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Supersymmetric fission at intermediate energy and production of neutron-rich nuclei with $A < 80$

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Abstract

The yields of neutron-rich Ni, Cu, Zn, Ga and Ge-isotopes were measured in 25 MeV proton induced fission of ^{238}U using the ion guide-based isotope separator technique. The results indicate enhancement for supersymmetric mass division at intermediate excitation energy of the fissioning nucleus and show the potential of this reaction for the production of neutron-rich exotic nuclei around $Z = 28$. © 1997 Elsevier Science B.V.

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Keywords: Nuclear fission; Production yields; Exotic nuclei

1. Introduction

A great deal of interest in the investigation of very asymmetric fission is connected with the search for the supersymmetric fission mode, and clarifying the perspectives to produce exotic neutron-rich nuclides. Information on extreme asymmetric fission of heavy nuclei, with the light fragment mass $A_L < 80$, has been scarce until recently, mainly due to very low yields. The supersymmetric mass division in the spontaneous fission of ^{252}Cf was reported for the first time in Ref. [1] and confirmed in the work of Ref. [2]. Later, interesting results were obtained in the region of very light fragment masses in thermal neutron-induced fission of ^{235}U [3], ^{239}Pu and ^{249}Cf [4]. The appearance of shell effects and odd-even variations in the

nuclear charge distribution with increasing asymmetry of the mass split was demonstrated for thermal neutron-induced fission. A shoulder in the mass yield curve near mass 70 had earlier also been observed in the reactor-neutron-induced fission of ^{238}U [5]. Recently, the element distribution for fragments with Z between 28 and 46 was measured in projectile-like fission reactions of ^{238}U at relativistic energies on Pb and Be targets [6].

The existence of a supersymmetric fission mode can, in principle, be inferred from the multimodal nature of nuclear fission [7]. In Ref. [7] this mode was introduced in the case of spontaneous fission of ^{252}Cf to explain the peculiarities in fission fragment mass and kinetic energy distributions at $A_L/A_H \approx 74/176$. A supersymmetric component in the primary fragment mass distribution was included in the theoret-

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Fig. 1. A gamma-ray spectrum recorded at the mass number $A = 78$ in coincidence with a thin plastic beta-detector. Only the short-lived ^{78}Zn ($T_{1/2} = 1.47\text{ s}$) and ^{78}Ga ($T_{1/2} = 5.12\text{ s}$) are seen because the implanted activity on the collection tape was moved away every 1.5 s.

ical model of Refs. [8,9] for calculating mass and independent fission product yields at intermediate energies. In this model the superasymmetric division is determined by the nuclear shells at $Z = 28$ and $N = 50$ and by the tail of the second standard asymmetric component and the symmetric component, whose contribution is changing with excitation energy of the fissioning nucleus.

The experimental data for fragment charge and mass distributions for $A_L < 80$ are necessary for determining the parameters describing the superasymmetric mode in the model [9]. The fission of heavy nuclei at moderate excitation energies can be used for the production of exotic nuclei, including the very rare ^{78}Ni [10]. For planning the experiments for the production and investigation of neutron-rich exotic nuclei at or below $Z = 28$, it is crucially important to characterize the properties of the superasymmetric fission mode. In this paper we wish to report a study on charge and mass distributions in highly asymmetric proton-induced intermediate-energy fission of ^{238}U . The experiment was performed using the ion guide isotope separation technique IGISOL [11,12]. The experimental results are analyzed in the framework of a new version of the theoretical model proposed earlier in Refs. [8,9].

2. Method and experimental results

The mass and charge distributions of fission products from 25 MeV proton-induced fission of ^{238}U were obtained from the independent yields extracted from the measured cumulative production yields, which were determined from the radioactivity of the mass separated nuclides. Fragments following the fission of the Np compound nuclei were slowed down as ions in high-pressure helium and transported by the flow through a differentially pumped electrode system into the acceleration stage of the mass separator. The target and the ion-guide systems used in this study are described in Ref. [12]. In order to maximize the production rate the 15 mg/cm² thick uranium-target foil was tilted to a small angle with respect to the beam axis, providing a ten-fold increase in the production rate of primary fission fragments. Radioactive ions of 40 keV energy were mass-analysed with a resolving power of 350 and implanted into a collection tape, which was directly viewed by a set of beta- and gamma-ray detectors. Typically the energy spectra were recorded in a saturation mode without moving the activity away from the implantation/counting position. In some cases the collected and built-up activities were moved away in regular short time intervals to enhance the sensitivity of detection for the rarest n -rich isotopes. The delay time of the ion-guide based

Table 1

Independent and cumulative fission product yields in atoms/s (errors in parentheses) for $Z = 28$ –32 and $A = 71$ –82 as measured at the focal point of the IGISOL separator. In the last column the elements which were used to determine the cumulative yields are indicated. The yields were measured in the reaction $25 \text{ MeVp} + {}^{238}\text{U}$ at a proton beam current of $3.0 \mu\text{A}$. The average masses, standard deviations and elemental yields were obtained by fitting with a Gaussian function

A	Ni	Cu	Zn	Ga	Ge	Cum. yield	element
71	5.7(17)	1.4(11)				7.1(8)	Cu
72		15(3)				15(3)	Cu
73		10(3)				10(3)	Cu
74		9(3)	16(5)			25(4)	Zn
75			69(8)			69(8)	Zn
76			55(5)	29(10)		84(8)	Ga
77			27(4)			155(9)	Ge
78			11(3)	92(12)	140(30)	320(60)	As
79			3.1(6)	96(17)	200(30)	460(60)	As
80				26(3)	234(20)	540(60)	As
81				13(5)	350(40)	790(60)	As
82					135(20)	1530(200)	As
A_{av}		72.7(6)	75.5(1)	78.2(2)	80.4(4)		
σ_A		1.2(7)	2.0(3)	1.3(2)	1.7(6)		
$Y(Z)$		40(18)	177(20)	330(50)	1200(300)		

mass separation is only of the order of ms. This allows the detection of the shortest-lived β -radioactivities of all elements without any decay losses. Due to the fact that primary thermalized ions are initially contained in and transported by the inert helium gas, no chemical selectivity influences the transport. The total efficiency of the ion guide depends on the kinematics of the reaction involved. For example, the light fission fragment is stopped less efficiently than the heavy one. However, in this work the yield measurements were performed for the fragments in the narrow mass range with masses between 70 and 80. Thus, within the overall experimental accuracy, no corrections for kinematic effects were performed.

The whole measurement series of the yields were performed with a constant proton-beam current of $3 \mu\text{A}$. The efficiency of the whole system was regularly monitored by the yield of ${}^{112}\text{Rh}$, which was typically 5000 ions/s. An example of a gamma-ray spectrum recorded at mass 78 in coincidence with a thin plastic beta-detector is shown in Fig. 1. The independent and cumulative yields extracted from similar measurements between $A = 71$ and 82 are shown in Table 1. The energies and branchings of the transitions used in extracting the yields from the measured spectra were obtained for $A = 74, 76$ from

Refs. [13, 14], and for the other masses from Ref. [15], except for ${}^{71}\text{Ni}$, which is reported in Ref. [16]. In the case of ${}^{71}\text{Cu}$ and ${}^{71}\text{Ni}$ the branchings have not yet been determined and the yields extracted represent the lower limit only. It is interesting to note that the drop in the yield from $A = 80$ down to about $A = 70$ is less than two orders of magnitude. This is drastically different from the one observed for the thermal neutron-induced fission [3], where the corresponding decrease in yield is about four orders of magnitude.

3. Discussion

The experimental results were analysed in the framework of a theoretical model proposed in Refs. [8, 9] and used here in a more advanced version. In this model the primary mass and charge distributions of the fission fragments and their excitation energies are calculated at the first step and then the process of emission of neutrons from excited fragments is considered. The influence of nuclear shells and charge polarization and their dependence on excitation energy of the compound nucleus are taking into account. In comparison with the previous version we introduced odd-even effects in the charge and mass

distributions and slightly changed the model parameters taking into account the new experimental data on very asymmetric fission. The formation cross section of a fission product with mass number A and charge number Z can be expressed in the form

$$\sigma_f(A, Z) = \sum_{A_c Z_c} \int Y_{\text{ind}}(A, Z, A_c, Z_c, E_c) dE_c \times \frac{d\sigma_f(A_t, Z_t, A_p, Z_p, E_p, A_c, Z_c, E_c)}{dE_c}, \quad (1)$$

where the subscripts t , p and c refer to target, projectile and compound nuclei, respectively, $d\sigma_f/dE_c$ is the partial fission cross section of the compound nucleus at the excitation energy E_c for different fission chances over which the summing is carried out and $Y_{\text{ind}}(A, Z, A_c, Z_c, E_c)$ is the independent yield. The independent yields Y_{ind} are defined as the yields of fission products after light particle emission from excited primary fragments. Here we shall consider only neutron emission because at the excitation energies up to about 100 MeV, the emission of protons and other charged particles is negligible with respect to the precision of available experimental data.

At low excitation energies, the primary fission fragment mass and charge distributions exhibit odd-even staggering. The primary distributions are presented in the factorisation form

$$P_{\text{pre}}(Z) = \tilde{P}_{\text{pre}}(Z) F_{\text{oe}}(Z), \quad (2)$$

$$Y_{\text{pre}}(A) = \tilde{Y}_{\text{pre}}(A) F_{\text{oe}}(A),$$

where $\tilde{P}_{\text{pre}}(Z)$ and $\tilde{Y}_{\text{pre}}(A)$ are smoothed distributions, and functions $F_{\text{oe}}(Z)$ and $F_{\text{oe}}(A)$ describe odd-even staggering. The calculation method of smoothed pre-neutron emission charge and mass distribution of fission fragments is described in Ref. [9]. The smoothed pre-neutron emission isobaric chain charge distribution is approximated by a Gaussian distribution, therefore odd-even structure can be described by the parameter defined as a third difference of the natural logarithms of the fractional yields [17]. We consider the proton and neutron odd-even effect separately and one can write

$$F_{\text{oe}}(Z) \sim \exp((C_p^1 + C_p^2) \delta_Z(A_c, Z_c, E_c)), \quad (3)$$

where C_p^1 and C_p^2 are defined by the parity of the proton number in the two fragments ($C_p = 1$ if Z is even

Fig. 2. Measured independent yields (points with error bars) and calculated mass yields (open circles) of fission products in the supersymmetric mass region in 25 MeV proton induced fission of ^{238}U , and the mass yields (solid circles) in the thermal neutron induced fission of ^{235}U from Ref. [3]. The theoretically calculated isotopic distributions are shown by thin solid and dashed lines.

and $C_p = -1$ if Z is odd). The proton odd-even difference parameter $\delta_Z(A_c, Z_c, E_c)$ is parameterised as a function of excitation energy, charge and mass number of compound nuclei in accordance with experimental data. Odd-even staggering in the primary mass distribution is described by the combination of proton and neutron odd-even effects. The proton and neutron odd-even difference parameters are taken to be proportional, i.e. $\delta_N(A_c, Z_c, E_c) = c \delta_Z(A_c, Z_c, E_c)$ ($c < 1$).

The independent fission product cross sections and the mass yield curves were calculated for the proton induced fission of ^{238}U and compared with the experimental data. The experimental yields, shown as points with error bars, for Ni, Cu, Zn, Ga and Ge isotopes are presented in Fig. 2. The calculated isotopic distributions as well as the derived mass yields, shown as open circles, are also shown in Fig. 2. Theoretical mass yields were obtained as the ratio of the calculated cross sections and the total fission cross section with 25 MeV proton energy ($\sigma_f = 1340$ mb). The experimental yields were obtained in units of atoms/s,

therefore they have to be normalized. The normalisation constant for each Z -chain of fission products is determined from the comparison of the sum of the measured independent yields to the calculated one for the same set of isotopes. The averaged normalisation constant of $C_{\text{norm}} = 0.02 \text{ mb}/(\text{atoms/s})$ was used for all Z -chains presented in Fig. 2. For comparison, the mass yields in the thermal neutron induced fission of ^{235}U from Ref. [3] is shown as solid circles. One can see that the yields in the supersymmetric mass region for fission of Np compound nuclei at intermediate excitation energy differ drastically from the ones in low energy fission of ^{236}U compound nuclei. In a very asymmetric mass division at intermediate excitation energy ($E_c \geq 20 \text{ MeV}$) there are contributions from three components: the tails from the symmetric and the second asymmetric modes and the supersymmetric mode. Our observation of the enhancement of the very asymmetric mass division at 25 MeV proton energy is supported also by the results reported in Ref. [16] for the yield of ^{71}Ni in fission of ^{238}U by 65 MeV protons.

Experimental elemental yields were obtained by Gaussian fitting of the independent yields for the isotope chains of Cu, Zn, Ga and Ge. Due to the lack of experimental data, the value for Ni was estimated using the experimental yield of ^{71}Ni only and the theoretical value of the average mass, $A_{\text{av}} = 71.23$ and the mass dispersion $\sigma_A = 1.47$, for the Ni-chain. The experimental (solid squares) and theoretical (solid line) elemental yields for Ni, Cu, Zn, Ga and Ge in 25 MeV proton induced fission of ^{238}U are shown in Fig. 3. The elemental yields in low energy fission of ^{238}U was measured with relativistic U-projectiles on a Pb-target [6] are presented in Fig. 3 for comparison. There is a reasonable agreement between the three data sets for supersymmetric fission. There is a deviation between the theoretical curve and the data from Ref. [6] for $Z > 32$, which may be due to the fact that the fissioning compound nucleus and the excitation energies are different. In particular, the theoretical Z -curve for 25 MeV proton-induced fission of ^{238}U did not exhibit odd-even staggering although the model reproduces odd-even differences observed in thermal neutron-induced and spontaneous fission.

From the comparison between the experimental yields and theoretical calculations we conclude that the contribution of the supersymmetric fission mode

Fig. 3. Measured elemental yields of fission products (solid squares) in comparison with theoretical predictions (solid line) in 25 MeV proton induced fission of ^{238}U ; the data for low energy fission of ^{238}U from Ref. [6] are shown by the dashed line.

near the nuclear shells $Z = 28$ and $N = 50$ is significant at intermediate excitation energies. Enhancement of the yields in the very asymmetric mass region proves that fission of heavy nuclei at intermediate excitation energy is a potential tool for the production of neutron-rich nuclei with $A < 80$. This has been recently confirmed in spectroscopic measurements of $A = 70$ nuclei using the improved ion guide system [18]. The maximum supersymmetric cumulative yields observed with a $20 \mu\text{A}$ beam current were as high as 100 ions/s for ^{70}Cu and 7000 ions/s for ^{80}As . This observation for the yield ratio between $A = 70$ and $A = 80$ is in a good agreement with the mass yield curve of Fig. 2 and the cumulative yields of Table 1. Moreover the analysis of the Z -dependence of the differences between the calculated average mass number of fission products and the mass number for the heaviest stable isotope shows that in the very asymmetric light mass region we can reach further from the most neutron-rich stable isotopes than in the other mass regions. Relying on our satisfactory description of the isotopic distributions shown in Fig. 2 we estimate the production cross section of ^{78}Ni in the

reaction under consideration to be 2.8 nb. This value is higher than that obtained in the work of Ref. [6].

4. Conclusion

Independent yields in 25 MeV proton induced fission of ^{238}U have been measured in the very asymmetric mass region $A = 71\text{--}82$ and $Z = 28\text{--}32$ by the on-line mass separator IGISOL. The enhancement of highly asymmetric mass and charge division in comparison with thermal neutron induced fission is observed. A comparison of the theoretical and the experimental yields supports the hypothesis of the superasymmetric fission mode at intermediate excitation connected with $Z = 28$ and $N = 50$ shells. To make this assertion more reliable, additional investigations are needed in particular on the dependence on the beam energy. One can conclude from the results obtained that the proton induced fission of heavy nuclei at intermediate energy is a promising tool for the production of exotic neutron-rich nuclides with $A < 80$.

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