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Production of lanthanide molecular ion beams by fluorination technique

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Abstract. Systematic off-line fluorination studies on all the stable lanthanide isotopes have been performed. The results are presented as a function of various parameters such as the target temperature, the type of ion source used (hot plasma or surface ionization) and the quantity of CF_4 introduced. The first on-line measurements allowed us to determine the optimal experimental conditions for producing radioactive lanthanide isotopes.

1. Introduction

The neutron-rich lanthanide (Ln) nuclei allow to study, in exotic nuclei, dynamical phenomena typical of mid-shell. They offer the opportunity to observe octupole excitations and their coupling to quadrupole excitations. They can provide information on the very puzzling nature of the 0^+ states lying at low excitation energy [1–5]. Moreover large quadrupole deformations are expected in the ground state of the neutron-mid-shell nuclei: for example, ^{170}Dy , having the largest number of valence particles among the $A < 208$ nuclei, is predicted to have the most deformed ground state [6]. Finally, these nuclei present an astrophysical interest. The small peak exhibited at $A = 160$ in the solar r-process abundance distribution is probably due to a subtle interplay between nuclear deformation and β decay then half-lives (or deformation) determinations via β decay measurements in the neutron-rich Ln nuclei can help to constrain the r-process freeze out conditions [7]. Until now the neutron-rich Ln nuclei have been little studied since they are difficult to produce. For example, in ISOL facilities, they are produced by fission induced in thick uranium-carbide (UC_x) targets from which they are slowly released due to their low volatility. In previous works, CF_4 injection in the target leads to form Ln fluorides that are more volatile than better released than the elements [8]. However, the Ln-fluoride release efficiency depends strongly on the impurities in the target, on the amount of CF_4 introduced and



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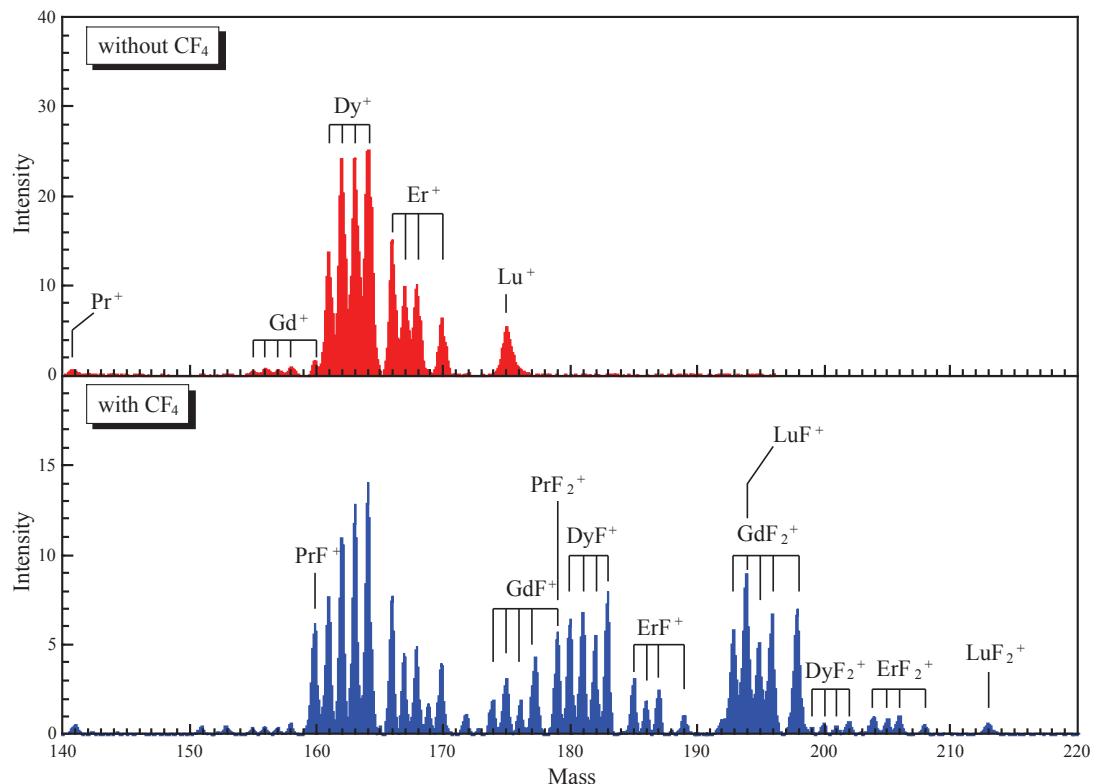


Figure 1. Mass spectra recorded in the focal plane of the off-line separator magnet.

on the temperature of the target [9,10]. In order to control and apply the fluorination technique at ALTO [11], we have performed systematic off-line fluorination studies on all the Ln stable isotopes. The experimental procedure developed, the off-line results as well as the first on-line measurements are detailed hereafter.

2. Experimental procedure

2.1. Off-line measurements

The fluorination tests on the Ln stable isotopes were performed with an off-line mass separator and either a hot plasma MK5 [12] or a surface ionization (SI) [13] ion source that are both commonly used at ALTO. Pellets containing uranium cannot be used on the off-line separator for safety reasons. The UC_x pellets used as targets at ALTO have graphite excess. Then in order to observe Ln diffusion in realistic conditions, we used pellets made of powder mixture of graphite with different sets of Ln oxides. The lanthanides were selected so that the molecular or elemental ions expected to be observed do not present any mass overlap: for example, La, Pr, Eu, Ho for the first series, La, Pr, Eu, Gd, Tb, Ho and Ce, Pr, Sm, Eu, Yb, for the second and third ones. Eu is one of the most volatile Ln elements and has only two stable isotopes, therefore it was used as a reference in all our measurements. Most of the Ln elements have been tested several times in order to control the reproducibility of the results. The powder mixture, put in a 13 mm diameter die, was pressed at 220 MPa. Each pellet weighed 275 mg and contained 50 mg of one of the Ln oxide mixes. For the tests, five pellets of a same mixture were placed inside the oven of the target and ion source system. In each case we measured the current collected in a Faraday cup located behind the separator magnet for the different masses corresponding to the Ln^+ , LnF^+ and LnF_2^+ ions as a function of the target temperature, the parameters of

the ion source and the quantity of CF_4 injected via a tunable calibrated leak. As a control, we registered the mass spectra obtained in the focal plane of the off-line mass-separator magnet. Figure 1 shows such spectra. For each element, one peak is observed for each stable isotope with relative intensities reflecting the isotopic ratio. Ln are observed as elemental ions without CF_4 and also as molecular ions when CF_4 is injected. The Pr and Gd intensities are very weak for the elemental ions and much stronger for the molecular ions, which proves that fluorination improves the Ln release from thick pellets.

2.2. On-line measurements

The first on-line measurement was performed with a MK5 ion source. For normalization with off-line results and in order to attest the proper running of the fluorination system, five graphite pellets doped with stable lanthanides, identical to that used in the off-line tests, were added to the UC_x pellets used as targets for the photo-fission reactions. No radioactive Ln isotopes were observed, making crucial a second experiment.

The second on-line measurement, without fluorination, was performed using a SI ion source, with a two-fold purpose. Firstly, in order to check the Ln cross sections by photo-fission, the yields of some isotopes of the most volatile Ln elements were measured. Secondly, to compare the Ln release from graphite and UC_x pellets, five UC_x pellets doped with the previous mixture of stable Ln oxides, in the same amount as in the first on-line experiment, were placed in the target oven. These UC_x Ln-doped pellets were made using a 14 mm diameter die and a pressure of 190 MPa.

The intensity of the stable ion beams was measured by a Faraday cup located behind the magnet of the on-line mass separator tuned on the Ln^+ , LnF^+ and LnF_2^+ masses. Mass spectra were recorded by scanning the field of the separator magnet. The yields measurements were carried out by γ and β - γ spectrometry.

3. Experimental results

3.1. From the off-line tests

Whatever the experimental conditions, the stable Ln beam intensity exhibits high sensitivity to the target temperature and the CF_4 flow injected. Figure 2 shows the results obtained for both ion sources with target temperatures around 2000 °C. The beam intensities measured with the SI ion source are clearly higher than those measured with the MK5 ion source. Other differences are observed between the two ion sources.

With the MK5 ion source:

- for $T_{\text{target}} < 1630$ °C, only Eu is observed, as Eu^+ and as EuF^+ when CF_4 is added; the other elements begin to appear as molecular ions when $T_{\text{target}} = 1630$ °C and a CF_4 amount of 6×10^{-3} mbar·l/s is introduced in the target and ion source system;
- for $T_{\text{target}} \geq 1900$ °C and in particular at 2030 °C (see fig. 2), the more CF_4 injected, the higher the intensity of the lanthanide beams ; the CF_4 maximum quantity compatible with good running conditions of the ion source was determined to be 6×10^{-3} mbar·l/s;
- the type of ions observed preferentially (Ln^+ , LnF^+ or LnF_2^+) appears to be highly correlated with the valence of the given lanthanide and thus with the type of fluoride formed (mono-, di- or tri-fluoride), for example La, Ce, Pr, Tb and Ho are preferentially observed as LnF_2^+ , Eu and Sm as LnF^+ , Yb as Ln^+ , except Gd observed preferentially in the Gd^+ form whereas GdF_2^+ was expected. The LnF_n^+ abundance distributions generally reflect the Ln ionization energy. These variations across the lanthanide series are correlated to the Ln specific electronic configurations [14].

With the SI ion source:

- for $T_{\text{target}} \geq 1900$ °C and without CF_4 , in addition to Eu, other Ln elements as Pr, Nd, Er, Lu, Dy and Tm are observed in the Ln^+ form, this is probably due to the higher ionization

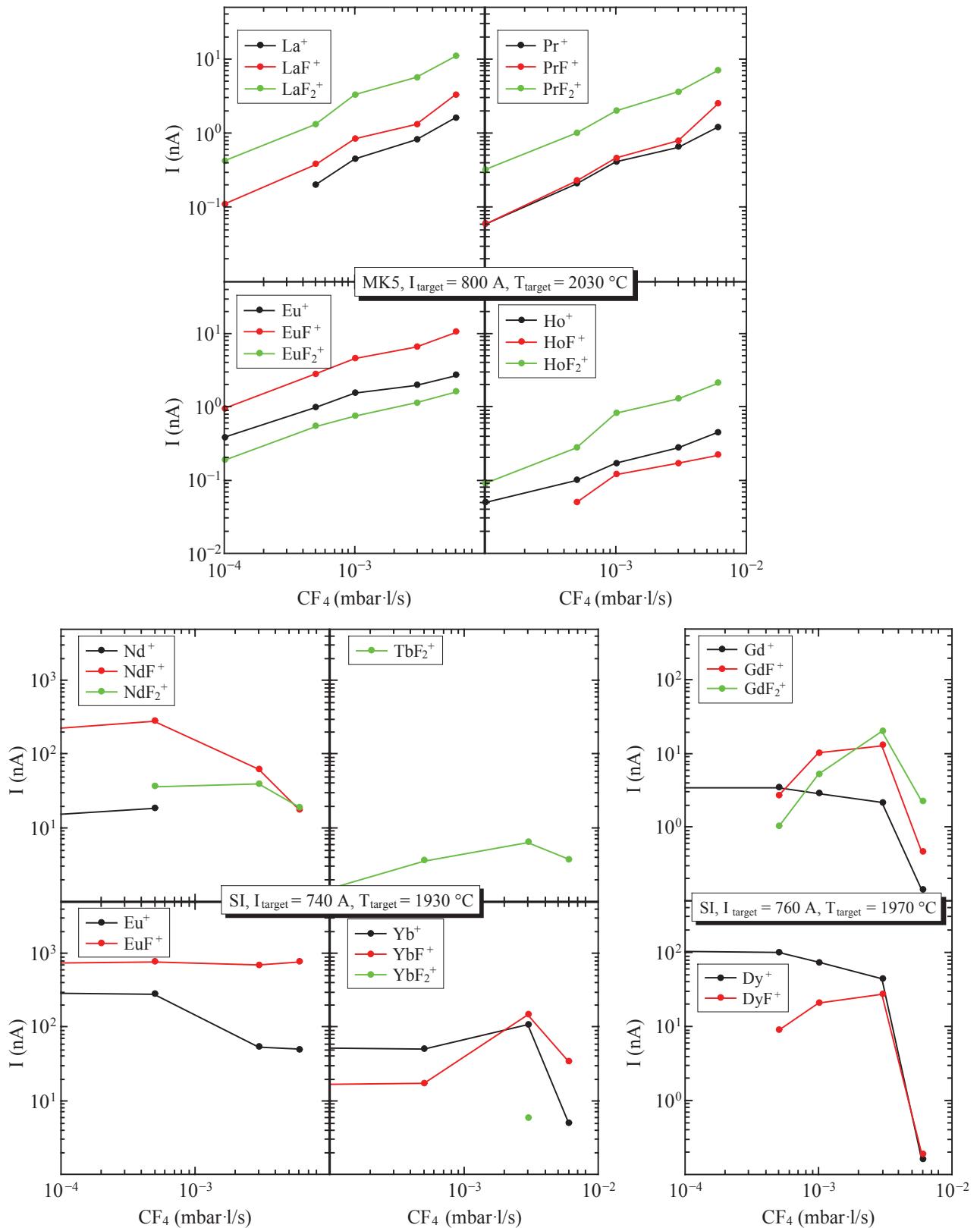


Figure 2. Ion beam intensity measured during the off-line tests as a function of the quantity of gas injected for various lanthanides in their elemental and molecular forms using MK5 or SI ion source.

efficiency of the SI ion source (compared to the MK5 one) for Ln; with CF_4 , when the gas flow is high the beam intensities collapse (see fig. 2), this feature is not well understood but has already been observed in other fluorination works [15];

– the type of ions observed preferentially (Ln^+ , LnF^+ or LnF_2^+) depends strongly on the CF_4 amount introduced; for instance, with a flow lower than 3×10^{-3} mbar·l/s, Yb and Dy are observed as Ln^+ , Nd as NdF^+ whereas with a CF_4 flow equal to 6×10^{-3} mbar·l/s Yb and Dy are observed as LnF^+ and Nd as NdF_2^+ (see fig. 2);

– the correlation between the valence of the given element and the type of ions preferentially observed (Ln^+ , LnF^+ or LnF_2^+) is only reached under extreme running conditions ($T_{target} \sim 2000$ °C and 6×10^{-3} mbar·l/s CF_4), as shown in fig. 2 for Yb and Gd.

One can reasonably assume that the diffusion of stable Ln isotopes in the graphite pellets and the formation of molecular compounds do not differ depending on whether the MK5 or SI ion source is used. On the contrary, the mechanisms of molecular-compound breaking and of molecular-ion formation seem to be different with the MK5 or SI ion source. The behavior of the Ln fluorinated compounds in the MK5 ion source appears to be simpler and quite consistent with what is expected from the Ln chemical properties. This is the reason why the MK5 ion source was used for the first fluorination on-line experiment.

3.2. From the on-line measurements

3.2.1. with the MK5 ion source

Figure 3 shows the stable La and Pr ion beam intensities released by the graphite pellets without and with injection of CF_4 in the target and ion source system. The LnF_2^+ beam intensities are significantly lower in the first measurement with CF_4 than in the next ones; this shows that a reaction time, equal to \sim half an hour, is required before obtaining stable running conditions. The results observed are very similar to those obtained off-line (compare figs. 2 and 3). Indeed when CF_4 is introduced, the beam intensities measured increase strongly, by a factor of up to 100. For both elements, the LnF_2^+ molecular ions are observed preferentially, with intensity very close to that observed off-line (see figs. 2 and 3). This shows that the gas inlet system was working properly. Nevertheless the electronic components of the tunable leak were more sensitive than expected, preventing us from measuring the flow of gas introduced, this part of the system remains to be improved. However, the running conditions of the target and ion source

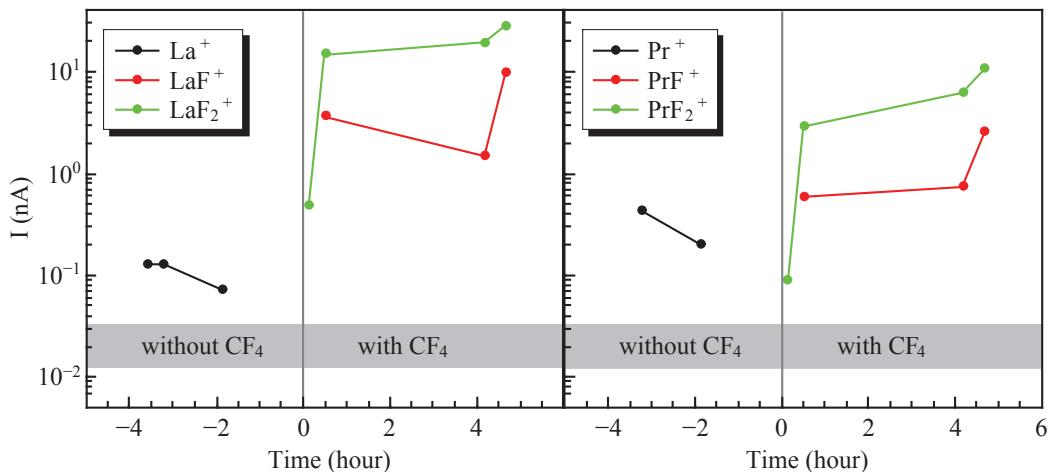


Figure 3. Ion beam intensity measured during the on-line experiment performed with a MK5 ion source for La and Pr stable isotopes without (elemental ions) and with (fluorinated ions) CF_4 . Time $t = 0$ indicates the beginning of the CF_4 injection.

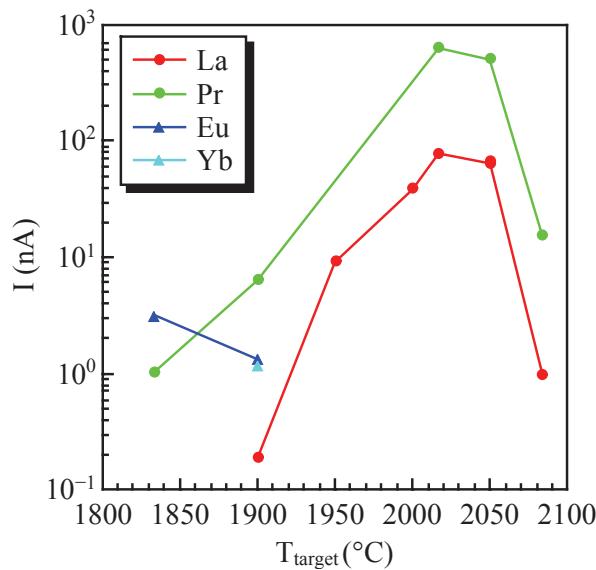


Figure 4. Ion beam intensity measured during the on-line experiment performed with the SI ion source for La, Pr, Eu and Yb stable isotopes released by UC_x pellets without CF_4 .

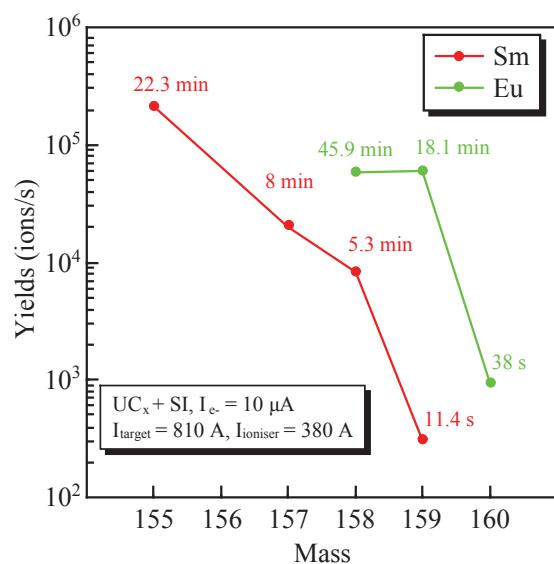


Figure 5. Yields measured for Sm and Eu isotopes released by UC_x pellets during the on-line experiment performed with the SI ion source.

system, especially the target temperature ($I_{target} = 750 A$, i.e. $T_{target} \sim 1950$ °C) but also the CF_4 flow, seemed to be close to that defined off-line as the optimal ones.

Concerning the radioactive species, the measurements performed to observe the Ln isotopes were all unsuccessful: not only those carried on the 158, 160 masses in order to observe some of the most volatile lanthanides, Eu and Sm, in their elemental form but also those on the 179, 182 and 191 masses in order to see ^{160}Eu , ^{144}La and ^{153}Pr isotopes in the mono and di-fluorinated molecular forms, respectively.

3.2.2. with the SI ion source

Figure 4 presents the La, Pr, Eu and Yb ion beam intensities released by UC_x pellets doped with stable Ln. It is worth noting that Eu and Yb, belonging to the most volatile lanthanides, were probably nearly totally released before the target temperature reached 1800 °C. On the contrary La and Pr were kept in the target until $T_{target} = 1800$ °C. Their beam intensity measured increases strongly with the target temperature. Between the two last measurements performed at $T_{target} = 2050$ and 2085 °C, the target was kept at high temperature ($T_{target} = 2050$ °C) during one week, then most of the stable lanthanides added in the UC_x pellets could have been released, which probably explains the intensity decrease from $T_{target} = 2050$ °C observed in fig. 4. The comparison with previous measurements indicates that the lanthanide release efficiencies are quite similar for graphite and UC_x pellets. Indeed, at a given target temperature, the intensities measured from UC_x are very similar to those obtained from graphite. Moreover the ionization efficiency of the surface ionization ion source is, depending on the element studied, greater by a factor of 50 to 100 than that obtained with the MK5 ion source.

Figure 5 shows the yields measured for the most volatile Ln isotopes released without fluorination by UC_x pellets heated at ~ 2050 °C. The spectroscopic data required to extract the yields from the γ spectra, in particular the half-lives and the absolute γ intensities, were taken from refs.[16–20]. This result proves that the Ln photo-fission cross sections are sufficient to allow

the observation of Ln mass-separated ion beams provided that the target temperature is high enough. The observation of low volatile Ln radioactive isotopes should then be possible at such high temperature with fluorination.

4. Conclusions

We performed the first systematic study on fluorination of lanthanides in SI and MK5 ion sources. The off-line tests carried out using graphite pellets doped with stable lanthanides showed that adding CF_4 in the target and ion source system increases the Ln release by a factor of 10 to 100 and that most of the lanthanides are preferentially observed as di-fluorinated ions except Sm, Eu and Yb that appear as mono-fluorinated ions. These features are more pronounced with the MK5 ion source than with the SI one, which seems to indicate that the processes involved in fluorination differ between the two ion sources and are simpler within the MK5 ion source. We showed that the release properties of lanthanides from thick uranium carbide pellets are similar to that obtained from graphite pellets. With the SI ion source the Ln ionization efficiency is found to be higher by a factor of 50 to 100 compared to that obtained with the MK5 ion source. The off-line tests and the first on-line experiments allowed us to determine the experimental requirements for producing lanthanide radioactive isotopes: the target temperature is really a key parameter and has to be as high as possible ($T_{\text{target}} \geq 2050$ °C); moreover we have to use the SI ion source in order to achieve maximum efficiency. In these conditions, we succeeded in measuring, without fluorination, the yields of some isotopes of the most volatile lanthanides. Before performing a new on-line fluorination experiment, it remains to improve the gas-inlet system using a well-chosen fixed calibrated leak in order to obtain more secure running conditions.

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