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# New Neutron Cross-Section Measurements on $^{19}$F, $^{39,41}$K, $^{55}$Mn, and $^{103}$Rh for Improved Nuclear Criticality Safety

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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 as well as for burnup credit studies involving the transportation of spent nuclear fuel. New data and evaluations including covariances are required for several stable fission products as well as for materials found in mixtures with uranium.

***KEYWORDS: neutron cross section, transmission, $^{19}$F, $^{39,41}$K, $^{103}$Rh, $^{55}$Mn***

## 1. Introduction

Many neutron cross-section evaluations in libraries such as U.S. Evaluated Nuclear Data File (ENDF/B-VI) or Japanese Evaluated Nuclear Data File (JENDL-3.2) exhibit deficiencies or do not cover energy ranges important for criticality safety applications. Deficiencies may occur in the resolved and unresolved-resonance regions. Therefore, some of these evaluated data may not be adequate for criticality calculations where effects such as self-shielding, multiple scattering, or Doppler broadening are important. Furthermore, many neutron cross section evaluations for nuclides having small neutron capture cross sections are erroneously large. Although their neutron capture cross sections are small, these nuclides can be important absorbers in many 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 U.S. Department of Energy (DOE)[**]. To obtain these data, several new measurements were

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[**] Cross –section measurements for $^{19}$F, $^{39,41}$K, and $^{55}$Mn have been performed for the National Nuclear Security Administration Nuclear Criticality Safety Program (NCSP). In addition, $^{55}$Mn measurements have been performed by the U.S. DOE Office of Environmental Management (EM). Measurements for $^{103}$Rh have been performed for the U.S. DOE Office of Civilian Radioactive Waste Management (RW) to support burnup credit applications involving the transport of spent nuclear fuel.

performed as collaborations between Oak Ridge National Laboratory (ORNL) and the Institute for Reference Material and Measurements (IRMM) of the Joint Research Institute of the European Community in Geel, Belgium, and between ORNL and Los Alamos National Laboratory (LANL).

For example, due to their large neutron cross sections and substantial uncertainties in previous measurements, new neutron capture and total cross section measurements are needed for $^{39,41}$K, $^{55}$Mn, and $^{103}$Rh. In addition, evaluations of $^{19}$F neutron inelastic cross sections have been performed for inclusion in the ENDF/B-VI as well as for the JENDL-3.2. The ENDF/B-VI evaluation is based on data of Morgan and Dickens [1], whereas the JENDL-3.2 evaluation is based on data from Broder et al. [2] The two evaluations for the two inelastic channels at 110 and 197 keV show differences as large as 50%. Although a new R-matrix resonance evaluation of $^{19}$F neutron cross sections up to 1 MeV recently was performed by the ORNL Nuclear Data Group [3], this large discrepancy for the inelastic cross section remains, and new measurements are essential for accurate criticality safety calculations for mixtures containing large amounts of fluorine (e.g., $UF_6$). Also, accurate knowledge of the inelastic cross section will result in a better determination of the $\gamma$-ray production cross section for fluorine.

Criticality safety calculations dealing with nuclear material outside reactors frequently encounter neutron spectra very different from the usual thermal or fast neutron spectrum. Indeed the neutron spectrum found in criticality safety calculations has a significant contribution in the epithermal energy region (keV region), where the inelastic cross section for fluorine is not well known.

## 2. Measurements

### 2.1 Inelastic Neutron Cross Section of $^{19}$F using GEANIE

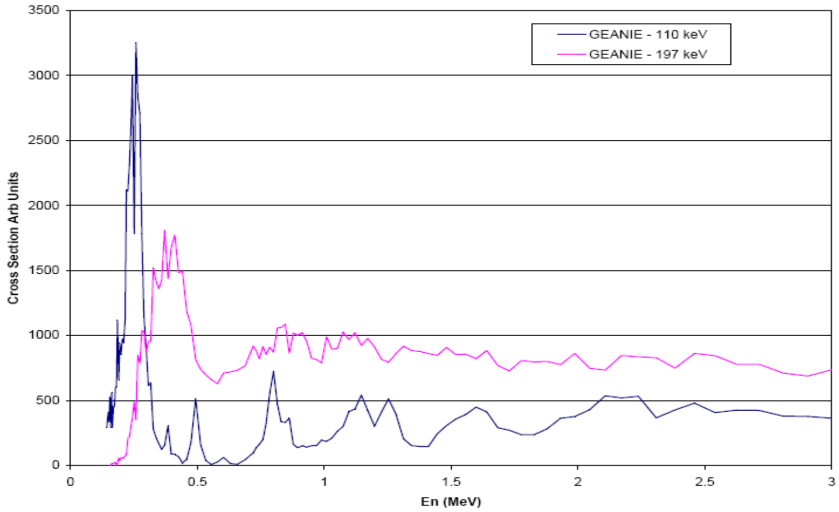
The $^{19}$F(n,n'$\gamma$) cross sections for the two inelastic channels at 110 and 197 keV were measured using the GEANIE spectrometer at the Weapons Neutron Research (WNR) facility of the Los Alamos Neutron Science Center at LANL. GEANIE was used in the “standard configuration” (10 planar plus 16 coaxial Ge detectors) to detect gamma rays from the two excited levels at 110 and 197 keV following inelastic neutron scattering. All detectors were configured for singles $\gamma$-ray cross section measurements with the front faces of the detectors about 15 cm from the sample. The sample utilized for this measurement was high-purity Teflon, which contains only carbon and fluorine.

Preliminary results of our new measurements (see Fig. 1) agree well with the cross section shape of Ref. [1]. These new inelastic data will be incorporated into an R-matrix analysis of $^{19}$F cross sections up to 1 MeV that is being performed by the ORNL Nuclear Data Group [3].

### 2.2 Neutron Cross Section Measurements of $^{103}$Rh and $^{55}$Mn at GELINA

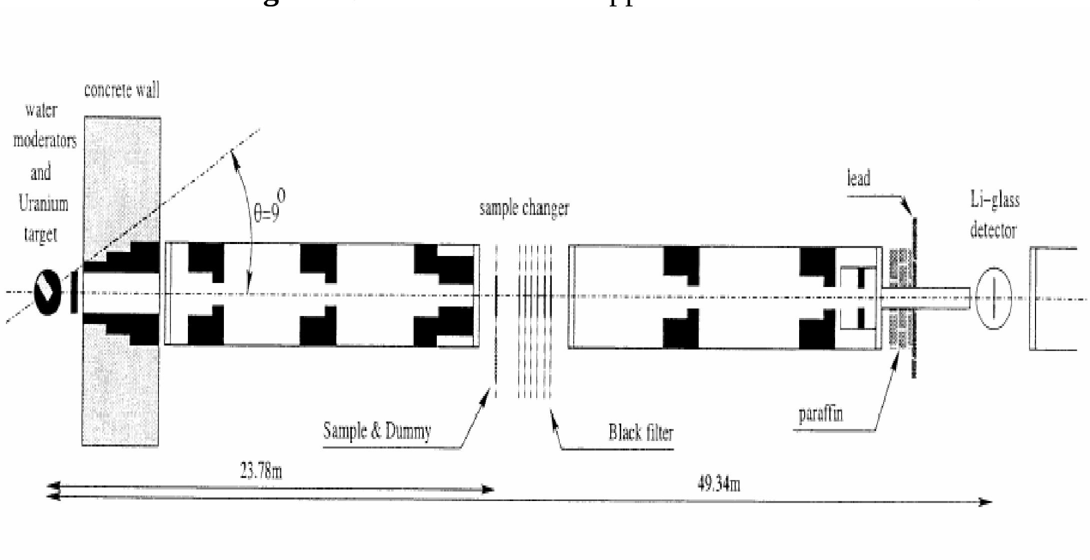
Neutron capture and transmission measurements for $^{103}$Rh and $^{55}$Mn were performed at the Geel Electron Linear Accelerator (GELINA) at IRMM. Due to a special compression system, the accelerated electron pulse at GELINA can be reduced to 1-ns pulse width without intensity loss, allowing for excellent neutron energy resolution using the time-of-flight technique. Neutron capture and total cross-section experiments were run on different flight paths using repetition rates of 800 and 400 Hz, respectively.

**Figure 1:** The inelastic cross section shape of fluorine for the two channels at 110 and 197 keV, respectively.



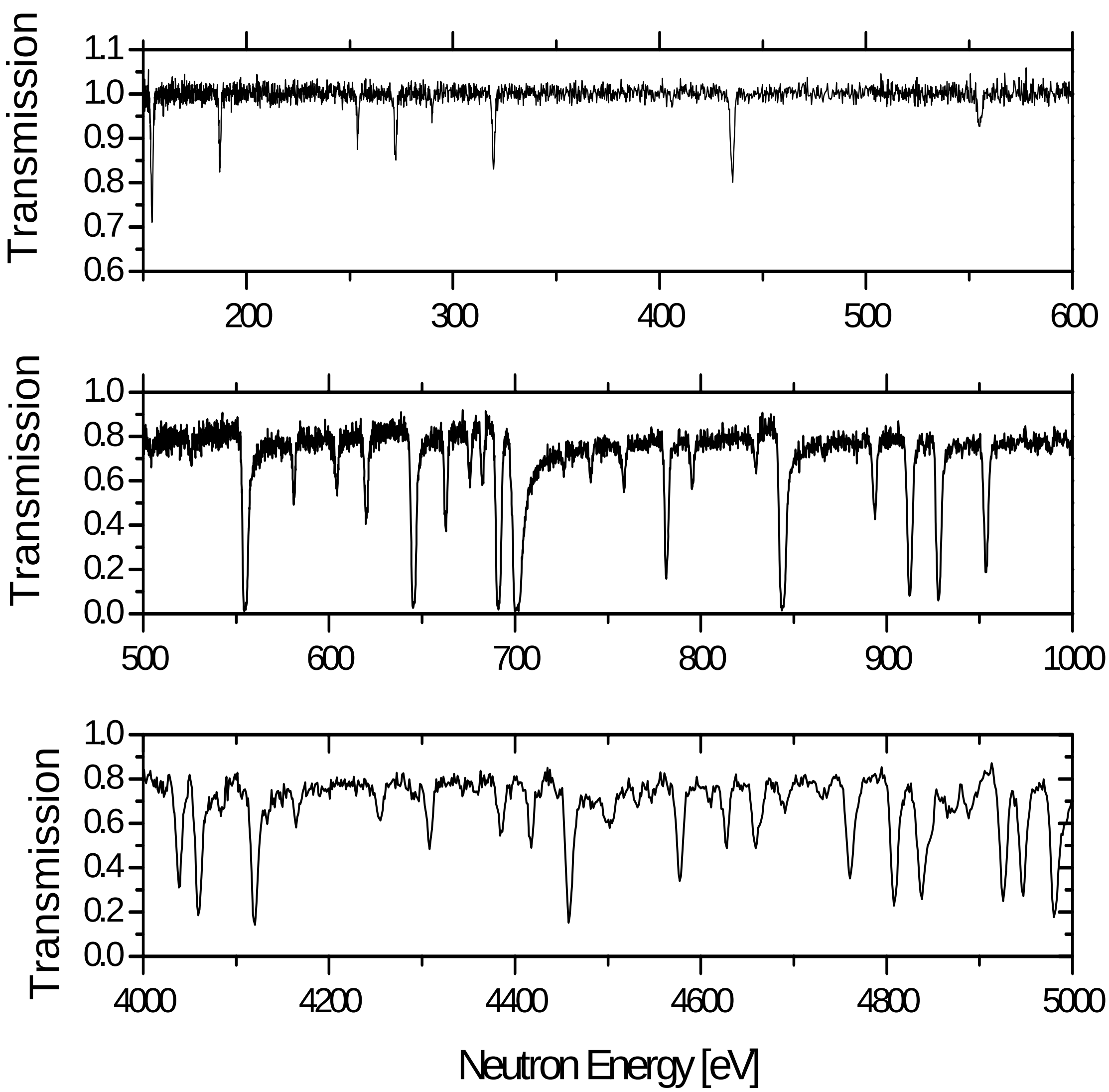

Transmission experiments were run using two different flight path lengths of 49.33 (FP 4, see Fig 2) and 26.45 m (FP 2), respectively. The detector used at the 26-m flight station was a 1.27-cm-thick $^{6}$Li-loaded glass scintillator viewed by two photomultipliers placed outside of the neutron beam. At the 49-m flight station, a 0.635-cm-thick $^{6}$Li-loaded glass scintillator viewed by one photomultiplier placed outside of the neutron beam was used to detect the neutrons. The neutron beam was collimated to 1.5-cm diameter at the sample position. Two $BF_3$ proportional counters served as neutron flux monitor. In addition to sample-in and sample-out measurements, runs were made with filters having blacking-out resonances to determine backgrounds. The useful neutron energy range covered in these experiments was from 100 eV to about 100 keV.

**Figure 2:** The transmission apparatus on FP4 at GELINA.



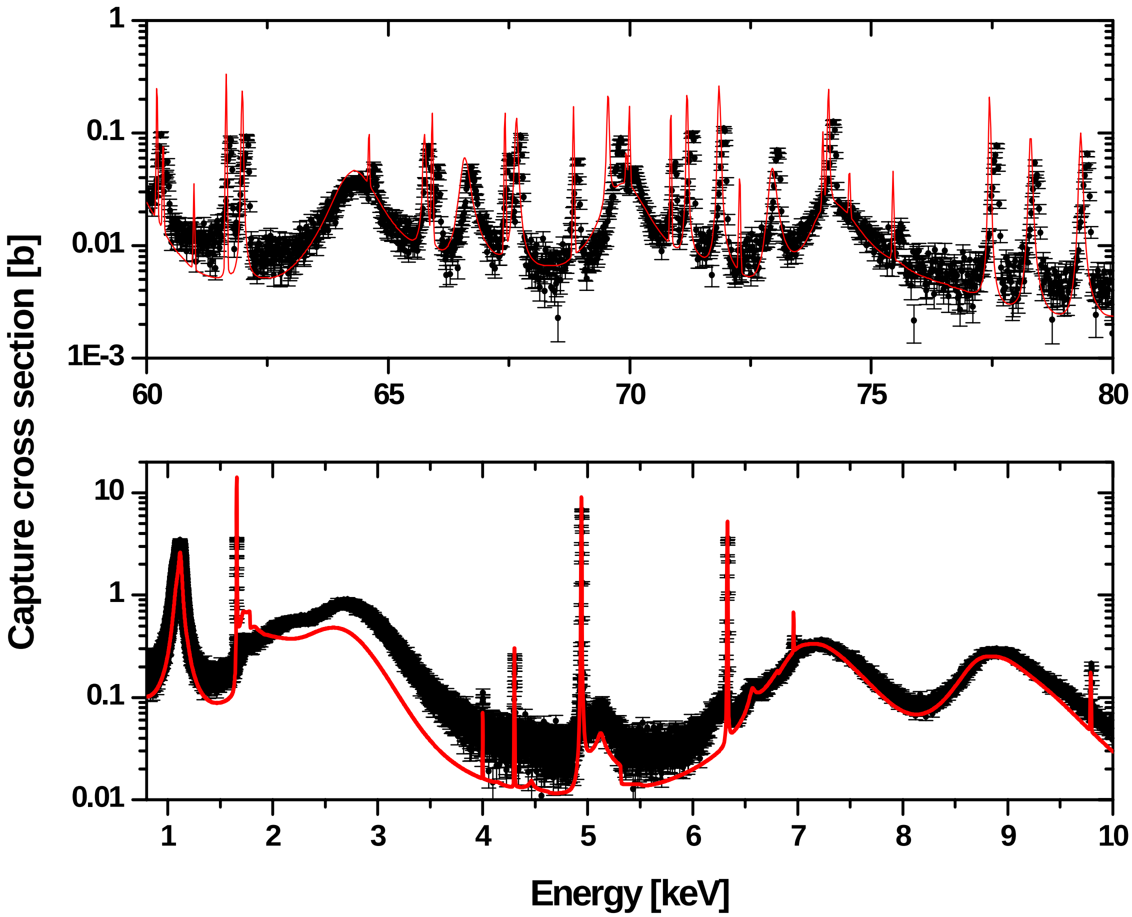
Two $^{103}$Rh samples of different thickness were used, a thin sample at 0.00033 at/b was used at flight path 2 and a thick sample at 0.0458at/b was placed at 23 m in FP4. Results from these measurements are displayed in Fig. 3 for the thin sample in the low neutron energy range and the thick sample at higher energies. The combination of these two experimental data sets will allow a resonance analysis beyond the most recent evaluation, that is, above 4 keV. The total cross section of $^{55}$Mn was measured using FP4 with a 0.118-at/b thick sample positioned at 10 m from the neutron production target.

**Figure 3:** Part of the $^{103}$Rh transmission data for the thin (top) and thick (middle and bottom) samples. Neutron resonances were well resolved up to 10 keV.



Neutron capture experiments were performed at two different flight paths. At flight path 15 with a flight path length of 28.69 m, two $C_6D_6$ detectors were used employing the pulse-height-weighting technique. The detectors are positioned at an angle of $90^O$ with respect to the neutron beam on each side of the sample. A $^{10}B$-loaded ionization chamber located 80 cm in front of the sample served as a neutron flux monitor. Additional measurements were made using an empty sample holder and with a $^{208}Pb$ sample to determine backgrounds from the sample holder and sample scattered neutrons, respectively. Filters with blacking-out resonances were put in the beam for supplementary background determinations, and a $^{10}B$ slab served as a frame overlap filter. The sample used in this experiment was a 7.8-cm-diameter metallic $^{103}Rh$ disc with a thickness of 0.00182 at/b.

**Figure 4:** The $^{55}Mn$ capture yield (black dots) obtained at the 60-m flight path compared to the calculated yield using the ENDF/B-VI parameters and preliminary resolution broadening.



For the $^{55}Mn$ neutron capture experiment, a 0.01899-at/b Mn disc was positioned in the neutron beam of GELINA at the 60-m flight station on flight path 14. The capture γ-rays were detected with four $C_6D_6$ detectors using the pulse-height-weighting technique. The detectors were placed in an angle of $125^O$ with respect to the neutron beam. In this geometry, any anisotropic effects of emitted γ-rays were minimized. A $^{10}B$-loaded Frisch grided ionization chamber located in front of the sample served as a neutron flux monitor. Additional measurements were made using an empty sample holder to determine backgrounds. The black resonance technique was used to determine supplementary backgrounds, and a $^{10}B$ slab served as a frame overlap filter. Fig 4 shows a comparison with the calculated yield using the parameters from the ENDF/B-VI evaluation.

### 2.3 $^{41}K$ Neutron Capture at ORELA

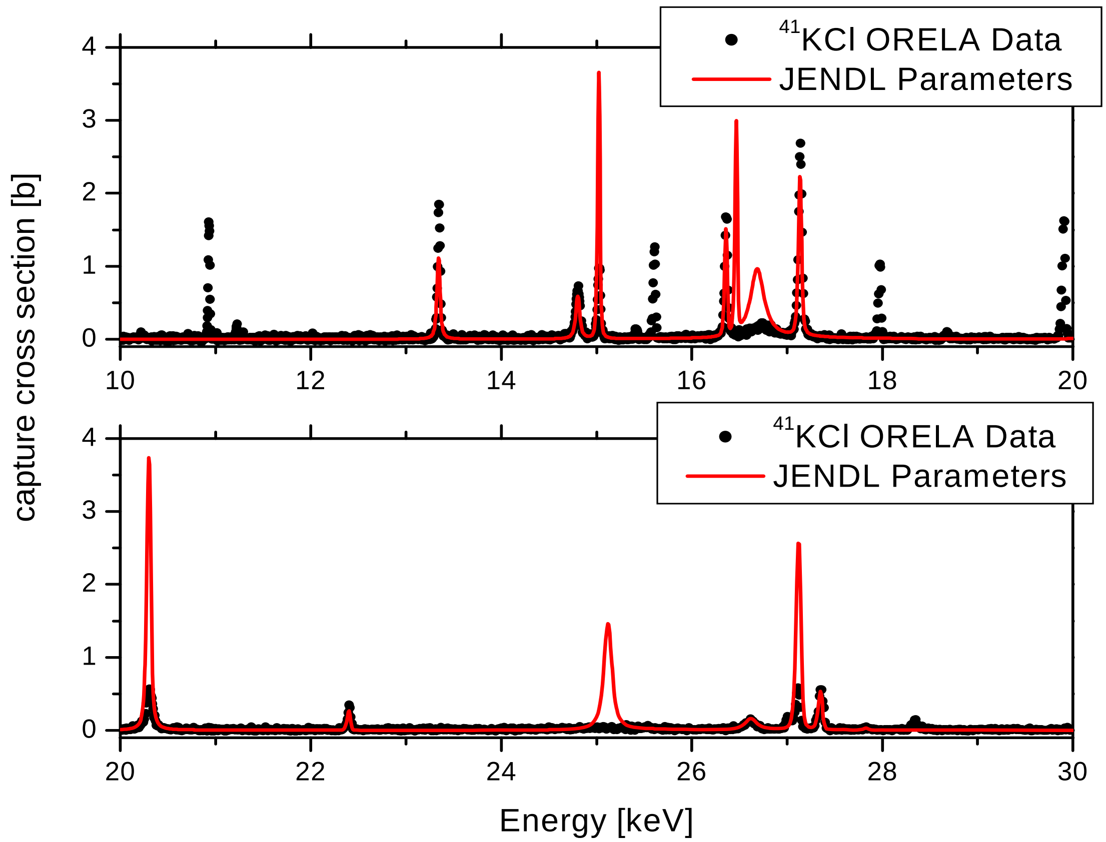
New $^{41}K$ neutron capture experiments using an enriched $^{41}KCl$ sample were performed at the Oak Ridge Electron Linear Accelerator (ORELA). These data will complete a series of measurements made at ORELA to be used in an evaluation of the neutron capture and total cross sections of $^{39}K$ and $^{41}K$.

The neutron capture experiments were performed at the 40-m flight station of ORELA utilizing flight path 7. To detect the capture γ-rays a pair of $C_6D_6$ detectors is used, applying the pulse-height-weighting method. Over the last couple of years, the system has been improved in many ways compared to the old ORELA apparatus [4]: First, most of the structural material surrounding the sample and detectors was greatly reduced to decrease the background from sample-scattered neutrons (neutron sensitivity). This was accomplished by replacing the massive Al-sample changer and beam pipe with a thin carbon fiber tube. The steel detector housings were replaced with reduced-mass detector mounts. Second, the $C_6F_6$ scintillator was replaced by $C_6D_6$, which has much lower neutron sensitivity. More details about these improvements can be found in the papers by Koehler et al. [4, 5]. Third, calculation of the detector weighting functions has been improved by using the Monte Carlo code EGS4, including the sample and all structural materials within 30 cm of the detectors, in the calculations.

A 1.27-cm-thick Pb filter was employed to reduce the γ-ray background from the neutron production target, and pulse overlap neutrons were eliminated with a 0.48-g/cm$^2$ $^{10}B$ filter. Absolute cross sections were determined using the saturated resonance technique, employing the 4.9-eV resonance in gold [6]. A 0.5-mm-thick $^6Li$-glass scintillator, placed 42.1 cm upstream of the sample position, was used to measure the energy dependence of the neutron flux.

The KCl sample used for this experiment was enriched to 99.17% in $^{41}K$ with a thickness of 0.005617 at/b. Even though the sample contains chlorine, it should be possible to extract reliable resonance parameters for $^{41}K$ using SAMMY [7] by including the most recent chlorine resonance parameters [8] in the analysis. The neutron capture cross sections obtained from this experiment shows that there are serious discrepancies compared to the cross section calculated using the resonance parameters from the JENDL-3.2 library. Fig 5 shows a small part of the data from this measurement, illustrating that many resonances for $^{41}K$ are missing and that several resonances in the JENDL-3.2 evaluation have been overestimated. The calculated cross section included all experimental effects such as self-shielding, multiple scattering, and Doppler and resolution broadening. Together with the total cross section data for potassium and capture data for a natural $K_2CO_3$ sample, it will be possible to obtain a more reliable resonance parameter set for potassium in the resolved and unresolved resonance range.

**Figure 5:** Neutron capture data for KCl using a sample enriched in $^{41}K$ compared to the cross section calculated from the JENDL-3.3 parameters.




## 3. Conclusion

In an effort to support nuclear criticality safety and burnup credit applications, we performed new neutron total and capture measurements in a multi laboratory collaboration over broad energy ranges. The measurement results will be analyzed using the multilevel R-matrix code SAMMY. In all measured 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 improved cross-section data for nuclear applications.

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