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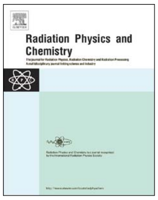

High precision measurements on fission-fragment de-excitation

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ABSTRACT

In recent years nuclear fission has gained renewed interest both from the nuclear energy community and in basic science. The first, represented by the OECD Nuclear Energy Agency, expressed the need for more accurate fission cross-section and fragment yield data for safety assessments of Generation IV reactor systems. In basic science modelling made much progress in describing the de-excitation mechanism of neutron-rich isotopes, e.g. produced in nuclear fission. Benchmarking the different models require a precise experimental data on prompt fission neutron and γ-ray emission, e.g. multiplicity, average energy per particle and total dissipated energy per fission, preferably as function of fission-fragment mass and total kinetic energy. A collaboration of scientists from JRC Geel (formerly known as JRC IRMM) and other institutes took the lead in establishing a dedicated measurement programme on prompt fission neutron and γ-ray characteristics, which has triggered even more measurement activities around the world. This contribution presents new advanced instrumentation and methodology we use to generate high-precision spectral data and will give a flavour of future data needs and opportunities.

1. Introduction

Nuclear fission is a rather complex process, which, even after almost 80 years since its discovery, is still not understood in all details. In recent years nuclear fission research has undergone a renaissance. Interest from the nuclear energy as well as the basic science community motivated several new measurement programmes. The first group is represented by the OECD Nuclear Energy Agency (), who expressed the need for more accurate fission cross-section and fragment yield data for safety assessments of Generation-IV reactor systems (Rullhusen, 2006). Concerning the second, modelling of the de-excitation mechanism of neutron-rich isotopes, as particularly produced in nuclear fission has recently advanced so much (Becker et al., 2013; Litaize et al., 2014; Talou et al., 2014; Serot et al., 2014; Stetcu et al., 2014; Schmidt et al., 2015; Vogt and Randrup, 2014) that precise experimental studies are necessary as benchmarks and references.

The energy released in nuclear fission is distributed in kinetic and excitation energy of the two fragments. The excitation energy manifests itself in fragment deformation and intrinsic excitation energy. The first step of fission fragment de-excitation takes place at a very early stage after scission through the successive emission of neutrons and γ rays (see the schematic in Fig. 1). In nuclear applications the prompt energy release by neutrons and γ rays accounts for about 5% of the total prompt heat set free in nuclear fission. At a later stage, the residual fragments are still unstable and release their excitation energy through β and subsequent γ-decay forming isotopes approaching the valley of nuclear stability. This delayed release of excitation energy contributes to the so-called decay heat. The knowledge about this particular quantity is related to the fission fragment yields and is highly relevant for the assessment of the nuclear fuel inventory and the eventual waste management.

Properties of the emitted particles prior to the onset of weak decays are important for the better understanding of the mechanism of fission-fragment de-excitation. Precise experimental data on prompt fission neutron and γ-ray emission, e.g. multiplicity, average energy per particle and total dissipated energy per fission, preferably as function of fission-fragment mass and total kinetic energy, are key input to benchmark nuclear fission models attempting to describe the competition between prompt neutron and γ-ray emission.

Some years ago, a collaboration of scientists from JRC Geel (the former JRC IRMM) and other institutes took the lead in establishing a dedicated measurement programme on prompt fission neutron (Kornilov et al., 2010; Göök et al., 2016) and γ-ray characteristics (Billnert et al., 2013; Lebois et al., 2015; Oberstedt et al., 2013a, 2016, 2014; Oberstedt et al., 2015; Oberstedt et al., 2015). They contributed with a number of precise measurements of prompt neutron and γ-ray

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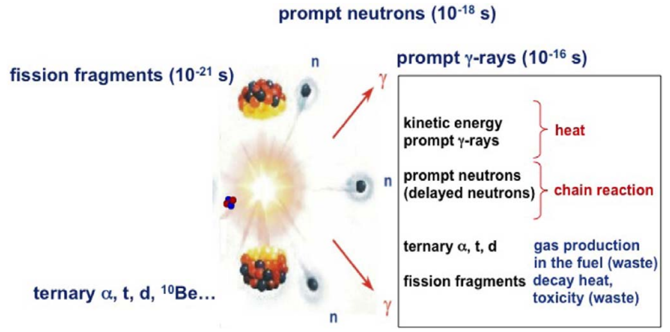

Fig. 1. Schematic of the nuclear fission process indicating the characteristic time frame for the transition of the compound nucleus from its ground-state to the formation of the fission fragments, occasionally accompanied by a light charged (ternary) particle and for the emission of prompt neutrons and γ-rays. In the box the impact of the different components on nuclear applications is given.

spectral data from spontaneous, thermal- and fast-neutron induced fission of several compound systems, which has triggered even more measurement activities around the world.

2. Challenges in nuclear (fission) physics research

The evidently progressive global warming requires a steady decrease of production of greenhouse gases. A considerable percentage must be attributed to energy production through the combustion of coal and gas. One economically viable way of essentially CO_2-free energy production is offered by nuclear fission of ^{233}U, ^{235}U and/or ^{239}Pu. Nuclear power reactors of the Generation-IV type are being designed under the condition to maximize the efficiency of the nuclear fuel and to minimize the production of long-lived radioactive waste as e.g. minor actinides. For reliable design studies and safety assessments of future nuclear reactors precise nuclear and in particular neutron-induced reaction data are an essential pre-requisite.

Despite the many decades of experimental and evaluation work devoted to neutron-induced reaction data the presently contemplated technologies cover a very different regime of incident neutron energies, calling for a new generation of nuclear data. As one example, in the domain of heat production in and close to reactor cores discrepancies between evaluated nuclear data and integral experiments became apparent some years ago (Rimpault, 2006) pointing to a lack of understanding of the heat production related to the reactor core (NEA Nuclear Energy Agency, 2006). The goal towards more efficiently reducing long-lived nuclear waste, e.g. by means of a fast-neutron spectrum and a higher burn-up, require measuring fission fragment yields as well as the neutron spectrum at relevant incident neutron energies. One may summarize important nuclear fission data needs in the following, although not exhaustive, list:

1. High mass-resolved fission fragment yield data for the main actinide isotopes from uranium and plutonium as well as from long-lived minor actinides as ^{237}Np, americium and curium isotopes
2. Prompt neutron multiplicity and spectrum characteristics in neutron-induced fission
3. Prompt fission γ-ray spectrum characteristics relevant for the heat production in the core and the surrounding of the Generation-IV nuclear reactors.

All these data are needed for incident neutron energies ranging from the so-called resolved-resonance region, i.e. the eV range, to that of fast neutrons up to 20 MeV kinetic energy, as the impact of excitation energy is illustrated in Fig. 2. Furthermore, those measurements reach out to the investigation of neutron-rich isotopes, far from nuclear stability and towards the neutron drip-line. They may help to reveal whether nuclear magic shells still exist at large deformation and strong neutron excess, which is relevant for astrophysical applications such as describing the nucleosynthesis in the universe by the s- and r-process.

3. Recent instrument developments

The generation of high-precision spectral neutron and γ-ray data was achieved by implementing advanced instrumentation and methodology. New technologies, concerning particle detectors and data acquisition as well as data analysis techniques, require benchmarking experiments to link present and new data to the past.

In the following we will discuss two major technical achievements, which were key to the latest advancements in the area of prompt particle measurements emitted in nuclear fission.

3.1. *A 4π position-sensitive double Frisch-grid ionization chamber (FGIC)*

In a typical experiment to measure prompt neutron emission in fission one needs a device to detect the fission event and a neutron detector to measure the emitted prompt neutron. The latter is typically made from a scintillating liquid, which allows distinguishing between incoming neutrons and γ rays, but does not possess a good pulse height resolution for γ rays.

For the purpose of acquiring nuclear data only, the measurement of the neutron spectrum characteristics in the laboratory frame, i.e. spectral shape, mean neutron energy per fission and average multiplicity, is sufficient.

However, if we want to learn about the underlying physics, we need to measure the fragments' kinetic energy and the emission angle of the emitted neutron relative to the fission axis in the centre-of-mass frame (see upper part of Fig. 3). A double Frisch-grid ionization chamber (FGIC) is often used to measure both the kinetic energy of each fission fragment (from which also its mass can be derived) and the angle relative to the chamber axis, as shown on the left part of Fig. 3 (Budtz-Jørgensen and Knitter, 1988; Al-Adili et al., 2012; Göök et al., 2014). FGIC are radiation resistant and have a geometrical efficiency of 4π.

The Frisch-grid is a mesh of wires with a typical diameter of 0.035 mm and a period of 0.5 mm. The chamber is usually operated with P-10, a counting gas consisting of 90% Ar and 10% CH4, at a pressure of 108.5 kPa and under a constant flow of about 80 ml/min. By placing the neutron detector along the chamber axis this angle coincides with the angle relative to the momentum direction of the detected neutrons. Hence, the projection of the neutron momentum on the fragments direction of travel is known, and the relevant kinematics in the fission fragment rest-frame can be reconstructed.

Employing an array of neutron detectors, necessary for collecting a significant number of events, each detector forms an axis of symmetry around which the fission fragment direction of travel needs to be known (see illustration in Fig. 3). As a consequence the traditional Frisch-grid ionization chamber is no longer sufficient to reconstruct the kinematics in the fragment rest-frame. Therefore, we have replaced the ionization chamber anode-plates by a position-sensitive read out structure, which allows determination of all three space-components $\{x, y, z\}$ of the fission fragments' direction of travel (Göök et al., 2016b). The position sensing structure consists of two parts, a plane of parallel wires and a strip anode. The wire plane is placed 4 mm above the Frisch-grid and the strip anode is placed 4 mm further above the wire plane, with the strips oriented perpendicular to the wires. Details and viewgraphs of the wire plane and the strip anode may be found in Göök et al. (2016b). Tungsten wires of 0.025 mm radius are soldered 2 mm apart to the support structure, made from a circular printed circuit board (PCB) of 17.76 cm diameter with a 10 cm×10 cm quadratic hole exposing the wires. In total 51 resistors of 100 Ω are surface-mounted in electrically insulated soldering pads forming a resistive charge-divider. The near and far-end of the charge divider is connected to