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Off-line production of intense $^{7,10}Be^{+}$ beams

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Abstract

^{7}Be and ^{10}Be were produced by 590 MeV proton bombardment of a graphite target at PSI. Parts of this graphite target were transferred into an ISOLDE target and ion source unit and ionized with the ISOLDE resonance ionization laser ion source. Thus intense radioactive ion beams of 300 nA of $^{7,10}Be^{+}$ were produced off-line.
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1. Introduction

Recently pure ion beams of ^{7}Be became popular for astrophysical applications. They allow e.g. to measure precisely the proton capture cross-section $^{7}Be(p,\gamma)^{8}B$ which is important to determine the expected neutrino flux from the sun [1]. The ^{7}Be beams can be used either for direct measurements in inverse kinematics [2] or by preparing an ion-implanted ^{7}Be target for a subsequent measurement in normal kinematics [3].

Being the lightest gamma-emitter with suitable half-life (53.3 days), ^{7}Be is also the favorite tracer nuclide for high-sensitivity wear analysis in materials sciences and medical applications [4,5].

At Paul Scherrer Institute graphite targets are routinely bombarded with 590 MeV protons at an average current of up to 2 mA for the production of pion and muon beams. Occasionally these targets have to be exchanged and are disposed of as radioactive waste. Since the dominant spallation and fragmentation produced activity remaining in the graphite is ^{7}Be, they are on the other hand a valuable source for an off-line use of this radioisotope. Here we report on the first off-line experiment with irradiated graphite from PSI which was used to produce intense ion beams of $^{7,10}Be$ with the ISOLDE resonance ionization laser ion source (RILIS).

2. Target activation

The PSI "E type" target wheel (4 cm long in beam direction) is made of fine grain high purity graphite (grade EK94/R6400 from SGL carbon) [6]. The latter has a density of 1.78 g/cm^3, an

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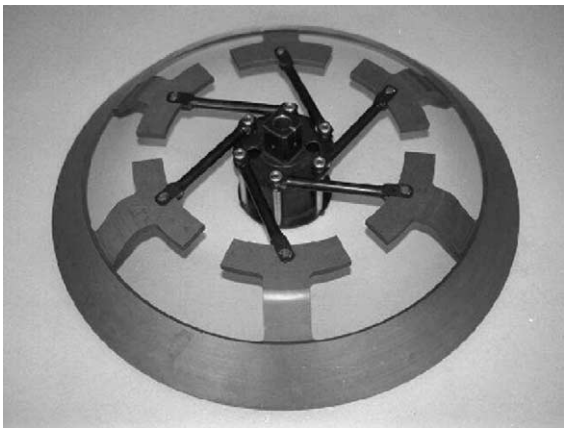


Fig. 1. PSI 'E type' target wheel.

average grain size of 10 μm, an open porosity of 14% with a pore size of 2.0 μm and an ash content of impurities of 20 ppm [7]. The 45 cm diameter conic wheel, attached via six spokes to a stainless steel hub, is rotating during irradiation for regular distribution of the beam power. Fig. 1 shows the target wheel.

The target was bombarded with 590 MeV protons from the PSI separated sector cyclotron with an integrated dose of 1.07 Ah over one month. During irradiation with up to 2 mA the target is heated by the proton beam up to 1700 K. After removal from the irradiation position it was stored for two months. The activated graphite parts were separated from the spokes and broken into pieces ranging from some cubic millimeters to some cubic centimeters. 25.4 g of the activated graphite with a ^{7}Be activity of about 84 GBq (5.6×10^{17} atoms) were then filled into a standard ISOLDE tantalum target container (20 mm inner diameter and 200 mm long) connected to a tungsten surface ionizer (3 mm inner diameter and 30 mm long). These operations were performed in the PSI shielded hot cell ("ATEC") using remote controlled manipulators. The target was then packed into a lead shielded radioactive transport container and brought to CERN.

3. Ion beam preparation

The target and ion source unit was put onto the front-end of the ISOLDE high resolution separator (HRS) and slowly heated in order to outgas and to release the ^{7}Li accumulated from ^{7}Be decay. While lithium (ionization potential 5.39 eV) is rather well thermally ionized in the tungsten cavity heated to 2000–2100 °C, beryllium (ionization potential 9.32 eV) was ionized with the ISOLDE RILIS [8]. The earlier developed [9] excitation scheme goes with two ultraviolet transitions (234.9 and 297.3 nm) from the atomic ground state to an autoionizing state. When heating the target container above 1400 °C, the beryllium beams ^{7,9,10}Be^{+} were observed. The extracted Be^{+} current could be easily adjusted by varying the temperature of the target container. At ≈1700 °C a ^{7}Be^{+} beam current of 300 nA was obtained and could be maintained for several hours.

From the ratio of the extracted amount of ^{7}Be to the original activity in the target we deduced a lower limit [1] of 8% for the on-line RILIS efficiency of ^{7}Be. Also beams of ^{9,10}Be^{+} with similar intensity could be produced. On 6.12.2001 the isotopic ratio of the beryllium beams was determined to

$$^{7}\mathrm{Be} : {}^{9}\mathrm{Be} : {}^{10}\mathrm{Be} = 56 : 100 : 54$$

The first-step laser was retuned in each case to compensate for the isotope shift. Within the experimental errors this ratio corresponds roughly to the ratio of the known production cross-sections for carbon bombarbed by 600 MeV protons [10]: ^{7}Be (mind the decay losses of ^{7}Be between irradiation and measurement): 11.0(11) mb, ^{9}Be: 5.3(7) mb and ^{10}Be: 2.8(4) mb.

It should be noted that the RILIS efficiency varies slightly with the mass of the atoms. Light atoms diffuse faster through the ionizer cavity and have therefore a slightly lower chance to interact with the laser light before escaping (see the effect of the mean thermal velocity $\bar{v}$ in Eq. (4) of [8]). Table 1 shows the calculated increase in RILIS efficiency for heavier beryllium isotopes relative to the efficiency for ^{7}Be. When taking into account the

[1] For a determination of the absolute efficiency it would be necessary to integrate the current until no more Be is left in the container. However, the experiment was stopped before in order to keep some ^{7,10}Be in the container for future experiments. Thus only a lower limit was obtained.

Table 1
Calculated RILIS efficiency relative to that of ^{7}Be

Isotope	^{7}Be	^{9}Be	^{10}Be	^{11}Be	^{12}Be	^{14}Be
Relative efficiency	1.00	1.10(2)	1.15(3)	1.19(4)	1.23(5)	1.31(7)

slightly higher RILIS efficiency for the heavier isotopes, the observed ratio fits perfectly with the expected one.

The surface ionized isobaric beam contamination $^{7}Li^{+}$ was of the order of 10 nA. The production cross-section of ^{6}Li from 590 MeV proton bombardment of carbon is 7.4(15) mb [10] compared to 5.6(11) mb for ^{7}Li, but the current on mass 6 was measured to be 8000 times lower. This shows that the directly produced ^{6,7}Li was already released during the irradiation at PSI where the target is heated by the proton beam up to 1400 °C. Thus the observed ^{7}Li stems entirely from the decay of the ^{7}Be still left in the target and, *under the assumption* that the observed ^{7}Li is in equilibrium with the ^{7}Be decay, an excellent thermal ionization efficiency for lithium of about 80% could be deduced.

For applications where particularly pure ^{7}Be beams are required, a fast beam chopper can be used to open the beamgate only during the laser ionized ion pulses and thus suppress the dc surface ionized background. With the standard tungsten ionizer cavity (1 mm wall thickness) used here the selectivity ^{7}Be/^{7}Li was thus increased by one order of magnitude, however at the cost of a reduced ^{7}Be current.

4. Beam applications

The ^{7}Be beam was used for different applications:

- A metallic ^{7}Be target was produced by 60 keV ion implantation into a copper matrix. Since only a makeshift [2] collection point was available at the HRS, only 50% of the beam could be focused through the 2 mm diameter diaphragm. Still 1.2×10^{16} ^{7}Be atoms were collected in the sample within some hours of beam time. This ^{7}Be target was used for a new measurement of the $^{7}\mathrm{Be}(p,\gamma)^{8}\mathrm{B}$ cross-section at Weizmann Institute (Rehovot) [11].
- Several samples of GaN semiconductors were implanted with ^{7}Be. The combination of classical semiconductor spectroscopy techniques (e.g. deep level transient spectroscopy) with a radiotracer doping (here ^{7}Be) allows one to uniquely assign band gap states to being related to the chemical element (here beryllium) when correlating the decrease (or increase respectively) of the observed signals with the decay of the radiotracer [12].
- ^{7}Be was also implanted at different energies (60–260 keV) into various samples (metal foils, graphite) to study off-line the beryllium release. Part of the results are reported in [13].

5. Summary

It has been shown that even radioactive waste can find a very useful application. This off-line method allows one to produce much more intense beams of long-lived radio-isotopes than could be produced on-line at ISOLDE since the primary proton beam at PSI is a factor thousand more intense than the average beam current presently available at ISOLDE.

It should be noted that for applications where the ^{7}Be or ^{10}Be is not necessarily required as an ion beam but only a radioactive sample is to be produced, one can replace the rather involved step of ionization and mass separation by a chemical separation and deposition [14,15]. The latter methods provide a higher total efficiency (well above 50%), but obviously do not give an isotopic separation. Hence, for an isotopically pure target produced by chemical means a selective production reaction

[2] Normally high activity collections are performed at the general purpose separator GPS. The latter not being available for this experiment, the collection chamber had to be mounted closely behind the focal plane of the second HRS magnet. The beamline elements upstream of this position did not provide enough degrees of freedom to obtain a better focusing.

like ^{7}Li(p,n)^{7}Be is required and "waste graphite" cannot be used.

6. Outlook

In this experiment we limited the ^{7}Be current deliberately at about 300 nA to avoid any possible efficiency reduction due to space charge effects in the ionizer cavity. [3] However, this was a very conservative approach and RILIS ion beams of 1 μA are certainly feasible with the present geometry.

A permanently irradiated PSI 'E type' target contains in saturation about 40 TBq of ^{7}Be, i.e. the macroscopic amount of about 3 mg of ^{7}Be! The entire activated part (about 340 cm^{3} of graphite) could be crushed and transferred into a special "large volume" tantalum container. Starting the off-line experiment soon (e.g. two weeks) after end of irradiation, a ^{7}Be^{+} beam of e.g. 1 μA could be produced and continuously provided for several weeks. However, the transport and remote handling of such a target (unshielded 0.3 Sv/h at 1 m) would become rather involved.

An "old" PSI target (6 cm long in beam direction [6]) irradiated with 10 Ah of protons contains about 5 mg of ^{10}Be. This could be used for many weeks of continuous 1 μA ^{10}Be^{+} beam without any particular shielding requirements. [4]

Alternative sources of ^{10}Be could be the primary cooling water of spallation neutron facilities which is directly exposed to the proton beam or the ternary fission products in reactor waste. Provided there is no contamination with stable beryllium, the isotopic ratio in the latter is more favorable (^{10}Be/^{9}Be ≈ 6–7 [16,17]), but the chemical separation of the beryllium (about 10 mg/ton of fuel with a burnup of 40 GWd/t) from all other fission products is much more involved. A still better isotopic purity of ^{10}Be is obtained by a dedicated production via ^{13}C(n_{fast}, α)^{10}Be [14].

In a future high intensity ISOL facility (EURISOL) it could be foreseen to use the ^{7}Be which is continuously produced in a graphite beamstop for a batch-mode production of ^{7}Be ion beams with a dedicated off-line separator.

[3] Note that the laser beam is pulsed with a repetition rate of 11 kHz, thus the peak ion density in the cavity reaches values of the order of 10^{10} cm^{-3} shortly after the laser pulse!

[4] Due to its long half-life of 1.5×10^{6} years and the low beta decay energy the dose rate is negligible even for larger amounts of ^{10}Be.

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References

[1] E. Adelberger et al., Rev. Mod. Phys. 70 (1998) 1265.
[2] L. Campajola et al., Z. Phys. A 356 (1996) 107.
[3] M. Hass et al., Phys. Lett. B 462 (1999) 237.
[4] M. Hoffmann et al., Nucl. Instr. and Meth. B 183 (2001) 419.
[5] P. Fehsenfeld, C. Eifrig, R. Kubat, Nucl. Phys. A 701 (2002) 235.
[6] G. Heidenreich, Scientific and Technical Report 2000, Vol. VI, Large Research Facilities, Paul Scherrer Institut, Villigen, Switzerland, 2001, p. 26.
[7] http://www.sgl-carbon.com.
[8] V. Fedoseyev et al., Hyp. Int. 127 (2000) 409.
[9] J. Lettry et al., Rev. Sci. Instr. 69 (1998) 761.
[10] P. Fontes, C. Perron, J. Lestringuez, F. Yiou, R. Bernas, Nucl. Phys. A 165 (1971) 405.
[11] L.T. Baby et al., Phys. Rev. Lett. 90 (2003) 22501.
[12] F. Albrecht, J. Grillenberger, G. Pasold, W. Witthuhn, N. Achtziger, Appl. Phys. Lett. 79 (2001) 961.
[13] K. Peräjärvi et al., Nucl. Instr. and Meth. B, these Proceedings. doi:10.1016/S0168-583X(02)01924-9.
[14] D.R. Goosman, Nucl. Instr. and Meth. 116 (1974) 445.
[15] A. Zyuzin et al., Nucl. Instr. and Meth. B 187 (2002) 264.
[16] A. Vorobyov et al., Phys. Lett. 40B (1972) 102.
[17] W. Baum, Ph.D. thesis, TH Darmstadt, 1992.