Measurement of prompt gamma-ray emission in fission

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Abstract

In this work we present results from the characterization of both LaCl₃:Ce scintillation and polycrystalline diamond detectors to be used for measurements of prompt gamma radiation emitted in fission. The properties of these new detector types, such as excellent timing resolution and improved energy resolution will allow to obtain fission data with higher accuracy than those available today.

Introduction

A particular challenge for the modelling of new generation reactor neutron kinetics is the calculation of the \( \gamma \)-heat deposition e.g. in steel and ceramics reflectors without UO₂ blankets, which is required to be known with an uncertainty as low as 7.5% [1]. The comparison of various benchmark experiments with calculated \( \gamma \)-heating shows a systematic underestimate ranging from 10 to 28% for the main fuel isotopes \(^{235}\text{U}\) and \(^{239}\text{Pu}\). This is attributed to deficiencies in \( \gamma \)-ray production data in evaluated nuclear data files. Requests for new measurements of prompt \( \gamma \)-emission in the reactions \(^{235}\text{U}(n,f)\) and \(^{239}\text{Pu}(n,f)\) have been formulated and included in the Nuclear Data High Priority Request List of the Nuclear Energy Agency [2].

Data found in modern nuclear-data libraries all date back to experiments performed in the early 1970's. In those experiments NaI scintillation detectors were used as \( \gamma \)-ray spectrometer with an ionisation chamber as fission trigger. A major difficulty in measuring the competition between neutron and \( \gamma \)-ray emission during fission fragment de-excitation is the suppression of background \( \gamma \)-rays induced by prompt fission neutrons in the \( \gamma \)-detector. A common method is to distinguish between \( \gamma \)-rays and neutrons by their respective different time-of-flight, which however is limited by the timing resolution of the detector (not better than 5 ns for NaI).

The characterization of recently developed Cerium-doped Lanthanum Chloride crystal scintillation detectors is the subject of an ongoing feasibility study at Institute for Reference Materials and Measurements, a Joint Research Centre of the European Commission. Three LaCl₃:Ce detectors have been characterized in terms of energy resolution, efficiency and timing resolution. The study, made in perspective of measuring prompt fission gamma-rays, has shown that the detectors are suitable for this purpose, having a much better timing resolution and efficiency than the NaI crystals previously used.

In the framework of a parallel project, a new highly time-resolving detector type based on polycrystalline CVD diamond has been introduced and tested. These detectors will be used in the two-arm time-of-flight spectrometer VERDI for the investigation of prompt and delayed neutron emission in neutron-induced fission that is presently being constructed and tested at IRMM. Among the advantages of this detector material is its very high radiation
tolerance compared to others (like e. g. Si) and its excellent timing properties. Using the fission trigger provided by the diamond detector in combination with the LaCl$_3$:Ce gamma detectors, measurements of fission fragments and prompt fission gamma-rays are possible with much higher accuracy.

1. Characterization of LaCl$_3$:Ce detectors

The detectors are cylindrical, with a size of 1.5"x1.5", and are doped with 5% Cerium. The crystals are attached to a photomultiplier of type Photonis XP2550/FB, and were purchased from Scionix Holland. A photograph of the detector is shown in Fig. 1.

The following features were investigated: the intrinsic activity, the energy resolution, the intrinsic efficiency and the timing resolution.

1.1. Intrinsic activity

It is known from previous studies [3] that this detector material possesses an intrinsic activity due to the decay of $^{227}$Ac (and all the daughters of its decay chain) and $^{138}$La. A characteristic spectrum illustrating this feature is shown in Fig. 2.

As can be seen, there are two possibilities of discarding the peaks coming from the intrinsic radioactivity: subtracting a scaled background spectrum from the measured spectrum or using a coincidence condition. The specific intrinsic activity of these particular detectors amounts to 1.3 Bq cm$^{-3}$, in reasonable agreement with previously reported results.

1.2. Energy resolution

The energy resolution of the LaCl$_3$:Ce detectors was investigated using standard calibration $\gamma$-sources up to an energy of 2.6 MeV and up to about 7 MeV by measuring $\gamma$-rays from excited states of $^{16}$O, produced in the reaction $^{19}$F(n,$\alpha$)$^{16}$O*. The FWHM of the peaks from the measured energy spectra is plotted as a function of $\gamma$ energy in Fig. 3. The results are satisfactory since the energy resolution for these detectors is 40% better than for NaI(Tl) scintillation detectors.
Fig. 2. Characteristic spectra taken with a LaCl₃:Ce detector. a). The black spectrum shows peaks from several γ-sources as well as the peaks caused by the intrinsic activity, as indicated on the peaks. The red spectrum is taken without any radioactive source and shows only the intrinsic activity peaks.
b). Background-subtracted spectrum (scaled).
c). Coincidence spectrum of two LaCl₃:Ce detectors.

Fig. 3. Energy resolution measured for energies ranging from 80 keV to 7 MeV. The grey dots represent the energies from the decay of excited states in ¹⁸O.
1.3. Intrinsic efficiency

The intrinsic efficiency was determined using several calibrated standard sources and compared with the efficiency of a NaI(Tl) detector of same size [4] and also with values obtained previously for other LaCl$_3$:Ce detectors [5]. The results are shown in Fig. 4. It can be observed that the efficiency of these detectors is 53% better than the one of NaI(Tl) detectors of the same size over the entire energy range.

Fig. 4. Intrinsic peak efficiency of two LaCl$_3$:Ce detectors measured in this work: full and empty dots; the dashed line represents results obtained in Ref. [4]. The full line represents the intrinsic efficiency for NaI(Tl) detectors of same size.

1.4. Timing resolution

In order to determine the timing resolution, two LaCl$_3$:Ce detectors were used to measure coincident gamma-rays emitted from $^{22}$Na and $^{60}$Co sources. A timing spectrum measured in this way is shown in Fig. 5. The analysis of the obtained spectrum results in a timing resolution of 890 ps, which corresponds to an intrinsic timing resolution of 630 ps for each detector.

Fig. 5. Timing spectrum measured using two LaCl$_3$:Ce detectors.
2. Characterization of polycrystalline diamond detectors

The use of artificial polycrystalline and single-crystal Chemical Vapour Deposited (CVD) diamond material is being constantly extended to different research areas due to its excellent timing characteristics, radiation hardness and easy operation. In nuclear physics, diamond detectors are used mostly in high-energy experiments as beam monitors and tracking devices, replacing the traditionally employed Si detectors, because they survive in high radiation environments, have very low leakage current and need no cooling [6].

The detector used in this work is a 100 µm thick polycrystalline CVD diamond film. The active area is 10 mm x 10 mm (Fig. 6). Electrical contact is made by a gold pad deposited on the growth side of the film.

In order to estimate the intrinsic timing resolution of the CVD diamond detector, the following experimental set-up was used: two detectors were placed in a test reaction chamber; a spontaneous fission $^{252}$Cf source with an activity of 250 fissions/s was attached to one of the detectors. The other diamond detector was placed at a distance of 9.5 cm from the first one. The pressure in the chamber was $2.5 \times 10^{-5}$ mbar. The detectors were biased to 180 V.

Over the whole duration of the experiment, the radiation dose amounted to $2.6 \times 10^9$ fission fragments, corresponding to $8 \times 10^{10}$ alpha particles and $8 \times 10^9$ neutrons, respectively. In these conditions the output signals of the detectors remained unchanged which shows their high radiation tolerance and stability. The measured time-of-flight spectrum was analysed and a Monte Carlo simulation was performed for the determination of the timing resolution. The time spectrum shown in Fig. 7 corresponds to an intrinsic timing resolution of 280 ps for each CVD detector.
3. Conclusions and outlook

Two different types of detectors were characterized in this work. The LaCl$_3$:Ce detectors intended for measuring gamma-rays were investigated and found to have 40% better energy resolution and 53% better intrinsic efficiency compared to the previously used NaI(Tl) detectors, and also better timing resolution. The intrinsic activity present in the crystals can be suppressed by either imposing a coincidence condition or by subtracting a scaled background spectrum.

The polycrystalline diamond detectors have shown very high resistance to radiation and stability. Also, they are easy to operate and have very good timing resolution.

Therefore, we conclude that both detectors are very suitable for their intended purpose and it should be possible indeed to achieve better quality data on prompt gamma emission in fission by using LaCl$_3$:Ce detectors as gamma-detectors and pcCVD diamond detectors as fission trigger. An experiment on thermal neutron-induced fission of $^{235}$U is scheduled for early 2010 in the framework of the transnational access programme EFNUDAT.

References

[1] G. Rimpault, Proc. Workshop on Nuclear Data Needs for Generation IV, April 2005 (Editor P. Rullhusen) Antwerp, Belgium, World Scientific, ISBN 981-256-830-1 (2006) 46
[2] Nuclear Data High Priority Request List of the NEA (Req. ID: H3, H4)
[3] A. Owens et al., Nucl. Instr. Meth. A574 (2007) 110
[4] G. Gilmore, Practical Gamma-Ray spectroscopy, John Wiley & Sons, ISBN 978-0-470-86196-7 (2008)
[5] B. Ayaz Maierhofer and T. A. DeVol, Nucl. Instr. Meth. A578 (2007) 410
[6] R. S. Wallny, Nucl. Instrum. and Meth in Phys. Res. A 582 (2007) 824 – 828