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LOS ALAMOS SCIENTIFIC LABORATORY OF THE UNIVERSITY OF CALIFORNIA O LOS ALAMOS NEW MEXICO

LEAKAGE NEUTRON SPECTRUM OF U²³³ CRITICAL ASSEMBLY

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LEAKAGE NEUTRON SPECTRUM OF U²³³ CRITICAL ASSEMBLY

by

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ABSTRACT

The leakage neutron spectrum of a U^{233} spherical critical assembly (Jezebel) has been measured using nuclear emulsions as radiator and detector. The spectrum obtained is compared with similar measurements on the U^{235} and Pu^{239} analogues of Jezebel U^{233} ; it is found to agree closely with that of the Pu^{239} assembly and to be harder than that of the U^{235} assembly. All three spectra are compared with theoretical spectra obtained from a numerical approximation to the neutron transport equation.

I. INTRODUCTION

The use of photographic plates for determining neutron spectra is a well-established practice. Special nuclear emulsions are readily available for use on the plates; as a consequence of their small size, these emulsions alter the neutron flux at their location very slightly. With specially-equipped microscopes, the pertinent information concerning the recoil proton tracks is obtained after development of the plates. Highspeed computers render feasible the direct translation of this information into the energy of the incident neutron (provided its initial direction is known). From the output of the computer the incident flux can be easily obtained.

High-speed computers also make possible numerical solution of such otherwise-forbidding equations as the neutron transport equation. Leak-age spectra obtained from Carlson's S_n method¹ for this equation are compared with the experimental spectrum of the U²³³ assembly and earlier measurements of the spectra of Godiva (U²³⁵) and Jezebel (Pu²³⁹).

II. EXPERIMENTAL PROCEDURE AND ANALYSIS

Ilford K2 nuclear emulsions containing extra plasticizer were exposed to the neutron flux approximately 125 cm from the surface of the

 U^{233} critical assembly, which, to minimize scattering, had been taken out-of-doors and set up some 13 feet above the ground; the emulsions were positioned somewhat higher. Several runs at different integrated flux levels were performed to optimize track density for subsequent analysis. Background runs in which a polyethylene block 33.6-cm long shielded the source from the detectors were also made. After development of the emulsions, tracks displaying horizontal and dip angles $\leq 16^{\circ}$ with respect to the direction of the incident beam were examined. A small area was scanned analyzing all tracks $\geq 3\mu$; to decrease the statistical uncertainty at high energies without undue expenditure of time, a larger area of the plate was examined with only tracks longer than a set minimum being recorded. From measurements of the ranges and angles of the tracks, an IBM 704 converted each track to an equivalent neutron energy. Details of development and analysis procedures may be found in Reference 2. The neutron flux indicated by the background plates was subtracted from the observed spectrum; the correction was of the order of 3% below 1 MeV, 1% from 1 to 2 MeV, 0.8% from 2 to 3 MeV, and less than 0.5% at higher energies.

Let $F(E_n \pm \frac{1}{2} \Delta E_n)$ be the time-integrated flux of neutrons with energies between $E_n - \frac{1}{2} \Delta E_n$ and $E_n + \frac{1}{2} \Delta E_n$ which produces N_p proton recoils that are observed in the emulsion. Then

$$F(E_n \pm \frac{1}{2} \Delta E_n) = \frac{4\pi}{\Omega \sigma_{n-p}(E_n)n A t} N_p P(E_p) T(E_n),$$

where

 $F(E_n \pm \frac{1}{2} \Delta E_n) = number of neutrons/cm^2$ in the energy interval ΔE_n ;

- N_p = number of protons observed at energy E_p and direction specified by ψ , where for a given collision $E_n = E_p \sec^2 \psi$, ψ being the angle between the direction of the proton and that of the incident neutron in the laboratory frame;
- $P(E_p) = \text{correction for protons leaving the emulsion and hence}$ not being counted (plotted against E_n in Fig. 1*), where use is made of the approximate relation $E_n = E_p \overline{\cos^2 \psi}$, with $\overline{\cos^2 \psi}$ being the "average" value of $\cos^2 \psi$ over the solid angle considered;**
- $T(E_n)$ = correction for attenuation of the neutron beam in traversing the emulsion, $\simeq 1$ since analysis was confined to a strip near the edge (0.2 to 0.5 cm);
- t = emulsion thickness before processing, cm;
- A = area of emulsion analyzed, in cm^2 ;
- n = number of hydrogen atoms/cm³ = 3.43 x 10^{22} , corresponding to a density of 0.057 gm/cm³;

 $\sigma_{n-p}(E_n) = n-p$ total scattering cross section at energy E_n (assumed isotropic in the center-of-mass system); $\Omega =$ solid angle of acceptance of tracks in the unprocessed

emulsion, but in the center of mass (see Appendix).

* Based on the experimental determination of Rosen, (Reference 2). ** See Rosen, Reference 2, for the evaluation of $\cos^2 \psi$.



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Fig. 1. Probability Correction P(E) versus Neutron Energy (angle criterion--16°)

After the analysis described above was performed using $\Delta E_n = 0.1$ MeV, the data were grouped into larger intervals of energy (to improve statistics) and smoothed using an average weighted as 1:2:2:1 for $E_n < 3.2$ and as 1:2:3:3:2:1 for higher energies, with the statistics weighted accordingly. Such smoothing is justified since the errors involved in determining the range and angles of a track are large enough to prevent accurate assignment to a 0.1 MeV interval.

The factor most limiting of accuracy in photographic-plate studies is delineation of the solid angle of acceptance of proton recoils. This is especially true for short tracks because of the difficulty in determining the dip and horizontal angle of such a track. The following estimates³ of the non-statistical errors encountered are probably not unreasonable:

$$σ_{n-p} = 2\%$$
 $P(E_p) = 3\%$
 $T(E_n) = 3\%$
 $n = 5\%$
 $Ω = 13\%$
 $t = 3\%$

The root-mean-square error is thus 15%. Of these errors, those in n and t do not affect the relative distribution of neutrons, and the error in Ω must in this connection be reckoned less than 13%. A reasonable estimate of the precision with which the relative neutron distribution was obtained is hence 10%.

III. RESULTS

The leakage neutron spectrum obtained for the U^{233} assembly is tabulated in Table I with statistical errors and plotted in Fig. 2. The leakage spectra of the similar critical assemblies Godiva (U^{235}) and Jezebel (Pu^{239}) have also been measured⁴ using a similar experimental arrangement; these spectra are tabulated in Tables II and III, where the errors shown are the result of counting statistics only. For Fig. 3 the three spectra have been normalized so that the area under each curve from 0.3 to 9.5 MeV is the same. The figure shows (see also Table IV) that the leakage spectra of the U^{233} and Pu^{239} assemblies are quite similar and that both are harder than that of the U^{235} assembly, observations which are in keeping with the known cross sections and fission spectra of these elements.

The discontinuity at 1.1 MeV is of questionable significance since the statistical uncertainties are fairly large in this region. Note, however, that there is an indication of such a discontinuity at the same energy in the Pu^{239} spectrum. Possibly some resonance phenomenon is indicated.

The average energy of the leakage neutrons from each of the three assemblies can be easily calculated. Using 0.3 MeV as a lower energy limit, the results are as follows:

Neutron Energy Interval, MeV	Neutrons/cm ² per MeV	Statistical Uncertainty, %	Neutron Energy Interval, MeV	Neutrons/cm ² per MeV	Statistical Uncertainty,
0.3 - 0.4 0.4 - 0.6 0.6 - 0.8 0.8 - 1.0 1.0 - 1.2 1.2 - 1.4 1.4 - 1.6 1.6 - 1.8 1.8 - 2.0	$646 \times 106520 \times 106391 \times 106306 \times 106324 \times 106299 \times 106230 \times 106209 \times 106182 \times 106$	11 10 7 7 7 8 8 8 10 11 10	3.2 - 3.6 $3.6 - 4.0$ $4.0 - 4.4$ $4.4 - 4.8$ $4.8 - 5.2$ $5.2 - 5.6$ $5.6 - 6.0$ $6.0 - 6.4$ $6.4 - 6.8$ $6.8 - 7.2$	94.3 \times 106 83.6 \times 106 57.9 \times 106 41.6 \times 106 42.6 \times 10 32.3 \times 106 23.5 \times 106 14.3 \times 106 13.6 \times 106 6.0 \times 10	9 10 12 14 15 15 22 30 30 30 50
2.0 - 2.2 2.2 - 2.4 2.4 - 2.6 2.6 - 2.8 2.8 - 3.0 3.0 - 3.2	166×10^{6} 157×10^{6} 148×10^{6} 138×10^{6} 119×10^{6} 108×10^{6}	10 10 10 10 11 12	7.2 - 7.6 7.6 - 8.0 8.0 - 8.4 8.4 - 8.8 8.8 - 9.2 9.2 - 9.6	$8.4 \times 106 4.4 \times 106 1.7 \times 106 5.2 \times 106 2.5 \times 106 3.1 \times 10^6$	40 60 100 60 90 80

leakage neutron spectrum of jezebel u^{233}

TABLE I



Fig. 2. Leakage neutron spectrum of Jezebel U^{233}

Neutron Energy Interval, MeV	Neutrons/cm ² per MeV	Statistical Uncertainty, %	Neutron Energy Interval, MeV	Neutrons/cm ² per MeV	Statistical Uncertainty, %
0.2 - 0.4 $0.4 - 0.6$ $0.6 - 0.8$ $0.8 - 1.0$ $1.0 - 1.2$ $1.2 - 1.4$ $1.4 - 1.6$ $1.6 - 1.8$ $1.8 - 2.0$ $2.0 - 2.2$ $2.2 - 2.4$ $2.4 - 2.6$ $2.6 - 2.8$ $2.8 - 3.0$	721 x 10^{5} 718 x 10^{5} 612 x 10^{5} 499 x 10^{5} 371 x 10^{5} 320 x 10^{5} 257 x 10^{5} 218 x 10^{5} 224 x 10^{5} 193 x 10^{5} 159 x 10^{5} 141 x 10^{5} 122 x 10^{5} 136 x 10^{5}	4 3 4 4 5 6 7 8 8 7 8 8 7 8 9 9 9 9	3.0 - 3.4 3.4 - 3.8 3.8 - 4.2 4.2 - 4.6 4.6 - 5.0 5.0 - 5.4 5.4 - 5.8 5.8 - 6.2 6.2 - 6.6 6.6 - 7.0 7.0 - 7.4 7.4 - 7.8 7.8 - 8.2 8.2 - 8.6 8.6 - 9.0	84.6×10^{5} 70.5×105 62.1×105 40.7×105 42.9×10^{5} 12.9×10^{5} 12.9×10^{5} 12.9×10^{5} 10.8×10^{5} 11.4×10^{5} 13.9×10^{5} 1.9×10^{5} 2.2×10^{5} 6.6×10^{5} 2.4×10^{5}	9 8 9 11 11 14 16 24 27 27 27 27 25 70 70 40 70

leakage neutron spectrum of godiva u^{235^a}

TABLE II

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^aTable from Reference 4.

TABLE	III
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LEAKAGE NEUTRON SPECTRUM OF JEZEBEL Pu^{239^a}

Neutron Energy	Neutrons/cm ²	Neutron Energy	Neutrons/cm ²
Interval, MeV	per MeV	Interval, MeV	per MeV
0.3 - 0.5 0.5 - 0.7 0.7 - 0.9 0.9 - 1.1 1.1 - 1.3 1.3 - 1.5 1.5 - 1.7	$\begin{array}{rrrrrrrrrrrrrrrrrrrrrrrrrrrrrrrrrrrr$	3.5 - 3.9 3.9 - 4.3 4.3 - 4.7 4.7 - 5.1 5.1 - 5.5 5.5 - 5.9 5.9 - 6.3 6.7	$50.3 \pm 5.2 \times 10^{6}$ $44.0 \pm 5.0 \times 10^{6}$ $33.9 \pm 4.5 \times 10^{6}$ $28.8 \pm 4.3 \times 10^{6}$ $16.6 \pm 3.3 \times 10^{6}$ $16.5 \pm 3.4 \times 10^{6}$ $8.2 \pm 2.5 \times 10^{6}$ 10^{6}
1.7 - 1.9 1.9 - 2.1 2.1 - 2.3	$\begin{array}{rrrrrrrrrrrrrrrrrrrrrrrrrrrrrrrrrrrr$	6.3 - 6.7 6.7 - 7.1 7.1 - 7.5	$\begin{array}{r} 11.2 \pm 3.0 \times 106 \\ 9.7 \pm 2.9 \times 106 \\ 4.7 \pm 2.1 \times 106 \end{array}$
2.3 - 2.5	$\begin{array}{rrrrrrrrrrrrrrrrrrrrrrrrrrrrrrrrrrrr$	7.5 - 7.9	$4.3 \pm 2.1 \times 106$
2.5 - 2.7		7.9 - 8.3	$2.1 \pm 1.5 \times 106$
2.7 - 3.1		8.3 - 8.7	$4.9 \pm 2.5 \times 106$
3.1 - 3.5		8.7 - 9.1	$1.2 \pm 1.2 \times 10$

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^aTable from Reference 3.





IV. S APPROXIMATION TO THE LEAKAGE SPECTRA

The neutron transport equation and its reduction to the S_n form are discussed in two reports by Carlson.^{1,5} Using Carlson's program in the S_8 approximation with 18 intervals of radius, Hansen⁶ has calculated the leakage spectra of several Pajarito assemblies, the cross sections employed being those in the report of Hansen and Roach.⁷

Among the output of the computer is the relative neutron leakage flux in each energy interval used. For comparison with the experimentally determined spectrum, the theoretical leakage spectrum is normalized so that

$$\sum_{g=3}^{6} N_g = 1,$$

and then N_g , the neutron flux in energy group g, is divided by ΔE_g , the width of the gth group in MeV. Here the energy groups are as follows:

Group	Energy, MeV
l	0-0.1
2	0.1-0.4
3	0.4-0.9
4	0.9-1.4
5	1.4-3.0
6	3.0-∞

(For group 6, ΔE_g is taken as 6.5 MeV; i.e., it is assumed that the contribution of neutrons with energies greater than 9.5 MeV is negligible.) This normalization yields neutrons cm^{-2}/MeV . The experimental spectrum is lumped into the same four groups in a similar manner, where now

$$N_{g} = \int_{\Delta E_{g}} F(E_{n}) dE_{n} / \int_{0.4 \text{ MeV}}^{9.5 \text{ MeV}} F(E_{n}) dE_{n}.$$

These quantities are displayed, along with the ratio of the calculated and measured quantities, in Table IV. Included in this table are the same quantities for the critical assemblies Jezebel Pu^{239} and Godiva U^{235} . The root-mean-square deviation of the theoretical from the experimental flux is 0.09 for Jezebel Pu^{239} , 0.08 for Jezebel U^{233} , and 0.03⁴ for Godiva U^{235} . In no case is a deviation sufficiently large to warrant condemnation of the cross sections, etc., employed in the S_n approximation.

TABLE IV

COMPARISON OF THEORETICAL AND EXPERIMENTAL LEAKAGE SPECTRA

	0.4-0.9 MeV	0.9-1.4 MeV	1.4-3.0 MeV	3.₀0-∞ MeV
Jezebel Pu ²³⁹				
Theoretical	0.481	0.390	0.216	0.034
Experimental	0.488	0.342	0.211	0.038
Ratio ^a	0.99	1.14	1.02	0.89
Jezebel U ²³³				
Theoretical	0.513	0.384	0.218	0.031
Experimental	0.510	0.372	0.203	0.036
Ratio ^a	1.01	1.03	1.07	0.86
Godiva U ²³⁵				
Theoretical	0.634	0.392	0.197	0.026
Experimental	0.658	0.392	0.189	0.027
Ratio ^a	0.96	1.00	1.04	0.96

(Neutrons cm⁻² MeV⁻¹)

^aTheoretical flux/Experimental flux

APPENDIX

CALCULATION OF Ω

Elementary considerations of collisions between equally massive particles yield the results,

$$d^{2}\Omega_{\text{LAB}} \equiv d^{2}\Theta = \sin \theta \, d\theta \, d\phi,$$
$$d^{2}\Omega_{\text{CM}} \equiv d^{2}\Omega = 4 \cos \theta \, d^{2}\Theta = 4 \cos \theta \sin \theta \, d\theta \, d\phi,$$

where all angles are measured in the laboratory reference frame. In general $z = \rho \cos \theta$, $x = \rho \sin \theta \cos \phi$, whence

$$\frac{x}{z} = \tan \theta \cos \phi.$$

It follows that along edge AB of Fig. 4, $\tan \theta \cos \phi = \frac{x}{z} = \tan \alpha$, where α is the maximum permissible value of the horizontal or dip angle (assumed equal in this analysis); i.e., α is the half angle of the square-based pyramid of acceptance. Thus

$$\theta = \tan^{-1} \left(\frac{\tan \alpha}{\cos \phi} \right)$$

along edge AB. Since the domain of integration in ϕ can be divided into 8 congruent segments,

$$\Omega = 8 \int_{0}^{\pi/4} \int_{0}^{\tan^{-1}} \left(\frac{\tan \alpha}{\cos \phi}\right)$$
$$\Omega = 8 \int_{0}^{\pi/4} \int_{0}^{\tan^{-1}} \left(\frac{\tan \alpha}{\cos \phi}\right)$$



Fig. 4. Illustration of the pyramid of acceptance

$$= 32 \int_{0}^{\pi/4} \int_{0}^{\cos^{-1}} \left(\frac{\cos^{-\phi}}{\sqrt{\cos^{2\phi} + \tan^{2}\alpha}} \right)$$
$$= 16 \tan^{2\alpha} \int_{0}^{\pi/4} \frac{d\phi}{\tan^{2\alpha} + \cos^{2\phi}}.$$

:

Evaluating the final integral,

$$\Omega = 16 \sin \alpha \tan^{-1} (\sin \alpha).$$

For the case at hand $\alpha = 16^{\circ}$ and $\Omega = 1.186$ steradians.

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