

Ion beam production from rare isotopes with GSI ECR Ion Sources

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Abstract

ECR ion sources (ECRIS) of CAPRICE-type, working at 14.5 GHz, are in use at the High Charge State Injector (HLI) of the accelerator facility at GSI for beam production and at a test bench for development work. The ECRIS is mostly used to produce ion beams from rare isotopes because of its high efficiency and low material consumption. Depending on their material properties beams of rare isotopes are produced from gases, gaseous compounds, solid materials or solid compounds. Gases can be used directly, while solids have to be transformed into the gaseous state for the ECR plasma which is achieved by using resistively heated ovens. As enriched materials are produced by isotopic separation processes their composition including contamination by impurities can be of importance for the handling in the evaporation process and can be detrimental for the beam user if the ion beam contains additional ion species. Characteristics and suitable treatment of materials and production processes are described. Experimental investigations with different sample materials and operational experiences are reported.

INTRODUCTION

Two preaccelerators are providing ion beams for the heavy ion Universal Linear Accelerator (UNILAC) at GSI which in turn delivers the ion beam to an experimental area with high duty cycle (pulses of typ. 5 ms length, 50 s⁻¹ repetition frequency) and to the Heavy Ion Synchrotron (SIS) with low duty cycle (pulses of typically 300 μs length, 1 s⁻¹ repetition frequency). The High Charge State Injector (HLI) is mainly used for the high duty cycle operation. In this mode the CAPRICE type ECR ion source (ECRIS) at the HLI is working in DC mode [1]. The appropriate input velocity for injection into the preaccelerator and the maximum mass/charge ratio of 8.5 determines the choice of ion charge state and extraction voltage, respectively. Typical ion charge states are Mg⁵⁺, Ni⁹⁺, and Xe¹⁸⁺. The respective extraction voltages are between 5 kV and 22 kV. The basic demand to the operation of the ECRIS at the accelerator is to provide a great variety of ion species in stable and reproducible long time operation. These requirements are well fulfilled for the operation with gases. For non gaseous elements which are more than 85% of all elements sometimes gaseous compounds of the desired element can be used. In all other cases the solids must be transformed into the gaseous state

at a suitable vapor pressure in the order of 10⁻³ mbar.

At GSI the oven technique has been preferred because it resembles closely the operation with gases. So it provides higher intensities in comparison with sputtering and lower contaminations compared to the MIVOC method (Metal Ions from Volatile Organic Compounds).

Many experiments at GSI request beams of specific isotopes which require highly enriched isotope materials. For rare isotopes the ECRIS is favorable due to its low material consumption and its high efficiency of conversion of sample material into the ion beam. Fig. 1 shows a statistical overview of the ion species produced for accelerator beam times from 2002 until August 2008. Without taking into account the C²⁺ beam time for cancer therapy the proportion of isotopically enriched sample material is exceeding 80 %.

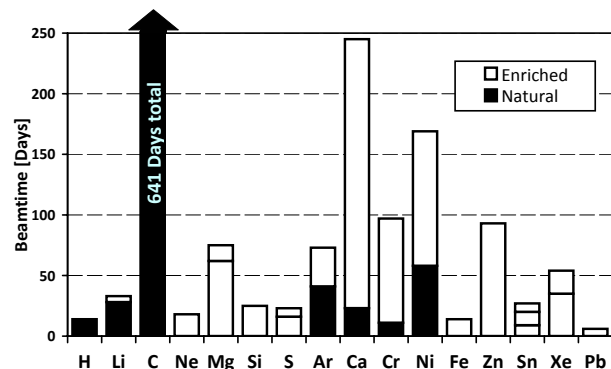


Figure 1: Ion beams produced for accelerator beam times from 2002 to August 2008.

ENRICHED MATERIALS

Natural sample material can be easily obtained in the desired forms and with high chemical purity. For isotopically enriched materials the enrichment processes as well as additional chemical treatments can result in different material properties compared to natural materials. These can be chemical composition including impurities, mechanical and structural characteristics. E. g. metal powders have a huge internal surface facilitating oxidation or hydration processes which are detrimental for the evaporation.

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Table 1: Ion species of rare isotopes produced from the CAPRICE ECRIS for accelerator injection, * designates values including material recycling.

ion species of accelerated charge state q	natural abundance (%)	isotope enrichment (%)	sample material	material consumption ($\mu\text{g/h}$)	average particle intensity ($\text{p}\mu\text{A}$)	efficiency all charge states ($\%/q_i\dots q_f$)	efficiency charge state q (%)
$^3\text{He}^{1+}$	0.00014	99.9	He				
$^6\text{Li}^{1+}$	7.5	95	LiF	1120			
$^{22}\text{Ne}^{4+}$	9.25	99.9	Ne				
$^{25}\text{Mg}^{4+}$	10	99	Mg	620	38	26/2...8	5.7
$^{26}\text{Mg}^{5+}$	11	99	Mg	310/170*	20	26/47*/2...9	6.4/11.5*
$^{30}\text{Si}^{6+}$	3.1	99.5	SiO	1090/580*	13.3	5/10*/3...9	1.3/2.6*
$^{34}\text{S}^{5+}$	4.2	80	SO ₂	660	12.4	14/2...9	2.5
$^{36}\text{S}^{5+}$	0.02	79	SO ₂	420	12.4	23/2...8	4
$^{36}\text{Ar}^{7+}$	0.34	99.5	Ar				
$^{48}\text{Ca}^{7+}$	0.19	96	Ca	210	6.6	40/3...12	5.7
$^{48}\text{Ca}^{10+}$	0.19	96	Ca	200	14.0	43/3...11	12.6
$^{50}\text{Cr}^{7+}$	4.35	96.5	Cr	2300			
$^{54}\text{Cr}^{7+}$	2.37	99	Cr	2300			
$^{58}\text{Fe}^{8+}$	0.28	> 90	Fe	3100/1440*			
$^{64}\text{Ni}^{9+}$	0.93	93	Ni	1300	4.5	4.5/4...13	0.8
$^{70}\text{Zn}^{10+}$	0.6	95	ZnO	3170	7.8	3.3/5...14	0.5
$^{112}\text{Sn}^{15+}$	0.97	99	Sn	890			
$^{114}\text{Sn}^{17+}$	0.65	87	Sn	1000			
$^{124}\text{Sn}^{16+}$	5.8	96	Sn	1200			
$^{124}\text{Xe}^{17+}$	0.1	99.9	Xe				
$^{136}\text{Xe}^{18+}$	8.9	99.9	Xe	550	2.2	50/6...25	2

As the range of usable charge states is strongly limited it may be a problem if the beam is contaminated by another ionic component from the ion source plasma. This can originate from residual gas components but also from the sample material itself.

One lot of ^{54}Cr sample material used for delivering a requested $^{54}\text{Cr}^{8+}$ beam contained several hundred ppm of Al. Due to the different vapor pressures of Cr and Al this was sufficient to generate a considerable superposition of Al^{4+} on top of $^{54}\text{Cr}^{8+}$ as shown in Fig. 2. As the Al impurities did not vanish within a tolerable time the only solution was to change the charge state from $^{54}\text{Cr}^{8+}$ to $^{54}\text{Cr}^{7+}$ in order to obtain a pure ^{54}Cr ion beam on target.

Contaminations can hinder the evaporation process if they reach high vapor pressure at lower temperatures than the bulk material. Thus they may cause an excessive drain current at the extraction. So these contaminations have to be heated off before evaporation of the bulk material can start. This can take a considerable time like for one kind of ^{70}ZnO sample material which started to evaporate at first S then followed by Cd (which was present in the sample material as a fraction of 800 ppm). It took 40 hours until regular evaporation of ^{70}ZnO was achieved. In some cases an additional treatment of the material like preheating in a separate vacuum vessel or under inert gas atmosphere

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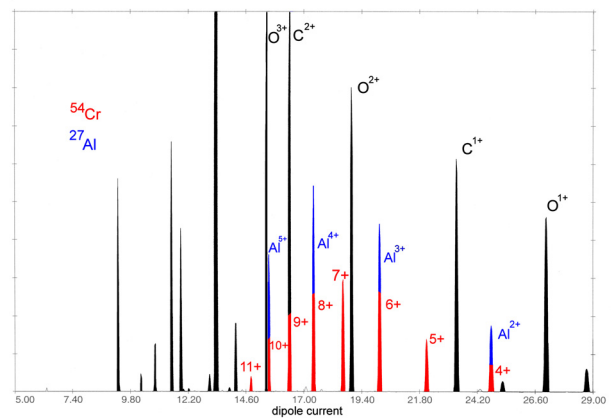


Figure 2: Charge state spectrum of $^{54}\text{Cr} + \text{He}$ with superposition of Al; 100 $e\mu\text{A}$ full scale.

before use in the ECRIS can help to get rid of impurities.

Another example is a ^{54}Cr sample which was pressed into pellets with the aid of paraffin by the provider. It caused such a high evaporation rate that it was not usable in the ion source. A complete heat treatment prior to the beamtime was necessary to get suitable operating conditions.

EFFICIENCY

Table 1 gives an overview of most of the rare isotopes produced from the CAPRICE ECRIS for accelerator injection. The values of efficiency given in the table are not corrected and include the beam losses in the low energy beam transport line to the RF accelerator.

The lowest material consumption rates and the highest efficiencies are obtained for the alkaline earth metals Mg and Ca because one can take advantage of immediate re-evaporation from a hot surface by using a cylindrical insert ("hot screen") [2] in the plasma chamber which is passively heated by microwave and plasma. Much lower efficiencies are observed for materials like Ni and ZnO which suffer from condensation on cold surfaces which cannot be completely avoided.

With increasing atomic number Z of the ions the number of electrons in the atomic shell increase. Hence the charge state distribution covers an increasing number of charge states, because in a steady state plasma the population of charge states is subject to an equilibrium between electron loss and electron capture processes. Thus in a complex atomic shell more electrons are participating in these processes. So the efficiency for ^{136}Xe can be as high as 50 % for all ionic charge states present in the extracted ion beam, but its charge state distribution covers charge states between 5+ and 25+ while the charge state of interest (18+) contains only a small fraction of 2 %.

A low material consumption rate of $210 \mu\text{g/h}$ was observed for ^{48}Ca when optimized on 7+. Up to 40 % of the evaporated ^{48}Ca could be analyzed behind the dipole magnet spectrometer as ions of the charge states 3+...12+ while 5.7 % of the material could be provided as ion beam of the requested charge state 7+. For $^{48}\text{Ca}^{10+}$ a long run of 67 days was characterized by exceptional stability at high intensity level. The lifetime of the oven exceeded 1500 hours while 3 fillings of the crucible were consumed. Taking into account the amount of material recovered from condensates a consumption of only $200 \mu\text{g/h}$ has been achieved which corresponds to a further improved efficiency with respect to $^{48}\text{Ca}^{7+}$ (see table 1).

For ^{26}Mg operation up to 26 % of the sample material is transformed into the ion beam distributed in the charge states 2+...9+. 6.4 % of the material are then analyzed as ions of the requested charge state 5+. The net efficiency increases to 11.5 % if the recovery of material from the plasma chamber and hot screen is taken into account. Fig. 3 shows a charge state spectrum of an analyzed ^{25}Mg beam which is already optimized on $^{25}\text{Mg}^{4+}$ and $^{25}\text{Mg}^{5+}$, respectively. The high intensity of almost $40 \mu\text{A}$ particles at moderate material consumption results in an even better efficiency of 5.7 % for the charge state 4+.

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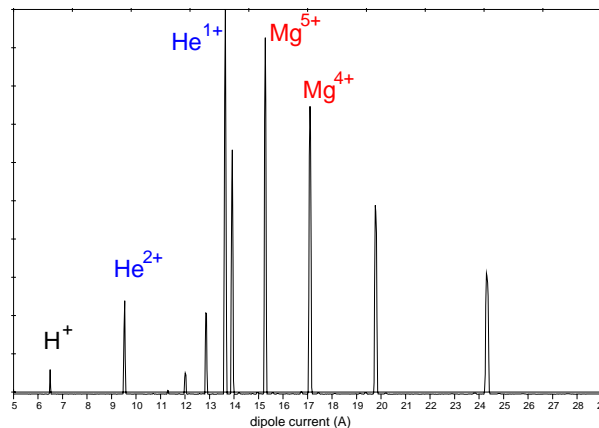


Figure 3: Charge state spectrum of $^{25}\text{Mg} + \text{He}$; $200 e\mu\text{A}$ full scale

EVAPORATION OF SOLID MATERIALS

The basic characteristics of the GSI standard oven (STO) have been described in detail before [3]. Several additional features have been applied to improve stability of evaporation, reduction of condensates and increase of efficiency. It is used for temperatures between 600°C and 1500°C .

For high temperature operation of the STO for materials of low vapor pressure like e. g. Ni, Fe or Cr, an additional heat shield was developed to get a more homogeneous temperature distribution in axial direction of the oven and to reduce the material condensation in its orifice. The heat shield consists of several layers of corrugated tantalum foil with the same aperture as the oven housing. Reliable long time operation of the modified STO could be approved showing the expected reduction of condensates. Thus the material consumption can be reduced and maintenance periods for the oven can be extended.

For low temperature operation of the STO for materials of high vapor pressure a parasitic heating of the oven has to be avoided. Generally, a complete decoupling of ion beam extraction and plasma generation would be desired. Additional aperture rings and enclosure of the complete oven in the injector tube are mechanical measures while the use of an accel-decel extraction system avoids parasitic heating of the oven front by electrons accelerated on axis from the ground electrode of the extraction system into the plasma towards the injection side [4].

COMPOUNDS

The use of gaseous or solid compounds is recommended if elements are not usable due to their physical properties. The use of chemical metal compounds as evaporation source can be an alternative if the vapor pressure of the pure metal is not matching the capabilities of the oven.

Initiated by a request of the Super Heavy Element pro-

gramme for a ^{30}Si beam tests were performed at the ECR injector test setup (EIS) using natural SiO to produce a $^{28}\text{Si}^{5+}$ beam by evaporation from the oven. As the use of ZnO has become a standard procedure for producing Zn ion beams SiO appeared to be the only choice, because the vapor pressure of elementary Si and SiO_2 are by far too low. After the successful test a beam of $^{30}\text{Si}^{6+}$ could be provided from the ECRIS to the accelerator. Highly enriched ^{30}SiO has been used to provide a beam of high stability and intensity for nearly 4 weeks.

PREPARATION OF SAMPLE MATERIALS

Some of the required enriched materials can be procured in suitable form while others have to be transformed prior to be used with the ECRIS. Metallic ^{48}Ca is obtained as $^{48}\text{CaCO}_3$ and is transformed into metallic ^{48}Ca by chemical reduction [5].

A $^{32}\text{S}^{5+}$ -beam has been produced from natural SO_2 gas at GSI several years ago while at GANIL enriched (63%) $^{36}\text{SF}_6$ has been used for ion beam production [6]. At GSI SF_6 was not chosen as sample material because of the high number of F-atoms in the molecule and because of the high chemical reactivity of F. Following the good experiences with CO_2 gas and with SO_2 gas for producing C ion beams and S ion beams, respectively, SO_2 was also chosen for the rare isotopes.

As the isotope material could only be obtained as elementary material it was necessary to do a conversion to SO_2 gas. A special procedure could be developed to convert at first 2500 mg of highly enriched elementary ^{34}S and then to convert 1950 mg of highly enriched elementary ^{36}S , respectively.

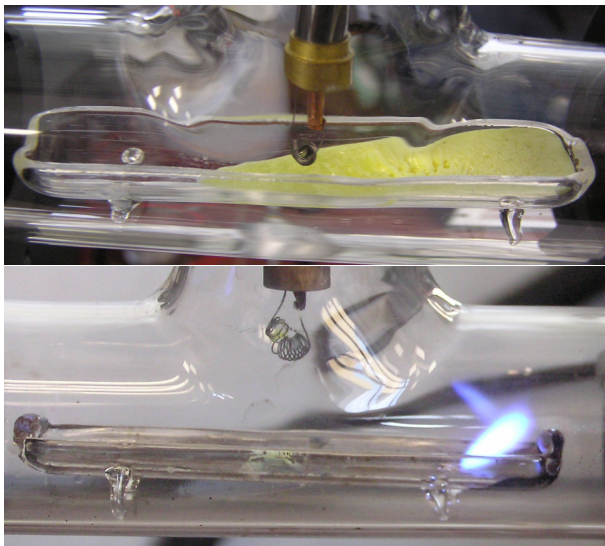


Figure 4: Quartz boat with elementary ^{36}S before and during the combustion process

The sulfur material (powder in elementary form, $\approx 80\%$ enrichment) was melted in a boat of quartz glass at a temperature of 160°C and then placed inside a glass "T" part with KF flanges DN25 as vacuum recipient. Through the top flange a sparking plug was introduced, and the other flanges were used as gas inlet and outlet. Three flasks made from stainless steel were connected in series with cooling mantles made from copper tubes. The flasks were covered inside with PTFE; each of them had two valves at both sides. The whole vacuum-tight system was first pumped to 10^{-3} mbar and then flushed with helium. The flasks were cooled down to -140°C by liquid nitrogen vapor flowing through the copper cooling tubes. Then helium flow was switched to oxygen gas flow of ≈ 50 ml/min. The oxygen gas was purified from water content by molecular sieves. The sparking plug was used to ignite a self supporting combustion of the sulfur material. The complete oxidation was finished after ≈ 10 min (see Fig. 4). At the exit from the third flask, the outgoing gas was bubbled through a SO_2 trap - a glass flask filled with NaOH solution. No sulfur has been found in the trap. The content of condensed $^{36}\text{SO}_2$ has been measured in all three flasks after the oxidation process. More than 80% of produced $^{36}\text{SO}_2$ have been found in the first flask, while 16% have been condensed in the second flask and 2–3% in the last one, respectively. The total conversion yield was $\approx 99\%$. Afterwards the flasks were heated up to equilibrate the gas pressure. Then each flask was closed from both sides and disconnected separately.

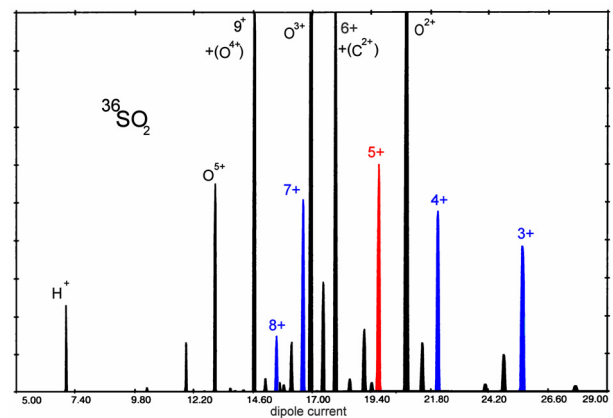


Figure 5: Charge state spectrum of $^{36}\text{S} + \text{O}$; $100\text{ e}\mu\text{A}$ full scale

A test run with $^{34}\text{SO}_2$ showed that the sample material is well suited for ECRIS operation. The long time stability of about $\pm 3\%$ with only a few deviations was demonstrated in a continuous run of 8 days. $^{36}\text{SO}_2$ was then used for an accelerator beam time delivering $^{36}\text{S}^{5+}$ under same conditions. As Fig. 5 shows $60\text{ e}\mu\text{A}$ of $^{36}\text{S}^{5+}$ could be obtained, which is the required charge state for acceleration. The average material consumption could be determined to be $420\text{ }\mu\text{g}$ of ^{36}S per hour.

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