New Neutron Cross Section Measurements at ORELA for Improved Nuclear Data

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The Oak Ridge Electron Linear Accelerator (ORELA) was used to measure neutron total and capture cross sections of aluminum, silicon, chlorine, fluorine, and potassium in the energy range from 100 eV to ~600 keV. These measurements were carried out to support the Nuclear Criticality Safety Program (NCSP). Concerns about the use of existing cross section data in nuclear criticality calculations have been a prime motivator for the new cross section measurements. Our results are substantially different from the evaluated nuclear data files of ENDF/B-VI and JENDL-3.2.

KEYWORDS: neutron capture cross section, neutron sensitivity, transmission, Al, Cl, Si, F, K

1. Introduction

Concerns about the use of existing cross-section data in the nuclear criticality calculations using Monte Carlo codes and benchmarks have been a prime motivator for the new cross section measurements. Most of the older neutron-induced cross section data used in libraries such as ENDF/B-VI or JENDL-3.2 show deficiencies or do not cover the neutron energy range which is important for a wide variety of applications. For example, many of the older measurements suffer from poor time-of-flight resolution and the description of some data in the neutron energy range above several tens of keV is crude. As a result, these data may not be sufficient for applying certain corrections such as self-shielding, multiple-scattering, or Doppler broadening of individual resonances. This impacts not only the resolved cross section region but also the unresolved region, and could lead to problems in the correct processing of data from data libraries.

The Oak Ridge Electron Linear Accelerator (ORELA) has been used to measure the total and capture neutron cross sections of several elements (Al, Si, Cl, F, and K) in the energy range from 100 eV to ~600 keV. ORELA, the only high power white neutron source with excellent time resolution still operating in the USA, is ideally suited for these experiments. In this paper we report the new findings of mostly significant smaller neutron capture cross sections compared to most recent evaluations.

2. Experimental Set Up

Neutron capture experiments were carried out using flight path 7 at a distance of 40 meters from the neutron production target using the pulse-height weighting technique with a pair of C_6D_6 detectors. Compared to the old ORELA setup [1], the capture system has been improved by minimizing the amount of structural material surrounding the sample and detectors in order to reduce the background due to sample-scattered neutrons (neutron sensitivity) and by calculating the detector weighting function using the computer code EGS4 [2].

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Normalization of the capture efficiency was carried out in a separate measurement using the "black resonance" technique by means of the 4.9-eV resonance from a gold sample [3]. A 0.5-mm thick ⁶Li-glass scintillator served as the neutron flux monitor. We used two extremely high purity (0.01520 a/b and 0.04573 a/b) samples for the aluminum neutron capture measurements. For the silicon capture measurements, we used a high purity natural silicon sample (0.07831 a/b). The neutron capture of chlorine was measured using a natural LiCl sample (0.09812 a/b). A Teflon sample (0.05086 a/b) was used for the fluorine capture experiment, and a natural K_2CO_3 sample (0.0088791 a/b) was used for the potassium measurement.

For an evaluation of the neutron capture cross section, total cross section data are indispensable. Therefore, we made corresponding total cross section measurements when needed. For the Al transmission measurements, the two samples (0.0189a/b and 0.1513 a/b) were mounted in the sample changer positioned at about 10 meters from the neutron target in the beam of ORELA. For the chlorine transmission measurement we used a natural CCl₄ (thickness for Cl 0.2075 a/b) sample. For the potassium transmission we used two metallic samples (0.013367 and 0.10517 a/b) mounted in a sealed brass holder. A pre-sample collimation limited the beam size to about 2.54-cm on the samples and allowed only neutrons from the water moderator part of the neutron source to be used. The neutron detector was an 11.1-cm diameter, 1.25-cm thick ⁶Li-glass scintillator viewed on edge by two 12.7-cm diameter photomultipliers and positioned in the beam at 79.815 meters from the neutron source. The samples were cycled periodically. Additional measurements with corresponding "compensators" in the open beam and with a thick polyethylene sample were used to determine the gamma-ray background from the neutron source.

3. Conclusion

Comparisons of our recent results for Si, K, Cl, and Al (Figures 1-4) with the evaluated nuclear data obtained from the ENDF/B-VI or JENDL-3.2 nuclear data bank reveal significant differences in the neutron capture cross sections. In many cases the capture widths were severely overestimated or resonances were missed due to large backgrounds.



Fig.1 Neutron capture cross section of natural Si compared to the ENDF/B-VI evaluation. Due to an undercorrected neutron sensitivity background the capture cross section were overestimated in the previous experiments on which the evaluation is based.



Fig.2 Neutron capture cross section of natural Potassium compared to the ENDF/B-VI evaluation.



Fig.3 Neutron capture cross section of natural Cl compared to the ENDF/B-VI evaluation.



Fig.4: Neutron capture cross section of Al compared to the JENDL-3.2 evaluation.

These discrepancies have two reasons (i) the use of improper weighting functions and (ii) the underestimated neutron sensitivity of the experimental setup. In order to reduce the neutron sensitivity the experimental setup was redesigned, i.e. the surrounding structural material was reduced by removing the massive Al-sample changer and beam pipe which was replaced by a thin carbon fiber tube. Also, the massive detector housings were removed and the two C₆F₆ scintillation detectors were replaced by less neutron sensitive C₆D₆ detectors with reduced mass detector mounts. For the determination of the weighting function the more sophisticated computer code EGS4 was used. For all structural materials within 30 cm of the detectors including the sample to be investigate, the electron and γ -ray interaction are to be included in the input of the EGS4 code [4]. The code was used to calculate the response functions of the detector for various monoenergetic γ -rays. The resulting pulse-height spectra were then broadened using a resolution function. The final weighting function was calculated from these broadened spectra using a least square fitting code. More details about these improvements can be found in [5, 6] where the impact of the neutron sensitivity was impressively demonstrated in a high-resolution TOF measurement for ⁸⁸Sr. In this case, the two prominent resonances at 289 and 325 keV with neutron widths $g\Gamma_n = 24,932$ and 22,082 eV, respectively, a reduction of capture widths by an average factor of five was reported.

The neutron total and capture cross section data were analyzed with the computer code SAMMY [7], applying all the corrections for the experimental effects. The resonance parameters obtained are the bases for an evaluation of the cross section performed by the ORNL Nuclear Data group. The final result was then checked for consistencies using criticality benchmark calculations. As an example the ORNL evaluation for Al is shown in Figure 5 which clearly demonstrates that the two evaluated files from ENDF/B-VI and JENDL-3.2 are inadequate.



Fig.5: ORNL evaluation of the neutron capture cross section for Al compared to the JENDL-3.2 and ENDF/B-VI evaluation.

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References

- 1) R. L. Macklin and B. J. Allen, Nucl. Instrum. Methods, 91, 565 (1971).
- 2) W. R. Nelson, H. Hirayama and D.W.O. Rogers, The EGS4 Code System, Stanford Linear Accelerator Center Report SLAC-265 (Stanford, Ca) (1985).
- 3) R. L. Macklin, J. Halperin, and R. R. Winters, Nucl. Instrum. Methods, 164, 213 (1979).
- F. G. Perey et al., Proc. Int. Conf. on Nuclear Data for Science and Technology, Mito, Japan, May 30-June 3, 1988, 379 (1988).
- 5) P. E. Koehler et al., Phys. Rev. C 54, 1463 (1996)
- 6) P. E. Koehler, et al., Phys. Rev. C 62, 055803 (2000).
- N. M. Larson, Technical Report No. ORNL/TM-9179/R6, Oak Ridge National Laboratory, 2003.