Preparation of $^{60}\text{Fe}$, $^{7}\text{Be}$, $^{44}\text{Ti}$ and other samples for nuclear physics experiments

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Abstract. Chemical separation procedures were developed aimed to isolate exotic long-lived radionuclides from proton-irradiated material for use in experiments in the fields of basic nuclear physics and astrophysics. Some examples for separation as well as results of first applications are presented.

1. Introduction
Several long-lived exotic radionuclides like $^{44}\text{Ti}$, $^{60}\text{Fe}$ and others play an important role in nuclear astrophysics topics. Experimental studies of processes and reactions in this scientific field require these isotopes to be used as targets, beams and other samples. In this context, the availability of sufficient amounts of these very rare isotopes is a precondition. One of the few possibilities for their production is the exploitation of activated components available from accelerator facilities at the Paul Scherrer Institute. There, the most powerful proton accelerator in Europe and also world-wide is operated with a proton energy of 590 MeV and a beam intensity of around 2.5 mA. In spallation reactions, a broad variety of exotic radionuclides is produced with high yield.

In the frame of the ERAWAST initiative [1], chemical separation procedures were developed which allow isolating the desired isotopes from several suitable exposed materials like beam dumps, targets, shielding equipment and cooling water. Due to the high radiation doses, usually the procedures have to be carried out using radio-protection equipment. The separation techniques are therefore very expensive and time-consuming.

Three selected examples concerning the separation and application of exotic radionuclides are presented in this report. One source for gaining these isotopes are samples from an irradiated copper beam dump, containing $^{44}\text{Ti}$, $^{60}\text{Fe}$ and others as well as $^{60}\text{Co}$ as the main contaminant. For the separation of $^{7}\text{Be}$ the cooling water of the spallation neutron source (SINQ) of PSI was used.

2. High-activity separation of $^{44}\text{Ti}$, $^{60}\text{Fe}$, $^{26}\text{Al}$ and others from a copper beam dump
The BMA station for pions cancer therapy was operated at PSI from 1980 to 1992. For this period the copper beam dump of the facility received a total dose of approximately 0.16 Ah of 590 MeV protons. After dismantling in 1993, it was cut and samples were taken for analytical purposes. The sampling collected 500 grams of high active copper chips that can be used for separation of exotic radionuclides [2]. The overall gamma analyses showed main nuclides present to be $^{60}\text{Co}$, $^{54}\text{Mn}$, $^{22}\text{Na}$, $^{65}\text{Zn}$ and long lived $^{44}\text{Ti}$ with a daughter nuclide $^{44}\text{Sc}$ [3]. Further analyses by LSC and AMS demonstrated that significant amounts of $^{55}\text{Fe}$, $^{63}\text{Ni}$, $^{26}\text{Al}$, $^{53}\text{Mn}$, $^{59}\text{Ni}$ and $^{60}\text{Fe}$ are present in the copper beam dump. The
analytical results estimate that about 100 MBq $^{44}$Ti, 500 MBq $^{53}$Mn, 7 kBq $^{26}$Al, 8 MBq $^{59}$Ni and 5 kBq $^{60}$Fe are available in the collected copper chips.

Due to the high activity of $^{60}$Co, approx. 4 GBq in total, the separation should be implemented using appropriate shielding, e.g. in a hot cell. The purpose of this work is to develop a simple, selective, efficient and easy to accommodate for remote manipulation procedure for the separation of $^{26}$Al, $^{59}$Ni, $^{53}$Mn, $^{44}$Ti and $^{60}$Fe from gram amounts of the copper beam dump.

The separation procedure, a combination of precipitation and ion exchange, is presented in Figure 1. Copper as the main matrix element interferes with the separation of all elements of interest. For this reason, after the copper dissolution in 7 M HNO$_3$ the solution is conditioned to 1 M HNO$_3$ and Cu(II) is precipitated by saturation with H$_2$S. At these conditions the copper precipitates selectively, while the exotic radionuclides remain in the solution. Further the solution is transferred in 12 M HCl and passed on a Dowex 1x8 anion exchange column. Al and Ni are not retained and are washed out of the column with 12 M HCl. The remaining ions are eluted consequently in the following way, Mn – 10 M HCl, Ti – 8 M HCl, Co – 5 M HCl, Cu – 2.5 M HCl and finally Fe with 0.5 M HCl [4]. Further Al is separated from Ni on a Dowex 50x4 cation exchange column. The Ni is purified with Eichrom Ni resin based on the traditional dimethylglyoxime precipitation chemistry.

The proposed separation procedure is easy for remote controlled implementation in a hot cell. The ion exchange separation of Ni, Al, Mg, Ti and Fe is complete and high decontamination factors for copper and cobalt were achieved.

A setup in a hot cell of a remotely controlled system for consecutive separation exotic radionuclides from gram amounts of copper beam dump is ongoing.

Figure 1. The flowchart of the exotic radionuclides separation procedure.
3. **Laboratory-scale separation of $^{60}$Fe from the copper beam dump and first applications**

The long-lived radionuclide $^{60}$Fe plays a key role in tracing the history of the Early Solar System (ESS) [5]. Attempts at reconstructing the inventory of radioactivity between Fe and Pb require, among others, the exact knowledge of the half-life and experiments aimed to determine the neutron capture cross section.

As a first attempt, two $^{60}$Fe samples were prepared from the copper beam dump in laboratory scale using around 3 g of material, respectively. Liquid-liquid extraction, precipitation and ion exchange are applicable to separate the impurities with a decontamination factor of $\sim 3 \times 10^8$ for $^{60}$Co and $\sim 5 \times 10^6$ for $^{44}$Ti. The chemical separation procedures to be applied depend on the specific requirements of the envisaged experiment.

The first sample was foreseen as target for the determination of the neutron capture cross section at stellar energies. For this application, the content of stable isotopes has to be as low as possible in order to avoid the production of the long-lived $^{59}$Fe. Therefore, precipitation was not possible; all separation steps were performed using liquid-liquid extraction. A detailed description of the target preparation is given in [6]. The results of the neutron capture experiment can be found in [7].

The second sample was used for a re-measurement of the half-life of $^{60}$Fe [8]. Here, the presence of stable carrier is mandatory because isotope ratios are measured by Inductively Coupled Plasma Mass Spectrometry (ICP-MS). On the other hand, for this method it is important to reduce the content of isobaric elements as much as possible. So, an additional purification from Ni had to be performed.

Parts from this very pure $^{60}$Fe sample were used to prepare standards for AMS measurement. Several grams of samples with isotopic ratios of $I=10^{-12}$, $10^{-10}$ and $10^{-8}$, respectively, are now available and can be provided.

4. **$^{7}$Be from SINQ cooling water**

$^7$Be as well as $^{10}$Be are key radionuclides for investigations of several astrophysical processes and phenomena.

One of the "hot topics" is the half-life of $^{10}$Be, where the literature values differ from 1.34 to 1.51 My [9,10]. Additional measurements are, therefore, urgently needed. One possibility is the use of Liquid Scintillation Counting (LSC) for the determination of the activity and ICP-MS for measuring the number of atoms. The calibration of the ICP-MS requires at least 2 mass points in known amounts, and since Be has only one stable isotope ($^6$Be), $^7$Be can serve as the second marker.

Another application of $^7$Be is the study of key reactions concerning the solar neutrino flux, in particular the reaction $^7$Be($p,\gamma)^8$B [11]. Highly-active $^7$Be targets in the range of several 100 GBq are required for such studies.

$^7$Be is produced in considerable amounts in the cooling water ($D_2O$) of the SINQ facility at PSI by spallation reactions on $^{16}$O with the generated fast neutrons. By-products can be nearly neglected, so that this cooling water establishes an ideal source for highly active $^7$Be-samples.

A filtration device shown in Fig.2. was installed as a by-pass for the cooling water into the cooling circle of SINQ for 6 weeks. The apparatus contains 1 litre of the mixed-bed ion exchanger LEWATIT (grain size 400 μm), which absorbs cations and anions as well.

5. **Conclusions**

First successful experiments with laboratory-scale produced amounts of exotic radionuclides like $^{60}$Fe or $^7$Be show the potential of the EAWAST project. With the development of a remote-controlled incell-equipment we will be able to provide a bright variety of these urgently needed rare isotopes in higher amounts.
Fig. 2: Drawing of the adsorption device
1,2 -stainless steel vessel; 3 - inlet; 4 - ring for filter fixing; 5 - filter; 6 - outlet

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