Experimental studies of prompt fission neutron energy spectra

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Abstract

Prompt fission neutron spectra were measured in the reactions \(^{238}\text{U}(n,f)\), \(^{235}\text{U}(n,f)\) and \(^{237}\text{Np}(n,f)\) at different incident neutron energies. The neutrons were detected using a coaxial doped \(p\)-terphenyl scintillation detector in coincidence with fission fragments and their time-of-flight was recorded. The properties of the neutron detector were determined and the results are presented in this work. A preliminary neutron detection efficiency was applied to data from the neutron-induced fission of \(^{238}\text{U}\) at \(E_n = 5.2\) MeV, leading to encouraging results.

Keywords: Prompt fission neutrons; time-of-flight; \(^{238}\text{U}(n,f)\); \(p\)-terphenyl; intrinsic neutron efficiency; pulse shape discrimination

1. Introduction

Prompt fission neutron energy spectra (PFNS) play an important role in the criticality and other properties of nuclear systems. However, experimental data for fast neutron-induced fission are scarce and the precision of our current knowledge of these observables is far less good than that of other fission observables, such as neutron multiplicity or fission cross-sections. In order to improve the numerical modeling of the present and future nuclear systems, it is essential to perform new precise measurements.

In connection with the coordinated research program launched by the IAEA (Capote et al., 2009), new experimental investigations on PFNS for fast neutron-induced fission of major actinides have been initiated in Buyères-le-Châtel. Measurements were performed for the fission of \(^{238}\text{U}\), induced by 5.2 and 15 MeV neutrons, and of both \(^{235}\text{U}\) and \(^{237}\text{Np}\), induced by 500 keV neutrons. Coincidences between neutrons and fission fragments were recorded and the kinetic energy of the neutrons determined by employing the time-of-flight technique. Below we report on the detector characterization, describe the experimental details and present first results from the neutron-induced fission of \(^{238}\text{U}\) at \(E_n = 5.2\) MeV.

2. Experimental details

In this section the involved detectors as well as the data acquisition system are presented and the experimental setup is described.
Fig. 1. Light output curve for the para-terphenyl detector used in this work (see text for details). The symbols represent experimental data, while the line corresponds to the result of a fit according to Eq. 1.

2.1. Fission chambers

The actinide targets used in this work are ionization chambers filled with P10 gas (10% methane, 90% argon) and containing several tens of parallel plate electrodes on which the actinides are deposited. In the following, they will be referred to as fission chambers. The voltage difference between two adjacent electrodes was about 300 V. The fission fragments ionize the gas, generating electron-ion pairs. The charge is collected, leading to an electrical signal, signing a fission event. This signal is fed via a preamplifier directly into the data acquisition system, constituting the start for the time-of-flight measurement.

2.2. Para-terphenyl detector

The neutron detector was manufactured by Cryos-Beta and consists of an organic crystal of doped p-terphenyl (C_{18}H_{14}) coupled to a photomultiplier (Photonis XP53X2). The crystal is a cylinder of 75 mm diameter for a 500 mm length and a density of 1.23 g cm\(^{-3}\). It was shown in the literature that this type of detector provides a better energy resolution, higher light-output and neutron efficiency than a NE213-equivalent neutron detector of same size (Lovchikova et al., 2004; Matei et al., 2012). The signal of a neutron (or \(\gamma\)-ray) hitting the detector is fed into the data acquisition system and is used as a stop for the time-of-flight measurement. Our characterization of the scintillation detector gave the following results:

- The energy resolution of the detector was determined using \(\gamma\)-sources in an energy range from 59.5 to 1836 keV. For \(^{137}\)Cs (662 keV) about 10% (FWHM) were obtained, following the expected \(E^{-1/2}\) behavior.
- The energy calibration for \(\gamma\)-rays was carried out by using the \(\gamma\)-sources mentioned above. The edges of the charge distribution of Compton-scattered electrons are associated with \(\gamma\)-energies, which allows calibration. In order to do the same thing for neutrons, the detector was irradiated with spontaneous fission neutrons from a \(^{252}\)Cf source. A time-of-flight spectrum was recorded and converted into energy. By analyzing the charge distributions of the recoil protons corresponding to different energy bins and determining the edges, one obtains a relation between a charge value and the incident neutron energy. For both neutrons and \(\gamma\)-rays a good linearity was observed. The equivalence of the maximum recoil proton energy (\(E_p\)) and the electron energy (\(E_e\)), leading to the same charge, is represented by the so-called light output curve of the detector, as depicted in Fig. 1. There, our experimental values are compared to the results of a fit to the function
\[ E_e = L_0 \frac{E_p^2}{E_p + L_1}, \]

which according to (Matei et al., 2012; Kornilov et al., 2009) describes the observed dependence. The obtained fit parameters are \( L_0 = (0.500 \pm 0.026) \) and \( L_1 = (1.95 \pm 0.34) \) MeV.

- The total neutron efficiency corresponds to the ratio between the numbers of detected and emitted neutrons and consists of two components: (a) the geometric acceptance \( \epsilon_{\text{geo}} \), corresponding to the solid angle, which during our experiment was in the order of \( \epsilon_{\text{geo}} = (2.73 \pm 0.29) \times 10^{-4} \), (b) an intrinsic component \( \epsilon_{\text{intr}} \), which is a function of the detected neutron energy. In order to determine the differential intrinsic efficiency of our detector, we carried out a PFNS measurement by using a point-like \(^{252}\text{Cf(SF)}\) source. The fission trigger was provided by prompt fission neutrons, which were detected using a NE-213-equivalent BC-501 detector. The measured neutron spectrum was then compared with a theoretical description of the emission spectrum, provided by the Mannhart evaluation (Mannhart, 1989), which is considered as standard for \(^{252}\text{Cf(SF)}\). The ratio of both gives the relative differential intrinsic efficiency, as shown in Fig. 2. Absolute efficiency values could not been determined so far because the fission rate was not precisely known.

### 2.3. Digital data acquisition system: FASTER

The data from the experiments presented in this work were recorded with the digital acquisition system FASTER (Fast Acquisition SysTem for nuclEar Research), which is currently being developed at LPC Caen (http://faster.in2p3.fr/). Some real time functions, implemented on FPGAs (Field Programmable Gate Array) process the signals so as to get the relevant information (CFD timing, charge integration, pulse shape discrimination, etc.). Also, logical operations, like coincidences, may be set by the software, or may be defined off-line, since all recorded events are assigned an absolute time value. All settings, like e.g. thresholds for constant-fraction discriminators (CFD), are defined directly on the acquisition computer by a graphical user interface, which also allows the monitoring of the stored data in real-time.

### 2.4. Experimental setup

The PFNS experiments were performed using Van de Graaff accelerators in Bruyères-le-Châtel and Geel. These machines were used to produce quasi-monoenergetic neutrons at 5.2 MeV and 500 keV through the reactions \( \text{D(d,n)}^3\text{He} \) and \( \text{^7Li(p,n)}^7\text{Be} \), respectively. The produced neutrons irradiate the actinide samples inside the fission chamber. The
para-terphenyl detector, which is sensitive to neutrons and γ-rays, was placed at a distance of about 1 m from the fission chamber, perpendicular to the ion beam axis. It was surrounded by a shielding cone so as to be protected from the room-scattered neutrons. It was also shielded from direct neutrons and γ-rays emitted by the neutron production target. This experimental setup is shown in Fig. 3.

2.5. Data analysis process

Events in the para-terphenyl detector are investigated in coincidence with the trigger provided by the fission chamber. As 238U is an α-emitter, a threshold is applied both during the acquisition and the off-line analysis so as to discard any random coincidences caused by α triggering. A time-of-flight distribution may be obtained, which exhibits a prompt γ-peak as well as a region where neutrons are expected, and finally a flat background corresponding to random coincidences with background neutrons and γ-rays uncorrelated with fission events. The position of the prompt fission γ-ray peak serves for time calibration and its width is a measure of the overall timing resolution.

Pulse shape analysis (Leo, 1987) is performed by comparison of slow and fast components of the waveforms using integrals over different time windows as depicted in Fig. 4. Here, the purple line corresponds to the discrimination function used in the analysis. Since both regions still overlap for low values of the total charge (corresponding to the deposited energy), only events above 500 keV neutron energy (indicated by the vertical dashed line) are considered. The resulting neutron time-of-flight distribution is shown in Fig. 5. The remaining constant background is subtracted and the time-of-flight is converted into energies. The spectrum is then normalized to the number of fission events and corrected with the neutron efficiency according to Fig. 2. This results in an experimental prompt fission neutron spectrum, as shown in the following section.

3. Results and discussion

The detected prompt neutron energy distribution from the reaction 238U(n,f) induced by 5.2 MeV neutrons has been corrected with the measured neutron efficiency presented in Fig. 2. The resulting PFNS is shown in Fig. 6. The error bars on the yield contain statistical uncertainties from both the measurement itself and the determination of the efficiencies, while the error bars on the energies refer to the widths of the respective energy bins. This result is compared with an earlier measurement (Trufanov et al., 2001), obtained in Obninsk in 2001 for 5 MeV neutrons, and to data libraries ENDF/B-VII and JEFF-3.1. A normalization factor has been applied to our data as an absolute efficiency has not yet been determined for our detector (cf. Sect. 2.2).
Figure 4. Pulse-shape discrimination between neutrons and γ-rays: the black dashed line corresponds to a neutron energy of 500 keV and the purple line to the applied discrimination function.

Figure 5. Neutron time-of-flight spectrum (data sample) after applied neutron-γ discrimination according to Fig. 4. Obviously, a small rest of the prompt γ-peak still remained.

Figure 6 shows a fair agreement between all data sets, however, our data exhibits some deviations from the others. For one, there is an apparent enhancement in the yield around 1 MeV; for the other, the high energy region of the spectrum above 4 MeV is slightly underestimated by our data. This is confirmed by the result of fitting Maxwellians to the experimental values, which gives a nuclear temperature parameter $T = (1.23 \pm 0.05)$ MeV for our work, while the corresponding value for Trufanov et al. is $T = (1.34 \pm 0.01)$ MeV. We believe that this discrepancy is caused by systematic errors on the neutron efficiency, which is the reason why major efforts are now dedicated to the precise determination of the latter. Hence, the presented PFNS has to be considered a preliminary one.

4. Conclusions and outlook

In this paper we have reported on the measurement of the prompt fission neutron spectrum (PFNS) from the neutron-induced fission of $^{238}$U at $E_n = 5.2$ MeV. We described the instrumentation and the setup of the experiment carried out at the CEA research center in Bruyères-le-Châtel. Our results of the thorough characterization of the 75 mm $\times$ 50 mm para-terphenyl detector are in good agreement with those published by (Matei et al., 2012), although it
was observed that similar detectors of this type may have quite different properties (Hambsch, 2012). Nevertheless, it may be concluded that these detectors, in conjunction with an appropriate data acquisition system like the one used here, may indeed constitute an excellent tool for measuring PFNS down to low energies.

Some very encouraging results have been obtained for the PFNS of $^{238}$U(n,f) induced by 5.2 MeV neutrons. Further investigations remain to be performed as for the precise determination of the neutron intrinsic efficiency of our para-terphenyl detector, both with a known continuous neutron source like $^{252}$Cf and with quasi mono-energetic neutrons produced in well-known reactions from light ions of certain energies. MCNP simulations will have to complement the experimental studies in order to assess the influence of the environment on the shape of the neutron spectra. And finally, data from other measurements on $^{235,238}$U and $^{237}$Np, already performed in Bruyères-le-Châtel and at the Institute for Reference Materials and Measurements (IRMM) in Geel, respectively, will be analyzed in the near future.

References


