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NEUTRON SPECTRA FROM MATERIALS USED IN FUSION AND FUSION-FISSION HYBRID REACTORS

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The neutron leakage spectra from pulsed spheres of Cu (1.0, 3.0 and 5.0 mean-free-path, mfp), Nb (1.0 and 3.0 mfp), ^{232}Th (1.0 mfp) and ^{238}U (0.8 and 2.8 mfp) have been measured using time-of-flight techniques. The neutron spectra from the above materials used in fusion hybrid reactors have been measured between 0.8 and 14 MeV using a stilbene scintillator, pulse shape discrimination and flight paths of around 10 meters. The measured spectra are compared with calculations carried out with TARTNP, a coupled neutron-photon Monte-Carlo transport code. The Lawrence Livermore Laboratory neutron and photon cross sections library (ENDL) and the ENDF/B-IV library have been used in these calculations. The calculated neutron spectra using the ENDL library are in fair agreement with the measurements (5 to 10% discrepancies for the 1-mfp spheres). Calculations for Cu, Nb and Th carried out with the ENDF/B-IV library, badly underestimate the neutron emission between 5 and 10 MeV. The discrepancies with the measurements in this energy region are as large as a factor of two. In order to facilitate the use of these data for calculational purposes by other laboratories, one-dimensional "experimental" neutron energy spectra have been generated for each of the above measurements and are available upon request.

Introduction

The optimum design of fusion and fusion-fission hybrid reactors requires the use of accurate codes and cross sections which correctly describe the interaction of 14-MeV neutrons with materials used in the reactor designs. These designs will be judged by their performance characteristics (tritium breeding ratio, energy multiplication, power density, fuel burnup, etc.), as predicted by the neutronic calculations. The accuracy of these latter calculations in turn is dependent upon the physical and mathematical assumptions and the correctness of the evaluated cross sections used in the transport codes. Because the complexity of the designs tends to obscure the sources of discrepancies between the calculations and any direct measurements, it is preferable to make measurements and calculations for a single material in a simple geometry [1]. Agreement between calculations and measurements for each constituent material of the reactor is then a necessary prerequisite for obtaining reliable hybrid reactor calculations.

As part of the pulsed sphere program at Livermore, measurements of the neutron and gamma emission spectra from a large number of materials ranging from Li to Pu have been carried out using the sphere transmission and time-of-flight (TOF) techniques. In the present paper the measurements on Cu and Nb (materials of interest to magnetic and laser fusion) and the measurements on ^{232}Th and ^{238}U (used in the fission blanket of the hybrid reactor) are presented.

Experimental Method

The neutron spectrum measurements have been carried out using pulsed spheres and TOF techniques. Nominal 14-MeV neutrons are obtained from the T(d,n)^{4}He reaction using the 400-keV D^+ beam from the Livermore Insulated-Core-Transformer (ICT) accelerator. The beam is swept and bunched with a repetition rate of 0.5 MHz and a burst width of ∿2 ns. The tritium-titanium target, having a tungsten backing 2.06 cm in diameter and 0.051 cm thick, is mounted at the end of a low mass assembly and is positioned at the center of the spherical targets. A solid state detector, positioned at 174° with respect to the D^+ beam line, monitors the neutron production by counting the associated ^{4}He particles. The neutron detectors, 5.08-cm diameter by 5.08-cm long NE 213 or stilbene scintillators, are located at around 10 meters from the center of the spheres at 26 and 120°. With stilbene, the neutron spectra are measured down to 0.8-MeV neutron energy while with the NE 213 detector, the low energy cutoff is 2 MeV. A detailed description of the low mass tritium target assembly, neutron detector packages, collimators and electronics used in these measurements is given in Ref. 2. The determination of the efficiency of the NE 213 and stilbene detectors has been published elsewhere [2,3]. A schematic drawing of the experimental setup is shown in Fig. 1.

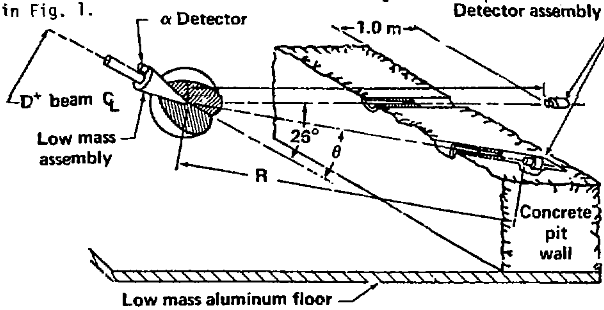

Figure 1. Schematic drawing of the experimental setup

1. Hansen, L. F., et al., "Measurements of the Neutron Spectra from Materials Used in Fusion Reactors and Calculations using the ENDF/B-III and IV Neutron Libraries," Nucl. Sci. Eng. 60, 27 (1976).
2. Wong, C., et al., "Livermore Pulsed Sphere Program: Program Summary Through July 1971," UCRL-51144, Rev. I, Lawrence Livermore Laboratory (1972).
3. Hansen, L. F., et al., "Neutron and Gamma Spectra from ^{232}Th, ^{235}U, ^{238}U and ^{239}Pu Bombarded with 14-MeV Neutrons," (to be published).

Experimental Results and Calculations

The neutron spectra have been measured at 26° and 120° with respect to the deuteron beam line using NE 213 detectors and have been remeasured using a stilbene detector at 26°. Because the spectra at the two angles are very similar and the stilbene measurement extended to lower energies (0.8 MeV compared to 2 MeV), only the stilbene results are presented. The calculations have been carried out with TARTNP [4], a coupled neutron-photon Monte Carlo transport code. For each neutron reaching the detector, the code calculates the energy, time-of- arrival direction cosines and the spatial coordinates. The Monte Carlo calculation of both the time-of-arrival and energy is very useful when large spheres of fissionable materials are being measured. High energy neutrons from secondary fission events in the large spheres will reach the detector at later times and they might be misinterpreted as low energy neutrons because of their apparently longer time of arrival. In this case, only the comparison between the measured and calculated TOF spectra is exactly correct, while the comparison between the Monte Carlo calculated energy spectrum and the experimental spectrum obtained by transforming the TOF spectrum directly to an energy spectrum will be incorrect. This latter transformation is done relativistically assuming a flight path (L) equal to the distance between the detector and tritium target. The transformation is only valid if the sphere radius is much smaller than L. This is the case for all the spheres discussed in this paper.

In order to compare the calculations with the measured TOF spectra, the efficiency of the detector as a function of neutron energy and the time resolution of the system are folded into the calculations. To properly account for time-dependent background effects due to scattering from the low mass tritium target assembly, the collimators, the materials surrounding the detectors and the air between the spheres and the detectors, all these sources of background scattering are included in the Monte Carlo calculations. In addition, the description [2] of the 14-MeV neutron source (the variations in energy and intensity of the neutrons from the $T(d,n)^4He$ reaction) and the time spread of the deuteron beam from the ICT accelerator are also included in the calculations.

Cu and Nb

The neutron emission spectra have been measured for 1.0 (32.6 g/cm^2), 3.0 (102.8 g/cm^2), and 5.0 (174.5 g/cm^2) mfp copper spheres and for 1.0 (31.3 g/cm^2), 3.0 (120.9 g/cm^2) mfp niobium spheres. Figure 2a shows the measured TOF spectrum for the 1-mfp Cu and Fig. 2b the 1-mfp together with the calculations

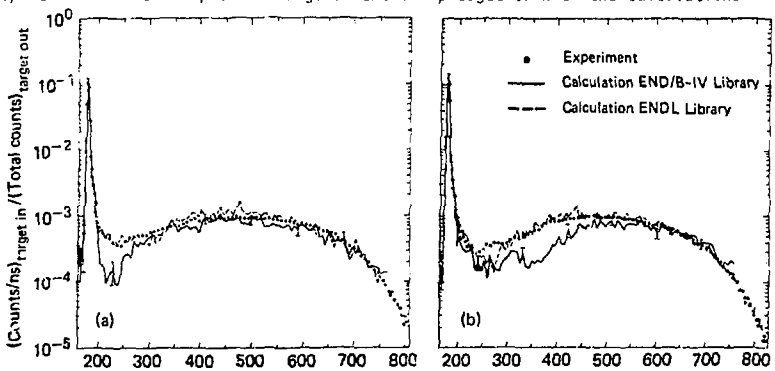

Figure 2. Measured TOF spectra for 1 mfp of Cu (a) and Nb (b). The calculated spectra are shown for the ENDF/B-IV (——) and the ENDL (----) libraries.

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4. Plechaty, E. F., et al., TARTNP Coupled Neutron-Photon Monte Carlo Transport Code, UCRL 50400, Vol. 14, Lawrence Livermore Laboratory, December 1975.

carried out with the ENDL [5] and the ENDF/B-IV [6] libraries. The large discrepancy observed for the Cu in the region of 5 to 12 MeV between the measurements and the ENDF/B-IV calculation is analogous to that observed earlier for iron [7] with version III of the ENDF/B library. The above ENDF/B calculations seriously underestimate the production of high energy neutrons from pre-equilibrium processes; the neutron emission from (n,n') inelastic scattering is assumed to be almost completely the result of a compound reaction mechanism. From a total of 650 mb assigned by ENDF/B-IV to the (n,n') reaction in Cu, 620 mb are described by a temperature model (T = 3 MeV for 14-MeV neutrons) and 40 mb are assigned to inelastic levels between 0.67- and 1.86-MeV excitation energy. The (n,n') cross section in the ENDL library (550 mb at 14 MeV) is represented by a continuous energy distribution peaking at 4 MeV and extending up to 14 MeV. The cross sections for Cu at 14 MeV for the two libraries are listed in Table I. The (n,2n) cross sections for Cu are close in magnitude but the temperature is 0.95 MeV in the ENDL and 0.35 MeV in the ENDF/B-IV.

Table I

Material	Library	σ_t	σ_{el}	$\sigma_{n,n'}$	$\sigma_{n,2n}$	$\sigma_{n,n'p}$	$\sigma_{n,n'\alpha}$	$\sigma_{n,p}$	$\sigma_{n,\alpha}$	$\sigma_{n,\gamma}$
Cu	ENDL	2.91	1.50	.55	.64	.112	.007	.087	.018	.0006
	B-IV	2.96	1.46	.66	.60	.113	.003	.079	.033	.0023
Nb	ENDL	3.98	2.20	.36	1.37	--	.0025	.040	.0092	.0008
	B-IV	3.98	2.30	.416	1.215	--	.0025	.040	.0092	.0008

For the 1-mfp Nb sphere (Fig. 2b) the ENDL library reproduces measurements better than the ENDF/B-IV library, which underestimates the neutron production between 0.8 and 10 MeV by more than 50%. It should be mentioned that both libraries originally had the same evaluations for Nb. However, the Nb cross section in the ENDL library was recently re-evaluated [8], resulting in the improved agreement shown in Fig. 2b. Table I shows that the Nb cross sections in the two libraries differ mainly in the magnitude of the elastic, the (n,n') and (n,2n) cross sections. In the ENDL library, the 360 mb assigned to (n,n') are represented by a continuous energy distribution between 3 and 13.9 MeV, while in the ENDF library, only 60% of the 416 mb assigned to (n,n') have the same representation between 2.49 and 13.9 MeV. The remaining 30% (124 mb) is described by a temperature spectrum (T = 0.94 MeV). The temperatures for the (n,2n) distributions are 0.85 MeV in the ENDL and 0.56 MeV in the ENDF/B-IV.

The general features of the calculations relative to the measurements shown in Fig. 2 persist in the comparisons for the larger size spheres. The measured and calculated integrals for the energy intervals 0.8-5, 5-10 and 10-15 MeV are tabulated in Table II. With the ENDL library, the discrepancies with measurements are not larger than 25% for Cu and Nb. The ENDF/B-IV underestimates by almost a factor of two the neutron production below 10 MeV for both materials.

Table II

		1 mfp			3 mfp			5 mfp		
Mat	ΔE	Exp ± 7%	ENDF ± 2%	B-IV ± 2%	Exp ± 7%	ENDF ± 2%	B-IV ± 2%	Exp ± 7%	ENDF ± 2%	B-IV ± 2%
Cu	0.8-5	0.313	0.343	0.281	0.465	0.430	0.379	0.285	0.238	0.210
	5.0-10	0.041	0.040	0.025	0.037	0.042	0.019	0.017	0.021	0.010
	10.0-15	0.642	0.627	0.634	0.236	0.221	0.224	0.082	0.078	0.075
Nb	0.8-5	0.308	0.333	0.235	0.422	0.402	0.265			
	5.0-10	0.031	0.025	0.018	0.033	0.029	0.021			
	10.0-15	0.707	0.686	0.698	0.265	0.233	0.238			

5. Howerton, R.J., et al., the LLL Evaluated Nuclear Data Library (ENDL), UCRL 50400, Vol. 15, Parts A to E, Lawrence Livermore Laboratory (1975-1978).
6. ENDF/B Summary Documentation BNL 17541 (ENDF-201), National Neutron Cross Section Center, Brookhaven National Laboratory (compiled by D. Garber).
7. Hansen, et al., "Measurements and Calculations of the Neutron Spectra from Iron Bombarded with 14-MeV Neutrons," Nucl. Sci. and Eng. 51, 278 (1973).
8. Howerton, R. J., Private Communication, Lawrence Livermore Laboratory, November 11, 1977.

^{232}Th and ^{238}U

Neutron spectrum measurements have been made for a 1 mfp (67.1 g/cm^2) Th sphere and for 0.8 mfp (69.2 g/cm^2) and 2.8 mfp (207.7 g/cm^2) ^{238}U spheres. Figure 3 shows the measured and calculated TOF spectra for the Th (a) and ^{238}U (b) 1 mfp spheres. For Th, the discrepancy between the ENDF/B-IV calculations and the measurements is similar to that observed for Cu. The 370 mb assigned by ENDF/B-IV to the (n,n') reaction at 14 MeV for Th (see Table III) is assumed to be totally ascribed to a compound reaction mechanism with a temperature of 0.54 MeV. The

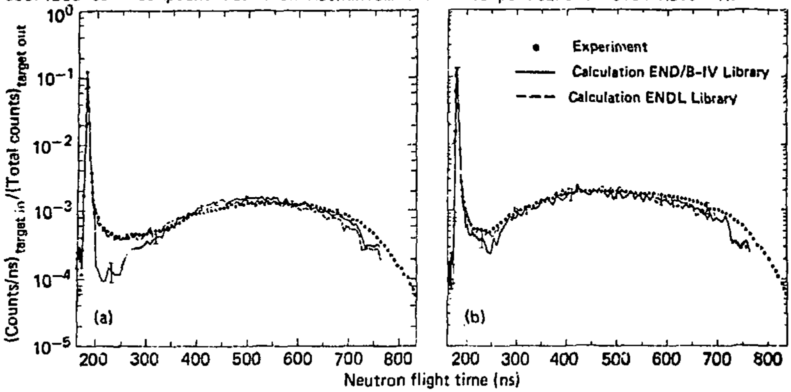

Figure 3. Measured TOF spectra for 1 mfp of Th (a) and 0.8 mfp of ^{238}U (b). The calculated spectra are shown for the ENDF/B-IV (——) and the ENDL (----) libraries.

ENDL library assigns 550 mb to the (n,n') on Th and the inelastic spectrum is represented by a continuous energy distribution extending up to 14 MeV. The (n,2n) cross sections on Th are 1300 mb (ENDL) and 1560 mb (ENDF/B-IV) with a temperature of 0.75 MeV. Although Table III shows large differences for the partial cross sections for Th for the two libraries, the total and non-elastic cross sections differ by less than 2%.

Table III

Material	Library	σ_t	σ_{el}	$\sigma_{n,n'}$	$\sigma_{n,2n}$	$\sigma_{n,3n}$	σ_{nf}	$\sigma_{n\gamma}$
^{232}Th	ENDL	5.71	2.85	.55	1.30	.65	.355	.0054
	B-IV	5.70	2.90	.37	1.56	.60	.36	.0053
^{238}U	ENDL	5.85	2.98	.403	.897	.434	1.129	.001
	B-IV	5.79	2.99	.313	.855	.495	1.140	.001

For ^{238}U, the shape of the measured spectrum is better represented by the ENDL calculation. The discrepancy between the ENDF/B-IV calculations and measurements in the 7- to 10-MeV energy region seen in Fig. 3, could be due to an incorrect description of the (n,n') cross section in the library. Four pseudo levels at 1.95, 2.95, 3.95 and 4.95 MeV account for 51% of the (n,n') cross section of 313 mb. The absence of levels above ~4-MeV excitation energy accounts for the smaller calculated integral around 8-10 MeV. The ENDL library has instead an (n,n') cross section of 403 mb, 99.3% of which is described by a continuous energy distribution.

The measured and calculated integrals in the energy intervals 0.8-5, 5-10 and 10-15 MeV obtained from the TOF spectra are tabulated in Table IV. For ^{232}Th, both libraries yield integrals in good agreement with the measurements for the regions 0.8 to 5 and 10 to 15 MeV. From 5 to 10 MeV, the ENDL library underestimates the neutron production by less than 3% while the ENDF/B-IV underestimates it by about 100%. For ^{238}U, the two libraries reproduce very well the integral in the 10-15 MeV interval while in the region of 5 to 10 MeV the ENDF/B-IV underestimates the magnitude of the integral by $\sim$25%.

Table IV

Mat	E	Exp. ± 7% (0.8 mfp)	ENDF ± 2% (0.8 mfp)	B-IV ± 2% (0.8 mfp)	Exp. ± 7% (1 mfp)	ENDF ± 2% (1 mfp)	B-IV ± 2% (1 mfp)	Exp. ± 7% (2.8 mfp)	ENDF ± 2% (2.8 mfp)	B-IV ± 2% (2.8 mfp)
^{232}Th	0.8-5				0.442	0.438	0.467			
	5.0-10				0.039	0.038	0.020			
	10.0-15				0.645	0.636	0.628			
^{238}U	0.8-5	0.670	0.644	0.595				0.803	0.905	0.846
	5.0-10	0.059	0.055	0.046				0.063	0.060	0.052
	10.0-15	0.669	0.639	0.645				0.232	0.225	0.237

Conclusions

The neutron emission spectra from Cu (1, 3 and 5 mfp), Nb (1 and 3 mfp), ^{232}Th (1 mfp) and ^{238}U (0.8 and 2.8 mfp) have been measured using pulsed sphere and TOF techniques. The calculations with the ENDL [5] and ENDF/B-IV [6] libraries reproduce well the neutron integrals between 10 and 15 MeV. The good agreement suggests that the non-elastic cross sections in both libraries for the above materials are correct. Below 10 MeV, calculations using the ENDL library are in fair agreement ($\lesssim$ 20%) with the measurements for all materials studied. The ENDF/B-IV library underestimates the neutron production between 5 and 10 MeV: for Cu and Th, the discrepancy is as large as a factor of two, 60 to 70% for Nb, and 30% for ^{238}U. This discrepancy arises mainly from assuming an incorrect energy distribution for the (n,n') neutrons. Specifically, the production of high energy neutrons from the pre-equilibrium process is not properly accounted for. Below 5 MeV the ENDF/B-IV underestimates the neutron production by 20 to 30% for the larger size spheres. A one-dimensional "experimental" equivalent spectrum has been generated for each of the measurements presented in this paper. These normalized spectra have been calculated using the code TARTNP with the following assumptions: 1) The targets are solid spheres with no conical apertures. 2) The 14-MeV source, positioned at the exact center of the sphere, is taken to be isotropic and monoenergetic. Because of the limited space available for this presentation, the tabulated data for these one-dimensional spectra are available upon request.

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