R-Matrix Evaluation of ^{16}O Neutron Cross Sections up to 6.3 MeV

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We have evaluated ^{16}O neutron cross sections in the resolved resonance region with the multilevel Reich-Moore code SAMMY. Resonance parameters were determined by a consistent analysis, including both Doppler and resolution broadening effects. To properly treat the α particle exit channel, an algorithm to calculate charged particle penetrabilities and shifts was incorporated into SAMMY.

KEYWORDS: resonance parameter evaluation, oxygen 16, cross section, alpha particle exit channel

I. Introduction

Over the years the nuclear community has developed a collection of evaluated nuclear data for applications in thermal, fast reactor, and fusion systems. In contrast to these systems, typical neutron spectra in criticality safety applications appear to peak in the epithermal energy range. Because nuclear data play a major role in the calculation of criticality safety margins, a thorough examination of the behavior of present nuclear data evaluations in criticality safety calculations is needed. For oxygen, the existing ENDF/B-VI.5 evaluation is expressed in terms of point-wise cross sections derived from the analysis of G. Hale.[1)] Unfortunately such an evaluation is not directly useful for resonance analysis of data from samples in which oxygen is combined with other elements; for that purpose, resonance parameters are needed. In this paper we describe a resonance parameter evaluation of ^{16}O neutron cross sections in the resolved resonance region with the multilevel Reich-Moore R-matrix formalism using the code SAMMY.[2)] A preliminary report of this work has been given previously.[3)]

II. Cross Section and Differential Elastic Data

An extensive search of standard nuclear databases and the open literature led to selection of total, reaction, and angle differential elastic cross section data sets for analysis; see **Tables 1** and **2**. The σ_{total} data include measurements by Johnson, et al.,[4)] on the 200-m flight path at the Oak Ridge Electron Linear Accelerator (ORELA); Cierjacks, et al.,[5)] on the 200-m flight path at the Karlsruhe cyclotron; Larson[6)] (ORELA 80 m flight path); Fowler, et al.,[7)] who utilized a 47-m flight path and a pulsed van de Graaff accelerator to produce neutrons; and Johnson, et al.,[8)] who made accurate measurements in the 2.35 MeV window region. In the energy range of overlap, 0.6 - 4.3 MeV, the σ_{total} values for Refs 4, 6, 7 are in good agreement, but the data of Cierjacks, et al.[5)] is about 3.5% lower. The Cierjacks data were normalized to the data of Johnson, et al.,[4)] by integrating between 3.45 and 3.72 MeV. A neutron energy transformation was applied to align the peak energies of Johnson, et al. with the higher resolution Cierjacks values.

The $^{16}\mathrm{O}(n,\alpha)^{13}\mathrm{C}$ channel opens at a laboratory neutron energy E_n = 2.36 MeV and contributes about 9% to σ_{total} at E_n = 4.18 MeV and about 25% at 5.07 MeV. Therefore,

Table 1 Cross Section Data Sets for ^{16}O Evaluation

Authors	Facility	Energy Range (MeV)	Atoms/barn
Johnson, et al.[4)]	ORELA	0.2 - 6.3	0.183
Cierjacks, et al.[5)]	KFK cyclotron	3.14 - 6.3	1.201
Larson[6)]	ORELA	2.0 - 6.3	0.549
Fowler, et al.[7)]	ORNL VDG	0.6 - 6.3	0.488
Johnson, et al.[8)]	ORELA	2.25 - 2.49	6.700
Ohkubo[9)]	Linac	0.01 - 0.9	
Bair, Haas[10)]	ORNL VDG	3.2 - 6.3	
Drotleff, et al.[11)]	Stuttgart	2.87 - 3.48	

Table 2 Angular Distribution Data Sets for ^{16}O Evaluation

Authors	Facility	Energy Range (MeV)	Θ_{CM} (deg)
Okazaki[12)]	Wisconsin	0.410 - 0.493	46 - 133
Fowler, Cohn[13)]	ORNL VDG	0.73 - 2.15	32 - 138
Phillips[14)]	LANL	3.0 - 6.0	22 - 152
Martin, Zucker[15)]	BNL	1.51 - 2.25	21 - 166
Hunzinger, Huber[16)]	Basel CW	2.00 - 4.11	41 - 147
Lister, Sayres[17)]	Columbia VDG	3.1 - 4.7	Leg. Coef.
Johnson, Fowler[18)]	ORNL VDG	3.266 - 4.200	20 - 147
Fowler, Johnson[19)]	ORNL VDG	1.833 - 3.441	20 - 146
Kinney, Perey[20)]	ORNL VDG	4.34 - 6.44	16 - 139
Drigo, et al.[21)]	Lignaro VDG	2.56 - 2.76	26 - 156

$\sigma_{n,\alpha}$ values deduced by reciprocity from $^{13}\mathrm{C}(\alpha,n)^{16}\mathrm{O}$ measurements by Bair and Haas[10)] were fit to obtain Γ_α values for several resonances. These data exhibit good α energy resolution of 2 to 5 keV over the energy range of interest for this evaluation. Energy transformations were applied to align narrow resonances with the more precise Cierjacks energies. Since the Bair-Haas data agree to better than 10% with the recent high-precision measurements of Drotleff, et al.[11)] in the region of overlap (3.20-3.48 MeV), we analyzed both data sets with a single normalization that was varied in the analysis.

In order to give a proper treatment for charged particles in an exit channel, an algorithm[3)] to calculate charged particle penetrabilities (CPP) and shifts was incorporated in SAMMY. A slightly modified version of the routine COULFG of Barnett[22)] is used to compute Coulomb wave functions and derivatives. Routines based on the CPP algorithm have been integrated into a prototype modification of the NJOY[23)] code and will be incorporated in the AMPX[24)] code.

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Angle differential elastic cross sections were computed with SAMMY using the set of resonance parameters obtained from analysis of the total and reaction cross section data. These predicted values were compared with angular distribution data[12–16,18–21] to confirm J values for several resonances. Predicted Legendre coefficients were compared with the corresponding experimental values of Lister and Sayres.[17]

III. Resonance Analysis and Results

Resonance parameters were determined by a consistent analysis in which both Doppler and resolution broadening effects were incorporated. Results from a preliminary ^{16}O evaluation have been reported previously.[3,25] Total and reaction data sets were analyzed sequentially so that each fit was connected to the previous fit by the SAMMY parameter covariance matrix, thereby yielding energies and widths for 37 resonances in the range $0 < E_n < 6.3$ MeV. Two negative-energy resonances were included to account for bound levels and 13 high-energy resonances were included to account for the effect of resonances above 6.3 MeV. Partial waves $s_{1/2}$ through $g_{9/2}$ were included in the analysis. The neutron channel radius, a_n , α channel radius, a_α , and the $\sigma_{n,\alpha}$ normalization factor, $F_{n\alpha}$, were varied; final values were a_n = 3.80 fm, a_α = 6.7 fm, and $F_{n\alpha}$ = 1.00.

Spin-parity assignments were based on fits to σ_{total} and $\sigma_{n,\alpha}$ data and on comparison of predicted and experimental $d\sigma/d\Omega$ values. Where fits were inconsistent with the data, several J^π values were tried to improve the fits. For most resonances our J^π values are identical to those reported in the compilation by Tilley, et al.[26] Exceptions are resonances at 5993 and 6076 keV. Energy resolution values for the differential elastic data vary over a wide range. Theoretical $d\sigma/d\Omega$ values were energy-broadened by the appropriate amount before comparison with the data. An example of the effect of energy broadening is given in **Fig. 1** for the data of Fowler and Johnson[19] for the 1834 keV $d_{3/2}$ resonance (ΔE = 13 keV, Γ = 7.8 keV), the 3211 keV $f_{5/2}$ resonance, and the 3442 keV $f_{5/2}$ resonance.

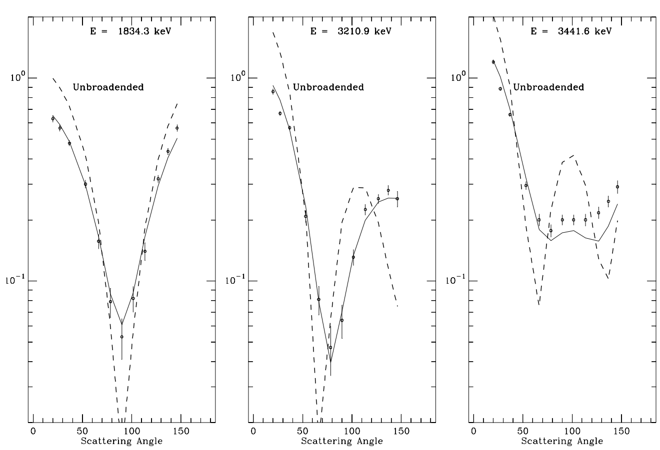

Fig. 1 Broadened (solid) and unbroadened (dashed) SAMMY predictions for $d\sigma/d\Omega$ data (points) of Fowler and Johnson.[19]

Figure 2 presents a global view of the final SAMMY fits to the total cross section data of Refs. 4–9.

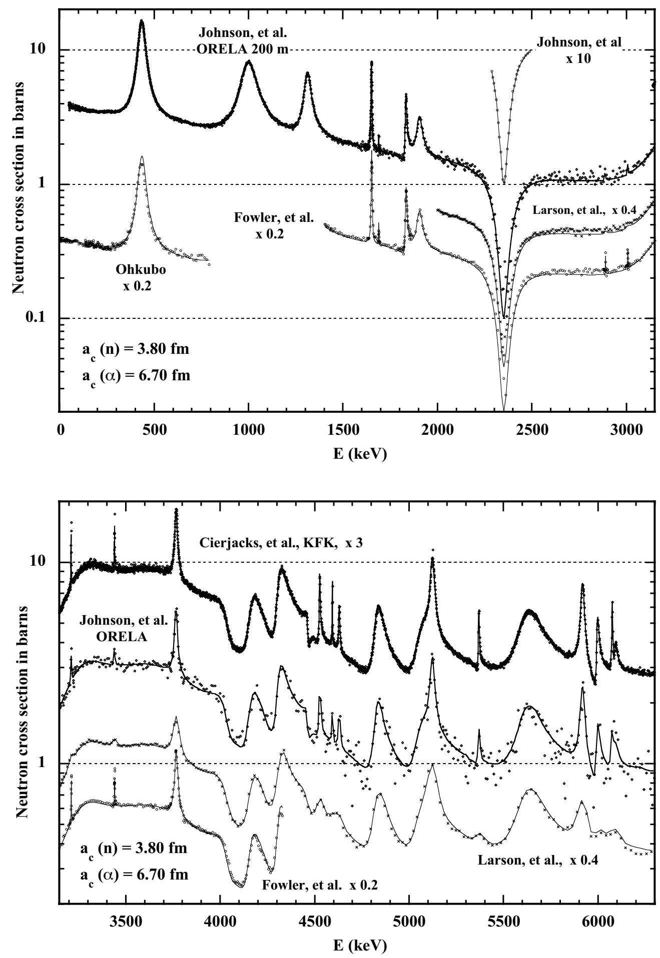

Fig. 2 Comparison of SAMMY predictions to ^{16}O σ_{total} data.[4–9] Data and predictions have been scaled for visual separation.

The (n, α) cross sections obtained by reciprocity from the (α, n) data of Bair and Haas[10] and Drotleff, et al.[11] are compared with the SAMMY fits in **Fig. 3**. A rather large α channel radius, 6.7 fm, was required in order to fit the (n, α) data because the 3291 keV $d_{3/2}$ resonance (Γ_n = 340 keV, Γ_α = 0.17 keV) introduces a significant background for $E_n > 4.5$ MeV. This is due to the exponential increase of the Coulomb penetrability, and hence Γ_α , with E_n. The agreement between predicted and experimental $\sigma_{n,\alpha}$ values is quite satisfactory over the fit range, 3.1 to 6.3 MeV. At lower energies, where the cross section is orders of magnitude smaller, the prediction underestimates $\sigma_{n,\alpha}$.

Examples of angle differential elastic data[18] are compared with SAMMY predictions in **Fig. 4**. Legendre coefficients given by Lister and Sayres[17] are compared with predicted values in **Fig. 5**. When uncertainties are taken into account, predicted and experimental coefficients are in satisfactory agreement. It is assumed that the extraction of $d\sigma/d\Omega$(nn) and Legendre coefficients from the experimental measurements was not affected by competition from the α channel.

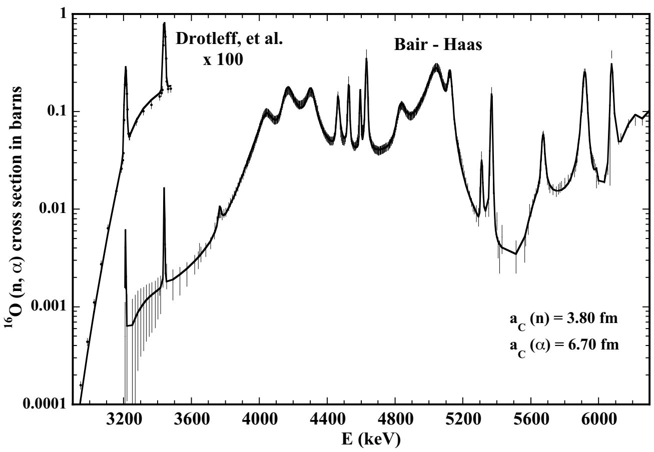

Fig. 3 Comparison of SAMMY predictions to $\sigma_{n,\alpha}$ data deduced by reciprocity from $^{13}C(\alpha,n)^{16}O$ data of Bair and Haas[10] and Drotleff et al.[11]

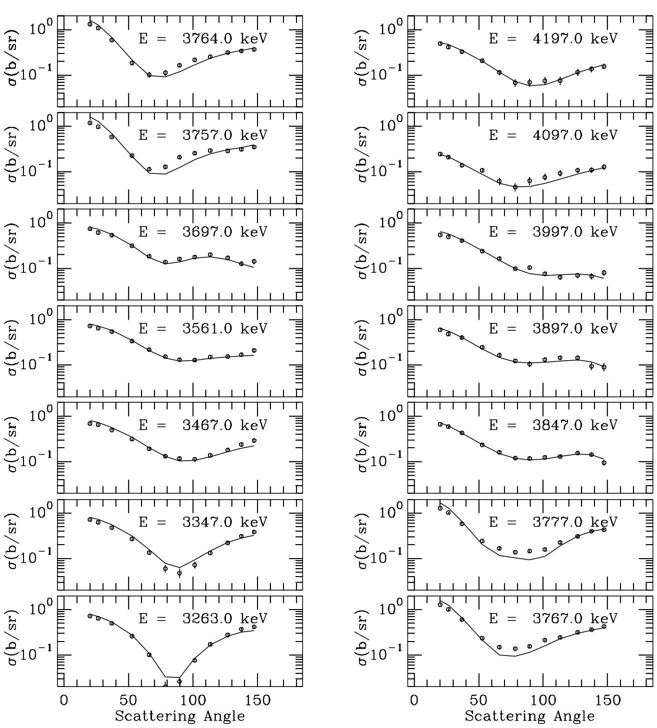

Fig. 4 Comparison of SAMMY predictions to differential elastic data of Johnson and Fowler.[18]

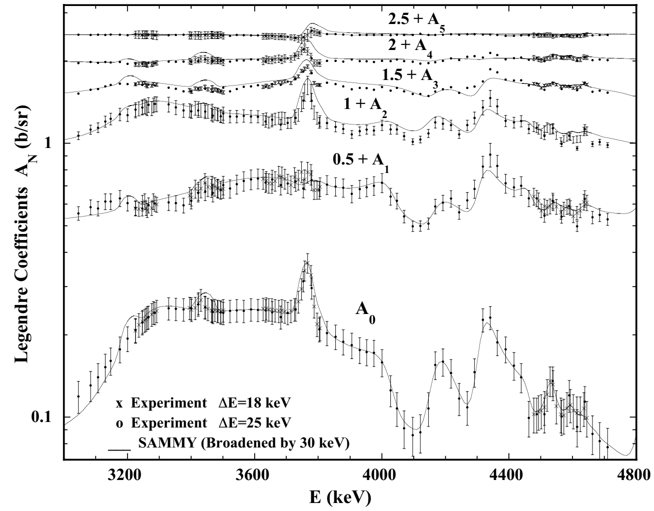

Fig. 5 Comparison of SAMMY predictions to Legendre coefficients of Lister and Sayres.[17]

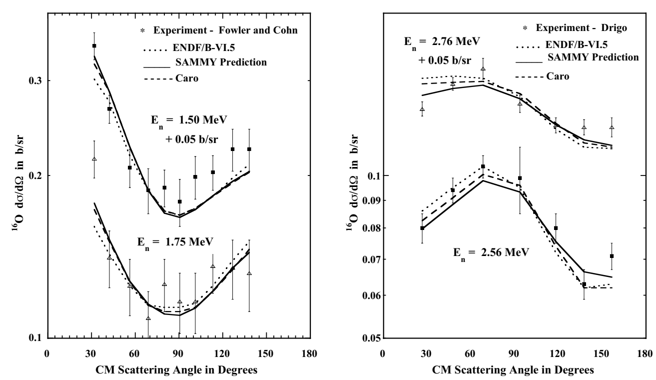

Fig. 6 Experimental[13,21) , ENDF/B-VI (dotted), Caro (dashed), and SAMMY (solid) $d\sigma/d\Omega$ for E_n = 1.5, 1.75, 2.56, and 2.76 MeV.

Caro[27] has reported an evaluation of ^{16}O using a resonance plus potential well model which, unfortunately, does not provide a Reich-Moore resonance parameter representation. In **Fig. 6**, we compare our predicted $d\sigma/d\Omega$ with Caro and ENDF/B-VI for four non-resonant energies: 1.50, 1.75, 2.56, and 2.76 MeV. At these energies the differences in predicted values are small except at forward angles, where the two recent evaluations give better agreement with experiment than does ENDF/B-VI.

The ^{17}O level excitation energy E_x, peak energy E_{peak}, resonance energy E_R, neutron width Γ_n, α width Γ_α, and J^π value are listed in **Table 3** for resonances included in the present evaluation. For a particular partial wave, e.g., $d_{3/2}$, peak energies are defined as those energies corresponding to maxima (l > 0) or minima (l = 0) in the unbroadened partial cross section for that partial wave. Excitation energies are computed from the separation energy and nuclear masses A_n and A_{O16} according to the relation: E_x = 4143.36 keV + E_R * $A_{O16}/(A_{O16} + A_n)$. The resonance energies E_R correspond to the eigenenergies determined by the Reich-Moore analysis with SAMMY with boundary conditions chosen so that the level shifts are zero.

1. Individual Resonance Discussion

Selected individual resonances are discussed here. More detailed results are presented in Ref 3.

For the 10 levels in the energy range $0 < E_n < 3100$ keV, the contribution to σ_{total} from $\sigma_{n,\alpha}$ is completely negligible. Parameters for resonances at 434, 999, 1312, 1651, 1834, and

Table 3 Energies and Widths for Resonances in ^{16}O (n,X)

J^π	$E_x(^{17}O)$ (keV)	E_{peak} (keV)	E_R (keV)	Γ_n (keV)	Γ_α * (keV)
$3/2^-$	4551.9 ± 1.5	434.60	434.31	44.41	
$3/2^+$	5084.2 ± 2.5	999.30	1000.22	100.36	
$3/2^-$	5375.1 ± 2.0	1312.70	1309.38	43.43	
$7/2^-$	5696.7 ± 2.0	1651.38	1651.38	4.10	
$(5/2^-)$	5732.3 ± 1.9	1689.15	1689.10	0.27	
$3/2^+$	5868.7 ± 2.0	1834.18	1834.09	7.79	
$1/2^-$	5932.0 ± 2.3	1905.78	1901.44	33.50	
$1/2^+$	6380.2 ± 3.3	2351.09	2377.88	162.37	
$(5/2^+)$	6860.7 ± 2.0	2888.87	2888.70	0.22	
$(7/2^-)$	6971.9 ± 2.0	3007.08	3006.90	0.16	
$5/2^-$	7164.6 ± 0.4	3211.76	3211.76	1.50	0.009
$3/2^+$	7239.1 ± 8.0	3299.68	3291.01	339.63	0.17
$5/2^+$	7378.2 ± 0.4	3438.83	3438.80	0.60	0.020
$5/2^-$	7380.8 ± 0.4	3441.56	3441.55	1.30	0.007
$3/2^-$	7446.9 ±20.0	3654.25	3511.91	660.21	0.026
$7/2^-$	7686.9 ± 0.4	3767.08	3767.00	18.53	0.026
$1/2^+$	7963.3 ± 2.2	4062.70	4060.82	105.58	5.23
$1/2^-$	7896.3 ± 6.0	4059.82	3989.64	276.19	19.15
$3/2^+$	8075.4 ± 2.1	4187.67	4180.04	92.38	9.80
$1/2^-$	8199.3 ± 4.5	4327.90	4311.70	43.52	- 0.44
$3/2^-$	8190.9 ± 2.5	4321.36	4302.79	54.30	5.77
$1/2^+$	8345.7 ± 0.6	4469.48	4467.36	16.89	3.72
$5/2^+$	8402.2 ± 0.2	4527.78	4527.36	4.99	0.86
$7/2^+$	8465.6 ± 0.2	4594.83	4594.83	1.39	0.44
$5/2^-$	8499.8 ± 0.3	4631.26	4631.21	3.20	3.88
$3/2^-$	8677.7 ± 1.5	4839.10	4820.33	58.40	2.74
$3/2^+$	8909.1 ± 4.0	5087.80	5066.30	94.50	- 34.36
$7/2^-$	8963.2 ± 0.5	5123.98	5123.74	23.35	2.75
$(1/2^-)$	9139.3 ± 6.0	5312.80	5311.00	0.50	4.00
$5/2^+$	9194.1 ± 0.4	5369.72	5369.27	2.78	1.25
$3/2^-$ a	9387.5 ±14.0	5637.20	5574.84	191.17	0.42
$5/2^-$	9479.5 ± 4.1	5672.84	5672.62	0.59	15.63
$7/2^+$	9710.9 ± 0.5	5919.05	5918.63	20.50	4.19
$3/2^-$ b	9781.1 ± 0.5	5998.90	5993.29	14.78	- 0.21
$9/2^+$ c	9859.1 ± 0.2	6076.20	6076.19	3.13	2.51
$(1/2^-)$	9869.7 ± 0.8	6094.20	6087.44	16.04	1.92
$5/2^+$	9983.0	6220.60	6207.95	4.97	109.23

Γ_γ = 2.7 eV for 434 keV resonance;
Γ_γ = 0.25 eV for all other resonances.

E_x = 4143.36 keV + E_R * $A_{O16}/(A_{O16} + A_n)$

* Minus sign means the reduced amplitude product $\gamma_n\gamma_\alpha < 0$.

a. Tilley, et al.[26] : E_R = 5610 keV, Γ = 120 keV; $J^\pi = 3/2^-$.
b. Tilley, et al. assign $3/2^+$.
c. Tilley, et al. assign $(5/2^-)$.

1905 keV were based mainly on fits to data of Refs. 4, 7. Our E_R , Γ_n , and J^π values are consistent with Tilley, et al.[26] Predicted $d\sigma/d\Omega$ values agree with the data.[12,13,19]

For the $1/2^+$, 2377.9 keV resonance, Γ_n and E_R are primarily determined by the high-precision data of Johnson, et al.[8] corrected for ^{17}O and ^{18}O. The predicted minimum σ_{total}, 0.1013 b, agrees with the experimental value, 0.1028 ± 0.018 b. As shown in **Fig. 7**, our evaluation fits the data much better than does ENDF/B-VI.

The broad $d_{3/2}$ [3291 keV] and $p_{3/2}$ [3512 keV] resonances are the primary components of σ_{total} for 3200 < E_n < 3700 keV. Each has a small Γ_α that is determined by the low-energy (n,α) data. The Γ_α are rather sensitive to a_α.

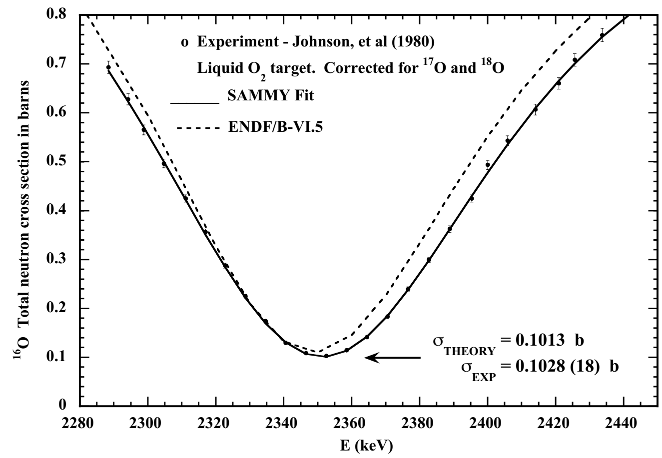

Fig. 7 Comparison of σ_{total} predictions to ENDF/B-VI.5 and data of Johnson et al.[8]

The 3800-5300 keV region is characterized by several broad overlapping resonances. In companion papers, Johnson[28] and Fowler, et al.[7] have reported R-matrix analyses of total[7] and reaction[10] cross sections leading to J^π assignments and interference patterns for pairs of $p_{1/2}$ and $d_{3/2}$ resonances. Using the more recent Cierjacks high-resolution data in addition to the data of Refs. 4, 7, 10, we have confirmed the J^π assignments and interference signs of Johnson. Some of our Γ_n and Γ_α values differ from those of Johnson since our formalism (Reich-Moore) is different from that of Johnson.

Acceptable fits to σ_{total} , $\sigma_{n,\alpha}$, and $d\sigma/d\Omega$ can be obtained for the 3990-4312 keV $p_{1/2}$ pair only if the sign of the interference term, $\gamma_n\gamma_\alpha$, is negative.

Good fits to σ_{total} , $\sigma_{n,\alpha}$, and $d\sigma/d\Omega$ were obtained for the strong 4180 keV resonance. The partial cross section due to the 4180-5066 keV $d_{3/2}$ pair is quite large for 4200 < E_n < 5000 keV. The sign of the interference term, $\gamma_n\gamma_\alpha$, must be negative in order to fit σ_{total} and $\sigma_{n,\alpha}$.

Good agreement with the σ_{total} data[4,5] was found in the region of the broad 5575 keV, $p_{3/2}$ resonance. The peak asymmetry is reduced owing to interference with the 5993 keV, $p_{3/2}$ resonance.

Table 4 compares our total widths with widths given by Refs. 19, 7, and 5 for 10 selected narrow resonances.

For the 3211, 3438, 3441, and 3767 keV resonances, we predict differential elastic cross sections in good agreement with the data (see Figs. 1, 4, and 5). Our J^π values agree with those of Refs 19, 7, and 26. Our Γ_n , as determined by fits to the σ_{total} data,[4,7] agree with values quoted by Fowler and Johnson[19] and Cierjacks, et al.[5] However, the Cierjacks data near the resonance maxima are 5 to 15% larger than theoretical values based on widths *quoted* by Cierjacks. The 3438 and 3441 keV peaks are well-resolved. The discrepancies in maxima are not due to variations in the time channel width. However, small background errors at transmission minima can

Table 4 Total Widths for Selected Narrow Resonances in ^{16}O (n,X)

J^{π}	E_R (keV)	Total Width (keV) Fowler and Johnson[19]	Cierjacks, et al.[5]	Present Evaluation
$5/2^-$	3211.76	1.4	1.45	1.51
$5/2^+$	3438.80	0.5	0.68	0.62
$5/2^-$	3441.55	1.1	1.02	1.31
$7/2^-$	3767.00	18.0*	15.4	18.6
$5/2^+$	4527.36		6.56	5.85
$7/2^+$	4594.83		2.26	1.83
$5/2^-$	4631.21		7.33	7.08
$5/2^+$	5369.27		3.75	4.03
$3/2^-$#	5993.29		12.4	15.0
$9/2^+$$	6076.19		4.26	5.64

* Fowler, et al.[7]
# Cierjacks, et al.[5] assign $J^{\pi} = 3/2^+$
$ Cierjacks, et al.[5] assign $J^{\pi} = 5/2^-$

produce large errors in σ_{total} maxima . For the Cierjacks thick (1.201 atoms/b) sample, transmission minima for these four resonances range from 0.0006 to 0.0022. Since Cierjacks, et al. normalized their thick sample data to a previous thin sample measurement,[29] they may have deduced widths from their thin sample data. Unfortunately, the thin sample data is not available; only a plot is given.[29] We note that σ_{total} maxima read from this plot are more consistent with our predicted values than with the thick sample data.[5]

On the basis of better fits to σ_{total} and $\sigma_{n,\alpha}$, we assign J^{π} = $3/2^-$ rather than $3/2^+$ as given by Tilley, et al.[26] for the 5993 keV resonance. Our total width is 15.0 keV as compared with 12.4 ± 0.3 keV by Cierjacks. Our peak energy, 5998.9 ± 0.5 keV, is not in agreement with Cierjacks (5995.68 ± 0.15 keV). Some of this difference is probably due to the large difference in phase shifts between $p_{3/2}$ and $d_{3/2}$.

For the 6076 keV resonance, $J^{\pi} = 9/2^+$ gives a much better fit to the σ_{total} and $\sigma_{n,\alpha}$ data than does $5/2^-$ as assigned by Tilley, et al.[26] From (α, n) angular distributions, Kerr, et al.[30] assigned $J^{\pi} = 9/2^+$, although they did not resolve the adjacent 6087 keV ($1/2^-$) resonance.

2. Thermal and Integral Quantities

Total and capture cross sections for E_n = 0.0253 eV and T = 300K are in agreement with the ENDF/B-VI values:

Cross Section	ENDF/B-VI.5	Present Evaluation	Ratio
Total	4.0138 b	4.0297 b	1.004
Capture	0.190 mb	0.196 mb	1.032

We used the experimental Γ_γ , 2.7 ± 0.5 eV, for the 434 keV resonance and Γ_γ = 0.25 eV for all other resonances to compute the resonance capture integral, I_γ. Our value, I_γ = 0.24 mb, agrees with the value of 0.27 ± 0.03 mb given by Mughabghab and Garber[31] but is smaller than the more recent value of 0.36 mb given by Mughabghab, et al.[32]

3. Integral Test: Thermal Reactor Benchmarks

Point-wise cross sections generated from our Reich-Moore resonance parameter representation were used for five thermal reactor benchmarks[33] consisting of three reflected and two bare spheres of highly enriched uranium as aqueous solutions of uranyl fluoride. These benchmarks are useful for testing fast scattering by H_2O as well as ^{235}U fission and capture in the thermal range. Calculated multiplication factors, k_{eff} , were obtained with the BONAMI-NITAWL-XSDRNPM sequence of the SCALE-4.3 system[34] using the 199-group VITAMIN-B6 cross section data library.[35] As shown below, k_{eff} values based on the present evaluation agree with k_{eff} values computed with ENDF/B-VI point-wise cross sections.

Benchmark	ENDF/B-VI.5	Present Evaluation	Δk_{eff}
L-7	1.0006	0.9995	-0.0011
L-8	1.0050	1.0047	-0.0003
L-9	1.0020	1.0021	0.0001
L-10	0.9986	0.9974	-0.0012
L-11	0.9997	0.9996	-0.0001

IV. Summary and Conclusions

We have evaluated ^{16}O neutron cross sections in the resolved resonance region with the multilevel Reich-Moore R-matrix formalism. To give a proper treatment for the α particle exit channel, an algorithm to calculate charged particle penetrabilities and shifts was incorporated into the SAMMY code. Routines based on the CPP algorithm have been integrated into a prototype modification of NJOY. An ENDF format revision will be proposed to accommodate this new feature.

When uncertainties are considered, there is good agreement between theory and experiment for ^{16}O total, (n, α) and differential elastic cross sections up to E_n = 6.3 MeV. New J^{π} assignments are proposed for levels with E_R = 5993 keV [$3/2^-$] and 6076 keV [$9/2^+$]. Point-wise cross section values based on our Reich-Moore resonance parameter representation have been used for several thermal reactor benchmark calculations; the predicted k_{eff} values are in excellent agreement with values computed using ENDF/B-VI.5 point-wise cross sections. Thermal values of total and capture cross sections agree with the corresponding ENDF/B-VI values.

For ^{16}O neutron cross section data, the present Reich-Moore evaluation gives an accurate, few-parameter representation that should be extremely useful for radiation transport calculations in criticality safety analyses. Since the present evaluation fits the 2.35 MeV "window" data much better than does ENDF/B-VI, it should give more reliable results for applications that are sensitive to σ_{total} in this energy region.

Acknowledgments

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