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Impact of new results of the neutron capture cross section measurements for odd gadolinium isotopes on thermal-spectrum systems

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Summary. — Light Water Reactors (LWRs) are frequently equipped with fuel pins in which UO_2 is mixed with Gd_2O_3. Gd odd isotopes have extremely high neutron capture cross sections at very low energies and are currently used as burnable poisons. For this reason, ENEA put forward a research proposal for an improvement of the Gd nuclear data accuracy by means of new experiments to be done in the framework of the n_TOF Collaboration. In 2016, new measurements were performed at the CERN, and subsequently ENEA in collaboration with IRSN, started to re-evaluate the neutron capture cross sections (XSs) of Gd odd isotopes. This paper presents the results of Monte Carlo simulations performed with the new measured data to estimate their impact on the criticality of a thermal-spectrum benchmark for which the value of k_{eff} is known. The outcomes demonstrate that the new data can produce a k_{eff} which is closer to the experimental one than that obtained using the currently available Gd evaluations.

1. – Scientific background

Capture XS of the Gd odd isotopes, despite their reactor safety-related importance, also stated by OECD/NEA that has included ^{155}Gd, ^{157}Gd in the High Priority Request List [1] have not been so widely studied up to now and are presently known with an accuracy not fully adequate to the needs of the present-day nuclear industry. For example, the ENDF/B-VII.1 Gd odd isotope XSs are associated with rather high ($5 \div 10\%$)

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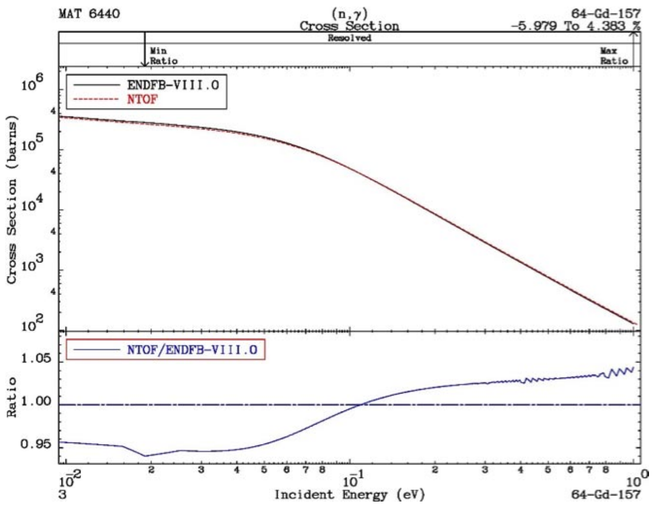

Fig. 1. – New n_TOF ^{157}Gd(n,γ) cross section and comparison with ENDF/B-VIII.0.

uncertainty values in the thermal range. Moreover, XSs experimental values reported in the scientific literature show sensitive deviations ($-16 \div +4\%$) once compared to the ENDF/B-VII nominal value [2]. In 2012, MCNP6 has been used to perform an extensive analysis of ENDF/B-VII.1, JENDL-4.0 and JEFF-3.1.1 evaluated data over several hundred reference cases of the International Criticality Safety Benchmark Evaluation Project (ICSBEP). Results have shown that Gd isotopes evaluations are not sufficiently adequate to represent experimental data from ICSBEP, including uncertainties [3]. Moreover, the use of a specially corrected ENDF/B-VII data library, which included experimental data on Gd by [4], has produced a worsening in the experimental effective multiplication factor (k_{eff}) results with respect to the reference experimental values for almost all the ICSBEP benchmarks considered. For a deep understanding of the role of ^{155}Gd, ^{157}Gd in nuclear fuels, ENEA performed a Sensitivity and Uncertainty analysis (S/U) on k_{eff} for several different Boiling Water Reactors (BWR) and Pressurized Water Reactors (PWR) Fuel Assemblies (FAs) at Beginning of Life (BOL) and in Hot-Full Power Conditions (HFP) using ORNL SCALE 6.1 code package [5,6]. Results revealed that Gd odd isotopes give the largest contribution on k_{eff} uncertainty after ^{235}U and ^{238}U.

2. – Experimental campaign

In 2015, the n_TOF collaboration decided to carry out new Gd odd isotopes (n,γ) cross section measurements based on scientific justification suggested by ENEA. These have been performed at CERN n_TOF facility in 2016 using the time-of-flight method [7]. Gd samples used in the experimental campaign were acquired in form of isotopically "quasi-pure" (i.e., ^{157}Gd and ^{155}Gd 91.75% enriched) self-sustaining metallic discs from Oak Ridge National Laboratory. After data analysis, preliminary capture cross sections at 0.0253 eV have been preliminary estimated to be 62.2 ± 2.2 kb for ^{155}Gd and 239.8 ± 9.3 kb for ^{157}Gd with an uncertainty of about 3.9% and up to 6% relative difference with respect to those reported in data libraries [8]. Figure 1 shows the new n_TOF ^{157}Gd XS compared with the ENDF/B-VII.0 evaluation [9]. The ratio between n_TOF data and

Fig. 2. – ZED-2 k_{eff} sensitivity per unit lethargy to ^{157}Gd(n,γ).

ENDF-VII.0 evaluation has been estimated to be, on average, about −3.5% in the 0.01 to 0.1 eV energy range, and about 2.5% in the 0.1 to 1.0 eV energy range.

3. – Impact of results of new measurements on thermal-spectrum systems

To assess the effect on reactivity of the new ^{157}Gd cross section, an MCNP6 full-core simulation of a reference thermal-spectrum experimental facility has been performed. The system considered is the ZED-2 research reactor at the Chalk River Laboratories (AECL) for which the bias of the reactivity has already been investigated [10]. The latter showed that the Gadolinium (Gd) capture cross sections are overestimated in the ENDF/B-VII.0 data library, and underestimated in ENDF/B-VII.1b, in which the standard Gd odd isotopes evaluations were replaced by data from [4]. The ENDF/B-VII overestimation is such that, for benchmark Case 2 (see below), the ideal compensating criticality gain necessary to attain the ZED-2 experimental criticality factor amounts to about +22 pcm ^{157}Gd capture XS at the thermal point is 254 kb in the ENDF/B-VII evaluation and 226 kb in the ENDF/B-VII.1 modified version, which uses data by [4]. Consequently, assuming the predominance of thermal neutrons in the behavior of the system, it can be inferred that real ^{157}Gd thermal capture XS should be between the two extreme values of 226 and 254 kb. Therefore, the new measured data is certainly a good estimate of the ^{157}Gd capture XS at the thermal point. MCNP6 has also been used to perform a sensitivity analysis on ZED-2 for different Gd concentrations diluted in the reactor moderator (i.e., Case 2: 0.5 ppm, Case 3: 1 ppm, Case 4: 1.5 ppm) as reported in Figure 2. Simulations have shown that the system is sensitive to ^{157}Gd mainly in the thermal region (i.e., $0.01 \div 0.1 eV$). The value of the sensitivity integral for Case 2 was found to be roughly -8.4×10^{-3}. An analogous sensitivity analysis has been performed for evaluating the sensitivity of k_{eff} on the Gd impurities in the ZED-2 graphite reflector: this was found to be negligible. A preliminary assessment of the new k_{eff} value, which would be achieved from the new ^{157}Gd data, has been done by evaluating the criticality gain on the ZED-2 system with respect to the k_{eff} obtainable by the ENDF/B-VII data. Eq. 1 − in which k is the calculated nominal value obtained using ENDF/B-VII data, S is the k_{eff} sensitivity integral value for ^{157}Gd capture XS, and $\Delta\sigma/\sigma$ is the relative difference of the ^{157}Gd XSs (i.e., with respect to ENDF/B-VII data) − has been used to perform the gain calculation.

$$\Delta k \approx kS\Delta\sigma/\sigma \quad (1)$$

Eq. 1 has been used adopting a 1-group theory here applied in an approximated mode assuming that the contribution to criticality is only due to the neutrons in the thermal energy range. Thereby, the contribution of the relative difference of the ^{157}Gd XS has been considered only with respect to the thermal zone (i.e., $0.0 \div 0.1$ eV). The obtained results can be considered a good estimate of the criticality gain for a system like ZED-2, in which the energy region outside the thermal range can be neglected (see Fig. 2). The difference in k_{eff} results to be about +29.2 pcm (adopted values: $k = 0.99766$, $S = -8.36E-03$, $\Delta\sigma/\sigma \approx -0.035$), only +7.2 pcm higher than the ideal compensating gain (i.e., +22 pcm, see above) necessary to compensate the effect of the wrong ENDF/B-VII data. In a more precise 2-groups (i.e., $0.0\div0.1$ and $0.1\div2.0\times10^7$ eV) framework, the correction of the k_{eff} value has been estimated similarly and found to be roughly equal to +28.1 pcm, 1.1 pcm closer to the optimal value than by using a 1-group approach.

4. – Conclusions

A preliminary analysis of the effect of the new ^{157}Gd capture XS on reference benchmarks at the ZED-2 research reactor revealed that the new XS has the potential to reproduce experiments much better than currently available evaluations. The new XS seems still to underestimate slightly capture by about 0.8% in the thermal range. MCNP calculations with a continuous-energy approach are needed to improve confidence on the performance of the new XS for ZED-2. For this reason, full MCNP6 calculations with the new Gd odd isotopes capture data will be accomplished soon for a more precise assessment. In the future, many other ICSBEP benchmarks will be used for further validation of the new product. New experiments with the same Gd samples used to produce the present capture XS are currently underway at the JRC GELINA facility in order to try to further reduce the uncertainty in the thermal energy region. Once fully validated, the n_TOF data could be used to produce new evaluations for the future JEFF4 evaluated data library.

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REFERENCES

[1] HPRL https://www.oecd-nea.org/dbdata/hprl/.
[2] ROCCHI F. *et al.*, *Eur. Phys. J. N*, **3** (2017) 21.
[3] VAN DER MARCK S. C., *Nuclear Data Sheets*, **113** (2012) 2935.
[4] LEINWEBER G. *et al.*, *Nucl. Sci & Eng.*, **3** (2006) 261.
[5] ROCCHI F. *et al.*, ENEA-ADPFISS-LP1-083 (2016).
[6] ORNL-TM-2005/39, Ver. 6.1 (2011).
[7] CASTELLUCCIO D. M. *et al.*, ENEA -ADPFISS-LP1-100 (2017).
[8] MASTROMARCO M. *et al.*, preprint arXiv:1805.04149 (2018).
[9] ROCCHI F. *et al.*, in Proceedings of the Fourth International Conference on Physics and Technology of Reactors and Applications, Marrakech, 17-19 September, Morocco (2018).
[10] CHOW J. C. *et al.*, *AECL Nuclear Review*, **1** (2012) 21.