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**CALCULATED NEUTRON EMISSION SPECTRA FROM SPHERICAL
SHELLS SURROUNDING A 0- TO 11-MeV NEUTRON
SOURCE: TESTS OF THE LLL EVALUATED
NUCLEAR DATA LIBRARY (ENDL)**

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CALCULATED NEUTRON EMISSION SPECTRA FROM SPHERICAL SHELLS SURROUNDING A 0- TO 11-MeV NEUTRON SOURCE: TESTS OF THE LLL EVALUATED NUCLEAR DATA LIBRARY (ENDL)

Abstract

Calculated neutron emission spectra from spherical shells of various materials surrounding an Am-Be source are compared with published experimental results. The evaluated neutron interaction data used for these calculations were those of the Lawrence Livermore Laboratory Evaluated Nuclear Data Library (ENDL), June, 1974, version. The quality of the compari-

sons varied from excellent to rather poor. The results for spherical shells of CH_2 , Be, and Mo are excellent. For tantalum and tungsten, the comparisons served as diagnostic and resulted in locating an error in the case of tantalum and an inadequacy in the case of tungsten. It is concluded that more microscopic measurements are required for niobium and lead.

Introduction

In recent years, integral experiments have been useful in assessing the reliability of neutron transport calculations. These experiments have uncovered deficiencies in evaluated microscopic data and bugs in the codes which are used to process these data and to calculate neutron transport. The processing and transport codes are now considered quite reliable. Therefore, the comparisons of integral experiments with calculations can focus on the evaluated microscopic data.

Several types of integral experiments are available to test evaluated data at a variety of neutron energies. In particular, neutron data below a few MeV are tested by both reflected and unreflected critical

assemblies of fissionable materials. The LLL 14-MeV pulsed spheres¹⁻³ are especially sensitive to 14-MeV data. Linac-produced neutrons can test data in and somewhat above the fission energy spectrum.

Results of integral experiments with another type of neutron source were published recently by Bogart, Shook, and Fieno⁴ of the National Aeronautics and Space Administration's Lewis Research Center. They performed an especially simple integral experiment. An ^{241}Am -Be neutron source, which produces a spectrum of neutrons from 0 to 11 MeV, is surrounded by a concentric spherical shell of the material to be investigated.

The energy spectrum of neutron-induced proton recoils is measured by an organic scintillator (NE213) placed 2 m from the center of the sphere. After an unfolding operation, the neutron energy spectrum emerging from the shell is obtained.

The energy range of this experiment is between those used in previous tests of the LLL Evaluated Nuclear Data Library (ENDL).⁵ This energy range

tests our knowledge of the n, n' cross sections and emission spectra. The spheres are sufficiently thick (see Table 1) that the calculations are also sensitive to the total cross sections and to the cross sections and angular distributions of elastic scattering. Because of these important sensitivities, we decided to calculate the neutron emission spectra using the ENDL and to compare them to the experimental results.

Calculations

The calculations were performed with the Monte Carlo neutronics code, TART,⁶ using the ENDL (June, 1974, version) as processed by CLYDE⁷ into 175 energy-averaged groups. The histories of 10^5 source neutrons were followed for each case.

The source was taken to be the so-called "virgin α, n source spectrum" given by Bogart et al.⁴ It was assumed to be uniformly distributed

throughout the ^{241}Am -Be sphere. This source sphere is described⁴ as 16.7 g of ^{241}Am and 66.8 g of beryllium powder which are intimately mixed and pressed into a spherical shape with a volume of 83.4 cm^3 . This source sphere was clad by two stainless steel shells, 1.40 and 1.52 mm thick, with a total outer diameter of 6.1 cm and an inner diameter of 5.46 cm.

Table 1. Composition and size of test spheres. Taken from Bogart et al.⁴

Material	Form	Density (g/cm ³)	Outside diameter (cm)	Container ^a (cm)	Shell thickness, total mean free paths at 4 MeV (cm)	Shell thickness, total mean free paths at 4 MeV
CH ₂	Polyethylene	0.957	45.72	—	19.81	4.5
Be	Solid metal	.84	20.32	—	7.00	1.7
Nb	Solid metal	8.4	25.4	—	9.61	2.0
Mo	Powder	3.73	22.96	(0.05)	8.38	0.8
Ta	Solid metal	16.60	24.13	—	9.02	3.4
W	Fine balls	11.87	22.96	(0.05)	8.38	2.0
W	Fine balls	12.26	44.24	(0.48)	18.29	4.5
Pb	Solid metal	11.35	38.9	—	15.9	4.0

^a Container material was stainless steel.

Table 1, which is extracted from Ref. 4, gives the dimensions of the spherical shells. Thin stainless steel shells around the molybdenum and small tungsten spheres were assumed negligible for the neutronics calculations. Details of the large tungsten sphere, which was not completely described in Ref. 4, were provided by Bogart.⁸ This sphere consisted of a non-negligible, stainless steel container filled with fine tungsten balls. The 44.24-cm outer diameter of this sphere is made up of the 6.10-cm source sphere diameter, a 1.4-mm clearance (taken twice), an inner container wall of 1.6 mm stainless steel (taken twice),

the 18.29-cm thickness of the tungsten-filled region (taken twice), and the outer wall of 4.8 mm stainless steel (taken twice).

In the calculation, ^{242}Am was substituted for ^{241}Am because the former had been evaluated and the latter had not. These isotopes have markedly different fission cross sections at thermal neutron energy. However, the spectra in spheres is well above thermal. Even for the largest sphere, very little neutron-induced fission is calculated; it is about 0.004 fissions per source neutron. Thus, we conclude that the error resulting from this substitution is less than 1%.

Results

AMERICIUM-BERYLLIUM SOURCE

Because the dimensions of the source are comparable to the mean free paths for the neutrons of interest, the emission spectrum had to be calculated for the clad Am-Be sphere itself. It was crucial to calculate the source sphere correctly before calculating the source sphere plus spherical shells. The results are shown in Fig. 1. The agreement in shape between the calculated and experimental values is quite good. This result indicates that the source is well described by the virgin source and the evaluated data for beryllium and (less importantly) for americium. The calculated spectrum was normalized to the experiment by setting the virgin source strength to $1.26 \cdot 10^8$ neutrons/s which yields, through a small calculated multiplication,

Fig. 1. Neutron leakage spectrum for the ^{241}Am -Be source sphere. Error bars are the experimental points. The histogram is the calculation using the June, 1974, version of ENDL.

$1.30 \cdot 10^8$ neutrons/s emitted from the macroscopic source sphere. The measured value for the macroscopic sphere is $1.30 (\pm 0.08) \cdot 10^8$ neutrons/s.

The calculational results for the various spherical shells surrounding the source follow.

BERYLLIUM

For beryllium, the calculation agrees quite well with the experimental results

Fig. 2. Neutron leakage spectrum from the beryllium spherical shell surrounding the ^{241}Am -Be source sphere. Error bars are the experimental points. The histogram is the calculation using the June, 1974, version of ENDL.

with a slight difference in normalization below 3 MeV. (See Fig. 2.)

CH_2

For CH_2 , the calculated and experimental spectra are in excellent agreement. (See Fig. 3.)

NIObIUM

For niobium, the calculated values are systematically about 20% lower than the experimental values for emitted neutron energies above 1.5 MeV. (See Fig. 4.) Some correction to the nonelastic cross section is indicated.

Fig. 3. Neutron leakage spectrum from the polyethylene spherical shell surrounding the ^{241}Am -Be source sphere. Error bars are the experimental points. The histogram is the calculation using the June, 1974, version of ENDL.

MOLYBDENUM

For molybdenum, the calculated values agree closely with the experimental values. (See Fig. 5.)

TANTALUM

For tantalum, the calculated values are lower than the experimental values below 6.5 MeV. (See Fig. 6.) This com-

parison pointed out a problem in several n, n' cross sections for discrete level excitation which were evaluated inconsistently with the n, n' continuum cross section, the elastic cross section, and the total cross section. The discrete n, n' cross sections were evaluated unreasonably large in the 1- to 6-MeV range. (See Further Calculations section for the correction.)

TUNGSTEN

Both the small and large tungsten spheres are calculated fairly well with the ENDL. (See Figs. 7 and 8.) A difference in shape below 4 MeV indicates that the rather high emission temperature in the

Fig. 4. Neutron leakage spectrum from the niobium spherical shell surrounding the ^{241}Am -Be source sphere. Error bars are the experimental points. The histogram is the calculation using the June, 1974, version of ENDL.

Fig. 5. Neutron leakage spectrum from the molybdenum spherical shell surrounding the ^{241}Am -Be source sphere. Error bars are the experimental points. The histogram is the calculation using the June, 1974, version of ENDL.

June, 1974, version of the ENDL is probably in error. See Further Calculations section for corrections.)

neutron energy and disagree between 1 and 5 MeV. (See Fig. 9.) Some inadequacy is indicated in the n, n' cross section.

LEAD

For lead, the calculated values agree with the experimental values above 5 MeV

Fig. 6. Neutron leakage spectrum from the tantalum spherical shell surrounding the ^{241}Am -Be source sphere. Error bars are the experimental points. The histogram is the calculation using the June, 1974, version of ENDL.

Fig. 7. Neutron leakage spectrum from the small tungsten spherical shell surrounding the ^{241}Am -Be source sphere. Error bars are the experimental points. The histogram is the calculation using the June, 1974, version of ENDL.

Fig. 8. Neutron leakage spectrum from the large tungsten spherical shell surrounding the $^{241}\text{Am-Be}$ source sphere. Error bars are the experimental points. The histogram is the calculation using the June, 1974, version of ENDL.

Fig. 9. Neutron leakage spectrum from the lead spherical shell surrounding the $^{241}\text{Am-Be}$ source sphere. Error bars are the experimental points. The histogram is the calculation using the June, 1974, version of ENDL.

A Comment

A disagreement between calculated and experimental values indicates that at least one is inaccurate. In the preceding section, it has been tacitly assumed that the experimental results were correct. However, there could be errors

in the detector efficiency or in the procedure of unfolding the proton-recoil spectra to give the neutron spectra.

Evidence that there are no gross errors comes from the comparison of calculated and experimental results for

the CH_2 and beryllium spheres. For these materials, the microscopic data have been better determined experimentally than the microscopic data for the other materials. Consequently, the

evaluations are expected to be more reliable. The good agreement between the calculated and experimental results for these materials gives us confidence that both are without serious error.

Further Calculations

In view of the above findings, the ENDL evaluations of tantalum and tungsten were redone. The error in the tantalum evaluation was corrected and the n, n'

emission spectrum for tungsten was altered to be more consistent with systematics of neighboring nuclei. The results are shown in Figs. 10 and 11. No relevant data for niobium or lead were available to guide re-evaluations. Consequently, the niobium and lead evaluations were not changed.

Fig. 10. Neutron leakage spectrum from the tungsten spherical shell surrounding the ^{241}Am -Be source sphere. The histogram is the revised ENDL calculation. Error bars are the experimental points.

Fig. 11. Neutron leakage spectrum from the small tungsten spherical shell surrounding the ^{241}Am -Be source sphere. The histogram is the revised ENDL calculation. Error bars are the experimental points.

Conclusions

The neutron spectra emitted from spherical shells of various materials surrounding a 0- to 11-MeV radioactive neutron source are valuable tests of evaluated nuclear data. We have found that the ENDL evaluations of H, Be, C, and Mo are adequate to calculate the experimental

data. The evaluations existing in June, 1974, of Nb, Ta, W, and Pb had some deficiencies. Review of the microscopic data for tantalum and tungsten allowed us to correct their deficiencies. Relevant microscopic data for niobium and lead are needed to improve these evaluations.

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