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ACTIVATION CROSS SECTION MEASUREMENTS NEAR THRESHOLD FOR THE $^{24}Mg(n,p)^{24}Na$ AND $^{27}Al(n,\alpha)^{24}Na$ REACTIONS

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Abstract—Differential cross sections have been measured for the $^{24}Mg(n,p)^{24}Na$ and $^{27}Al(n,\alpha)^{24}Na$ reactions in the neutron energy range from near-threshold to approx. 10 MeV using ^{238}U fast-neutron fission as a cross section standard. The present data generally support previous work, although the cross sections tend to be somewhat larger for $^{27}Al(n,\alpha)^{24}Na$, particularly in the 8–9 MeV range. These data contribute significantly to reducing the uncertainty in contemporary knowledge of the cross sections for these reactions in the threshold region.

1. INTRODUCTION

The $^{24}Mg(n,p)^{24}Na$ and $^{27}Al(n,\alpha)^{24}Na$ reactions are both important neutron dosimetry processes for fission and fusion energy applications (e.g. Cheng, 1986). For fusion, knowledge of the cross sections in the vicinity of 14 MeV is naturally of paramount importance since the neutrons incident upon the first wall of a fusion reactor operating on the D–T fuel cycle will be predominantly of this energy. However, elsewhere in such a device, e.g. in the T breeding blanket, the neutron energy spectrum will be considerably degraded so that knowledge of dosimeter cross sections at lower energies is also quite important. In fission systems the interest is generally in the neutron spectra at energies below 10 MeV. For example, an examination of the integral response functions for both reactions in a pure ^{252}Cf spontaneous-fission neutron spectrum (Mannhart, 1987) clearly shows that most of the ^{24}Na activity generated in Mg and Al dosimeter samples placed in fission-neutron fields is produced by neutrons with energies below 10 MeV.

A survey of the literature indicates that substantial experimental data-bases exist for both of these reactions (e.g. CINDA, 1935–1987; CSISRS, 1987). Furthermore, both data-bases have been subjected to comprehensive evaluations within the last decade (e.g. Tagesen *et al.*, 1979; Tagesen and Vonach, 1981). The data-bases used in these evaluations exhibit a familiar pattern, namely a plethora of experimental information in the 13–15 MeV energy range and substantially less information at both lower and higher energies. The result is small predicted evaluation uncertainties (generally below 1%) in the vicinity of 14 MeV, but significantly larger ones elsewhere. Near threshold, the uncertainties in these contemporary evaluations are larger than is desirable for dosimetry applications. Furthermore, since the evaluated values near threshold are derived from relatively limited experimental data-bases, it is quite probable that they are afflicted to some extent by undiscovered systematic uncertainties. Consequently, there is a need for additional differential information in this lower-energy region in order to better establish the evaluated cross sections there and to reduce their overall uncertainties. The present experiment was undertaken with this objective in mind since very few relevant results had become available for either reaction since the evaluations by Tagesen and coworkers nearly a decade ago.

2. EXPERIMENTAL AND DATA ANALYSIS PROCEDURES

The experimental techniques used in this work are well-documented in earlier publications from this laboratory (e.g. Smith *et al.*, 1984a; Kanno *et al.*, 1984; Smith *et al.*, 1984b provide an overview of the procedures).

Nearly monoenergetic neutrons above 5 MeV were produced by bombarding a small D_2-gas target cell with deuterons [the $^2H(d,n)^3He$ reaction]. Deuteron beams in the energy range 2.25–7.00 MeV were produced at the Argonne National Laboratory Fast-Neu-

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tron Generator (FNG) which has been described by Cox and Hanley (1971). The relevant procedures for neutron production at this facility have been documented by Smith and Meadows (1974). The charged-particle energy scale was determined to within an uncertainty of about 5 keV while the average energies of the neutrons incident at the sample/monitor position were defined to within a conservative uncertainty of about 20% of the full-width–half-maximum (FWHM) resolutions, i.e. to within 27–48 keV. The samples used in the activation measurements were Mg and Al metal disks of natural isotopic composition (Mg: $99.92 \pm 0.08\%$ purity by weight, 2.61 cm dia $\times$ 0.35 cm thick; Al: $99.20 \pm 0.8\%$ purity by weight, 2.54 cm dia $\times$ 0.32 cm thick). These were attached to a neutron fluence monitor (as shown in Fig. 1 of Smith *et al.*, 1984b) for the irradiations. The fluence monitor was a low-mass ionization chamber which contained a calibrated thin film of depleted U (essentially 100% ^{238}U). Procedures for fabricating and calibrating such U deposits are described by Meadows (1972), Smith and Meadows (1975) and Poenitz *et al.* (1979). The irradiated Mg and Al samples were counted for ^{24}Na activity with a Ge detector which was calibrated using standard γ-ray sources and the techniques described by Meadows and Smith (1984) and Geraldo and Smith (1989). The particular sample-irradiation and activity-counting procedures of this experiment were carried out with careful attention to the properties of the ^{24}Mg(n,p)^{24}Na and ^{27}Al(n,α)^{24}Na reactions, and the decay of ^{24}Na, as indicated in Table 1 (e.g. see Lederer and Shirley, 1978; Tuli, 1985). Detailed calibration measurements were carried out only for the Mg samples. Monte-

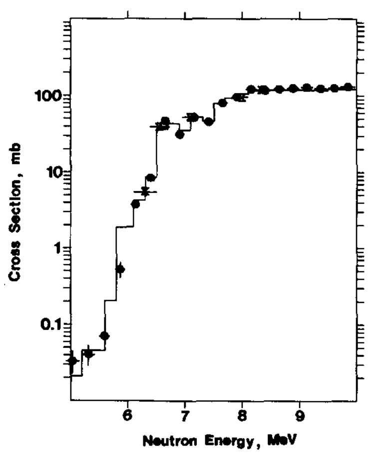

Fig. 1. Differential cross sections for the ^{24}Mg(n,p)^{24}Na reaction: ●, present results; ⌶, Mostafa (1976); solid line denotes the evaluated results of Tagesen *et al.* (1979) in group format.

Table 1. Pertinent features of the ^{24}Mg(n,p)^{24}Na and ^{27}Al(n,α)^{24}Na reactions

	^{24}Mg(n,p)^{24}Na	^{27}Al(n,α)^{24}Na
Reaction Q-value[a]	-4.732 MeV	-3.132 MeV
^{24}Na decay half-life[a]	15.020 ± 0.007 n	
Prominent ^{24}Na γ-rays[b]	1.369 MeV (100%)	
	2.754 MeV (100%)	

[a] Tuli (1985).
[b] Lederer and Shirley (1978).

Carlo calculations were employed to show that the difference in counting efficiency for the Al samples was negligible. However, an additional error of 0.8% is included for those measurements involving the Al samples owing to the indirect manner in which the γ-ray counting efficiency was established in this instance.

The experimental data were analyzed using methods discussed by Smith and Meadows (1974) and Smith (1979). The γ-ray count data were corrected for decay, detector efficiency, finite sample geometry, nonuniform sample activity and the effects of secondary neutrons from the gas target cell. The fission yield data were corrected for lost fission events masked by α-decay, proton recoil and noise pulses, for the fission fragments emitted near 90° which were lost in the U deposit and for the effects of secondary neutrons from the gas target cell. These data were further adjusted using calculated neutron multiple-scattering correction factors, as described by Smith and Meadows (1973, 1977). Finally, the adjusted data were processed with a computer code which calculated activation cross sections relative to the standard ^{238}U neutron fission cross section. Additional corrections for geometry, neutron-source properties, neutron absorption and specific characteristics of the U deposit and the Mg and Al samples were also determined using this code. Incident-neutron distributions were computed according to the method of Smith (1979), thereby providing detailed energy- and angular-resolution information for the cross-section results.

The known sources of experimental error in this work were examined in detail. Table 2 provides a list of the various error contributions which were considered and an indication of the magnitudes involved. Specific comments on the systematic-error correlations appear in footnotes to this table. The partial errors and their correlations were combined to form a covariance matrix, according to procedures documented in papers by Smith (1981, 1987a, b).

3. EXPERIMENTAL RESULTS

The results of the present experiment are summarized in Tables 3–6 and Figs 1 and 2.

Table 2. Error sources for the measured cross section ratios

	^{24}Mg(n,p)^{24}Na	^{27}Al(n,α)^{24}Na
Random errors (%)[a]		
Fission event statistics	0.3–1.7	0.3–1.8
γ-ray counting statistics	0.4–4.6	0.4–1.5
Geometric effects	0.1–1.5	0.1–0.3
Systematic errors (%)[b]		
U deposit mass	0.6	0.6
Fission extrapolation correction	0.3	0.3
U-deposit thickness correction	0.9	0.9
Sample atom content	0.1[c]	0.8[c]
Calibration for γ-ray counting	1.6[d]	1.8[d]
Inhomogeneity of sample γ-ray activity	N[e]–0.2	<0.1
Neutron source properties	2.0[f]	2.0[f]
Neutron energy scale	N–32.7[g]	1.1–7.9[g]
Neutron absorption and multiple scattering	0.7–1.0[f]	0.8–1.0[f]
Effects of ^{2}H(d;n,p)^{2}H neutrons	N–2.1[g]	N–2.1[g]

[a] Uncorrelated errors.
[b] 100% correlated errors unless otherwise specified.
[c] 100% correlated errors for a specific sample type.
[d] A 100% correlated error component of 1.6% exists for all data points. An additional 100% correlated error of 0.8% exists only for data points involving Al samples.
[e] N = Negligible.
[f] (100–10ΔE)% correlated errors for the same reaction. ΔE = data-point neutron energy difference (MeV).
[g] 50% correlated errors for the same reaction.

The measured cross section ratios for ^{24}Mg(n,p)^{24}Na relative to ^{238}U neutron fission are given in Table 3. Evaluated values for the ^{238}U neutron fission cross section obtained from Poenitz (1988) were employed to convert these ratios to the reaction cross sections which are plotted in Fig. 1. The results of Poenitz (1988) were used as the cross section standard rather than equivalent values from ENDF/B-V (1979) since it is likely that the former will be incorporated in ENDF/B-VI which is soon to be released. Uncertainty correlations for the present experimental ratios are provided in Table 5. The specific error components for each data point had to be analyzed in order to derive this matrix. The detailed information is too extensive to be included in this paper, but if required it can be obtained by contacting one of the authors (DLS). The only other available experimental differential cross sections near threshold which were apparently not included in the evaluation of Tagesen *et al.* (1979) are from the work of Mostafa (1976). They are also plotted in Fig. 1. Finally, the evaluated cross sections of Tagesen *et al.* (1979) are plotted in Fig. 1 for comparison with these recent data points. They are represented by a histogram because this evaluation is provided in a group format.

Table 3. Experimental results for the ^{24}Mg(n,p)^{24}Na reaction

Data point	Neutron[a] energy (MeV)	Neutron[b] resolution (MeV)	Cross section ratio	Ratio error (%)	Standard[c] fission σ (mbarn)	Reaction σ (mbarn)
1	5.038	0.225	6.195(−5)[d]	33.2	540.8	3.350(−2)
2	5.313	0.205	7.584(−5)	30.3	543.5	4.122(−2)
3	5.592	0.178	1.299(−4)	26.2	557.1	7.234(−2)
4	5.866	0.153	9.116(−4)	22.7	587.0	0.5351
5	6.133	0.145	5.745(−3)	9.9	662.3	3.805
6	6.395	0.148	1.088(−2)	8.6	776.8	8.450
7	6.654	0.142	5.431(−2)	9.0	872.9	47.41
8	6.908	0.146	3.400(−2)	6.7	924.0	31.42
9	6.910	0.136	3.468(−2)	6.3	924.0	32.04
10	7.164	0.138	5.439(−2)	3.7	955.4	51.96
11	7.415	0.151	4.695(−2)	7.8	975.3	45.79
12	7.664	0.154	8.163(−2)	5.3	987.5	80.61
13	7.912	0.166	9.753(−2)	4.6	992.6	96.81
14	8.159	0.171	0.1235	3.1	995.6	123.0
15	8.403	0.175	0.1171	2.9	998.7	116.9
16	8.648	0.179	0.1225	3.1	999.4	122.4
17	8.888	0.188	0.1252	3.4	998.4	125.0
18	8.890	0.174	0.1270	3.0	998.4	126.8
19	9.130	0.193	0.1305	3.5	996.5	130.0
20	9.371	0.198	0.1267	3.8	993.8	125.9
21	9.611	0.214	0.1269	3.6	991.1	125.8
22	9.851	0.242	0.1327	4.0	988.5	131.2

[a] Mean energy of the incident neutrons.
[b] Full-width–half-maximum (FWHM) of incident-neutron energy distribution.
[c] Standard ^{238}U neutron fission cross sections obtained from Poenitz (1988). Uncertainties are ≅ 1%.
[d] 6.195(−5) signifies 6.195×10^{-5}.

Table 4. Experimental results for the $^{27}Al(n,\alpha)^{24}Na$ reaction

Data point	Neutron[a] energy (MeV)	Neutron[b] resolution (MeV)	Cross section ratio	Ratio error (%)	Standard[c] fission σ (mbarn)	Reaction σ (mbarn)
1	5.869	0.163	1.948(−3)[d]	7.6	587.7	1.145
2	6.399	0.150	6.212(−3)	8.6	778.6	4.837
3	6.914	0.138	1.690(−2)	5.1	925.0	15.63
4	7.419	0.145	2.699(−2)	4.3	975.6	26.33
5	7.917	0.160	4.351(−2)	3.8	992.7	43.19
6	8.408	0.178	5.366(−2)	3.6	998.8	53.60
7	8.895	0.188	6.807(−2)	3.5	998.3	67.95
8	9.379	0.198	8.265(−2)	3.9	993.7	82.13
9	9.379	0.198	8.240(−2)	4.2	993.7	81.88
10	9.859	0.231	8.675(−2)	4.1	988.4	85.74

[a] Mean energy of the incident neutrons.

[b] Full-width-half-maximum (FWHM) of incident-neutron energy distribution.

[c] Standard ^{238}U neutron fission cross sections obtained from Poenitz (1988). Uncertainties are $\cong 1\%$.

[d] 1.948(−3) signifies 1.948×10^{-3}.

The corresponding results for the $^{27}Al(n,\alpha)^{24}Na$ reaction appear in Table 4 (measured ratios and derived reaction cross sections) and Table 6 (correlation matrix) and they are plotted in Fig. 2. Again, only one other available experimental differential cross section set near threshold was found which was not included in the evaluation of Tagesen and Vonach (1981). This data set originates from the work of Enz et al. (1985).

4. DISCUSSION

For $^{24}Mg(n,p)^{24}Na$, overall agreement between the present results, the data of Mostafa (1976) and the

Table 5. Uncertainty correlation matrix for the measured cross sections of $^{24}Mg(n,p)^{24}Na$ relative to the ^{238}U neutron fission standard[a]

	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18	19	20	21	22
1	1																					
2	0.49	1																				
3	0.50	0.50	1																			
4	0.49	0.49	0.50	1																		
5	0.49	0.50	0.50	0.50	1																	
6	0.49	0.49	0.50	0.50	0.54	1																
7	0.49	0.49	0.50	0.50	0.54	0.55	1															
8	0.47	0.47	0.48	0.49	0.54	0.56	0.56	1														
9	0.47	0.47	0.48	0.48	0.54	0.56	0.56	0.58	1													
10	0.36	0.37	0.38	0.39	0.51	0.54	0.53	0.60	0.62	1												
11	0.48	0.48	0.49	0.49	0.54	0.54	0.55	0.56	0.56	0.56	1											
12	0.45	0.46	0.46	0.47	0.54	0.56	0.56	0.59	0.60	0.66	0.58	1										
13	0.42	0.42	0.44	0.44	0.53	0.55	0.55	0.59	0.60	0.70	0.57	0.65	1									
14	0.24	0.25	0.26	0.27	0.41	0.44	0.44	0.52	0.54	0.78	0.48	0.63	0.70	1								
15	0.07	0.08	0.10	0.11	0.26	0.30	0.29	0.39	0.41	0.71	0.34	0.51	0.60	0.89	1							
16	0.16	0.17	0.18	0.19	0.32	0.36	0.36	0.44	0.47	0.71	0.40	0.55	0.63	0.86	0.89	1						
17	0.13	0.14	0.15	0.16	0.28	0.31	0.31	0.39	0.41	0.64	0.35	0.49	0.56	0.77	0.81	0.80	1					
18	0.13	0.14	0.15	0.17	0.30	0.34	0.33	0.42	0.44	0.70	0.38	0.53	0.61	0.85	0.89	0.88	0.83	1				
19	0.06	0.07	0.08	0.09	0.21	0.24	0.24	0.32	0.34	0.57	0.28	0.41	0.49	0.72	0.78	0.77	0.74	0.82	1			
20	0.08	0.09	0.10	0.11	0.22	0.25	0.24	0.31	0.33	0.54	0.28	0.40	0.46	0.66	0.71	0.71	0.67	0.75	0.72	1		
21	0.10	0.11	0.12	0.13	0.23	0.27	0.26	0.33	0.35	0.56	0.30	0.42	0.49	0.68	0.73	0.73	0.71	0.78	0.77	0.71	1	
22	0.21	0.21	0.22	0.23	0.32	0.35	0.35	0.41	0.42	0.57	0.38	0.48	0.53	0.65	0.65	0.68	0.65	0.72	0.70	0.65	0.72	1

[a] Indices correspond to those appearing in Table 3. The uncertainties in the standard cross section are excluded.

Table 6. Uncertainty correlation matrix for the measured cross sections of $^{27}Al(n,\alpha)^{24}Na$ relative to the ^{238}U neutron fission standard[a]

	1	2	3	4	5	6	7	8	9	10
1	1									
2	0.56	1								
3	0.59	0.58	1							
4	0.58	0.57	0.70	1						
5	0.55	0.53	0.69	0.76	1					
6	0.51	0.50	0.67	0.75	0.82	1				
7	0.48	0.47	0.64	0.71	0.79	0.83	1			
8	0.44	0.42	0.58	0.64	0.71	0.75	0.79	1		
9	0.40	0.39	0.53	0.59	0.65	0.69	0.73	0.73	1	
10	0.36	0.35	0.49	0.56	0.62	0.66	0.72	0.73	0.67	1

[a] Indices correspond to those appearing in Table 4. The uncertainties in the standard cross section are excluded.

evaluation of Tagesen *et al.* (1979) is excellent. This encouraged us to pursue an evaluation procedure in which the two new experimental data sets were merged with the evaluated values of Tagesen *et al.* (1979) using the least-squares code GMA (Poenitz, 1981; Sugimoto, 1987). The intent was not to generate a comprehensive updated evaluation but rather to examine the degree to which the new information permits a reduction in the uncertainties of the differential cross sections over a limited energy interval. Analysis was restricted to the neutron energy interval 6.4–9.8 MeV. The adjustments produced by introducing the new experimental data were generally well within the uncertainties of the earlier evaluation although the general tendency was toward slightly lower cross sections. The quoted uncertainties for the evaluated values of Tagesen *et al.* (1979) over this energy interval range from 3.9–7.9% (average 5.2%). The GMA procedure predicts revised uncertainties in the range 2.1–4.7% (average 3.3%), clearly a significant reduction. Since the values of Mostafa (1976) carry errors in the range 7.0–12.5%, their contribution to the reduction of error is small relative to that provided by the present data set. An additional differential cross-section data set covering the energy interval 8–15 MeV was reported at a recent conference by Boerker *et al.* (1988), but only a reprint with plotted values is currently available. Numerical values were estimated from these plots. They were found to be in good agreement with the present results between 8–9 MeV, but they tend to be somewhat larger at higher energies. These results were not included in the GMA analysis described above since the exact numerical information was not available.

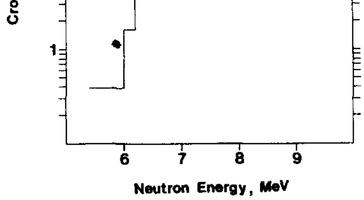

Fig. 2. Differential cross sections for the $^{27}Al(n,\alpha)^{24}Na$ reaction: ●, present results; □, Enz *et al.* (1985); solid line denotes the evaluated results of Tagesen and Vonach (1981) in group format.

The present results for $^{27}Al(n,\alpha)^{24}Na$ agree with those of Enz *et al.* (1985) within the uncertainties, though this is not a particularly significant result given that the quoted errors from that work are all in excess of 10%. The present results are also consistent with the evaluation of Tagesen and Vonach (1981), within the errors, but they tend to be systematically higher, especially two experimental points in the vicinity of 8–9 MeV which are noticeably high (beyond the combined error bars). Boerker *et al.* (1988) have also provided a data set for this reaction, however, as discussed in the preceding paragraph, exact numerical results are not yet reported. Numerical values estimated from a figure provided in their conference paper appear to be in very good agreement with the present experimental points, except possibly above 9.5 MeV where they are slightly higher. The evaluation procedure described in the preceding paragraph was also applied to the available information for $^{27}Al(n,\alpha)^{24}Na$, again excluding the data of Boerker *et al.* (1988) for the reasons given above. The energy interval addressed in this exercise was 6.3–9.3 MeV. The adjustments to the evaluation of Tagesen and Vonach (1981) which came about from inclusion of the new data were within the uncertainties of this earlier evaluation, except near 8 MeV where the

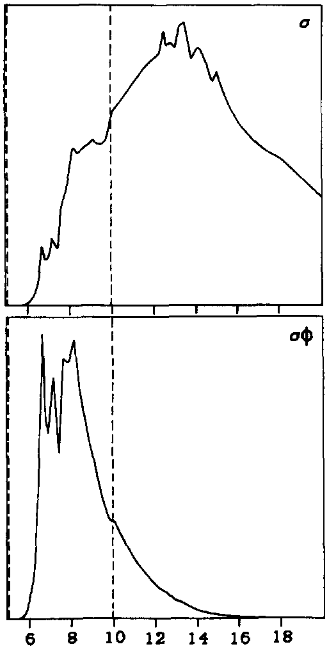

Fig. 3. Response profile for the $^{24}Mg(n,p)^{24}Na$ reaction in the ^{252}Cf spontaneous-fission neutron spectrum. Identity of symbols: σ [differential cross section based on the evaluation of Tagesen *et al.* (1979) in point format] and $\sigma\phi$ (response function). The neutron spectrum ϕ is based on the evaluation of Mannhart (1987). The vertical dashed lines indicate the energy range of the present experiment.

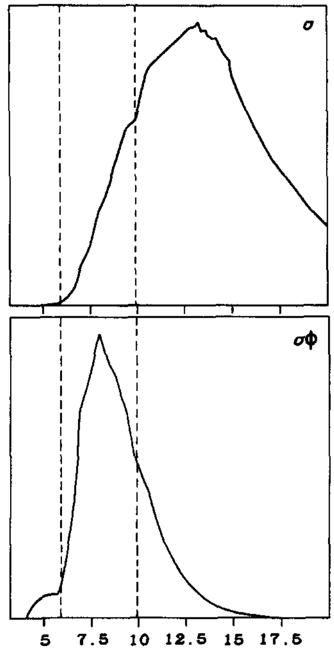

Fig. 4. Response profile for the $^{27}Al(n,\alpha)^{24}Na$ reaction in the ^{252}Cf spontaneous-fission neutron spectrum. Identity of symbols: σ [differential cross section based on the evaluation of Tagesen and Vonach (1981) in point format] and $\sigma\phi$ (response function). The neutron spectrum ϕ is based on the evaluation of Mannhart (1987). The vertical dashed lines indicate the energy range of the present experiment. The small pedestal seen at around 5 MeV in the plot of $\sigma\phi$ is a numerical artifact which can be attributed to the fact that the group widths near threshold are rather large for the evaluation of Tagesen and Vonach (1981).

adjusted results are just above the error limits of the old evaluation. In general, the GMA adjusted cross sections tend to be systematically larger than those predicted by the earlier evaluation. The uncertainties for the previous evaluation were in the range 2.7–8.9% from 6.3–9.3 MeV (average 5%). Inclusion of new information, dominated by the present results, reduces the corresponding errors to the range 2.3–6.9% (average 3.4%). While this represents a significant overall reduction in uncertainty, this procedure showed that most of the improvement is for energies >8 MeV.

Mannhart (1985, 1987) has compiled and evaluated the available integral activation cross section data for both of these reactions in the standard ^{252}Cf spontaneous-fission neutron field. He also calculated the corresponding values using the results of his own evaluation for the Cf spectrum (Mannhart, 1987) and evaluated differential cross sections for $^{24}Mg(n,p)$ ^{24}Na from Tagesen *et al.* (1979) and for $^{27}Al(n,\alpha)$ ^{24}Na from Tagesen and Vonach (1981). The response profile for $^{24}Mg(n,p)$ ^{24}Na which corresponds to this analysis is shown in Fig. 3, while the corresponding information for $^{27}Al(n,\alpha)$ ^{24}Na is presented in Fig. 4. From these profiles, it was determined that the present differential experiment for $^{24}Mg(n,p)^{24}Na$ addressed 79% of the response range for the Cf neutron field while that for $^{27}Al(n,\alpha)^{24}Na$ covered 73% of the response range. Mannhart (1985, 1987) deduced the following spectrum average cross sections for the Cf neutron field: $^{24}Mg(n,p)$ ^{24}Na [evaluated experimental result, 1.998 mbarn ($\pm$2.4%); calculated result, 2.101 mbarn] and $^{27}Al(n,\alpha)^{24}Na$ [evaluated experimental result, 1.017 mbarn ($\pm$1.5%); calculated result, 0.9886 mbarn]. The corresponding calculated and integral results appear to be reasonably consistent within the indicated combined experimental and calculational errors. It is clear that inclusion of new differential information will not lead to major changes in the earlier evaluations for these reactions. However, it is also evident from the preceding discussion that inclusion of the new data should result in adjustments to the earlier evaluations from Tagesen and coworkers that will provide improved agreement between calculated and experimental integral results (C/E) for both reactions.

5. CONCLUSIONS

The present investigation provides new experimental differential cross section sets of good accuracy for the $^{24}Mg(n,p)^{24}Na$ and $^{27}Al(n,\alpha)^{24}Na$ reactions. These results are generally found to be consistent with prior information, within the errors, although for $^{27}Al(n,\alpha)^{24}Na$ the present cross sections tend to be systematically higher than those predicted by an earlier evaluation. The new information provided by this experiment appears to offer an opportunity to reduce the evaluated cross section errors for these reactions to values in the vicinity of 3% over a substantial portion of the threshold region, thereby increasing the usefulness of these processes for precision neutron dosimetry applications in fission and fusion energy systems.

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