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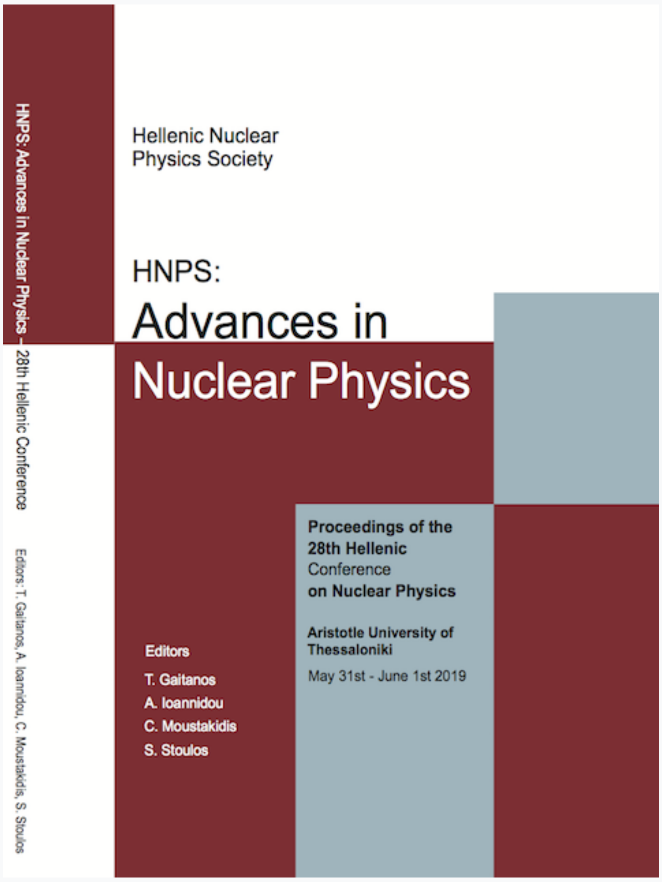

237Np reactions with fast neutrons: a phenomenological study

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^{237}Np reactions with fast neutrons: a phenomenological study

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Abstract *Neptunium presents various opportunities as nuclear fuel, especially in deep–space mission power generators. As it is part of the nuclear spent fuel in PWR, waste management concerns due to ^{237}Np long α-emitting half-life have attracted some attention recently. The scarcity of experimental data in the fast neutron energy range highlights the necessity to investigate the radiative neutron capture and neutron–induced fission cross sections of this radioisotope. In the present work, statistical modeling of these reactions is performed using TALYS in an extended range of neutron energies between 10 keV and 20 MeV. In total, 72 different combinations of code parameters were selected to study the level density and γ-strength function dependence of the cross section in ^{238}Np. Preequilibrium and compound nucleus formation phenomena are also examined. Theoretical calculations are compared to available experimental total cross section data found in literature in an attempt to investigate any discrepancies between experiment and theory and validate statistical uncertainties.*

Keywords *^{237}Np, fast neutron, cross section, TALYS*

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INTRODUCTION

Neptunium presents various opportunities as nuclear fuel, especially in deep–space mission power generators. Neutron activation of ^{237}Np leads to the production of ^{238}Np which can subsequently produce ^{238}Pu via β–decay:

$$^{237}Np + n \rightarrow {}^{238}Np + \gamma \xrightarrow{\beta-} {}^{238}Pu$$

The latter has a half–life of 87.7 a, which is considered sufficient for space missions inside the solar system. Furthermore, ^{237}Np is part of the nuclear spent fuel of PWR, raising waste management concerns due to its long half-life as an α-emitter. In addition, Gen–IV nuclear power reactors are designed to operate in the fast–neutron energy spectrum, and ^{237}Np has been proposed as a potential fuel candidate.

The scarcity of experimental data [1] in the fast neutron energy range highlights the necessity to investigate the radiative neutron capture and neutron–induced fission cross sections of this isotope. Radiative capture reactions of neutrons are largely unexplored for several nuclei around uranium, while the need for more detailed and evaluated neutron–induced fission data has been highlighted in recent years.

In the present work, theoretical estimation of cross sections of radiative neutron–capture reactions is performed using the Hauser–Feshbach statistical model.

METHODOLOGY

For the theoretical calculations, TALYS [2] was employed in an extended range of neutron energies between 10 keV and 20 MeV. The default options offered by TALYS for the Optical Model

Potential (OMP), the nuclear level densities (NLD) and the gamma–strength functions (γSF) can be combined to a total of 96 different combinations. In the present calculation, a preliminary, gross-energy step, calculation was performed at the beginning. Some combinations resulted in indistinguishable results, therefore they were dropped from the subsequent, more detailed, calculation (energy step was selected as ~1% of the E value).

Table 1 *Various options of OMP, NLD and γSF offered by TALYS that were employed in the present study. In total 72 combinations were used to provide results in energies 10 keV–20 MeV.*

OMP	NLD	γ-SF
• Global OMP (Koning & Delaroche) • Local OMP (Koning & Delaroche) • Semi-microscopic optical model (JLM)	•Constant Temperature & Fermi Gas Model •Back-shifted Fermi Gas Model •Generalized Superfluid Model •Microscopic LD (Skyrme) Goriely's tables •Microscopic LD (Skyrme) Hilaire's tables •Microscopic LD (TD HFB,Gogny) Hilaire's tables	•Kopecky-Uhl generalised Lorentzian •Brink-Axel Lorentzian •Hartree-Fock BCS tables •Hartree-Fock-Bogolyubov tables •Goriely's hybrid model •Goriely T-dependent HFB •T-dependent RMF •Gogny D1M HFB+QRPA

In total 72 combinations of code parameters (Table 1) were selected [3–5] to study the level density and γ-strength function dependence of the cross section in ^{238}Np. Preequilibrium and compound nucleus formation phenomena are also examined in addition to radiative neutron capture reactions. Theoretical calculations have been compared to all available experimental total cross section data found in literature [1,6] in an attempt to investigate any discrepancies between experiment and theory and validate statistical uncertainties. Table 1 summarizes the various choices of models employed in our study.

RESULTS AND DISCUSSION

Results from the reported calculations can be summarized in the following figures (Figs. 1-4). Cross sections have been calculated for all 72 combinations. All calculations for the neutron-capture radiative reactions show an excellent agreement within each other and with the experimental data at the lower energy range of the present study, up to ≈100 keV. This can be attributed to two reasons: first, experimental data in the lower neutron energies have been available since the dawn of neutron science; second, OMPs which essentially define the behavior of the cross section at low energies have been fine tuned over the years to provide reliable descriptions of the experimental data. The latter is not true for the higher energies, therefore large deviations can be observed in the calculations both from the experimental data and each other.

The complete absence of data for the (n,γ) reaction channel for energies above ≈3 MeV is mostly responsible for the large deviations observed. A shaded are in Figs. 1-4 shows the range of calculated cross sections defined by the minimum and maximum of each of the 72 combinations used at each particular energy value used in the calculation. The shaded area becomes wider and wider as the energy increases. Each of the 72 combinations have been examined in comparison with the others

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visually for the quality of data representation. Although this simple procedure can determine combinations that provide prominently poor agreement with the data, it can not be considered safe to decide on the "best" choice. For that purpose, an additional criterion was introduced. The "best" choice was selected by the best agreement between calculation and experimental cross sections, *simultaneously* for the following reaction channels: total, elastic, (n,γ), (n,2n) and induced fission (see Fig. 1). In the end, a "best" combination of OMP, NLD and γSF is reported here:

OMP: Global Koning–Delaroche
NLD: Constant Temperature & Fermi Gas Model
γSF: Kopecky–Uhl

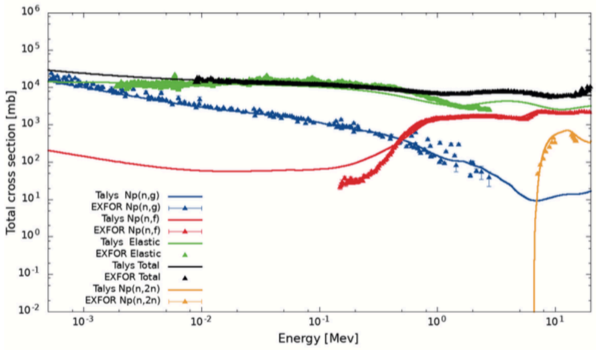

Figure 1 *The results of the calculations simultaneously describing best all neutrons reactions with ^{237}Np, using the Koning–Delaroche OMP, the constant temperature model NLD and the Kopecky–Uhl γSF.*

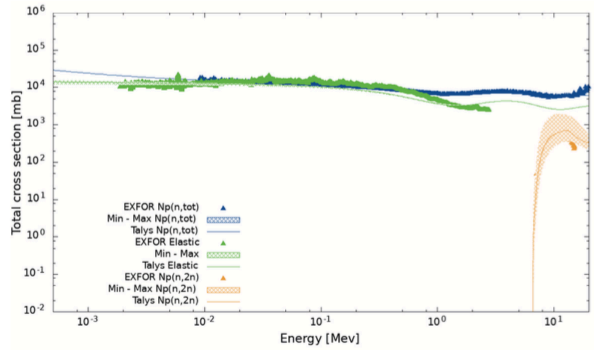

Figure 2 *Calculations for the elastic, total and (n,2n) reactions compared to available experimental data.*

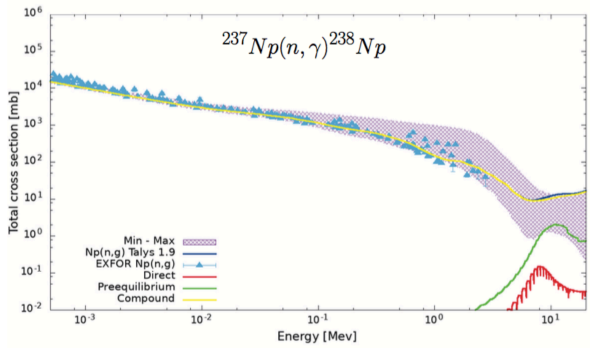

Figure 3 *The range of all calculations performed for the ^{237}Np(n,γ) reaction is shown with the shaded area. Experimental data found in [1] are also shown.*

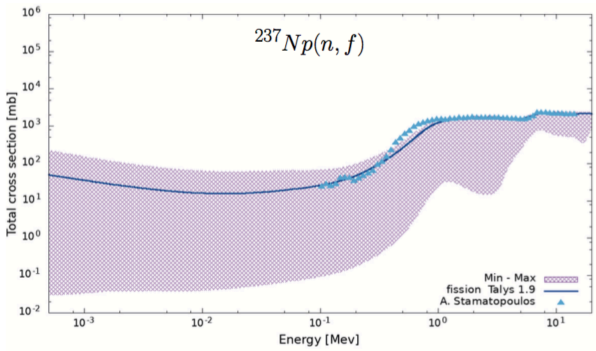

Figure 4 *As in Fig. 3, but for the (n,f) reaction channel.*

For the "best" combination, the neutron–induced fission in the fast neutron spectrum has been calculated and compared to recently reported experimental data by Stamatopoulos et al. [7]. Despite some small deviation from the experimental values in ≈0.3–0.8 MeV, the calculation agrees rather well in trend and magnitude (Fig. 4) up to 20 MeV. It is important to stress that the large spread of the shaded area (as discussed earlier) exceeds four order of magnitudes. Hence it seems to underpin the need for evaluation of the available experimental data and the optimization of the parameters used in TALYS. Fission requires a "double hump" potential for a reliable description. This is not the case for the OMPs listed in Table 1.

CONCLUSIONS

A detailed calculation with TALYS was performed to provide theoretical values for cross sections in reactions of neutrons with the minor actinide ^{237}Np. The focus was on the radiative capture reaction channel. From the 72 different default combinations available in TALYS, a “best” choice of OMP+NLD+γSF was determined and used to provide predictions for cross sections in the fast spectrum. Further work is required to provide an objective determination of that “best” choice, based on residuals and fine-tuned parameters in the models employed in the present study. In any case, the results highlight the need for more experimental area, as well as further theoretical work, especially in the case of neutron–induced fission of ^{237}Np.

Acknowledgements

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