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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 (D_2O) 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 byproducts 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 D_2O 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 activitites of several hundred GBq with a very high purity. However, it has to be mentioned that both stable ^{9}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 utilizied. Although a thorough discusion 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}Be radioactive target. Previous attempts to measure this reaction cross section using a ^{7}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 Astrophysics) collaboration has started an experiment, whose aim is to determine the cross section of the ^{7}Be$(p, \gamma)^8$B reaction exploiting the ^{7}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 $E_{cm} = 0.4$–1.0 MeV. The required ^{7}Be is presently produced at ATOMKI *via* the ^{7}Li$(p, n)^7$Be reaction following the procedure described in [9]. The SINQ spallation source could represent an additional, alternative source of ^{7}Be, considering the high amount of ^{7}Be needed and the relatively long time required for its production and radiochemical separation by means of proton activation of ^{7}Li targets.

2.2 The problem of primordial ^{7}Li and the ^{7}Be$(n, \alpha)\alpha$ and ^{7}Be$(n, p)^7$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}Li, predicted by the standard theory of Big Bang Nucleosynthesis (BBN) [10], and the value inferred from observations. New and accurate measurements are necessary in order to investigate the destruction of ^{7}Be during BBN *via* the ^{7}Be$(n, \alpha)\alpha$ and ^{7}Be$(n, p)^7$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}Be for wear analysis

Radioactive tracer techniques are nowadays routinely used for non-contacting, on line wear and corrosion measurements and represent an extremely powerful tool in material 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 application depends on the material to be tested and it is limited to those materials that exhibit a high activation cross section 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 references 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 energy during the implantation in a given material allows a quite wide range of possible radio-isotope depth distributions. 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 powerful tool for comparative studies. The main problem of this technique is the availability of a suitable RIB. A beam of ^{7}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 systems can be seen in Fig. 1. The source is cooled by three separate cooling loops, consisting of a target window cooling (1), target cooling (2), and the moderator tank (3) for slowing down the neutrons to thermal energies.

Determination of the radionuclide inventory was performed using γ-ray spectrometry and liquid scintillation

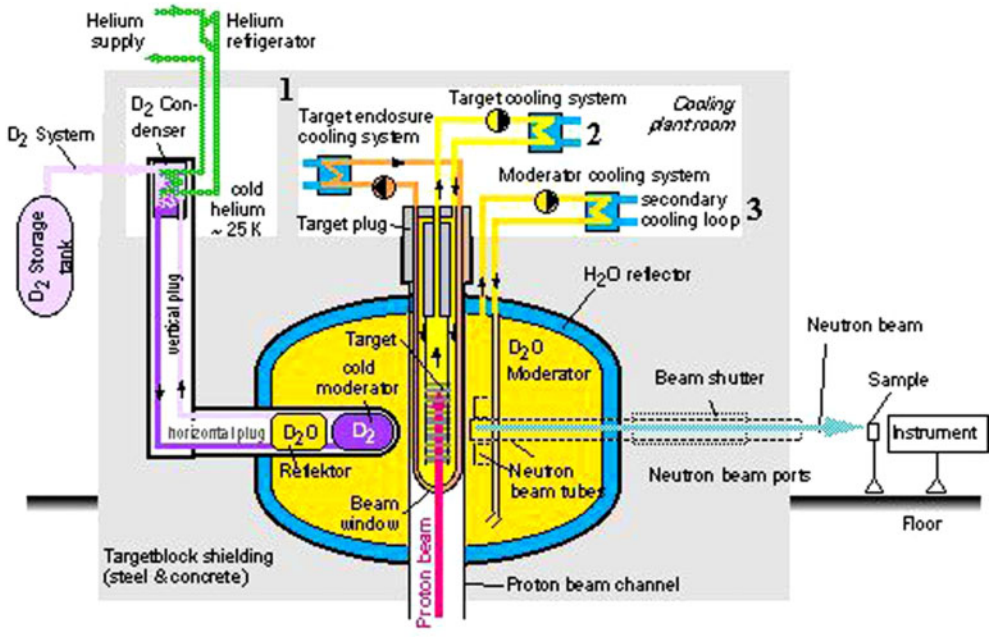

Fig. 1. Vertical cut of the SINQ target station and its cooling circuits (figure taken from [13]).

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

Sample	^{7}Be [Bq/g]	^{3}H [MBq/g]	^{22}Na [Bq/g]	^{110m}Ag [Bq/g]	^{88}Y [Bq/g]
Target window	480	24	0.4	–	–
Target	73	25	–	–	0.2
Moderator	535	21	–	1.2	–

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 γ-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_2O is poured out, the filter is dried with air and the device is dismantled.

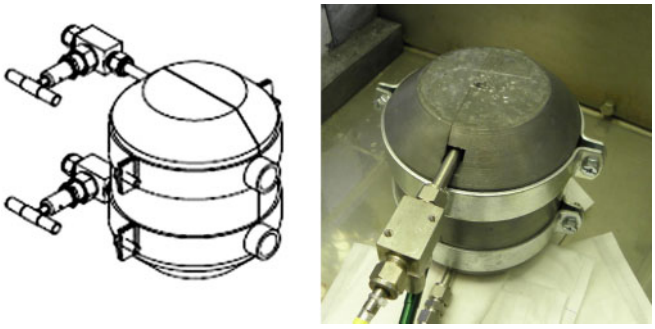

Fig. 2. Drawing and photo of the filter device.

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-

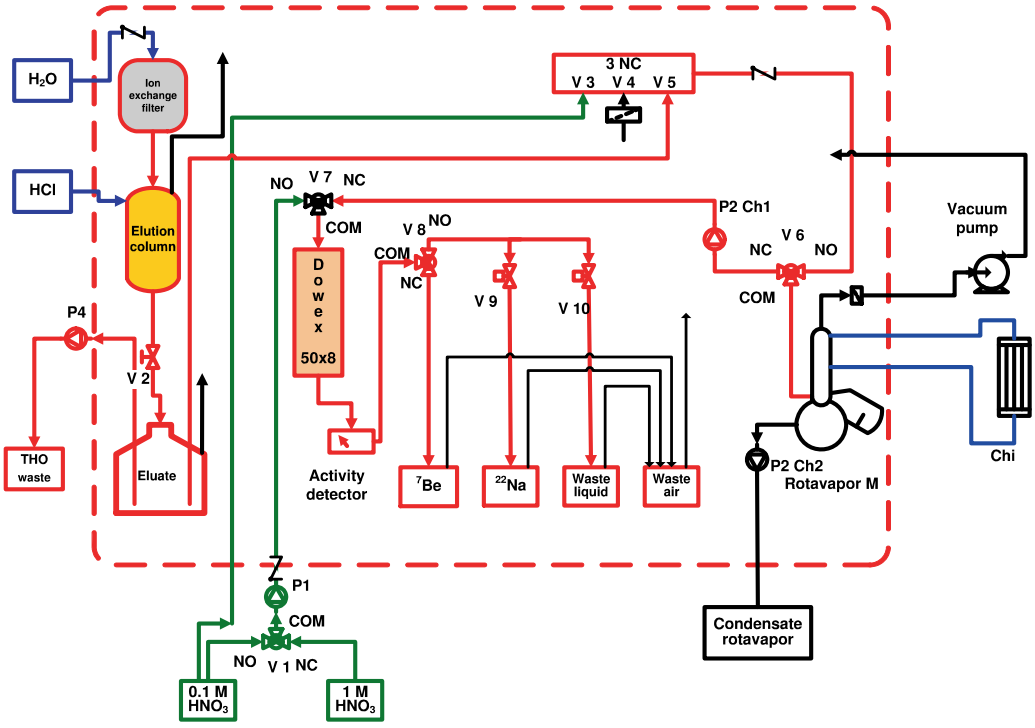

Fig. 3. Remote controlled system for separation of ^{7}Be. * All the components of the system that are inside the red box are installed in the hot cell.

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 ^{7}Be

The purification of ^{7}Be from the decay product ^{7}Li and impurities such as ^{22}Na, ^{110m}Ag, ^{88}Y *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 HNO_3 and passed on a Dowex 50×8 cation exchanger. With 0.1 M HNO_3, ^{22}Na as well as boron in form of borate is washed out and collected in a 100 mL plastic vessel. Then ^{7}Be is eluted with 1 M HNO_3 in a 100 mL plastic vessel shielded in a lead container. Containers with ^{22}Na and ^{7}Be 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.* ^{54}Mn, ^{110m}Ag, ^{88}Y. 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 ^{7}Be 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

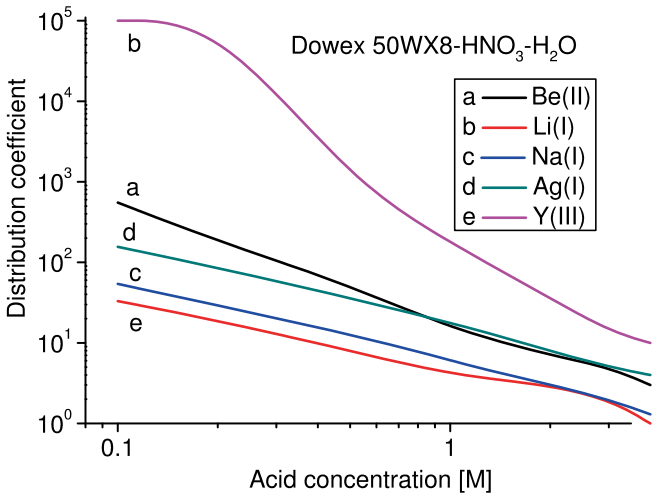

Fig. 4. Dependence of the distribution coefficients of mono-, bi- and tri- valent cations on the acid concentration in the system H_2O/HNO_3 – DOWEX 50×8.

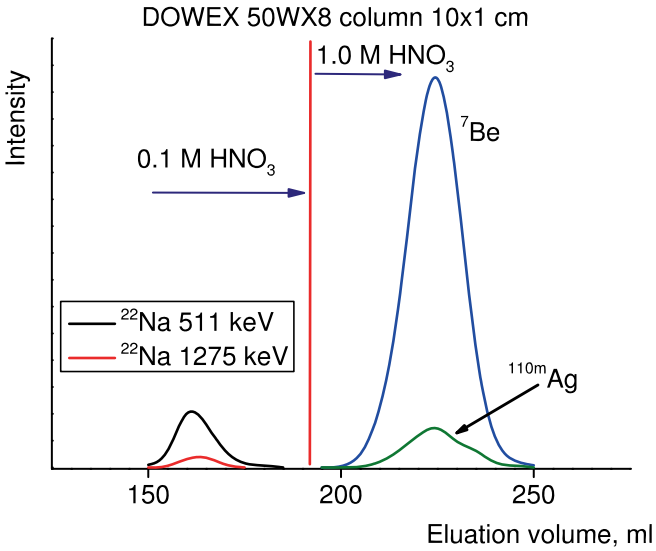

Fig. 5. Elution behaviour of mono- and bi- valent cations in the system H_2O/HNO_3 on DOWEX 50×8.

DOWEX 50W×8 in diluted HNO_3 solutions is of interest. Boron forms anionic borate $(BO_3)^{3-}$, not being adsorbed on cation exchangers. Fig. 4 shows the distribution coefficients for the mentioned cations, taken from literature [14]. It can be seen, that at relatively low HNO_3 concentrations the differences in the K_d values are the biggest, and a separation should be possible in the region from 0.1–0.3 M.

This system was tested with tracers and a column filled with DOWEX 50×8 and dimensions of 1×10 cm, similar to that foreseen for the equipment in the cell. The results of this test experiment are shown in Fig. 5: with 0.1 M HNO_3, a complete elution of Na and – very probably also Li as monovalent ion – was achieved. Increasing the acid concentration to 1 M allows for elution of the entire ^{7}Be-activity within a volume of ~ 50 mL. Silver was coeluted with ^{7}Be in these test experiments, but could not be found in the real samples, because the pre-treatment of the loaded LEWATIT exchanger with 4 M HCl leads to unsoluble AgCl which is retained on the LEWATIT column.

4.2 Performance of the first collection and separation campaign

In March 2011, the first campain for the ^{7}Be 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 ^{7}Be, which could be nearly quantitatively obtained after purification, also 20 MBq ^{22}Na were separated. The activity was transferred into the required chemical forms – diluted HCl and HNO_3. 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.

^{22}Na	^{3}H	^{54}Mn	^{88}Y	^{7}Be
~ 1 MBq	< 1 GBq	traces	traces	100 MBq

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}Li$(p, n)^{7}$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 HNO_3 solution. That should allow to obtain cathodes that contain BeO, produced by decomposition of $Be(NO_3)_2$ at 200 °C. The presence of BeO in the cathode could, as shown in [15], enhance the yield $^{7}BeO^{-}$.

However, due to its oxidizing chemical properties, HNO_3 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 $BeCl_2$. 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 ^{10}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 $BeCl_2$ 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 HNO_3 solution), or evaporated on a backing. The amounts achieved will be sufficient for several scientific experiments, like the neutron induced nuclear reactions ^{7}Be$(n, \alpha)\alpha$ 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 the disturbing stable ^{9}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}Li$(p, n)^{7}$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 shall be developed.

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