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Production and release of ISOL beams from molten fluoride salt targets

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ABSTRACT

In the framework of the Beta Beams project, a molten fluoride target has been proposed for the production of the required 10^{13} ^{18}Ne/s. The production and extraction of such rates are predicted to be possible on a circulating molten salt with 160 MeV proton beams at close to 1 MW power. As a most important step to validate the concept, a prototype has been designed and investigated at CERN-ISOLDE using a static target unit. The target material consisted of a binary fluoride system, NaF:LiF (39:61 mol.%), with melting point at 649 °C. The production of Ne beams has been monitored as a function of the target temperature and proton beam intensity. The prototype development and the results of the first online tests with 1.4 GeV proton beam are presented in this paper.

1. Introduction

Pure and intense radioactive ion beams (RIBs) are of great demand in numerous fields such as nuclear physics and astrophysics, weak interactions, solid state physics and life sciences. These characteristics are particularly relevant for the Beta Beams project [1] where the required rates exceed by several orders of magnitude the intensities presently delivered to nuclear physics and astrophysics communities.

The isotopes of interest are produced in a nuclear reaction between an accelerated primary beam and a stationary target. One of the main production mechanisms is the isotope separation online (ISOL) [2] method where a thick target is bombarded with accelerated high energy primary particles leading to the production of radioactive ions. The products of this process are extracted, ionized, electromagnetically mass-separated and distributed to the experimental apparatuses. Typically, the ISOL technique allows obtaining pure and intense RIBs. Nevertheless, the intensities of short-lived species are strongly affected by decay losses due to time delay between in-target production and ion beam extraction. Accounting for the need of high intensities of short-lived isotopes, the target and ion source units must be developed in order to shorten diffusion lengths for fast diffusion release and fast transport to the ion source.

In particular, molten targets [3,4] can provide the highest intensities for isotopes of certain elements due to the high material density. However, these targets suffer from long diffusion times which limit their use on the production of short-lived species. Moreover, these targets are also limited in the deposited beam powers. In order to cope with these limitations, molten target loops with a diffusion chamber are required with the purpose of decreasing the diffusion times and to accommodate high beam powers.

Within the Beta Beams project, a molten salt target loop was proposed for the production of the high rates of ^{18}Ne ($T_{1/2}$ = 1.67 s) [5,6]. To validate the concept, a prototype static unit has been tested at ISOLDE with a 1.4 GeV proton beam. In this paper, the prototyping and the results of the online tests are presented.

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2. Static molten salt target design and commissioning

The choice of the target material has been performed by considering the ^{18}Ne production rates, thermal stability and low vapour pressures of the material at the operating temperatures. Target materials containing Na, F or Mg are the best candidates for the production of ^{18}Ne, which benefits from the (p,X) (or (^{3}He,X)) reactions [7] onto these elements. Thus, binary systems containing sodium fluoride (NaF) were considered as candidates for the present application.

A rate of 10^{13} ^{18}Ne/s, extracted from the target, could be obtained using 160 MeV proton beam impinging on two binary fluoride systems: NaF:ZrF_4 (60:40 mol.%) and NaF:LiF (39:61 mol.%). The production yield of ^{18}Ne has been estimated using the following equation:

$$I = \frac{N_A \Phi H}{A} \int \sigma(E) \left(\frac{\partial E}{\partial(\rho x)} \right)^{-1} dE \quad (1)$$

where N_A is the Avogadro number, A the mass number of the target element, H the target nucleus abundance, $\sigma(E)$ the cross-section at energy E, Φ the projectile current and $\partial E/\partial(\rho x)$ the stopping power. The stopping powers have been calculated using Stopping and Range of Ions in Matter (SRIM) software [8] while the cross-sections have been obtained using the TALYS code [9]. The required 10^{13} ^{18}Ne/s for the Beta Beams project can been obtained at 160 MeV, 6 mA incoming proton beam for NaF:ZrF_4 and 7 mA for NaF:LiF (Table 1).

The binary system NaF:ZrF_4 has been first investigated as material for the molten salt loop target [5,6] due to its lower melting point. However, its high vapour pressure (5 mmHg at 900 °C) [10] combined with the high reactivity of ZrF_4 hampered its safe use in the present application. In contrast, the binary system composed by the mixture of NaF and LiF has shown thermal stability and has been chosen as target material for this prototype. This binary system presents an eutectic point at 649 °C and vapour pressure of the order of 0.1 mmHg at 900 °C [10].

For the synthesis of the NaF:LiF system, stoichiometric quantities of high purity NaF and LiF (Sigma Aldrich, 99.9% and 99% pure, respectively) have been mixed and heated up to about 50–100 °C above the melting point of the system. Due to the hygroscopicity of the fluoride salts, all handling has been carried out in dry glove boxes (purity levels: H_2O < 1 ppm, O_2 < 1 ppm), under argon atmosphere, to prevent oxide and moisture contamination.

To test the proposed concept, a prototype inspired from the standard ISOLDE static target units has been constructed. A schematic representation of the target unit is depicted in Fig. 1. The different parts of the unit are shown in the pictures of Fig. 2.

The NaF:LiF salt has been inserted in a 21.6 cm long and 2 cm diameter hexacylindrical container as shown in Fig. 2(a). Accounting for the corrosive behaviour of fluoride salts at high temperatures, the container was built in a nickel-rich alloy (Haynes 242 [11,12]). In addition, the dimensioning of the salt target container has been done accounting for the material's mechanical and heat transfer properties [13] and difficulties in the machining of the alloy. The container was filled with the NaF:LiF melt up to 3/4 of its volume allowing a free surface for the isotopes to diffuse out of the target. A temperature controlled condensation helix, shown in Fig. 2(b), was inserted in a 7.5 cm vertical chimney on top of the horizontal container. The previous elements were connected to a versatile arc discharge ion source (VADIS) [14] via a temperature controlled transfer line (shown in Figs. 2(c) and (d), respectively), suited for the production of noble gases, in the so-called VD7 configuration. The transfer line, named here as cold line, aimed at the condensation of less volatile elements that could be released as contaminants for the beams of interest. For a systematic study of the effect of temperature on the production and release of the beams of interest, the unit has been equipped with three type K thermocouples: one located at the centre of the container, a second one at the top of the condensation chimney and a third one at the bottom of the transfer line. The final arrangement of the static molten salt target, equipped with the thermocouples, is shown in Fig. 3.

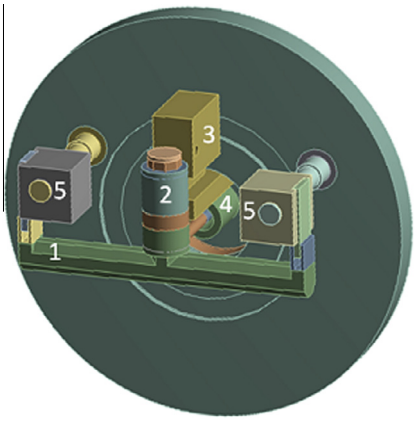

Fig. 1. Prototype molten salt target unit and its different components: (1) salt container, (2) condensation chimney, (3) cold transfer line, (4) ion source and (5) copper blocks for the heating of the container.

The VADIS ion source has been setup with an injection gas composed by a mixture of He, Ne, Ar, Kr and Xe (20% each by vol.) via a calibrated leak. The efficiencies required to compute the ion production from the measured beam intensities have been obtained with these stable tracers by measuring the ion current in a faraday cup.

3. Online measurements at ISOLDE

The release and production yields of Ne and other low-Z elements have been assessed at CERN-ISOLDE with 1.4 GeV proton beam from the PSB accelerator. The intensity of the proton pulses has been varied from 1×10^{12} to 6×10^{12} protons. Alike the standard procedure for molten metal targets at ISOLDE, the staggered extraction mode ('STAGISO' [15]) has been used with extraction intervals of 16 and 20 μs, reducing the energy density of the proton beam and, thus, decreasing shock waves and target ageing. In addition, the proton gaussian beam size has been set to 4.2 × 4.4 mm FWHM.

The radioisotopes of interest have been assessed at the ISOLDE tape station. The radioactive beam was collected in the tape for 100 ms and after the tape was moved from the collection point

Table 1
Summary of relevant properties of NaF:ZrF_4 (NZF) and NaF:LiF (NLF) binary systems and respective ^{18}Ne production yield. The production yields have been calculated for 160 MeV, 6 mA and 7 mA incoming for NaF:ZrF_4 and NaF:LiF, respectively.

Salt	Composition (% mol.)	Melting point (°C)	Density (g/cm^3)	Vapour pressure at 900°C (mmHg)	Ion production (ions/s)
NZF	60:40	500	3.14	5	1×10^{13}
NLF	39:61	649	2.75	0.1	1×10^{13}

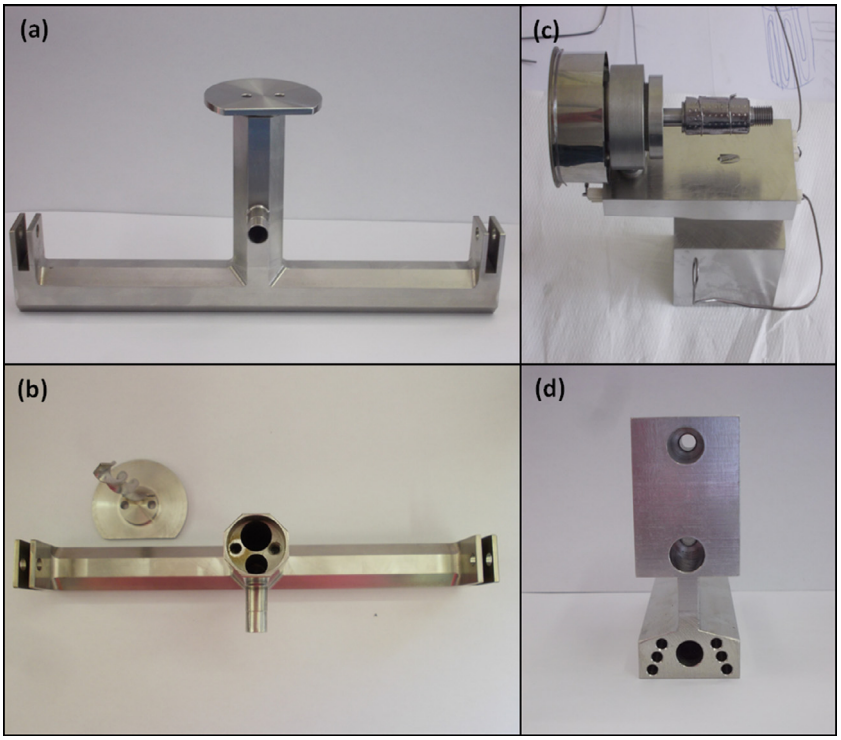

Fig. 2. Pictures of the molten salt target unit components: (a) salt container and chimney, (b) top view of the static container, chimney and condensation helix, (c) VADIS ion source connected to the transfer line, and (d) cold transfer line.

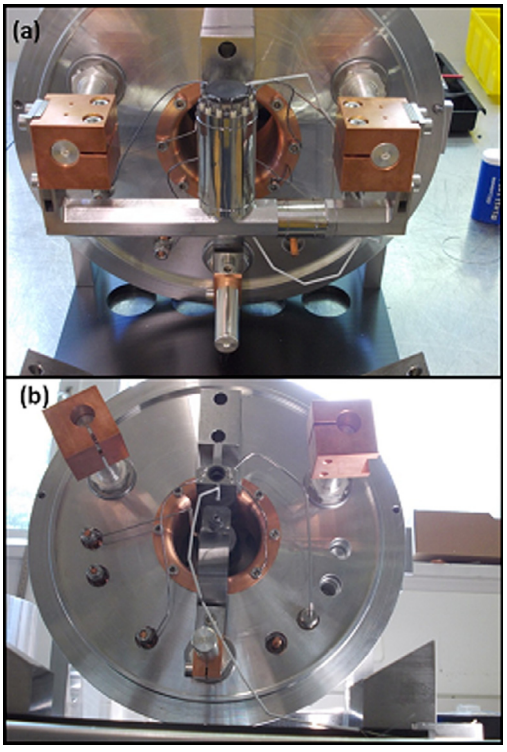

Fig. 3. Picture of the static molten salt target unit: (a) detail of the location of the thermocouples at the centre of the container and top of the chimney, and (b) thermocouple at the bottom of the cold transfer line.

towards the detectors (with transport time of 900 ms), where the count rate has been measured for 1 s. Following this procedure, the activity was sampled at regular intervals after switching off the proton beam allowing the measurement of the release curves.

The extraction efficiency was obtained from integration of the release curves, which can be approximated by a four parameter function [3]:

$$P(t) = \frac{1}{N}\left(1 - e^{-\frac{\ln 2 \cdot t}{t_r}}\right)\left(\alpha e^{-\frac{\ln 2 \cdot t}{t_f}} + (1-\alpha)e^{-\frac{\ln 2 \cdot t}{t_s}}\right) \quad (2)$$

where $P(t)$ is the probability of release of a particle, t_r, t_f and t_s are, respectively, the exponential rise, fast fall and slow fall times and α is a weighting factor between the slow and fast components. The factor $\frac{1}{N}$ is used for normalizing $P(t)$ to unity for $t \to \infty$. To obtain the yield of a certain isotope, the release curve is integrated, being the normalized yield expressed in units of ions/μC.

The release function $P(t)$ is the probability density for an atom generated at $t = 0$ of a given element to be released at a time t. A pulse of protons on the target gives a current of radioactive ions which initially rises from zero to a peak and then falls. The release curve can contribute to the understanding of the fundamental processes occurring in the target such as diffusion and effusion [16].

The RIBs of interest have been investigated as a function of the target temperature and proton beam intensities. The target unit has been kept above its melting point during the experimental run.

4. Production and release of ^{18}Ne

Fig. 4 shows representative data of the dependence of ^{18}Ne ($T_{1/2}$ = 1.67 s) production yields on the target temperature. A continuous increase of the ^{18}Ne yield with increasing target temperature with values ranging between 1.4×10^4 (at 700 °C) and 4.2×10^4 ions/μC (at 745 °C) of incident protons is observed. A larger effect of the proton beam intensity can be observed with an increase of 62% of the yield with a factor of six increase on the proton beam intensity. When reducing the proton bunch separation to 16 μs, a further increase on the yield has been observed with a maximum obtained value of 5.7×10^4 ions/μC at 770 °C.

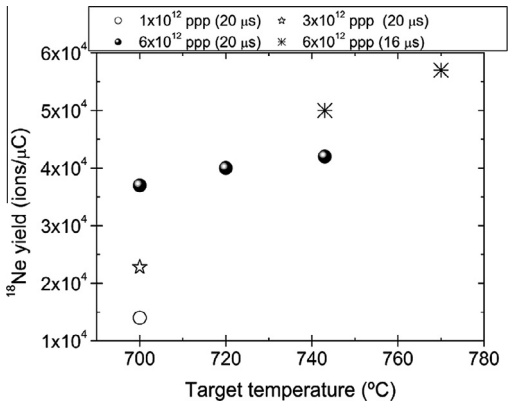

Fig. 4. ^{18}Ne experimentally measured yields as a function of the target temperature and proton beam intensity.

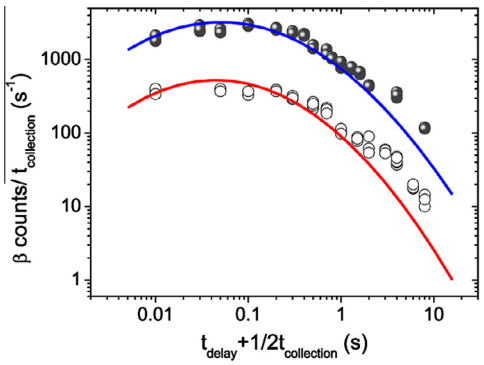

Fig. 5. ^{18}Ne release curves and respective fits measured at 700 °C (open dots) and 720 °C (closed dots).

The release of Ne has been investigated by measuring release curves at different target temperatures. Table 2 summarizes the release time constants obtained by fitting the experimental data with the function presented in Eq. (2). Representative release curves and respective fits to the experimental data are shown in Fig. 5.

Looking at the release time parameters, one can see that the values do not present a clear dependence on the target temperature. This result shows that these parameters may not be easily related to effusion and diffusion phenomena, which are known to have a strong dependence on the temperature. The rise times are lower than 100 ms showing a faster release than in CaO targets tested at ISOLDE with rising times of the order of 180 ms [17]. Still, the measured yields are below the best figures recorded at ISOLDE with 6.9×10^5 ions/μC using a CaO oxide target [18].

To characterize the release characteristics, the release efficiency, ε_{rel}, of ^{18}Ne has been extracted from the experimental data. The release efficiency is dependent on the material, operating temperature and unit geometry. This parameter can be simply obtained from the ratio between the experimentally observed yield, Y_{obs}, and the in target production yield, Y_{prod}, following the equation:

$$\varepsilon_{rel} = \frac{Y_{obs}}{Y_{prod} \cdot \varepsilon_{is}} \tag{3}$$

where ε_{is} is the ion source efficiency. Y_{prod} was obtained through simulations using ABRABLA code [19] with 32.7 g/cm^2 target thickness. Table 3 summarizes the release efficiencies obtained for ^{18}Ne as a function of the target temperature and proton beam intensity.

Table 4
Measured yields of carbon isotopes in molecular form as a function of the target temperature.

Isotope	$t_{1/2}$ (s)	Target temperature (°C)	Y_{prod} (ions/μC)
^{10}C + ^{16}O	19.26	720	3×10^5
^{11}C + ^{16}O	1223	700	6.7×10^8
^{11}C + ^{16}O	1223	720	7.7×10^8
^{11}C + ^{16}O	1223	740	6.0×10^8
^{11}C + ^{16}O + ^{16}O	1223	740	2.0×10^7
^{15}C + ^{16}O	2.449	720	1.2×10^4

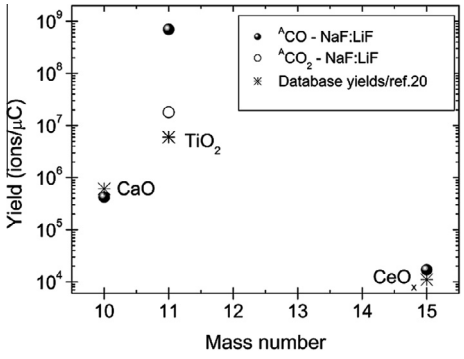

Fig. 6. Comparison between the extracted carbon isotopes from the molten salt target and the ISOLDE database yields.

One can observe that the release efficiency of ^{18}Ne increases slowly with the temperature and more data points at higher temperatures

Table 2
Release time constants obtained by fitting the data with Eq. (2) for ^{18}Ne.

Temperature (°C)	Proton intensity (ppp)	^{18}Ne yield (ions/μC)	t_r (s)	t_f (s)	t_s (s)
700	1×10^{12}	1.4×10^4	0.183	0.185	4.11
700	3×10^{12}	2.3×10^4	0.051	0.293	3.64
700	6×10^{12}	3.7×10^4	0.060	0.425	4.29
720	6×10^{12}	4.0×10^4	0.092	0.169	4.85

Table 3
Calculated release efficiency for ^{18}Ne. Y_{prod} was calculated using ABRABLA for a target thickness of 32.7 g/cm^2.

Temperature (°C)	Proton intensity (ppp)	ε_{is} (%)	Y_{prod} (ions/μC)	Y_{obs} (ions/μC)	ε_{rel} (%)
700	1×10^{12}	4	4.2×10^7	1.4×10^4	0.8
700	3×10^{12}	4		2.3×10^4	1.4
700	6×10^{12}	4		3.7×10^4	2.2
720	6×10^{12}	4.4		4.0×10^4	2.2
740	6×10^{12}	4.7		5.0×10^4	2.5

would be needed to well characterize this trend. Nevertheless, an increase of the release efficiency with temperature is expected due to an increase of the rates of effusion and diffusion.

The production of ^{18}Ne from the NaF:LiF molten target allowed the validation of this type of materials using the ISOL method. The prototyped unit showed a stable operation throughout the run without drops in the measured yields over time. A maximum yield of 5.7×10^4 ^{18}Ne/μC has been obtained for a target operating temperature of 770 °C. In order to improve the release properties, a molten target loop with a diffusion chamber is envisaged. One shall note that the present data has been obtained with 1.4 GeV, 0.1 μA proton beam whereas the required 10^{13} ^{18}Ne/s, for the Beta Beams project, are expected with 160 MeV, 7 mA incoming proton beam.

5. Production of carbon isotopes

Following the calculated in-target yields in this target material, carbon isotopes are expected to be produced in measurable quantities. The yields of carbon beams have been evaluated by taking release curves for ^{10}C, ^{11}C and ^{15}C in the molecular form. A summary of the obtained carbon yields as a function of the target temperature is presented in Table 4.

The temperature dependence of the carbon yields has been assessed only for ^{11}C and the results do not allow to infer a clear trend of an increase of the yield with temperature. More data would be needed to confirm the evolution of the yields at higher temperatures.

Fig. 6 shows a comparison between the carbon yields from the molten salt unit and the best values obtained at ISOLDE. One can see a clear improvement on the ^{11}C yield, being 110 times higher than the present records at ISOLDE. This result is consistent with the fact that the high density of the molten targets allows to obtain higher intensities, specially in longer lived species alike the case of ^{11}C. Also the production cross-sections for carbon isotopes in the sodium fluoride targets are higher than in TiO_2, CeO_x or CaO targets [18,20]. Nevertheless, the presence of oxygen in the unit was not expected and its origin might arise from structural materials or the presence of traces of water in the system.

The increase on the yields is observed in both monoxide and dioxide forms, being the maximum values held by the extraction of this isotope in monoxide form. Also in the case of ^{15}C, it is observed a small improvement of the yield in a factor of 1.5 times. Taking into account that the release times in molten targets are typically higher than the solid materials, one can expect an improvement of the carbon beams by using a molten salt target loop with a diffusion chamber. A decrease in the diffusion times coupled to high production rates may open the possibility of measuring more exotic carbon species.

6. Summary

A molten salt target unit has been proposed for the production of the required rates of ^{18}Ne for the Beta Beams project. A prototype of a static molten salt bath has been developed and tested at ISOLDE with 1.4 GeV protons. These initial results with a static molten salt target are promising and form a basis for future improvement of this method. Moreover, record yields have been measured for carbon beams opening the possibility of the use of molten fluoride salts as target material for the ISOLDE physics program.

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