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Tests of high-density UC targets developed at Gatchina for neutron-rich radioactive-beam facilities

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Abstract

Production of on-line mass-separated neutron-rich isotopes using fission induced by 1 GeV protons on high-density uranium targets (typically 11 g/cm^3) has been investigated for several years at the IRIS facility. Here we review some of the achievements during the years up to 2006 and the perspectives. In particular, we present a comparison of yields in p and n-induced fission, enhancements by secondary neutrons, a comparison with lower-density targets used at PARRNe and ISOLDE for Rb and Cs isotopes and preliminary results obtained with a new UC target material.

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1. Introduction

The demand for targets containing large amounts of uranium has been increased by the new projects concerning radioactive-beam facilities of the next generation based on the ISOL method [1–3]. The use of a high-density target material reduces the volume of the target unit and thus the effusion time. We developed high-density uranium carbide targets, of density close to the theoretical mono-crystal value, and carried out extensive tests at the IRIS facility located at PNPI-Gatchina, Russia, in the framework of the PNPI–LNL–IPN Orsay–GANIL (PLOG) collaboration. These works have been published in [4–10] and summarized in an activity report covering the measurements until 2006 [11]. Modelling the perturbation of release curves due to decay of precursors in the target is also published elsewhere [12,13]. Measurements of very short-lived francium isotopes and of more difficult elements like Ag and Sn are reported in other contributions to this conference [14]. Here we present some measurements with the fission products of Rb and Cs.

2. The IRIS facility at PNPI

IRIS (investigation of radioactive isotopes at synchrocyclotron) is a ISOL on-line isotope separator facility using a

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proton beam of 1 GeV with currents of about 0.1 μ A. Except for comparison of n- and p-induced fission yields the proton beam is sent directly to the target. The 10 mm size of the proton beam spot corresponds to the diameter of the targets. The number of fissions is a few 10^{10}/s. The deposited heat (0.1 μA times 4 MeV lost in 10 g/cm^2) is negligible against the external 1.8 kW required to keep the target at 2000 °C. Thermal condition in the target is thus not affected by beam pulsing.

The atoms of the produced radioactive nuclei are released from the target, ionized in the ion-source, mass-separated and then implanted on a movable tape. The collected activity is transported after 1.25 s in front of a γ or a β-radiation detector for counting. Yields are measured in a steady state to allow for equilibrium to be reached in the target, whereas the recording of release curves requires proton beam pulsing.

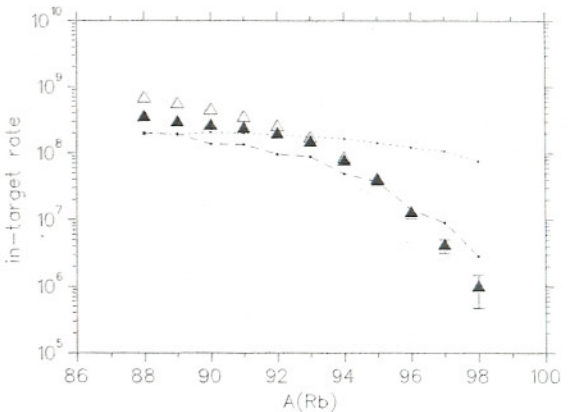

Fig. 1. Rates of rubidium isotopes produced in a 19.8 g/cm^2 UC_x target by 100 nA of 1 GeV protons, based on cross-sections (triangles, full if only Rb is considered, open if all precursors decay in the target), a calculation with an early version of MCNPx (dotted line) [5] and a recent one using CEM.2.6.b (dashed line) [18], in which all fissions induced by secondary particles are included.

3. Release times

The length of collection/counting cycles is chosen to be short with respect to characteristic release times and radioactive half-lives. The number of counted decays is therefore closely proportional to the average ion current during collection. Many such cycles are included in a beam pulsing cycle, starting with an irradiation and followed with the measurement after the proton beam has been switched off.

Release curves are analysed as sums of terms with time dependence given by $e^{-(\mu_k+\lambda)t}$ and $e^{-(v+\lambda)t}$ factors, where μ_k refers to a term of the diffusion series [15], v to the effusion term and λ is the decay constant. Our release curves exhibit a slow and a fast component. This decomposition allows a much lower χ^2 than the shape corresponding to the convolution of diffusion out of spherical grains and effusion. An open question is what the origin of these components could be. If we express the release function as

$$R(t) = a_1\mu_1 e^{-\mu_1 t} + a_2\mu_2 e^{-\mu_2 t},$$

with normalisation implying $a_1 + a_2 = 1$, the observed curve during beam-off after a long-lasting irradiation is

$$R(t) = C\left(a_1\frac{\mu_1}{\mu_1+\lambda}e^{-(\mu_1+\lambda)t} + a_2\frac{\mu_2}{\mu_2+\lambda}e^{-(\mu_2+\lambda)t}\right)$$

and the release efficiency is

$$\varepsilon = a_1\frac{\mu_1}{\mu_1+\lambda} + a_2\frac{\mu_2}{\mu_2+\lambda}.$$

Variation of efficiency versus lifetime of isotopes is thus an indirect measurement of release-time. However, the mere calculation of efficiency can be biased by assumptions. Experimental cross-sections for 1 GeV protons are available [16], but the impact of neutrons produced in the target is more difficult to estimate. Furthermore, fission creates neutron-richer isobars of the nucleus of interest, a fraction of which may decay in the target before escaping, see [13] for a detailed treatment. The actual in-target distribution is thus intermediate between those shown in Fig. 1. In addition, the calculated experimental ion current depends on the intensities of γ transitions per decay. They are not always trustable enough. An unusually severe mistake was done in the decay of ^{92}Rb, see [17], but scaling errors within a factor of 1.5 seem to be common. In our experimental conditions the efficiency is a quick method to evaluate the release-time performance of a target, but cannot teach much about the details of the processes.

4. The HDR material

A high-density rod (HDR) UC_x uranium carbide ($\rho = 11.2$ g/cm^3) with an average grain size of about 200 μ has been used for the early tests. Release properties of targets made of this material, be they as a rod or powder, are very comparable and no dependence on thickness (at least up to 20 g/cm^2) has been observed. They are thus dictated by the grain, and must be associated with diffusion. Release curves can be decomposed into about 1 min (fraction a_k of 0.2–0.3) and 25 min components. If they had been recorded with a short-lived isotope (say a few seconds), the slow component could only have been noticed from the discrepancy of experimental and calculated efficiencies.

4.1. Proton versus neutron-induced fission

In fission induced by neutrons, the production of neutron-rich isotopes far from stability is enhanced with respect to proton-induced fission. We investigated this effect experimentally. The target material was a powder with an effective density of 7.36 g/cm^3 and a thickness of 19.8 g/cm^2. Both the UC target and the ion-source were operated at 2100 °C. It turned out that the deduced efficiencies (defined as the observed yield divided by the in-target production rate calculated with MCNPx) for long-lived

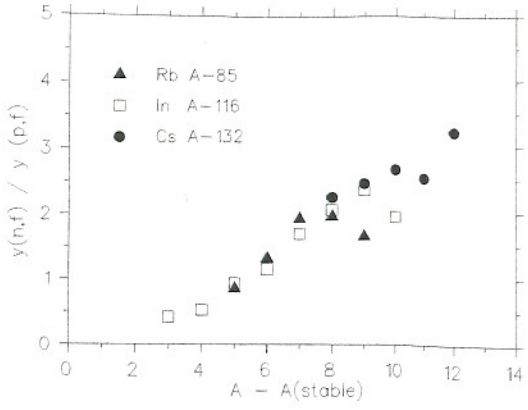

Fig. 2. Ratio of yields per neutron (resp. proton) for neutron (resp. proton)-induced fission versus the distance from the line of stability for neutron-rich isotopes of Rb, In and Cs. Error bars due to counting are smaller than the symbol size.

nuclei were a few times higher in (n,f) than in (p,f). We consequently lowered them by the efficiency ratios of 3.3 (Rb), 2.0 (In) and 2.7 (Cs) for drawing Fig. 2 (this correction was not done in [11]).

4.2. Enhancement due to target thickness

The rate of fissions induced by secondary particles increases with the square of the thickness. These secondary particles are mostly neutrons produced via (p,xn) or accompanying fission. They add extra cross-section to fission with a (Z,A) distribution shifted towards the most neutron-rich isotopes. Fig. 3 shows ratios of yields for the HDR as a rod of 8.2 g/cm^2 thickness and as a powder of thickness of 1.9 g/cm^2 obtained by smashing the rod (thus the rod target is 4.3 times thicker than the powder). There is indeed a slight enhancement of yields of the most n-rich isotopes, which cannot be accounted for by invoking a shorter release-time since the releases curves measured for rod and powder targets are quasi identical.

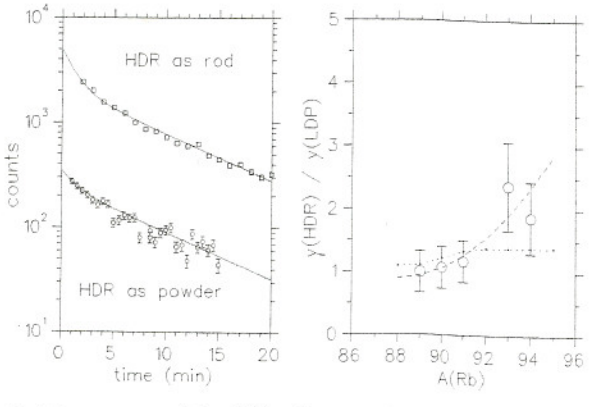

Fig. 3. Enhancement of the yields of very neutron-rich Rb isotopes with target thickness. l.h.s: Release curves at $T = 2050$ °C recorded with ^{139}Cs ($T_{1/2} = 9.27$ min). r.h.s: Yield ratios arbitrarily normalised. The dashed line merely gives a trend, whereas the dotted line is the calculated ratio of efficiencies, based on release times.

4.3. Long-lasting uranium carbide target

Carbonization of the tungsten outer target container leads to fast destruction. The long-lasting-target (LLT) unit differs from the early versions by the thicker wall of the W target container (from 0.25 mm to now 0.50 mm) and the fact that its contact with the inner graphite container is avoided. The tested target was 9.2 g/cm^2 thick. Yields and release curves were measured before the target was off-line continuously heated during three months at 2030 °C, and after. They remained the same. The less than 2% increase of target resistance suggests that the heating could have continued further without damage.

5. HDR versus PARRNe target material

The release-times measured for the HDR remain definitely longer than those reported by the PARRNe group at IPN-Orsay. In order to see whether this is an artifact of the different measurement method or not, the 'low-density' target material used at PARRNe (LDT, $\rho = 2.32$ g/cm^3, grains of 25–35 μ) was prepared according to the proper specifications and tested at IRIS. A new construction unit, in which ionisation only takes place in the container, was built for the purpose of comparison in the same geometry of HDR (now 6.7 g/cm^2) and LDT (2.8 g/cm^2 distributed among 8 pellets each of 1.2–1.6 mm). The HDR higher mass results in higher production for all nuclei measured, see Fig. 4. Yields of long-lived nuclei scale up more than target thickness, which again hints at fissions induced by secondary neutrons. The decrease of the excess for shortest-lived isotopes confirms that the HDR target has slower release than the LDT.

6. Small grain uranium carbide target

The above comparison suggested the use of a smaller grain material. The new high-density pellet (HDP, grain size 20 μ, $\rho = 12$ g/cm^3, thickness 6.3 g/cm^2 shared on

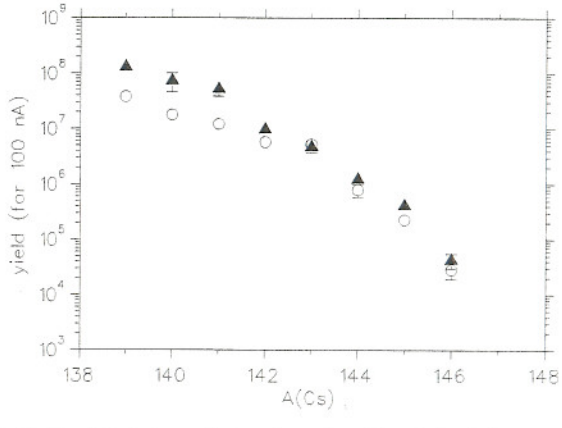

Fig. 4. Yields of Cs isotopes in ions/s produced by the ionizing target with HDR (6.7 g/cm^2) shown as triangles, LDT (PARRNE-like) at IRIS (2.8 g/cm^2) shown as circles.

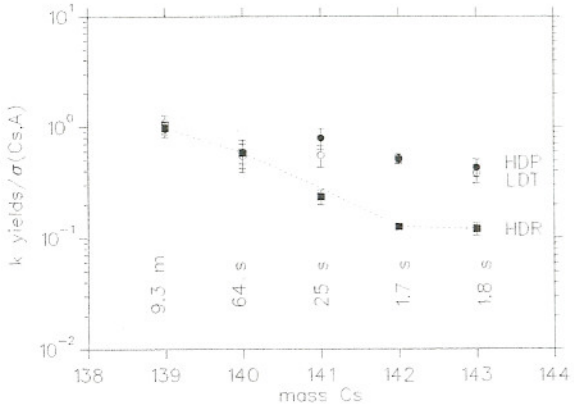

Fig. 5. Comparison of release efficiency for the reference HDR (closed squares), the LDT PARRNe-like (open circles) and the new HD Pellet target (closed squares). Note that experimental cross-sections [16] have been used and the scale has been adjusted to unity for the long-lived isotope.

3 pellets of thickness about 1.6–1.9 mm each) has been delivered in spring 2006. The target has been operated at 1700 °C due to a technical problem. Low ionisation efficiency explains why yields of long-lived nuclei were about 30 times lower than the best ones with the HDR target.

The release efficiency of the three targets compared in the same geometry is shown in Fig. 5. It was necessary to multiply by 2 all the yields for ^{142}Cs in order to get a smooth behaviour. This might point to a problem with spectroscopic input. The enhancements of neutron-rich isotopes due to secondary neutrons produced in the targets cannot be very different as the thicknesses of the HDP and LDT used for this test are comparable. Therefore, the almost overlapping trends of relative yield versus half-life suggest that the new HDP and standard LDT have very similar release properties.

A direct release-time measurement was made for HDP. The flattening, instead of a steeper rise, near the origin of time of the release curve of ^{130}Cs^{m} was fitted by the convolution of sequential diffusion and effusion with half-time constants of 5.4 s (representing either diffusion or effusion) and about 3 s (representing the other one), see [12]. This result suggests that one starts to see the effect of effusion, as diffusion has become much faster than with the original HDR material.

7. Outlook

We studied yields, efficiencies and release-times of UC_x high-density target material of grain size about 200 μ operated at temperatures of typically 2100 °C. This target was proved to function after 3 months of off-line heating without loss of its performance and allows higher yields than the low-density target in use at PARRNe-Orsay and CERN-ISOLDE for all Rb and Cs isotopes with half-life longer than one second, but its release is slower. This drawback is removed through a newly manufactured high-density UC material, now with a grain size of 20 μ. This material shall be further tested to characterize it fully.

Our lack of insight in the mechanism of release prevents us to predict the properties of a 1 kg target. Longer effusion path and increase of the number of secondary fissions work in opposite ways. Thus, in June 2006 the first on-line experiment with a thickness of uranium of 91 g/cm^2 of the new material was carried out. There was little loss of efficiency even for activities as short as a few seconds and this target was successfully tested for its long-term stability.

Acknowledgments

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