New Resonance Parameter Evaluation of Cl Neutron Cross Sections

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Abstract. Better measurements and evaluations are needed for many elements where the existing evaluations or the underlying nuclear cross section data are not sufficiently accurate for reliable calculation of criticality safety margins. Deficiencies in the existing ENDF/B-VI data evaluation for Cl led to our resonance parameter evaluation of Cl neutron cross sections in the resolved resonance region with the multilevel Reich-Moore R-matrix formalism. Our evaluation takes advantage of recent high-resolution capture and transmission measurements at the Oak Ridge Electron Linear Accelerator (ORELA) as well as older total cross section measurements at Karlsruhe (KFK) to extend the resolved resonance energy range to 1.2 MeV with much more accurate representation of the data than previous evaluations.

1. INTRODUCTION

Improved measurements and evaluations are needed for elements where the existing evaluations or the underlying nuclear cross section data are not sufficiently accurate for reliable criticality safety calculations. Chlorine is important in applications where chlorides are present in significant amounts; e.g., polyvinyl chloride pipe is 57% Cl by weight. Several deficiencies in the existing ENDF/B-VI data evaluation [1] for Cl have been noted previously [2]. Herein we describe a resonance parameter evaluation in the resolved resonance region with the multilevel Reich-Moore R-matrix formalism using the code SAMMY [3]. Recent high-resolution capture and transmission measurements at ORELA allowed us to extend the resolved resonance energy range to 1.2 MeV with much more accurate representation of the data than previous evaluations. A previous report [4] includes tabulations of the resonance parameters as well as more details of the evaluation procedures and fit results.

2. EXPERIMENTAL DATA

The total cross section data include measurements by Guber et al. [2] and Good et al. [5] on the 80-m flight path at ORELA; Cierjacks et al. [6] on a 57-m flight path at the Karlsruhe Isochronous Cyclotron; Singh et al. [7] on the 200-m flight path at the Columbia synchrocyclotron; Brugger et al. [8], who utilized a crystal spectrometer and also the MTR fast chopper with a flight path of 45 m; Kiehn et al. [9] with the Rockefeller generator; and Newson et al. [10] at the Duke Van de Graaff facility. Also included in the evaluation were the high-resolution capture cross section data ($0.1 < E_n < 500$ keV) of Guber et al. [2] and the older, low-resolution capture data ($0.02 < E_n < 1.0$ keV) of Kashukeev et al. [11]. In some cases, normalizations and energy transformations were applied to achieve consistent backgrounds and peak energies. The ^{35}Cl(n,p)^{35}S cross section data of Koehler [12] and Druyts et al. [13] were also included in the evaluation.

3. ANALYSIS AND RESULTS

Resonance parameters were determined by a consistent analysis that included corrections for Doppler broadening, resolution broadening, multiple scattering, and other experimental effects. 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 67 s-wave and 319 p-wave resonances in the range $0.2 < E_n < 1200$ keV. Of these 386 s- and p-wave resonances, 248 were assigned to ^{35}Cl and 138 to ^{37}Cl. Below 160 keV, the capture data of Macklin [14] for a sample enriched to 98.2% in ^{37}Cl were used to identify several ^{37}Cl resonances. Two negative-energy resonances were included to account for bound levels, and several high-energy resonances were included to account for the effect of resonances above 1200 keV. J^π values were assigned to 40 levels in ^{35}Cl and 8 levels in ^{37}Cl on the basis of detailed shape and area analysis of capture and transmission data.

In order to give a proper treatment for charged particles in an exit channel, an algorithm [15] to calculate charged-particle penetrabilities and shifts was incorporated in the SAMMY code. The radii used to compute hard-sphere phase shifts were allowed to vary, and different radii were allowed for s- and p-waves.

For $E_n < 1$ keV, Fig. 1 shows a global view of the final fits to the total cross section data of Refs. [2, 8], the ^{35}Cl(n,p) cross section values of Ref. [12], and the low-resolution capture data of Ref. [11]. These data were analyzed sequentially to obtain parameters for the bound state at −180 eV.

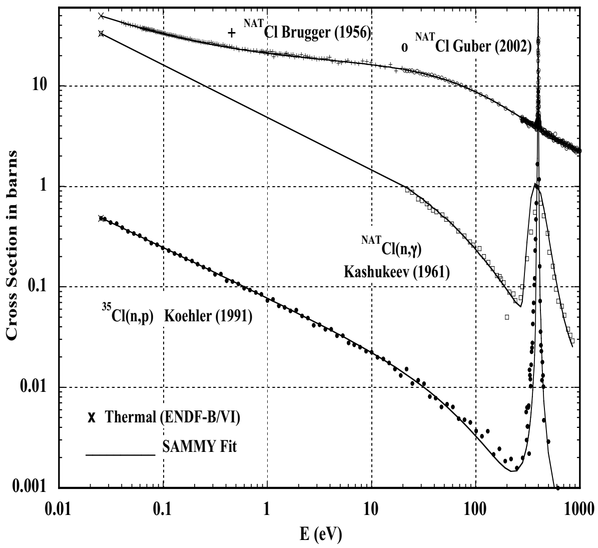

FIGURE 1. Comparison of SAMMY fits (solid lines) to the ^{nat}Cl total cross section data of Ref. [8] (+ symbols) and Ref. [2] (open circles); the ^{nat}Cl capture data of Ref. [11] (open squares); and the ^{35}Cl(n,p) data of Ref. [12] (solid circles). The x symbols denote the ENDF/B-VI thermal values.

Guber et al. [2] measured the transmission of a natural CCl_4 sample (thickness for Cl 0.2075 atoms/b); these data exhibit much higher energy resolution and lower background than the older data sets. This high resolution is shown in Fig. 2, where fits are compared with data for $50 < E_n < 400$ keV. We also fit the thin (0.00812 atoms/b) sample data of Good et al. [5] to obtain an accurate neutron width for the 398-eV resonance.

3.1. (n,p) Cross Section Analysis

The ^{35}Cl(n,p)^{35}S data were analyzed with SAMMY, and fits are compared with the data of Koehler in Fig 1. Fits for peak regions are also in good agreement with the data of Druyts et al. [13].

A wide range of ^{35}Cl(n,p) thermal cross section values has been reported [16] from both activation and proton-emission experiments. In the ^{35}Cl(n,p) analysis we tried data normalizations that corresponded to varying the thermal (n,p) cross section from 440 to 483 mb. For resonances at 0.398 and 4.251 keV, Γ_p is a significant fraction of the total width, hence σ_{total} and σ_γ are sensitive to Γ_p . In addition, Γ_p depends on the resonance strengths ω = $g\Gamma_n\Gamma_p/\Gamma$ deduced by Koehler and Druyts et al. from peak area analysis. Thus, Γ_p and normalization values must give resonance strengths consistent with experimental peak areas as well as satisfactory fits to the transmission, capture, and ^{35}Cl(n,p) data. We could not find acceptable fits to all the data with a normalization significantly lower than $\sigma_{thermal}$ = 483 mb.

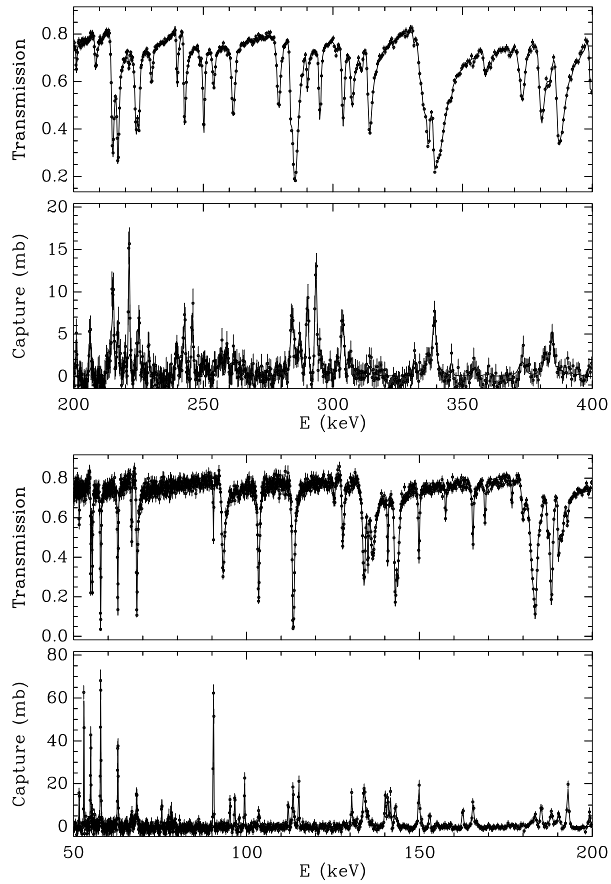

FIGURE 2. Comparison of SAMMY fits (solid lines) to the ^{nat}Cl capture (lower) and transmission (upper) data of Guber et al. [2] for $50 < E_n < 400$ keV.

Widths used in our evaluation are given in Table 1 for the 0.398- and 4.251-keV resonances, and resonance strengths are compared with those of Refs. [12, 13, 17].

3.2. Capture Cross Section Analysis

Guber et al. [2] measured the neutron capture of chlorine up to 500 keV using a natural LiCl sample of thickness 0.09812 atoms/b and the ORELA capture system, which had been re-engineered [18] to minimize structural material near the sample and detectors.

TABLE 1. Proton Widths and Resonance Strengths ω = g$\Gamma_n\Gamma_p/\Gamma$ for ^{35}Cl(n,p) from the present evaluation compared with the data of Druyts et al. [13], Koehler [12], and Gledenov [17]. All data are normalized to a thermal cross section of 483 mb.

E (eV)	J	Γ_γ (meV)	Γ_n (meV)	Γ_p (meV)	ω (meV)	ω[Druyts] (meV)	ω[Koehler] (meV)	ω[Gledenov] (meV)
397.8	2	665	50.5	322 $\pm$ 21	9.8 $\pm$ 0.9	9 $\pm$ 1	10	10.8 $\pm$ 1.6
4250.8	1	472	628	230 $\pm$ 22	40.8 $\pm$ 5.1	42 $\pm$ 3	35	40.0 $\pm$ 8.0

To calculate accurate correction factors for experimental effects of neutron capture data from thick samples, reliable neutron widths are needed. Initial Γ_n values were obtained by fitting the transmission data; using these newly determined Γ_n values, corrections for self-shielding and multiple scattering were calculated with SAMMY and used to determine capture widths. Several iterations of fitting the transmission and capture data produced final resonance parameters that yielded calculated average cross sections that were rather different from those in ENDF/B-VI. This difference was attributed to underestimated neutron sensitivity in the older measurements as well as an improved calculation of the weighting function.

In Fig. 2 we compare the capture cross section from our resonance parameters with the Guber data.

In nuclides where the (n,γ) cross section is small, the direct capture (DC) is often a significant fraction of the cross section. DC calculations [2] with the code TEDCA [19, 20] predict a small DC effect for ^{35}Cl ($\sigma_{thermal}$ = 43.6 b) but a large DC component (72%) for ^{37}Cl ($\sigma_{thermal}$ = 0.433 b). In this evaluation, we deduced a set of resonance parameters, including external level parameters, that reproduce the resonant part of the capture cross section. To this resonant part, one must add the DC contribution to obtain the overall capture cross section. The calculated thermal value of the DC cross section is 0.16 $\pm$ 0.05 b for ^{35}Cl and 0.31 $\pm$ 0.16 b for ^{37}Cl.

3.3. Results

Our elastic, capture, (n,p), and total cross sections for E_n = 0.0253 eV and T = 0 K agree with the corresponding ENDF/B-VI quantities [16]. Resonance capture integral values also agree with the ENDF values.

In Fig. 3 we plot the ^{35}Cl total cross section for T = 300 K as given by the ENDF/B-VI parameters and by the present evaluation. Between resonances, there are large differences ($\approx$ 10% for 30 eV $< E_n <$ 2 keV and $\approx$ 20% for 2 keV $< E_n <$ 200 keV) between the two calculations. The ENDF/B-VI representation above 226 keV, based on calculations utilizing Hauser-Feshbach statistical theory, is clearly inadequate.

Only one spin group (the ^{35}Cl J = 2, s-wave group) contains a sufficient number of resonances for meaningful statistical comparisons; the distribution of neutron widths for $0 < E_n < 1$ MeV agrees well with the Porter-Thomas distribution [21].

Our resonance parameters gave the following neutron strength function values for the range $0 < E_n < 1$ MeV:

^{35}Cl:	$10^4\ S_0 = 0.59 \pm 0.12$	45 resonances
	$10^4\ S_1 = 1.11 \pm 0.12$	172 resonances
^{37}Cl:	$10^4\ S_0 = 0.20 \pm 0.07$	19 resonances
	$10^4\ S_1 = 0.65 \pm 0.09$	115 resonances

The rather small value of S_0 for ^{37}Cl suggests that some tentatively assigned p-wave resonances for ^{37}Cl may, in fact, be s-wave resonances. For the expected 2J + 1 distribution of the number of resonances, the p-wave/s-wave ratio is 3/1. For ^{35}Cl the ratio is 3.8, whereas it is about 6 for ^{37}Cl. The ^{38}Cl structure could also play a role in strength reduction; for example, there are only a few known positive parity levels in ^{38}Cl.

Uncertainties were obtained for resonance energies, capture widths, and neutron widths. The energy uncertainties include fitting uncertainties taking into account correlations between the energy and width(s) of a particular resonance and correlations between energies of neighboring resonances. Also included was the uncertainty in the energy scale, which we estimate to be given by $(dE)_s = 1.5 \times 10^{-4} E\sqrt{1 + 5.32 \times 10^{-6} E}$ where $(dE)_s$ and E are in eV. Width uncertainties include both fitting uncertainties and systematic uncertainties related to background, normalization, etc. Correlations between widths were also taken into account. For each resonance, several SAMMY calculations with different width values were performed and overlaid with the data. Both the overlay plots and the variation in χ^2 with width were used to determine final uncertainties that were, in most cases, significantly larger than the SAMMY values.

Both ^{35}Cl and ^{37}Cl have ground state spin 3/2 and positive parity. Thus s waves give two spin groups: J^π= 1^+ and 2^+; p waves give six spin groups: J^π= 0^-, 1^-, 2^- for channel spin 1 and 1^-, 2^-, 3^- for channel spin 2. We assumed that, for the energy region of interest ($E_n <$ 1.2 MeV), d waves could be neglected. However, for 1 MeV neutrons the ^{35}Cl penetrabilities for s-, p-, and d-waves are 1.030, 0.530, and 0.087, respectively. Thus, some of the weak, high-energy resonances may be d-waves.

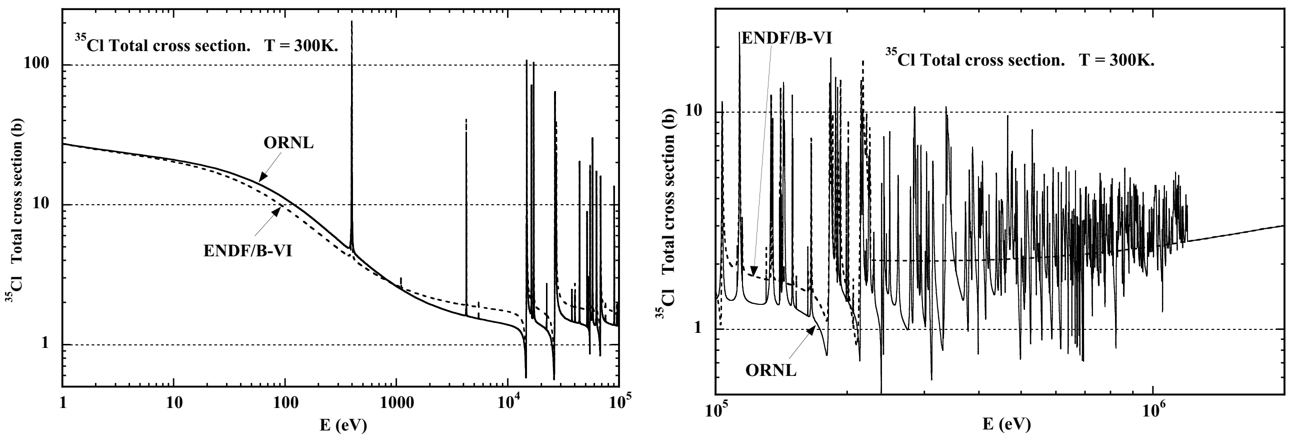

FIGURE 3. Comparison of ^{35}Cl total cross sections from ENDF/B-VI and the present evaluation.

4. SUMMARY AND CONCLUSIONS

The Cl data used in this evaluation include recent ORELA high-resolution capture and transmission measurements as well as several older data sets. Since the ^{35}Cl(n, p)^{35}S reaction yields a significant contribution to the total cross section from thermal energies up to about 10 keV, the ^{35}Cl(n, p) data were fit to obtain proton widths for several resonances. The proton widths are significant fractions of the total widths for resonances at 398 and 4251 eV. When uncertainties are considered, there is good agreement between our resonance parameter calculations and experiment for ^{nat}Cl total cross sections up to E_n = 1200 keV, for ^{35}Cl(n,p) cross sections up to E_n = 100 keV, and for ^{nat}Cl capture cross sections up to 500 keV. Our thermal cross section values agree with the corresponding ENDF/B-VI quantities.

The present evaluation provides resonance energies and widths for 386 s- and p-wave resonances in the range 0.2 to 1200 keV. Of these resonances, 248 were assigned to ^{35}Cl and 138 to ^{37}Cl. Values for J^{π} were assigned to 40 levels in ^{35}Cl and 8 levels in ^{37}Cl. Our evaluation fits the data much better than does ENDF/B-VI.8. This new representation should lead to more reliable criticality safety calculations for systems where Cl is present.

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