Fission studies using multinucleon transfer reactions

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Abstract. It is shown that the multinucleon transfer reactions is a powerful tool to study fission of exotic neutron-rich actinide nuclei, which cannot be accessed by particle-capture or heavy-ion fusion reactions. Identification of fissioning nuclei and of their excitation energy is performed on an event-by-event basis, through the measurement of outgoing ejectile particle in coincidence with fission fragments. Fission fragment mass distributions (FFMDs) are measured for each transfer channel, in selected bins of excitation energy. It was found that the mass distributions for all the studied nuclides maintain a double-humped shape up to the high energies that the shell effects are expected to smear out. From a comparison with the dynamical calculation based on the fluctuation-dissipation model, this behavior of the mass distributions was unambiguously attributed to the effect of multichance fission.

1. Introduction
Induced nuclear fission is a unique decay process which may be described by the interplay of macroscopic (collective) and microscopic (single particle) degrees of freedom in a nucleus [1]. Such a description of the fission process allows for studies of nuclear shell structures, nuclear viscosity and their excitation energy dependence at extreme values of deformation. In particular, fission fragment (FF) mass distributions and their sensitivity to the excitation energy and isospin provide a deep insight into the mechanism of the process of fission.

Since the discovery of fission [2], fission process was intensively studied in capture reactions with light particles (e.g. neutrons, protons) and γ-rays, as well as with spontaneously fissioning nuclei [3]. Also, complete-fusion reactions with heavy ions and also few-nucleon direct transfer reactions of e.g. (d,pf), (3He,pf) or (6Li,df) and similar types started to be exploited. Often, such studies concentrated mostly on the measurements of fission probabilities and their excitation energy dependence, see e.g. [4, 5, 6, 7, 8, 9, 10, 11, 12]. Fission fragments mass distribution (FFMD) measurements via the direct few-nucleon transfer was reported in [13] for the isotopes 227,228Ac, studied with the 226Ra(3He,df) and 226Ra(3He,pf) reactions, respectively. The FFMD of the 239Pu(d,pf) reaction via the superdeformed β vibrational resonance was reported [14]. As the available target materials are limited to stable or long-lived radioactive nuclides, nuclei to be studied in neutron-rich nuclei were rather limited.

The scope of the present work is to explore the potential of the multinucleon transfer (MNT) reactions to measure FFMDs and their excitation energy dependence for neutron-rich nuclei, which cannot be accessed by particle-capture and/or heavy-ion fusion reactions. The MNT channels of the 238U+12C reaction in inverse kinematics were recently used to study fission of excited transactinide nuclei with the help of the large-acceptance magnetic spectrometer VAMOS@GANIL [15, 16, 17].
At the JAEA tandem facility in Tokai, on the contrary, we have developed a system to investigate fission of excited nucleus by the use of $^{18}$O induced MNT reactions [18, 19]. An obvious advantage of this method is a relatively easy possibility to change the projectile and/or the target nuclei. In particular by using targets of the rarest highly-radioactive neutron-rich isotopes heavier than $^{238}$U (e.g. Cm and Cf), nuclei to be studied can be extended to isotopes far heavier than uranium, which cannot be used as a beam at the accelerator facilities for the inverse kinematics experiments.

2. Experimental method and results
The experiments were performed at the JAEA tandem accelerator facility using a 157.0–162.0 MeV $^{18}$O beam with an intensity of $\sim$0.5 pnA. The target was prepared by electrodeposition of a radioactive target material (e.g. $^{232}$Th, $^{238}$U, and $^{237}$Np) on a 90–300 $\mu$g/cm$^2$ nickel backing. The setup consists of a multidetector $\Delta$E-E silicon telescope, to detect ejectiles, and four multiwire proportional counters (MWPCs), to detect fission fragments as shown in Fig.1. Details of the experimental setup is given in [18, 19].

Figure 1. Schematic detection set-up (left) and expanded view of the silicon $\Delta$E-E detector telescope (right).

Figure 2. $\Delta$E-$E_{\text{tot}}$ spectrum for ejectiles measured by one pair of the $\Delta$E-E detectors, obtained in the $^{18}$O+$^{238}$U reaction ($E_{\text{beam}}$=157.5 MeV). The curves corresponding to different ejectiles are marked with the respective isotopes.
Figure 3. Fission events registered on the mass and excitation energy plane for (a) $^{236}\text{Np}^*$, (b) $^{238}\text{Pu}^*$, and (c) $^{240}\text{Am}^*$, obtained in the $^{18}\text{O}+^{237}\text{Np}$ reaction.

Specific particle-transfer channels were determined by identifying the ejectiles using the \(\Delta E\)-E silicon telescope. An ejectile passing through one of the twelve \(\Delta E\) detectors (75 \(\mu\)m thick) is stopped in the 16-strip annular E detector (300 \(\mu\)m thick) to measure the residual energy \(E_{\text{res}}\). Thus, the ejectile kinetic energy \(E_{\text{ejectile}}\) is represented by \(\Delta E + E_{\text{res}}\). The direction of a scattered ejectile was determined by the combination of a \(\Delta E\) segment and one of the strips in the E detector. As the \(\Delta E\) detectors have a good energy resolution, achieved using silicon wafers of highly uniform thickness (<1.3% variation), it allowed us to distinguish not only the ejectiles of different elements (e.g. F, O, N, C, B, Be, Li), but also different isotopes of each element, as shown in Fig. 2. Considering the binary kinematics, the method allows to determine the total excitation energy of the exit channel \(E^*_{\text{tot}}\), as being the sum of the excitation energy of the fissioning nucleus and of the ejectile. It is assumed that all the excitation energy is given to the recoiled fissioning nucleus \((E^*)\), without exciting the ejectile nucleus; thus the excitation energies quoted in this study should be considered as the upper limit.

The coincident FFs resulting from the fission of excited nuclei (after the MNT) are detected by four 200×200 mm$^2$ position-sensitive MWPC (Fig. 1). The flight-time difference between two fragments was measured to determine the pre-neutron emission masses.

Figure 3 shows the fission events for $^{236}\text{Np}^*$, $^{238}\text{Pu}^*$ and $^{240}\text{Am}^*$ presented in the plane of the fission-fragment mass \(A\) and excitation energy of fissioning nucleus \(E^*\). For $^{236}\text{Np}^*$, we clearly see the threshold for fission at \(E^*\sim 6.3\) MeV, corresponding to the fission barrier height (see Ref.[20] for the derivation of the fission barrier height). It is also evident that the light- and heavy-fragment groups are clearly separated at the low excitation energies below \(E^* = 30\) MeV, which tends to smear out toward higher excitation energies. A similar excitation energy dependence is visible also for $^{238}\text{Pu}^*$. For $^{240}\text{Am}^*$, the light- and heavy-fragment groups are not clearly distinguishable already at an excitation energy of \(\sim 30\) MeV, in contrast to $^{236}\text{Np}^*$ and $^{238}\text{Pu}^*$.

The FFMDs for 23 nuclides, $^{234–237}\text{U}^*$, $^{236–239}\text{Np}^*$, $^{238–241}\text{Pu}^*$, $^{240–243}\text{Am}^*$, $^{242–245}\text{Cm}^*$, and $^{244–246}\text{Bk}^*$, and their \(E^*\) dependence are systematically shown in Fig. 4. Here, the excitation-energy binning is chosen to be \(\Delta E^* = 10\) MeV as a compromise between the available statistics and a reasonable increment of \(E^*\). For the lowest energy, the range was set to 7.0–10.0 MeV. At this lowest energy, the FFMDs show a predominantly asymmetric shape for $^{235–237}\text{U}^*$, $^{236–239}\text{Np}^*$, $^{238–241}\text{Pu}^*$, $^{240–241}\text{Am}^*$. The double-peak structure of the FFMDs gradually smears out at higher excitation energies, and the shape becomes nearly mass-symmetric with a Gaussian-like distribution.

As can be seen in Fig. 4, there is no visible difference in the FFMDs among the studied...
Figure 4. Fission fragment mass distributions for $^{234-236}\text{U}$*, $^{236-239}\text{Np}$*, $^{238-241}\text{Pu}$*, $^{240-243}\text{Am}$*, $^{242-245}\text{Cm}$*, $^{244-246}\text{Bk}$* obtained in the multinucleon transfer channels of the $^{18}$O+$^{238}$Np reaction. Excitation energy of the compound nucleus is shown on the right-hand side.

Figure 5. (a) A conceptional view of MCF for the case of $^{240}\text{U}$*. (b) Calculated FFMDs for all the MCF steps up to 6th-chance fission (dashed curves) are shown for the initial compound nucleus $^{240}\text{U}$ at excitation energies 40–50 MeV. The sum of them is shown by the thin solid line. For comparison with the experimental data (open symbols with error bars), the calculated sum of FFMDs was broadened by the experimental mass resolution, the resulting FFMD is shown by the red thick curve.

isotopes at the low excitation-energy of $E^*$ =10–20 MeV. The isotope dependence, however, is evident at the relatively higher excitation energies of $E^*$ =30–40 MeV, where the peak-to-valley (P/V) ratio of the double-humped shape of the FFMD systematically changes. As the fissioning nuclei move to larger atomic numbers, the P/V ratio decreases, as can be seen by the smearing of the double-peak structure. For curium and berkelium isotopes, almost single Gaussian-like shape is found at $E^*$ =30–40 MeV. Concerning the isotope dependence for a certain element, the P/V ratio in this excitation-energy range increases with the mass of the fissioning nucleus, as can be seen for uranium, neptunium, plutonium and americium isotopes. These trends can be interpreted as due to the effects of multi-chance fission (MCF), i.e., fission after consecutive neutron evaporation from the initial compound nucleus, as explained below.
In order to discuss the effects of MCF on FFMDs, calculation based on the fluctuation-dissipation fission model [21] is introduced. In this model, the evolution of a nuclear shape, defined by three parameters (charge-center distance, mass-asymmetry and fragment deformation), is traced by solving the Langevin equations to the scission point, and the FFMD is obtained by accumulating different trajectories. The potential energy is defined as the sum of the liquid-drop part and excitation-energy ($E^*$)-dependent shell-correction energy, $\delta W(0)\exp(-E^*/E_d)$, where $\delta W(0)$ is the zero-excitation shell correction energy. The shell-damping energy ($E_d$) was chosen to 20 MeV as in [21].

As a first step in the present calculations, the MCF is not taken into account, which means that the FFMD is formed only by the initial excitation energy. The results are shown in Fig. 6, where experimental data for fission of uranium, neptunium and plutonium isotopes are taken from the MNT channels in the reactions of $^{18}$O+$^{232}$Th [18], $^{18}$O+$^{238}$U [19], and $^{18}$O+$^{237}$Np [22]. The results are shown by the blue curves in Fig. 6. Under this assumption, FFMDs for all the isotopes are reproduced only at the lowest excitation energy of $E^* \sim 20$ MeV. At the highest energy, the calculation shows structure-less symmetric fissions in contrast to the measurement, where experimental data shows the double peak-structure up to the highest energy region of 50 MeV.

Figure 5(a) is a conceptional view of MCF for the case of initial compound nucleus, $^{240}$U*. The highly-excited $^{240}$U* can decay either by the first-chance fission, or by a neutron emission, leading to the less-excited $^{239}$U. The residual excited nucleus can decay again either by fission (second-chance fission) or neutron evaporation; the higher-order chance fission successively occurs until the excitation energy drops below the fission barrier of the corresponding nucleus. Shape of the FFMD at each chance fission is also shown in this panel. Figure 5(b) shows the influence of MCF in the fission of $^{240}$U at the initial excitation energy 40–50 MeV. Each MCF component is shown by the dashed curve with different color, where the fraction of each chance fission is determined by the GEF code [23]. At each MCF, the potential energy surfaces for the respective compound nucleus was adopted. The sum of all the MCF yields, shown by the thin curve, are broadened with the experimental mass resolution as indicated by the thick solid curve. The calculation

3. Discussions

Figure 6. Experimental FFMDs (points with error bars) of the U, Np and Pu isotopes and their dependence on the excitation energy in the range of $E^* = 10–50$ MeV. The experimental FFMDs are compared with the Langevin calculations without (blue curves) and with (red curves) the inclusion of the multi-chance fissions.
well reproduces the measured FFMD properties, the mass asymmetry and the peak-to-valley ratio. The results of Fig. 5(b) clearly shows that the mass-asymmetric fission preserved at the high excitation energies originate from the 4th, 5th, and 6th chance fissions ($^{235,236,237}$U).

The same calculation procedure was applied for all the cases shown in Fig. 6, and the results are shown by red curves in Fig. 6. In contrast to the calculation without MCF (blue curves), the calculation with MCF well reproduce the experimental data, both for the mass-asymmetry and the P/V ratio. The calculation also demonstrates the decreasing peak-to-valley ratio of FFMDs for heavier elements (from uranium to plutonium), observed for example in the excitation energies 30–40 MeV, whereas the analysis without MCF predicts almost the same distributions through the isotopes. For a fixed element isotopes, the heavier isotopes gives larger P/V ratio, due to increasing number of neutrons emitted before fission. Consideration of MCF also validate that the shell effects responsible for mass-asymmetry in fission disappears around $E^* = 30–40$ MeV (blue curves in Fig. 6). It is evident that theoretical interpretations of FFMDs without the MCF consideration leads to a misinterpretation of the high-energy fission data. The effects are especially important for lighter-element and neutron-rich fissioning nucleus as the neutron emission probability becomes larger.

4. Conclusions
Even though MCF is a well-established concept in several fission observables (e.g., fission probability), so far its role for fission-fragment mass distributions has not been experimentally investigated in detail. This is mainly due to the absence of systematic data on the FFMDs in a large span of excitation energies and fissioning nuclides. We overcame this difficulty by exploiting the novel approach of multinucleon transfer reactions. Fission of a multitude of nuclides studied in a broad range of excitation energies has allowed us to show that the apparent asymmetric shape of FFMDs for a given initial excitation energy originates from fission of less excited lighter isotopes produced via a chain of MCF. In particular, this finding means that asymmetric shapes in the FFMDs measured at high excitation energies should no longer be interpreted as signatures of survival of shell effects in the initial compound nucleus, which would incite one to reexamine existing experimental data measured at high excitation energies. Ignoring multichance fission, the asymmetric structure of FFMD observed at high excitation energy would introduce an unexpectedly higher shell-damping energy than the conventional $E_d$-value of 20 MeV which was also used in this work. The shell-correction energy at high excitation energy is also important for other fields, for example, heavy-ion fusion reaction for the synthesis of superheavy elements. This is because only the shell-correction energy forms the fission barrier of a compound nucleus the height of which significantly alters its survival probability in the competition between neutron evaporation and fission.

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