## Fragment mass dependence of angular anisotropy in 15 MeV proton-induced fission of $^{244}\mathrm{Pu}$

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**Abstract.** Angular distributions of fission fragments with mass number A=97-159 have been measured by the radiochemical recoil-catcher method in the proton-induced fission of  $^{244}$ Pu with the incident energy of 15 MeV. Angular anisotropies of extreme asymmetric mass division products even up to the fragment mass ratio of  $A_H/A_L \sim 1.85$  are found not any different from those of the typical asymmetric mass division products with  $A \sim 138$ , which indicates that no clear evidence is observed for the existence of an additional saddle point configuration in the extreme asymmetric mass division. The correlation between the saddle point state evaluated from the angular anisotropy and the mass division mode is discussed.

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The correlation between the saddle point state and the mass division mode has been studied by observing angular distributions of fission fragments as a function of fragment mass number and it is found that the fragment anisotropy is quite different for the typical symmetric and the asymmetric mass division products in the low-energy particleinduced fission of light actinides [1–6]. In the previous work [3], the two kinds of angular anisotropies obtained in the 15 MeV proton-induced fission of  $^{232}$ Th were interpreted by the existence of two kinds of saddle point nuclear temperatures  $(T_{saddle})$ . The analysis took into account the contributions of the multiple chance fission in the asymmetric and symmetric mass division modes but it assumed the same saddle shape  $\Im_{eff}$ , effective moment of inertia, for both the symmetric and asymmetric saddle configurations. The result indicated that the excitation energy at the saddle point becomes higher for the symmetric mass division than that for the asymmetric one: high- $T_{saddle}$  for the symmetric and low- $T_{saddle}$  for the asymmetric mass division.

On the theoretical side, a reflection asymmetric saddle was first pointed out to be energetically more favored than the symmetric one at the 2nd saddle [7,8], and its possible connection with the final mass division inferred from the shape of the deformed nucleus near the saddle was calculated by both static potential and dynamical paths [7,9]. The aim of the present work is to further the understanding of the correlation between the saddle point state evaluated from the fragment angular anisotropy and the mass division mode by using the 15 MeV proton-induced fission of medium actinide target <sup>244</sup>Pu. The angular anisotropies of the products up to  $A_H/A_L \sim 1.85$  are also measured to examine the existence of an additional fission mode in the extreme asymmetric mass division, where  $A_H$  and  $A_L$  denote heavy and light fragment mass number.

The enriched <sup>244</sup>Pu (97.867%) target was prepared by electrodeposition onto a 2.7 mg/cm<sup>2</sup> thick aluminum foil [10]. The thickness of the target estimated by  $\alpha$ spectrometry was 110  $\mu$ g/cm<sup>2</sup>. Angular distributions of fission fragments were measured by the recoil-catcher technique. An aluminum catcher foil of 4.9 mg/cm<sup>2</sup> thickness was mounted over the angular ranges of 90° to 177° on the innerwall of the cylindrical irradiation chamber having a length of 60 mm and an inner diameter of 70 mm. The target was placed at the center of the chamber.

Bombardment was performed for about 10 h at the JAERI (Japan Atomic Energy Research Institute) tandem accelerator using the proton beam with the incident energy of 15 MeV. The beam was collimated by a tantalum collimator having a diameter of 3 mm, placed at the entrance of the chamber. The beam current, monitored

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**Table 1.** Angular anisotropies of the products obtained in the 15 MeV proton-induced fission of  $^{244}$ Pu

Nuclide	Anisotropy $(^{244}Pu)$
$^{97}\mathrm{Zr}$	$1.29 \pm 0.05$
$^{99}Mo$	$1.32 \pm 0.04$
$^{103}\mathrm{Ru}$	$1.29 \pm 0.02$
$^{105}$ Rh	$1.26 \pm 0.03$
$^{112}\mathrm{Pd}$	$1.18\pm0.03$
$^{127}\mathrm{Sb}$	$1.23 \pm 0.04$
$^{128}\mathrm{Sb}$	$1.22 \pm 0.03$
$^{129}Sb$	$1.22\pm0.03$
$^{131}I$	$1.22 \pm 0.03$
$^{132}$ Te	$1.26 \pm 0.03$
$^{133}I$	$1.30\pm0.03$
$^{135}I$	$1.31 \pm 0.06$
$^{140}Ba$	$1.33\pm0.03$
$^{141}\mathrm{Ce}$	$1.31\pm0.07$
$^{142}$ La	$1.29\pm0.04$
$^{143}\mathrm{Ce}$	$1.35\pm0.04$
$^{147}$ Nd	$1.31\pm0.06$
$^{149}$ Nd	$1.32\pm0.03$
$^{151}$ Pm	$1.26\pm0.04$
$^{153}Sm$	$1.30\pm0.04$
$^{156}$ Sm	$1.26\pm0.04$
$^{157}$ Eu	$1.32\pm0.06$
$^{159}$ Gd	$1.31\pm0.07$

with a Faraday cup connected to a current integrator, was typically 1.5  $\mu \mathrm{A}.$ 

After the bombardment, the aluminum catcher foil was removed from the irradiation chamber and cut into strips of appropriate widths that correspond to the angular width  $\Delta\theta$  around the mean angle  $\theta$  with respect to the direction of the proton beam. The relative radioactivity of each nuclide at a specified angle was determined through the  $\gamma$ -ray spectrometry. Low yield rare earth nuclides produced by extreme asymmetric mass division were chemically separated from other fission products with the computer-controlled rapid ion-exchange separation system [11] before  $\gamma$ -ray measurements. Details of the separation procedures have been reported elsewhere [12].

Fragment angular distributions  $W(\theta)$  were fitted to the equation  $W(\theta) = a + b \cos^2 \theta$  by the weighted least squares analysis. The angular anisotropy of each product is defined as  $W(180^{\circ})/W(90^{\circ})$ , i.e., (1 + b/a), and is given in Table 1. Products in the symmetric region of  $A \sim 122$  were not observed in the present study because of no appropriate nuclides available for radiochemical measurements. The correlation between the angular anisotropy and the mass distribution is shown in Fig. 1 as a function of the heavy fragment mass number. The mass distribution of primary fragments obtained by a double velocity time-offlight method for the 15 MeV proton-induced fission of  $^{244}$ Pu [13] is shown by the solid line in Fig. 1b.

The anisotropies of the asymmetric mass division products are essentially the same and lie within the error limit of  $W(180^{\circ})/W(90^{\circ})_{asym} = 1.30 \pm 0.04$  (average of 15 nuclides with  $A \leq 103$  and  $A \geq 135$  shown in Table 1). It



**Fig. 1. a** Angular anisotropy of the fission products as a function of heavy fragment mass number for the 15 MeV protoninduced fission of <sup>244</sup>Pu. The *dashed line* shows the deduced anisotropy at the symmetry (see text). **b** The *solid line* shows the mass distribution for the 15 MeV  $p+^{232}$ Pu fission taken from [13]. The decomposed mass yields obtained by Zhao et al. [13] are shown by the *long-dashed* and the *dashed lines* corresponding to the compact and elongated scission configurations, respectively

is found that the anisotropies observed for the extremely asymmetric mass division products are not any different from the average value of the other asymmetric mass division products. The recent precise measurements on the angular anisotropy in the 15 MeV proton-induced fission of <sup>232</sup>Th confirmed the constancy of the anisotropy value for the extreme asymmetric mass division products up to  $A_H/A_L \sim 2$  [14]. Thus, we can conclude that even the extreme asymmetric mass division experiences the same saddle point in the course of deformation as other fission events that lead to the typical asymmetric mass division  $(A \sim 140)$ .

It is seen that the anisotropies of the products with mass number around A=130 are smaller than those of the asymmetric mass division products with  $A \ge 140$  as shown in Fig. 1a, as also reported in the 15 MeV proton-induced fission of  $^{232}$ Th [3]. In the latter fission, the intermediate values have been interpreted as the result of mixing of the symmetric and asymmetric mass division modes [14]. In [13] the mass yield curve observed by the double TOF method for the proton-induced fission of  $^{244}$ Pu was decomposed in the mass region around A=130 from the intensity ratio of the high TKE to the low TKE as shown in Fig. 1b. Using the relative degree of contribution of the two compo

nents, the anisotropy values for symmetry were calculated to be  $W(180^{\circ})/W(90^{\circ})_{sym} = 1.14 \pm 0.6$  in which the error includes the ambiguity arisen from the mass difference between the secondary mass (post-neutron) in Fig. 1a and the primary mass in Fig. 1b.

In summary, the angular distributions of the fission products were measured by the radiochemical recoilcatcher technique in the proton-induced fission of <sup>244</sup>Pu with the incident energy of 15 MeV. The angular anisotropies of the asymmetric mass division products were found about the same even up to such extreme mass division as  $A_H/A_L \sim 1.85$ . This observation indicated that the extreme asymmetric mass division experiences the same saddle point configuration as the typical asymmetric mass division with no special saddle point state. From the angular anisotropy data, the existence of two kinds of saddle point configurations for each symmetric and asymmetric mass division mode was also confirmed for the protoninduced fission of <sup>244</sup>Pu.

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