Neutron induced fission of ^{237}Np – status, challenges and opportunities

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Abstract. Nowadays, there is an increased interest in a complete study of the neutron-induced fission of ^{237}Np. This is due to the need of accurate and reliable nuclear data for nuclear science and technology. ^{237}Np is generated (and accumulated) in the nuclear reactor core during reactor operation. As one of the most abundant long-lived isotopes in spent fuel ("waste"), the incineration of ^{237}Np becomes an important issue. One scenario for burning of ^{237}Np and other radio-toxic minor actinides suggests they are to be mixed into the fuel of future fast-neutron reactors, employing the so-called transmutation and partitioning technology. For testing present fission models, which are at the basis of new generation nuclear reactor developments, highly accurate and detailed neutron-induced nuclear reaction data is needed. However, the EXFOR nuclear database for ^{237}Np on neutron-induced capture cross-section, σ_γ, and fission cross-section, σ_f, as well as on the characteristics of capture and fission resonance parameters (Γ_γ, Γ_f, $\sigma_\mathrm{o}\Gamma_\mathrm{f}$, fragments mass-energy yield distributions, multiplicities of neutrons ν_n and γ-rays ν_γ), has not been updated for decades.

1 Introduction

For basic (fundamental) and applied nuclear science, ^{237}Np continue to be amongst the most important and interesting isotopes (minor actinides) for experimental investigation with neutrons of different energies. This is because, as it is a long-lived ($T_{1/2} = 2.144 \times 10^6$ years) isotope, which is present in spent fuel of light-water nuclear reactors, it contributes significantly to the long-term radio-toxicity of nuclear waste. For its successful incineration (transmutation, $\sigma_\mathrm{f} \sim 0.01$–$10$ barn), neutrons of different energies can be used [1–7]. In this case, neutron-induced reaction cross sections (particularly for fission) should be known with high precision for a broad neutron energy range (from thermal to fast) for estimating the effectiveness of the transmutation process.

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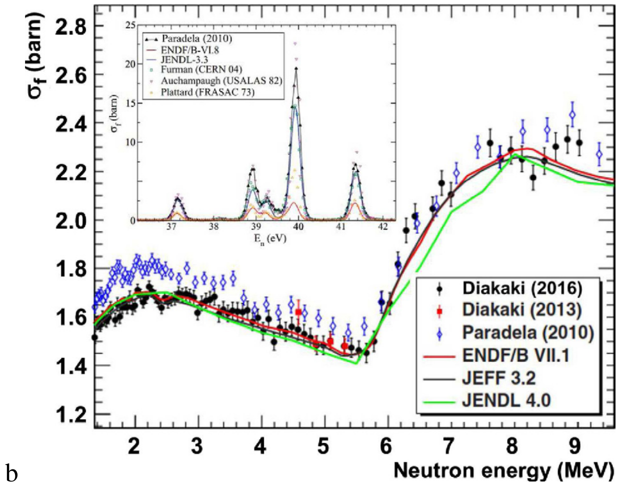

Figure 1. ^{237}Np(n,f) cross-section in the vicinity of the first resonance cluster around E_n = 40 eV (Fig. 6 [12]) and in the neutron energy range between 1.5 MeV and 9 MeV (Fig. 7 [14]); the depicted data are from different experiments, as well as from different evaluated nuclear data libraries (see the references in [12–14]).

According to one of the contemplated scenarios, a high-flux thermal reactor ($\sigma_f^{th} \sim 20$ mb) can be used as a transmutation device [7], because the majority of the operating commercial reactors in the world are based on a thermal spectrum.

2 Neutron-induced fission cross section of ^{237}Np

As ^{237}Np is a major component of spent nuclear fuel, the accurate knowledge of the capture and fission cross-sections is needed for studies of waste transmutation and advanced nuclear facilities (reactors, ADS, etc.). However, significant discrepancies exist in neutron-induced reaction experimental data repositories and in the present evaluated data libraries, based on the different sets of experimental data (see Fig. 1 and Ref. [8–14]). Similar discrepancy between different data libraries exists for fission-resonance parameters below the fission threshold [10]. Although some data sets for fast-neutron induced fission are consistent within the claimed accuracy, usually about 3–4%, some significant discrepancies show up between more recent data, when compared to several previous measurements. For example, several experiments based on the time-of-flight (TOF) technique were dedicated to the measurement of fission cross section data in a wide neutron energy range (0.7 eV–1 GeV), using different fission-fragment detectors [12–14].

While being in good agreement with previous measurements and evaluated data files at energies below and around the fission threshold, the data exhibit an excess of about 6–7% above E_n = 1 MeV [8–13]. The more recent measurements [14] were not able completelly to resolve the situation. That is why, to study the ^{237}Np(n,f) and ^{238}U(n,f) cross sections in a comprehensive manner, two experiments were performed at two different Van de Graaf (VDG) facilities: IRMM and NPL. Analysis of both experiments is in progress. Some preliminary results were reported at the ND2016 Conference on Nuclear Data for Science and Technology [15].

Very recently, two simultaneous experiments with PPACs (at EAR1) and Micromegas detectors (at EAR2) were conducted at the n_TOF facility to re-measure the ^{237}Np(n,f) cross-section [16], but the results have not published yet.

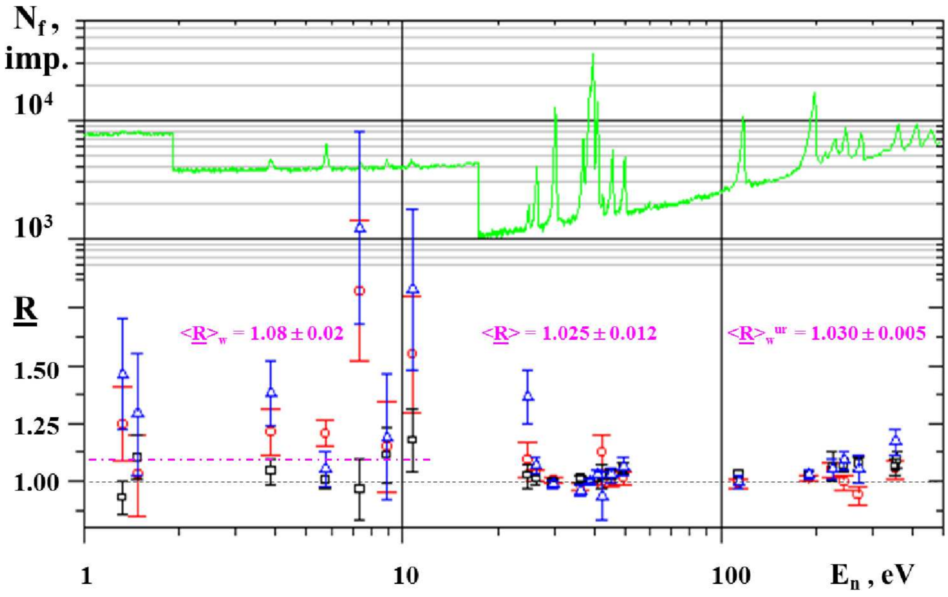

Figure 2. *Upper graph:* fission count rate in neutron energy interval from 1 eV up to 500 eV. The steps indicate the different value used for a single time-channel in the neutron time-of-flight (TOF) fission spectrum; *Lower graph:* variation of the relative (to the strongest resonance around 40 eV) fission γ-ray yield $\underline{\mathrm{R}}$ from resonance to resonance at different thresholds of γ-quanta registration E_γ: ~0.2 MeV (circles), ~0.4 MeV (squares), ~0.6 MeV (triangles). $<\underline{\mathrm{R}}>$, $<\underline{\mathrm{R}}>_\mathrm{w}$ and $<\underline{\mathrm{R}}>_\mathrm{w}^\mathrm{ur}$ are explained in the text.

3 The (n,γf)-reaction

The investigation of the resonance neutron induced fission of ^{237}Np and the (n, γf) reaction provide important information about the compound nucleus (CN) internals:

- the height and structure of fission barriers;
- the transition states to fission and the degree of their damping;
- the strength of the coupling between the collective mode of motion with the internal excitations;
- the influence of the quantum characteristics of the excited states on the properties of the fission fragments;
- the scheme of γ-transitions between highly excited states and fission resonances;
- the connection strength between states with different deformations (classes I and II).

It aims at refining the quality of nuclear data, such as σ_γ and σ_f. The reaction (n, γf) leads to significant changes in Γ_γ, σ_γ, Γ_f and σ_f of the order of 10–15% (for e.g. ^{239}Pu at $E_n = 1$ keV) on σ_t and $\alpha = \sigma_\gamma/\sigma_f$, especially, at low excitation energy region of the CN. On the other hand, the database on fission resonance parameters for ^{237}Np is meagre (only few experimental datasets available in the EXFOR database).

The nature of the coupling between 1st and 2nd class states, as well as the existence and location of class II resonances remains unclear. The existence of very weak resonances in the sub-threshold fission of ^{237}Np, with $\Gamma_\mathrm{f} \sim 1$ μeV and, consequently, relatively long lifetimes of excited states in the ^{238}Np* compound nucleus (t∼1 ns) suggest that the probability of emission of a pre-fission g-quantum might be larger than the probability for this reaction to occur in the fission resonances of ^{235}U and ^{239}Pu. A detailed review on the investigation of the ^{235}U(n,γf) and ^{239}Pu(n,γf) until 1990 is given in reference [18].

Until now, the only experiment on the investigation of fluctuations of the γ-ray yield from resonance to resonance in ^{237}Np(n,f) (Fig. 2, [18–20]) shows that this fluctuation might be partly explained assuming the existence of a ^{237}Np(n, γf) reaction channel. In Fig. 2, $< \underline{\mathrm{R}} >$ is the average relative γ-ray yield from the resonances in the first cluster around 40 eV, $< \underline{\mathrm{R}} >_{\mathrm{w}}$ is the weighted average relative value from the resonances below 10 eV and $< \underline{\mathrm{R}} >_{\mathrm{w}}^{\mathrm{ur}}$ is the weighted average relative value from the unresolved resonance above 100 eV.

4 Conclusion and outlook

The EXFOR nuclear database for the ^{237}Np(n,f) reaction does not provide experimental data with uncertainties appropriate for nuclear reactor applications. This is true for all of the available incident neutron energy regions. The data from the sole experiment on the fluctuation of fission γ-ray yields from resonance to resonance does not exclude a (n,γf) reaction channel in the sub-threshold fission of ^{237}Np [20]. New experiments can be conducted, e.g. at the high-resolution neutron time-of-flight facility GELINA [24], with a focus on measuring $< \nu_{\mathrm{n}} >$ and $< \nu_{\gamma} >$, prompt n/γ-ray competition, correlations r(<TKE>, Γ_{f}^{-1}) [21], n- and γ- yields as a function of resonance neutron energy. The assessment of those quantities in correlation with fission fragment properties (A, TKE), i.e. fission fragment mass-dependent γ- and n-emission is recommended but requires a highly efficient experimental set-up.

To minimize the influence of the statistical and systematic uncertainties, new experiments need to be conducted on the different existing resonance-neutron TOF facilities [22–24], using fission fragment and multi n-γ detector spectrometers.

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