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Fission Experiments at *n*ELBE

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Abstract

A ^{235}U and a ^{242}Pu parallel-plate fission ionization chamber will be used to investigate fast neutron-induced fission cross sections at the Center for High-Power Radiation Sources at Helmholtz-Zentrum Dresden-Rossendorf. To optimize the chamber parameters extensive Geant4 simulations with GEF code generated fission observable inputs have been used. Pile-up effects had to be included due to the high α-activity of the plutonium targets. For the determination of targets surface density and homogeneity an α-spectroscopy setup was developed and simulations related to that will also be presented.

Selection and peer-review under responsibility of Joint Research Centre - Institute for Reference Materials and Measurements

Keywords: Neutron-induced fission cross sections; Stopping power of fission fragments using Geant4; Minor actinide target characterization; *n*ELBE

1. Introduction

For the simulation of transmutation in innovative reactor and accelerator driven systems (ADS) accurate nuclear data is required (Salvatores and Palmiotti, 2011). Sensitivity studies (Working Party on International Evaluation Cooperation , 2011; Working Party on International Evaluation Co-Operation, 2008) show that a reduction of the total uncertainty of cross section data below 5 % is needed to perform reliable neutron physical calculations. Especially neutron-induced fission cross sections of plutonium and minor actinides show high uncertainties in the high-energetic neutron range. For example, available ^{242}Pu data are discrepant by about 21 % (cf. Fig. 1), uncertainties related to target properties in the order of 7 %.

Improved background conditions (low-scattering environment) and beam power, paired with the adequate spectral shape of the new *n*ELBE neutron beam (Beyer et al., 2013) will provide excellent conditions to face the challenging task of reducing nuclear data uncertainties at Helmholtz-Zentrum Dresden-Rossendorf (HZDR).

2. Design of the *n*ELBE fission chambers

Two similar parallel-plate fission ionization chambers are under development at HZDR. Both chambers will measure fission fragments from thin minor actinde layers with more than 90 % detection efficiency. The first chamber will

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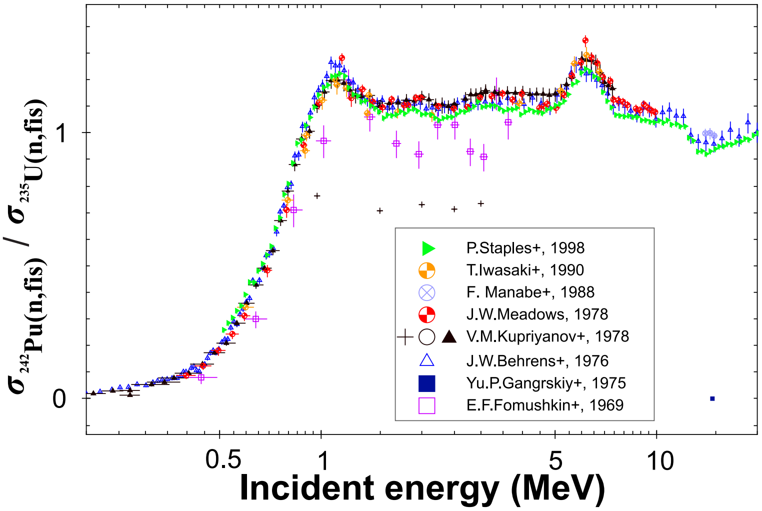

Fig. 1. Fast neutron-induced fission cross sections of ^{242}Pu normalized to the fisson cross section of ^{235}U. The data presented was taken from the EXFOR database (Nuclear Reaction Data Centres Network, 2008) (graph taken from Janis 3.4 (OECD/NEA, 2013)). Between 1 and 6 MeV large discrepancies between the different data sets are clearly visible.

be filled with totally 50 mg of plutonium (the isotopic composition is shown in Tab. 1(b)) distributed on 8 samples, the other one will contain 140 mg of uranium (Tab. 1(a)). Since the neutron-induced fission cross section of ^{235}U is

Table 1. Projected isotopic composition of the targets.

(a) Uranium target

Isotope	Mass-ratio
^{234}U	0.0100
^{235}U	87.9650
^{236}U	0.0039
^{238}U	12.0261

(b) Plutonium target

Isotope	Mass-ratio
^{238}Pu	0.003
^{239}Pu	0.005
^{240}Pu	0.022
^{241}Pu	0.009
^{242}Pu	99.959
^{244}Pu	0.002

known very accurate, the second chamber will determine the incoming neutron flux very precisely.

Choosing a target diameter (74 mm) larger than the neutron beam avoids uncertainties related to beam profile effects. On the other hand side this requires very homogeneous targets. Using the molecular plating technique in combination with 400 μm thick titanium coated silicon wafer backings (Vascon et al., 2011, 2012) this requirements could be fulfilled by our colleagues from University of Mainz. The resulting surface density will be about 150 μg/cm^2 for the plutonium and 400 μg/cm^2 for the uranium samples.

Standard vacuum components (cf. Fig. 2) will be used for the construction of the chamber housing. To protect against incorporation of target material, especially the plutonium, the chamber will be a metal-sealed vacuum chamber. In addition the chamber is operated below atmospheric pressure in combination with valves and High-Efficiency Particulate Air (HEPA) filters to prevent against leakage.

A critical design parameter is the choice of the counter gas. Typical counter gas mixtures are combinations of noble and polyatomic gases. The former neither forms anions nor contains components, which attracts electrons. Polyatomic gases, like methane and other organic gases deexcite mostly without the emission of photons (non-radiative modes). They are added to absorb photons created by the noble gases in secondary Townsend avalanches (L'Annunziata, 2003).

According to Ref. (Beringer et al., 2012, Tab. 31.5) the heavier noble gases krypton and xenon have a lower energy

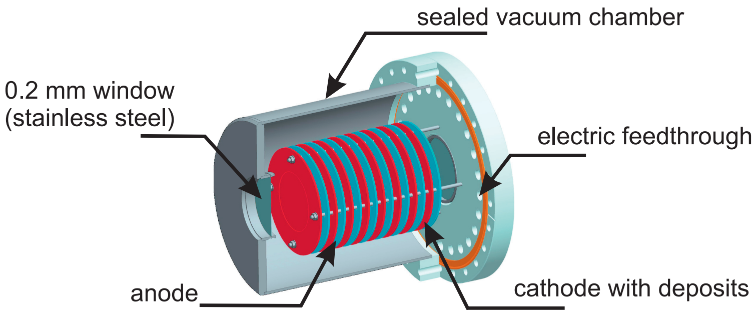

Fig. 2. Computer-aided design of the nELBE fission chamber.

threshold for the production of electron-ion pairs compared to argon. This results in a larger number of primary and secondary charge carriers and further in a better energy resolution. But due to their higher density the electron drift velocity is smaller (Peisert and Sauli, 1984), which leads to longer signal times and weaker time-of-flight resolution. However, the formation of charge carriers in denser noble gases is quite more localized, which maybe could lead to a compensation of this effect or even to an improvement of the time-resolution. Considering all above mentioned properties and additionally the handling and the costs of the gases, three counter gas mixtures have been selected for further investigation:

1. 90 % argon + 10 % methane (P10)
2. 90 % krypton + 10 % tetrafluoromethane
3. 90 % xenon + 10 % isobutane

3. Simulations of energy deposition in different counter gases

To handle the high specific α-activity of the Pu targets, a combination of fast preamplifiers and digital signal processing has been developed to suppress pile-up effects. A fast charge-sensitive preamplifier was developed at HZDR that produces total signal times in the order of 300 ns and shows identical performance in terms of time and energy resolution compared to conventional preamplifiers with μs discharge times. The signals will be sampled by an Acqiris DC-282 10 bit fast digitizer. A fast ROOT-based DAQ with Qt graphical user interface was developed to record complete signal waveforms. Offline and online digital signal processing including pile-up rejection, charge-integration and digital constant fraction triggering (to only present a small part of it functionality) is provided. Very good acquisition performance and stability was achieved by using multithreading.

Nevertheless, pile-up events related to the α decay will influence the measurement. The α-decay rate per sample is expected to be 1.51 million per second. Occurring within a time window of typical signal rise-times of 110 ns the probability of higher (2nd, 3rd and even 4th) order pile-up is not negligible. This could lead to a misinterpretation of fission events. To optimize the target thickness and total mass, simulations have been performed using the Geant4 framework (Allison et al., 2003).

To use more realistic fission fragments properties in the Geant4 simulation, the charge, mass and kinetic energies of the fission fragments were simulated using the General Description of Fission Observables (GEF) code (Schmidt and Jurado, 2011). Accurate data describing the α decay of plutonium was provided by the radioactive decay package of Geant4 (G4RadioactiveDecay).

The probability P_n of detecting n additional α particles to the primary particle is given by

$$P_n(R, \tau) = \frac{(R\tau)^n e^{R\tau}}{n!}. \quad (1)$$

Thereby R denotes the expected detection rate and τ the time window, in which these events should occur. The fission rate was scaled with respect to a measurement at nELBE using the ^{235}U fission chamber H19 (Nolte et al., 2007) of

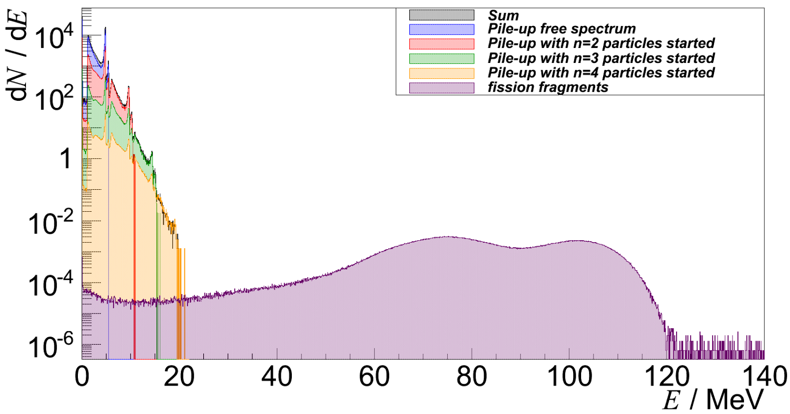

Fig. 3. GEANT4 simulated energy deposition of the decay products and fission fragments from neutron-induced fission of the *n*ELBE plutonium target material in 90 % xenon + 10 % isobutane counter gas.

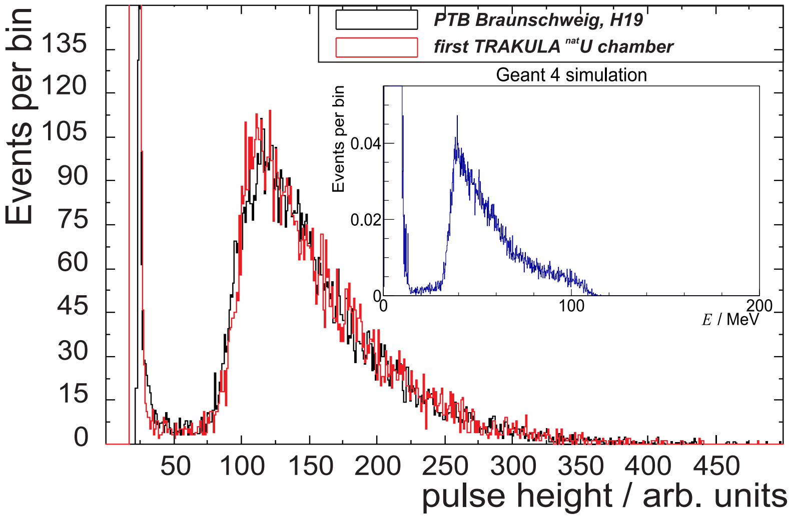

Fig. 4. Comparision of simulated (inset) and measured pulse-height distribution of the PTB ^{235}U fission chamber H19.

the Physikalisch-Technische Bundesanstalt (PTB) Braunschweig. Within the simulation pile-up up to the 3^{rd} order was considered. The outcome of this procedure is shown in Fig. 3, which only reflects the energy deposition in the counting gas.

With 50 mg of plutonium and the intended configuration a separation of α-induced background events from the main part of the fission fragment distribution could be achieved. The amount of fission events below the threshold in the simulation is less then 0.8 % in the xenon case. Results of the other counter gas mixtures are similar. For the experimental determination of this fraction the shape of the fission fragment distribution below the threshold is interesting. The distribution drops firstly linearly to rise again below 10 MeV.

Since no measurement exists to validate the results of the performed simulation so far, a description of the H19 fission chamber was implemented into the code and the outcome of this simulation was compared with a measurement of the pulse-height spectrum of this chamber at PTB (cf. Fig. 4).

The results show a good agreement of simulation and measurement. However, discrepancies are shown in the plateau and in the high-energetic tail of the fission fragment distribution. For a detailed description of the fission fragment yield in this regions simulations including the signal generation process (Plompen et al., 2011) are ongoing.

4. Target characterization

It is planned to determine the homogeneity of the minor actinide targets by two different methods. Due to their high specific activity the number of fissionable plutonium atoms per unit area will be determined by a spatially resolved α-spectroscopy. This could be a complementary approach to radiographic imaging of the targets performed at University of Mainz. The required setup (cf. Fig. 5) was optimized using Geant4 simulations.

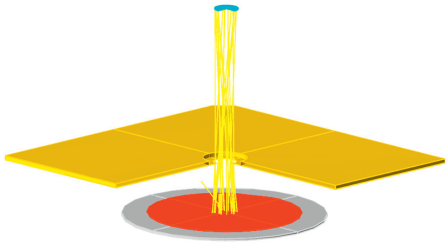

Fig. 5. Geant4 model of the planned α-spectroscopy setup. The aluminum collimator is displayed in yellow, the target material in red. For a better visualization one part of the structure was cut to get a better view on the drilling and the thin aluminum foil. Emitted α particles from the active area impinging the ion implanted silicon detector (cyan) and generating the signal are shown in yellow lines.

A combination of 50 µm aluminum foil with 1 cm diameter aperture and a 5 mm aluminum supporting structure will lead to a defined solid angle. The contribution of scattered α particles at the edge of the foil was determined to be less than 10^{-9}. With the nominal energy resolution of the ion-implanted silicon detector (15 keV at 5.4 MeV) the α full energy peaks of Pu isotopes could only partially be resolved, but clearly discriminated from X-rays, β, δ and scattered α particles (cf. Fig. 6).

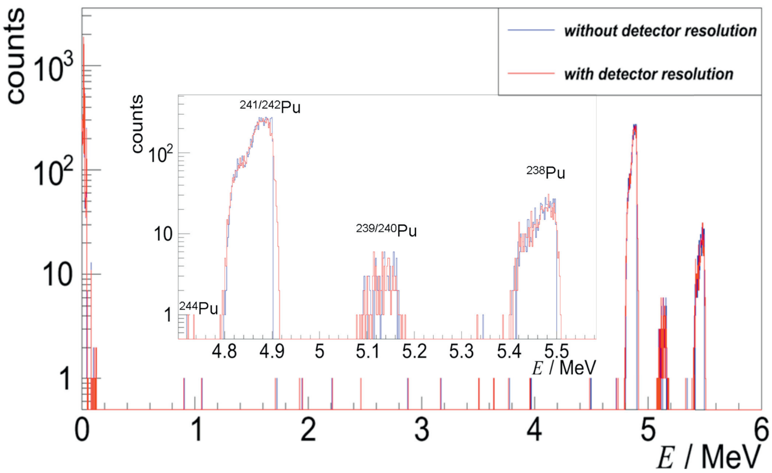

Fig. 6. Deposited energy of decay products of the nELBE plutonium target material in an ion-implanted silicon detector calculated using Geant4.

For the low-active uranium targets it is planned to determine the homogeneity in a fission chamber with a collimated neutron beam at PTB Braunschweig.

5. Conclusions

Fast neutron-induced fission experiments on ^{235}U and ^{242}Pu will be performed at the neutron time-of-flight facility *n*ELBE in the near future. Fission cross sections will be examined using a parallel-plate fission ionization chamber. Different chamber parameters have been optimized by using extensive Geant4 simulations. For the announced 50 mg of plutonium and the resulting target thickness the loss of fission events below the trigger threshold is acceptable low and the calculated neutron-induced fission rate is high enough to perform experiments with sufficient statistics in less than one week. The investigation of the fission yield distribution in the very low energy range is ongoing.

The usage of different counter gas mixtures has been investigated. The gain in energy resolution using xenon instead of methane is rather small (< 0.2 ‰). The electron drift velocity in the xenon mixture has to be at least half the velocity in P10 gas, to get a comparable timing-resolution. Drift velocity measurements in xenon-methane mixtures allows hypothesizing that it will be much lower.

A setup for the determination of the plutonium target areal density and homogeneity was developed. The feasibility could be demonstrated in a Geant4 based simulation.

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References

Allison, J. et al., 2003. Geant4 - a simulation toolkit, Nucl. Instrum. Method. A 506, 250 - 303.
Beringer, J. et al., 2012. J. Beringer et al., Review of particle physics, Phys. Rev. D 86, 010001.
Beyer, R. et al., 2013. Characterization of the neutron beam at nELBE, Nucl. Instrum. Method. A, to be published.
L'Annunziata, M. F., 2003. Handbook of Radioactivity Analysis, 2nd Edition, Academic Press Inc.
Nolte, R. et al., 2007. Cross sections for neutron-induced fission of U-235, U-238, Bi-209, and Pb-nat in the energy range from 33 to 200 Mev measured relative to n-p scattering, Nucl. Sci. Eng. 156, 197 - 210.
Nuclear Reaction Data Centres Network, 2008.EXFOR formats description for users, Tech. rep., IAEA.
OECD/NEA, 2013. A new java-based nuclear data display program.
Peisert, A. and Sauli, F., 1984. Drift and Diffusion of Electrons in Gases: A Compilation, Tech. Rep. 84-08, CERN - Experimental Physics Devision 139.
Plompen, A. et al., 2011. Method Developing and Testing for Inelastic Scattering Measurements at the GELINA Facility, J. Korean Phys. Soc. 59 (2) 147, 1581 - 1584.
Salvatores, M. Palmiotti, G., 2011. Radioactive waste partitioning and transmutation within advanced fuel cycles: Achievements and challenges, Progr. Part. Nucl. Phys. 66, 144 - 166.
Schmidt, K.-H. and Jurado, B., 2011. Thermodynamics of nuclei in thermal contact, Phys. Rev. C 83, 014607.
Vascon, A. et al., 2011. Toward large-area targets for "TRAKULA", Nucl. Instrum. Method. A 655, 72 - 79.
Vascon, A. et al., 2012. Elucidation of constant current density molecular plating, Nucl. Instrum. Method. A 696, 180 - 191.
Working Party on International Evaluation Co-operation (WPEC), 2011. The High Priority Request List for Nuclear Data (HPRL).
Working Party on International Evaluation Co-Operation (WPEC), 2008. Uncertainty and target accuracy assessment for innovative systems using recent covariance data evaluations, Tech. rep., OECD / NEA.