Abstract

At present, more precise data on capture and fission cross-sections and on fluctuation of prompt fission neutron and gamma-ray yields are needed for nuclear industry. A new experimental setup for investigating the resonance neutron induced capture and fission of $^{239}\text{Pu}$ has been constructed at the Frank Laboratory of Neutron Physics. It consists of 2 rings of 12 NaI(Tl) detectors with variable diameter and distance between both rings. Such a setup makes possible to measure the multiplicity, energy and angular distribution of prompt fission gammas. The signals from the 24 detectors are recorded simultaneously in digitized form and stored on the computer hard disks for further off-line analysis.

© 2013 The Authors. Published by Elsevier B.V. Open access under CC BY-NC-ND license. Selection and peer-review under responsibility of Joint Research Centre - Institute for Reference Materials and Measurements

Keywords: Nuclear Reactions; Resonance Neutron; Nuclear Fission; Plutonium-239; Prompt Fission Gamma-ray Emission
1. Introduction

The neutron induced fission of $^{239}$Pu is still of big interest because of its importance for the nuclear power industry. New more precise measurements of the characteristics of the reaction products are needed for modelling the new generation nuclear power reactor core as well as for utilizing of the spent fuel (Rimpault et al., 2006). The fission process is a quite complicated phenomenon because it can go different ways. According to nowadays knowledge, the capture of a $s$-wave neutron (spin $s=1/2$) of thermal ($E_{th} \sim 0.0253$eV) or resonance kinetic energy ($E_n < 500$eV) by $^{239}$Pu (“ground” state spin and parity $I^e = 1/2^+$), forms a compound nucleus (CN) of $^{240}$Pu mainly in 2 states (Bohr, 1955) with $J^e = 1^+$ and $J^e = 0^+$, which belong to two well separated ($\sim 1.25$MeV) transition state bands with $K^e = 0^+$ and $K^e = 1^+$ (Fig. 1).

![Diagram](image-url)

Fig. 1. $^{240}$Pu compound nucleus fission barrier and different channels leading to fission.

The de-excitation of the CN to its ground state can be by irradiating of one or more gamma-quanta (capture-reaction), or the CN can split into two or more fragments (fission-reaction). The process of fission is accompanied by releasing (on average) 2-3 prompt fast neutrons and ~7-8 gammas. There are some other possible channels of CN-disintegration which occur with less probabilities, but still of big importance when calculating the reaction energetics. One of them is the so called ternary fission in which light energetic particles are irradiated. The other is the $(n,\gamma f)$-reaction (Lynn, 1965), which was experimentally proven also to take place in the resonance neutron induced fission of $^{239}$Pu (Shackleton et al., 1972; Ryabov et al., 1973; Fréhaut and Shackleton, 1974; Trochon, 1978, Trochon, 1979; Scherbakov, 1990).

The multiplicity, total energy and spectrum shape of prompt $\gamma$-emission from fission of $^{239}$Pu (and $^{235}$U)
could be the major source of uncertainty in the prediction of \(\gamma\)-ray heating in thermal or fast cores of reactors, loaded with either uranium oxide (UOx) or mixed oxide (MOx) fuel.

To reduce the present uncertainties, these characteristics need to be measured with an accuracy of about 7.5% in thermal and fast neutron induced fission of \(^{239}\text{Pu}\) (and \(^{235}\text{U}\)). In the resonance neutron energy region quite large fluctuations of the fission mean prompt neutron multiplicities \(\langle n_p \rangle\) were measured (Shackleton et al., 1972). Such fluctuations have a significant impact on the reactivity coefficient of advanced water reactors (Fort et al., 1988), but their origin is still not quite clear. They correlate with mass yields \(A\) and mean total kinetic energy fluctuations \(\langle \text{TKE} \rangle_{\text{exp}}\) (Fig. 2) (Hambsch et al., 2012), but in less extend than in the \(^{235}\text{U}(n,f)\)-reaction (Hambsch et al., 1989), probably because of the viscosity effects and the only channel with \(J^*K=(0^+0)\) open to fission (Hambsch et al., 2012).

![Fig. 2. (a) Fluctuation of the fission relative \(\gamma\)-ray yields \(R/\langle R \rangle\) (Dermendjiev et al., 1991) and \(\langle\nu_{\gamma}\rangle\); (b) Fluctuation of the fission fragment \(\langle\text{TKE} \rangle\) from resonance-to-resonance (Hambsch et al., 2012).](image)

Similar fluctuations were obtained in the independent fission-fragment yields (Polhorský et al., 1993), too. From energy conservation point of view, the fluctuations of the \(\langle\nu_{\gamma}\rangle\) have to anti-correlate with those of the \(\langle n_p \rangle\). Data of Ref. (Ryabov et al., 1973) (shown in Fig. 2a, bottom) are explained as manifestation of the \((n,\gamma f)\)-reaction, which acts as a concurrent to the direct fission. And particularly, it concerns the fission resonance at \(E_n=44.53\text{eV}\), with a smaller fission width, where the contribution of the \((n,\gamma f)\)-reaction is expected to be more ‘visible’ than at the other \(1^+\) resonances.

The Fig. 2a-data (up part) were obtained in a short term measurement conducted at the IBR-30 neutron time-of-flight spectrometer (TOFS). The fluctuations of 3 or more gammas, measured in coincidence with the IC fission-fragment pulses, were found to be in the range of the experimental data uncertainties and did not show such a well pronounced pattern as at the experiments reported in Ref. (Shackleton et al., 1972; Ryabov et al., 1973; Fréhaut and Shackleton, 1974), in the whole neutron energy interval up to \(\approx 200\ \text{eV}\). The relatively large error-bars of the experimental data and the moderate neutron energy resolution of the IBR-30 TOFS, together with the need for data with better precision, trigger us to start preparation of a similar experiment at the recently commissioned IREN “white” spectrum pulsed neutron source, using NaI(Tl) multi-detector arrays and the multiplicity method for separation of the contribution of capture gamma-rays from that of fission gamma-rays (and neutrons).
2. The experimental setup

The experimental setup consists of: IREN as a “white” spectrum pulsed neutron source; vacuum tube collimated beam-line; a multi-sample parallel-plate gas ionization (fission) chamber (IC) loaded with $^{239}$Pu samples, as a charge particle detector; NaI(Tl) arrays as detector-spectrometer of gamma-rays (and neutrons) and a computerized system for multichannel data acquisition and analysis. As an alternative to the IC as a charge particle detector-spectrometer, a “sandwich” formed by $^{239}$Pu layers and Si-semiconductor charge particle detectors is considered, too.

2.1. The Intense Resonance Neutron Source (IREN)

A linear electron accelerator LUE-200 is used as the driver of IREN. It consists of a pulsed electron gun, accelerating system, RF power supply system based on klystrons with modulators, electron beam focusing and transport system, including a wide aperture magnetic spectrometer and a vacuum system. The accelerator is allocated vertically inside a 3-floors building. The IREN parameters are listed in Table 1.

Table 1. The main IREN parameters

| Parameter                      | Achieved     | New Project |
|--------------------------------|--------------|-------------|
| Maximal electron energy, MeV   | 50           | 200         |
| Pulse e-beam peak current, A   | 1.5          | 3           |
| Electron beam power, kW        | 0.4          | 10          |
| Pulse repetition rate, Hz      | 25           | 150         |
| Neutron producing target       | $^{238}$U    | $^{239}$Pu  |
| Integral neutron yield, n/s    | $3 \times 10^{11}$ | $3.4 \times 10^{13}$ |
| Neutron pulse width, ns        | 20-100       | 20-200      |

2.2. The charge particle detector

As a detector of charge particles, alphas or fission fragments (FF), a multi-plate gas IC will be used. The IC and the disposition of its electrodes are shown in Fig. 3. It is loaded with three thin (~0.14 mg/cm$^2$) reactor grade plutonium (~88% $^{239}$Pu) layers of total mass ~2.1 mg, deposited on ~20 |m| thick Al backings. Spots’ diameter is ~2.5 cm. As a working gas a mixture of 95%Ar + 5% CO$_2$ will be used at a constant pressure less than 1 atm. The same chamber (filled with P-10 gas) was used in the experiment, described in Ref. (Dermendjiev et al., 1991) at the IBR-30 pulsed reactor, using a large 6-sections 210/l liquid scintillation detector for gamma-ray detection (Malecky et al., 1972).

2.3. The multi-detector array for gamma-ray spectrometry

A new multi-detector gamma-ray spectrometry system was designed (Ruskov et al., 2012) and constructed (Fig. 3). A more detailed description of the spectrometer with associated electronics and data acquisition system (DAQ) will be published elsewhere. Here only a brief description is given. It consists of 2 rings (arrays) of 12 Amcrys-H NaI(Tl) detectors (Amcrys-H, Hamamatsu) both with variable ring diameter and distance between them. Such setup gives the possibility to measure not only the multiplicity, energy and
angular anisotropy of the prompt fission gammas, but also to separate the contribution of the prompt fission neutrons by their longer time-of-flight (TOF) from the fissile targets to the detectors. The construction allows as many as 24 detectors to be arranged in a concentric ring of diameter up to ~1 m. The signals from all the 24 detectors are recorded simultaneously in digitized form and are stored on a separate personal computer (acting as a file-server) for further off-line analysis by the AFI Data acquisition system ADCM16-LTC (AFI-ADCM). A computer screenshot with a typical signal from a single NaI detector is shown in Fig. 3 (lower right corner).

Fig. 3. Prompt fission gamma-ray spectroscopy system: 1 - Two arrays by 12 NaI(Tl) detectors, 2 - PMT+HV generator, 3-5V DC for powering the HV generator, 4 - parallel plate gas ionization chamber, 5 - computerized 32 channel ADC, 6 - Data acquisition software ADCM.

2.4. The data acquisition system

The data acquisition system (DAQ) of AFI-Dubna consists of 2x16 channel data acquisition board ADCM16-LTC and a software package, which includes a kernel module (driver), a control program, and a reconstruction program. The ADCM16-LTC, 16-channel 14-bit 100 MHz Analog-to-Digital Converter (ADC)
board with a signal processing core, is used together with a CCB-PCIe carrier board and utilizes one PCI slot of the PC. To drive the 24 detector gamma-ray spectrometry system and 4 signals from fission fragments detector, two ADCM16-LTC boards are connected together to form a system of 32 channels 14-bit. The theory of the ADCM operation is described in detail in Ref. (AFI-ADCM). The DAQ is designed for a direct digitizing of signal pulses coming from gamma detectors and fission chamber. It has a built-in trigger circuitry with three modes of operation: time-driven, single-channel, and double (gamma-gamma) coincidences. So, software wise is possible accurately to reconstruct the amplitude and time-mark of any signal and to form amplitude and/or time-spectra. The information from the 24 NaI(Tl) detectors and the fission chamber is collected and stored on separate computer hard-disks simultaneously. Two typical amplitude spectra from standard $^{137}$Cs+$^{60}$Co sources, used for energy calibration, are shown in Fig. 4.

The energy resolution of a single NaI detector is ~7% for 662 keV gammas from $^{137}$Cs. On the same Fig. 4 the coincidence spectrum between the pulses of a single NaI and NE213 detectors is shown. The time resolution of a single NaI was found to be ~3ns.

The neutron capture and fission gammas will be separated by their different multiplicities: the mean fission gamma-ray emission multiplicity is about 7-8 gammas/fission, when that from the capture process is about 2-3 gammas/capture. Using a standard $^{252}$Cf source the system can be properly calibrated. This way the neutron capture and fission processes can be simultaneously investigated and the capture-to-fission ratio coefficient fluctuations can be determined. The mean prompt capture and fission gamma ray multiplicities can be determined following the approaches described in (Theobald et al., 1972) and (Muradyan et al., 1976).

The fission fragments counts from IC can be used as an additional (to the multiplicity) constrains to separate neutron capture and fission gamma induced events and to determine the corresponding reaction cross-sections. Other possible uses of the new build multi-detector system are outlined in (Janeva et al., 2010).

3. Summary

In order to fulfil the Nuclear Community requests for improvement of the existing knowledge and quality of nuclear data, needed for modelling of the Generation IV nuclear facilities and, in particular, on the prompt fission gamma ray emission in neutron induced fission of $^{235}$Pu, new experiments are under preparation at the IREN facility. This work was partially supported by a grant of the Plenipotentiary Representative of the Government of Republic of Bulgaria in JINR.
Acknowledgements

One of us (I.R.) gratefully acknowledges the support of the JRC-IRMM for making it possible to attend this scientific workshop as a lecturer.

References

AFI-ADCM, A digital pulse processing system for nuclear physics experiments ADCM16-LTC, AFI-Electronics, http://afi.jinr.ru/ ADCM, http://afi.jinr.ru/ ADCM16-LTC.

Amcrys-H, Scintillation Materials and Detectors, www.amcrys-h.com.

Bohr, A., 1955, Proc. Int. Conf. on Peaceful Use of Atomic Energy 2, Geneva, New York: UN, 1956, p. 151.

Dermendjiev, E., Goverdovsky, A.A., Furman, W.I., Pikelner, L.B., Ruskov, Iv., Siegler, V., Zamyatin, Yu.S., 1991, Proc. Int. Conf. on Nuclear Data for Science and Technology, Jülich, 13-17 May, Berlin: Springer-Verlag, 1992, pp. 147-149.

Fort, E., Fréhaut, J., Tellier, H. and Long, P., 1988, Nucl. Sci. Eng. 99, p. 375.

Fréhaut, J., Shackleton, D., 1974, Proc. III Symp. on Physics and Chemistry of Fission 2, Vienna: IAEA, p. 201.

Hamamatsu, Scintillation Detectors Photomultiplier Tubes, http://www.hamamatsu.com/jp/en/index.html.

Hambsch, F.-J., Ruskov, I., Dematté, L., 2012, Resonance neutron induced fission of $^{239}$Pu, THEORY-1 Proc. Sci. Workshop on Nucl. Fission Dynamics and the Emission of Prompt Neutrons and Gamma Rays, 27-29 Sept, Sinaia, Romania, JRC:EUR 24802 EN, 2011, p. 41. http://publications.jrc.ec.europa.eu/repository/handle/111111111122054.

Hambsch, F.-J., Knitter, H.H., Budtz-Jørgensen C., Theobald, J.P., 1989, Nucl. Phys. A491, p. 56.

Janeva, N.B., Grigoriev, Yu.V., Gundorin, N.A., Lukyanov, A.A., Kopatch, Yu.N., Koyumdjiev, N.T., Pikelner, L.B., Ruskov, I.N., Shvetsov, V.N., Sedyshev, P.V., Zeinalov, Sh., 2010, Future neutron data activity on the neutron source IREN, Proc. Int. Conf. on Nuclear Measurements, Evaluations and Applications-NEMEA-6, Krakow, 25-28 Oct., France: OECD-NEA, 2011, p. 153. www.oecd-nea.org/science/wpec/nemea6/, www.oecd-nea.org/science/wpec/nemea6/NEMEA6_web.pdf.

Lynn, J.E., 1965, Phys. Lett. 18, p. 31.

Malecky, K., Pikelner, L.B., Rodionov, K.G., Salamatin, I.M., Sharapov, E.I., 1972, Report JINR13-6609, Dubna: JINR.

Muradyan, G.V., Schepkin, Yu.G., Adamchuk, Yu.V., 1976, Report IAE-2634. Moscow: Kurchatov IAE.

Pohlowski, V., Kliman, J., Ktrištiak, J., Bogdzel, A.A., Gundorin, N.A., Popov, A.B., Gohs, U., 1993, Proc. Int. Workshop on Dynamical Aspects of Nuclear Fission, ČSSR, p. 258.

Rimpault, G., Courcelle, A., Blanchet, D., 2006. Needs for accurate measurements of spectrum and multiplicity of prompt $\gamma$ emitted in Fission, OECD HPRL, France: NEA, http://www.oecd-nea.org/dbdata/hprl/tmp/HPRLgammapfission.pdf.

Ruskov, I., Kopatch, Yu.N., Panteleev, Ts., Skoy, V.R., Shvetsov, V.N., Dermendjiev, E., Janeva, N., Pikelner, L.B., Grigoriev, Yu.V., Mezentseva, Zh.V., Ivanov, I., 2012, Prompt Gamma Emission in Resonance Neutron Induced Fission of $^{239}$Pu, Physics Procedia, 31, pp. 35-42, http://www.sciencedirect.com/science/article/pii/S187538921201923.

Ryabov, Yu., Trochon, J. and Shackleton, D., 1973, $\gamma$-ray multiplicity in $^{239}$Pu fission induced by resonance neutrons: experimental evidence for the (n,$\gamma$) reaction, Nucl. Phys. A216, p. 395.

Scherbakov, O., Sov. J., 1990, Part. Nucl. 21, p. 177.

Shackleton, D., Trochon, J., Fréhaut, J., Le Bars, M., 1972, Prompt neutron and gamma-ray yields in fission induced in $^{239}$Pu by resonance neutrons: existence of the (n,$\gamma$) reaction, Phys. Lett. 42B, p. 344.

Theobald, J.P., Wartena J.A., Kolar W., 1972, Report EUR-4829 E, Geel: CBNM.