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RIB PRODUCTION WITH PHOTOFISSION
OF URANIUM

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Introduction

In the synthesis of superheavy elements and the study of heavy nucleus fission (collective dynamics of many-body systems) in any target-ion combination we do not reach the closed neutron shell $N=184$, in which gigantic changes in the nuclear structure are expected, and hence, in the heavy nucleus stability. Even when using the most heavy ^{244}Pu , ^{248}Cm or ^{249}Cf targets and ^{48}Ca beams, some nuclei are formed which in their ground states are 7-9 a.m.u. away from the region enhanced of stability [1]. The use of a radioactive beam of ions with neutron excess higher than in the stable ^{48}Ca nucleus could solve the problem.

Reactions of cold fusion between massive nuclei with masses and charges close to those of fission fragments [2] may turn to be promising for these purposes. To demonstrate the inverse fission reaction, Fig. 1 shows the results of low excitation energy asymmetric fission of ^{224}Th nuclei [3] due to the contribution of the neutron shells $N=50$ and $N=82$ and the cross sections of the fusion reaction $^{86}\text{Kr} + ^{136}\text{Xe} \rightarrow ^{222}\text{Th}$ at different excitation energies of the compound ^{222}Th nucleus [4]. As it follows from Fig. 1, of all investigated reactions involving different nuclei used as targets and projectiles, the inverse fission leads to formation of the compound ^{222}Th nucleus with a minimal excitation energy. Fission fragments of the ^{132}Sn , ^{133}Sb and ^{134}Te nuclei which, as known, determine the character of fission of practically all actinide nuclei at low excitation energies, are of obvious interest from the point of view of the synthesis of nuclei heavier than Th, and also for the synthesis of isotopes of transactinide elements. That is why the acceleration of fission fragments in the vicinity of the doubly magic nucleus ^{132}Sn is perhaps the most ambitious goal of the existing RIB projects.

The idea of producing a beam of neutron-rich isotopes which are formed in the fission of uranium and thorium nuclei have existed for many years. At the new generation accelerator complexes of radioactive nuclei they use the reaction of ^{238}U fission with high energy protons ($E_p=0.6\div 50$ GeV; CERN,KEK), with fast neutrons generated by a deuterium beam with $E_d=20$ MeV (the PARNE project, Orsay-GANIL), with thermal neutrons in the reaction $^{235}\text{U}(n_{th},f)$ – the PIAFFE project (Grenoble) and a new MAFF project (Munich).

To make the picture complete, note that it is possible to use extremely expensive, but very convenient spontaneous fission – the isotope ^{252}Cf , produced in high-flux reactors (Oak-Ridge, Dimitrovgrad), as well as photofission of ^{238}U . The latter deserves separate consideration.

Photofission of uranium

As known, in the interaction between electrons having an energy E_e and converter material (W or U), γ -radiation with a continuously falling spectrum down to $E_\gamma^{\max} = E_e$ is generated. Fission of heavy nuclei induced by γ -rays with different energies is determined by the region of giant dipole resonance (GDR). In the case of

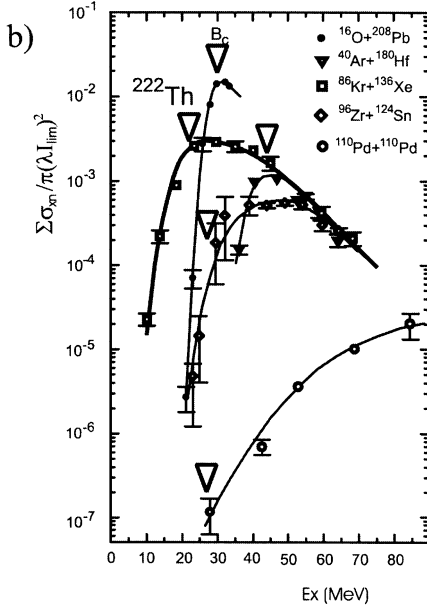
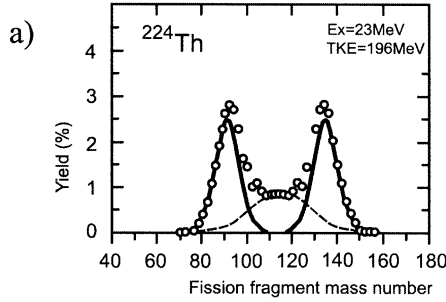


Fig. 1. a) Mass distribution of ^{224}Th ($E_x = 23$ MeV) fission fragments produced in the reaction $^{16}\text{O} + ^{208}\text{Pb}$. b) Formation cross section of $^{220-224}\text{Th}$ nuclei in the fusion reactions. The thick curve corresponds to the reaction $^{86}\text{Kr} + ^{136}\text{Xe}$. The arrows show the position of the Coulomb barrier of the reactions.

^{238}U nuclei, the fission cross section at the GDR energies reaches 0.16 barn [4]. The dependence of the uranium fission cross section $\sigma_f(E_\gamma)$ in fact determining the GDR structure is presented in Fig. 2. The yield of γ -quanta in the GDR region ($E_\gamma=10-17$ MeV) depends on the energy of electrons as it is shown in Fig. 2. Proceeding from this it is easy to derive the dependence $\sigma_f(E_\gamma)$ and estimate the probability of uranium fission in the optimal size target per every electron reaching the target with an

energy E_e . From Fig. 3 it follows that the fission fragment yield sharply grows with an increase in E_e up to $E_e=30$ MeV and then continues to grow smoothly up to $E_e=50$ MeV and higher. At $E_e=50$ MeV the calculated fission yield is about 0.6% per electron.

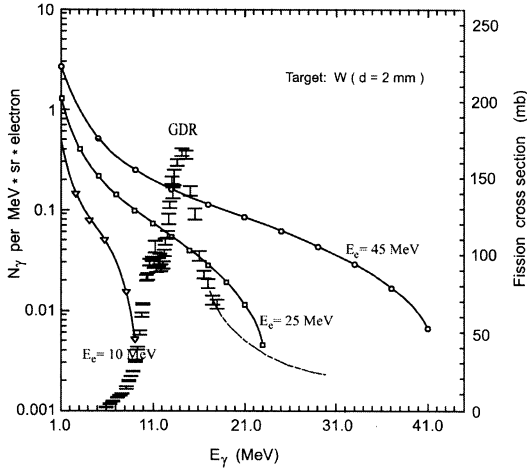


Fig. 2. The solid lines (the left-hand scale) is the γ -quanta spectrum produced by electrons with various energies (indicated in the figure). The experimental points (the right-hand scale) correspond to the ^{238}U photofission cross section (according to the data from [4]).

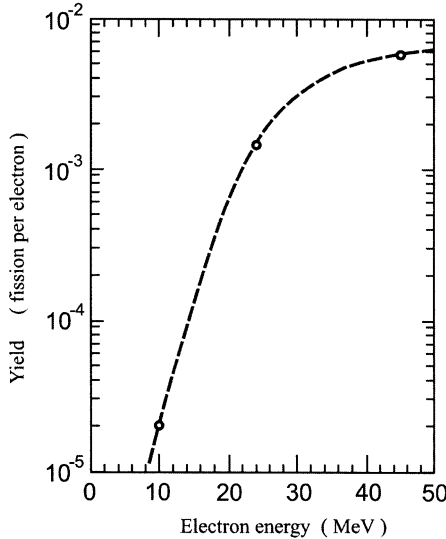


Fig. 3. The fission yield per electron for ^{238}U as a function of the electron energy.

Experiments with a 25 MeV electron beam

The yield of photofission ^{238}U fragments was determined experimentally at a beam of electrons produced by the compact accelerator of the microtron type MT-25 [5].

The facility MT-25 consuming about 20 kW produces a beam of electrons with an energy $E_e = 25$ MeV and intensity of $20 \mu\text{A}$ (the power of the electron beam is 0.5 kW). A Ta plate 2.5 mm in thickness was used as a converter.

In the first experiment a metallic uranium plate 35 g/cm^2 in thickness was placed behind the converter (the total weight of the target was 100 g). The intensity of neutrons "born" at the target and emitted in a solid angle of $4\pi\text{-sr}$ was $8 \cdot 10^{11}/\text{s}$. According the data on the partial cross sections $\sigma_{\gamma n}$ and $\sigma_{\gamma f}$ for ^{238}U it was found that 59.5% of the neutrons were produced due to the photofission of uranium. At an average number of neutrons per fission $\bar{\nu} = 3.6$ ($E_x \sim 15$ MeV), it corresponds to $1.3 \cdot 10^{11}$ of fission events per second [6]. At this energy the ratio $N_f/N_e \approx 10^{-3}$. Another experiment was aimed at the determination of the effective target size. The angular spread of the beam of γ -quanta depends on the energy E_γ and in first approximation is determined by the relation $\alpha \approx m_0 c^2 / E_\gamma$. For the γ -quanta in the GDR region $\alpha \approx 2 - 2.5^\circ$. Due to the narrow angular spread of the γ -rays the radial size of the U target is determined by the radial size of the electron beam which in our experiment was $d_e = 6$ mm.

Reaching the U-target the intensity of the γ -rays which produce fission fragments is getting weaker due to absorption in the layer by means of electron-positron pair production. For the determination of the γ -quanta beam configuration an experiment was performed as is schematically shown in Fig. 4a.

A standard "sandwich" consisting of a thin U_2O_3 layer 0.3 mg/cm^2 in thickness deposited on an Al ($20 \mu\text{m}$) backing and a plastic track detector (Mylar – $50 \mu\text{m}$) was used for the fission fragment registration. Pb plates of various thicknesses imitating the solid U-target were placed in between the "detector sandwiches". Fig. 4b shows radial distributions of fission fragment tracks produced with the use of Pb absorbers of different thicknesses. The effective longitudinal and transverse sizes of the U-target were derived from the dependences presented in Fig. 5.

More than 80% of the γ -quanta with the GDR energy range are absorbed by metallic uranium of about 30 g/cm^2 in thickness. For the electron beam with $d_e = 6$ mm (the experiment was performed under this condition) the effective target volume was in the form of a cylinder with $d = 10$ mm and $l = 15$ mm (matter weight ~ 22 g). If an electron beam size of down to $d_e = 3$ mm is focused onto the converter, the target weight decreases to 10 g. The total fission fragment yield in the effective volume is about $1.5 \cdot 10^{11}/\text{s}$ which is in good agreement with the previous experimental data on measuring the neutrons in the photofission of uranium. For increasing the fission fragment diffusion from the target it is supposed to use composite material in which the uranium atom is implanted into the structure UC_x ($x = 5 - 7$); the density is about 1.5 g/cm^3 .

According to technology used at the Institute of Nuclear Physics (Gatchina, St.Petersburg) such structure may contain up to 80% of uranium by atomic weight [7]. In this case for the electron beam with $d_e = 5$ mm the target configuration will be in the

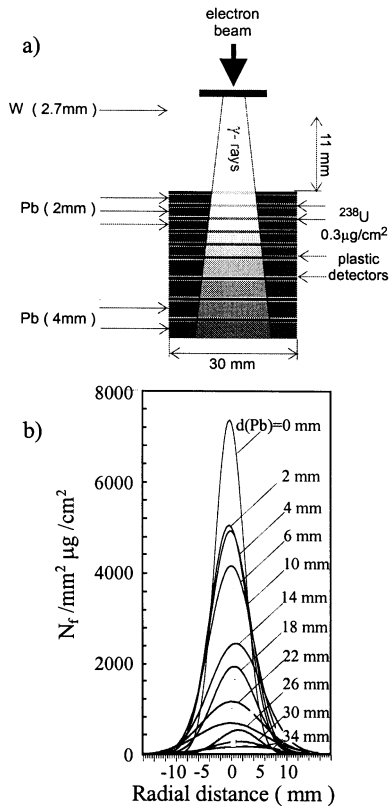


Fig. 4. a) Layout of the experiment b) radial distributions of γ -quanta in the GDR region after the absorption in Pb of various thicknesses (indicated in the figure).

form of a truncated cone, its small and large diameters are 7 and 20 mm, respectively, and the length is about 120 mm. The loss in the intensity of the γ -rays due to the absorption in the carbon ballast do not exceed 10%. Naturally, for producing relatively long-lived radioactive atoms of the above mentioned magic nuclei ^{132}Sn ($T_{1/2}=40 \text{ s}$); ^{133}Sb (2.5 min); ^{134}Te (42 min), the target may be manufactured of a more dense composition of the uranium carbide type (density of about $12 \text{ g}/\text{cm}^3$) which decreases the volume to $1.5 \div 2 \text{ cm}^3$. Fig. 6 is the map of yields of different nuclei formed as fission fragments in the ^{238}U photofission. In the calculation we used the data obtained from the ^{238}U and ^{237}Np fission induced by neutrons with the average energy of about 2 MeV [8,9] and ^{238}U photofission at the electron energy of 20–22 MeV [10,11]. As seen from Fig. 6, the mass spectra of fission fragments look like asymmetric distributions typical for low-energy fission of uranium.

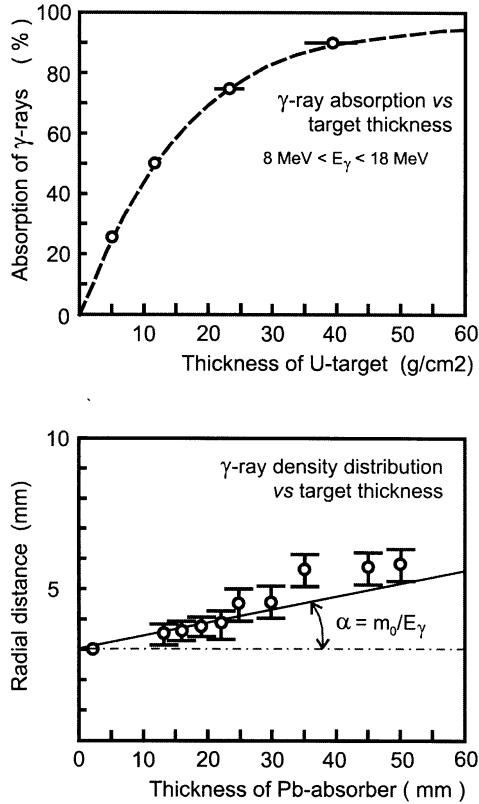


Fig. 5. The choice of the U-target effective size.

Table 1 shows the yields of neutron rich isotopes of Sn and Xe produced in the $^{238}\text{U}(n,\gamma)$ reaction at the rate of $1.3 \cdot 10^{11}$ fission/s.

Two experiments aiming at the determination of the yields of ^{132}Sn and ^{142}Xe isotopes in the $^{238}\text{U}(\gamma,f)$ -reaction were performed. The yield of ^{132}Sn nuclei ($T_{1/2}=39.7$ s) was determined through the intensity of γ -rays ($E_{\gamma}=8.99$ keV directly during a series of short exposures. For the measurement of the Xe yield the nuclei which recoiled from a thin U layer were collected on a cold catcher ($T=63^{\circ}\text{K}$) where the decay daughter products, in particular, ^{142}La ($T_{1/2}=92.5$ min) were measured.

For the 10-g uranium target and the electron beam $P_e=0.5$ kW, the experimentally determined yields amounted to: $^{132}\text{Sn} - 1.7 \cdot 10^9 \text{ s}^{-1}$ and $^{142}\text{Xe} - 2 \cdot 10^9 \text{ s}^{-1}$, which agree with the expected one within the limits of measurement accuracy. Now we can compare different reactions of uranium fission in view of RIB production.

PRODUCTION OF NEUTRON-RICH NUCLEI

$^{238}\text{U}(\gamma, f)$ -reactor.

Electron beam energy: 25 Mev

intensity: 20 μA

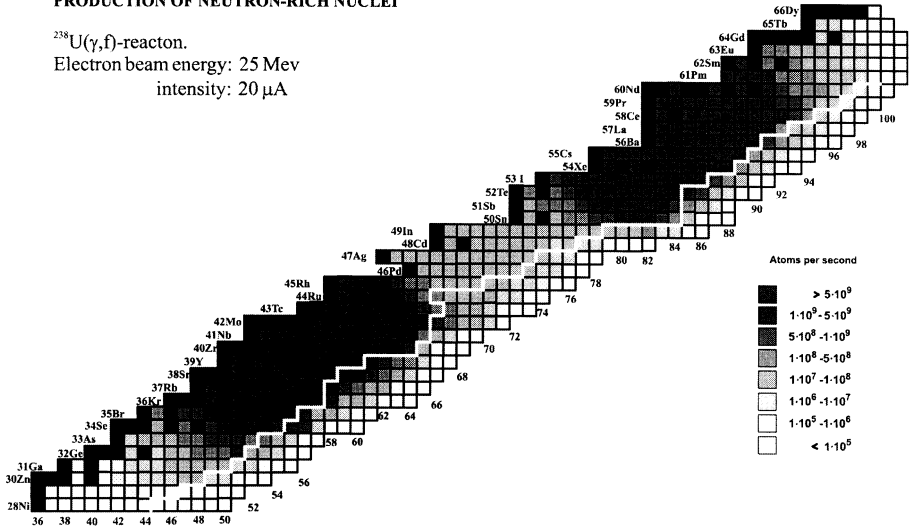


Fig. 6. The yield of ^{238}U photofission fragments. The white line corresponds to nuclides with $T_{1/2} \approx 1$ s.

Table 1. The yields of Sn and Xe isotopes produced in the $^{238}\text{U}(n, f)$ and $^{238}\text{U}(\gamma, f)$ reactions (at the rate $1.3 \cdot 10^{11}$ fission/s)

| Sn-isotopes | | | | Xe-isotopes | | | |
|-------------|-----------|------------------------------------|-------------------------------------|-------------|-----------|------------------------------------|-------------------------------------|
| A | $T_{1/2}$ | $^{238}\text{U}(n, f)$ ENDF 349 | $^{238}\text{U}(\gamma, f)$ exp. | A | $T_{1/2}$ | $^{238}\text{U}(n, f)$ ENDF 349 | $^{238}\text{U}(\gamma, f)$ exp. |
| 130 | 3.7 m | $2.3 \cdot 10^9/s$ | | 140 | 13.6 s | $6.9 \cdot 10^9/s$ | |
| 131 | 50 s | $2.6 \cdot 10^9/s$ | | 141 | 1.72 s | $3.6 \cdot 10^9/s$ | |
| 132 | 39.7 s | $2.1 \cdot 10^9/s$ | $1.7 \cdot 10^9/s$ | 142 | 1.24 s | $1.7 \cdot 10^9/s$ | $2 \cdot 10^9/s$ |
| 133 | 1.44 s | $6.5 \cdot 10^8/s$ | | 143 | 0.30 s | $2.6 \cdot 10^8/s$ | |
| 134 | 1.05 s | $1.3 \cdot 10^8/s$ | | 144 | 1.15 s | $5.2 \cdot 10^7/s$ | |

Table 2 shows the yields of ^{137}Xe (for some reason isotope has been chosen for comparing the fission fragment yields in the RIB project). The yield of neutron-rich isotopes depends on the excitation energy which is taken by neutron emission before and after fission.

It is known that post-scission neutrons are emitted by fission fragments with large neutron excess. That is why the fission of weakly excited nuclei is preferable for producing neutron-rich fission fragments.

Table 2. Fission sources used for the production of the isotope ^{137}Xe with rate $5 \cdot 10^9$ atoms/s.

| Projectiles | Target | Beam intensity |
|-----------------------------------|---------------------------|--------------------------------------|
| Protons (600 MeV) | ^{238}U (100 g) | 50 μA |
| Spontaneous fission | ^{252}Cf (0.2 g) | |
| Thermal neutrons | ^{235}U (0.4 g) | $1.6 \cdot 10^9$ n/cm ² s |
| Fast neutrons (deutrons 130 MeV) | ^{238}U (33 g) | 500 μA |
| γ -rays (electrons 25 MeV) | ^{238}U (10 g) | 20 μA |

This situation is demonstrated by Fig. 7, in which the calculated yields of Sn isotopes in different reactions induced by protons, deuterons and ^{238}U nuclei (A.R.Junghans, GSI 1998) [12] are shown as well as the data on ^{238}U photofission obtained earlier.

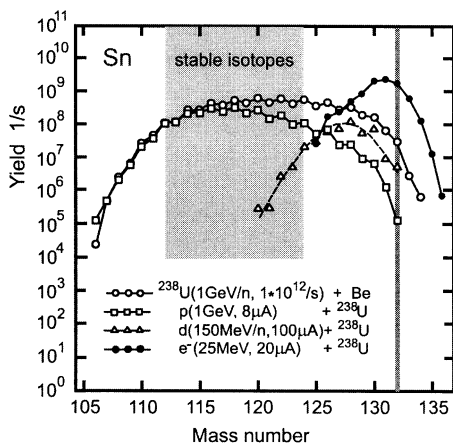


Fig. 7. The yield of Sn isotopes in different reactions (indicated in the figure).

Conclusion

Photofission of ^{238}U nuclei will be used for producing beams of neutron-rich nuclei in the DRIBs project (Dubna Radioactive Ion Beams). The MT-25 microtron, allowing a 0.5 kW electron beam, in combination with the cyclotron complex ECR-4M + U400 will make it possible to produce beams of the ^{132}Sn and ^{142}Xe type with an intensity of up to 10^7 s⁻¹ and energy from 5 to 18 MeV/n. This variant is realized in comparatively mild radiation conditions using acting accelerator facilities and allows operation without any complicated radiation protection systems during a long period of time.

Naturally, parameters of the compact microtron are far from those of modern electron accelerators operating with medium energies. An increase in the power of the electron beam up to 1 MW ($E_e=50$ MeV, $I=20$ mA), which is quite feasible at modern accelerators, will allow increasing the intensity of ion beams by $5 \cdot 10^3$ times. Note that such set-up is at the same time an intensive compact neutron source ($\sim 3 \cdot 10^{15}$ n/s) and it can be employed in the breeders used for different purposes.

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Получение радиоактивных пучков фоторасщеплением урана

Рассматривается процесс фотоделения урана электронными пучками с энергией 20–50 МэВ для получения осколков деления. Показано, что при взаимодействии между электронным пучком (с энергией 25 МэВ и интенсивностью 20 мкА), полученным на компактном электронном ускорителе микро-тронного типа, и урановой мишенью толщиной около 40 г/см² генерируется в среднем $1,5 \cdot 10^{11}$ делений/с.

Согласно расчетам и экспериментальным результатам, полученным в настоящей работе, это соответствует выходу изотопов ¹³²Sn и ¹⁴²Xe приблизительно $2 \cdot 10^9$ /с.

Представляются результаты экспериментов по оптимальной конструкции урановой мишени. Обсуждаются проблемы, связанные с разделением изотопов и изобар для их дальнейшего ускорения на циклотроне У-400 до энергий 5–18 МэВ/нуклон.

Реакции фотоделения тяжелого ядра сравниваются с другими методами получения радиоактивных пучков ядер средней массы.

Работа выполнена в Лаборатории ядерных реакций им. Г.Н.Флерова ОИЯИ.

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RIB Production with Photofission of Uranium

The process of uranium photofission with electron beams of 20 ÷ 50 MeV is considered in terms of the production of fission fragments. It is shown that in the interaction between an electron beam (25 MeV in energy and 20 μA in intensity), produced by a compact accelerator of the microtron type, and a uranium target of about 40 g/cm² in thickness, an average of $1.5 \cdot 10^{11}$ fission events/second is generated.

According to the calculations and test experiments, this corresponds to the yield of ¹³²Sn and ¹⁴²Xe isotopes of approximately $2 \cdot 10^9$ /s.

The results of experiments on the optimal design of the U-target are presented. Problems are discussed connected with the separation of isotopes and isobars for their further acceleration up to energies of 5–18 MeV/n.

The photofission reactions of a heavy nucleus are compared with other methods of RIB production of medium mass nuclei.

The investigation has been performed at the Flerov Laboratory of Nuclear Reactions, JINR.

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