

RESEARCH ON METALLIC ION BEAM PRODUCTION WITH ELECTRON CYCLOTRON RESONANCE ION SOURCES

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Abstract

Many experiments in nuclear physics request the production of metallic ion beams. All elements from Lithium up to Uranium are of interest and most of them are required as a specific isotope which demands commonly enriched materials. Depending on the material properties, beams of rare isotopes can be produced from solid materials or solid compounds. In this report the results of experiments carried out under a collaboration of JINR and iThemba LABS on the production of metallic ions from Electron Cyclotron Resonance (ECR) Ion Sources using resistive oven evaporation, Metal Ions from Volatile Compounds (MIVOC) method and sputtering technique will be presented.

INTRODUCTION

Several methods for the production of ions from solid materials have been developed. Solid materials can be evaporated from a resistor or inductive oven inserted into a source chamber [1, 2]. Refractory metals can be sputtered by plasma ions [3] or inserted into plasma with subsequent heating by energetic plasma electrons (“insertion technique”) [4, 5]. Another way of producing ions of solids is to feed plasma of an organometallic compound using the MIVOC method [6]. The selection of the best method to feed solids into ECR ion sources strongly depends on specific properties of materials.

OVEN EXPERIMENTS

Development of the oven evaporation method for production of ions of solids for FLNR JINR ECR ion sources was stimulated by the requirements of production of intense ^{48}Ca beam, which is the key ingredient in the experiments on synthesizing of new heavy nuclei.

To solve this problem, a new method for the solid material feed into the ECR source was developed. The combination of a micro oven with a hot tantalum liner inside the discharge chamber allowed the production of intense beams of ions of metals with relatively low evaporation temperature (Li, Mg, Ca, Bi) [7]. This development allowed long-term experiments on synthesis of super heavy elements during last 20 years and led to discovery of new super heavy elements with $Z = 113-118$ [8].

The experience of FLNR ECR ion source group was successfully applied in a collaboration of JINR and iThemba LABS on the production of metallic ions from

ECR ion sources at iThemba LABS.

The experiments on production of ions of solids by oven evaporation method were performed with the GTS2 ECR ion source [9]. The layout of the beam line used for the experiments is shown in Fig. 1. In all experiments the source was operated with 14 GHz frequency.

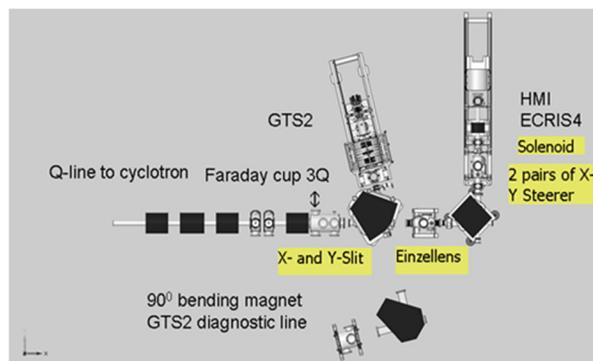


Figure 1: Experimental layout for the oven and MIVOC experiments.

The GTS2 ECR ion source is equipped with two ports through which electrically heated ovens can be introduced into the plasma chamber. The oven used for the measurements was designed and manufactured by the ion source group at FLNR. The design is based on the micro oven [10], which has been successfully used for several years for the production of ^{48}Ca and Li ion beams. The present design of the oven allows using a crucible with an inner volume of 480 mm^3 , which is about 6 times more than that of the original design. The calibration for the oven inner temperature as a function of the oven electrical heating power is shown in Fig. 2. The oven was mechanically and electrically connected to the oven support of the GTS2. The oven is positioned inside the plasma chamber in a way that the tip of the oven has a distance of 30 mm to the bias disc. In addition, a liner made from 0.1 mm stainless steel sheet was installed inside the plasma chamber. The liner has folds on both ends to keep it in 1 mm distance to the plasma chamber wall to reduce the thermal contact. This results in a higher temperature of the liner by means of microwave and plasma heating thereby preventing the condensation of the oven material.

The beam extracted from GTS2 was either analysed with Faraday cup 3Q behind the 104° -bending magnet in the Q-line or with Faraday cup Q2 in the diagnostic beam line behind the 90° -bending magnet (see Fig. 1).

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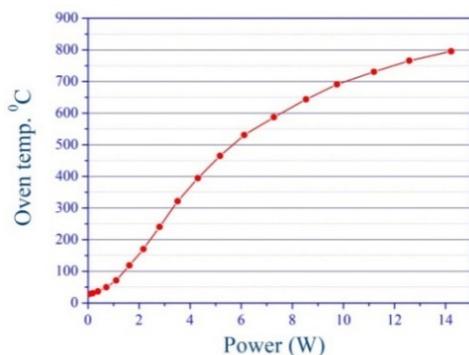


Figure 2: Oven inner temperature vs electrical heating power.

Calcium Experiments

The stainless steel crucible was loaded with approximately 500 mg of natural Ca granulate (97% ^{40}Ca). The granulate surface was prepared with a smooth file. The oven and the liner were conditioned for 15 hrs with 100 W of RF power with He support gas and oven heating power of 1.1 W ($\sim 100^\circ\text{C}$).

The source was optimized for the extraction of $^{40}\text{Ca}^{9+}$ ions. Fig. 3 shows the spectrum of extracted Ca ions at extraction voltage of 10 kV, RF power $P_{\text{rf}} = 250$ W, horizontal slit width $\text{SLX} = 20$ mm, vertical slit width $\text{SLY} = 30$ mm, oven heating power $P_0 = 5.6$ W. For this experiment He gas was used as a supporting gas. A $^{40}\text{Ca}^{9+}$ ion current of 100 μA was measured in Faraday cup 3Q.

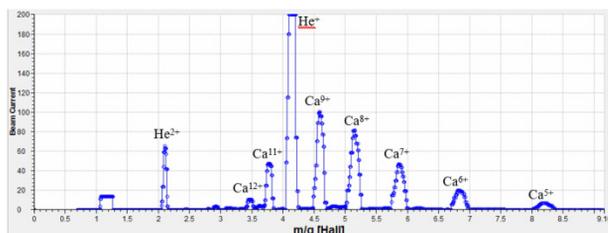


Figure 3: Spectrum of the extracted Ca ion beam.

The Faraday cup current for $^{40}\text{Ca}^{9+}$ ions was monitored for constant source settings over 12 hours in which the current dropped by 20%.

Lithium Experiments

After experiments with Ca the crucible was loaded with approximately 200 mg of Li. For Li operation it is essential that the oven opening is closed with a small ball twirled of 0.2 mm Ta wire because the material's melting point is below its operation temperature. The oven was then moved into the source at the same position as described before. The oven was conditioned by gradually increasing the oven temperature to 100°C . The source was then started up, optimized for Li^{2+} and after one hour of operation the spectrum shown in Fig. 4 was measured in Faraday cup 3Q at extraction voltage $U_{\text{ex}} = 10$ kV, RF power $P_{\text{rf}} = 200$ W, horizontal slit width $\text{SLX} = 20$ mm, vertical slit width $\text{SLY} = 30$ mm, oven heating power $P_0 = 3.8$ W ($\sim 350^\circ\text{C}$). The source is also operated with He supporting gas.

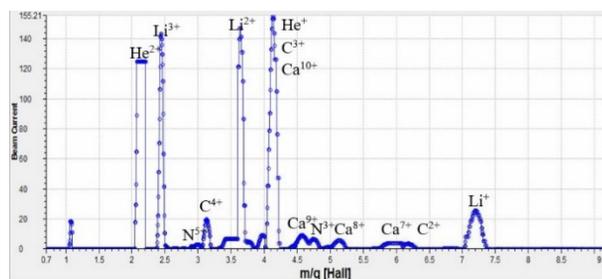


Figure 4: Spectrum of the extracted Li ion beam.

Three observations from the spectrum are evident: firstly, there is still some Ca left in the source from the previous experiment (appears as Ca^{7-10+} peaks). Secondly, C ions are detected which were not measured in the Ca experiment (see Fig. 4). This contamination can be explained by the fact that the Li material was stored in oil. Thirdly, the source is very well conditioned which is obvious from the low intensities of the N^{3+} and N^{5+} peaks. This is important for the determination of the Li beam intensity because all charge states of Li ions are interfered by Nitrogen ions with corresponding double charge states.

Intensities measured for Li ions with charge state 2+ and 3+ were more than 100 μA . Next the beam extracted from the source was directed into the diagnostic beam line for long-term experiments. The transmission through this beam line is roughly half of that of the Q-line. The 90° -bending magnet was adjusted for Li^{2+} .

The source was operated with Li beam for a total period of 6 days. During this period of time the Li^{2+} beam current dropped from 55 to 40 μA . The short term stability was better than $\pm 5\%$. Only once the gas flow of the supporting gas was slightly adjusted to improve the source stability.

The source was then tuned for Li^{3+} production and the beam was transported through Q-line to the entrance of the Solid Pole Cyclotron (SPC2). For cyclotron injection the slits in the Q-line were closed to 10 mm (horizontal) and 20 mm (vertical) resulting in a beam current of 17 μA measured in Faraday cup in front of SPC2. This beam was then injected into SPC2 and accelerated to 10 MeV. The Li^{3+} current extracted from SPC2 was 2.3 μA .

Magnesium Experiments

The stainless steel crucible was loaded with approximately 350 mg of natural Mg (79 % ^{24}Mg , 10 % ^{25}Mg , 11 % ^{26}Mg). The material was chipped with a knife from a solid disc. The chips were compressed inside the crucible. The oven was conditioned with 1.1 W ($\sim 160^\circ\text{C}$) of oven heating power for 12 h. In addition, the source was operated with 60 W of RF power with He as support gas. The source was then optimized for the extraction of $^{24}\text{Mg}^{7+}$ ions. The spectrum shown in Fig. 5 was obtained at following source settings: extraction voltage $U_{\text{ex}} = 10$ kV, RF power $P_{\text{rf}} = 430$ W, oven heating power $P_0 = 3.8$ W (the oven temperature is approximately 390°C), horizontal slit width $\text{SLX} = 5$ mm, vertical slit width $\text{SLY} = 15$ mm. Furthermore, beside the main peaks

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for the ^{24}Mg peaks for the isotopes 25 and 26 can be identified.

A maximum $^{24}\text{Mg}^{7+}$ ion current of $40\ \mu\text{A}$ was measured in Faraday cup 3Q. When the extraction voltage was increased to 15 kV, the $^{24}\text{Mg}^{7+}$ current amounted to $90\ \mu\text{A}$. This can be either due to extraction conditions (Child-Langmuir) and/or due to a higher transmission through the bending magnet into the Q-line. The Faraday cup current for $^{24}\text{Mg}^{7+}$ ions was monitored for constant source settings over 12 hours in which the intensity varied by less than 5%.

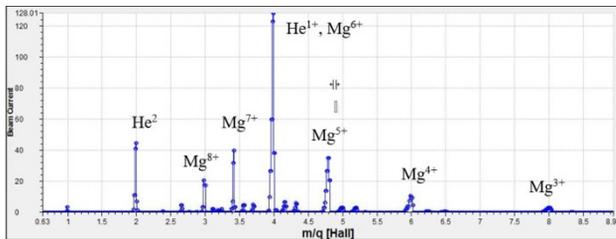


Figure 5: Spectrum of the extracted Mg ion beam.

Bismuth Experiments

The crucible was loaded with approximately 2 g of Bi. For Bi operation it is essential that the oven opening is closed with a small ball twirled of 0.2 mm Ta wire because the material's melting point ($270\ \text{°C}$) is below its operation temperature ($630\ \text{°C}$). The oven was then moved into the source at the same position as described before. The oven was conditioned at a temperature of $200\ \text{°C}$ for 12 hrs.

The source was optimized for Bi^{27+} . The spectrum, shown in Fig. 6 shows the intensities of 23 and $16\ \mu\text{A}$ for Bi^{27+} and Bi^{30+} , respectively. The source parameters are: extraction voltage $U_{\text{ex}} = 15\ \text{kV}$, RF power $P_{\text{rf}} = 460\ \text{W}$, horizontal slit width $\text{SLX} = 15\ \text{mm}$, vertical slit width $\text{SLY} = 35\ \text{mm}$, oven heating power $P_0 = 5.7\ \text{W}$ ($510\ \text{°C}$).

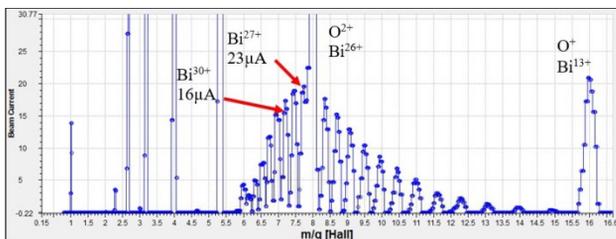


Figure 6: Spectrum of the extracted Bi ion beam.

The results so far suggest that the oven position might be too close to the plasma for Bi operation. This is because maximum beam intensities were obtained at lower oven heating powers than expected. Experiments at FLNR gave best Bi operation at oven temperature of $630\ \text{°C}$ corresponding to oven heating powers of 8.4 W. The heating power of 5.7 W used in our experiments corresponds to only $510\ \text{°C}$, the additional oven heating could be from plasma heating. Further experiments in which the oven position will be varied need to be carried out.

MIVOC EXPERIMENTS

The MIVOC technique was first developed in Finland in 1994 [6]. In the MIVOC technique, solid state volatile metallic compounds can produce vapor gas having metallic atoms even at room temperature. Compounds are placed in a separate vacuum chamber connected to the plasma stage of the ECR ion source.

In FLNR the investigations on MIVOC method were motivated by the demand on production of intense ^{50}Ti ion beam for further progress in synthesis of super heavy elements. The series of experiments with natural and enriched compounds of Ti ($\text{CH}_3)_3\text{C}_5\text{Ti}(\text{CH}_3)_3$ were performed at the ECR test bench leading to production of intense beams of Ti ions ($80\ \mu\text{A}\ ^{48}\text{Ti}^{5+}$ and $70\ \mu\text{A}\ ^{48}\text{Ti}^{11+}$) [11]. Experiments on the production of Co, Cr, Ni, V, Ge and Hf ion beams were also performed at the test bench of ECR ion sources.

This development allowed long-term experiments with accelerated ^{50}Ti and ^{54}Cr ion beams at the U-400 cyclotron.

The experience obtained during MIVOC method development in FLNR was applied in a collaboration of JINR and iThemba LABS for production of Ni ion beam from isotopes with low natural abundance. The experiments were performed with ECR4 [12] ion source in experimental layout shown in Fig. 1.

The experiments were performed with commercial Nickelocene ($\text{Ni}(\text{C}_5\text{H}_5)_2$) with natural abundance, in-house produced 99 % enriched 60-Nickelocene and in-house produced 98 % enriched 62-Nickelocene. The procedure of Nickelocene synthesis and details of the experiments are described in [13]. Currents of $30\ \mu\text{A}$ for charge state 8+ were obtained.

From the performed experiments we can conclude, that the drift length of the Nickelocene molecules into the plasma chamber should be reduced by excluding the insulator of the injection system, and the injection system should be kept on source potential to prevent possible gas discharges which might interact with the Nickelocene flow. The other possible way to increase the conductivity of the injection system and thus increase the beam current is the modification of the ECR4 ion source like similar CAPRICE-type ECR source mVINIS [14].

SPUTTERING EXPERIMENTS

The experiments on production of ions of solids by sputtering method were performed using the DECRIS-SC ion source [15] at the CI-100 cyclotron [16].

The CI-100 cyclotron is mainly tuned for acceleration of ions with A/Z ratio close to 5 ($^{132}\text{Xe}^{26+}$, $^{86}\text{Kr}^{17+}$ and $^{40}\text{Ar}^{8+}$) for the energy of about 1.2 MeV/n. To avoid the changing of RF system tuning $^{51}\text{V}^{10+}$ ion was chosen for the first experiments. The DECRIS-SC is equipped with two ports, one of these ports can be used for insertion of oven or sputtering sample. The experimental equipment includes a negatively biased metal sample positioned close to the ECR plasma. The metal sample of V was supported by water cooled

supporting rod. The position of rod was remotely controlled. The rod position and high voltage for sputtering were optimized for maximizing the beam intensity of V^{10+} ions. Ar gas was used as a sputtering gas. One of the spectrum is shown in Fig. 7, where the position of V^{6+} , V^{7+} , V^{8+} and V^{10+} peaks are indicated.

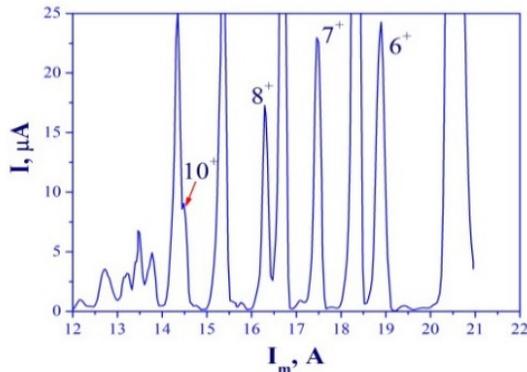


Figure 7: Spectrum of the extracted V ion beam. Positions of V^{6+} , V^{7+} , V^{8+} and V^{10+} peaks are indicated.

Due to the pure resolution of the injection system the tuning of the source was performed with accelerated beam at the final radius of cyclotron. The intensity of accelerated beam was 50 – 60 nA at the final orbit. The beam was produced at an extraction voltage of 14.65 kV, 250 W of RF power. The sputtering voltage and current amounted to 5 kV and 8 mA, respectively.

The accelerated beam was used for irradiation of ceramics samples during 125 h. The energy of accelerated beam was measured and constitutes of 62.4 MeV. The material consumption was measured as 1.32 mg/h.

In future experiments we plan to produce $^{181}\text{Ta}^{35+}$ ($A/Z = 5.17$) and study the effect of argon and oxygen as sputtering/mixing gases. Also, similar system can be applied at the GTS2 source.

CONCLUSION

Over the past few years, notable results and significant progress have been achieved by FLNR JINR and iThemba LABS collaboration in the production of intense multiply charged metal ion beams in ECR ion sources. Stable beams of Li, Mg, Ca and Bi were produced with a resistive oven from the GTS2 ion source. The Li^{3+} ion beam was accelerated to 10 MeV with SPC2; the extracted current was 2.3 μA . Further optimisation of this method is required from the point of view of oven position and preventing oven overheating by plasma. The MIVOC method allows the production of intense ion beams with long-term stability. The further increase in beam intensity can be achieved by optimization of the injection line and remote tuning of compound flow.

The first experiments with sputtering technique were performed at FLNR JINR, and the accelerated beam of V^{10+} was produced at CI-100 cyclotron. This method can be also applied at the GTS2 source.

REFERENCES

- [1] D. J. Clark and C. M. Lyneis, “The production of beams from solid materials at the LBL ECR source”, *J. Phys. Colloq.* vol. 50, pp. 759-766, 1989.
- [2] R. Harkewicz, “Efficient production of a ^{48}Ca beam from oxide material in an electron cyclotron resonance ion source using a low power miniature oven”, *Rev. Sci. Instrum.*, vol. 67, pp. 2176–2178, 1996.
- [3] R. Harkewicz *et al.*, “Ion plasma sputtering as a method of introducing solid material into an electron cyclotron resonance ion source”, *Rev. Sci. Instrum.*, vol. 66, pp. 2883–2887, 1995.
- [4] R. Geller, P. Ludwig, and G. Melin, “Metal ion production in ECRIS”, *Rev. Sci. Instr.*, vol. 63, pp. 2795–2800, 1992.
- [5] T. Nakagawa *et al.*, “Recent developments of RIKEN ECR ion sources”, in *Proc. 10th Int. Workshop on ECR Ion Sources*, ORNL, Oak Ridge, USA, 1990, pp. 163-172.
- [6] H. Koivisto, J. Arje, and M. Nurmi, “Metal ions from the volatile compounds method for the production of metal ion beams”, *Rev. Sci. Instrum.*, vol. 69, no. 2, pp. 785–787, 1998. doi:10.1063/1.1148539
- [7] V. B. Kutner, S. L. Bogomolov, A. A. Efremov *et al.*, *Rev. Sci. Instrum.*, vol. 71, no. 2, pp. 860-862, 2000. doi:10.1063/1.1150290
- [8] Yu. Ts. Oganessian, A. Sobiczewski, and G. M. Ter-Akopian, “Superheavy nuclei: from predictions to discovery”, *Phys. Scr.*, vol. 92, p. 023003, 2017. doi:10.1088/14024896/aa53c1
- [9] R. Thomae *et al.*, “First commissioning results with the Grenoble test electron cyclotron resonance ion source at iThemba LABS”, *Rev. Sci. Instrum.*, vol. 83, p. 02A323, 2012. doi:10.1063/1.3669790
- [10] A. Efremov *et al.*, “Performance of the ion source DECRIS-14-2”, *Rev. Sci. Instrum.*, vol. 69, no. 2, pp. 662 – 664, 1998. doi:10.1063/1.1148567
- [11] S. L. Bogomolov, A. E. Bondarchenko, A. A. Efremov *et al.*, “Production of intense metal ion beams from ECR ion sources using the MIVOC method”, *Phys. Part. Nucl. Lett.*, vol. 12, no. 7, pp. 824–830, Dec. 2015. doi:10.1134/S1547477115070043
- [12] P. Sortais *et al.*, “ECRIS development at GANIL”, *Rev. Sci. Instrum.*, vol. 61, p. 288, 1990. doi:10.1063/1.1141324
- [13] N. Kheswa, R. Thomae, F. Nemulodi *et al.*, “Production of high intensity nickel-ion beams with high isotope purity with the metal ions from volatile compound (MIVOC) method”, *AIP Conf. Proc.*, vol. 2011, p. 040007, 2018. doi:10.1063/1.5053281
- [14] A. Efremov *et al.*, “Upgrading of the CAPRICE type ECR ion source”, *AIP Conf. Proc.*, vol. 2011, p. 040017, 2018. doi:10.1063/1.5053291
- [15] A. Efremov *et al.*, “Status of the ion source DECRIS-SC”, *Rev. Sci. Instrum.*, vol. 77, p. 03A320, 2006. doi:10.1063/1.2165268
- [16] B. Gikal, S. Dmitriev, G. Gulbekian *et al.*, “Cyclotron based complex IC-100 for scientific and applied research”, in *Proc. 18th Int. Conf. on Cyclotrons and their Applications (Cyclotrons’07)*, Giardini Naxos, Italy, Oct. 2007, pp. 27–32.