

New neutron cross section measurements from ORELA and new resonance parameter evaluations

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Abstract. A series of new measurements has been undertaken in response to deficiencies identified in nuclear data libraries of crucial importance to the Nuclear Criticality Safety Program. New data and evaluations, including covariances, are required for several materials found in mixtures with uranium. For this purpose we performed neutron capture and total cross-section measurements on natural potassium, ⁴¹K, and manganese.

1 Introduction

Many neutron cross-section evaluations in libraries such as US Evaluated Nuclear Data File (ENDF/B-VI) or Japanese Evaluated Nuclear Data File (JENDL-3.2) show deficiencies or do not cover energy ranges important for modern criticality safety calculations. Inadequacies may occur in the resolved and unresolved resonance regions. Thus, some of the evaluated data may be insufficient for criticality calculations where effects such as self-shielding, multiple scattering, or Doppler broadening are important. Moreover, for nuclides with small neutron capture cross sections, the evaluations report erroneously large values due to underestimated neutron sensitivity of the experimental set up. Although their neutron capture cross sections are small, these nuclides are sometimes absorbers in criticality calculations, and accurate cross-section data are essential. Such concerns about existing nuclear data have been the prime motivator for new cross-section measurements for the Nuclear Criticality Safety Program (NCSP) of the US Department of Energy. Criticality safety calculations involving nuclear material outside reactors have significant contributions in the epithermal energy region (keV region). Many neutron cross sections are not known with sufficient accuracy. For example, due to their uncertainties in previous measurements, new neutron capture and total cross section measurements are needed for ^{39,41}K and ⁵⁵Mn.

2 Measurements

Neutron capture and total cross-section measurements for ^{39,41}K and ⁵⁵Mn were performed at the Oak Ridge Electron Linear Accelerator (ORELA). ORELA was operated at a repetition rate of 525 Hz, a burst width of 8 ns, and a power of 5 to 6 kW. The combination of short pulse width and long flight path resulted in a neutron energy resolution superior to any previously reported measurements for these nuclides.

Neutron capture cross sections for ^{39,41}K and Mn were measured on flight path 7 at a distance of 40 m from the

neutron-producing target using a pair of deuterated benzene (C₆D₆) detectors by employing the pulse-height-weighting method. This apparatus [1] has been improved in several ways, compared to that used in previous measurements at ORELA for these nuclides. Firstly, the original C₆F₆ scintillator was replaced by C₆D₆, which is much less sensitive to backgrounds from scattered neutrons. Secondly, much of structural material surrounding the sample and detectors was removed to reduce the neutron sensitivity even further. For example, the massive Al-sample changer and beam pipe were replaced by a thin carbon fiber tube. Additionally, the massive detector housings were replaced with reduced-mass detector mounts. Thirdly, the appropriate detector weighting function for each experiment is now calculated using the more sophisticated computer codes EGS4 and MCNP. All structural materials within 30 cm of the detectors, including the sample, are incorporated into these calculations. More details about these improvements can be found in the papers by Koehler et al. [1, 2].

The neutron flux was measured using a 0.5-mm-thick ⁶Li-glass scintillator at a distance of 39.695 m from the neutron target. The final normalization of the capture efficiency was carried out in a separate measurement using the “saturated resonance” technique by means of the 4.9-eV resonance from a gold sample [3].

The neutron capture cross section of ⁵⁵Mn was measured using a solid metallic ⁵⁵Mn sample with dimensions of 2.54 × 5.08 × 0.317 cm. The experimental data are shown in figure 1 and compared to the cross section calculated using SAMMY [4] with ENDF/B-VI resonance parameters. This calculation included experimental effects such as self-shielding and multiple scattering. There are substantial differences between ENDF and the data, including missing resonances in the evaluation.

For ³⁹K we used a natural potassium carbonate sample with a thickness of 0.0088 at/b. The ⁴¹K neutron capture experiments using ⁴¹KCl were performed with a sample enriched to 99.17% in ⁴¹K and a thickness of 0.005617 at/b. Even though the sample contained chlorine, it will be possible to extract reliable resonance parameters for ⁴¹K using SAMMY by including the most recent chlorine resonance parameters [5] in the analysis. The neutron capture cross sections obtained from this experiment show that there are discrepancies compared

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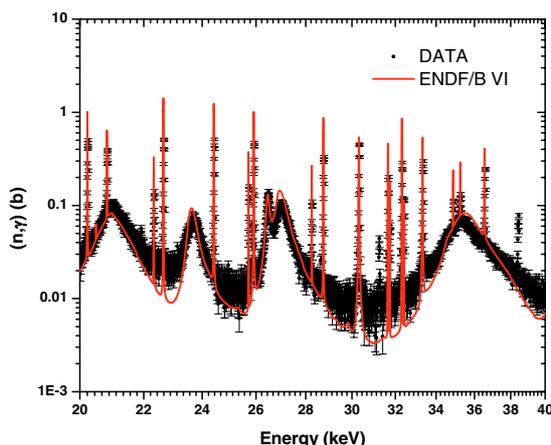


Fig. 1. New ORELA neutron capture data for Mn compared to the calculated cross section using ENDF/B VI parameters.

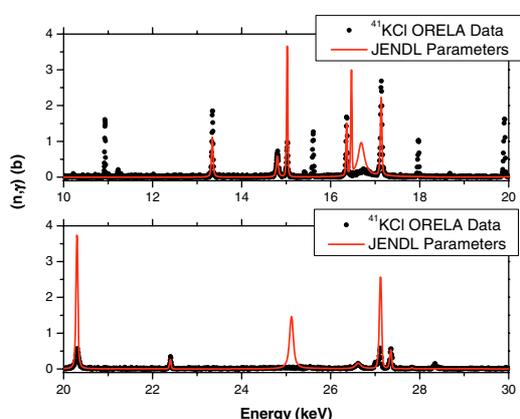


Fig. 2. New ORELA neutron capture data for ^{41}KCl compared to the calculated cross section using the JENDL 3.2 parameters.

to the cross section calculated using the resonance parameters from the JENDL 3.2 library. Figure 2 shows a small part of the data from this measurement, illustrating that many resonances for ^{41}K are missing from the evaluation and that several resonances in the evaluation have been overestimated. The calculated cross section included experimental effects such as self-shielding, multiple scattering, and Doppler and resolution broadening.

Usually neutron capture experiments are performed with a fairly thick sample; therefore, the corrections for self-shielding and multiple scattering can be sizeable. To apply the most accurate corrections for these experimental effects, we made corresponding total cross-section measurements. These data also can be useful in making isotopic assignments as well as for observing some resonances not visible in the (n,γ) data. For the total cross-section measurements on Mn, we used a metallic powder sample having a thickness of 0.119 at/b. It was mounted in a computer-controlled sample changer positioned at about 10 m from the neutron target in the beam of ORELA. The natural potassium transmission measurement was carried out using two metallic samples (0.013367 and 0.10517 atom/b). For ^{41}K we used the same ^{41}KCl material

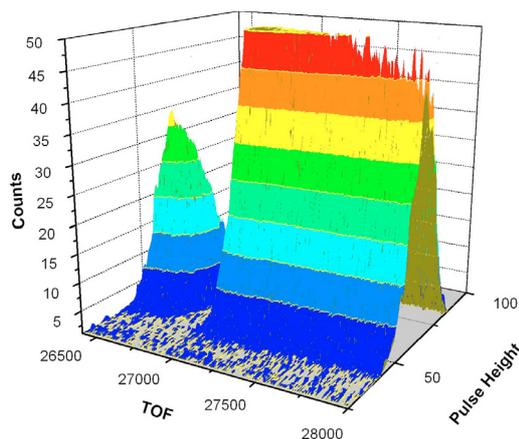


Fig. 3. Transmission data for ^{41}KCl acquired with the new data acquisition system, which records pulse-height vs TOF.

as in the capture experiments after it was reshaped to yield a thickness of 0.0155 at/b. A presample collimator limited the beam size to 2.54 cm at the sample position and allowed only neutrons from the water-cooled part of the neutron source to be seen by the detector. As a neutron detector we used an 11.1-cm-diameter, 1.25-cm-thick ^6Li -glass scintillator positioned in the beam at the 80-m flight station at a distance of 79.815 m from the neutron source.

The total cross section was measured using a modernized data acquisition apparatus. Two channels of an Acqiris digitizer were used to digitize signals from the ^6Li -glass detector and determine both time-of-flight (TOF) and pulse height for each event. The data were written to disk in event mode as well as displayed online in histograms. By using the digitizer many of the previous data acquisition electronics were removed; therefore the new system with fewer components should be much more reliable. A small portion of the two-dimensional data, TOF vs pulse-height, is shown in figure 3 for the ^{41}KCl transmission measurements.

3 Evaluation

Resonance analyses were carried out with the computer code SAMMY, which utilizes Bayes' method, a generalized least-squares technique. Doppler and resolution broadening, multiple scattering corrections, and other effects were taken into account. Our evaluation for K includes our new data mentioned above as well as previous total cross-section data for enriched KCl samples [6]. A measurement from the Research Center at Karlsruhe (FZK) was included to extend the resolved resonance energy range up to 1.0 MeV [7].

In figure 4 a preliminary SAMMY fit to neutron capture data using the K_2CO_3 sample is shown. Because multiple isotopes can be included in SAMMY, it is possible to analyze mixtures of isotopes and even chemicals as it is the case for the KCl samples. To fit those samples, the most recent chlorine resonance parameters were included in the parameter file and kept fixed. This feature in SAMMY makes it possible to extract the resonance parameters of the isotope of interest. Preliminary fits to the transmission data for $^{39,41}\text{KCl}$ samples

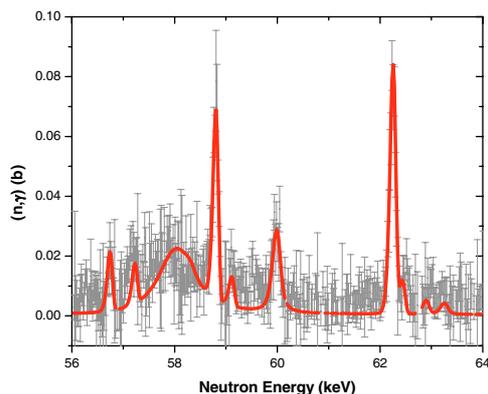


Fig. 4. Preliminary SAMMY fit to the neutron capture data for K_2CO_3 taken at ORELA.

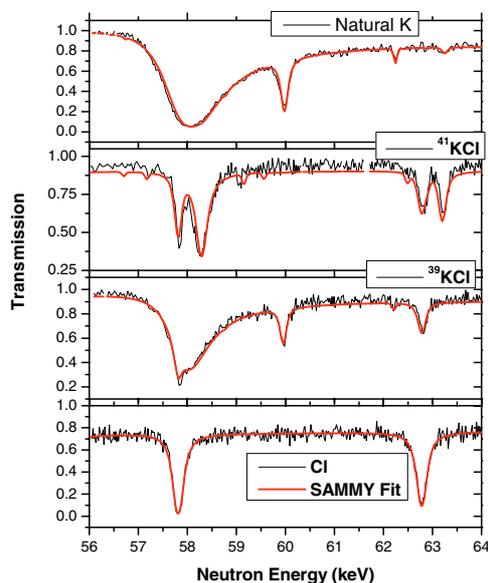


Fig. 5. Preliminary SAMMY fit to the transmission data for natural K, ^{41}KCl , ^{39}KCl , and Cl.

are shown together with the Cl transmission data in figure 5. We expect that our new data and resonance analysis will result

in improved resonance parameters for K and hence improved criticality safety calculations for systems containing K.

4 Conclusion

To support the Nuclear Criticality Safety Program, we performed new neutron total and capture measurements over broad energy ranges for K and Mn. These data will complete a series of measurements made at ORELA to be used in an evaluation of the neutron capture and total cross sections of $^{39,41}K$ and Mn. With these new data it will be possible to obtain improved resonance parameter sets for potassium and manganese in the resolved and unresolved resonance range using the multilevel R-matrix code SAMMY. In all cases we were able to extend the resolved resonance region to much higher energies compared to the existing evaluations. These new evaluations should lead to much more reliable nuclear criticality calculations.

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