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Photofission for the production of radioactive beams: Experimental data from an on-line measurement

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Abstract. A PARRNe 1 experiment (Production d'Atomes Radioactifs Riches en Neutrons) aimed at the production of neutron-rich radioactive noble gases produced by photofission has been performed at CERN. The LEP Pre-Injector (LPI) has been used to deliver a 50 MeV electron beam. The results obtained show clearly that the use of an electron beam to produce neutron-rich fission fragments for futur RNB facilities is an option that should not be neglected.

PACS. 25.85.Jg Photofission – 29.25.Rm Sources of radioactive nuclei – 25.20.-x Photonuclear reactions

1 Introduction

The availability of intense neutron-rich isotope beams opens many new applications, *e.g.* detailed nuclear-structure studies in a yet unexplored region [1,2]. Several laboratories are focusing on studies aimed at producing high enough intensities to warrant a new generation of experiments. Facilities are planned to provide these radioactive beams, *e.g.*, the SPIRAL-II project at GANIL. Fission is a very powerful mechanism to produce a number of such beams. A large effort is done to investigate the production of neutron-rich isotopes in fission reaction [3, 4] or photofission [5]. As one possibility to achieve the highest possible luminosities without dissipating too much power in the fissioning target, high-energy neutrons can be produced by breaking up an intense deuteron beam in a dedicated and well-cooled converter and induce fission in a thick ^{238}U target.

The program PARRNe (Production d'Atomes Radioactifs Riches en Neutrons) at IPN Orsay is aimed at studying the parameters to optimize the production of radioactive beams from ^{238}U fission. The PARRNe 1 set-up was developed in order to measure online production of rare gases. These measurements can give relative and empirical information on the production of radioactive beams. In order to study the production of the other elements as well as the optimization of the target ion source unit, PARRNe 2, an isotope separator on-line has then been installed at the Tandem of IPN Orsay [6,7].

The influence of the energy of the deuterons (20 to 130 MeV) along with the nature of the converter (Be, C, U) has been studied within the European RTD program SPIRAL II. These "PARRNe 1" measurements were done at Orsay (20 and 26 MeV) [8], Louvain la Neuve (50 MeV), KVI (80 and 130 MeV) with an UC_x target [6] and at Orsay at 26 MeV with a molten U target [9]. The main advantage of the PARRNe 1 measurements is the use of an identical set-up at various accelerators. These experiments have demonstrated that fast-neutron–induced fission is a highly interesting method for future RNB facilities. However, this method requires the development of very intense primary beams (*i.e.* deuterons or protons) for the neutron production.

An interesting alternative to neutron-induced fission would be to use the bremsstrahlung-induced fission of uranium. Diamond [10] has calculated that $5 \cdot 10^{13}$ fissions/s could be produced in an optimal ^{235}U target or

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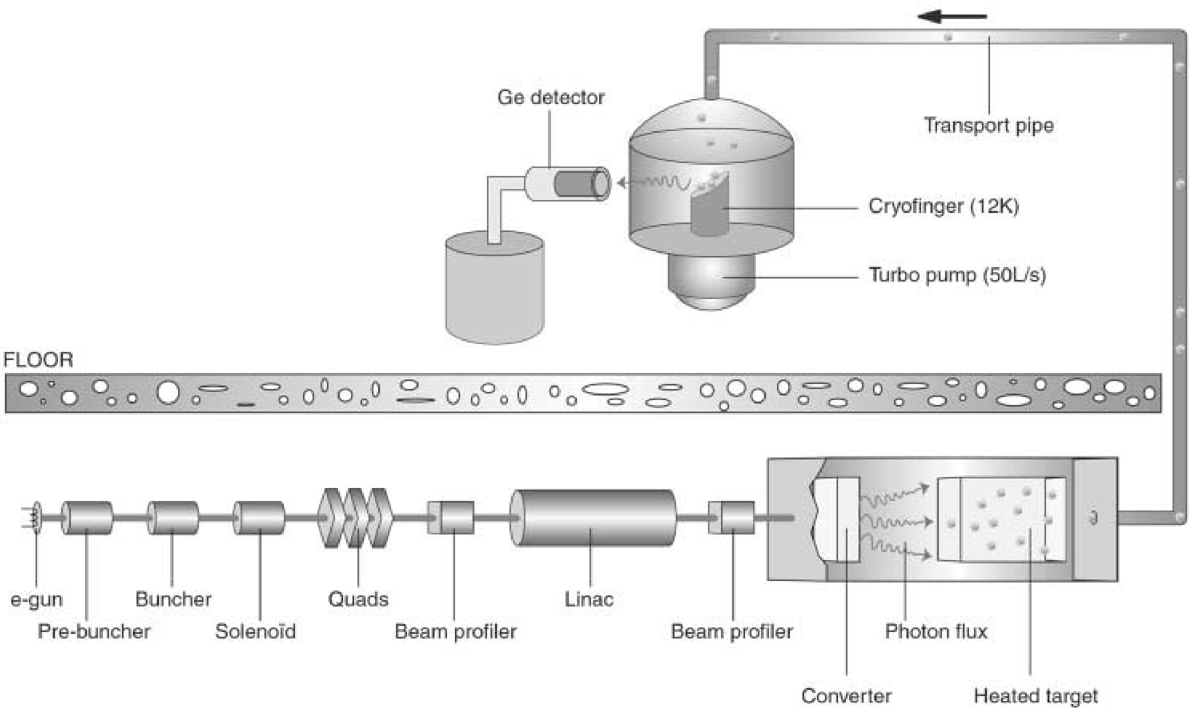

Fig. 1. Schematic view of the PARRNe 1 experimental set-up at LPI.

about 60% of that amount in a natural uranium target using a 100 kW beam of 30 MeV electrons. Oganessian *et al.* [11] report first encouraging off-line results with a low-energy (25 MeV) electron beam. The relatively low cost of a high-intensity electron accelerator compared to other primary-beam accelerators would, indeed, make this alternative very attractive.

2 Experimental procedure

Fission may be induced by photons exciting the Giant Dipolar Resonance (GDR) of the nucleus. In the case of ^{238}U, the fission cross-section at the GDR energy, which is around 15 MeV, reaches 0.16 barn [12]. The most common way for producing high gamma fluxes is the bremsstrahlung induced by passage of electrons through matter. The LEP Pre-Injector (LPI) at CERN has been used to deliver a 50 MeV electron beam. The beam current was supervised during the measurements using a gamma-ray monitor placed near the target. The electron beam of 50 nA hits a thin tungsten converter target of 4 mm thickness. The bremsstrahlung γ-rays irradiate the primary production target after a short path through the air. The target material was uranium carbide. This UC_x target has been prepared at ISOLDE-CERN with the routinely used carburation technique [13]. It contains 23 g of ^{238}U and was kept during the experiment in a ohmically heated graphite oven at 1800° C. The density of the target was 3.6 g/cm^3, with a ratio of 4 carbon atoms for 1 uranium atom. The 67 UC_x pills with 14 mm diameter were contained in a 106 mm long graphite container. The products of the uranium fission induced by γ-ray absorption are slowed down, stopped and become neutral atoms inside the thick target. Only the gaseous elements are able to diffuse out of the target, to be transferred through a 8 m stainless-steel tube and be collected on a cryogenic cold finger (12 K). A germanium detector was placed directly in front of the cryogenic finger and the decay of the collected radioactive noble gases has been measured by γ spectroscopy. Figure 1 presents the experimental set-up installed after the first section of the LPI at CERN. This experimental set-up is not dedicated to the measurements of fission cross-sections. Moreover, the PARRNe 1 measurements are dedicated to the study of noble gases which are not the best species to answer the important question like the role of the temperature of the fissioning nucleus on the final production of fission residues [14]. Nevertheless, the relative information obtained with these measurements are of primary importance for the production of radioactive beams.

3 Results

The measurements have been performed with the 4 mm tungsten converter in different positions (8 cm from the target and 4 cm from the target) (50 nA primary beam) and one measurement has been performed without converter; the 10 nA primary electron beam hitting directly the UC_x target. The idea was to obtain some information about the opening angle of the gamma-rays of interest, and the possible gain in the production of fission fragments with the irradiation geometry. Indeed, around 15 MeV, the main contribution to gamma absorption are e^+e^- pair production and the photonuclear reactions (γ, f), (γ, n) and $(\gamma, 2\mathrm{n})$. A pair production reaction may in a thick

Table 1. Production rate obtained for noble-gases nuclides with the converter placed at 8 cm (exp 1) or 4 cm (exp 2) from the target (primary-beam current 50 nA) and without converter (exp 3) (primary-beam current 10 nA).

Nucleus	Half-life	exp 1 /μC	exp 2 /μC	exp 3 /μC
^{89}Kr	189 s	1.9×10^6	3×10^6	1.1×10^7
^{90}Kr	32.3 s	1.1×10^6	1.6×10^6	7.9×10^6
^{91}Kr	8.6 s	2.6×10^5	3.3×10^5	1.3×10^6
^{92}Kr	1.8 s	4×10^4	4.8×10^4	2×10^5
^{137}Xe	229.1 s	1.9×10^6	3.1×10^6	9.2×10^6
^{138}Xe	844.8 s	2.1×10^6	3.3×10^6	1.4×10^7
^{139}Xe	39.7 s	6×10^5	8.9×10^5	3.9×10^6
^{140}Xe	13.6 s	1.9×10^5	3.1×10^5	2.1×10^6

target eventually lead to a fission through the resulting photon produced. In the same manner, the neutrons produced by (γ, n) and $(\gamma, 2\mathrm{n})$ reactions can also induce fission. All our measurements have been performed irradiating the target with the gamma-rays during one minute and then waiting 9 minutes for the decay of the short-lived nuclei on the cryogenic finger. The cycle was then repeated in order to obtain enough statistics.

All these experiments have been performed in the same experimental conditions so that the extraction efficiencies are the same in the different measurements. The release of different elements from a uranium carbide target as from a molten uranium target is described in ref. [7]. The results, corrected for the detection efficiency, obtained during this experiment are summarized in table 1. The data shown in this table were measured at the cryogenic finger and normalized as production rate per second per 1μA electrons (*i.e.* per μC of electron beam hitting the converter or the target, respectively). An average gain factor of 1.5 ± 0.2 have been obtained between the experiment performed with the converter placed at 4 cm from the target and the experiment performed with the converter at 8 cm from the target. The gain is even much larger when the experiment is performed without converter. The production is now 3.9 ± 0.6 times larger without converter than with the 4 mm thick tungsten converter placed at 4 cm from the target. Figures 2 and 3 show the results obtained during the three measurements for the Xe isotopes and the Kr isotopes, respectively. These results are compared with the results obtained at KVI with the same experimental set-up and conditions (*i.e.* with the same conditions of release for the noble gases) and with a deuteron beam of 80 MeV (50 nA primary beam). In this experiment, the deuteron-to-neutron converter was made of a 20 mm thick Be target and was placed at 8 cm from the UC_x target.

The formalism of ref. [15] was adapted to estimate the rate of fission events N_f produced in the target as a function of the initial electron beam intensity and the set-up geometry. The main changes can be briefly reviewed in the following. The number of fission events induced by the photons in the UC_x target using a 4 mm tungsten converter with 50 MeV electron beams is obtained by com-

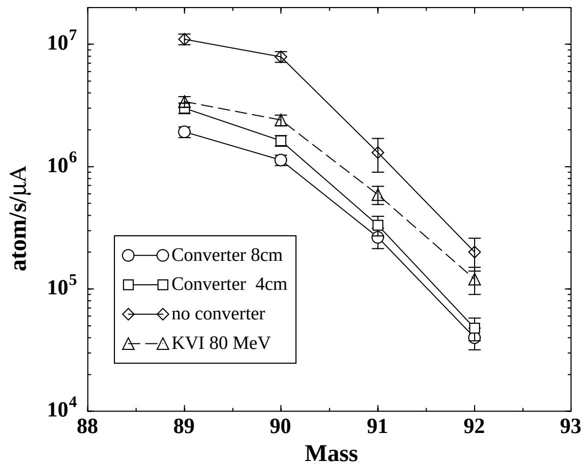

Fig. 2. Production rates obtained for Kr isotopes at LPI in different configurations compared with the results obtained at KVI with deuterons of 80 MeV (Be converter, 8 cm).

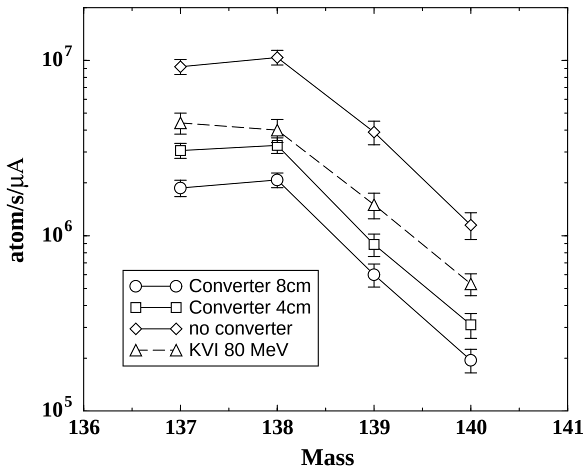

Fig. 3. Production rates obtained for Xe isotopes at LPI in different configurations compared with the results obtained at KVI with deuterons of 80 MeV (Be converter, 8 cm).

puting the following value:

$$N_\mathrm{f} = \int_{xy\theta\varphi} N_\gamma(\theta)\bar{\sigma}(\theta)\frac{[1-\exp(-\mu z)]}{\mu}\mathrm{d}x\mathrm{d}y\sin(\theta)\mathrm{d}\theta\mathrm{d}\varphi\,, \tag{1}$$

where

$$\bar{\sigma}(\theta) = \frac{1}{N_\gamma(\theta)}\int_{E_\gamma} N_\gamma(E_\gamma,\theta)\sigma(E_\gamma)\mathrm{d}E_\gamma \tag{2}$$

defines the mean value of the photofission cross-section at an angle θ,

$$N_\gamma(\theta) = \frac{1}{M_\mathrm{U}S_\mathrm{b}}\int_{E_\gamma} N_\gamma(E_\gamma,\theta)\mathrm{d}E_\gamma \tag{3}$$

is the number of photons emitted from the converter at an angle θ, $N_\gamma(E_\gamma,\theta)$ is the angular and energetic distribution of the bremsstrahlung radiation produced by the electron beam onto a tungsten converter deduced from

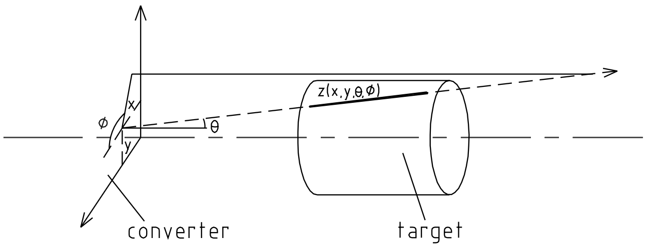

Fig. 4. Cylindrical geometry as used for the calculations of the rate of fission events N_f.

ref. [16], z is the path length inside the target of a photon emitted from a point (x, y) of the converter in a direction (θ, φ) (see fig. 4), S_b is the beam section, $\sigma(E_\gamma)$ is the cross-section for photofission obtained from ref. [10], M_U denotes the ^{238}U mass and μ is the γ absorption coefficient in UC_x extracted from ref. [11] and is taken as 0.061 cm^2/g.

The calculations have been performed for the three positions of the experiment. The calculation corresponding to the measurement referred without converter has been performed taking into account the same tungsten material in contact with the UC_x target. A gain by a factor of 2 has been calculated between the experiment with converters at 4 and 8 cm, and by a factor of 3 between the experiment with no converter and the experiment with a converter at 4 cm. From these results, the main contribution in the measured gain factors between the three different situations seems to originate from geometrical reasons. At this point, one can draw an important conclusion: for RNB production with photofission, it should be important to place the converter at a distance as short as possible from the target or even better, to hit the target directly with the electron beam. In order to compare the production of noble gases with rapid neutrons and photofission, we have estimated with the same code the rates in the best geometrical conditions (*i.e.* the converter placed in contact with the UC_x target). A gain of only 40% is obtained with the 80 MeV deuteron beam as compared to photofission.

4 Conclusion

The obtained results suggest photofission as an interesting alternative to rapid neutron-induced fission for the production of radioactive nuclear beams. However, in a complete comparison many other details have to be taken into consideration:

- The mass yield curve of fast-neutron–induced fission is much wider and has a smaller peak-to-valley ratio. Thus, it provides considerably higher yields for nuclei in the symmetric fission region. For the asymmetric fission region the yields are also higher for fast-neutron–induced fission at least for the less exotic nuclei, but this feature is not so clear for the most exotic ones since a colder process (*i.e.* photofission) minimizes neutron evaporation [17].
- High-energy neutrons can moreover be used for other applications, *e.g.*, for (n, xp) reactions which provide a unique tool to produce neutron-rich nuclei “south” of a variety of targets [18].
- While the deuterons are completely stopped in a converter and only the neutral neutron beam hits the ISOL-type target, in the case of photofission much more energy is deposited in the ISOL-type target, either directly by the primary electron beam (when running without converter) or by pair creation from the secondary photon beam (when using a converter).
- To reach the highest number of fissions in the ISOL-type target, other interesting possibilities exist (*e.g.*, neutrons from a spallation neutron source) which have to be investigated. A detailed comparison of different methods to attain, *e.g.*, 10^{15} fissions per second is presently being done in the frame of the EURISOL project [19].

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