THE INVESTIGATION OF THE TOTAL NEUTRON CROSS SECTION OF ^{237}NP

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Abstract

Within the framework of a collaboration between the Institute for Reference Materials and Measurements (IRMM) and the Commissariat à l'Énergie Atomique (CEA), a project has been started to study neutron cross sections for nuclear waste transmutation purposes. We describe the measurements of the total neutron cross section of ^{237}Np, performed at the pulsed white neutron source GELINA of the IRMM. We used the time of flight method to obtain neutron spectra in the energy range from 0.3 eV to 2 keV. A preliminary list of new resonances under 120 eV is presented here. Besides these measurements, the Doppler effect has also been investigated for NpO_2. We will show how the use of an harmonic crystal model for Doppler broadening can explain some unexpected effects in the fit of the resonances.

Introduction

The nucleus ^{237}Np with a half-life of 2.144 .10^{6} years is one of the most important minor actinides produced in conventional nuclear power plants: about 4 tons each year world-wide. After neutron capture, it becomes ^{238}Np which has a thermal fission cross section of 2000 b, much higher than the 0.02 b of the initial ^{237}Np, making it a good candidate for transmutation by neutron induced fission in thermal reactors.

Several measurements of the resonance parameters have been performed in the past [1,2,3,4] and several evaluations exist [5]. Nevertheless there is a strong demand for more precise resonance parameters [6].

Due to several large resonances at very low energy (starting at 0.49 eV), ^{237}Np is also a good candidate for Doppler effect studies.

Experimental set-up

The total cross section measurements of ^{237}Np were performed at the Geel Linear Electron Accelerator GELINA, using the time-of-flight technique. Three NpO_2 samples, with thicknesses 0.2, 1.0 and 2.0 g/cm^2 of ^{237}Np, have been used for the transmission measurements. For the two thinnest samples, the experiment covers the energy range from 0.3 eV to 40 eV at three different temperatures (15K, 50K and 290K) in order to investigate the Doppler effect. The neutron flight distance was 26.45 m. GELINA was providing a 15 ns electron bursts of 100 MeV average energy, a repetition frequency of 100 Hz and an average beam current of 12 μA. With a similar set-up, but at 49.33 m, we measured the transmission of the thickest sample covering the energy range from 0.3 to 120 eV at 290 K. The same sample was also used for the higher energy range from 45 eV to 2.6 keV at room temperature (290 K), but with GELINA running at 800 Hz, 70 μA and with a 2 ns pulse width.

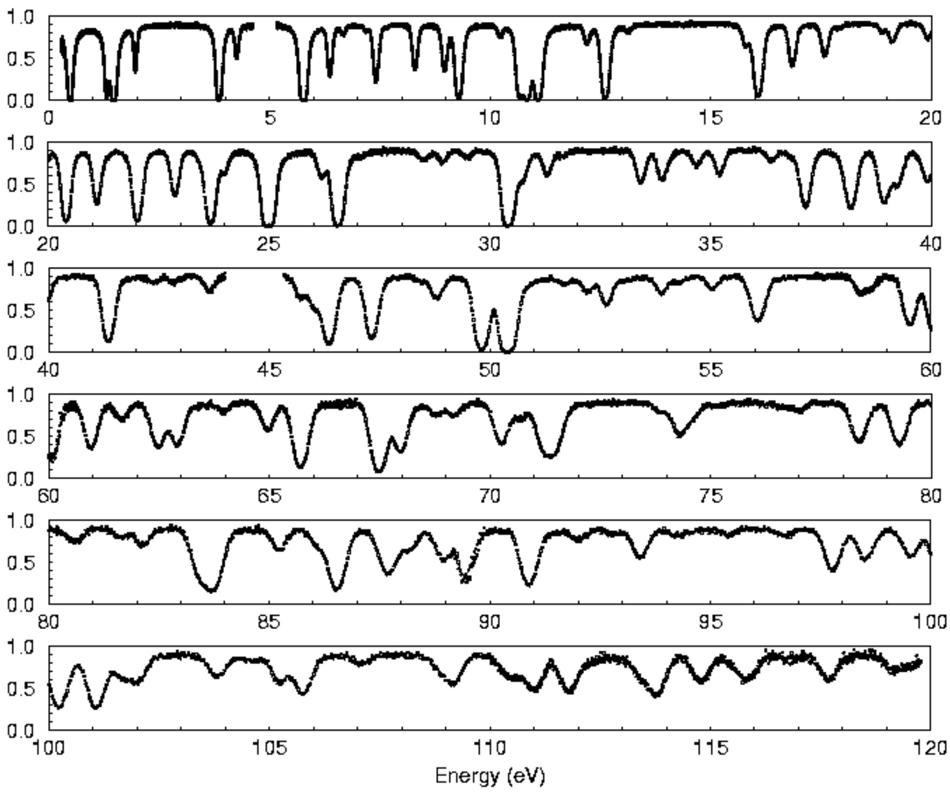
The neutrons, produced via bremsstrahlung by the electron beam hitting a rotating uranium target, were moderated by water canned in two beryllium containers (4 cm thick). The neutrons enter into the flight paths through evacuated aluminium pipes of 50 cm diameter and are confined by collimators consisting of borated wax, lead and copper. In order to absorb slow neutrons, that otherwise overlap with the following GELINA pulse, a filter of cadmium (for experiments at 100 Hz) or a filter of ^{10}B (for the experiment at 800 Hz) was placed in the neutron beam.

The NpO_2 sample was mounted in an automatic sample changer at 12 m (for the experiments with the two thinnest samples) and at 23 m (for the thick sample). To determine the background, filters absorbing neutrons at specific energies ("black resonances") were moved in and out of the beam with a sample changer controlled by the data acquisition system. In order to reduce systematic errors, these measurements sequences were recorded sequentially within a cycle lasting about one hour in total. The neutrons passing through the sample were further collimated and then detected by a NE912 lithium-glass detector. The signals from $^{6}Li(n,\alpha)$ reaction were processed to provide the neutron time-of-flight and then stored by the data acquisition system FAST, upgraded at IRMM [7].

Analysis and results

The raw time-of-flight spectra were corrected for deadtime and background. The transmission was calculated as the ratio of these corrected spectra and normalised to the ratio of the time-integrated incoming neutron fluxes of the sample «in» and «out», measured by neutron monitors located in the target hall. The in-house developed data processing package AGS [8,9] was used to carry out the various spectrum manipulations. A part of the transmission spectrum is shown in Figure 1.

Figure 1. **Transmission data of the 2 g/cm^2 thick NpO_2 sample measured at 49.33 m and at 290 K vs the neutron energy**



The R-matrix shape fitting program REFIT [10] was used in order to determine the resonance parameters like the resonance energy E_0, the neutron width Γ_n and the radiative width Γ_γ, which includes in our approximation the small fission width.

The given values of Γ_γ and Γ_n are still preliminary and demand further investigations. We find a good agreement between our Γ_n values and those of Auchampaugh [4] for the thinnest samples, but we have a systematic discrepancy of about 3% with the thick sample. This may come from an error on the quantity of neptunium. Therefore we only give in the following table the energies of the resolved resonances below 120 eV, those which are not present in the existing evaluations, all based on the results of D. Paya [1] or L.W. Weston [3]. In the table, our results are compared with the those of Auchampaugh given in the “BNL” [11]. Note that the two underlined resonances are only given in the evaluations and not in the reference 11.

Table 1. **Energies, in eV, of the resonances not given in the existing evaluations and comparison with the reference 11**

BNL	This work	BNL	This work	BNL	This work	BNL	This work
3.05		44.23	**44.29**	66.80	**66.80**	89.94	
7.68	**7.66**	44.95	**44.94**	69.35		91.02	
14.41	**14.38**	48.87	**48.90**	72.30		94.57	**94.53**
15.94	**15.92**	49.23	**49.29**	73.08		94.92	**95.08**
	<u>17.05</u>	50.34		75.66			**<u>102.07</u>**
17.94	**17.93**	56.15	**56.14**	76.21	**76.24**	103.40	
24.78	**24.79**	56.57	**56.55**	77.55	**77.58**	104.05	**103.96**
28.63	**28.61**	56.87	**56.87**	77.83			**106.91**
32.49	**32.49**	57.40		78.50	**78.50**	108.28	
34.07	**34.09**	61.67		79.90		109.84	**109.84**
38.04	**38.04**	62.49	**62.51**	82.39		112.65	**112.64**
39.02	**39.03**	63.42	**63.42**	83.80	**83.84**	113.03	**113.02**
43.20		66.40	**66.40**	87.79	**87.77**		

Doppler effect study

In the code REFIT, the Doppler broadening of the resonances is taken into account by the use of a “high-energy” Gaussian approximation to the free gas model with an effective temperature instead of the thermodynamic temperature.

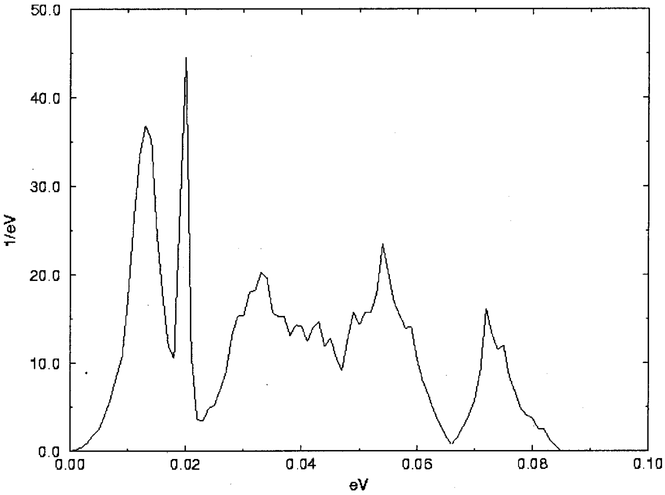
The fit of the transmission data show that REFIT is unable to reproduce exactly the shape of the resonances, mainly at low energy. This effect is highlighted by the shape of the residual (see second spectrum of Figure 3) which are the ratio (fit-data)/σ_{data}.

A second observation is the behaviour of the effective temperature as function of the energy. If we adjust during the fitting procedure the effective temperature, this is lower than the thermodynamic temperature for the resonances at low neutron energy, as shown in Figure 6. Such a result is not allowed in the theory of the free gas model.

To determine if the cause of these disagreements is the description of the Doppler effect, we use the program DOPUSH developed by D. Naberejnev [12,13] which describe the Doppler broadening based on a more realistic model derived from the Lamb’s crystal model [14].

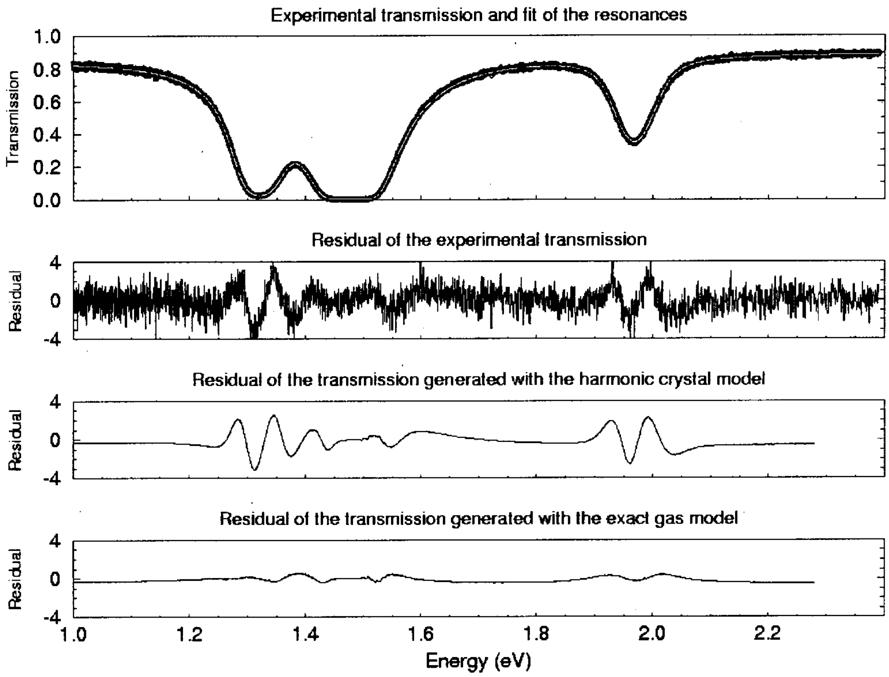
As we are missing information on the phonon spectrum of the NpO_2 crystal, we are using the known phonon spectrum of UO_2 (Figure 2), which has a similar crystal structure and mass.

Figure 2. **The UO_2 crystal phonon spectrum**



With a given set of resonance parameters E_0, Γ_n, Γ_γ, two Doppler broadened cross sections are calculated by the DOPUSH programme, one with the harmonic crystal description, the other using the exact free gas model. Then the corresponding transmissions are deduced, for a given flight distance, adopting experimental errors bars. Then REFIT is used to fit these transmissions with its free gas model. In Figure 3, we compare the residuals obtained from the experimental transmission and the two simulated transmissions. As a result, the residual of the data "generated" with the harmonic crystal model show the same behaviour as the residuals of the experimental data. On the other hand, the residuals of the data "generated" with the exact gas model do not produce a significant structure. In this indirect way we could infer that the data "generated" with harmonic crystal model reproduces the experimental data better than with the free gas model.

Figure 3. **Comparison between the residuals calculated by REFIT**



To verify this hypothesis, the following procedure of a "manual fit" has been developed:

For an isolated resonance with known parameters coming from the evaluations, a transmission spectrum is "generated" using DOPUSH with the harmonic crystal model. This spectrum is then fitted by REFIT with its gas model and the resulting parameters are compared to the initial parameters. The resulting differences are then used to correct the REFIT resonance parameters obtained from the analysis of the experimental data. The results are then assumed to be "real" resonance parameters which DOPUSH uses with the harmonic crystal model to generate a new transmission. Figure 4 shows in the residuals that the use of the "real" resonance parameters within the harmonic crystal model of DOPUSH creates a much better representation of the experimental data.

In Figure 5, the description of the resonance shape by REFIT is compared to the "generated" transmission of DOPUSH (with adjusted resonance parameters and harmonic crystal model). The harmonic crystal model seems to be able to reproduce the right shape of the resonances, because of its more accurate description of the Doppler broadening.

Figure 4. **Comparison between the fit by Refit and the simulated fit with the "generated" transmission based on the harmonic crystal model for Doppler broadening**

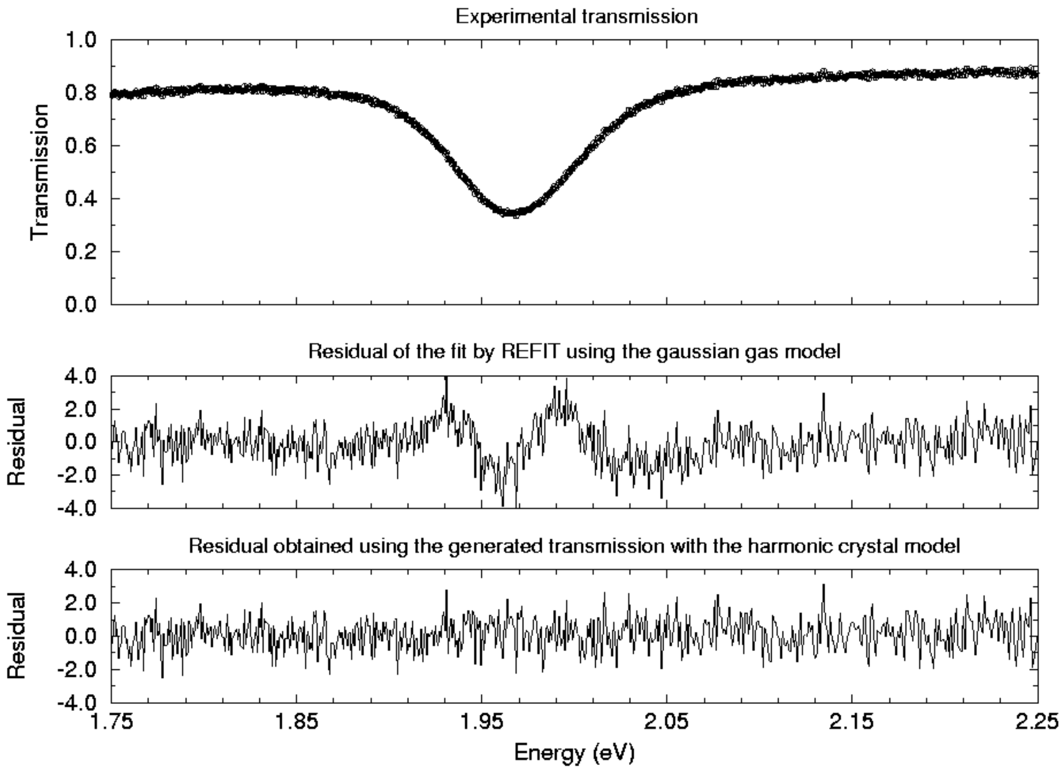
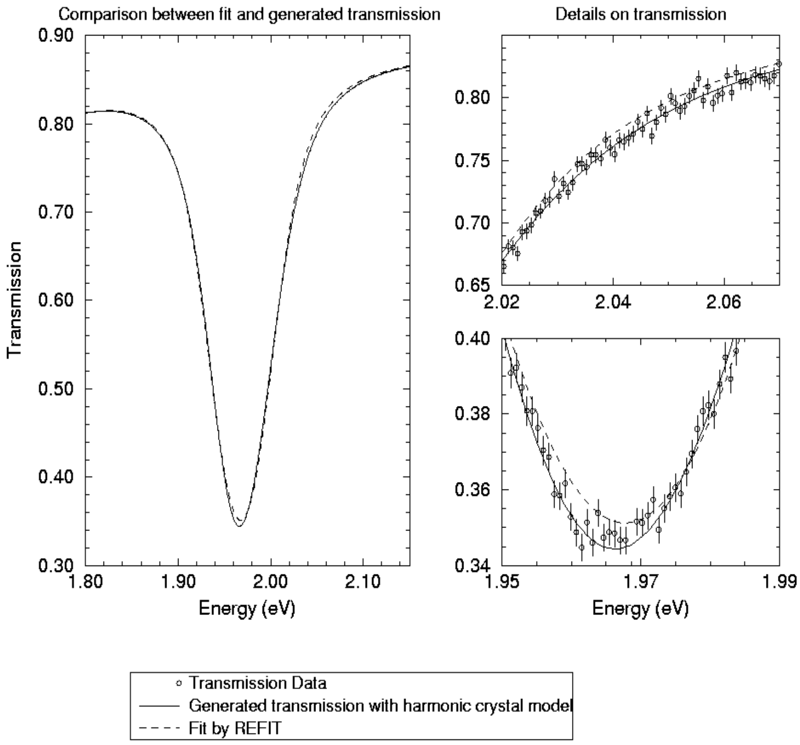


Figure 5. **Comparison between the fit by Refit and the "generated" transmission based on the harmonic crystal model for Doppler broadening with adjusted resonance parameters**



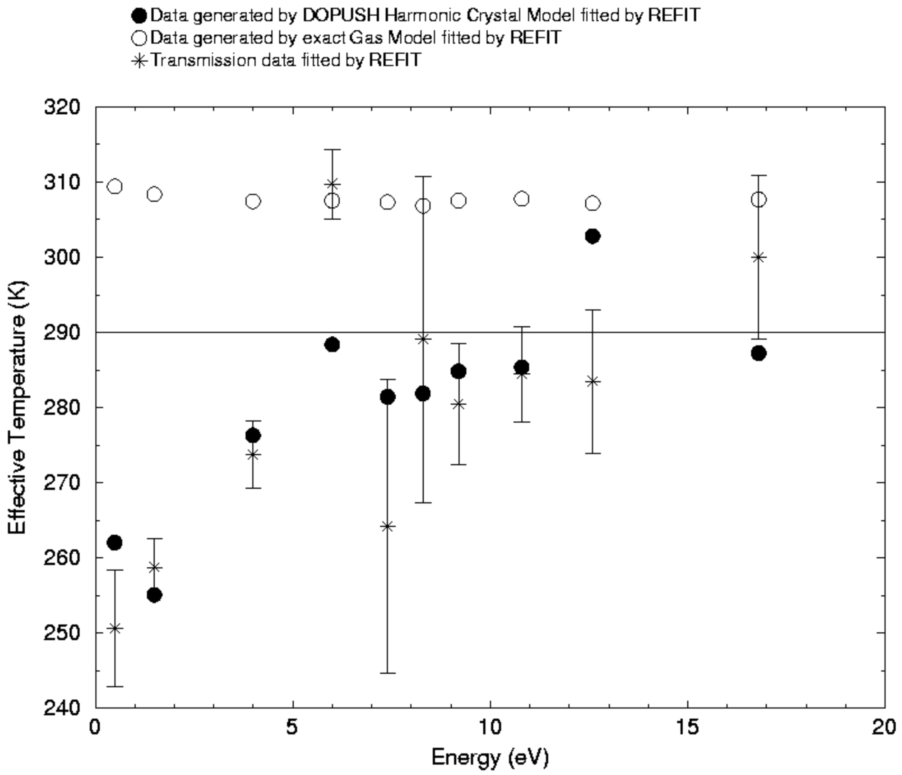
By adjusting the effective temperature on the transmission "generated" with the exact gas model, we observe in Figure 6 that the effective temperature stays constant as expected.

On the other hand, the effective temperature resulting from the transmission data "generated" by DOPUSH with the harmonic crystal model and fitted by REFIT shows the same behaviour as the fit to the experimental data.

We conclude that for molecules like NpO_2 and in the temperature/energy range under investigation, the structures seen in the residuals of the fits and the variation of the effective temperature in function of the neutron energy may be attributed to the the gas model, which is inadequate to describe the physical process of the Doppler broadening of the resonances.

The harmonic crystal model should be implemented in REFIT in order to improve the description of Doppler broadening for chemical compounds as NpO_2.

Figure 6. **Variation of the effective temperature adjusted on different data by Refit for a real temperature of 290 K**



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