On the feasibility of online terbium extraction at ISOL@MYRRHA

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Abstract

Terbium is an element that has four isotopes with interesting properties for medical applications, $^{149, 152, 155, 161}$Tb. These radioisotopes are however far from being sufficiently accessible, thereby hindering the pursuit of research on radiolabelling as well as clinical or preclinical investigations. Their lack of market availability is explained by difficulties in producing these radioisotopes with high purity and specific activity. While ^{161}Tb can be produced using neutron capture in nuclear reactors, for $^{149, 152, 155}$Tb, a production route involving the ISOL technique is under study within the Tb-IRMA-V project. The ongoing R&D towards the production and extraction of these isotopes from an ISOL target at the ISOL@MYRRHA facility is reported in this contribution.

Introduction

This short contribution addresses a first-order study of Tb-isotopes production at ISOL@MYRRHA[1] as illustrated in the poster presented at EMIS[2] conference. Four Tb isotopes have been identified with interesting properties for medical applications, $^{149, 152, 155, 161}$Tb [1]. These radioisotopes are however not sufficiently accessible, thereby hindering the pursuit of research on radiolabelling as well as clinical or preclinical investigations. The lack of market availability of these isotopes stems from difficulties in their production with high purity and specific activity [2]. While ^{161}Tb can be produced using neutron capture in nuclear reactors the other isotopes require other production pathways. For $^{149, 152, 155}$Tb, a production route involving the ISOL technique is under study within the Tb-IRMA-V project. Tb-IRMA-V is a collaboration between SCK CEN, KU Leuven and CERN with the aim of producing terbium based radiopharmaceuticals. A first study for the ISOL production of these isotopes with 100-MeV protons and on-line release from the target material is discussed here.

Production

In the ISOL method, nuclides of interest are produced from nuclear reactions induced by the impact of projectiles on a target and subsequently released through diffusion and effusion from the target [3]. The released nuclides undergo ionization and mass separation processes that allow to select a specific nuclide. The presence of selective release processes marks a difference between the ISOL method and a classical direct production where radiochemical separation is not able deal with the co-produced radioisotopes. Difficult to obtain, enriched targets are therefore needed e.g. ^{152}Gd(p,4n)^{149}Tb and ^{155}Gd(p,4n)^{152}Tb [4, 5].

The first objective of this study was to select the target material best suited for production of the desired Tb isotopes. In the framework of ISOL@MYRRHA phase 1, the following parameters are to be accounted for : 1- 100-MeV proton beam, 2- On-line extraction. To produce Tb with a 100 MeV proton beam this means that candidate targets are limited to Gd, Tb (for the release of precursors), Dy, Ho and Er. These are all lanthanides close to Tb, the element of which radioisotopes are to be produced. Using the evaluated nuclear cross-section data from TENDL [6] the production of the desired Tb isotopes is compared between possible target material candidates (see Figure 1). The energy range of interest is defined as 70 ~100 MeV, stemming from the heat dissipation capacity of targets and is a range where experimental cross sections are scarce. Analyzing the cross-section data in this energy range leads first to narrowing down the list of candidates. Indeed, cross-section values for Ho and Er are two to three orders of magnitude lower than for the other candidates.

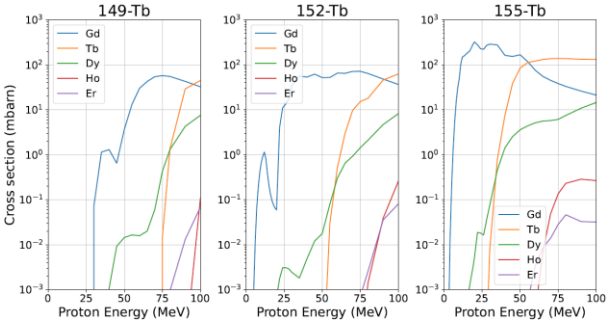

Figure 1: Production cross sections for candidate target materials

It is also notable that Gd and Tb display the highest cross sections but the release of Tb isotopes from a Tb target is not an option. However, dysprosium and Holmium precursors can be released prior to them decaying into the isotopes of interest. In light of this, the shortlisted targets ie. Gd, Tb and Dy have been compared, considering the production path that is the most suited for each candidate target-material. From a Gd target, this means the release of the Tb isotopes, from a Tb target, the release of Dy precursors and from a Dy target, this means the release of Ho precursors (Figure *2*).

[1] ISOL@MYRRHA is a radioactive ion beam facility under construction at SCK CEN in Mol, Belgium, as part of the MYRRHA ADS. In phase 1, the facility will operate with proton beams of 100 MeV and intensities up to 500 µA.

[2] International Conference on Electromagnetic Isotope Separators and Related Topics https://indico.ibs.re.kr/event/469/

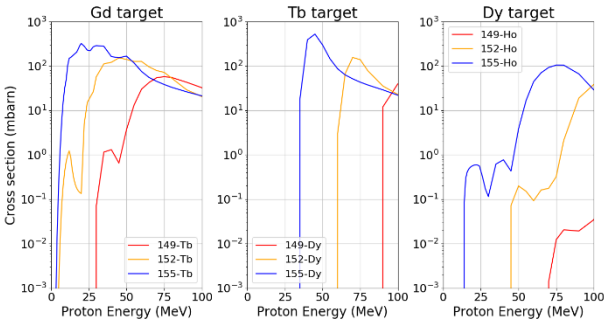

Figure 2: Production cross sections for the most suited path in each shortlisted candidate target-material

At this point, taking the different production paths into account, Gd and Tb still appeared to be the most interesting target-materials. Then, looking a bit further one realizes that most of the $^{149,\ 152}$Tb produced from a Gd target is expected to be in a metastable state (see Table 1) that decays significantly to nuclides other than the corresponding ground state. On the other hand, production in a metastable state is only expected for ^{149}Dy from a Tb target and 99.3 % of ^{149m}Dy ($t_{1/2}$ = 490 ms) decays to ^{149g}Dy ($t_{1/2}$ = 4.2 min) which in turns decays 100% to ^{149g}Tb. Besides 78.9% of the produced ^{152}Dy decays to ^{152g}Tb. This gives an edge to the Tb target, but not for ^{149}Tb due to the low production of ^{149}Dy.

Table 1: Cross section values for different production paths integrated between 70 and 100 MeV[6].

Production route	State	Integrated cross section (mbarn.Mev)
^{nat}Gd → ^{149}Tb	g	125
	m	838
^{nat}Gd → ^{152}Tb	g	480
	m	1126
^{nat}Gd → ^{155}Tb	g	1073
^{nat}Tb → ^{149}Dy	g	2.69
	m	0.519
^{nat}Tb → ^{152}Dy	g	2109
^{nat}Tb → ^{155}Dy	g	1158

An estimation of the in-target production rates has been conducted for the 500 µA proton beam foreseen at ISOL@MYRRHA on a 7 g/cm^2, 2 cm radius and 20 cm long target of either material (Table 2). These results indicate that, with an extraction efficiency of ~1%, the collection time for single treatments quantities of $^{149,\ 152}$Tb is 1~5 hours. This derives from literature references for single treatment quantities, namely 6 MBq of ^{149}Tb for mice [7] and 140 MBq of for a human [8]. The release of the produced isotopes from the target material is the next point of attention as this is one of the limiting steps of the extraction. Besides, in this case, the target material and the isotopes of interest belong to the same chemical group and thus, feature similar chemical properties.

Table 2: Estimated in-target production rates (FLUKA [9]) with the 500 µA proton beam foreseen at ISOL@MYRRHA with statistical uncertainties lower than 10^{-4} %)

Isotope	In-target Prod. Rate (#/s)
Gd target	
^{149}Tb	2.6 10^{12}
^{152}Tb	5.6 10^{12}
^{155}Tb	3.8 10^{12}
Tb target	
^{149}Dy	8 10^{8}
^{152}Dy	9 10^{12}
^{155}Dy	4.5 10^{12}

Release

The release of isotopes from an ISOL target is influenced by several factors (target material structure, pellet dimension, target container volume, transfer line, temperature,...) [10]. However, before proposing a concept of the target and performing the detailed analysis, the specific challenge of releasing a lanthanide from a target material made of another lanthanide is addressed in this contribution. Due to the lower primary-beam energies, refractory target materials (e.g. Ta [11]) are not suitable at ISOL@MYRRHA for the production of Tb isotopes, and one is limited to the use of lanthanides, which offer appropriate cross sections.

Molecular sideband extraction

Owing to a similar volatility of lanthanides, a selective release out of either of the two promising target-materials is not granted. Improving the release selectivity through molecular side bands has therefore been studied. Fluoride and chloride being literature recommendations for lanthanides [12], they have been considered. However, these recommendations are derived from the return of experience at ISOLDE where the use of a high energy proton beam allows the target material and elements of interest to have dissimilar chemical properties. In this case, both are lanthanides and a very similar affinity to fluorine and chlorine was expected. Thermochemical simulations based on the minimization of the systems Gibbs energy have been run [13] with as input the inventory derived from FLUKA [9] simulations of the target materials.

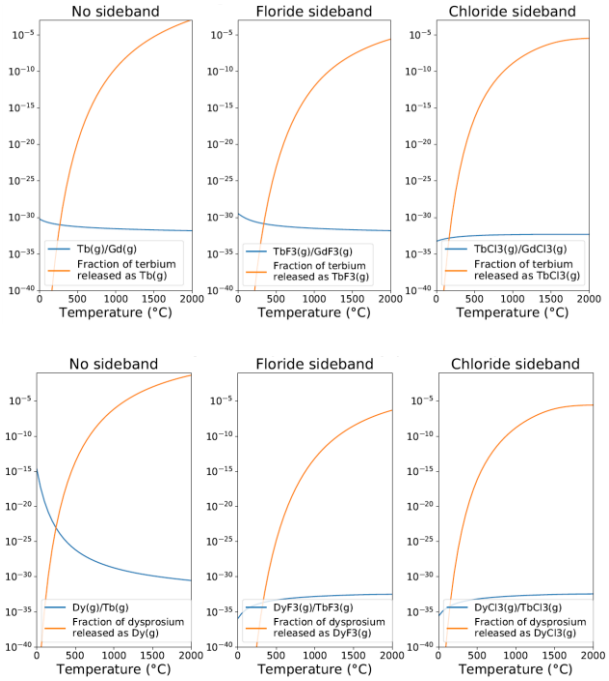

Figure 3: Simulated equilibrium composition of systems representative of the promising targets chemistry. (top) Gadolinium, (bottom) Terbium. Orange curves indicate the fraction of different compounds released in gas form. Blue curves indicate the ratio in the gas of the compounds of interest to contaminants from target evaporation.

Compared to the case with no sideband, the results (Figure 3) indicate no improvement of the release selectivity. The fraction of Tb (respectively of Dy) in the gas form from a Gd (respectively a Tb) target is actually higher in the case with no sideband, indicating a lower volatility of the corresponding fluoride and chloride molecules.

In an attempt to find a better suited sideband, a broader and more systematic search has been conducted. All the compounds identified in literature and in the HSC database that involve gadolinium, terbium and dysprosium were collected and compared. The objective of the comparison was to identify compounds uniquely reported for either the target or the element to be released. The result of this study was that only for two compounds of Gd (i.e. Gd_2O_3*WO_3, Gd_2O_3*$2WO_3$) there was no corresponding compound found with Tb. These are however complex and thus impractical since many simpler oxides of both Gd and Tb form and dominate in a chemical system involving oxygen. An alternative technique has thus been considered.

Temperature control of the transfer-line

The control of the transfer-line temperature is another technique considered for improving the release selectivity out of the promising target-materials. This is prompted by differences in adsorption enthalpy mainly between Tb ($\Delta H_{ads}^{SiO_2}$ ~-324 kJ/mole [13, 14], ΔH_{ads}^{Ta} ~-572 kJ/mole [15]) and Dy ($\Delta H_{ads}^{SiO_2}$ ~-241 kJ/mole [13, 14], ΔH_{ads}^{Ta} ~-472 kJ/mole [15]). Tailoring the transfer-line temperature allows to delay the migration of the most adsorptive species.

Based on the adsorption enthalpy values, the transport of Tb and Dy particles inside a transfer tube has been simulated with a Monte Carlo code to assess the separation capacity of the technique. Simulating a temperature gradient tube for a time period much longer than both the mean sticking time and the flight time, the obtained results are deposition curves (Figure *4*).

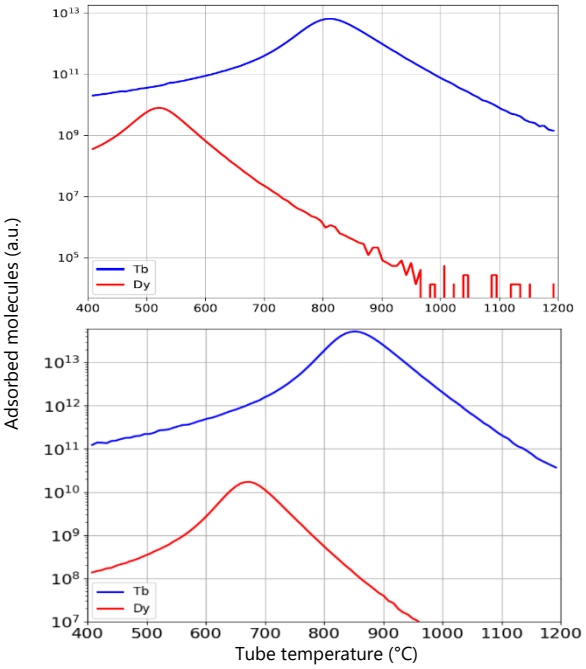

Figure 4: Surface adsorption of Tb & Dy, (Top) on SiO_2 and (bottom) on Ta

The curves indicate that the deposition peak of Tb is located at ~850°C while that of Dy is at ~670°C on a tantalum surface. Yet, because there will be ~10^4 times more target atoms of Tb than Dy, the tail of the Tb peak overwhelms the Dy peak. This is less the case with a SiO_2 surface where the peaks are further apart. Though SiO_2 is the better choice, both transfer tube materials lead to a purity improvement if the temperature of transfer-line tip can be set to a value in-between the deposition peaks.

Conclusion

Online separation of lanthanides at medium energy proton beam facilities is highly challenging. Promising target materials for the production of $^{149,\ 152,\ 155}$Tb isotopes have been identified. Improving the release selectivity through molecular sidebands appears unlikely at this point. However, combined with the right transfer tube material, temperature control of the transfer-line constitutes a more promising alternative.

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