

FAST NEUTRON LEAKAGE SPECTRA FOR ASSEMBLIES
CONTAINING THORIUM, D₂O AND NATURAL URANIUM

by

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Dissertation submitted to the Graduate Faculty of the
Virginia Polytechnic Institute and State University
in partial fulfillment of the requirements for the degree of
DOCTOR OF PHILOSOPHY
in
Physics

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November, 1973
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ACKNOWLEDGMENTS

A particular debt of gratitude is due to Dr. Andrew Robeson who conceived of this research project and guided it to its completion.

Thanks are due to Dr. Dale Long who assisted with the Van de Graaff accelerator runs. The author is indebted to Dr. [redacted] for helping with the spectrometer calibration and for several fruitful discussions pertaining to the physical nature of the spectrum unfolding procedure.

The [redacted] of the [redacted] National Laboratory provided the ANISN transport code and the DLC-2 cross-section library, and provided advice in their application.

The assistance and cooperation of the [redacted] reactor operations staff and [redacted] support personnel is gratefully acknowledged.

TABLE OF CONTENTS

<u>CHAPTER</u>	<u>PAGE</u>
I. INTRODUCTION	1
II. LITERATURE REVIEW	5
III. EXPERIMENTAL TECHNIQUES	10
A. The ³ He Spectrometer	10
B. Fission and Dummy Sources	14
C. The Experimental Assemblies.	16
1. Thorium/Heavy Water Lattices	16
2. Natural Uranium Assembly	18
3. Heavy Water Sphere	19
4. Measurements in Heavy Water and Thorium Nitrate Cylinders.	22
IV. THE CALCULATIONAL METHOD	25
A. The General Technique.	26
1. Multigroup Cross-Sections	26
2. Cross-Section Data Input and Codes	27
3. Multigroup Neutron Transport Calculations.	28
B. The Transport Code.	34
C. Details of the Calculations.	38
D. Test of the Transport Code Application	39
V. COMPARISON BETWEEN CALCULATION AND EXPERIMENT	44
VI. CONCLUSIONS	50

TABLE OF CONTENTS

(Continued)

<u>CHAPTER</u>	<u>PAGE</u>
VII. BIBLIOGRAPHY	52
VIII. VITA.	56

LIST OF TABLES

<u>TABLE</u>		<u>PAGE</u>
1.	Source Description	15
2.	Thorium/Heavy Water Lattice Descriptions.	16
3.	Broad Energy Group Boundaries for the Transport Calculations.	40

LIST OF FIGURES

<u>FIGURE</u>	<u>PAGE</u>
1. Experimental Arrangement and Neutron Spectrometer	11
2. Spectrometer Calibration	13
3. Arrangement of the Thorium/D ₂ O Assemblies	17
4. Arrangement of the Natural Uranium Assembly	20
5. D ₂ O Sphere	21
6. The Multigroup Transport Code Calculation	36
7. Central Neutron Spectrum of JAERI I-5 Fast Assembly	42
8. Neutron Leakage Spectrum for JAERI I-5 Fast Assembly	43
9. Neutron Leakage Spectra for Thorium/D ₂ O Lattice of Pitch = 3.97 cm (1 - 9/16-inch).	46
10. Neutron Leakage Spectra for Thorium/D ₂ O Lattice of Pitch = 4.45 cm (1 - 3/4-inch)	47
11. Neutron Leakage Spectra for Natural Uranium Assembly	48
12. Neutron Leakage Spectra for Heavy Water Sphere.	49

I. INTRODUCTION

An accurate knowledge of microscopic cross-section data for neutron behavior in matter is an essential requirement for the design of a nuclear reactor or for the analysis of a nuclear assembly. Basic neutron cross-sections for materials of importance to reactor physics have in many cases, been measured by differential techniques. Neutron cross-sections may be determined in the differential method from the attenuation or scattering of neutrons in a beam interacting with a target, or from the intensity of the secondary radiation produced.

Data resulting from the many differential techniques are not always in agreement, and a consistent set of cross-sections to describe a real nuclear system may be difficult to obtain. To obtain more consistent cross-sections, the integral technique of testing cross-section data is frequently employed. Integral methods for determining cross-sections are based on overall neutron interactions in a medium, including distribution in space, energy and time. In integral experiments, the effects of the cross-sections on the neutron behavior are measured, and calculational methods using existing cross-section data files are then employed to form the basis of a comparison. The advantage of the integral testing method is that it affords an opportunity to interpret the agreements (or disagreements) between experiment and calculations in terms of specific cross-section adequacies (or deficiencies).

The measurements reported in this dissertation were conducted to provide integral measurement tests of the validity of the differential data of the ENDF/B-II files for thorium, natural uranium, and heavy water. The experimental results were compared with the results obtained by using the ENDF/B-II data as basic data in a multigroup neutron transport computer code calculation.

The ^3He proportional counter has been used as a neutron spectrometer for spectrum measurements in fast reactors and assemblies^{1,2}. While the ^3He counter has several advantages as a neutron spectrometer, most of its past use has been as a neutron detector. The cost of ^3He limited its use until recent years. Natural helium contains ^3He with an abundance of about $1.3 \times 10^{-4}\%$. The ^3He isotope has, however, now become available by irradiation of ^6Li in reactors³. The resulting tritium decays with a half-life of 12.3 years to the daughter product ^3He .

The ^3He proportional counter has a relatively large $^3\text{He} (n,p) ^3\text{H}$ reaction cross-section which varies smoothly with neutron energy. The large Q-value of this reaction ($Q=764$ keV) permits discrimination of the pulses due to neutrons from those due to gamma rays.

The recorded pulse-height spectrum for a ^3He spectrometer is due to the deposition of energy from all possible charged particles from several reactions and their wall effects in the detector. The wall effects are due to the cutting short of the tracks of the charged particles of these reactions by the counter wall. These effects depend on the range of the ionizing track and the location of the event in the

counter. The energy dependent reactions are as follows:

- a. ${}^3\text{He} (n,p){}^3\text{H}$ for neutrons of all energies.
- b. ${}^3\text{He} (n,n){}^3\text{He}$ for neutrons of all energies.
- c. ${}^3\text{He} (n,d){}^2\text{H}$ for neutrons of energy ≥ 4.36 MeV.
- d. ${}^3\text{He} (n,p,n){}^2\text{H}$ for neutrons of energy ≥ 7.32 MeV.
- e. ${}^3\text{He} (n,p,2n){}^1\text{H}$ for neutrons of energy ≥ 10.3 MeV.

For most reactor applications, the contributions of the (n,d), the (n,p,n), and the (n,p,2n) reactions are negligible because of the high threshold energies and small cross-sections.

Analytical procedures have been developed to determine the neutron spectrum from the pulse-height distribution^{2,4,5}. These procedures utilize digital computer routines to unfold the pulse-height spectrum. The effects of the pulses due to ${}^3\text{He}$ recoils and due to the wall effects for the (n,p) and (n,n) reactions are computed and then eliminated. The actual or true neutron energy spectrum is defined as the spectrum recorded by the proton and triton induced pulse-heights, without wall effects, in the ${}^3\text{He} (n,p) {}^3\text{H}$ reaction.

Certain characteristics of the ${}^3\text{He}$ proportional counter need be considered for neutron spectrometer applications. The large relative value of the ${}^3\text{He} (n,p){}^3\text{H}$ cross-section for thermal neutrons (5460 barns at 0.0253 eV) complicates spectral measurements of systems with large thermal neutron backgrounds. In fast neutron experiments, neutrons may be degraded by scattering from the walls and floor of the room to epithermal and thermal energies. These room-return neutrons react

very rapidly with ^3He , liberate an appreciable energy in the detector, and produce a background which must be considered. The background effect and uncertainties in the values of the energy dependent (n,p) reaction cross-sections limit the reliability of the ^3He spectrometer for neutron energy measurements below about 100 keV^{6,7}. T. Iijima et. al.² note that the cross-sections of the (n,p) reaction strongly influences the resultant spectrum. They report² typical uncertainties in the values of the (n,p) reaction as 1% at 1 keV neutron energy, 3% at 10 keV, and 10% at 100 keV.

The ^3He spectrometer has been applied to fast neutron leakage spectra measurements for assemblies containing thorium metal, natural uranium metal, heavy water and thorium nitrate. The experimental results are compared to the results of multigroup transport calculations obtained with the discrete ordinates code, ANISN⁸.

II. LITERATURE REVIEW

A number of recent publications have cited the results of neutron physics measurements and neutron transport calculations for spectra in various materials and assemblies. References¹ and⁹ through²⁰ are representative of recent efforts in this area. Neutron energies in these fast - neutron spectra measurements are determined by several techniques. Time - of - flight, threshold and resonance foils, and neutron energy - sensitive detectors are employed.

The ^3He proportional counter has been used as a neutron spectrometer for spectrum measurements in fast reactors and subcritical assemblies^{1, 22}. In addition, it has been used in other neutron physics measurements requiring a neutron energy - sensitive spectrometer. A. Naeem⁵ and S. Shalev²¹ have employed the ^3He proportional counter as a neutron spectrometer for the measurement of delayed neutrons from fast fission. Y. Segal et. al.²² have reported the results of measurements of the albedo for fast neutrons from plane scatterers using a ^3He spectrometer. A Van de Graaff accelerator using the $^3\text{H}(p,n)^3\text{He}$ reaction was the neutron source. A tritium target was used with the accelerator.

Experimental nuclear data, modern computers, and transport codes using numerical procedures make possible a relatively rigorous solution of the transport equation for the prediction of fast neutron spectra. Practical considerations of storage and running time in computer solutions lead to approximations in coding procedures and in

the input data which should be justified by comparison with experimental results. In general, neutron spectra for simple systems can be computed by inserting into the Boltzmann transport equation appropriate cross-section data with the geometrical arrangement of the materials.

Generally, solutions of the equation require sophisticated numerical methods with a digital computer. One such code developed to provide a numerical solution of the Boltzmann equation is the one - dimensional discrete S_n transport code ANISN⁸. The ANISN code accepts microscopic neutron cross-section data of the well known ENDF/B data set catalogued at the Brookhaven National Laboratory. Cross-section data preparation codes^{23, 24, 25} have been developed at the Oak Ridge National Laboratory which provide a cross-section library in data format appropriate for input to the ANISN code with optional data storage configurations to prevent potential problems with computer core allocations.

The time independent form of the Boltzmann Equation describes the steady-state behavior of the neutron population in the assemblies of this investigation. This equation is derived in detail in several excellent texts such as those by Weinberg and Wigner²⁶, Meghreblian and Holmes²⁷, Ferziger and Zweifel²⁸, and Bell and Glasstone²⁹. This equation is:

$$\underline{\Omega} \cdot \nabla \phi(\underline{r}, E, \underline{\Omega}) + \sum_T(\underline{r}, E) \phi(\underline{r}, E, \underline{\Omega}) = \quad (1)$$

$$S(\underline{r}, E, \underline{\Omega}) + \int dE' \int d\Omega' \Sigma(\underline{r}, E' \rightarrow E, \underline{\Omega}' \rightarrow \underline{\Omega}) \phi(\underline{r}, E', \underline{\Omega}')$$

where:

$\Sigma_T(\underline{r}, E) = \Sigma_a(\underline{r}, E) + \Sigma_s(\underline{r}, E)$ = the energy dependent total cross-section at point \underline{r} . Σ_a represents the macroscopic absorption cross-section and Σ_s represents the macroscopic scattering cross-section.

$\Sigma(\underline{r}, E' \rightarrow E, \underline{\Omega}' \rightarrow \underline{\Omega})$ = represents the transfer cross-section, and denotes the scattering kernel whereby a neutron at position \underline{r} scatters from energy E' while travelling in direction $\underline{\Omega}'$ to an energy between E and $E + dE$ and direction $d\underline{\Omega}$ about $\underline{\Omega}$ after collision.

$S(\underline{r}, E, \underline{\Omega})$ = characterizes the neutron source distribution.

$\underline{\Omega}$ denotes a unit vector specifying the direction of motion of the neutron. It is defined by the polar angle θ and the azimuthal angle ϕ . The Cartesian components of $\underline{\Omega}$ are $\Omega_x = \sin\theta \cos\phi$, $\Omega_y = \sin\theta \sin\phi$, and $\Omega_z = \cos\theta$. The element of solid angle, $d\Omega$, is the surface element on the unit sphere normal to $\underline{\Omega}$, i.e., $d\Omega = \sin\theta \, d\theta \, d\phi$

$\phi(\underline{r}, E, \underline{\Omega})$

is the time - independent angular neutron flux defined such that the number of neutrons in a volume element d^3r at \underline{r} with energies between E and $E + dE$ and with velocity vectors in the solid angle element about the unit vector

$\underline{\Omega}$ is given by

$$n(\underline{r}, E, \underline{\Omega}) d^3r dE d\Omega = \frac{1}{v} \phi(\underline{r}, E, \underline{\Omega}) d^3r dE d\Omega$$

where v is the neutron speed.

In principle this neutron distribution problem could be solved by inserting into the transport equation a proper set of the appropriate cross-sections together with the geometrical arrangement of the materials in the experimental assembly. However, the transport equation is so intractable that solution is not possible by analytical means except in the simplest of geometrical cases. For one thing, energy dependent cross-sections are not completely known, and, for another, the geometrical arrangement of the materials in most assemblies prohibits solution of the transport equation in a reasonable time even with a computer. However, in many cases solutions of the transport equation can be obtained by numerical methods suitable for computer programming.

The use of one method for solution of the transport equation known as the method of discrete ordinates was suggested by G. C. Wick³⁰ in 1943. More extensive development of the method was carried out by S. Chandrasekhar^{31, 32, 33}. Further refinements to this method were later proposed by B. G. Carlson³⁴ in his Sn method. Development of this Sn method led to reducing the transport equation to a system of coupled differential equations suitable for solution by difference techniques on a digital computer.

The details of the Sn method, formulation of the difference equation, and numerical procedures are given in the references by B. G. Carlson^{34, 35}. C. E. Lee³⁶ has presented a detailed investigation describing the Sn method for solution of the multigroup two - and three - dimensional transport equations.

III. EXPERIMENTAL TECHNIQUES

All the measurements reported here have been made using the UTR-100 research reactor of the Virginia Polytechnic Institute and State University (V.P.I. & S.U.). The experimental setup with a sphere of D_2O is shown in Figure 1. Each assembly was placed in front of a collimated thermal neutron beam from the V.P.I. nuclear reactor. With collimators and shielding in place the flux at the center of the experimental assembly was 3.4×10^6 n/(cm² sec), with a cadmium ratio (total activity/epicadmium activity) of 6.0.

The collimated neutron beam was allowed to impinge on a small target. In each spectrum measurement, except those for the thorium nitrate cylinder and the heavy water cylinder, the target was located at the center of the experimental assembly. For the thorium nitrate and heavy water cylinders, the targets were placed on the surface of each container. Two types of targets, ^{235}U fission and dummy (mock) sources, were employed. Details of these targets are given in a separate section. Measurements have been made in a number of assemblies and these are described in subsequent sections.

A. THE 3He SPECTROMETER.

Fig. 1 includes a block diagram of the fast neutron spectrometer used in all experiments. A 6-atmosphere 3He proportional counter (Texas Nuclear Model 9321) was used. The pulses from the 3He counter were amplified and fed to a multichannel analyzer where the pulses were stored in 128

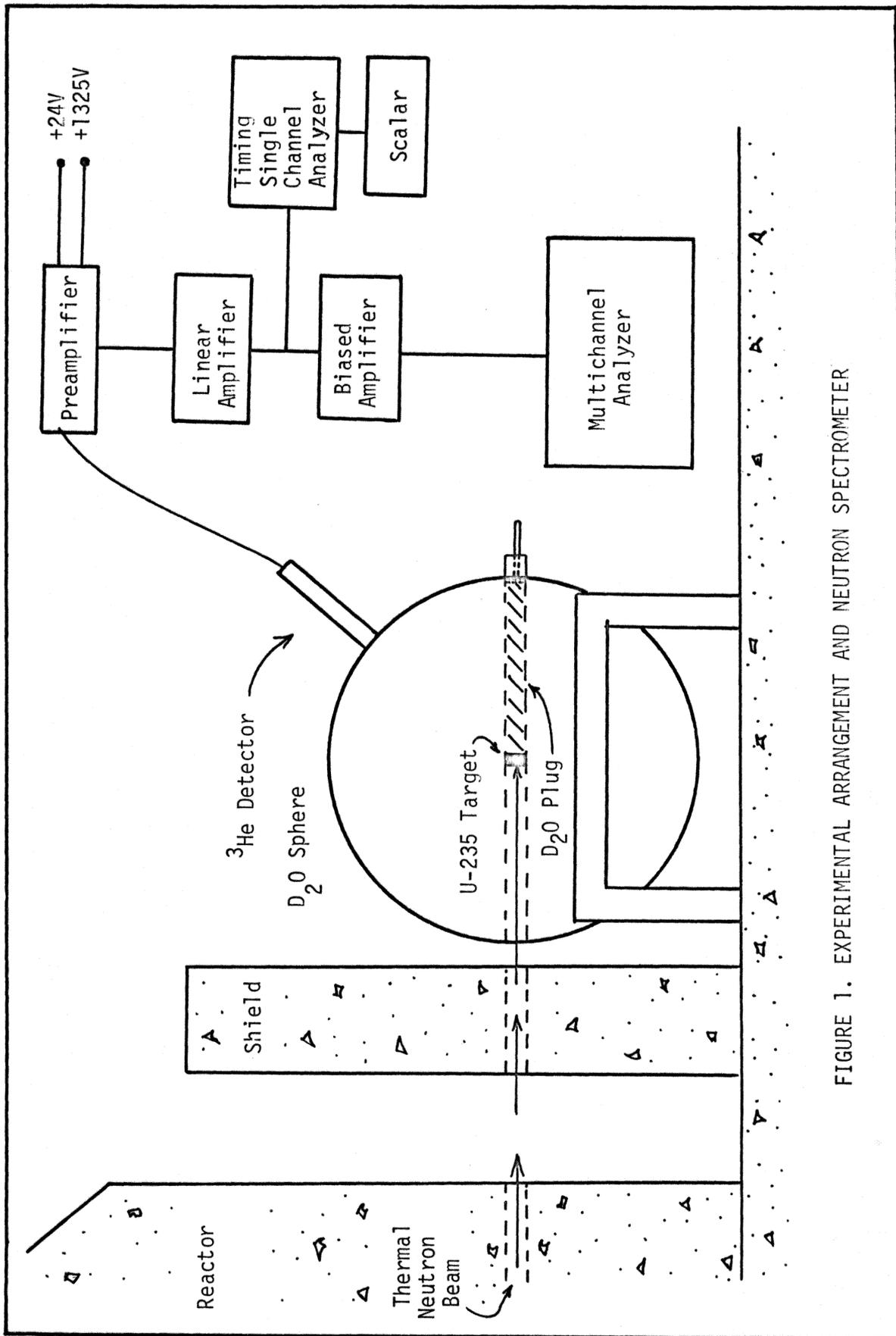


FIGURE 1. EXPERIMENTAL ARRANGEMENT AND NEUTRON SPECTROMETER

channels.

The spectrometer was calibrated with monoenergetic neutrons produced in the $^3\text{H}(\text{p},\text{n})^3\text{He}$ reaction with the V.P.I. Van de Graaff accelerator. Fig. 2 represents the calibration for neutron energy versus channel number. The calibration curve was quite linear. The measured resolution for this spectrometer was 24% for neutrons of 0.995 MeV, based on an energy increment of 13.5 keV per channel. The resolution is defined as the ratio of the full width of the peak at half maximum to the position of the peak midpoint, expressed in percent. The detail shown in fast neutron spectra is dependent on the resolution of the spectrometer.

Certain simplifying assumptions were made when unfolding the detector response for modelling the assembly and detector positioning. In the efficiency calculation for the ^3He detector response, standard shielding techniques, as described by T. Rockwell³⁷, were utilized. For the efficiency calculation, each assembly was modeled as a homogeneous spherical source of equivalent volume. The spherical source was then described in the efficiency calculation as a disk source of the same radius as the sphere. Self absorption distances were found using the empirical graphs in Reference 9 to give the relative source - detector positioning in the efficiency calculation for the spectrum unfolding code⁵.

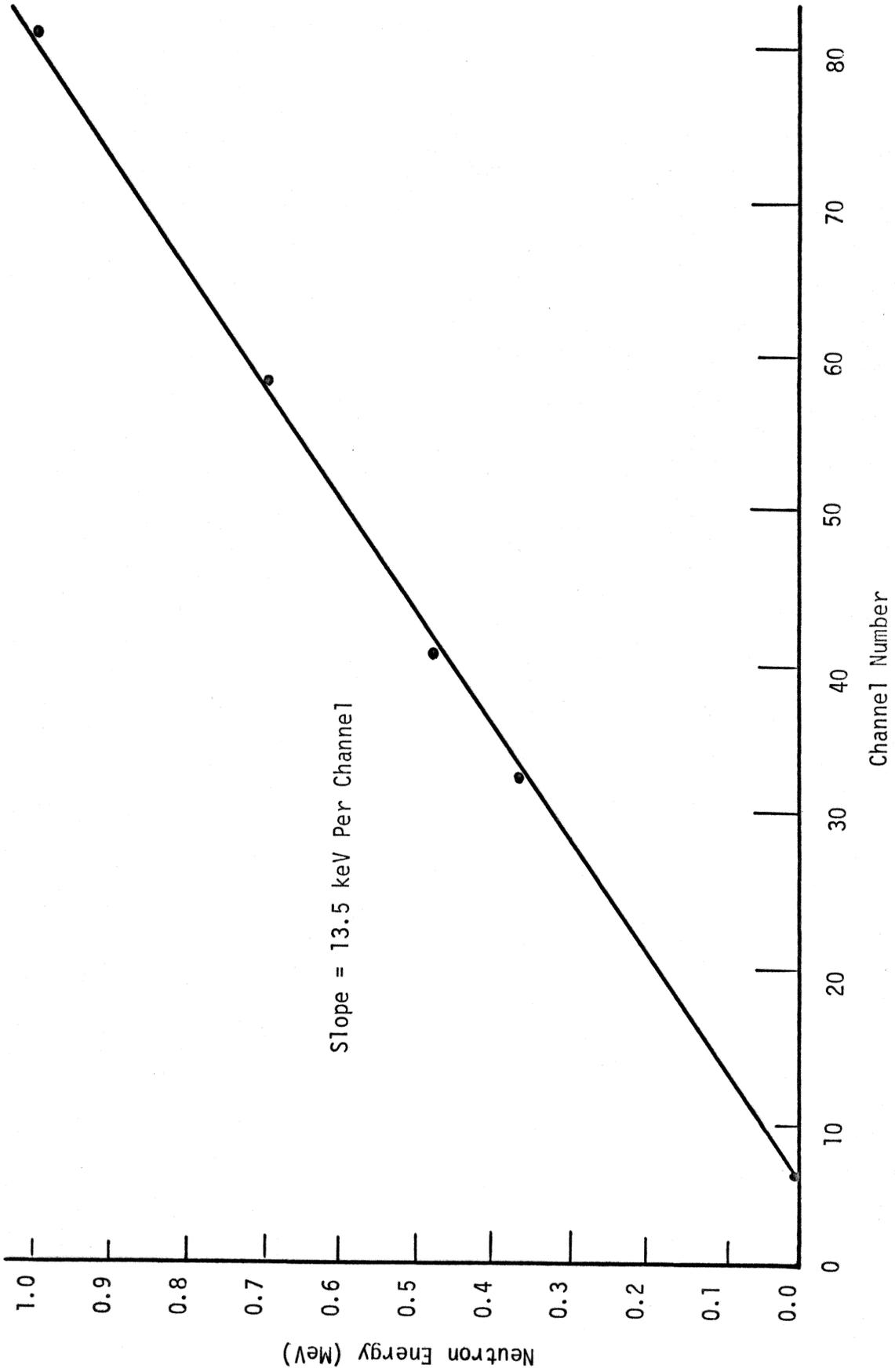


FIGURE 2. SPECTROMETER CALIBRATION -- NEUTRON ENERGY VERSUS CHANNEL NUMBER

Rockwell³⁷ concludes that maximum errors will not exceed -5 percent and +15 percent if the point where the flux is to be calculated is at a distance from the surface of the sphere equal to, or greater than, the radius of the sphere.

B. FISSION AND DUMMY SOURCES

The determination of leakage spectra in any one assembly involved two sets of measurements, one with a ^{235}U fission source and the other with a dummy source. Primary reactor epithermal neutrons may be scattered by the ^{235}U target or captured in its resonances. Background measurements with a dummy source were made for reactor produced epithermal neutrons that penetrate the assembly and for scattered gamma-ray components of the beam from the reactor.

Two different sized ^{235}U fission disks were employed with physical dimensions appropriate to the source tube size of the assembly under investigation. Dummy targets of both lead and lutetium - oxide were used. Lead has scattering properties similar to uranium and lutetium has a resonance structure similar to that of ^{235}U . D. M. Roberts and W. G. Pettus³⁸ of Babcock and Wilcox have used lutetium for a mock source because of its resonance structure. No detectable difference attributable to choice of dummy source was found in this investigation. Table 1 gives details of the two fission sources and the two dummy sources.

TABLE 1
SOURCE DESCRIPTIONS

TARGET TYPE	MATERIAL	DETAILS
Small fission disk	5 grams of ^{235}U Metal	25 one-mil foils, 0.90 inch diameter, and encapsulated in a thin walled cylindrical aluminum box.
Large fission disk	15 grams of ^{235}U in U_3O_8 form	U_3O_8 pressed into pellet form and encapsulated in a thin walled nickel container, 1.5 inch in diameter.
Small mock source	4.3 grams of Lu_2O_3	Lu_2O_3 encapsulated in a thin walled nickel container, 1.0 inch in diameter.
Large mock source	15 grams of Pb	Bare lead disk, 1.5 inch in diameter.

C. THE EXPERIMENTAL ASSEMBLIES

1 - THORIUM/HEAVY WATER LATTICES

Figure 3 gives a schematic representation for the thorium/heavy water assemblies. The leakage spectra of fast neutrons on two different lattices of thorium and heavy water was measured. The assemblies were composed of thorium slugs arranged in lattices of square pitch and immersed in heavy water (98.5%) in a 11-3/8 inch (28.89 cm) radius cylindrical aluminum tank. Aluminum clad thorium slugs of the MARK VII-T type were used. These slugs have a 1.207 inch outer diameter, a 0.495-inch inner diameter, and a 6-5/8 inch length. Forty mil aluminum cladding is used. The slugs were stacked end-to-end three deep, giving a lattice height of 19-7/8 inches (50.48 cm). The two lattices had different pitch. Table 2 gives further details of these assemblies.

TABLE 2

THORIUM/HEAVY WATER LATTICE DESCRIPTIONS

Square Lattice Pitch	Number of Thorium Slugs	D ₂ O/Metal	VOLUME RATIOS	
			D ₂ O/Thorium	D ₂ O/Aluminum
1-9/16 Inch	444	2.18	2.78	10.11
1-3/4 Inch	360	2.92	3.72	13.57
Height =	19-7/8 Inches		Diameter =	22-1/2 Inches

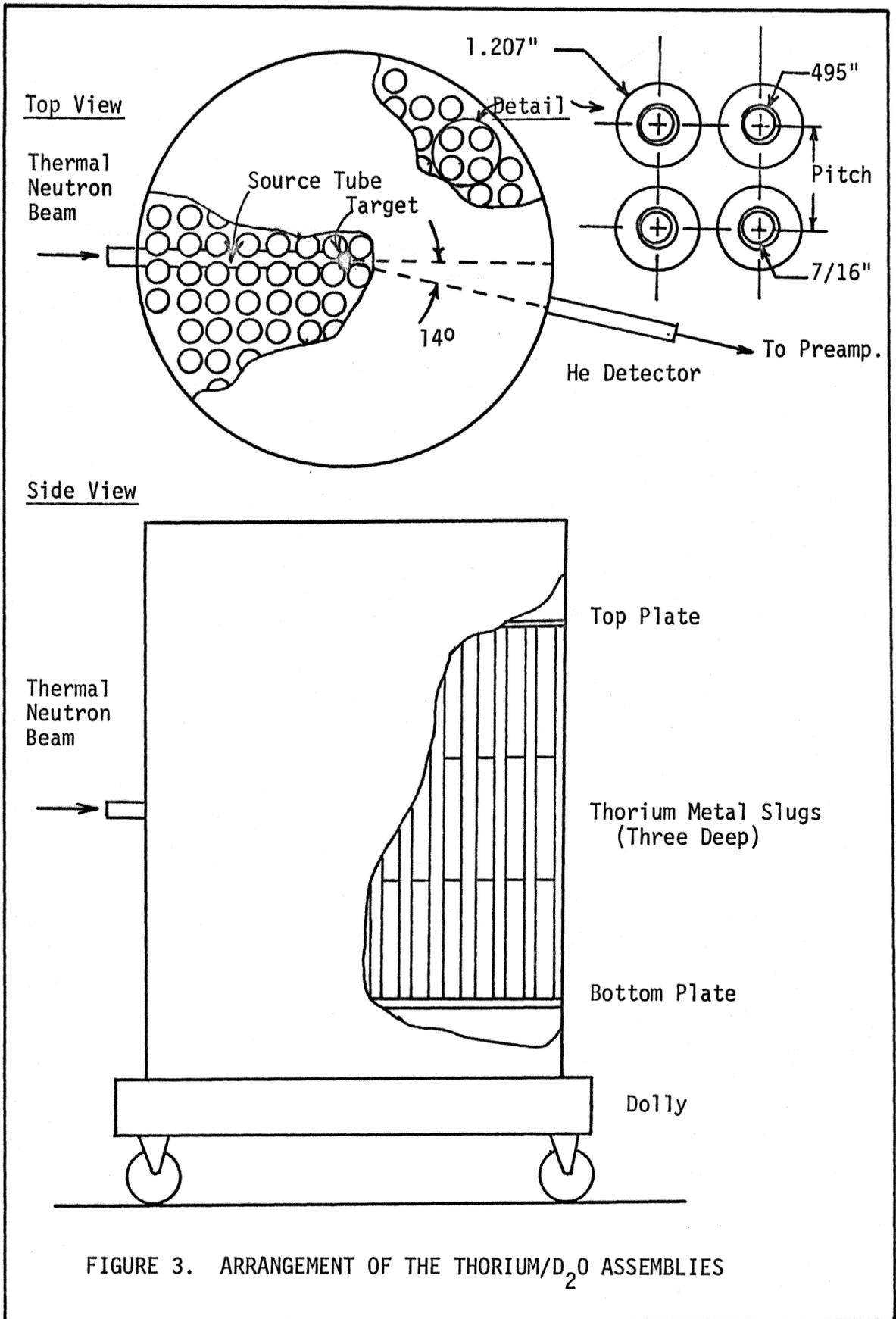


FIGURE 3. ARRANGEMENT OF THE THORIUM/D₂O ASSEMBLIES

The thermal neutron beam from the reactor entered the tank through a 1-inch aluminum tube which penetrated to the center of the lattice. The small fission disk and the small mock source, described in Table 1, were mounted at the end of the tube.

The purity of the heavy water was determined using an Abbe refractometer. The index of refraction of water - heavy water mixtures decreases with increasing percentages of heavy water. A plot of index of refraction vs. percent heavy water was made by G. N. Salaita³⁹ using samples of known mixtures of heavy and light water. Measurement of the index of refraction together with this plot, provided the determination of the percent heavy water of the sample. The purity thus determined was 98.5 ± 1.0 percent.

2 - NATURAL URANIUM ASSEMBLY

The natural uranium assembly is shown in Fig. 4. The assembly consisted of aluminum clad natural uranium slugs placed in a 8-7/8-inch (22.54 cm) radius cylindrical aluminum container. The slugs were 8-3/8-inches high by 1-inch in diameter with 60-mil thick cladding. The lattice of 434 slugs was generally hexagonal close-packed with a very slight anomaly due to a 1-1/16-inch OD source tube that penetrated completely the center of its mid-plane. This facilitated alignment. The slugs were stacked two deep.

One additional slug served as a material plug in the source tube to prevent neutron streaming in the forward direction. The volume ratio for natural uranium to air was 3.17 for this assembly. The volume ratio for natural uranium to aluminum was 3.09. The small ^{235}U source and the small mock source, described in Table 1, were used for the foreground and background measurements respectively. The sources were placed at the center of the assembly.

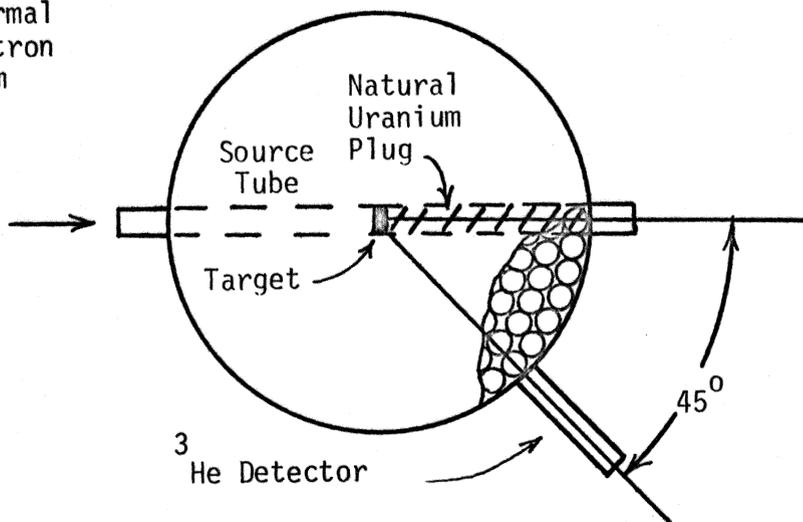
3 - HEAVY WATER SPHERE

The arrangement for this assembly is shown in Fig. 5. The 27-inch diameter sphere was constructed of 65-mil thick aluminum in two hemispheres joined at their mid-plane. A 1-9/16-inch I.D. source tube passed completely through the sphere facilitating alignment at the thermal neutron beam. The sphere was filled with 98.5% pure heavy water. A 1-1/2-inch O.D. thin walled aluminum cylinder, filled with heavy water, formed a material plug to prevent neutron streaming along the source tube in the forward direction.

The large fission disk and the large mock source, described in Table 1, were inserted in the assembly for foreground and background measurements. These sources were mounted on the end of the plug and placed at the center of the sphere.

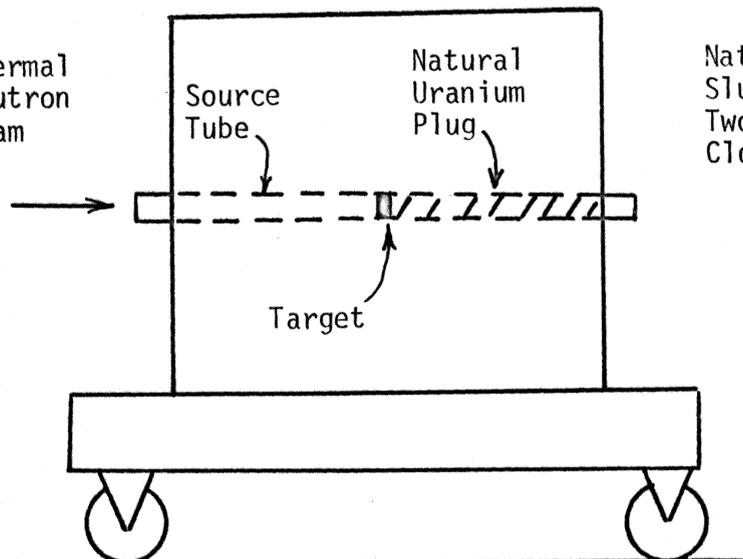
Top View

Thermal
Neutron
Beam



Side View

Thermal
Neutron
Beam



Natural Uranium
Slugs Arranged
Two Deep In A
Close-Packed Array

Dolly

FIGURE 4. ARRANGEMENT OF THE NATURAL URANIUM ASSEMBLY.

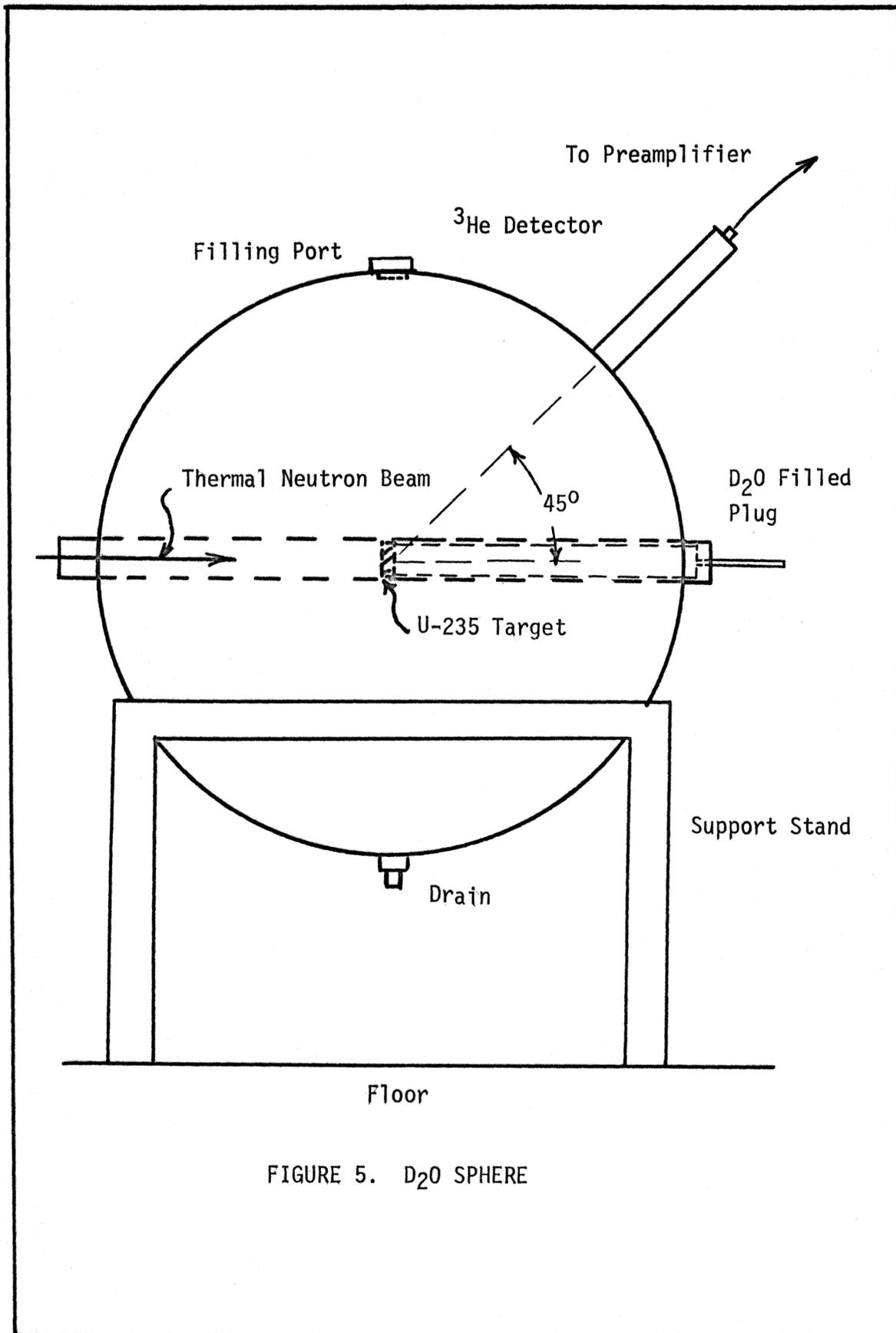


FIGURE 5. D₂O SPHERE

4 - MEASUREMENTS IN HEAVY WATER AND THORIUM NITRATE CYLINDERS

Two preliminary spectra measurements were made to aid in the selection of appropriate materials for the investigation. This resulted in the selection for parts 1-3 above and provided insight regarding the effects of source - detector positioning. Judgements made on the basis of these preliminary measurements resulted in the selection of the materials and the source-detector positioning used in the investigation reported in Sections 1 through 3 above.

Measurements were made of the fast neutron leakage from thorium nitrate and heavy water cylinders. These materials were in containers used in shipping and for permanent storage. The ^{235}U fission and mock sources were located on the outside of the containers. The ^3He detector was positioned at the mid-plane of the cylinder in a manner similar to that shown in Fig. 3 for the thorium/heavy water assemblies.

The results of these preliminary investigations indicate that large room return neutron backgrounds occur with the sources positioned on the outside of the containers. The background is due to fission and beam scattered neutrons that enter the detector after leaving the floor, walls, and shield with degraded energy. These background neutrons had a significantly high thermal component which complicated accurate unfolding of the detector response and made it

impossible to determine the true zero for neutron energy.

These measurements were important in that conclusions can be made regarding heavy water and thorium nitrate as candidate materials for comprehensive spectrum measurements. The conclusions reached for each material are as follows:

a. Heavy Water Cylinder

Based on the measurements of the fast neutron leakage spectrum for a 55 gallon shipping container, it was concluded that heavy water should prove suitable for the investigations which were to follow. The spectrum indicated that the greater portion of the leakage neutrons were within the energy range of the spectrometer. The height of the sample was 31.9 inches and the radius was 11.0 inches. A conclusion drawn from this measurement was that successful measurements could be made with assemblies containing heavy water, provided the perturbing influence of the thermal neutron background could be reduced by locating the sources at the center of the assemblies.

b. Thorium Nitrate Cylinder

Measurements of the leakage spectrum for thorium nitrate were made for a 1-cubic foot flat cylindrical sample. The measurements showed that

the spectrum was relatively hard with a significant proportion of neutrons with energies above the effective energy range of the spectrometer. This hard spectrum was attributed to small sample size. In addition the measurements were made with the fission source and the mock source on the outside surface of this small sample, and the fast neutron background was high. The decision was thus reached to carry out the investigation for thorium containing materials using larger sample sizes. This led to the experiments using thorium metal/heavy water lattices as described in Part C, 1, above.

IV. THE CALCULATIONAL METHOD

The steady state neutron behavior for the experimental assemblies is determined by distribution in space and energy. The fast neutron transport is described by the space - and energy - dependent angular neutron flux $\phi(\underline{r}, E, \underline{\Omega})$ which satisfies the Boltzmann equation:

$$\underline{\Omega} \cdot \nabla \phi(\underline{r}, E, \underline{\Omega}) + \Sigma_T(\underline{r}, E) \phi(\underline{r}, E, \underline{\Omega}) = \quad (1)$$

$$S(\underline{r}, E, \underline{\Omega}) + \int dE' \int d\underline{\Omega}' \Sigma(\underline{r}, E' \rightarrow E, \underline{\Omega}' \rightarrow \underline{\Omega}) \phi(\underline{r}, E', \underline{\Omega}')$$

- where:
- $\Sigma_T(\underline{r}, E)$ = the macroscopic total cross-section for neutrons with energy E at position \underline{r} .
 - $\Sigma(\underline{r}, E' \rightarrow E, \underline{\Omega}' \rightarrow \underline{\Omega})$ = the transfer cross-section which represents the macroscopic differential scattering cross-section for neutron transfers at position \underline{r} from energy E' while traveling between E and E + dE and direction $d\underline{\Omega}$ about $\underline{\Omega}$ after collision.
 - $S(\underline{r}, E, \underline{\Omega})$ = the neutron source distribution.

An objective of this research was to test the validity of the differential data of the ENDF/B-II data files for the materials of the assemblies by comparing calculated leakage spectra with measurements. In principle the neutron distribution could be solved by inserting into the transport equation appropriate ENDF/B-II data files with the geometrical arrangement of the materials. However, solution of the transport equation is not possible by analytical means except in the simplest of geometrical uses. Generally, solutions of the neutron transport equation require sophisticated numerical methods with a digital computer.

A. THE GENERAL TECHNIQUE

1. Multigroup Cross-Sections

Numerical solutions of the transport equation can be obtained by a multigroup treatment of the cross-section data. In the multigroup method the first step is division of the entire range of neutron energy into N groups. These groups need not be of equal size. The multigroup method does not infer any restrictions on the energy dependent cross-sections. These cross-sections are often very complicated functions of energy.

In multigroup theory the neutron energy range of interest, i.e., $E_{\min} \leq E \leq E_{\max}$, is divided into a finite number, N , of intervals separated by the energies E_i , where $i = 1, 2, 3, \dots, N$.

The numbering sequence is such that as i increases, the

energy decreases, i.e., $E_i \geq E_{i+1}$.

Typically, the procedure for solving the multigroup equations proceeds by solving first the equation for group 1, then for group 2, and so forth. Neutrons may disappear from a group either in an absorption interaction or as the result of elastic or inelastic scattering which decreases their energy (increases their lethargy) to that of another group. Lethargy for a neutron is defined by the expression $u = \ln E_0/E$ where E_0 is an upper energy reference level and E is the energy of the neutron.

For an accurate multigroup calculation the neutron energy is divided into a large number of groups. Care must be taken when choosing the energy range for each group to insure that the variation of important cross-sections within each group is kept reasonably small.

2. Cross-Section Data Input and Codes

For numerical computations the DLC - 2 data set²³ provides a basic library for the ANISN code. DLC - 2 provides multigroup transport code data input based on the ENDF/B Version II cross-section set. Neutron transport calculations can be performed for non-thermal energies with DLC - 2 data.

The DLC - 2 data was generated at the Oak Ridge National Laboratory using the PSR - 13/SUPERTOOG data retrieval program. This data was averaged over each

specified group width by the SUPERTOG code. DLC - 2 consists of fine group constants including reaction data (absorption and fission), P_g elastic scattering data, and inelastic and (n, 2n) scattering data which were generated, combined, and written on magnetic tape as card images in the ANISN format.

DLC - 2 data represents a P - 8 approximation for elastic scattering angular distributions. The data has a 100 group fine structure. The neutron energy range covers from 14.92 MeV to 0.414 eV.

3. Multigroup Neutron Transport Calculations

For the steady state problem, i.e., $\frac{\partial \Phi}{\partial t} = 0$, the neutron angular flux in group g may be defined by:

$$\Phi_g(\underline{r}, \underline{\Omega}) = \int_{E_g}^{E_{g-1}} \Phi(\underline{r}, E, \underline{\Omega}) dE \quad (2)$$

Thus, from the second term of equation 1,

$$\Sigma_g(\underline{r}, \underline{\Omega}) = \left\{ \int \Sigma_T(\underline{r}, E) \Phi(\underline{r}, E, \underline{\Omega}) dE \right\} / \Phi_g(\underline{r}, \underline{\Omega}) \quad (3)$$

The group cross-section, $\Sigma_T(\underline{r}, \underline{\Omega})$, has a dependence on $\underline{\Omega}$. Generally, solutions can be obtained by first assuming a form for the angular flux and then integrating over energy. The first step, therefore, in a multigroup approximation, is to represent the angular dependence of the flux by an expansion. To make calculations possible, the expansion is terminated

after a finite number of terms. In this manner an approximation is introduced.

In one class of multigroup methods, known variously as the discrete ordinates, discrete S_n , or simply S_n method, the transport equation is solved by evaluation of the angular distribution of the neutron flux in a number of discrete directions. Angular intervals are then approximated by sums over discrete directions and derivatives by differences. The accuracy of the S_n method can be improved simply by increasing the number of discrete directions, n , without otherwise changing the method of solution.

The S_n multigroup equations are differential equations. They can be converted into a system of algebraic equations for machine computation by introducing a space mesh and approximating derivatives by differences. With fast digital computers, the multigroup equations are capable of yielding results of high precision. In these calculations the main uncertainties are related to the values of the group constants (group cross-sections). Other uncertainties arise because of approximations associated with the degree of the angular expansion for the input cross-sections, in the energy spacing and number of groups, and in the space mesh chosen.

The discrete ordinates method gives a solution to the transport equation involving the variables of position \underline{r} , direction $\underline{\Omega}$, and energy E , and, dependent on these, and the neutron flux $\phi(\underline{r}, E, \underline{\Omega})$. It is basically a quadrature method over $\underline{\Omega}$ and furnishes a method of replacing $\phi(\underline{r}, E, \underline{\Omega})$ by a set of functions $\phi_m(\underline{r}, E)$, with $m = 1, 2, 3, \dots, M$. The ϕ_m 's are the ordinates in the quadrature and n specifies the order of precision. The integro - differential form of the transport equation is thus transformed into a set of much simpler differential equations in the ordinate fluxes ϕ_m .

The basic feature of the S_n approximation is that the transport equation is reduced to a system of linear differential equations with the discrete treatment of the angular (direction) variable. In spherical coordinates, μ , the direction cosine for the neutron relative to the radius vector, is represented by a set of discrete directions $\{\mu_j\}$.

The S_n method can be illustrated by simple example. Consider the case of neutron transport for a source free medium with plane symmetry and isotropic scattering. The vector flux $\phi(x, \underline{\Omega})$ is replaced by $\phi(x, \mu) = 2\pi \phi(x, \underline{\Omega})$. In isotropic systems the total scattering cross-section does not depend on the flight direction of the neutron, so:

$$\int_{4\pi} \int_0^{\infty} \Sigma(x, \underline{\Omega}' \rightarrow \underline{\Omega}, E' \rightarrow E) d\Omega dE = \Sigma_S(x, E') \quad (4)$$

The Boltzmann transport equation, equation 1, reduces for the one - velocity case to:

$$\mu \frac{\partial \Phi(x, \mu)}{\partial x} + \frac{\Sigma}{T}(x) \Phi(x, \mu) = 1/2 \Sigma_S(x) \int_{-1}^1 \Phi(x, \mu') d\mu' \quad (5)$$

Instead of the continuous direction variable μ , discrete directions μ_j , where $\mu_j = 1, 2, 3, \dots, n$, are introduced and $\Phi(x, \mu_j)$ is represented by:

$$\int_{-1}^1 \Phi(x, \mu') d\mu' \approx \sum_{i=1}^n W_i \Phi(x, \mu_i) \quad (6)$$

where the W_i are suitably chosen quadrature weights (or weighting factors).

This gives a set of n coupled differential equations which can be solved by difference techniques once the boundary conditions are specified. The equations thus formed are:

$$\mu_j \frac{\partial \Phi(x, \mu_j)}{\partial x} + \frac{\Sigma}{T}(x) \Phi(x, \mu_j) = 1/2 \Sigma_S(x) \sum_{i=1}^n W_i \Phi(x, \mu_i) \quad (7)$$

The accuracy with which the solution to these equations represents the actual solution of equation 5 depends, to a large extent, on the choice of quadrature weights, W_i , the quadrature directions μ_i , and n . With large n , the solution is practical only with the help of an electronic computer.

The use of discrete ordinates in transport theory was suggested by G. C. Wick³⁰ in 1943. More extensive development of the method was carried out by S. Chandrasekhar^{31, 32, 33}. In Wick's method, the directions, μ_i , and the weight factors, W_i , are chosen so that the Gauss integration method can be applied to the numerical integration.

A different procedure was proposed by B. G. Carlson³⁴ in his original S_n method. In this S_n method, the interval $-1 < \mu < 1$ is usually divided into equal parts and the vector flux is approximated by a linear function of μ in each part. The system of differential equations are then integrated by numerical methods with specified boundary conditions for the flux, $\phi(x, \mu_j)$.

In multigroup form, the transport equation (Equation 1) can be re-written as:

(8)

$$\Omega \cdot \nabla \phi_g(\underline{r}, \Omega) + \Sigma_g(\underline{r}) \phi_g(\underline{r}, \Omega) = S_g(\underline{r}, \Omega) + \sum_{j=1}^G \int d\Omega' \Sigma_{j \rightarrow g}(\underline{r}, \Omega' \rightarrow \Omega) \phi_j(\underline{r}, \Omega')$$

where $j = 1, 2, 3, \dots, G$ is the group index associated with the neutron energy E' of Equation 1. The index g is associated with the energy E and the terms are basically the same as previously defined with Equation 1. To illustrate certain aspects of the solution, consider the case of the transport equation in spherical geometry. The scattering cross-section can be expanded in Legendre polynomials. The expansion is:

$$\Sigma_s^{j \rightarrow g}(r, \mu_0) = \sum_{\ell=0}^{\infty} \frac{2\ell+1}{2} \Sigma_{s\ell}^{j \rightarrow g}(r) P_{\ell}(\mu_0) \quad (9)$$

and according to the addition theorem of Legendre polynomials

$$P_{\ell}(\mu_0) = P_{\ell}(\mu)P_{\ell}(\mu') + 2 \sum_{m=1}^{\ell} \frac{(\ell-m)!}{(\ell+m)!} P_{\ell}^m(\mu) P_{\ell}^m(\mu') \cos m(\phi - \phi') \quad (10)$$

where μ and μ' are the direction cosines and ϕ and ϕ' are the azimuthal angles specifying the directions $\underline{\Omega}$ and $\underline{\Omega}'$, respectively.

On insertion of Equation 10 into Equation 9 and the result into Equation 8, the terms containing $\cos m(\phi - \phi')$ will vanish upon integration over ϕ' ; then

$$\mu \frac{\partial}{\partial r} \Phi_g(r, \mu) + \frac{1-\mu^2}{r} \frac{\partial}{\partial \mu} \Phi_g(r, \mu) + \Sigma_T^g(r) \Phi_g(r, \mu) = S_g(r, \mu) \quad (11)$$

$$+ \sum_{\ell=0}^{\infty} \frac{2\ell+1}{2} P_{\ell}(\mu) \sum_{j=1}^G \sum_{s \ell}^{j \rightarrow g}(r) \int_{-1}^1 \Phi_g(r, \mu') P_{\ell}(\mu') d\mu'$$

In the above procedure the introduction of the scattering cross-section in Legendre polynomials is not an essential feature of the discrete ordinates method, but it does facilitate the determination and handling of group cross-sections since these have a dependence on μ .

The details of the method, formulation of the difference equation, and numerical procedures are given in the references by B. G. Carlson^{34, 35}. C. E. Lee³⁶ has presented a detailed investigation describing the S_n method for solution of the multigroup two and three - dimensional transport equations.

B. THE TRANSPORT CODE

One code developed to provide a numerical solution of the Boltzmann equation is the one-dimensional discrete S_n transport code ANISN⁸. ANISN is frequently used in neutron shielding calculations. It was used in the research reported herein for several reasons:

1. Options in the code permit solution of the Boltzmann transport equation for the spherical and cylindrical geometries of the experimental assemblies.

2. This code accepts microscopic neutron cross-section data of the ENDF/B-II data set as input. The code permits general anisotropic scattering (i.e. an L^{th} order P_L Legendre expansion of the scattering cross-sections).
3. The code was designed to solve neutron penetration problems in which angle-dependent spectra are calculated in detail. One feature that makes ANISN suitable for neutron transport work is its optional data-storage configurations which allow execution of small, intermediate, and relatively large problems.

A flow chart of the computer calculational technique is given in Fig. 6. The basic nuclear data is input from the DLC-2 format²³ for the ENDF/B-II differential scattering cross-section data. The DLC-2 format data was developed at Oak Ridge National Laboratory using the SUPERTOG²⁴ retrieval program. The procedures of the calculations used in this analysis are described by the following steps which are shown in the figure:

- (STEP 1): A binary representation of the 100-group DLC-2 data was created using the DLC-2 Data Retrieval Program compatible for input to ANISN.
- (STEP 2): The special purpose program TAPEMAKER²⁵ provided the capability of performing the multi-group calculations one group at a time to prevent potential problems with computer storage.

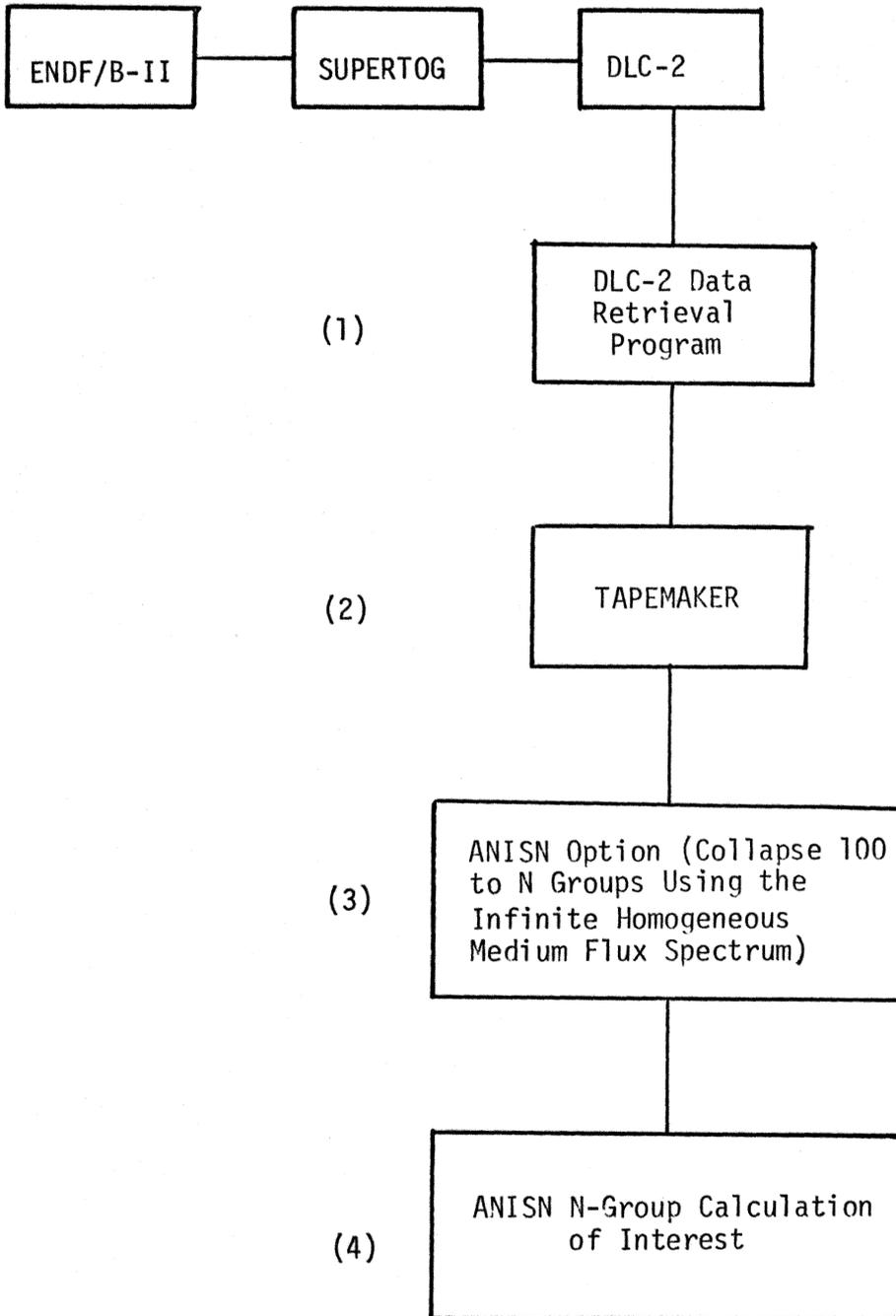


FIGURE 6. THE MULTIGROUP TRANSPORT CODE CALCULATION

These potential problems are due to the complex requirements for computations involving several different elements with a fine group, high order Legendre polynomial expansion of the anisotropic scattering cross-section data, and the high order numerical S_n technique for describing the discrete directions for scattering. The functioning of the TAPEMAKER program is described as follows:

- (a) Only the cross-section data for a single group is stored in the memory while the calculations for that group are performed.
- (b) Data for the next group is read through TAPEMAKER (replacing the data stored from the previous group) before calculations for that group are performed.

(STEP 3): An option of the ANISN code is used to collapse the 100-group cross-section set to a 27-group set. As a weighting function, the flux spectrum in an infinite homogeneous medium containing a spatially uniform source is used.

(STEP 4): Once these flux weighted 27-multigroups cross-sections are prepared by the above procedure, the discrete ordinate S_n transport calculation of the neutron leakage spectra was performed.

Eighty (80) spatial mesh intervals were used and the ^{235}U fission source was assumed to occupy the 1st mesh interval (center of the assembly) in each case. The assemblies were treated as homogeneous materials of the appropriate isotopes for each case.

C. DETAILS OF THE CALCULATIONS

In the calculations of the predicted leakage spectra the scattering cross-sections were expressed as expansions in Legendre Polynomials to order P_3 . This was done for each of the assemblies to represent the angular dependence of the neutron flux in the calculations. The transport equation was solved with the angular distribution of the neutron flux evaluated in a number of discrete directions. For cylindrical geometry the calculation for each assembly was to order S_8 . For the heavy water sphere, S_{16} quadrature order was used. The change from S_8 for calculations for the cylinders to S_{16} for the heavy water sphere was made to provide a more accurate angular quadrature without using significantly more computer core or requiring a longer computation time. This change was possible because S_n transport calculations require significantly fewer angular cosines and weights in the angular quadrature for spheres for the same order n . (that is: $(n + 1)$ angular cosines and weights are required for spheres and $n(n + 4)/4$ angular cosines and weights are required for

cylinders).

The 27-broad group energy widths for the transport calculation are given in Table 3. An option in the ANISN code was used to produce the group reduction of the cross-sections from 100-fine groups to 27 - broad groups as described in Section A, above. The multigroup structure is for the neutron energy range from 14.9 MeV to 0.414 eV. Groups 6 through groups 27 span the effective energy range of the spectrometer. The energy range for groups 19 through 27 covers the approximate range of the first spectrometer channel, i.e. 13.5 keV. The 27-broad group boundaries were selected to provide approximately the same lethargy width for each broad group.

D. TEST OF THE TRANSPORT CODE APPLICATION

Since the ANISN Code was to be used for fast neutron spectrum analysis, a test of its applicability was made. The Japanese Atomic Energy Research Institute (JAERI) I-5 fast critical assembly has a relatively hard central neutron spectrum for which ^3He spectrometer measurements have been reported by T. Iijima et. al.². This 20% enriched uranium assembly provided a good subject for the proposed test. The ^3He measurements give spectrum measurements which can be used for comparison with the transport calculation and, at the same time, an assessment of the ENDF/B-II cross-sections for the materials of the assembly can be made.

TABLE 3

BROAD ENERGY GROUP BOUNDARIES FOR THE TRANSPORT CALCULATIONS

<u>GROUP</u>	<u>UPPER BOUNDARY (eV)</u>	<u>GROUP</u>	<u>UPPER BOUNDARY (eV)</u>
1	1.492×10^7	15	1.500×10^5
2	9.048×10^6	16	1.111×10^5
3	6.065×10^6	17	5.248×10^4
4	4.066×10^6	18	3.183×10^4
5	2.466×10^6	19	1.503×10^4
6	1.827×10^6	20	3.335×10^3
7	1.353×10^6	21	5.830×10^2
8	1.003×10^6	22	1.013×10^2
9	7.427×10^5	23	2.902×10^1
10	6.081×10^5	24	1.068×10^1
11	4.978×10^5	25	3.059×10^0
12	4.076×10^5	26	1.125×10^0
13	3.020×10^5	27	4.140×10^{-1}
14	2.024×10^5		

Since the purpose of the test was to provide a qualitative check on the flexibility and usefulness of the code, a low order polynomial, P_2 , with low order angular quadrature, S_4 , calculation was performed using 27 energy groups. Eighty (80) spatial mesh intervals were used in the calculation.

The central flux is plotted versus energy in Figure 7. ^3He data points are from the T. Iijima et. al.². The ^3He data was normalized in the energy interval 407keV - 498 keV which represents the energy interval for the spectrum peak in the ANISN transport calculations. The results of the comparison show good agreement for the central spectrum. The experimentally determined spectrum is slightly harder than the spectrum determined by the transport calculation using ENDF/B-II cross-section data.

The significant population of measured neutrons above about 600 keV confirms the observations presented in Part IV and Part V regarding the ^{238}U ENDF/B-II data file. Fig. 8 shows the results for a $P_2 - S_4 - 27$ group transport calculation. No results for ^3He spectrometer measurement were presented by T. Iijima et. al.² for neutron leakage from the I-5 assembly.

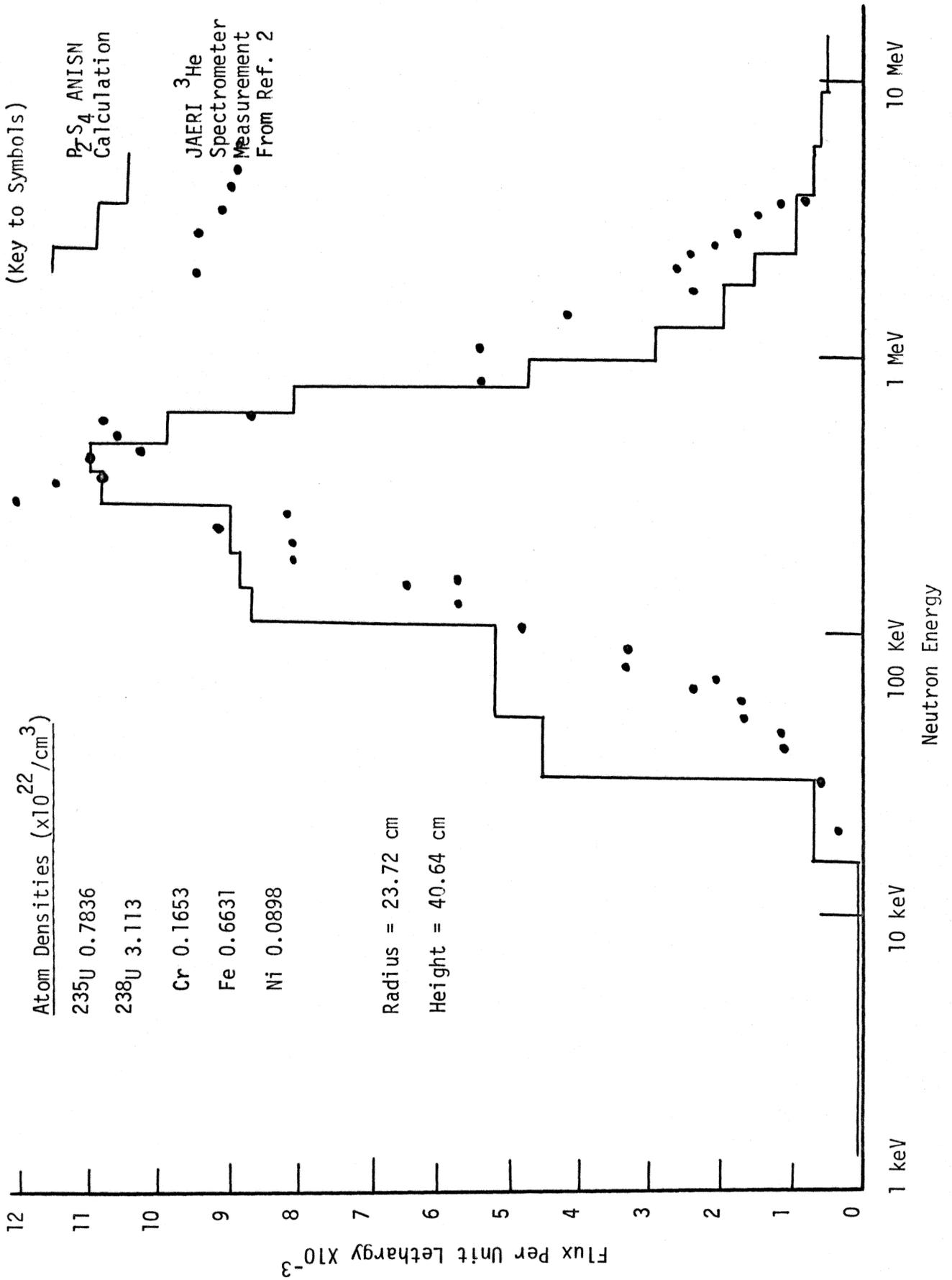


FIGURE 7. CENTRAL NEUTRON SPECTRUM OF JAERI I-5 FAST ASSEMBLY

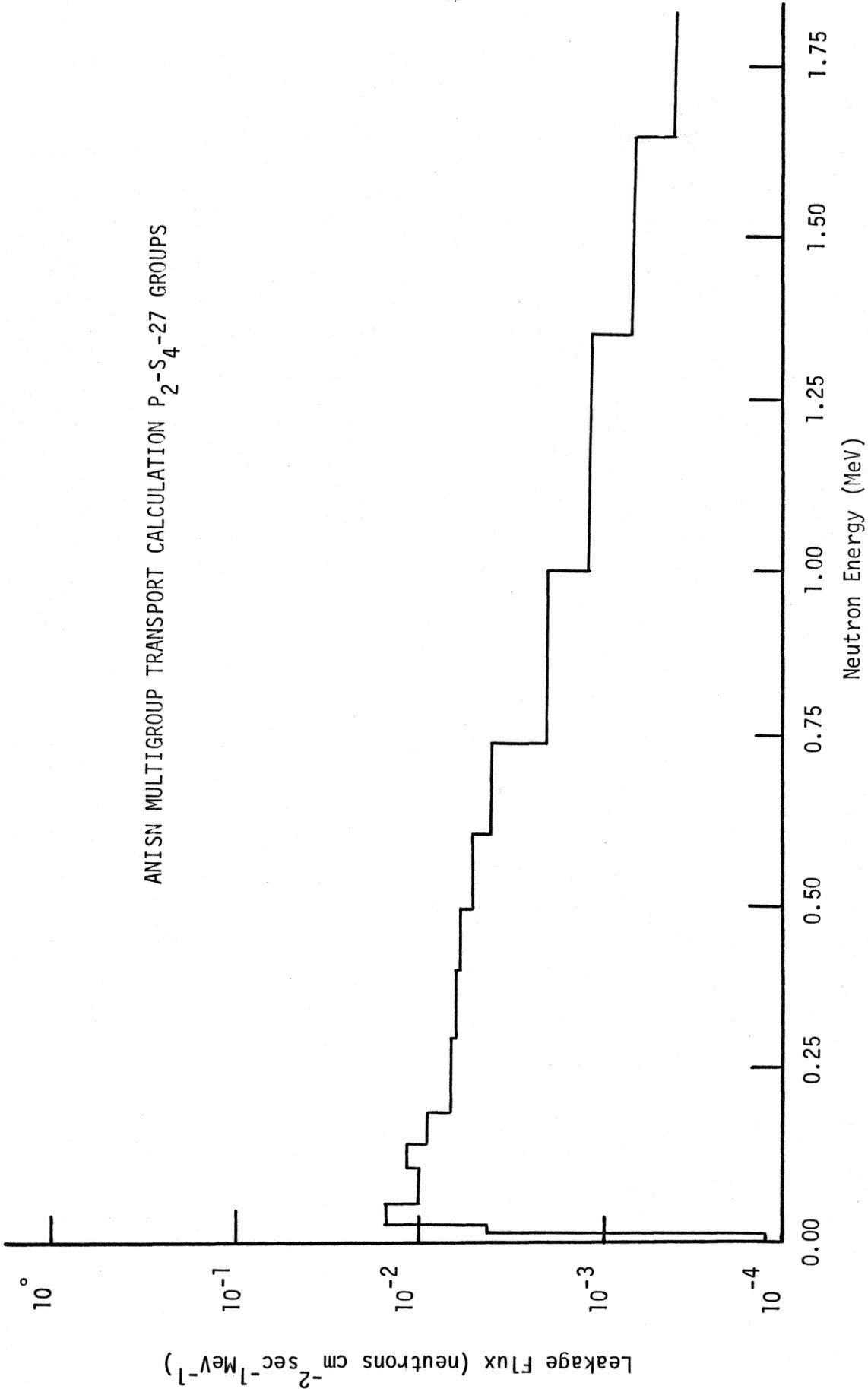


FIGURE 8. NEUTRON LEAKAGE SPECTRUM FOR JAERI I-5 FAST ASSEMBLY

V. COMPARISON BETWEEN CALCULATION AND EXPERIMENT

Figures 9 through 12 show comparisons of the grouped experimental spectrum to the calculated spectrum for each of the thorium/D₂O assemblies, the natural uranium assembly, and the heavy water sphere. The comparisons between the experimental data and the results of the transport calculations are grouped according to the 27-broad groups as listed in Table 3.

The ANISN transport calculation multigroup #9, which has the neutron energy range from 608.1 keV to 742.7 keV, was chosen for the normalization since this group is near the middle of the energy range of the experimental investigation. The ratio of the experimental flux to the calculated flux is unity in this group.

For each spectrum comparison there is a rather large difference between the experimental and calculated leakage below about 100 keV. This is attributed to room return low energy background and to uncertainties in the $^3\text{He} (n,p)^3\text{H}$ cross-sections used in the spectrum analysis previously discussed in Section I.

The agreement between the experimental and calculated spectrum is generally good for each comparison. Since the measured resolution of the spectrometer was about 24%, the sharp oxygen resonance at 433 keV was not resolved as expected for those assemblies containing D₂O.

For natural uranium, the results of the comparisons shown in Figure 11 indicates that between about 0.743 MeV and the upper limit

of the investigation, 1.65 MeV, the calculated leakage spectrum significantly underpredicts relative to the measured spectrum. These results using ENDF/B-II cross-section data tend to confirm the observations of N. N. Kaushal, B. K. Malaviya, et. al.¹⁶ and H. Alter⁴⁰ who suggest inadequacies in the ²³⁸U data files. Kaushal and Malaviya have observed discrepancies among various standard data files, including ENDF/B-II, for the inelastic slowing down effectiveness in the range 1 to 2 MeV. H. Alter, in a systematic comparison of calculations against integral experiments for critical assemblies, has suggested errors in the transport cross-sections for ²³⁸U in the ENDF/B-II data file.

Further evidence to support this view is given in Part IV., D, for the comparisons of results of the JAERI I-5 fast assembly.

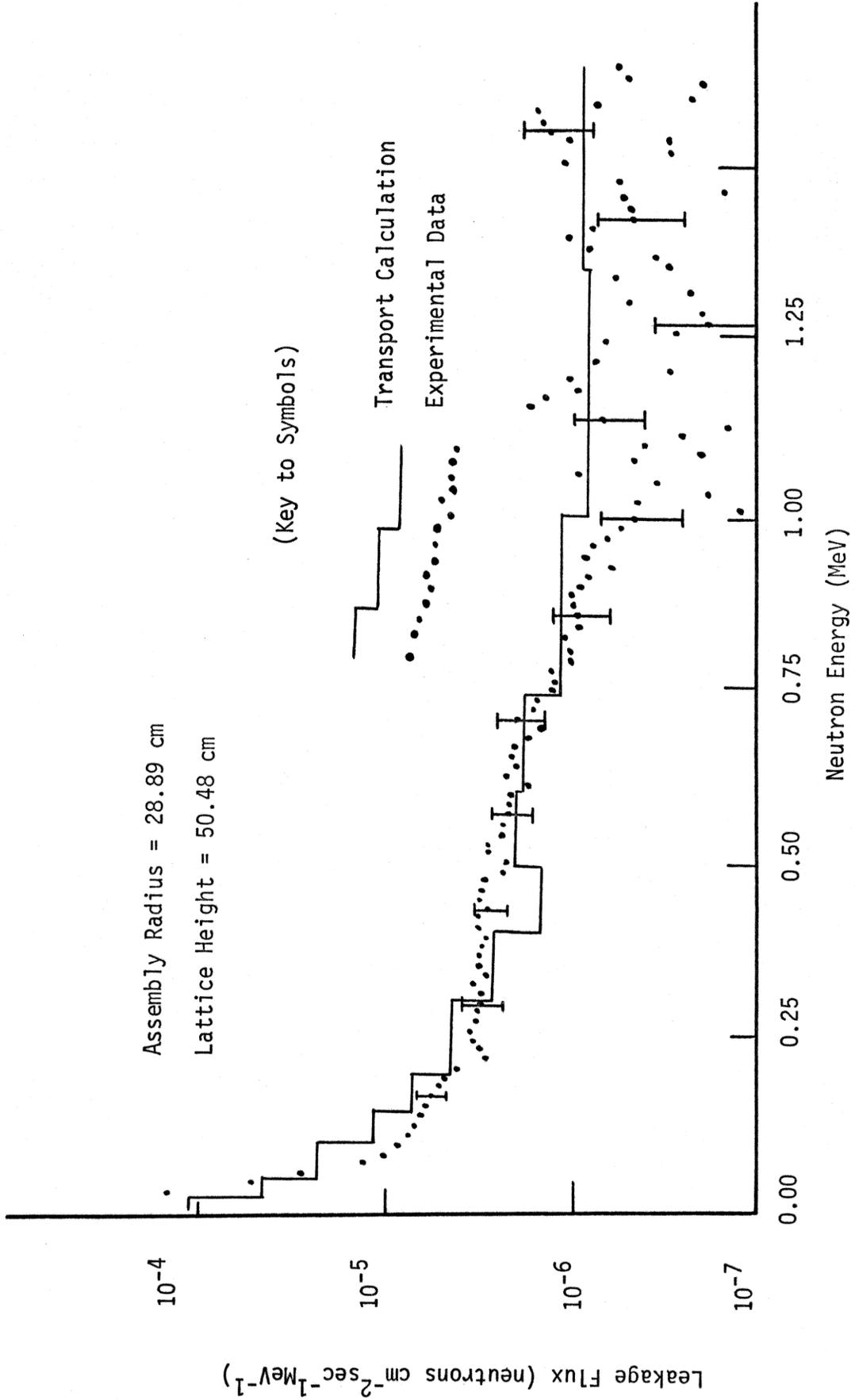


FIGURE 9. NEUTRON LEAKAGE SPECTRA FOR THORIUM/D₂O LATTICE OF PITCH = 3.97 cm (1-9/16-inch)

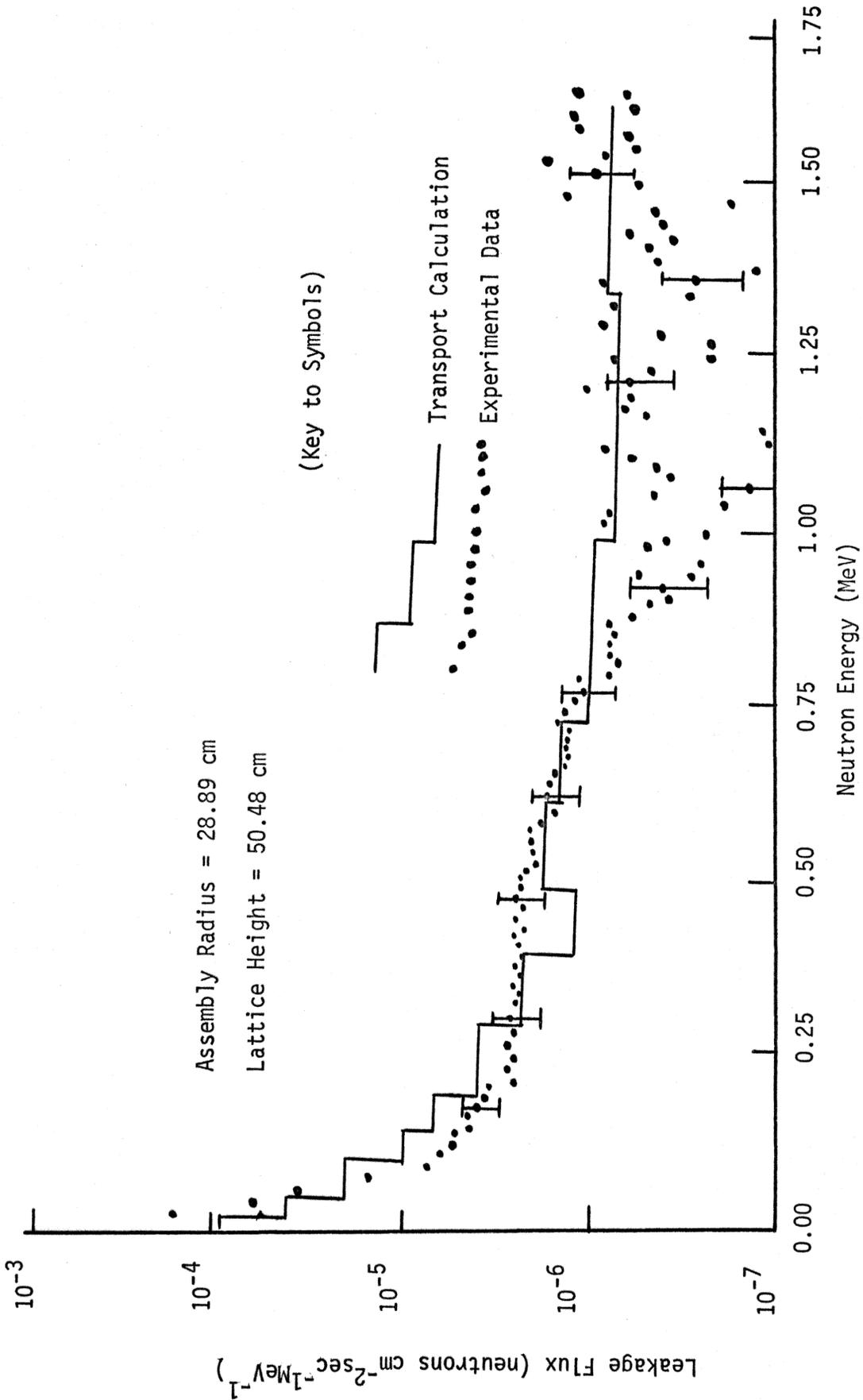


FIGURE 10. NEUTRON LEAKAGE SPECTRA FOR THORIUM/D₂O LATTICE OF PITCH = 4.45 cm (1-3/4-inch)

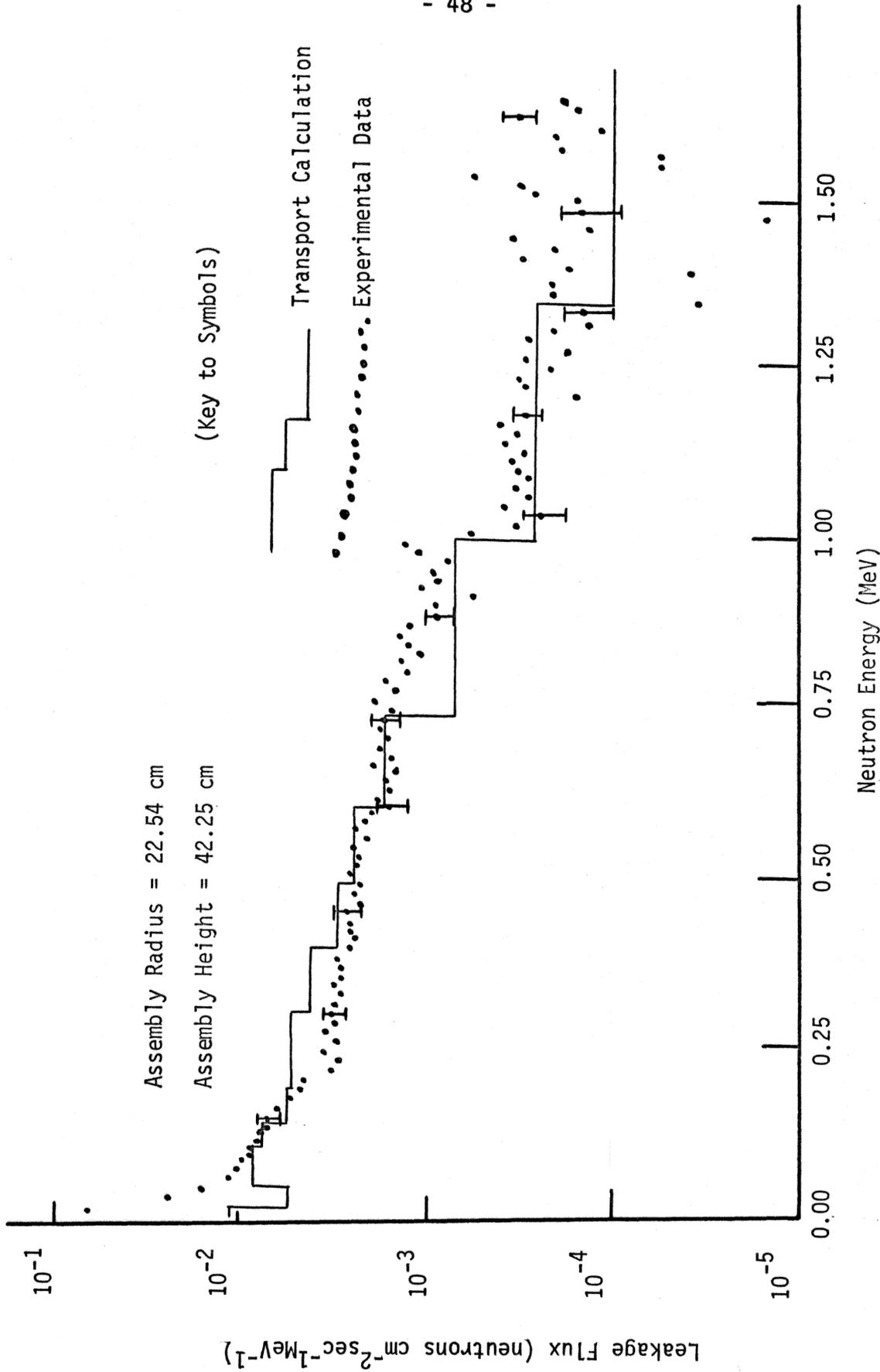


FIGURE 11. NEUTRON LEAKAGE SPECTRA FOR NATURAL URANIUM ASSEMBLY

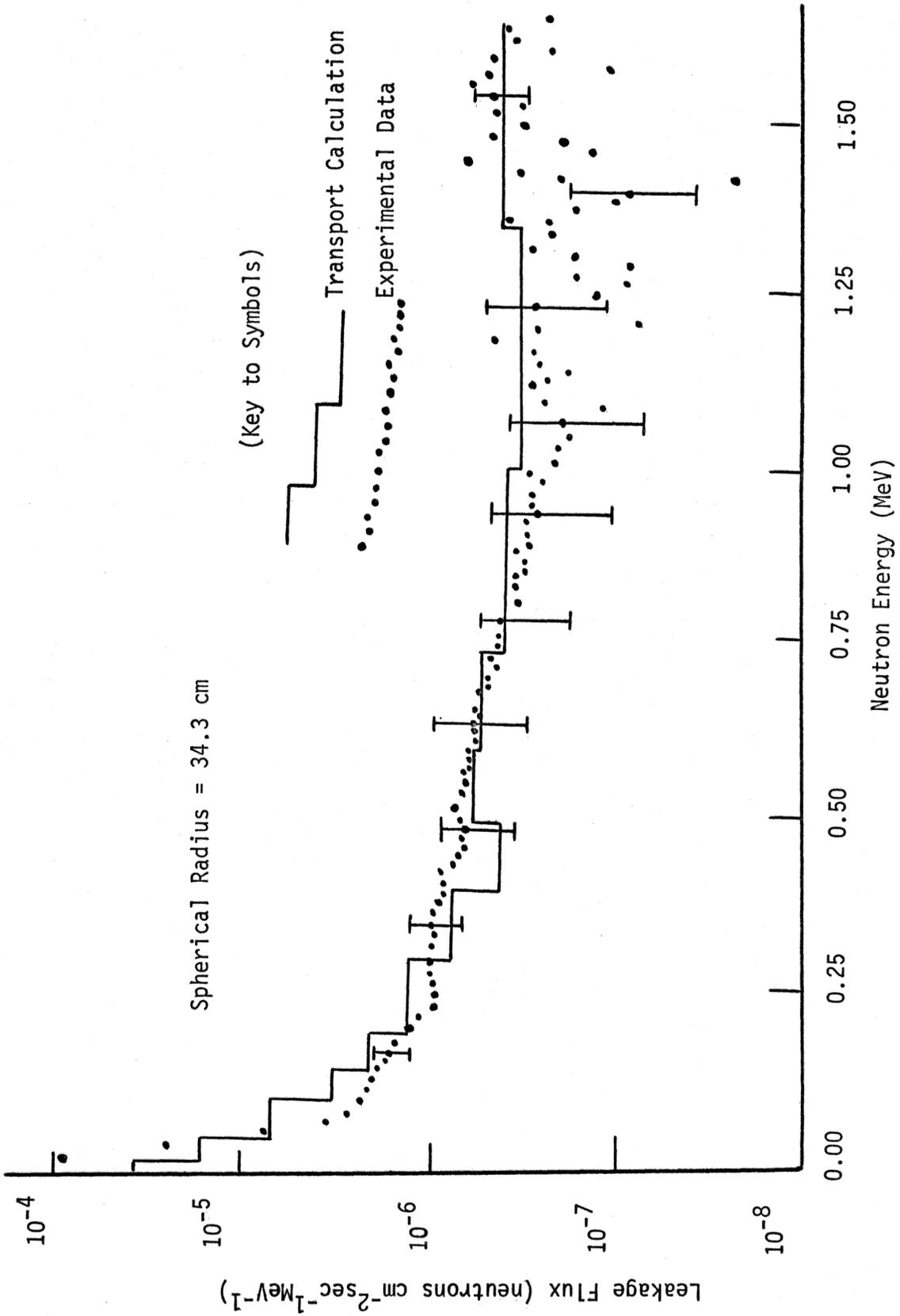


FIGURE 12. NEUTRON LEAKAGE SPECTRA FOR HEAVY WATER SPHERE

VI. CONCLUSIONS

The leakage spectra calculated with ENDF/B-II cross-section and measured with the ^3He spectrometer are in reasonable agreement over the neutron energy range from 100 keV to 1.65 MeV. For the natural uranium assembly the measurement shows significantly more neutrons above about 0.743 MeV than are predicted with the ANISN transport calculation.

Other investigators have found differences between calculations of integral parameters using ENDF/B-II uranium cross-sections and experimental results. H. Alter⁴⁰ has suggested that the results of systematic evaluations at Atomic International indicate that the ENDF/B-II neutron cross-sections for ^{238}U and ^{235}U are not adequate for the needs of the LMFBR Program. Testing was performed against a set of 10 integral experiments for assemblies with mean fission-energy ranging from a low of about 200 keV to a high of about 1.06 MeV. N. N. Kaushal, B. K. Malaviya, et. al.¹⁶ conclude that results of cross-section tests at R.P.I. suggest that uranium ENDF/B-II data is in need for revision for satisfactory spectra prediction.

For the two thorium/heavy water assemblies, a harder spectrum was obtained for the assembly with the greater metal/heavy water ratio as expected.

The ANISN multigroup transport code⁸, a standard code used in fast reactor analysis, proved useful. The flexibility afforded by the

several operational options in this code made possible the solution of a relatively demanding problem for a university size computer. In performing ANISN calculations, the limitations imposed by computer core data storage space must be considered by the user. The definition of the spatial, angular, and energy mesh should be sufficiently fine to give the desired resolution in the predicted spectra and to provide sufficient accuracy.

It is concluded that the ^3He spectrometer method is useful for measurements of the leakage spectrum for fast assemblies with an applicable energy range from 100 keV to 2MeV. A variety of ^3He detectors are available making possible for the investigator, a selection which can be tailored to the specific physical nature of his experiment. The resolution of ^3He detectors is degraded by increasing pressure. However, the higher pressure models offer decreased wall effects and higher efficiency. In spite of the relatively poorer resolution of the higher pressure detectors (4-10 atmospheres) they are to be preferred for spectrometry purposes because of their decreased wall effects and higher efficiency. For intermediate energy or fast neutron detection the highest pressure consistent with the required resolution is recommended. ^3He detectors are also available which have a significant partial pressure of Krypton stopping gas. Krypton containing ^3He detectors offer reduced wall effects at appreciably lower cost than a higher pressure detector with equivalent energy range.

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FAST NEUTRON LEAKAGE SPECTRA FOR ASSEMBLIES
CONTAINING THORIUM, D₂O AND NATURAL URANIUM

by

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(ABSTRACT)

Integral measurements with a ³He spectrometer have been made to test the validity of the differential data of the ENDF/B-II files for thorium, natural uranium and heavy water. The experimental measurements of fast neutron spectra have been in two lattices of thorium metal and D₂O, a cylindrical array of close-packed natural uranium metal slugs in air, a D₂O sphere and cylinder, and a cylinder of thorium nitrate. A ²³⁵U fission disc, irradiated by a beam of thermal neutrons from the V.P.I. & S.U. reactor, provided a fast neutron source. Neutron leakage spectra were taken over the energy range between 100 keV and 1.65 MeV. The spectrometer was calibrated using neutrons from the ³H(p,n)³He reaction in a Van de Graaff accelerator.

Experimental results were compared with leakage spectra calculated with the multigroup transport code, ANISN. For the thorium metal and D₂O lattices and for the D₂O sphere and cylinder, agreement between calculated and measured spectra was attained confirming the validity of the ENDF/B-II cross-sections and pointing to the usefulness of the ³He spectrometer for fast neutron leakage spectra determinations.

For the natural uranium assembly, the calculated leakage spectrum between about 0.743 MeV and 1.65 MeV was significantly below the measured spectrum. This discrepancy supports a recent suggestion by H. Alter of Atomics International that the ENDF/B-II data files contain significant errors in the transport cross-sections for ^{238}U and that the suspect data are in need of closer examination and revision.

As a further test of the applicability of the calculational procedures and ^{238}U cross-sections, transport analysis using the ANISN code was applied to predict the central spectrum of the Japanese Atomic Research Institute (JAERI) I-5 fast assembly. These calculations were compared with the results of published ^3He spectrometer measurements of the fast neutron spectrum for this 20% enriched uranium assembly. The results of the comparison reveal a measured spectrum slightly harder than the spectrum determined by the calculations using ENDF/B-II cross-section data, and gives further evidence to support the conclusions regarding the suspect ^{238}U data files.