. The main reason for the peaking of the flux in the theoretical spectrum is the large anti-resonance in the oxygen cross section at this energy.

IV. EXPERIMENT

A. Physical Properties of the Reactor Core

The Ames Laboratory Research Reactor is fueled with M.T.R. type fuel elements containing enriched Uranium-235. Positions are provided

for a maximum of 24 fuel elements. The reactor is moderated by heavy water which is maintained at a D_2 isotopic concentration of approximately 99.6%. The geometrical arrangement of the core is shown in Fig. 5.

The core configuration used for exposing the nuclear emulsions consisted of a 24 element core poisoned with 32.9 g of boron inserted in the fuel elements. This allowed the control rods to be almost completely withdrawn and also simulated an "end of life" core.

The nuclear emulsions placed in the center of the core of the reactor were separated from the nearest fission source of neutrons by approximately 2 cm of heavy water. The emulsions placed in beam port 9 were at the radial edge and on the center line of the core. These emulsions were about 5 cm from the nearest fission source and separated from it by heavy water.

B. Physical Properties of the Nuclear Emulsions

The emulsions used in this experiment are Ilford type K-2. The emulsions are $200\ \mu$ thick and are wrapped in black photographic paper. This package is contained within a 20 mil thick cadmium case which is used to screen out thermal neutrons (see Fig. 6). Since the range of interest is above .8 MeV, the low energy neutrons would only cloud the emulsion with unmeasurable dots. Because the emulsions are rather sensitive, the reactor was run at low power ($\sim .5$ watt) with a clean core to avoid over-exposure and clouding by gamma-ray tracks.

In order to perform the flux calculations, the hydrogen density in the emulsions is assumed to be 0.0534 g/ml of emulsion volume.⁷

Fig. 5. Reactor Core Cross Section

Fig. 6. Nuclear Emulsion Cutaway

The holders used for the emulsions are shown in Fig. 7. The holder on the left was used in the horizontal beam tube, and the holder on the right was used in the vertical thimble.

C. Development Technique for the Nuclear Emulsions

Developer - Kodak Developer D-19 mixed as directed and diluted by mixing 200 ml of the mixed developer with 1000 ml of distilled H_2O .

Stop Bath - 15 ml acetic acid plus 1000 ml of distilled H_2O .

Acid Fixer - Kodak acid fixer mixed as directed and used as is.

Procedure.

1. Soak the plates in cold distilled H_2O until thoroughly soaked. This means at a temperature between 6° and $9^\circ C$ for 50 to 60 min.
2. Soak the plates in cold developer until thoroughly soaked. This means at a temperature between 5° to $7^\circ C$ for 50 to 60 min.
3. Soak the plates in warm developer until fully developed. This means at a temperature around $9^\circ C$ for about 60 min.
4. Soak the plates in a stop bath. This is at a temperature around $15^\circ C$ for about 60 min.
5. Soak the plates in a $15^\circ C$ fixer solution for about 6 h.
6. Wash the plates overnight in running distilled H_2O .
7. Soak the plates in 10 ml of glycerine dissolved in 1000 ml of distilled H_2O for 50 min.
8. Soak the plates in 10 ml of glycerine mixed with 500 ml distilled H_2O and 500 ml absolute alcohol.
9. Soak the plates in 10 ml of glycerine mixed with 250 ml distilled H_2O and 750 ml absolute alcohol.
10. Dry the plates in air overnight.

Fig. 7. Emulsion Holders

V. RESULTS AND CONCLUSIONS

The nuclear emulsion analysis indicates that the neutron spectrum has its peak at 1 MeV in the center thimble. A peak at about 2.3 MeV is indicated for beam port 9. At higher energies, the spectra of both positions tend to follow more nearly the Watt fission spectrum characteristic of U-235.

Over 6000 proton recoil tracks were recorded for each spectrum obtained. A minimum of 5000 tracks should be counted to achieve acceptable statistics.²

In the range of interest (.8 to 5.0 MeV), oxygen has several resonances in its cross section between 1 and 2 MeV (see Appendix A). There is also a region of low cross section called an "anti-resonance" between 2 and 3 MeV. It would seem that both the experimental and theoretical spectra are characteristic of what one might expect from the cross sections of oxygen in this energy range.

The data for beam port 9 was analyzed by two different scanners (Appendix C). The peak in both cases is about 2.3 MeV. However, the abrupt low energy upturn of the spectrum obtained in the summer of 1965 is attributed to an error in scanning techniques for short track lengths. The spectrum obtained in the summer of 1966 was the result of improved scanning techniques of the same data.

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Appendix A. Cross Section Data

Appendix B. Proton Recoil Spectra

Appendix B. (Cont.)

Appendix B. (Cont.)

Appendix C. Experimental Neutron Spectra

Appendix D. Theoretical Neutron Spectra