**R-Matrix Evaluation of Cl Neutron Cross Sections up to 1.2 MeV**

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**ABSTRACT**

We have performed an evaluation of $^{35}\text{Cl}$, $^{37}\text{Cl}$, and $^{\text{nat}}\text{Cl}$ neutron cross sections in the resolved resonance region with the multilevel Reich-Moore R-matrix formalism. Resonance analyses were carried out with the computer code SAMMY, which utilizes Bayes’ method, a generalized least-squares technique. Because SAMMY now has the ability to calculate charged-particle penetrabilities, it was possible to include a proton exit channel in our evaluation. Our resonance parameter representation describes the data much better than does ENDF/B-VI.8 and should lead to improved calculations for systems where Cl is present.
INTRODUCTION

Many shielding and criticality safety applications rely on existing evaluations of nuclear data. 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 calculations. Chlorine is important in shielding and criticality safety 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 the Oak Ridge Electron Linear Accelerator (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.

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.\textsuperscript{(9)} with the Rockefeller generator; and Newson et al.\textsuperscript{(10)} at the Duke Van de Graaff facility. Also included in the evaluation were the high-resolution capture cross section data of Guber et al.\textsuperscript{(2)} and the older, low-resolution capture data of Kashukeev et al.\textsuperscript{(11)} In some cases normalizations and energy transformations were applied to achieve consistent backgrounds and peak energies.

The $^{35}\text{Cl}(n,p)^{35}\text{S}$ cross section data of Koehler\textsuperscript{(12)} and Druyts et al.\textsuperscript{(13)} were also included in the evaluation. The proton widths are significant fractions of the total widths for resonances at 398 and 4251 eV.

RESONANCE 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 $^{35}\text{Cl}$ and 138 to $^{37}\text{Cl}$. Below 160 keV the capture data of Macklin\textsuperscript{(14)} for a sample enriched to 98.2% in $^{37}\text{Cl}$ were used to identify several $^{37}\text{Cl}$ 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. Values for $J^\pi$ were assigned to 40 levels in $^{35}\text{Cl}$ and 8 levels in $^{37}\text{Cl}$ 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 (CPP) 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 and 8, the \(^{35}\text{Cl}(n,p)\) cross section data of Ref. 12, the low-resolution capture data of Ref. 11, and the ENDF/B-VI thermal values. These data were analyzed sequentially to obtain parameters for the bound state at \(-180\) eV.

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 transmission data for \(0 < E_n < 500\) keV. We also fit the thin (0.00812 atoms/b) sample transmission data of Good et al.\(^{(5)}\) to obtain an accurate neutron width for the 398-eV resonance.

When uncertainties are taken into account, our resonance parameter fits are in good agreement with the total cross section data of Ref. 6 for 500 to 1250 keV, Ref. 8 for 0.04 to 3 eV, and Ref. 7 for 4 to 400 keV, as well as the newer transmission data of Ref. 2 for 0.02 to 1250 keV.

**\((n,p)\) Cross Section Analysis**

The \(^{35}\text{Cl}(n,p)^{35}\text{S}\) 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.

A wide range of \(^{35}\text{Cl}(n,p)\) thermal cross section values has been reported\(^{(16)}\) from both activation and proton-emission experiments. In the \(^{35}\text{Cl}(n,p)\) 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, $\Gamma_p$ is a significant fraction of the total width, hence $\sigma_{\text{total}}$ and $\sigma_\gamma$ are sensitive to $\Gamma_p$. In addition, $\Gamma_p$ depends on the resonance strengths $g\Gamma_n\Gamma_p/\Gamma$ deduced by Koehler (12) and Druyts et al. (13) from peak area analysis. Thus, $\Gamma_p$ and normalization values must give resonance strengths consistent with experimental peak areas as well as satisfactory fits to the transmission, capture, and $^{35}\text{Cl}(n,p)$ data. We could not find acceptable fits to all the data with a normalization significantly lower than $\sigma_{\text{thermal}} = 483$ mb. Proton, neutron, and capture widths are given in Table 1 for the 0.398- and 4.251-keV resonances. Resonance strengths $\omega = g\Gamma_n\Gamma_p/\Gamma$ are compared with corresponding values of Refs. 12, 13, and 17. For the eight resonances from 14.8 to 103.5 keV, $\Gamma_p$ values were computed from the $\omega$ values of Ref. 13.

**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 the amount of structural material surrounding the sample and detectors. To calculate accurate correction factors for experimental effects of the neutron capture data, reliable neutron widths are needed since the sample was fairly thick. Initial $\Gamma_n$ values were obtained by fitting the transmission data; using these newly determined $\Gamma_n$ 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. 3 we compare the capture cross section computed from our resonance parameters with the data of Guber et al. (2).

In nuclides where the $(n, \gamma)$ 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
$^{35}\text{Cl}$ ($\sigma_{\text{thermal}} = 43.6\text{ b}$) but a large DC component ($\sim 72\%$) for $^{37}\text{Cl}$ ($\sigma_{\text{thermal}} = 0.433\text{ b}$). In this evaluation, we have deduced a set of resonance parameters, including the 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\textsuperscript{(2)} thermal value of the DC cross section is $0.16 \pm 0.05\text{ b}$ for $^{35}\text{Cl}$ and $0.31 \pm 0.16\text{ b}$ for $^{37}\text{Cl}$.

**Comparison with ENDF/B-VI**

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

In Fig. 4 we plot the $^{35}\text{Cl}$ total cross section for $T = 300\text{ K}$ as given by the ENDF/B-VI parameters and by the present evaluation. Between resonances, there are large differences ($\sim 10\%$ for $30\text{ eV} < E_n < 2\text{ keV}$ and $\sim 20\%$ for $2\text{ keV} < E_n < 200\text{ keV}$) between the two calculations. These differences reflect the more accurate recent ORELA transmission measurements. The new ORELA measurements and the older KFK measurements enabled us to extend the resonance parameter representation to $1.2\text{ MeV}$; the ENDF/B-VI representation above $226\text{ keV}$, based on calculations utilizing Hauser-Feshbach statistical theory, is clearly inadequate.

**Level Statistics**

Ideally the distribution of neutron widths corresponding to a particular spin group, e.g., $J^\pi = 2^+$, is expected to follow the hypothesis of Porter and Thomas,\textsuperscript{(21)} and the nearest-neighbor level spacings are expected to be apportioned according to the Wigner distribution.\textsuperscript{(22)} For our Reich-Moore representation of $^{35}\text{Cl}$ and $^{37}\text{Cl}$ resonance parameters, only one spin group (the $^{35}\text{Cl} J = 2$, s-wave group) contains a sufficient number of resonances for meaningful statistical comparisons. The distribution of $J = 2$, s-wave
$^{35}$Cl neutron widths in the energy range $0 < E_n < 1$ MeV agrees well with the Porter-Thomas distribution, considering that there are only 26 resonances.

For the energy range $0 < E_n < 1$ MeV, our distribution of $J = 2$, s-wave level spacings for $^{35}$Cl contains more closely spaced resonances than does the Wigner distribution. However, some of these closely spaced resonances are rather weak, and spin assignments are not definite for these resonances.

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

$^{35}$Cl: $\ 10^4 S_0 = 0.59 \pm 0.12 \ \ (45 \text{ resonances})$ $\ 10^4 S_1 = 1.11 \pm 0.12 \ \ (172 \text{ resonances})$

$^{37}$Cl: $\ 10^4 S_0 = 0.20 \pm 0.07 \ \ (19 \text{ resonances})$ $\ 10^4 S_1 = 0.65 \pm 0.09 \ \ (115 \text{ resonances})$

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. One expects the number of resonances to be distributed roughly as $2J + 1$, which implies a p-wave/s-wave ratio of 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 neutron strength reduction; for example, there are only a few known positive-parity levels in $^{38}$Cl.

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 contributes significantly to the total cross section from thermal energies up to about 10 keV, the $^{35}$Cl(n,p) data$^{(12,13)}$ 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 experimental data 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 elastic, capture, (n,p), and total cross sections are in good agreement 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 $J^\pi$ 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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REFERENCES


Table 1. Proton widths and resonance strengths $\omega = g\Gamma_n \Gamma_p / \Gamma$ for $^{35}\text{Cl}(n,p)$ from the present evaluation compared with the data of Druyts et al., Koehler, and Gledenov. Results are normalized to a thermal cross section of 483 mb.

<table>
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<th>E (eV)</th>
<th>J</th>
<th>$\Gamma_Y$ (meV)</th>
<th>$\Gamma_n$ (meV)</th>
<th>$\Gamma_p$ (meV)</th>
<th>$\omega$ (meV)</th>
<th>$\omega$ [Druyts] (meV)</th>
<th>$\omega$ [Koehler] (meV)</th>
<th>$\omega$ [Gledenov] (meV)</th>
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<td>397.8</td>
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<td>4250.8</td>
<td>1</td>
<td>472</td>
<td>628</td>
<td>230 ± 22</td>
<td>40.8 ± 5.1</td>
<td>42 ± 3</td>
<td>35</td>
<td>40.0 ± 8.0</td>
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Fig. 1. Comparison of SAMMY fits (solid lines) with the $^{35}\text{Cl}$ total cross section data of Brugger et al. (open circles); the $^{35}\text{Cl}$ capture data of Kashukeev et al. (open squares); and the $^{35}\text{Cl}(n,p)$ data of Koehler (solid circles). The x symbols denote the ENDF/B-VI thermal values.
Fig. 2. Comparison of SAMMY fits (solid lines) with the $\text{natCl}$ transmission data of Guber et al.\textsuperscript{2}
Fig. 3. Comparison of SAMMY fits (solid line) with the \( ^{nat}\text{Cl} \) capture cross section data of Guber et al.\textsuperscript{2}
Fig. 4. Comparison of $^{35}\text{Cl}$ total cross sections from ENDF/B-VI with the present evaluation.