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STUDY OF POST-NEUTRON MASS AND CHARGE YIELDS FOR $^{232}U(n_{th},f)$ and $^{238}Pu(n_{th},f)$

M. Haddad, M. Asghar

I. Ph. USTHB BP 32, El Alia, Alger, Algérie

J. Crançon, G. Lhospice

GEN/G, DRF/SPh/PhAN, 85X, 38041 Grenoble Cedex, France

Abstract : The post-neutron mass yields for $^{232}U(n_{th},f)$ and $^{238}Pu(n_{th},f)$ have been studied, for the first time, through γ-ray spectroscopy. A rabbit system was used to irradiate the encapsuled fissile material. Because of small quantities (<0.1 μg) of 'fission targets', an extreme care was taken as to the purity of the environment and the handling material as to the contamination from ^{235}U.

In the case of $^{232}U(n_{th},f)$, the yields of 36 fission products in 27 mass chains were determined. They represent ≈ 60% and ≈ 77% of the surface of the light and heavy mass peaks with $\langle A_L \rangle = 92.1 \pm 0.1$ and $\langle A_H \rangle = 137.9 \pm 0.1$ leading to $\langle \nu_T \rangle = 2.8 \pm 0.3$. The mean proton odd-even effect δ_p from 6 mass chains = $(24.6 \pm 3.0)\%$. For $^{238}Pu(n_{th},f)$, the yields of 51 fission products in 36 mass chains were obtained. They represent ≈ 62.5% and ≈ 82% of the surface of the light and heavy mass peaks with $\langle A_L \rangle = 93.1 \pm 0.1$ and $\langle A_H \rangle = 137.2 \pm 0.1$ - leading to $\langle \nu_T \rangle = 2.5 \pm 0.2$. The mean δ_p from 9 mass chains = $(14 \pm 3)\%$. These data will be compared with those for the other fissioning systems.

We have studied the post-neutron mass distributions resulting from thermal neutron-induced fission of $^{232}U(\sigma n_{th},f=75.2\ b)$ and $^{238}Pu(\sigma n_{th},f=16.5\ b)$. The fragment yields were obtained through γ-ray spectroscopy. The post-neutron mass yields were determined for 36 fission products in 27 mass chains for $^{232}U(n_{th},f)$ and for 51 fission products in 36 mass chains for $^{238}Pu(n_{Th},f)$. Furthemore, we managed to deduce the proton odd-even effect δ_p for ^{232}U and ^{238}Pu for 6 and 9 mass chains respectively. To our knowledge, these are the first results for the thermal neutron post-neutron mass distributions for these nuclei. Moreover, it is the first time that δ_p has been determined for $^{238}Pu\ (n_{th},\ f)$.

Experimental procedure

The nitric solutions containing higly pure ^{232}U and ^{238}Pu were obtained from AERE-Harwell. A drop of the solution containing ≈ 0.04 μg (≈ 0.6 μg) of ^{232}U(^{238}Pu) was transfered with a pipette to the bottom of a polyethylene capsule of 7 mm dia. and 15 mm height, dried and covered with a 2 mm thick layer of a special glue ; this thickness is greater than the range of fission fragments produced in the target. A lid of the top of the capsule through heating precautions were undertaken to ensure that no gaseous fission products escape from the capsule. As the "fission targets" consisted of very small quantities of fissile material (≈ 0.04 μg and ≈ 0.6 μg), the ^{235}U contamination of the environment in the laboratory posed a serious obstacle for this work. Drastic precautions had to be undertaken in order to overcome this problem. Tests were made with different blank capsules and handling material (pipettes...) to be certain that they did not contain ^{235}U at a detectable level of ≈ 10^{-9} g. It should be noted that ≈ 10^{-8} g of ^{235}U in the capsule produces as much fission activity as the target itself !

Three irradiation times : 90sec, 15 min and 2 h, were used. The 15 min and 2 h irradiations were done by sending the capsule via a rabbit system to a point inside a D_2O container of 1 m^3 volume placed near the core of the CEN-Grenoble Mélusine reactor with

$$\Phi_{th}=1.6\text{x}10^{13}\text{n/cm}^2\text{.sec},\quad \text{and}\quad \frac{\Phi n_{th}}{\Phi e_{pi}} > 2.5 \times 10^3.$$

The 90 sec irradiation was done in the Grenoble high-flux reactor at a point where

$$\Phi_{th}= 10^{14}\text{n/cm}^2\text{.sec},\quad \text{and}\quad \frac{\Phi n_{th}}{\Phi e_{pi}} > 5 \times 10^3.$$

These

measurements consisted of 1 - 2h. irradiation, 7 - 15min. irradiations and 29 - 90 sec. irradiations for ^{232}U ; and 1 - 2 h irradiation, 8 - 15 min irradiations and 58 - 90 sec irradiations for ^{238}Pu. A well-shielded and energy -and- efficiency calibrated 63 cm^{3}HP Ge detector was used to count and follow the decay of different γ-ray lines in order to identify the fission products emitting them and to ensure their purity. For the optimal functioning of the counting system, appropriate cooling and counting times were chosen for different irradiation times. The dead time corrections were obtained with a pulse generator. The fission product γ-ray energies and their emission probabilities were taken from ref.1.

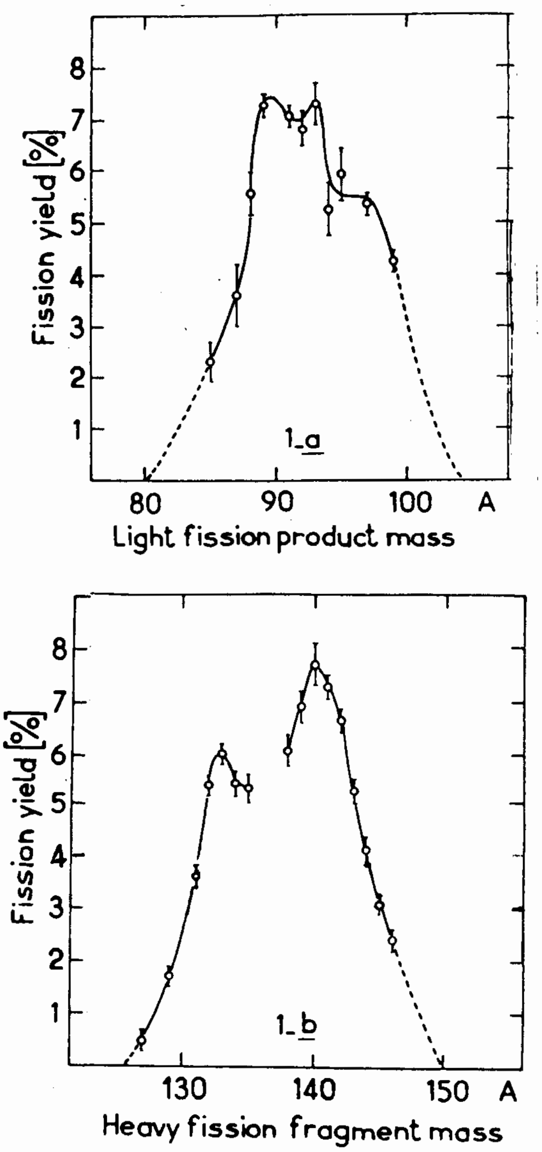
Results and discussion

a) The post-neutron mass yields for $^{232}U(n_{th},f)$.

As said before, to our knowledge, this is the first time that the post-neutron mass yields have been measured for thermal neutron induced fission of ^{232}U. However, the pseudo-mass yields have already been determined for this nucleus with the double-energy method^2.

The light fragment and the heavy fragment yields are plotted in Figs. 1a and 1b. The sum of the yields measured in this work represents ≈ 60.7% and ≈ 77.3% of the surface of the light fragment peak and of the heavy fragment peak respectively. The mean light and heavy fragment masses for the distributions are $\langle A_L \rangle = 92.1 \pm 0.1$ and $\langle A_H \rangle = 137.9 \pm 0.1$ respectively. The mean number of prompt neutrons emitted per fission from these masses is $\langle \nu_T \rangle = 233 - (\langle A_L \rangle + \langle A_H \rangle) = 2.8 \pm 0.3$; this value is a little lower than the experimental value of 3.13 ± 0.06 /3/.

The mass distributions of Figs. 1 a and 1 b show fine structures in the peak regions at masses ≈ 90, 93 and 97 (only shoulder) for the light peak, and at masses ≈ 133 and 140 for the heavy peak. With $\langle \nu_T \rangle = 3$, the mass pair 90, 140 and 97, 133 are complementary. However, we cannot check the complementarity of the structure at 93, because of the absence of measured yield in this region of the heavy peak. The understanding of these fine structures in terms of fragment shells has been discussed in details in ref.2.

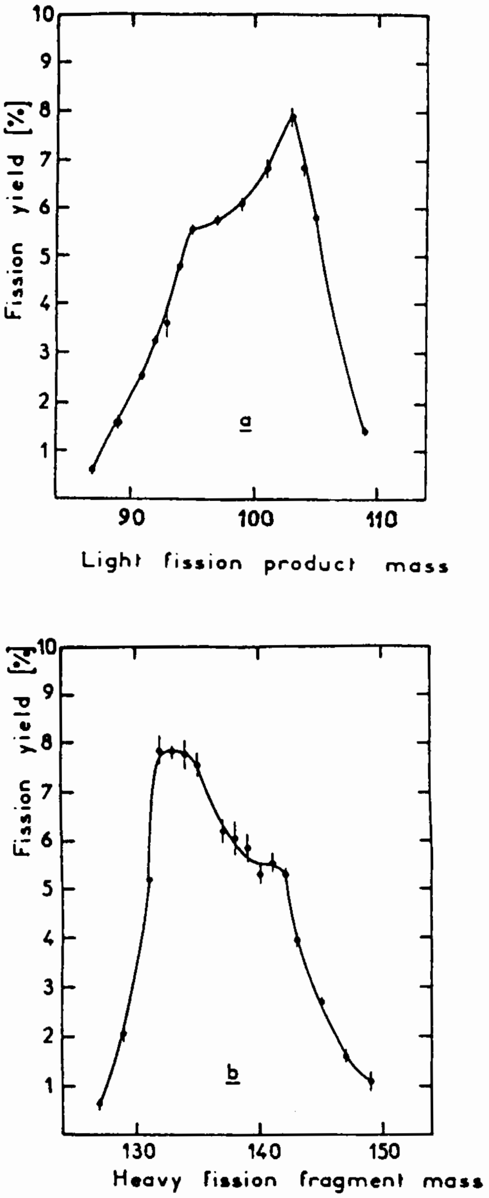


Figs. 1a and 1b - The post-neutron mass distributions (normalised to 100%) for the light and heavy groupes for $^{232}U(n_{th},f)$.

b) Post-neutron mass yields for $^{238}Pu(n_{th},f)$

To our knowledge, this is the first time that the post-neutron mass yields have been measured for this nucleus. However, the pseudo-mass yields have been determined for ^{238}Pu with the double-energy method^4 . This work shows that the mass distributions in the thermal neutron-induced fission of ^{238}Pu, ^{239}Pu and ^{241}Pu are quite similar. Therefore we compared the present post-neutron mass distribution of ^{238}Pu with the existing post-neutron data for ^{239}Pu and found them to be rather similar.

The light fragment and the heavy fragment yields are plotted in Figs. 2a and 2b. The sum of the yields measured in this study represent ≈ 62.5% and 82% of the surface of the light fragment peak and of the heavy fragment peak respectively. The mean light and heavy fragment masses for the distribution are $\langle A_L \rangle = 99.3 \pm 0.1$ and $\langle A_H \rangle = 137.2 \pm 0.1$ respectively. The mean number of prompt neutrons emitted per fission from these masses is $\langle \nu_T \rangle = 239 - (\langle A_L \rangle + \langle A_H \rangle) = 2.5 \pm 0.2$; this number is a little lower than the experimental value of 2.892 ± 0.027 /3/.



Figs. 2a and 2b - The post-neutron mass distributions (normalised to 100%) for the light and heavy groupes for $^{239}Pu(n_{th},f)$.

The mass distributions of Figs. 2a and 2b show fine strutures in the peak regions, at masses ≈ 95 and 103 for the light peak, at masses ≈ 133 and 142 for the heavy peak. If one considers that the mass pairs 95, 142 and 103, 133 are complementary, one gets $\nu_T = 2$ and 3 respectively for these pairs and $\langle \nu_T \rangle = 2.5$ for them ; this is close to the $\langle \nu_T \rangle$ value averaged over all the masses just given above.

c) Proton odd-even effect δ_p

The proton odd-even effect δ_p represents the excess of the yields of even-Z fission products over the yields of odd-Z fission products and is defined as

$$\delta_p[\%] = \frac{\Sigma\, Y_{even} - \Sigma\, Y_{odd}}{\Sigma\, Y_{even} + \Sigma\, Y_{odd}} \times 100$$

where Y_{even} and Y_{odd} are the yields of even-Z and odd-Z fission products respectively.

In order to get δ_p for different mass chains, one needs the independent yields of different isobars of these chains. The different methods to get them have been discussed in ref.5. These independent yields along with the Wahl "normal" distribution for independent yields help to determine δ_p. The width σ_z of the normal distribution was taken to be 0.5 and Z_p, the most probable nuclear charge for a given mass chain, was obtained using the semi-empiral relation of Coryell et al.[6]. This treatment of data helped us to get δ_p for 6 mass chains : 89, 94, 134, 135, 138 and 139 for $^{232}U(n_{th},f)$, and for 9 mass chains : 89, 94, 131, 133, 135, 137, 138, 139 and 142 for $^{238}Pu(n_{th},f)$. The mass-chain-weighted mean values of proton odd-even effect for $^{232}U(n_{th},f)$ and $^{238}Pu(n_{th},f)$ are $\delta_p = (24.5 \pm 3)\%$ and $= (14 \pm 3)\%$ respectively. The value of δ_p for $^{232}U(n_{th},f)$ is consistent with $\delta_p = (21 \pm 3)\%$ determined for this nucleus through the direct physical methods[7]. However, this is the first time that proton odd-even effect has been determined for $^{238}Pu(n_{th},f)$. In the case of ^{240}Pu, though 1.45 MeV excitation energy is available at the last saddle, yet very little pair breaking takes place there. This situation is similar to ^{239}Pu, where the fissioning nucleus has ≈ 0.0 MeV excitation energy above the barrier. Hence, it seems that, in the thermal neutron fission of nuclei at least up to Pu, most of the pair breaking - hence, quasi-particle excitation - comes about dynamically, when the fissioning nucleus moves from saddle to scission and ruptures into fragments.

Conclusions

The main contribution of this work is a follows :

a) For the first time, the yields of 36 fission products in 27 mass chains for ^{232}U(n_{th},f) , and the yields of 51 fission products in 36 mass chains for ^{238}Pu(n_{th},f) have been determined through γ-ray spectroscopy.

b) The post-neutron mass distribution for ^{232}U(n_{th},f) shows peaks and/or shoulders at masses ≈ 90, 93 and 97 for the light group and at masses ≈ 133 and 140 for the heavy group ; for ^{238}Pu(n_{th},f) they come up at masses ≈ 95 and 103 for the hight group and at masses ≈ 133 and 142 for the heavy group.

c) The mean values of neutrons per fission for ^{232}U(n_{th},f) and ^{238}Pu(n_{th},f) are $\langle \nu_T \rangle = 2.8 \pm 0.3$ and 2.5 ± 0.2 respectively.

d) The average proton odd-even effect values for ^{232}U(n_{th},f) and ^{238}Pu(n_{th},f) are $\delta_p = (24.5 \pm 3)\%$ and $(14 \pm 3)\%$ respectively.

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