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Radiochemical separation of $^7$Be from the cooling water of the neutron spallation source SINQ at PSI

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Summary. $^7$Be is a key radionuclide for investigation of several astrophysical processes and phenomena. In addition, it is used as a tracer in wear measurements. It is produced in considerable amounts in the cooling water (D2O) of the Spallation Induced Neutron Source (SINQ) facility at PSI by spallation reactions on $^{16}$O with the generated fast neutrons. A shielded ion-exchange filter containing 100 mL of the mixed-bed ion exchanger LEWATIT was installed as a bypass for the cooling water into the cooling loop of SINQ for three months. The collected activity of $^7$Be was in the range of several hundred GBq. Further, the $^7$Be was separated and purified in a hot-cell remotely-controlled using a separation system installed. With the exception of $^{10}$Be, radioactive by-products can be neglected, so that this cooling water could serve as an ideal source for highly active $^7$Be-samples. The facility is capable of producing $^7$Be with activities up to 1 TBq per year. The $^7$Be sample preparation is described in detail and the possible uses are discussed. In particular some preliminary results of $^7$Be ion beam production are presented.

1. Introduction

The radionuclide Beryllium-7 has gained over the last decades an increasing interest in both basic and applied research. Its relatively long half life (53.29 d) allows it to be used in the production of ion beams utilizing standard ion sources and accelerators, as well as in the preparation of targets for different experiments.

$^7$Be is produced by proton irradiation of $^7$Li through the nuclear reaction $^7$Li$(p,n)^7$Be. The cross section for this reaction is reported by several authors to be around 0.5 barn in the maximum of the excitation function at 2.25 MeV [1, 2]. However, several weeks of beam time are necessary to obtain the required high amounts of up to hundred GBq. An additional disadvantage of this production route is the macroamount of stable $^7$Li, which has to be chemically separated with a very high efficiency because the lithium can eventually disturb the foreseen experiments.

An alternative production mode is spallation on light target nuclei with highly energetic particles. The neutron spallation source SINQ at PSI represents one of the most powerful facilities of its kind world-wide. With 590 MeV protons, highly energetic neutrons are produced, which induce a considerable amount of $^7$Be in the D2O of its cooling loops via the $^{16}$O($n,x)^7$Be and $^{16}$O($p,x)^7$Be processes, approximating 10 mbarn [3]. With 4000 L of cooling water in total, TBq amounts of $^7$Be are produced within one operation cycle (typically lasting for 8–9 months). Partial filtration of the cooling water represents a cheap – because no dedicated beamtime is required – and very effective method for gaining $^7$Be activities of several hundred GBq with a very high purity. However, it has to be mentioned that both stable $^7$Be and radioactive $^{10}$Be (half-life 1.386 × 10$^6$ yr [4]) are produced during the nuclear reaction in comparable number of nuclides like the $^7$Be.

2. Need of $^7$Be

Recently several experiments have been proposed, where the $^7$Be would be utilized. Although a thorough discussion is beyond the scope of the present paper, a short review is presented here. A special interest appears to be in those applications which involve the use of a radioactive ion beam. A test production using material obtained from the neutron spallation source SINQ is presented below.

2.1 Measurement of the $^7$Be$(p,\gamma)^8$B cross section

Precise measurements of solar neutrino fluxes and the discovery of the neutrino oscillation call for a very precise determination of all relevant nuclear cross sections in order to confirm solar models. In particular, $^7$Be$(p,\gamma)^8$B is a key reaction for the high energy component of the solar neutrinos. The results of the experiments done over the last decades show discrepancies in both the energy dependence and the absolute value of the cross section. Unfortunately this holds also for the more recent measurements. A review

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of this issue can be found in [5]. It is worth noting that all
direct experiments producing useful results used the same
approach, i.e. a proton beam impinging onto a \(^7\text{Be}\) radioac-
tive target. Previous attempts to measure this reaction cross
section using a \(^7\text{Be}\) ion beam [6, 7] could not produce results
of useful precision due to the very low ion beam intensity.
The ERNA (European Recoil separator for Nuclear Astro-
physics) collaboration has started an experiment, whose aim
is to determine the cross section of the \(^7\text{Be}(p, \gamma)\(^8\text{B}\) reaction
exploiting the \(^7\text{Be}\) radioactive ion beam that can be produced
at the CIRCE accelerator laboratory [8]. The objective is to
determine the total reaction cross section in the energy range
\(E_{\text{cm}} = 0.4–1.0\ \text{MeV}\). The required \(^7\text{Be}\) is presently produced
at ATOMKI via the \(^7\text{Li}(p, n)^7\text{Be}\) reaction following the pro-
cedure described in [9]. The SINQ spallation source could
represent an additional, alternative source of \(^7\text{Be}\), consider-
ing the high amount of \(^7\text{Be}\) needed and the relatively long
time required for its production and radiochemical separa-
tion by means of proton activation of \(^7\text{Li}\) targets.

2.2 The problem of primordial \(^7\text{Li}\) and the \(^7\text{Be}(n, \alpha)\alpha\)
and \(^7\text{Be}(n, p)^7\text{Li}\) reactions
Another important unresolved problem of astrophysics is
the so-called “Cosmological Lithium problem”. It refers to
the large discrepancy between the abundance of primordial
\(^7\text{Li}\), predicted by the standard theory of Big Bang Nucle-
osynthesis (BBN) [10], and the value inferred from obser-
vations. New and accurate measurements are necessary in
order to investigate the destruction of \(^7\text{Be}\) during BBN via the
\(^7\text{Be}(n, \alpha)\alpha\) and \(^7\text{Be}(n, p)^7\text{Li}\) reactions. Target material
of several hundred GBq will be necessary to perform reliable
studies on this subject. This amount of activity can only be
produced by the PSI method.

2.3 \(^7\text{Be}\) for wear analysis
Radioactive tracer techniques are nowadays routinely used
for non-contacting, on line wear and corrosion measure-
ments and represent an extremely powerful tool in mate-
rial science and engineering. Presently, two radioactive
tracer techniques are typically used: bulk activation (BA)
and surface-layer activation (SLA).

In both cases, radio-isotopes are produced directly in
the sample by irradiating it with neutrons, for BA, or light
charged particles (protons, deuterons and alphas), for SLA.
A common problem of these techniques is that their appli-
cation depends on the material to be tested and it is limited
to those materials that exhibit a high activation cross sec-
tion and, in particular in the case of SLA, strong resistance
to radiation damage.

Radioactive ion implantation (RII) was suggested as
a possible solution to such problems ([11, 12] and refer-
ences therein). The implantation depth is determined by the
beam energy and the stopping power of the radioactive ion
in the sample: therefore a proper modulation of the ion beam
during the implantation in a given material allows
a quite wide range of possible radio-isotope depth distribu-
tions. The drastic reduction of the radiation damage (up to
a factor \(10^6\)) in comparison with SLA allows application of
RII in principle to any material, therefore providing a pow-
ful tool for comparative studies. The main problem of this
technique is the availability of a suitable RIB. A beam of \(^7\text{Be}\)
ions could be ideal for such applications.

3. Experimental
3.1 Radiochemical analysis and selection of the most
suitable cooling cycle
A schematic view of the SINQ facility and the cooling sys-
tems can be seen in Fig. 1. The source is cooled by three
separate cooling loops, consisting of a target window cool-
ing (1), target cooling (2), and the moderator tank (3) for
slowing down the neutrons to thermal energies.

Determination of the radionuclide inventory was per-
fomed using \(\gamma\)-ray spectrometry and liquid scintillation

![Fig. 1. Vertical cut of the SINQ target station and its cooling cir-
cuits (figure taken from [13]).](image)
Radiochemical separation of $^7$Be from the cooling water of the neutron spallation source SINQ at PSI

Table 1. Typical content of radionuclides in the three cooling circuits of the SINQ.

<table>
<thead>
<tr>
<th>Sample</th>
<th>$^7$Be [Bq/g]</th>
<th>$^3$H [MBq/g]</th>
<th>$^{22}$Na [Bq/g]</th>
<th>$^{110m}$Ag [Bq/g]</th>
<th>$^{40}$Y [Bq/g]</th>
</tr>
</thead>
<tbody>
<tr>
<td>Target window</td>
<td>480</td>
<td>24</td>
<td>0.4</td>
<td>−</td>
<td>−</td>
</tr>
<tr>
<td>Target</td>
<td>73</td>
<td>25</td>
<td>−</td>
<td>−</td>
<td>0.2</td>
</tr>
<tr>
<td>Moderator</td>
<td>535</td>
<td>21</td>
<td>1.2</td>
<td>−</td>
<td>−</td>
</tr>
</tbody>
</table>

counting (LSC) with standard measurement technique. Typical radionuclide contents of the three cooling systems are shown in Table 1.

While the tritium production is comparable in all three loops, the target circuit shows the lowest content of $^7$Be, and the target window cooling contains additional impurities of trace elements like Zn, As and others in small quantities, which are not shown in the table. We selected therefore the moderator loop for filtering, because it contains the highest amount of $^7$Be, produced per gram water and only a low amount of contaminations from other isotopes, especially $^{22}$Na, which has a long half-life of 2.6 yr and induces an additional dose rate due to the $\gamma$-line at 1275 keV.

3.2 Design of the filter device

The separation is performed by use of a stainless steel filter device (Fig. 2) containing 100 mL ion exchanger (LEWATIT mixed bed; Bayer, Canada). The filter is mounted into the moderator circuit of the SINQ cooling facility. The vessel is shielded by 5 cm lead and the total weight of the device is around 40 kg. The content of $^7$Be accumulated on the ion-exchanger can be roughly correlated with the measured dose rate. At the end of the sampling and after the necessary cooling time, the D$_2$O is poured out, the filter is dried with air and the device is dismantled.

3.3 Processing of the filter for separation of $^7$Be

The separation of $^7$Be from the SINQ cooling water filter device and the purification of the final product is implemented in a hot cell. The scheme of the $^7$Be separation system is presented in Fig. 3.

3.3.1 Filter transfer into the hot cell

The filter with the embedded shielding is transferred manually and positioned in upright position. The valves of the filter are replaced by hose-barb connectors and tubing for ion-exchange transport. Then, the hot cell is closed and all following procedures are performed remotely controlled.

3.3.2 Transfer of ion exchanger into the elution column and $^7$Be elution

The ion-exchanger from the filter device is transported to an elution column (see Fig. 3) with water injected manu-
ally (syringe with 3-way valve). The water used to transport the resin (500 mL) is collected in a receptacle vessel (called “Eluate”; see Fig. 3) and then transported out of the hot cell by means of a pump (P4).

Tritium activity is measured and if necessary the water is disposed as waste. The column containing the ion exchange resin is washed with 3 M HCl transported by injection (syringe with 3-way valve) and the eluate is collected also in the receptacle vessel (Eluate).

3.3.3 Purification of 7Be

The purification of 7Be from the decay product 7Li and impurities such as 22Na, 110mAg, 88Y etc. is performed by cation exchange. The system consists of two main parts – a Rota-evaporator and a cation exchange column. In the Rota-evaporator the beryllium eluate solution is evaporated and concentrated. Water vapours are condensed and collected in a way that no tritium can escape the system. Then the sample is conditioned in 0.1 M HNO3 and passed on a Dowex 50 × 8 cation exchanger. With 0.1 M HNO3, 22Na as well as boron in form of borate is washed out and collected in a 100 mL plastic vessel. Then 7Be is eluted with 1 M HNO3 in a 100 mL plastic vessel shielded in a lead container. Containers with 22Na and 7Be are sealed and removed from the hot cell for further use. The cation exchange column is removed as well and the ion-exchanger disposed as solid waste carrying the other radionuclides i.e. 54Mn, 110mAg, 88Y. Waste packing can be done in the hotcell to reduce the dose rate for personnel.

4. Results

4.1 Development of optimal separation conditions

Depending on the requirements of the final use, the 7Be activity produced should be free of contaminants as much as possible. In particular, the sorption behaviour of Be as well as Li, Na, Ag and Y on the cation exchanger

\[ \text{Be(II)} \]
\[ \text{Li(I)} \]
\[ \text{Na(I)} \]
\[ \text{Ag(I)} \]
\[ \text{Y(III)} \]

Fig. 4. Dependence of the distribution coefficients of mono-, bi- and tri-valent cations on the acid concentration in the system \( \text{H}_2\text{O}/\text{HNO}_3 \) – Dowex 50 × 8.

4.2 Performance of the first collection and separation campaign

In March 2011, the first campain for the 7Be production with the developed device was started. About 200 GBq were adsorbed on the filter during a collecting time of 3 month. Besides the desired 7Be, which could be nearly quantitatively obtained after purification, also 20 MBq 22Na were separated. The activity was transferred into the required chemical forms – diluted HCl and HNO3. The radioactive waste produced is shown in Table 2. It consists mainly of about 50 g ion exchanger and 0.5 L of aqueous waste.

Table 2. Summary of radioactive waste produced within one campaign.

<table>
<thead>
<tr>
<th></th>
<th>22Na</th>
<th>3H</th>
<th>54Mn</th>
<th>88Y</th>
<th>7Be</th>
</tr>
</thead>
<tbody>
<tr>
<td> </td>
<td>~ 1 MBq</td>
<td>&lt; 1 GBq</td>
<td>traces</td>
<td>traces</td>
<td>100 MBq</td>
</tr>
</tbody>
</table>
4.3 Development of a first beam at the CIRCE accelerator laboratory

A $^7$Be beam is routinely produced at the 3 MV Pelletron tandem accelerator at CIRCE (Center for Isotopic Research on Cultural and Environmental heritage) of INNOVA-Seconda University of Naples, Caserta, Italy, using the radioactive product obtained via the $^7\text{Li}(p,n)^{10}\text{Be}$ reaction.

In order to test the beam extraction procedure, two test cathodes were produced using two different samples produced at PSI. The first one, 0.5 GBq $^7$Be in a 0.5 M HCl solution, was similar to the one that is obtained by the chemical separation routinely used at CIRCE, in order to have a reference to the results of usual cathodes. The second sample consisted of 0.5 GBq in a 0.5 M HNO₃ solution. That should allow to obtain cathodes that contain BeO, produced by decomposition of Be(NO₃)₂ at 200 °C. The presence of BeO in the cathode could, as shown in [15], enhance the yield $^{10}\text{Be}$.

However, due to its oxidizing chemical properties, HNO₃ is not compatible with the standard cathode material, Cu. Therefore, the solution was dried at low temperature, dissolved in 0.5 M HCl, dropped in the cathode and then heated to 200 °C. This process results in at least a partial transition of the nitrate into BeCl₂. The cathode production took place about three months after the end of sample collection at PSI, and the final cathode activity was about 100 MBq.

$^7$Be, $^9$Be and $^{10}$Be beams were produced from BeO molecular ions selecting mass 23, 25, and 26, respectively. A terminal voltage HV = 1.726 MV corresponding to 4 MeV was chosen for $^7$Be. The terminal voltage was scaled for $^9$Be and $^{10}$Be in order to have the same velocity in the terminal and hence the same charge state. An Ar windowless gas cell was used to produce an energy shift of the isobaric contaminants $^7$Li and $^{10}$B from $^7$Be and $^{10}$Be, respectively. The beam energy spectrum was obtained by means of a Si detector inserted on the beam axis after tight collimation.

The first cathode yielded a current of about 20 pA $^7$Be, with no $^7$Li contamination observed. On the contrary, analysed mass = 10 beam turned out to be composed of both $^{10}$Be (10%) and $^{10}$B (90%). The high content of $^8$B is caused by the use of glass vessels consisting of boron silicate. Since the chemical separation itself guarantees a nearly complete separation of boron [16], this disadvantage can be avoided by use of plastic vessels if necessary. The deduced $^{10}$Be current was 100 pA. Finally, a $^9$Be beam current of about 2 nA was observed, with no contamination. The second cathode gave, within the experimental uncertainty, the same results, in spite of the different starting material. That is most likely due to the transition of nitrate to BeCl₂ mentioned above. Since the ion source conditions changed during the test, no accurate information can be gained about the isotopic composition of the sample. However, results support the presence of similar amounts of $^7$Be and $^{10}$Be, with a significantly larger amount of $^9$Be.

5. Discussion

It could be demonstrated, that the separation of $^7$Be from SINQ cooling water allows for delivery of activity up to several hundred GBq. The collection campaign can be repeated up to 4–6 times a year. The $^7$Be activity can alternatively be transformed into a customer-compatibel form, for instance as a solution (diluted HCl or HNO₃ solution), or evaporated on a backing. The amounts achieved will be sufficient for several scientific experiments, like the neutron induced nuclear reactions $^7$Be($n$,α)$^4$He and $^7$Be($n$,p)$^7$Li, which are planned in the near future at n_TOF CERN (Switzerland) and SARAF (Israel). For these two experiments, targets will be necessary, which contain mainly $^7$Be, without disturbing stable $^8$Be and radioactive $^{10}$Be. This can be realised by an implantation of $^7$Be using the offline radioactive beam technique at ISOLDE CERN into a suitable backing material.

For the application at CIRCE – in comparison to that produced via the $^7\text{Li}(p,n)^{10}\text{Be}$ reaction – the $^7$Be from SINQ has the big advantage, that the delivered solution is ready for use, i.e. no additional chemical separation is necessary. Since the activity is produced by spallation, and a careful chemical purification had been performed, the solution has no other by-products (with the exception of $^{10}$Be) and is nearly free of the isobaric isotope $^7$Li. However, the relatively high content of $^{10}$Be, which cannot be avoided – although not a problem for the beam development itself, because the mass separation in the device is highly efficient – could cause radiation safety problems due to its long half-life. A $^{10}$Be/$^7$Be ratio as low as possible can be reached by using the sample as soon as possible after end of collection. For a routine use of the $^7$Be from SINQ, for instance for the wear analysis, a method for recovery of $^{10}$Be from the cathodes should be developed.

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