MICROSCOPIC INTEGRAL CROSS SECTION MEASUREMENTS IN THE Be(d,n)
NEUTRON SPECTRUM FOR APPLICATIONS IN NEUTRON DOSIMETRY,
RADIATION DAMAGE AND THE PRODUCTION OF LONG-LIVED
RADIONUCLIDES*

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ABSTRACT

Integral neutron-reaction cross sections have been measured, relative to the U-238 neutron fission cross-section standard, for 27 reactions which are of contemporary interest in various nuclear applications (e.g., fast-neutron dosimetry, neutron radiation damage and the production of long-lived activities which affect nuclear waste disposal). The neutron radiation field employed in this study was produced by bombarding a thick Be-metal target with 7-MeV deuterons from an accelerator. The experimental results are reported along with detailed information on the associated measurement uncertainties and their correlations. These data are also compared with corresponding calculated values, based on contemporary knowledge of the differential cross sections and of the Be(d,n) neutron spectrum. Some conclusions are reached on the utility of this procedure for neutron-reaction data testing.

KEY WORDS: Be(d,n) neutron field, neutron-induced reactions, integral cross sections, integral/differential comparisons, spectrum adjustment

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INTRODUCTION

Integral measurements in continuum neutron fields can provide valuable information for the testing of differential neutron cross section data. In fact, when the yield of activity from a particular reaction is low, integral measurements may very well offer the only possibility for acquiring experimental information. Such measurements are often made in reactors, where degraded fission-neutron spectra with relatively low average energies are encountered. However, the intense thermal and epithermal neutron fluences found in reactors can generate strong interfering activities which complicate the measurement of weaker processes. A variety of fast-neutron fields (with relatively small low-energy components) can be conveniently produced using accelerators. We have explored applications for the neutron field produced by bombarding thick Be-metal targets with 7-MeV deuterons. This work was first reported at the Sixth ASTM-Euratom Symposium (hereafter referred to as Paper I) [1]. You are referred to Paper I (and references therein) for details of the method. The present paper documents the experimental results which have been obtained from this project.

EXPERIMENTAL PROCEDURE

The facility and methodology employed for these measurements is described more fully in Paper I and references therein. Nevertheless, a brief summary is provided here for completeness. A compact, continuum source of neutrons is produced by directing a collimated beam of 7-MeV deuterons onto a Be-metal target which is sufficiently thick to stop the deuterons. The maximum neutron energy is ≈ 11.5 MeV. Recent neutron time-of-flight measurements with a fission chamber have improved our knowledge of this spectrum at low energies [2]. Relatively few neutrons with energies < 200 keV are observed in this spectrum when a low-mass Be-target assembly is used and the presence of extraneous scatterers in the irradiation environment is kept to a minimum [1,3-5]. Activation samples are affixed to a fission chamber which also serves to measure neutron fluence via the U-238 neutron-fission cross-section standard. The neutron-induced activities are deduced using conventional techniques [4,6,7]. Samples of fissionable material (assayed by low-geometry alpha counting [8]) are placed either singly or back-to-back in a fission chamber. Fission yields are measured in real time during the irradiation [4,8]. The measurements reported here were carried out in three separate experiments. Accordingly, the 27

accumulated data points are grouped into three distinct sets: Set I (Nos. 1-5), Set II (Nos. 6-12) and Set III (Nos. 13-27).

DATA ANALYSIS AND EXPERIMENTAL ERRORS

A number of corrections were applied to the raw experimental data to convert them to integral cross section ratios. Relevant details are provided in Refs. 4 and 8, and other documents cited therein. Table 1 lists the reactions which were investigated in this work and also gives the decay half lives and detected radiations [9,10]. The principle data corrections account for: i) low-energy fission events masked by noise, alpha particles and gas-recoil pulses; ii) loss of fission fragments in the deposit; iii) decay of the induced sample activities; iv) radiation branching parameters; v) detector calibrations; vi) neutron absorption and multiple scattering; vii) geometry effects; and, viii) anisotropic emission of the Be(d,n) neutrons.

Experimental errors were estimated as described in Refs. 4 and 11. Table 2 summarizes the principle sources of error which were considered in this work. These errors were combined to produce a data covariance matrix, following the procedure described in Ref. 12.

EXPERIMENTAL RESULTS

The measured integral cross section ratios appear in Table 1. Corresponding errors (standard deviations) and their correlations are found in Tables 1 and 3, respectively. These measured ratios can be converted to integral cross sections by employing the absolute integral cross-section (for the present Be(d,n) spectrum) of the standard reaction (U-238 neutron fission). The value 0.46097 barn was used for this purpose. It was calculated using the ENDF/B-V [13] evaluated differential U-238 fission cross section and a representation for the Be(d,n) spectrum which was derived as described in the following section.

NEUTRON SPECTRUM ADJUSTMENT

Prior to the present work, knowledge of the thick-target Be(d,n) neutron spectrum for 7-MeV incident deuterons was based entirely on data reported in Refs. 3,5 and 14, and on unpublished results from this laboratory [2]. The first step taken to improve

this situation was to generate a hybrid zero-degree spectrum representation which incorporated the best features of earlier time-of-flight measurements (taken with a scintillation detector for neutron energies above 4 MeV [14] and with a fission chamber for lower energies [2]). This spectrum served as the prior for a spectrum adjustment procedure (utilizing code STAYS'L [15]) which incorporated all 27 reactions from Table 1. This procedure required uncertainty information for both the trial spectrum and for the differential cross sections. Uncertainties for the spectrum were deduced from statistics and systematic errors associated with the time-of-flight measurements. Uncertainties for the differential cross sections were derived from ENDF/B-V covariance files (where available) or were estimated. This analysis led to a modest increase in the neutron fluence at energies below 4.5 MeV (< 5%), a modest decrease in the range 4.5-5.5 MeV (< 5%), and a significant decrease at higher energies (5-25%). As a result, the calculated integral cross sections for low-threshold reactions changed by only small amounts whereas significant reductions were observed for the high-threshold reactions. The adjustment procedure produced a chi-square of 1.36 per degree of freedom which signifies rather good overall consistency between the integral and differential information. A point-wise representation for the resulting normalized Be(d,n) spectrum is given in Table 4.

COMPARISON OF MEASURED AND CALCULATED CROSS SECTIONS

A comparison has been made between the experimental (E) integral cross sections (see Table 1 and the discussion above) and those calculated (C) using the spectrum given in Table 4 (see preceding section). ENDF/B-V [13] values (generally obtained from the Dosimetry File) were employed for this analysis except as noted for the following reactions: Ti-47(n,p)Sc-47 [16]; Co-59(n,p)Fe-59, Co-59(n,2n)Co-58 and Co-59(n, α)Mn-56 [17]; Zn-64(n,p)Cu-64 [18]; Cu-65(n,p)Ni-65 [19]; In-113(n,n')In-113m [19]; and In-115(n,n')In-115m [20]. The resultant C/E comparisons are shown schematically in Fig. 1, except for Co-59(n,2n)Co-58 which is not shown because the agreement is poor (C/E = 2.38). Rough estimates were made of the errors in the calculated results (they are not given in this paper) in order to assist in making a judgement on the consistency of the integral and differential information. Overall, the agreement is found to be quite good. Eighteen reactions out of 27 (67%) exhibit acceptable consistency. The C/E comparisons were marginally consistent with unity (only slightly beyond the combined errors) for four

reactions (15%): Al-27(n, α)Na-24, U-234(n,fission), In-113(n,n')In-113m and In-115(n,n')In-115m. Only five reactions (18%) were indisputably discrepant: Li-7(n,n't)He-4, Al-27(n,p)Mg-27, Ni-60(n,p)Co-60, Co-59(n, α)Mn-56 and Co-59(n,2n)Co-58. Most of the integral response for these reactions comes from neutron energies above 5 MeV where the neutron spectrum uncertainty is the greatest. The differential cross section for Li-7(n,n't)He-4 is poorly known near threshold while that for Ni-60(n,p)Co-60 is not very well established at any energy. It is not surprising that the agreement should be very poor for Co-59(n,2n)Co-58 since its reaction threshold is quite high (10.633 MeV). There is no explanation for the poor agreement observed for Al-27(n,p)Mg-27 and Co-59(n, α)Mn-56. Their differential cross sections were thought to be fairly well established in the energy range of maximum response.

CONCLUSIONS

It is concluded from the results of this work that the accelerator neutron field produced by 7-MeV deuteron bombardment of thick Be metal targets is now reasonably well established and, furthermore, is quite useful for the integral testing of differential neutron cross-section data, so long as the effective reaction threshold is of the order of a few MeV or less. The relatively high yield of neutrons from this process (about 3×10^9 n/sr/ μ C at zero degree [21]) adds to its utility for investigating certain low-yield reactions at a small accelerator facility, relative to alternative techniques. However, for reactions involving higher effective thresholds the Be(d,n) source loses much of its appeal due to anisotropic neutron emission (necessitating large corrections) and to the relatively low yield of neutrons above 6 MeV when the incident deuteron energy is limited to 7 MeV. Naturally, this is not a fundamental limitation to the basic method discussed in this paper. The Be(d,n) source can be used very effectively to study higher-threshold reactions when greater deuteron bombarding energies can be obtained (e.g., see Ref. 22).

It is clear that future refinements in the characterization of the Be(d,n) spectrum (particularly for neutron energies above 6 MeV) and improvements in the differential cross sections will lead to a significant reduction in the measurement and computational uncertainties, and therefore to improvement in the C/E comparisons for those particular reactions studied in this work. However, the present status already offers sufficient

confidence in the basic method to encourage applications of this approach to the study of certain exotic reactions which are not as well known as most of those considered here, e.g., to various processes which yield long-lived activities implicated in low-level nuclear waste management technologies.

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Table 1: Reaction parameters and experimental results

Data Point	Reaction	Half ^a Life	Detected ^b Radiation	Measured ^c Ratio	Error ^d (%)
Set I:					
1	Li-7(n,n't)He-4	12.33 y	β^- [t]	0.1217	4.0
2	Al-27(n,p)Mg-27	9.462 m	γ	0.02380	3.9
3	Al-27(n, α)Na-24	15.020 h	γ	0.006155	5.9
4	Ni-58(n,p)Co-58	70.916 d	γ	0.5298	4.1
5	Ni-60(n,p)Co-60	5.271 y	γ	0.01750	3.7
Set II:					
6	Th-232(n,fission)	Various	FF	0.2579	2.2
7	U-233(n,fission)	Various	FF	4.053	1.7
8	U-234(n,fission)	Various	FF	2.894	1.6
9	U-235(n,fission)	Various	FF	2.646	1.4
10	U-236(n,fission)	Various	FF	1.646	1.7
11	Np-237(n,fission)	Various	FF	3.296	1.5
12	Pu-239(n,fission)	Various	FF	3.995	2.0
Set III:					
13	Ti-46(n,p)Sc-46	83.83 d	γ	0.07371	4.6
14	Ti-47(n,p)Sc-47	3.345 d	γ	0.08769	6.7
15	Ti-48(n,p)Sc-48	43.7 h	γ	0.002526	6.5
16	Fe-54(n,p)Mn-54	312.5 d	γ	0.4205	4.8
17	Fe-54(n, α)Cr-51	27.704 d	γ	0.007011	10.5
18	Fe-56(n,p)Mn-56	2.5785 h	γ	0.007349	5.6
19	Co-59(n,p)Fe-59	44.496 d	γ	0.009159	5.6
20	Co-59(n,2n)Co-58	70.916 d	γ	0.0003028	14.8
21	Co-59(n, α)Mn-56	2.5785 h	γ	0.001192	6.2
22	Cu-63(n, α)Co-60	5.271 y	γ	0.004122	7.3
23	Cu-65(n,p)Ni-65	2.520 h	γ	0.003770	11.6
24	Zn-64(n,p)Cu-64	12.701 h	γ	0.1745	4.0
25	Nb-93(n,2n)Nb-92m	10.15 d	γ [IT]	0.004780	9.8
26	In-113(n,n')In-113m	1.658 h	γ [IT]	0.5248	6.3
27	In-115(n,n')In-115m	4.486 h	γ [IT]	0.6173	3.7

^a Reaction product half lives from Ref. 9. Fission produces many half lives. Units: m (minute), h (hour), d (day), y (year).

^b Detected radiations: β^- [t], tritium beta rays; γ , gamma rays; γ [IT], isomeric-transition gamma rays; FF, fission fragments.

^c Integral cross-section ratios in the Be(d,n) neutron field for the indicated reaction relative to U-238 neutron fission. Multiply these ratios by 0.46097 barn to obtain absolute integral cross sections (see text).

^d Total error (random and systematic) in the measured ratio.

Table 2: Sources of experimental error

Description of Error	Magnitude (%) ^a
Random:	
1. Fission counting (statistics and spectrum extrapolation)	0.3 - 2.4
2. Beta- and gamma-ray counting statistics	0.3 - 3.0
3. Random geometry effects	N - 1.4
4. Measurement reproducibility (unidentified random errors)	N - 10.3
Systematic:	
1. Number of atoms in fission monitor deposit	0.5 - 2.0
2. Effects of fission deposit thickness, detector deadtime and thermal background	0.3 - 1.5
3. Half-life uncertainty	N - 0.4
4. Radiation detector efficiency calibration	0.6 - 3.1
5. Number of atoms in the sample	0.1 - 4.7
6. Radiation decay branching parameter	N - 5.9
7. Neutron absorption	0.1 - 0.5
8. Neutron multiple scattering	N - 1.5
9. Systematic geometry effects	N - 1.4
10. Neutron source properties	N - 10.1

^a N = Negligible.

Table 3: Experimental error correlations^a

	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15
	16	17	18	19	20	21	22	23	24	25	26	27			
1	100														
2	34	100													
3	28	55	100												
4	30	63	43	100											
5	41	62	52	46	100										
6	5	6	12	2	12	100									
7	1	2	1	1	2	44	100								
8	7	8	17	3	16	49	69	100							
9	2	2	1	2	2	40	71	63	100						
10	6	7	16	3	15	47	60	68	53	100					
11	7	8	18	3	17	53	65	68	67	60	100				
12	1	1	1	1	1	43	66	74	54	64	63	100			
13	14	21	25	16	29	9	1	12	2	11	13	1	100		
14	7	11	9	10	13	2	1	3	1	3	3	1	22	100	
15	18	24	42	13	44	19	1	26	1	25	28	1	45	18	100
16	10	16	13	14	18	3	1	4	1	4	4	1	28	18	24
17	100	7	10	14	7	15	5	1	7	1	7	8	0	18	9
18	20	100	18	25	40	15	43	18	1	24	1	22	26	1	44
19	29	24	100	12	17	21	13	25	7	1	10	1	10	11	1
20	20	14	34	100	12	16	32	7	31	15	0	21	0	20	22
21	12	15	48	25	100	12	15	48	25	100	0	20	22	0	26
22	18	24	40	14	42	18	1	24	1	23	26	1	40	16	64
23	22	22	62	38	50	100	16	21	37	12	38	17	1	23	24
24	22	21	59	30	45	57	100	22	21	59	30	45	57	100	22
25	6	8	10	6	12	4	1	5	1	5	5	0	13	7	16
26	10	7	17	13	12	18	15	100	10	7	17	13	12	18	15
27	13	20	18	17	24	4	1	6	2	6	7	1	31	19	29
28	26	14	31	30	18	34	26	15	100	17	22	43	10	43	21
29	17	22	43	10	43	21	1	28	1	27	30	1	39	13	71
30	19	22	67	32	54	66	63	15	24	100	19	22	67	32	54
31	7	12	9	11	13	1	1	2	1	2	2	1	17	11	13
32	15	8	15	17	8	17	12	8	23	10	100	12	20	15	18
33	12	20	15	18	22	2	2	3	2	3	4	1	28	19	23
34	26	13	25	29	13	28	20	14	39	17	25	100			

^a Correlations between errors given in Table 1 (in percent).
Matrix rows and columns labelled by data point number.

Table 4: Energy spectrum for Be(d,n) neutrons near zero degrees^a

E ^b	ϕ ^b	E	ϕ	E	ϕ	E	ϕ
< 0.1826	≈0.0	4.5600	.13930	6.4000	.02161	8.7400	.01341
0.2238	.09214	4.6500	.13519	6.4700	.01996	8.8500	.01362
0.2693	.10542	4.7300	.13315	6.5500	.01848	8.9700	.01316
0.3178	.10773	4.8500	.13168	6.6200	.01721	9.0900	.01279
0.3662	.11597	4.9700	.13088	6.7000	.01664	9.2100	.01231
0.4373	.11234	5.0400	.12816	6.8100	.01604	9.3300	.01096
0.5328	.11993	5.0900	.12347	6.9700	.01610	9.4600	.01024
0.6302	.12479	5.1400	.11960	7.0900	.01558	9.5900	.00939
0.7262	.13649	5.1900	.11802	7.1700	.01629	9.7200	.00848
0.8220	.18599	5.2400	.11233	7.2600	.01642	9.8500	.00776
0.91856	.18047	5.3000	.10482	7.3500	.01634	9.9900	.00756
1.05474	.17289	5.3500	.10209	7.4300	.01659	10.130	.00728
1.24055	.18021	5.4300	.09985	7.5200	.01726	10.270	.00720
1.4313	.18697	5.5500	.09753	7.6100	.01647	10.420	.00662
1.61385	.19302	5.6300	.09538	7.7100	.01583	10.560	.00603
1.7949	.20674	5.7200	.09522	7.8000	.01469	10.720	.00536
2.1208	.19354	5.8400	.09355	7.9000	.01454	10.870	.00462
2.5803	.19934	5.9400	.09020	8.0000	.01424	11.030	.00416
3.03595	.19832	6.0000	.08644	8.1000	.01448	11.190	.00356
3.25723	.18923	6.0600	.07440	8.2000	.01371	11.350	.00313
3.4785	.18034	6.1300	.05065	8.3000	.01386	11.370	.00302
3.9009	.17493	6.2000	.03200	8.4100	.01407	11.450	.00137
4.3500	.14546	6.2600	.02596	8.5100	.01380	11.550	.00012
4.4600	.14340	6.3300	.02333	8.6200	.01372	≥11.650	≈0.0

^a Neutron radiation field produced by 7-MeV deuterons incident on a thick Be-metal target. Energy (E) in MeV. Energy-differential flux (ϕ) in neutron/MeV. The flux is normalized to yield unit total fluence (a single neutron) when integrated over the complete energy spectrum.

Figure 1: Comparison of experimental (E) and calculated (C) integral cross sections, as described in the text. All experimental values are centered on unity (horizontal bars indicate their range based on the errors) while the calculated quantities (solid squares) exhibit the deviations of C/E ratios from unity. No errors are provided for the calculated results.