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YIELDS OF TERNARY PARTICLES FROM THE REACTIONS ^{229}Th(n_{th},f), ^{233}U(n_{th},f) AND ^{239}Pu(n_{th},f)*

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Ternary particles (TP's) from the thermal neutron induced fission of ^{229}Th, ^{233}U and ^{239}Pu have been studied. Their energy distributions and yields have been determined using the mass separator Lohengrin at the high flux reactor of the Institut Laue-Langevin (ILL). Special emphasis was put on the search of the heaviest ternary particles. As a new result it has been found that the mass and/or charge spectra of the heaviest TP's at the limit of detectability of present techniques are strongly correlated with the mass and/or charge of the fissioning compound nucleus. The actual limit of detection reached is about 1 TP in 10^{10} fission reactions. At this limit, in Th-fission the heaviest TP's are Ne-isotopes, while for ^{239}Pu-fission the observable TP's are shifted to Si-isotopes. The energy distributions and yields of several TP's having so far not been reported from the reactions ^{229}Th(n_{th},f), ^{233}U(n_{th},f) and ^{239}Pu(n_{th},f) are given. The dependence of the yields of the TP's on the fissility of the fissioning compound nucleus for actinides ranging from Th to Cf is shown. Experimental yields are compared to predictions in the framework of the Halpern model.

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1 Introduction

Ternary fission is a process where at scission of the disintegrating nucleus three particles are formed. First evidence for such a process was found more than fifty years ago [1, 2]. In thermal neutron induced fission ternary particles are produced in about two out of thousand fissions. In these processes one obtains two fission fragments and one light particle, the ternary particle (TP). The TP is in about 92% of these cases an α-particle. Particles with a nuclear charge of more than two are very rare. They can be observed in only about 1% of all cases of ternary fission. An overview of ternary fission is given in [3, 4]. For the ^{229}Th(n_{th},f) reaction only α- and triton-particles have been determined in the past [5]. For the ^{233}U(n_{th},f) reaction TP's up to ^{10}Be [6] and for the ^{239}Pu(n_{th},f) reaction TP's up to ^{20}O [7] are known from literature.

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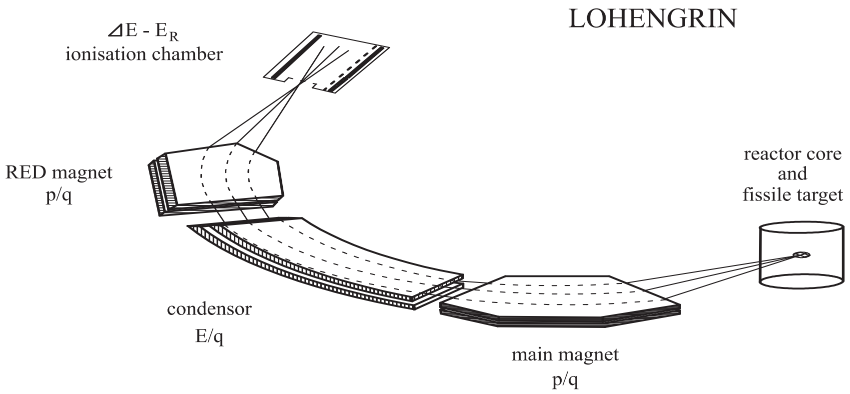

Fig. 1. Experimental setup: mass separator Lohengrin with RED-magnet and ΔE-E ionisation chamber.

The aims of the present experiments were the search for heavy ternary particles as well as the determination of their energy distributions and yields. Of special interest was the comparative study of the heaviest TP's as a function of the fissioning compound nucleus, which are detectable with the present techniques. Since in the meantime also ternary fission of ^{242}Pu*, ^{243}Am*, ^{246}Cm* and ^{250}Cf* [8–10] has been investigated, a broad range of actinides is now available for comparison.

2 Experimental Setup

For the determination of the energy distributions and yields of the TP's the mass separator Lohengrin [11] at the Institut Laue-Langevin (ILL) was used (see Fig. 1). Fission in targets with specific thickness of about 300μg/cm^2 is induced by thermal neutrons from the reactor core. The flux at the target position is 5,3·10^{14} n/(cm^2·s). Fission products emerging from the target reach the mass separator Lohengrin. In this spectrometer they are separated according to their mass over ionic charge A/q as well as their kinetic energy over ionic charge $E_{\rm kin}/q$ ratio by a magnet and an electrostatic condenser. An additional RED-magnet (**r**everse **e**nergy **d**isperson-magnet) focuses all particles leaving the exit slit of the mass spectrometer on the entrance window of an ionisation chamber. In this $\Delta E - E$ ionisation chamber the fission products are identified as to mass and energy and their nuclear charge is measured.

Determination of Yields

For the determination of the yields it is necessary to know the ionic charge distribution $F(v,q)$ and the energy distribution $Y(E_{\rm kin})$ of the ternary particles. The separation of the particles depends on their ionic charge q. Due to limited beam time it is not possible to perform a measurement for every ionic charge and energy. For a TP with the velocity v the ionic charge distribution $F(v,q)$ represents the relative contribution of the ionic charge q to all possible ionic charges. With the knowledge of $F(v,q)$ it is sufficient to perform only one measurement at one ionic charge for each energy. In Fig. 2 the $F(v,q)$ distributions of ^{10}Be are shown. Each marker represents one measurement, the curves are calculated. The ionic charge distribution is parameterized by a

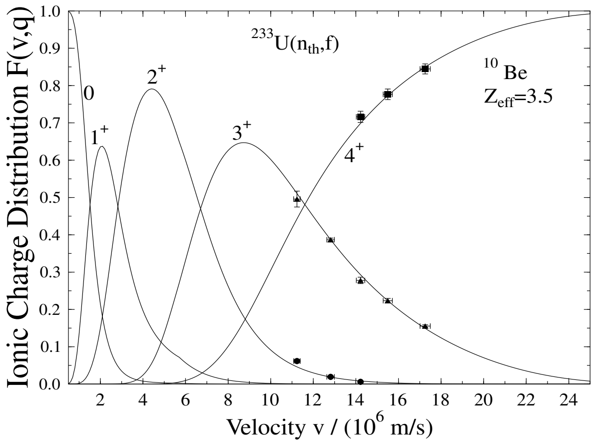

Fig. 2. Ionic charge distribution of ^{10}Be. The curves are calculated, each marker represents one measurement. $Z_{\rm eff}$ is a fit parameter.

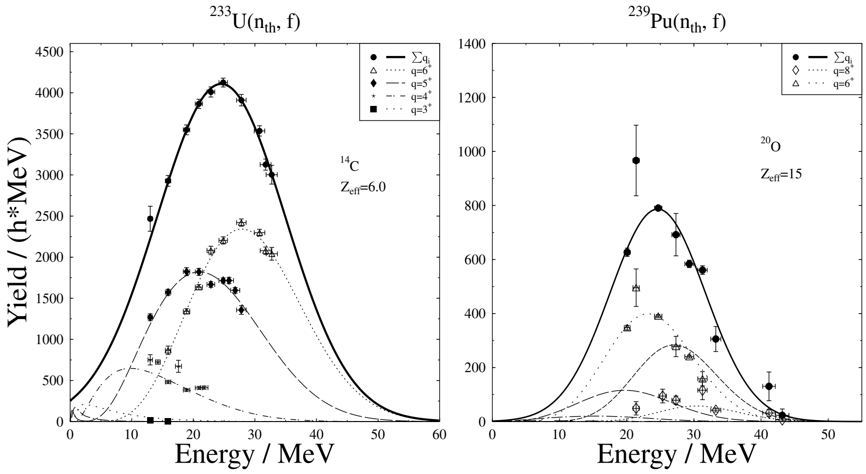

Fig. 3. Energy distributions of the ternary ^{14}C and ^{20}O from the reactions ^{233}U($n_{\rm th}$,f) and ^{239}Pu($n_{\rm th}$,f).

modified reduced χ^2 function. $Z_{\rm eff}$ is a parameter which is used to fit the modified reduced χ^2 function to the experimental data. For further details see [12].

Since the functions $F(v,q)$ fix the relative probability of the ionic charges the global yield of a TP as a function of its energy can be found. In Fig. 3 some energy distributions are shown. Global yields (summed over all ionic charges) are indicated by full points, thick lines represents the global energy distributions. These distributions are found from Gaussian fits to the data. With the help of $F(v,q)$ the global energy distributions are decomposed into the energy distributions at given ionic charges. These energy distributions for individual ionic charges are indicated in Fig. 3 by thin lines.

Table I. ^{229}Th(n_{th},f)
Mean energy $\overline{E}$, FWHM and yield of the TP's of the reaction ^{229}Th(n_{th},f). The yield is given relative to the yield of the ternary α-particle which is set to 10^4.

TP	This Measurement			Wagemans [5]		
	$\bar{E}$ / MeV	FWHM / MeV	Yield / $Y(\alpha)\cdot 10^{-4}$	$\bar{E}$ / MeV	FWHM / MeV	Yield / $Y(\alpha)\cdot 10^{-4}$
^{3}H				—	—	392±71
^{4}He				—	9.4±0.4	10^4
^{7}Li	—	—	10.4±1.7			
^{9}Be	—	—	3.8±2.3			
^{10}Be	18.4±0.5	17.5±1.2	75.7±7.1			
^{12}Be	—	—	1.1±0.5			
^{14}C	—	—	20.9±2.4			
^{15}C	23.8±0.9	15.9±1.8	4.3±0.4			
^{18}N	—	—	$(1.2\pm0.9)\cdot10^{-1}$			
^{20}O	22.9±2.8	21.5±3.8	1.8±0.5			
^{21}O	24.8±3.8	12.9±12.1	$(1.4\pm0.6)\cdot10^{-1}$			
^{21}F	—	—	$(2.5\pm2.0)\cdot10^{-2}$			
^{24}Ne	—	—	$(2.9\pm2.4)\cdot10^{-2}$			

The yields of the TP's are finally determined by an integration over the global energy distributions.

3 Results

3.1 ^{229}Th(n_{th},f)

For the ^{229}Th(n_{th},f) reaction 17 different TP's were observed, of which the heaviest particle was ^{24}Ne. The count rate for this ternary particle in the different settings of the mass separator Lohengrin was about 1 event/day. In practice this count rate sets a limit to the detectability of rare isotopes from ternary fission. Due to poor statistics the yields of the TP's ^{6}He, ^{9}Li, ^{12}B, ^{15}B, ^{18}C and ^{20}N could not be evaluated. In Table I mean energies $\overline{E}$ and FWHM of kinetic energy distributions, and yields of the ternary particles from the reaction ^{229}Th(n_{th},f) are shown. In case no energy data are indicated in Table I this means that not sufficient measurements were available to establish the energy distributions from experiment. For the determination of yields, $\overline{E}$ and FWHM had to be guessed by extrapolation from neighboring TP's and /or other compound nuclei. The yield is given relative to the yield of the ternary α-particle which is set to 10^4.

3.2 ^{233}U(n_{th},f)

For the ^{233}U(n_{th},f) reaction 25 different TP's were observed. It was possible to determine the yields of 17 ternary particles [12]. The heaviest TP searched for was ^{27}Na. However, only an upper limit was obtained with no positive detection.

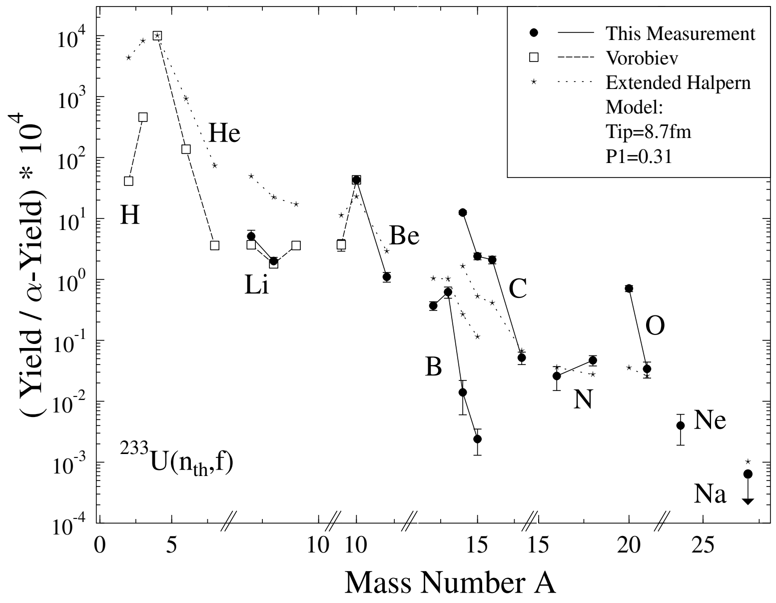

Fig. 4. Comparison between the experimental yields of TP's from this measurement [12] and Vorobiev [6] with the calculated yields of the extended Halpern model. In the case of ^{24}Ne the calculated and experimental yield is "by chance" equal. The ^{27}Na data point represents an upper limit.

The dependence of the ternary yield on mass or charge of the particles was analyzed in the framework of the extended Halpern model. In this model [13] the energy costs which are necessary to switch from binary to ternary fission are calculated. By weighting the energy costs with the binary fragment distribution it is taken into account that for a given TP many different mass and charge splits of the fragments may contribute. The distance between the tips of the fragments directly after scission is called the tip-distance. The tip-distance can be estimated from the total kinetic energy of the fragments. The fragments with the smallest kinetic energy originate from scission configurations with the largest deformations or equivalently with the largest tip distance between undeformed fragments. From such configurations ternary particles are thought to be emitted. High energy costs are expected to be unfavourable for ternary fission. The ternary yield is hence parameterized by an exponential function $Y \sim \exp(-E_{\text{cost}} \cdot P1)$ with only one fit parameter $P1$.

In Fig. 4 experimental yields from this and Vorobiev's [6] measurement are compared with the calculated yields from the extended Halpern model. As seen from Fig. 4 the general trend as a function of mass is quite well reproduced while the odd-even effect appears to be grossly underestimated.

3.3 ^{239}Pu(n$_{\text{th}}$,f)

For the ^{239}Pu(n$_{\text{th}}$,f) reaction 36 different TP's were found, with 23 of them having never been reported before from this reaction. The new TP's range from the light ^{6}He up to the heavy ^{33}Si. For this reaction the data are still under evaluation. In Table II the mean energies $\overline{E}$, FWHM and

Table II. ^{239}Pu(n_{th},f)
Mean energy $\overline{E}$, FWHM and yield of TP's from the reaction ^{239}Pu(n_{th},f). The yield is given relative to the yield of the ternary α-particle which is set to 10^4. For ^{34}Si an upper limit is given, it was not observed.

TP	This Measurement $\overline{E}$ / MeV	FWHM / MeV	Yield / $Y(\alpha)\cdot 10^{-4}$
^{10}Be	16.4±0.1	19.2±0.6	49±1
^{17}N	24.2±0.9	17.6±0.9	$(1.9\pm0.1)\cdot10^{-1}$
^{20}O	24.6±0.2	16.7±0.5	$(6.8\pm0.3)\cdot10^{-1}$
^{21}F	—	—	$(2.5\pm0.4)\cdot10^{-2}$
^{24}Ne	28.8±1.3	20.1±1.7	$(2.5\pm0.3)\cdot10^{-2}$
^{27}Na	—	—	$(2.0\pm0.5)\cdot10^{-3}$
^{30}Mg	—	—	$(2.8\pm1.5)\cdot10^{-3}$
^{32}Al	—	—	$(1.6\pm1.6)\cdot10^{-4}$
^{34}Si	—	—	$\leq 3.5\cdot10^{-4}$

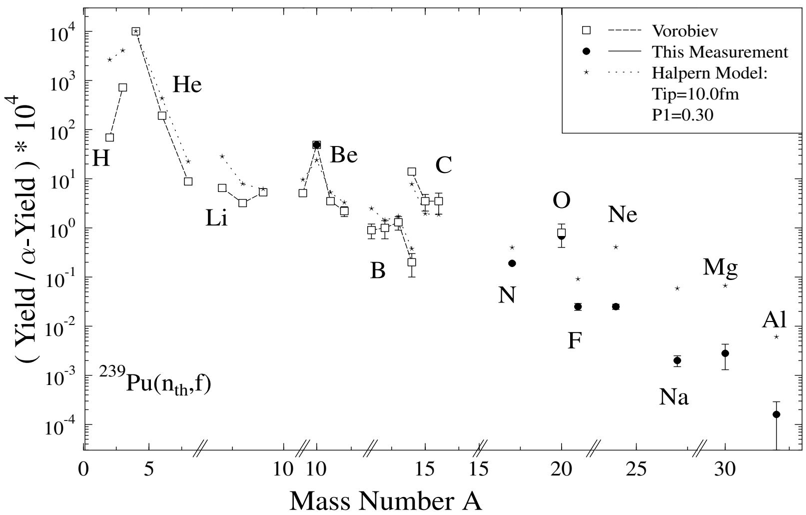

Fig. 5. Comparison between the experimental yields of TP's from this measurement and Vorobiev [7] with the calculated yields of the Halpern model. In the case of ^{20}O the calculated and experimental yield is "by chance" equal.

yields of the TP's which were evaluated until now are shown.

Figure 5 shows a comparison of experimentally obtained data and calculated yields for the reaction ^{239}Pu(n_{th},f). Experimental data are from this measurement and from Vorobiev [7]. The yields are calculated with the Halpern model. In contrast to the yields calculated for the ^{233}U(n_{th},f) reaction only one mass and charge split of the fragments is taken into account. The heavy fragment is set to be the doubly magic ^{132}Sn and it is assumed that the ternary particle is emitted by the complementary light fragment. Without weighting of the energy costs with the

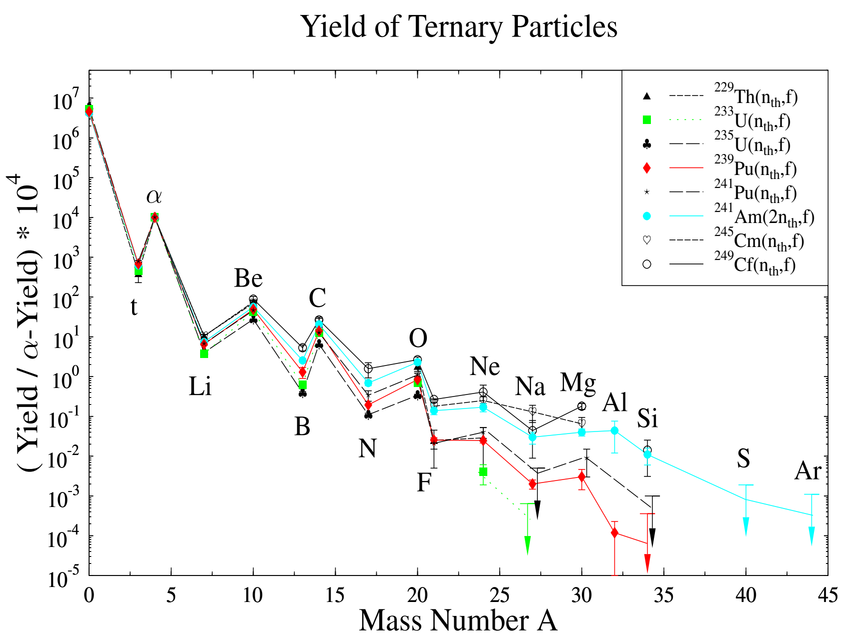

Fig. 6. Yields of ternary particles as a function of their mass number *A* for several actinides. The yields are given relative to the α-particle, whereby this yield is given as 10^4. Arrows indicate upper limits.

binary fragment distribution a better agreement is obtained between experimental and calculated yields, at least for ternary particles up to fluorine. For still heavier TP's, however, there is a violent disagreement.

4 Discussion

In Fig. 6 the yields of ternary particles for several thermal neutron induced fission reactions are compared. For this survey only the yields of the most abundant isotopes for each element have been retained. The yields are given relative to α-particles, with the α-particle yield being arbitrarily set to 10^4. The yields are shown as a function of the mass number *A* of the isotopes in question. For $A = 0$ the binary fission yield normalized to the above α-ternary yield is included. One can observe a dramatic decrease of the TP yields with their mass. Furthermore, systematically a higher yield for even charge numbers of TP's is seen. This odd-even effect is smoothed for the heavier elements. The yields of the lighter TP's from the different reactions lie close together (e.g. t, ^{7}Li, ^{10}Be). For higher masses of the particles, however, a widening spread in the yields for the different reactions is observed. As a rule, the heavier the compound nucleus the larger the ternary yields, especially for the heavy TP's. The mass or charge number of the heaviest TP's at the limit of detectability of present techniques is strongly correlated with the mass and/or charge of the fissioning compound nucleus. In Th- and U-fission the heaviest TP's are Ne-isotopes, in Pu-fission Al-isotopes and in Am- and Cf-fission the heaviest TP's are shifted to Si-isotopes. At a comparable level of sensitivity the lightest fragment from binary Am-fission has a mass of

$A = 68$ [14]. This means that in the mass spectrum of particles emitted in the fission process induced by thermal neutrons, there is at a 10^{-9} level in yield still a gap in observation between the heaviest ternary TP's and the lightest fragments from binary reactions.

The systematics in the limits of the heaviest TP's as a function of the compound system might be explained by recent evidence in an experimental study of spontaneous fission of ^{252}Cf [15]. In this experiment it was discovered that the total average excitation energy $\overline{\mathrm{TXE}}$ still left in the ternary fissioning system actually decreases rapidly with increasing mass and/or charge of the TP. Also a remarkably clear correlation between the TP yield and $\overline{\mathrm{TXE}}$ emerges. Extrapolating this correlation down to the yield levels measured for Si-isotopes e.g. in ^{243}Am-fission, one is led to the conclusion that in these limiting cases the system runs out of energy. Stated otherwise, for the heaviest observed TP's, ternary fission is a cold process. Having identified the limit of TP emission as the cold frontier of phase space, it is finally seen why the limit of TP emission is shifting with the mass and/or charge of the compound nucleus undergoing fission. Since the mean neutron number in fission increases with the mass of the fissile nuclei and since this number is a measure of $\overline{\mathrm{TXE}}$, it is concluded that more energy $\overline{\mathrm{TXE}}$ is available in the heavy fissioning systems. Hence the limit of TP emission with ever increasing energy costs shifts to heavier fissioning systems providing more energy.

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