Report on experimental results for metal-ion beams

1. Research and development of evaporation oven technique

Induction oven: The first version of the JYFL induction oven developed during ISIBHI/EURONS/FP6 was very promising. However, it had two drawbacks: part of the RF-power was coupled to the surrounding structures and the control unit was fairly complicated to use. Further development was carried out during this task in order to resolve these problems. The power coupling was minimized by using triaxial design shown in Fig. 1. This remarkably decreased the total power of unit needed for the heating of crucible placed inside the induction coil. The lower power of the unit will naturally affect also its life-time. The tuning effort was minimized by using the autoresonance property of the control unit. The first experiment with the improved induction oven was successful and the oven was capable of reaching 2000˚C. The oven structure has been completed but the control unit has to be developed further for the safety of operation. The project suffered from the unexpected problem: the specialist who developed the control unit left the group for the private market and no replacement able to complete the unit was found.

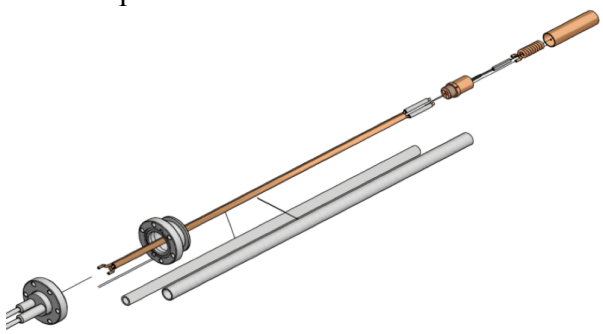

Figure 1: Triaxial design used to minimize the coupling of RF-power to surrounding structures.

Foil oven: The first version of the foil oven was developed during ISIBHI/EURONS/FP6. The oven was capable of reaching 2000˚K but it was not reliable in operation. The main reason was that its operation temperature had fairly strong variation between different experiments. The oven was developed further during FP7 and special attention was paid on the mechanical connections in the high temperature region. Three critical areas, shown in Figure 2, were pinpointed as a result of these considerations: a) locking/connection system inside the region A was improved (see Fig. 2). In this area the structure 5) is connected to the tube 6) for return current, b) support structure between the vacuum lock (shown in Figure 3) and part indicated in figure was added to make assembly more rigid. It was found that after these modifications the variation of the oven resistivity between the different oven assemblies decreased. The oven operation was tested with the evaporation of Cr where the intensity of 7.6 µA for Cr^{8+} beam was achieved. The current through the 20 µm heating foil was 59 A. According to the temperature calibration curve this corresponded to the oven temperature of slightly more than 1500˚C. In addition to the afore-mentioned it was found out that the connection between parts 1) and 2) has to be changed. The coefficient of thermal expansion of molybdenum is smaller than in the case of copper and as a result the connection between these components decreases during the heating process. In order to solve this problem the Co rod has to be inside the Mo support structure. Further experiments will be performed after this modification and the maximum operation temperature will be defined.

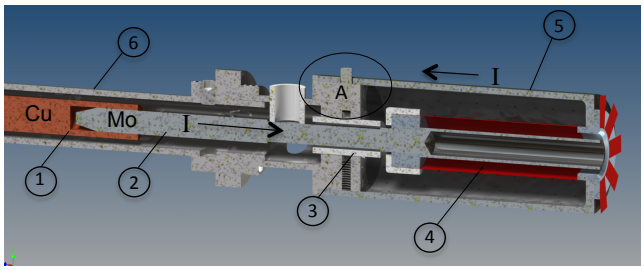

Figure 2: The structure of the foil oven: 1) copper rod, 2) Mo support structure, 3) Al_2O_3 insulator, 4) Ta foil (20 µm in thickness), 5) outer Mo oven structure for return current, 6) SS tube for return current.

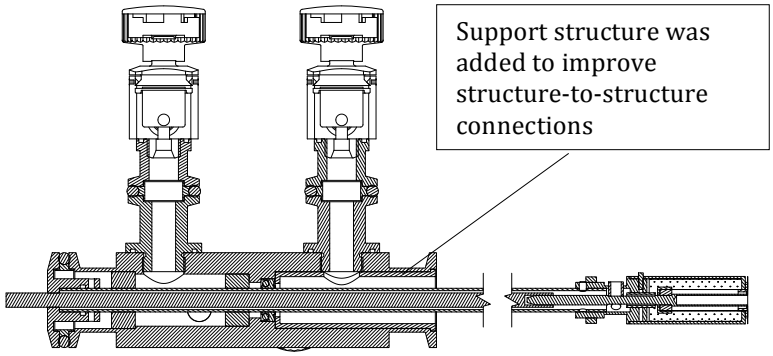

Figure 3: Modification to improve the mechanical structure of the foil oven. Surface-to-surface current conductivity improved and variation in the total resistance decreased.

The original plan was to perform movable oven experiments using the foil oven. According to the hypothesis the production efficiency could increase when the distance between the outlet of the oven and plasma decreases. In other words, the probability of atom to enter the cold plasma chamber wall decreases and consequently the production efficiency increases. In order to insert the oven into the plasma chamber side the opening for the oven needs to be increased (from 12 mm to 21 mm). As Figure 4 reveals the oven would be very close to the plasma chamber wall and would expose the permanent magnets to strong heat radiation load. This program was cancelled as a result of the damage of the JYFL plasma chamber caused by the sputtering experiments. The sputtering work is described in section dedicated for "*Research and development of sputtering technique.*"

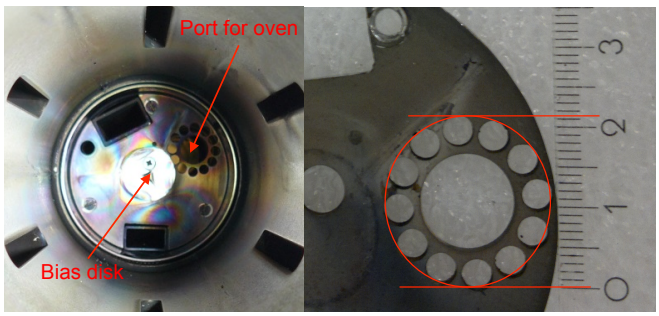

Figure 4: Injection side of the JYFL 14 GHz ECRIS. The figure shows the bias disk, waveguides and oven port. The oven, 20 mm in diameter, is typically placed behind the plate highlighted in the right-hand picture.

Standard oven at GANIL: The SPIRAL 2 program sets very high requirements regarding the production of highly charged metal ion beams. As an example, the beams of $^{48}Ca^{16+}$ and $^{58}Ni^{19+}$ are required with the unprecedented beam intensities (≈ 280 µA and 160 µA for Ca^{16+} and Ni^{19+} ion beams, respectively). The MIVOC method is usually the best option for the production of Ni ion beams but the evaporation oven was found to be more efficient in the case of the highest charge states like $^{58}Ni^{19+}$. During the program strong attention was paid to define the most suitable operation parameters. The experiments were performed using STO for evaporation (standard oven) and the geometry of the experimental setup shown in Figure 5. As an example Table 1 shows the optimization result for the $^{40}Ca^{14+}$ and $^{40}Ca^{16+}$ ion beams. As is shown by the table the nitrogen gas mixing turned out to be the most efficient for the production of highly charged calcium ion beams.

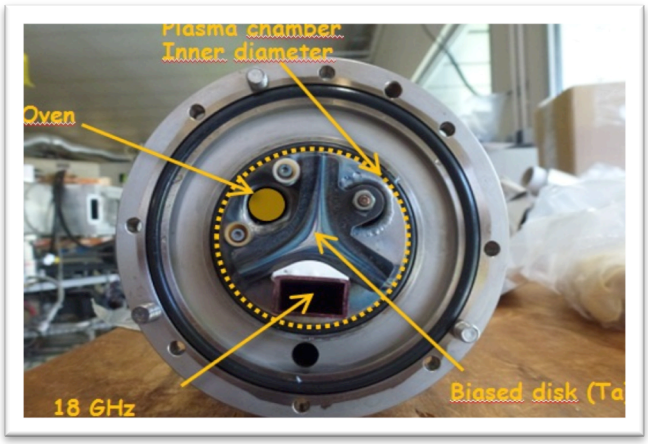

Figure 5: The injection side of the Phoenix-V2. The figure shows the location of the oven, waveguide, bias disk and plasma flux area.

During the experiments two interesting phenomena was observed: RF heating of the evaporation oven and getter effects both negatively affecting the production of highly charged calcium ion beams. Typically the charge state distribution improves as a function of RF power (microwave power) used for the plasma heating. For example in the case of $^{58}Ni^{19+}$ ion beam the power of 1.7 kW was used to get the beam intensity of about 20 µA without observing the saturation. However, in the case of calcium ion beam production the evaporation of material increases although the gas feeding rate and the power used for the oven heating is kept constant. This indicates that the oven temperature is increased via other heating channel: via direct RF heating or via plasma flux. Same effect has been observed when the gas flow is increased while the other source parameters haven been kept constant. Despite of a very low electrical power of the oven, the Ca evaporation becomes too high above the microwave power of 560 W (0.9 kW/L) and leads to a degraded charge state distribution. Therefore, to further increase the Ca^{16+} ion beam intensity (i.e. increase the RF power), a technical solution has to be found to reduce interaction between the oven and the plasma/RF. Research and development work focusing on low temperature ovens could help to reach this goal.

Table 1: Results of mixing gas studies.

	He support gas		O_2 support gas		N_2 mixing gas	
	I [µA]	Stability	I [µA]	Stability	I [µA]	Stability
$^{40}Ca^{14+}$	10	Stable with a fine tuning of He	20	Not stable	35	Stable
$^{40}Ca^{16+}$	1		6		16	

Using oxygen buffer gas an intensity of 6 µA for $^{40}Ca^{16+}$ has been obtained, but the beam was very unstable. A strong getter effect was observed leading in some cases to a complete disappearance of the O_2 mixing gas. The use of the nitrogen mixing gas shaded off the getter effect observed with O_2. It was possible to keep the beam stable over 1 day with an intensity of 16 µA. The chemical getter effect was much less pronounced in the case of nickel and consequently oxygen-mixing gas could be used without affecting the beam stability. Moreover, due to low vapor pressure of nickel, the RF could be increased to the maximum power available leading to a record intensity of 19 µA for charge state Ni^{19+}

High Temperature oven (HTO) at GSI: Contrary to the STO the design of the HTO avoids any ceramics in the hot volume. Instead it utilizes a free heater helix supported only at its ends. All parts in the hot volume are made from W or W compounds. This circumvents the specific problems arising from the use of ceramics and so the application of the high temperature oven turned out to be the most promising approach. In order to optimize the HTO for routine operation its modular construction was improved in mechanical dimensional accuracy for more precise and easier mounting. The use of a special W compound containing 2% of La_2O_3 for crucibles and for furnace facilitates their ductility and machinability. However, its drawback is the evaporation of La out of the metal matrix at very high temperatures, which implies the use of W crucibles under such conditions.

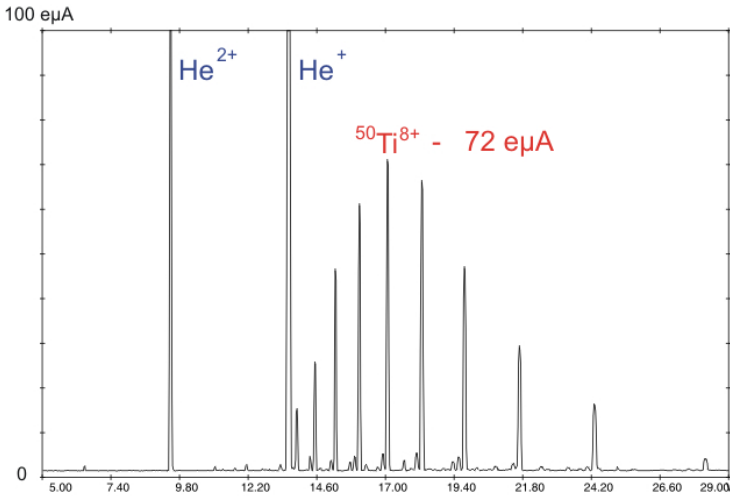

Figure 6: Mass/charge spectrum of natural ^{50}Ti produced at GSI using HTO.

Various experiments were performed with the HTO using natural Ti for most runs. For final confirmation, however, ^{50}Ti sample material had to be used. In addition the evaporation of TiO_2 was investigated, which avoids the problems related to the chemical reactivity of pure Ti at high temperatures. According to the required vapour pressure for the evaporation of TiO_2 the temperature limit of the high temperature oven could be successfully extended to 2300 °C corresponding to 560 W of electrical heating power. However, this high operating temperature reduces the lifetime of the oven. Therefore it appeared to be more favourable to use pure titanium as sample material at evaporation temperatures between 1750 °C and 1800 °C. Figure 6 shows the high intensity ^{50}Ti ion beam spectrum produced at GSI using the high temperature oven (HTO) developed during the program.

Oven for the production of Mo beams: For a stable evaporation of molybdenum in vacuum a temperature of 2469°C would be necessary being too high value for any resistive oven. Luckily, molybdenum is the only metal forming a volatile oxide, MoO_3 reaching the vapour pressure value of 1 mbar at 800°C. This element was tested first time in January 2013 by using MoO_3 with natural isotopic Mo abundances: ^{92}Mo (14.84 %), ^{94}Mo (9.25 %), ^{95}Mo (15.92 %), ^{96}Mo (16.68 %), ^{97}Mo (9.55 %) and ^{98}Mo (24.13 %) natural abundances shown in the parenthesis. A problem that immediately arose was the resolution of the bending dipole after the LEGIS source (LEGnaro ecrIS) - it was not able to separate the different isotopes of heavy elements of requested charge states. This problem was solved by using enriched $^{92}MoO_3$ compound. During the Mo experiment the LEGIS source was operated with the oxygen plasma. The tuning of the source and oven were difficult due to the fact that three oxygen atoms enter the plasma per each molybdenum atom changing continuously the equilibrium of the plasma and resulting in a less stable beam with respect to the other metal ion beams. Figure 7 shows the ^{92}Mo spectrum when the microwave power of 300 W was used. The Pantechnik oven was loaded with 150mg of $^{92}MoO_3$ enriched to 95%. The oven heating current of only 0.8 A was used (3.7W).

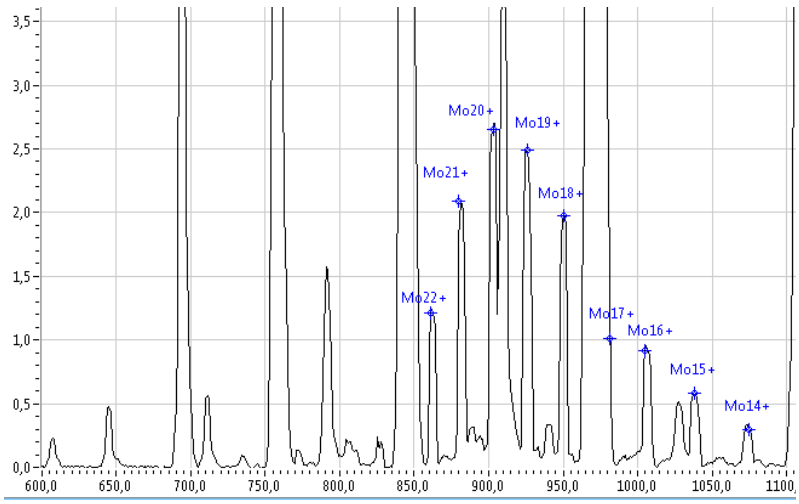

Figure 7: Charge state distribution of Molybdenum ions extracted from the LEGIS source.

2. Research and development of sputtering technique

The radial sputter technique has been applied at JYFL for the production of titanium and zirconium ion beams. In this method the sputter sample is inserted through the radial port to the level of plasma chamber surface (see Figure 4). In one experiment the record high Ti^{11+} ion beam intensity of 20 µA was extracted from the JYFL 14 GHz ECRIS. This intensity exceeded the normal level at least by a factor of 2 and was much higher than could be anticipated from the known sputter yields. In this experiment the sample was inserted, presumably, slightly deeper than normally. It was observed that the Ti^{11+} ion beam intensity did not disappear although the sputter voltage was turned OFF. This was a clear indication that the titanium sample was evaporated via resonant or plasma heating. After the experiment it was discovered a remarkable deterioration of source performance and the measurement confirmed the local damage of nearby hexapole permanent magnet. As a result of this the cooled sputter sample presented in Figure 8 was designed. The experiments with the new design confirmed that the abnormally high intensity was partially achieved as a combination of evaporation and sputtering. In this experiment the intensity level was restored to the level of 5-10 µA for both titanium and zirconium. The research and development work utilizing the

combined heating should be continued with a dedicated test stand. The tight research program of each infrastructure set strict boundary conditions for the ion source continuously operated for the research program (limiting their availability).

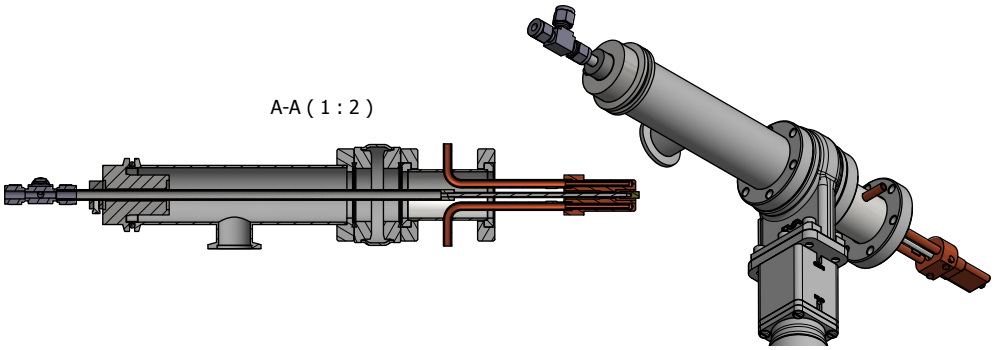

Figure 8: Modified design for the cooled sputter sample.

The research and development work was continued by developing sputter configuration making the axial insertion possible. The sputter sample presented in Figure 9 was inserted using the oven port through the injection plate shown in Figure 4. The structure allowed the movement of 40 mm into the plasma chamber. In this position the distance between the sample and resonance zone (for cold electrons) is about 6 cm. It should be also noted that the rod is situated between the radial magnetic poles – in other words not inside the plasma flux area. The intensities of zirconium ion beams were lower than anticipated: at the position of 40 mm (inside the plasma chamber side) the intensity of Zr^{12+} beam was only 0.5 µA. The explanation can be given by looking at the values presented in Table 2.

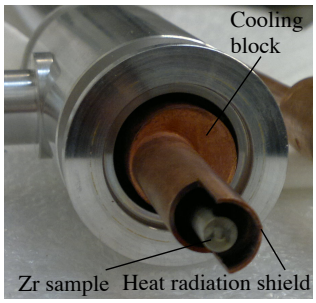

Figure 9: Configuration for axial sputtering. The device was placed into the oven port shown in Figure 4. The read piece indicated by red ring was removed to make the insertion of sample into the plasma chamber possible.

Table 2: Sputter current as a function of position. Zero position corresponds to the level of injection plate presented in Figure 4.

Position [mm]	Sputter voltage [kV]	Sputter current [mA]
-10	3	0.21
-20	3	0.45
-20	4	0.52
-40	4	1.04

Typical sputter current value in the case of radial sputtering is 1-2 mA while the sputter voltage of 1-2 kV is used (depending on the sputtered material and its sputter yield). These sputter voltage/current values typically produce the intensity of around 10 µA. The tendency shown in Table 2 indicates that the sputter sample was too far from the plasma. As a next step the device has to be modified to make longer movement of sample possible. The table also

reveals another interesting detail: the sputter current of 1 mA should cause enough sputtering of the sample in order to reach the intensity level of around 10 μA. Consequently, it can be assumed that the sputter products do not enter the plasma. If it is assumed that the average charge state of plasma is 3 (oxygen mixing gas) this would allow the sputter yield of about 0.6 at the projectile energy of 12 keV (4 kV sputter voltage). As a result of this it can be estimated that the production efficiency of Zr^{12+} ion beam was as low as 0.02 % supporting the assumption that the direction of the sputter products do not support efficient production process.

The axial sputtering was also tested and developed at ATOMKI (Figure 10). In this case the sputter sample was on the axis of the chamber and promising results were obtained. As an example the intensity of 1 μA was produced for the Au^{20+} ion beam. This beam was developed for the implantations needed for medical applications.

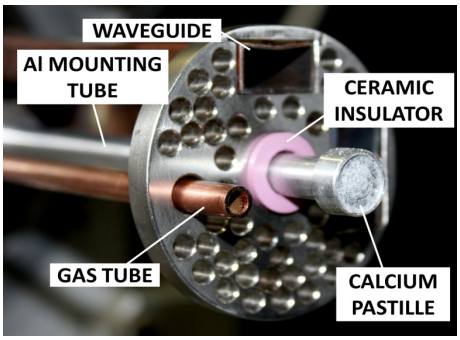

Figure 10: Axial sputtering at ATOMKI.

3. Development of beams produced by the MIVOC method

The JYFL ion source group produced very intensive titanium ion beams in 2002 with the aid of the MIVOC method. Since then, several MIVOC titanium beams have been accelerated worldwide by using a commercially available organometallic titanium compound: trimethyl-pentamethylcyclopentadienyltitanium. Several attempts to synthesize the compound using enriched titanium, especially ^{50}Ti, were carried out before the successful work performed by IPHC-Strasbourg group. As a result,the enriched compound has successfully been used to produce $^{50}Ti^{10+}$ and $^{50}Ti^{11+}$ ion beams for the nuclear physics experiments at JYFL and GANIL. In both cases the stable beam intensity or around 20 μA was maintained for the duration of at least of two weeks. Figure 11 shows the spectrum produced at GANIL.

The total ionization efficiency for titanium was around 10%. Unfortunately, this ionization efficiency could not be compared to the oven method since the titanium ion beams have not been produced yet at GANIL by oven. Nevertheless, it has been done for nickel, and the ionization efficiency is 3-4 times higher with the MIVOC method. It is also worth of mentioning that the used compound is very sensitive to the air and light, and consequently special care has to be taken for its handling. A gloves box under argon atmosphere and an IR light are used during the transfer of compound into the MIVOC chamber.

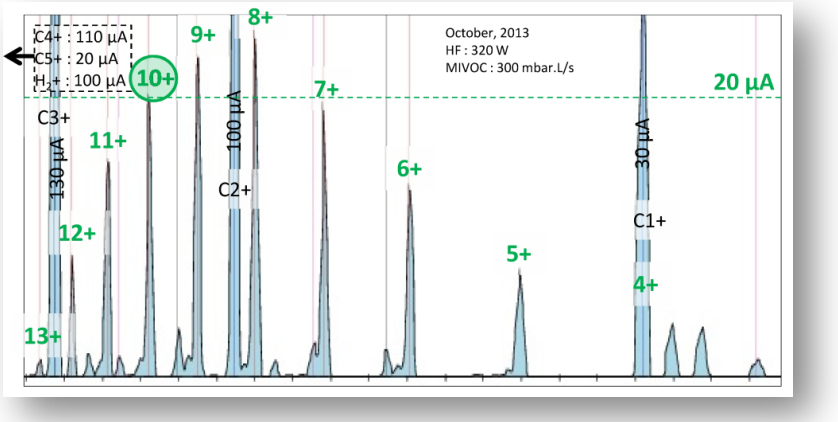

Figure 11: ^{50}Ti spectrum produced at GANIL using the MIVOC method.

4. Production efficiency

The production efficiency experiment and its results have been reported in milestone report "JRA01-ARES-MS84" and its appendix. In this task different experimental conditions and production methods were compared in order to define parameters affecting the production efficiency. Figure 12 shows the production efficiencies of some elements (Ti, Mn and Kr) as a function of charge state. The titanium ion beams were produced with the aid of the MIVOC method and the manganese ion beams were produced by using the JYFL miniature oven. As figure shows the production efficiencies of about 7 % is obtained for the Kr^{16-18+} ion beams. Surprisingly high production efficiency values have been reached with the MIVOC method – about 3 % for the Ti^{9-11+} ion beams. Remarkably lower efficiency is obtained with the evaporation oven. The plasma dynamics and ionization processes regarding the titanium and manganese should be approximately identical, i.e. same ionization efficiency could be achieved for Ti and Mn. This indicates that the main difference in the production efficiency can be linked to the method used to feed the solid element into the plasma and the subsequent capture process of the element. In the case of the MIVOC method the material is not heated and consequently no condensation of the gaseous molecules on the plasma chamber walls takes place before the dissociation of molecules. In the case of the evaporation ovens the atoms, which are not captured by the plasma, will be lost via condensation on the plasma chamber walls. The data presented here strongly indicates that the production efficiencies, regarding the oven methods, can be improved as a result of further development work. The oven geometry should be designed such a way that the evaporated metal atom has a very limited possibility to enter the cold plasma chamber wall. In addition, the method to guide the evaporated elements directly into the plasma should be considered.

Similar tendency was observed as a result of production efficiency experiments performed at GANIL as is presented in Figure 13. As figure shows the ion beam production efficiency associated to the use of the MIVOC method is considerably higher than in the case of the oven. This strongly supports the result obtained at JYFL.

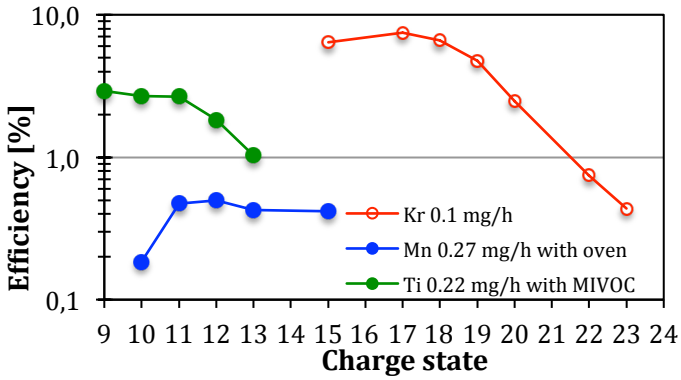

Figure 12: *Production efficiencies of Ti, Mn and Kr ion beams as a function of charge states. Manganese ion beams were produces with the evaporation oven and Ti ion beams by the MIVOC method.*

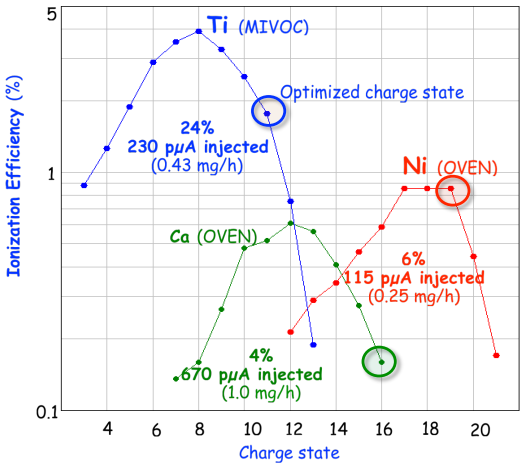

Figure 13: Production efficiencies of Ca, Ti and Ni ion beams.

4. Hot liner experiments

As was described in the previous chapter the production efficiency of ion beams is much lower for elements being solid at room temperature. This efficiency can be ten times less in the case of metal ion beams and is playing an important role in the case of rare and expensive elements like ^{48}Ca. Especially in the case of the ovens, the sticking of the hot vapors on the surface of the plasma chamber leads to high material consumption rates. For elements like Ca, a tantalum liner inserted inside the chamber can be used to limit this phenomenon. The hot liner decreases or even prevents the condensation of element on the plasma chamber walls and consequently makes the recirculation of material possible and thus improves the possibility to extract the element as an ion beam. This effect, earlier developed and tested in Joint Institute for Nuclear Research at Dubna, has been studied at GSI and LNL-INFN.

Temperature of the liner is a relevant parameter affecting the sticking probability of the element. The code COMSOL-multiphysics was used for temperature distribution simulation of the tantalum liner. The overall geometry of ion source with inner tube (liner) from tantalum is taken from the original design of the plasma chamber. The temperature distribution along the tantalum liner as a function of the heating power is presented in Figure 14. According to the graph, the liner temperature of around 950 K is reached using the heating power of 250 W. It is worth of mentioning that the melting and boiling points of calcium are 1115 K and 1757

K, respectively.

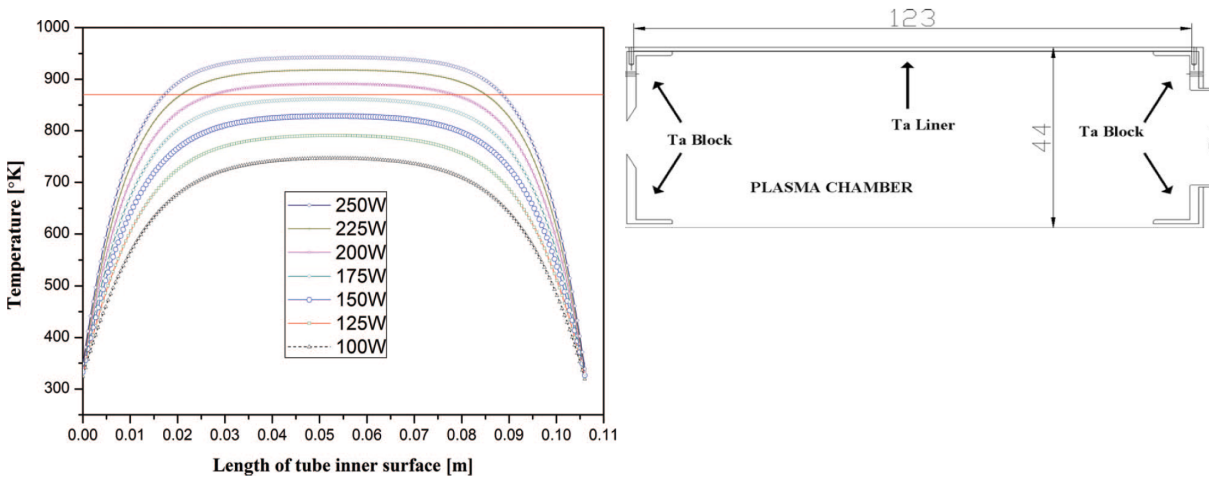

Figure 14: Temperature distribution of tantalum liner as a function of the heating power. Simulations were performed using the LNL-INFN ECRIS (LEGIS) geometry and the liner support structure shown in the figure.

The liner effect for the production efficiency of Ca ion beams was tested at LNL using LEGIS and at GSI using Caprice ECR ion source. At LNL the microwave power of 250 W was used and the source was tuned for Ca^{11+} ion beam (≈3.2 μA). The experiment, which was performed with and without the liner, showed the improvement of 30 % in consumption rate (from 0.23 mg/h to 0.16 mg/h) while the beam intensity was kept unaltered. The afore-mentioned values give the production efficiency of 0.27 % for the Ca^{11+} ion beam. At the same time the production efficiency of 0.4 % was reached for Ca^{9+} ion beam. Similar experiments were performed at GSI using the microwave power of 600 W (Fig. 15). In both cases He was used as a mixing gas. The source was tuned for Ca^{10+} and the production efficiency of 13 % was achieved. This efficiency is similar to the production efficiency of gases and proves the power of the liner method. As a next step its applicability will be tested with much higher charge state (Ca^{16+}) needed for SPIRAL2 operation.

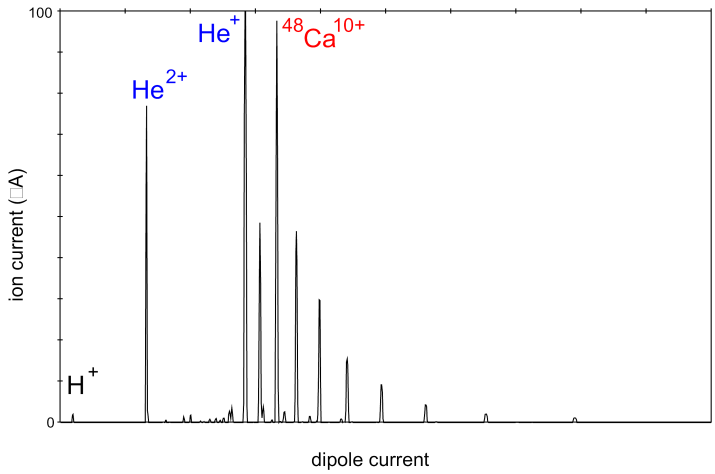

Figure 15: Charge state distribution of ^{48}Ca produced at GSI with Caprice ECRIS.