HIGH POWER MOLTEN TARGETS FOR RADIOACTIVE ION BEAM PRODUCTION: FROM PARTICLE PHYSICS TO MEDICAL APPLICATIONS

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Abstract

Megawatt-class molten targets, combining high material densities and good heat transfer properties are being considered for neutron spallation sources, neutrino physics facilities and radioactive ion beam production. For this last category of facilities, in order to cope with the limitation of long diffusion times affecting the extraction of short-lived isotopes, a lead-bismuth eutectic (LBE) target loop equipped with a diffusion chamber has been proposed and tested offline during the EURISOL design study. To validate the concept, a molten LBE loop is now in the design phase and will be prototyped and tested on-line at CERN-ISOLDE.

This concept was further extended to an alternative route to produce 10^{13} ^{18}Ne/s for the Beta Beams, where a molten salt loop would be irradiated with 7 mA, 160 MeV proton beam. Some elements of the concept have been tested by using a molten fluoride salt static unit at CERN-ISOLDE. The investigation of the release and production of neon isotopes allowed the measurement of the diffusion coefficient of this element in molten fluoride salts.

INTRODUCTION

New findings in the fields of particle and nuclear physics are expected to come from progresses in accelerator technology. The need for intense secondary beams is the driving factor behind the development of new facilities exploiting primary beams with megawatts of beam power and, consequently, target interfaces that can withstand such large powers. In the last years, research and development work in high power targets has experienced important advances. Special attention has been devoted to megawatt-class molten targets for future neutron spallation sources, neutrino physics and radioactive ion beam (RIB) facilities [1-3] and new systems have been proposed and developed, notably during the EURISOL project [1].

In particular for RIB production, molten materials are extremely attractive since they can provide the highest intensities for isotopes of certain elements due to high material density [4]. Moreover, these materials are not subject to radiation damage and exhibit good heat transfer properties, which allow the simplification of the cooling system. Notwithstanding these advantages, targets based on molten materials suffer from long diffusion times, which strongly affect the extraction of short-lived isotopes. In this context, molten target loops, equipped with a diffusion chamber, are proposed in order to reduce the diffusion times and accommodate high beam powers [5]. Figure 1 shows a schematic representation of such proposal.

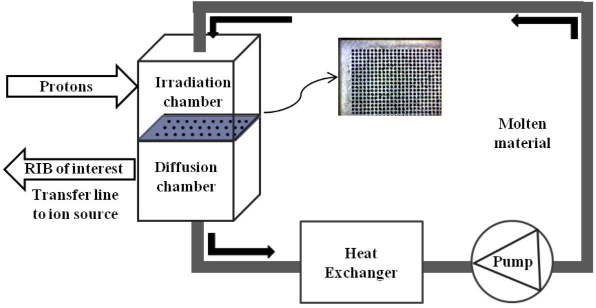

Figure 1: Schematic representation of a molten target loop: the material is irradiated in a dedicated volume; it goes through a metallic grid where a shower is created, facilitating the diffusion of the produced RIBs in a diffusion chamber. The material is kept circulating with a pump. The loop also includes a heat exchanger.

Table 1: Proposed molten target loops, driver beam characteristics and respective RIBs of interest

Material	Energy [GeV]	Power [kW]	RIBs	Project
LBE	1	100	$^{177-186}$Hg	EURISOL
LBE	1.4	2.8	$^{177-186}$Hg	LIEBE
NaF:LiF	1.4	1.1	^{18}Ne,^{11}C	Beta Beams/ PET isotopes
NaF:LiF	0.16	1100	^{18}Ne	Beta Beams

In the direction of increasing primary driver beam intensities, a lead bismuth eutectic (LBE) target loop has been proposed and tested offline at IPUL, Latvia, by Noah and collaborators [1], during the EURISOL design study. The proposed design would allow accommodating 1GeV, 100 kW CW proton beam. A molten LBE loop is now developed within the project LIEBE (Liquid Eutectic Pb Bi target loop for EURISOL). This target loop is in the design phase and will be prototyped and tested on-line at CERN-ISOLDE with 2 µA, 1.4 GeV pulsed proton beam.

A molten loop has also been proposed to produce high intensity ^{18}Ne beams [2,3] for the Beta Beams [6], a large scale facility aiming at the production of pure and collimated ultra-relativistic beams of electron (anti)-neutrinos (ν_e) from accelerated beta decaying radioactive ions. The production and extraction of 10^{13} ^{18}Ne/s are predicted to be possible on a circulating molten fluoride salt target with 7 mA, 160 MeV pulsed proton beam [2,3]. Finally, this type of targets is also proposed in an innovative method to produce isotopes relevant in nuclear medicine [7].

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ISBN: 978-3-95450-132-8 doi:10.18429/JACoW-IPAC2014-WEPRO080

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In this paper, the application of the molten target loop approach using different materials (summarized in Table 1) is described, aiming at the production of RIBs in different fields of application.

MOLTEN FLUORIDE SALTS FOR RADIOISOTOPES PRODUCTION

In the framework of the Beta Beams [6], a molten fluoride target loop has been proposed for the production of high rates of the neutrino (ν_e) emitter ^{18}Ne [2,3]. As an important step for the validation of the concept, a prototype static target unit has been designed and tested on-line at the CERN-ISOLDE facility with 1.4 GeV pulsed proton beam [8].

Molten salts have been used only once in the production of RIBs [9] due to the challenges inherent to corrosion and thermal instability of these compounds. In this work, the target material consisted of a fluoride eutectic salt binary system, NaF:LiF (39:61% mol) and several challenges related to the corrosive nature and hygroscopicity of the fluoride salts have been overcome by a proper engineering, dimensioning and construction of the unit [8]. A special corrosive resistant nickel-rich alloy, Haynes242, was procured for the target unit manufacturing, requiring special tooling due to the hardness of the material. The prototype validated the use of the chosen salt and allowed the assessment of Ne intensities used as input for the design of the diffusion chamber.

Primary focus has been given to the production and release of ^{18}Ne ($T_{1/2}$=1.67s), which have been assessed at the ISOLDE tape station, equipped with a 4π-beta detector, as a function of the target temperature. The obtained intensities for ^{18}Ne have been found to be increasing with the target temperature with values ranging between 3.7×10^4 (at 700ºC) and 5.0×10^4 ions/μC (at 740ºC) [8]. Furthermore, the efficiency of release (ε_{rel}) of Ne ions, expressed as the relative amount of ions that survive from the moment of their production in the target to the moment of their extraction from the ion source, was found to be in the range of 2.2-2.5% [8].

The release efficiency depends on the diffusion and desorption processes occurring in the target unit. Therefore, the analysis of the release characteristics allows the determination of the diffusion coefficient (D) [10], an essential parameter for production rate assessment in a diffusion chamber. A first estimation for Ne in NaF:LiF, a previously unknown parameter, has been obtained in this work. The diffusion coefficient is equal to 6.7×10^{-3} mm^2/s at 740ºC, in the same range as the coefficients of other noble gases (Xe, Kr) in fluoride salts [11].

Accounting for the measured diffusion coefficient, the production rates of ^{18}Ne in a diffusion chamber could be calculated. The diffusion chamber has the function of reducing the diffusion length by fragmenting the material into droplets, jets or films. This increases the release efficiency of exotic RIBs. Figure 2 shows a comparison between the release fractions for different shapes (droplets, jets, films) as a function of their sizes, taking the measured diffusion coefficient of 6.7×10^{-3} mm^2/s. The released fractions have been calculated using an analytical model for the diffusion from a material in the form of spheres, cylinders and films derived from Fick's law [12]. Figures as high as 96% are predicted when ^{18}Ne is diffusing out of a droplet of 100 μm radius, which translate into yields of the order of 1.9×10^7 ions/μC at 740ºC at ISOLDE. This figure shows a predicted improvement of ^{18}Ne yields up to 1×10^{13} ions/s for 160 MeV, 1 MW proton beam, as a possible version of the upgraded Linac4 at CERN [2,3].

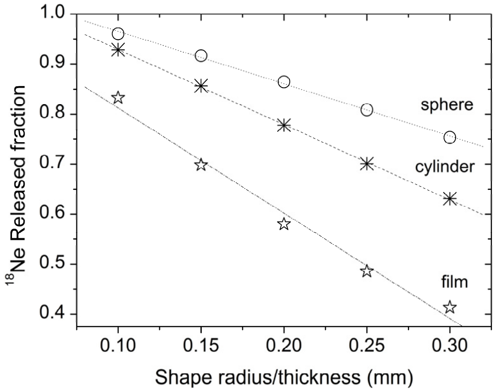

Figure 2: Calculated released fractions of ^{18}Ne diffusing out of droplets, jets or films as a function of dimension of the shape, for 130 ms residence time in the diffusion chamber. Lines are guides to the eye.

Opportunities for medical applications with molten fluoride salt targets

In addition to the measurement of ^{18}Ne beam intensities, carbon beams have been investigated [8]. Particular attention has been given to ^{11}C beams due to its relevance in medical imaging as a PET (positron emission tomography) imaging probe or in hadrontherapy replacing stable ions [7]. The evaluation of the yields as a function of temperature has been done in the atomic and molecular form since the extraction in the latter form is typically favoured. A record yield of 8×10^8 ^{11}CO/μC has been measured [8] showing an improvement by two orders of magnitude in comparison to the previously reported values. In fact, the combination of molten salt targets with the ISOL technique could be used as an alternative and innovative method to produce radiotracers for nuclear imaging and/or treatment [7]. To reach more interesting in-target production yields, the molten salt unit could be combined with a more appropriate proton driver (such as commercial cyclotrons delivering 30 MeV, 1.2 mA proton beams).

MOLTEN LEAD-BISMUTH EUTECTIC LOOP

Research and development work in lead bismuth eutectic (LBE) has been undertaken by several institutes due to the favourable properties for its use in accelerator driven systems (ADS), as coolant for nuclear reactors or as spallation material for neutron sources. In RIB production, LBE is a good candidate in the production of neutron-deficient mercury isotopes, which are of great interest in nuclear shape coexistence investigations. A new project, defined under the acronym LIEBE, has been initiated in 2012 and proposes the use of LBE in a circulating loop as target for production of exotic neutron-deficient mercury beams [14]. This proposal is inspired by the LBE target loop, proposed and tested offline during the EURISOL design study [1,5]. As an important step for the validation of the concept, the LBE loop compatible with the ISOLDE robot handling and front-end is now in the design phase.

The molten LBE will be irradiated in a dedicated volume, then will pass a metallic grid where the fragmentation of the material will occur, reducing the diffusion length and, thus, increase the released fraction of the produced radioisotopes. The LBE will be kept circulating with an electromagnetic pump. Furthermore, the loop will include a heat exchanger to cope with the temperature gradients from the diffusion chamber (heated up to 600ºC) and the pump. As a most important part of the design of such target unit, the dimensioning of the diffusion chamber shall be performed accounting for the predicted production rates and respective release efficiencies, which are strongly dependent on the diffusion and desorption processes. A first assessment of the diffusion released fractions as a function of the time in the diffusion chamber has been performed using analytical models. The calculated released fractions as a function of the time in the diffusion chamber, for different isotope half-lives, are shown in Fig. 3. As an example, for exotic neutron-deficient mercury species such as ^{177}Hg ($T_{1/2}$=130 ms) the predicted released fraction by diffusion out of a 100 µm radius droplet is about 40% for a residence time in the diffusion chamber of 200 ms. The assessment of the production rates accounting for the diffusion out of the material shall be complemented by calculations including desorption and transfer to the ion source, which are dependent on the material size and geometry as well as on the geometry of the diffusion chamber.

The first on-line tests are foreseen in 2015 where the LBE loop will be irradiated with a 1.4 GeV, 2 µA pulsed driver beam. An improvement by several orders of magnitude is expected for exotic mercury species such as $^{177\text{-}178}$Hg. First estimations indicate that the fragmentation to droplets of 100 µm radius will improve the release fraction of ^{177}Hg from 1.8% [4] up to 40%. Further improvements are predicted with an upgrade to 2 GeV, 6 µA proton driver, under consideration for HIE-ISOLDE.

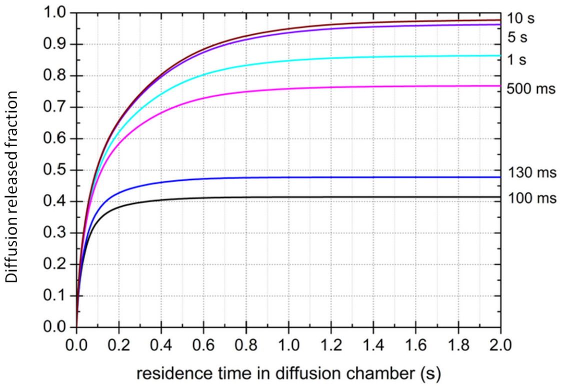

Figure 3: Calculated released fractions by diffusion out of a 100 µm droplet as a function of the residence time in the diffusion chamber, for different isotope half-lives.

CONCLUSION

Molten materials are extremely appealing due to the combination of high material densities and good heat transfer properties, which makes them attractive candidates for high power applications. In the particular case of RIB production, the combination of target loops with material fragmentation in a diffusion chamber leads to a great improvement of the beam intensities.

Following the results obtained during the tests of a molten salt static unit, the feasibility of using this material in the production of pure and intense RIBs with a broad range of applications was validated, from particle physics to nuclear medicine. The upgrade into a circulating loop will allow withstanding high beam powers (MW class) and consequently improve the RIBs production rates.

The feasibility of the target loop concept will be tested using a prototype molten LBE loop. The unit is currently in the design phase and on-line tests are foreseen to take place in 2015 at the CERN-ISOLDE facility.

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