Systematic Studies of Asymmetric Mass Distributions in Proton-induced Fission of Actinides

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Received: March 22, 2002; In Final Form: May 11, 2002

In order to clarify the relationship between a mass distribution and shell structure of the fission fragments in nuclear fission, the fragment mass and the kinetic energy distributions in the proton-induced fission of plutonium isotopes, ^{239,242,244}Pu, were precisely measured using a double time-of-flight system. It was found that the position of the light side of the heavy asymmetric mass distributions shifts to a larger fragment mass number according to the neutron-proton ratio, N_f/Z_f, of the fissioning nucleus. The result is qualitatively explained by the change of the most probable mass number of fission fragments for $Z = 50$ proton shell.

1. Introduction

In the low-energy fission of actinides, particularly in the thermal-neutron-induced fission and the spontaneous fission, a number of studies of the mass distributions have been performed for a long time. Comparing these distributions,[1] it can be seen that the peak of the heavy asymmetric mass distribution locates at $A \sim 140$ in any system and that the position of the light peak shifts with the mass number of the fissioning nucleus. These features have been qualitatively explained by the fragment shells of $Z = 50$ and/or $N = 82$ and the deformed shell of $N = 88$. Until now, however, direct evidence of the shell effects has not been presented.

Recently, in the low-energy proton-induced fission of actinides, it has been demonstrated that there exist at least two independent deformation paths for fission process; one leads to a symmetrically elongated scission configuration, and the other leads to a compact scission configuration with reflection asymmetry.[2] From the detailed analysis with respect to the asymmetric mass distributions in the proton-induced fission of ^{232}Th, ^{238}U, ^{244}Pu, and ^{248}Cm, it has been found that the position of the light side of the heavy asymmetric mass distributions converges on $A = 126$–128, whereas the heavy side of those distributions broadens with the increase of fissioning nuclear masses.[3] These trends are consistent with the assumption that the heavy fragments of $Z = 50$ or the light fragments of $N = 50$ are produced preferentially.

If the systems have different neutron to proton ratios, N_f/Z_f, the fragment mass number corresponding to $Z = 50$ and $N = 50$ should change with the ratios under the unchanged charge distribution (UCD) hypothesis. Therefore, it is expected that the features of the asymmetric mass distribution will be affected by the N_f/Z_f ratios. On the basis of this expectation, the mass distributions in the proton-induced fission of uranium isotopes, ^{233,235,238}U, have been studied previously.[4] As a result, it was found that the shell effects of fission fragments, particularly at $Z = 50$, affect the mass division in the nuclear fission. To study on this matter more systematically, in this work, the mass distributions in the proton-induced fission of ^{239,242,244}Pu were measured, and the influence of the shell effects of the fragments on the mass division was discussed.

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2. Experimental

A proton beam with energy of 13 MeV was supplied from the JAERI (Japan Atomic Energy Research Institute) tandem accelerator. An average beam current was 1.0–1.5 μA. The ^{239,242,244}Pu ($\sim$60 μg cm^{-2}) targets were prepared by electrodeposition on 0.1 μm Ni foils. Fission fragment velocities were measured by the double time-of-flight (double TOF) system. The start and stop detectors of the first telescope (TOF1) placed at $\theta_{lab} = 50°$ were composed of a gold-evaporated thin organic film and a microchannel plate (MCPD). A Si surface barrier detector (SSD) located behind the stop MCPD was used for the stop and energy signals. The second telescope, TOF2, was set around $\theta_{lab} \sim 128°$ at the opposite side of the beam direction in order to detect complementary fragments. The start and stop signals of TOF2 were taken by a MCPD and a thin (1 mm) plastic scintillator, respectively. The flight paths of the telescopes were about 96 and 110 cm and their solid angles, 0.23 and 4.6 msr, respectively. More than 2×10^5 fission coincidence events were accumulated in each fissioning system. Detector calibration was performed by using 230-MeV ^{127}I beam on calibration targets, ^{89}Y, ^{nat}Ag, ^{nat}In, ^{141}Pr, and ^{159}Tb, which have atomic and mass numbers in the regions of the fission products. Each target of them was made by vacuum evaporation on 30 μg cm^{-2} thick carbon foil, and each thickness was 30–40 μg cm^{-2}. The time-of-flight and the energy of the elastically scattered iodine ions and the recoiling target nuclei were measured for calibration. A time resolution of approximately 600–700 ps was achieved and the resulting overall resolution of mass and total kinetic energy (TKE) were estimated to be $\Delta M \sim 2.0$ amu and $\Delta TKE \sim 2.5$ MeV, respectively, which include the effects of energy loss of the particles passing through the target, the backing foil, and the foils of MCPDs.

3. Results and Discussion

Based on the laws of the momentum and the mass conservation in the nuclear fission, the primary mass (A_1) and the TKE of each fission fragment were evaluated from the measured velocities of a pair of fragments on the assumptions that no neutron was emitted from the compound nucleus prior to fission and that neutrons were isotropically emitted from primary fragments and had no influence on the initial fragment velocity on the average. The fragment mass and the total kinetic energy were calculated

Published on Web 6/30/2002

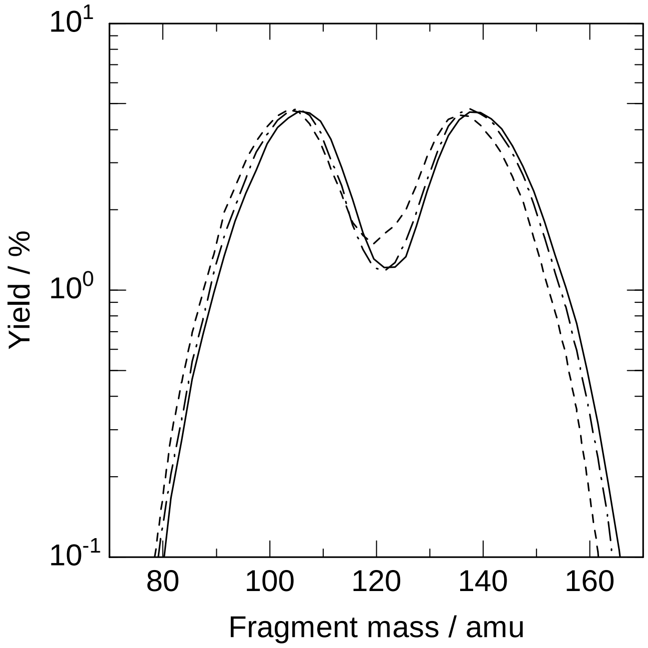

Figure 1. Fragment mass distributions in the 13-MeV proton-induced fissions of ^{239}Pu (dashed line), ^{242}Pu (dot-dashed line), and ^{244}Pu (solid line).

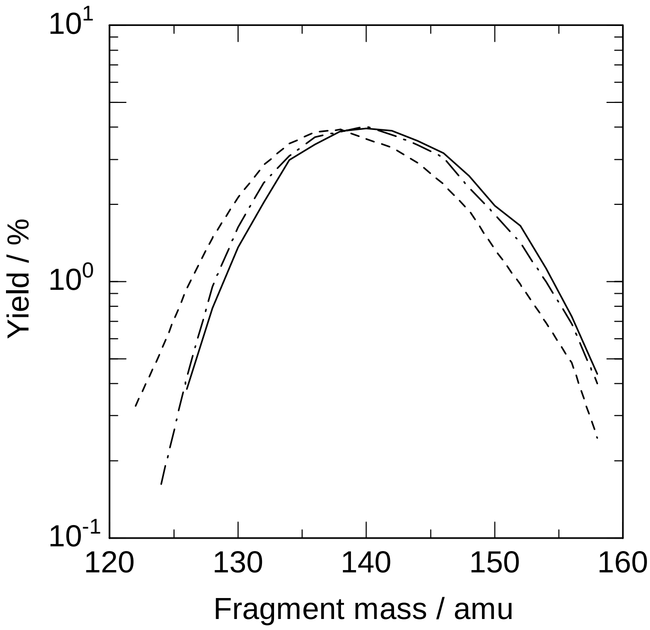

Figure 2. Decomposed heavy asymmetric mass distributions for the 13-MeV proton-induced fissions of ^{239}Pu (dashed line), ^{242}Pu (dot-dashed line), and ^{244}Pu (solid line).

by the following equations:

$$A_1 = A_f \frac{1}{1 + v_{1,\mathrm{cm}}/v_{2,\mathrm{cm}}} , \tag{1}$$

and

$$TKE = \frac{1}{2} A_f \, v_{1,\mathrm{cm}} \, v_{2,\mathrm{cm}} , \tag{2}$$

where $v_{1,\mathrm{cm}}$ and $v_{2,\mathrm{cm}}$ are the velocities in the center-of-mass system for the complementary fragments A_1 and A_2, respectively, and A_f is the mass of the fissioning nucleus with a relation of $A_f = A_1 + A_2$.

The obtained mass distributions are demonstrated in Figure 1. A general feature of the mass distribution in asymmetric fission, as the mention above, is that the position of the light peak changes with the mass number of fissioning nucleus, whereas that of the heavy peak remains approximately constant. However, a detailed examination for the present fissioning systems shows that the heavy peak slightly shifts to the heavy mass side

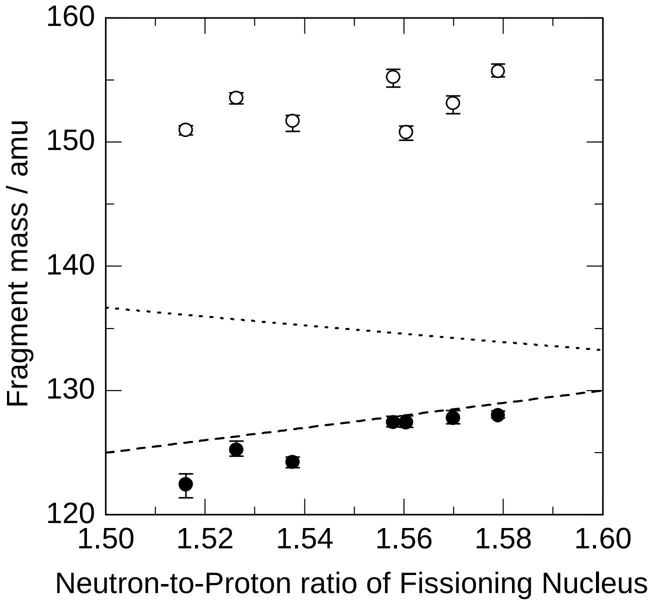

Figure 3. Correlation between N_f/Z_f of the fissioning nucleus and the mass number at the "one-fifth" yield of the peak for the heavy asymmetric mass distribution. Closed symbols represent the position of the light side, and open symbols the heavy side. Expectation from the UCD hypothesis for $Z = 50$ (dashed line) and $N = 82$ (dotted line) is also shown.

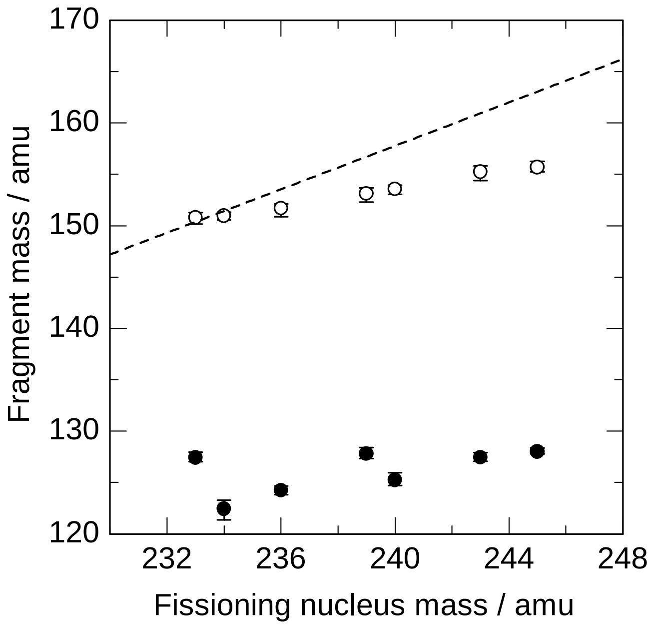

Figure 4. Correlation between the mass of fissioning nucleus and the mass number at the "one-fifth" yield of the peak for the heavy asymmetric mass distribution. Closed symbols represent the position of the light side, and open symbols the heavy side. Expectation from the UCD hypothesis for the complementally light fragment of $N = 50$ (dashed line) is also shown.

with the increase of the mass number of fissioning nucleus. To examine the features of the asymmetric mass division more precisely, it is necessary to subtract the component corresponding to symmetric mass division from the total mass distribution, although the contribution from the symmetric fission is not expected to be large in the the present systems as seen in the figure. The two component analysis in the TKE distribution for each fragment mass was done according to Reference 4.

Figure 2 shows the heavy asymmetric mass distributions which resulted from the two component analysis. It was found that the position of the light side of the heavy peak shifts to the heavy mass side with increasing A_f, the same as the results in the system of uranium isotopes. To discuss this trend quantitatively, the mass number at the "one-fifth" yield of the peak for both sides of the heavy peak was plotted against N_f/Z_f and A_f of the fissioning nucleus for all systems including the system studied previously, $p + {}^{232}$Th and $p + {}^{233,235,238}$U (Ref. 4).

The results were depicted in Figures 3 and 4. The errors were come from the ambiguity in the fit of TKE distributions for the two component analysis. From these figures, it was found that the position of the light side of the heavy peak shifts certainly with increasing N_f/Z_f although the data disperse somewhat, while that of the heavy side shifts depending on the mass number of fissioning nucleus, A_f.

In Figure 3, for the light side of the heavy asymmetric mass distribution, the dotted and dashed lines denote the trend of the mass number expected from the UCD hypothesis for $N = 82$ and for $Z = 50$, respectively. It was demonstrated in Figure 3 that the position of the light side of the heavy peak follow the most probable mass of $Z = 50$. On the other hand, as shown in Figure 4, the position of the heavy side shifts differently from the expectation of UCD. Namely, the heavier the fissioning nucleus mass, the larger the difference. This indicates that the yield of the light fragment with $N = 50$ decreases with A_f. Therefore, the shell effect of $N = 50$ in the asymmetric fission is not so strong as that of $Z = 50$.

4. Conclusions

To study the correlation between the fragment mass distributions and the shell structures of fission fragments, the fragment mass and the energy distributions in proton-induced fissions of plutonium isotopes, ^{239,242,244}Pu, were precisely measured using a double time-of-flight method.

Obtained fragment mass distributions were decomposed into symmetric and asymmetric mass division components by analyzing the total kinetic energy distributions. The results indicate that the position of the light side of the heavy asymmetric mass distribution shifts to the heavy mass side with the N_f/Z_f value of the fissioning nucleus. This trend is qualitatively explained by the change of the most probable mass number of fission fragments with the $Z = 50$ proton shell.

On the other hand, the position of the heavy side of the heavy peak depends not on N_f/Z_f but on the fissioning nucleus mass A_f. However, the positions deviate from the expectation of UCD with A_f. This indicates that the shell effects of $N = 50$ for fission fragments are not so strong as that of $Z = 50$.

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