FP6

CONTRACT N°: RII3-CT-2004-506065

ISSUE CERTIFICATE

EURONS
EUROpean Nuclear Structure research

Activity JRA07 : ISIBHI

Identification: D-J07-5 **Revision: 0**

ISIBHI
Report on metallic ion production

Dissemination level: *PP*
Issued by: *JYFL/JYU*
Reference: *EURONS-D-J07-5*
Status: *Final*

Summary:

In this report the work performed for the production of metal ion beams is described. During the project two different kinds of evaporation ovens were successfully developed to make the evaporation of metals at temperature of about 2000˚C possible. This work increases the variety of metal ion beams available for the nuclear physics program in partner laboratories of EURONS. Additionally, the MIVOC technique was studied in order to improve the ionization efficiency and to minimize the carbon contamination. As a result of the work the carbon contamination can be decreased by an order of magnitude with careful ion source tuning without compromising the intensity of highly charged ions.

31/10/2008	Hannu Koivisto JYFL/JYU	Giovanni Ciavola, INFN-LNS	Alex C. Mueller, CNRS
	Hannu Koivisto	Ciavola	
DATE	RESPONSIBLE Name/Company Signature	WP LEADER Name/Company Signature	COORDINATOR Name/Company Signature

ISIBHI

Report on metallic ion production

1. INTRODUCTION

Task 5 had an important place in the project due to the increased demands for the metal ion beams. For example the ECOS (European Collaboration On Stable ion beams) working group has impressed their concern as follows: "*In order to meet the requirements of the future experiments with high-intensity beams, further development is needed, especially in the production of metal-ion beams.*" In this task both the oven and the MIVOC technique was studied and improved in order to increase the variety and availability of the metal ion beams. Consequently the task was divided into two subsections dealing with the MIVOC method and the evaporation ovens. The development work was started already in the design phase of the ISIBHI/EURONS/FP6 and continued through the whole duration of the project.

2. DEVELOPMENT WORK FOR MIVOC METHOD:

The MIVOC method makes it possible to produce ion beams from some refractory elements without heating. It is very efficient especially for the production of iron and nickel ion beams. The method has actively been used for example at RIKEN where it has been applied for the studies and production of super-heavy elements. The well-known drawback of the MIVOC method is the unwanted carbon contamination, which decreases the intensity of highly charged ion beams. The focus of this research and development work was to study parameters affecting the contamination and cleaning of the plasma chamber. This work was started in the design phase of the EURONS.

2.1. Minimization of carbon contamination

The objective of this study was to find parameters affecting the carbon contamination of the plasma chamber. The surface material of the plasma chamber strongly affects the secondary electron emission, which should be as high as possible in order to maximize the performance of ECRIS. Clean and oxidized aluminum surface is an excellent source of secondary electrons, which coefficient factor can be close to 10, while the factor can be as low as 0.5 for carbon surface. Consequently, it is of great importance to minimize the carbon contamination.

The carbon contamination is certainly affected by the feed rate of the MIVOC compound into the plasma chamber of ECR ion source. However, a certain amount of material has to be fed in to the plasma in order to meet the intensity requirement of the ion beam of interest. With a given material feed rate the contamination can be minimized if the confinement of carbon in the plasma is maximized. This can be done by the gas mixing. In the gas mixing technique the thermal energy of the heavier ion decreases in the collision with the lighter ion (having the same initial thermal energies). The ion cooling mechanism can be effective only if the average ion temperature of the plasma decreases. Therefore lighter ions with lower average charge have to transfer the energy out of the plasma effectively. This would mean that their confinement time in the plasma decreases indicating that their flux to the plasma chamber wall increases. This increases the wall contamination caused by the lighter plasma species, i.e. by carbon in the case of MIVOC compound where the element of interest is the heavier element (for example, $Fe(C_5H_5)_2$, $Ni(C_5H_5)_2$). Here it is assumed that the carbon contamination can be decreased by adding gaseous element into the plasma. The mass of the element has to be approximately in the range of carbon. This decreases the collisions of carbon ions with the heavier plasma species like iron or[1] nickel for example. This would decrease the losses of carbon ions to the wall of the plasma chamber.

The studies were carried out with ferrocene ($Fe(C_5H_5)_2$), which is used for the production of iron ion beams. The consumption of ferrocene was calibrated to correspond to a certain partial pressure in the plasma chamber. Then the JYFL 6.4 GHz ECRIS was tuned for maximum output of a Fe15+ ion beam with a pure ferrocene plasma. The production efficiencies of iron and carbon ion beams were subsequently measured. The measurements were repeated after adding helium, nitrogen or oxygen to the plasma. The optimal ferrocene feed rate for the production of Fe^{15+} ions was found to be 1.7 mg/h for a pure ferrocene plasma and 0.2-0.3 mg/h for gas mixing plasmas. Figure 1 shows the fraction of consumed carbon extracted from the plasma with different mixing gases and mixing gas feed rates.

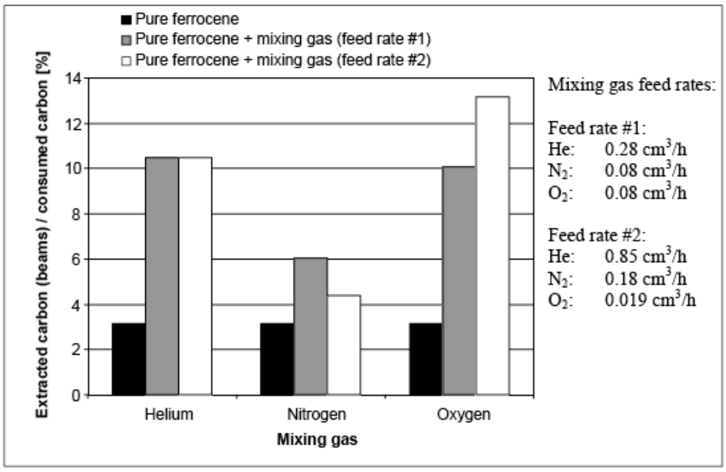

Figure 1: *Fraction of the consumed carbon that was extracted with different mixing gases and mixing gas feed rates.*

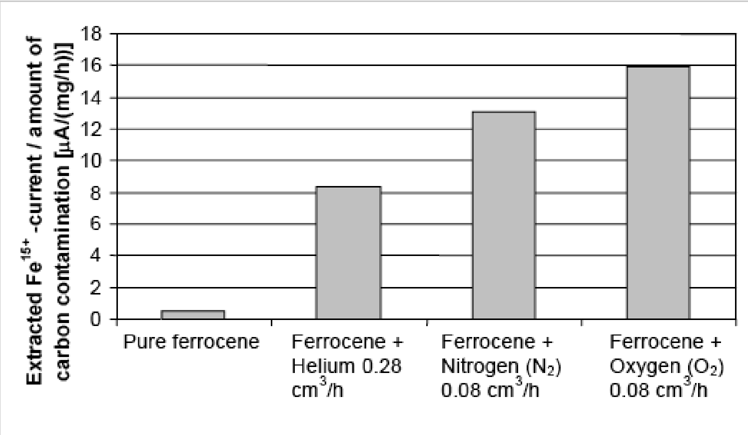

Figure 2: *The extracted Fe^{15+} current compared to the level of accumulated carbon contamination with different mixing gases.*

As figure 1 shows the amount of extracted carbon can be affected significantly with the gas mixing. In the case of oxygen gas mixing the percentage of extracted carbon was fourfold compared to pure ferrocene feed. However, the effect in overall contamination was remarkable because with the aid of gas mixing the same intensity level – or even higher – was reached with order of magnitude smaller feed rate (from 1.7 mg/h to 0.2 mg/h). This effect is demonstrated in figure 2, which shows the extracted current of Fe^{15+} ions compared to the carbon accumulated on the walls of the plasma chamber [µA/(mg/h)] with different mixing gases. The figure shows clearly that the most effective mixing gas for the production of highly charged iron ion beams (with the MIVOC-method) and for minimizing the carbon contamination is oxygen.

According to the observations, low-charge state oxygen and carbon ions can carry the kinetic energy of ions away from the plasma more effectively than the carbon ions alone. This process eventually leads to enhanced confinement of highly charged iron ions. Another explanation for the reduction of carbon contamination with oxygen could be based on chemical reactions. As oxygen is a reactive element it can be expected to form compounds (CO and CO_2) with the carbon accumulated on the plasma chamber walls. These compounds can drift back into the plasma and become ionized again. The probability of extracting carbon as an ion beam increases, which reduces the amount of the contamination. It was observed, for example, that the intensity of the $(CO)^+$ ion beam doubled as oxygen was fed into a ferrocene plasma, which supports this conclusion.

2.2. Removal of carbon

Regardless of minimization of contamination the most of the carbon fed into the plasma will accumulate on the plasma chamber walls (> 80 %). Because of that the parameters affecting the removal of the carbon contamination were studied. For the experiments the carbon contamination was produced with the CO_2 plasma. Approximately the same amount of contamination was produced by keeping the ion source parameters, feeding rate and feeding time identical.

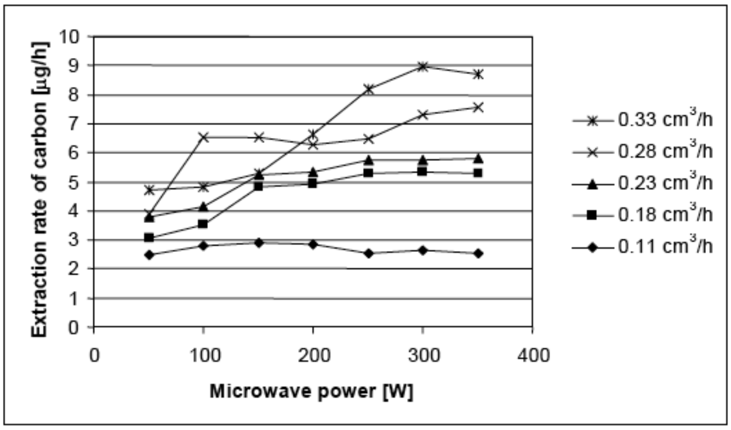

Figure 3: The extraction rate of carbon with nitrogen plasma.

The parameters affecting the cleaning process of the plasma chamber was studied with nitrogen and oxygen plasmas. The extraction rate ($\dot{m}$) of carbon ions was determined by measuring the extracted currents of different carbon ion beams. Figures 3 and 4 show carbon removal rates with nitrogen and oxygen, respectively, as a function of microwave power and gas feed rate. As figures 3 and 4 shows the removal rate of carbon provided by the oxygen plasma is factor of two higher than in the case of nitrogen. However, it has to be noticed that the accumulation rate of carbon is ten times higher than the removal rate.

The distribution of the carbon contamination was also studied. For that purpose a plasma electrode with a thickness of 0.5 mm was constructed. After producing the contamination with a ferrocene plasma the electrode was cut into small pieces for thermal analysis showing the level of carbon at different points on the surface of the electrode. According to the results, the contamination has a well-defined area, which quite precisely follows the magnetic field lines (obviously ions follow the electron flux). Consequently the contamination of plasma chamber can be avoided if the local shielding on the magnetic poles can be used.

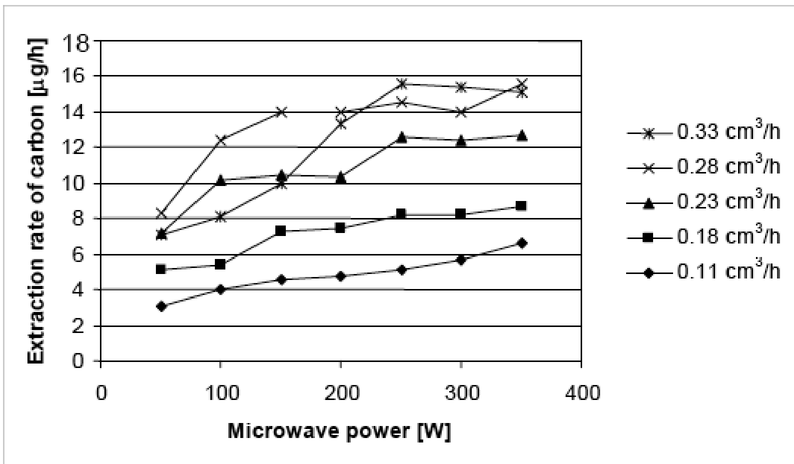

Figure 4: *The extraction rate of carbon with oxygen plasma.*

3. DEVELOPMENT WORK FOR EVAPORATION OVENS

In many cases the evaporation oven is the most appropriate method to produce metal ion beams. It is fairly simple in use and does not cause carbon contamination. However, it has to be noticed that some metallic elements can cause similar contamination effect as carbon. The effect of the most metals on the performance of ECR ion source is unknown and needs to be studied. The main problem relating to evaporation ovens is their maximum operation temperature. In many cases it is limited up to about 1500˚C. The reason for the limitation can be found from the typical structure of the oven, which is shown in figure 5.

Typically the heater of the oven is insulated from the metallic body in order to avoid a short-circuit. Alumina is the most commonly used – and maybe the most suitable - insulating material used with the resistively heated ovens. Its melting point is 2050˚C, which is sufficient for the operation close to the temperature of 2000˚C. However, the limitation comes from its insulating property, which declines steeply between 1000˚C and 1900˚C, being $1 \cdot 10^8\ \Omega$ at 830˚C, $1 \cdot 10^6\ \Omega$ at 1095˚C and 22 Ω at 1875˚C for the sample used as an example. The distance between the heater (typically resistively heated wire) and the insulator has to be adequate in order to keep the temperature of the insulator low enough. This can be done with a careful design or by using induction technique. Both approaches have been studied and developed in task 5. In both cases the objective was to make the reliable oven operation of one week close to 2000˚C possible.

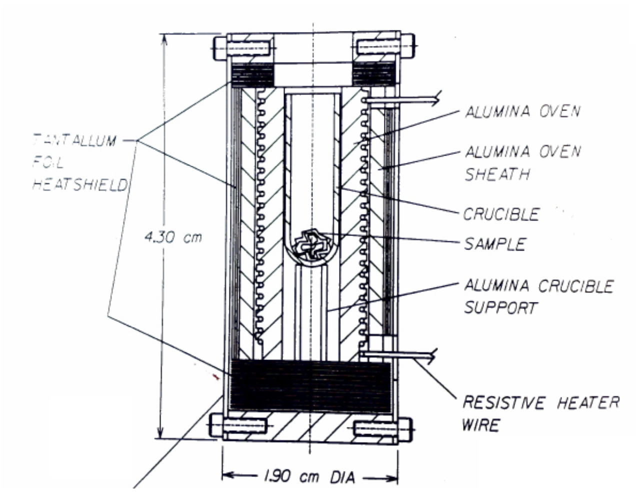

Figure 5: *Typical resistively heated oven (MSU miniature oven).*

3.1. Resistively heated foil oven

Figure 6 shows the principle of the foil oven. The most important parts of the first oven version have been described in figure captions. The current is conducted along the copper rod (12) and crucible stem (5) through a 25 µm thick Ta foil (8). Due to high current density in the foil (up to 10 kA/cm^2) its temperature easily exceeds 2000˚C. The crucible (6), where the material to be evaporated is mainly heated by the heat radiation from the foil. The heat radiation is the main heat transfer mechanism at the temperature close to 2000˚C. The foil was chosen instead of commonly used wire due to its larger area – compared to a wire - and consequently more effective heating. On the other hand the foil requires higher current because of its lower resistance.

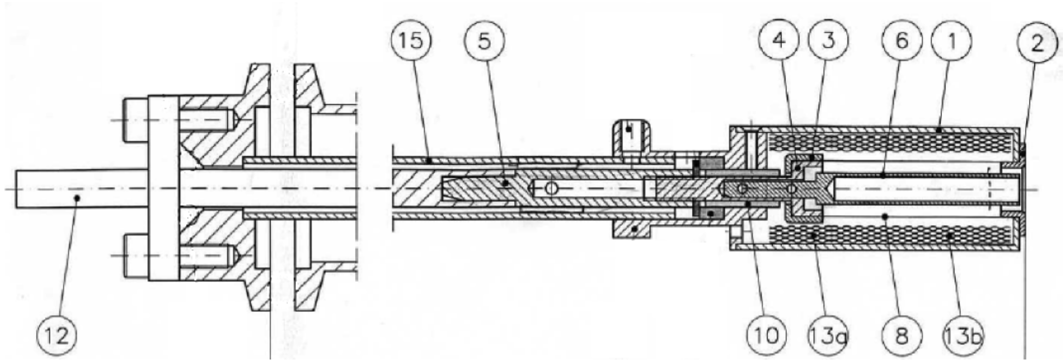

Figure 6: *Resistively heated foil oven: 1) Mo body, 2), 3) and 4) Mo foil holder, 5) crucible stem, 6) crucible, 8) Ta foil, 13) Ta radiation shielding, 10)* Al_2O_3 *insulator, 12) copper rod.*

Figure 6 shows the location of alumina insulator (10). The distance between the heater (foil) and insulator is relatively short and consequently the safety of the insulator was studied by simulations. Figure 7 shows the temperature distribution of the oven when the power of 500 W was fed into the foil. The temperature of the foil and the crucible is about 2000˚K and the temperature of the alumina insulator about 1200˚K. The simulation confirmed that the temperature of the insulator will be low enough to ensure adequate electrical insulating properties.

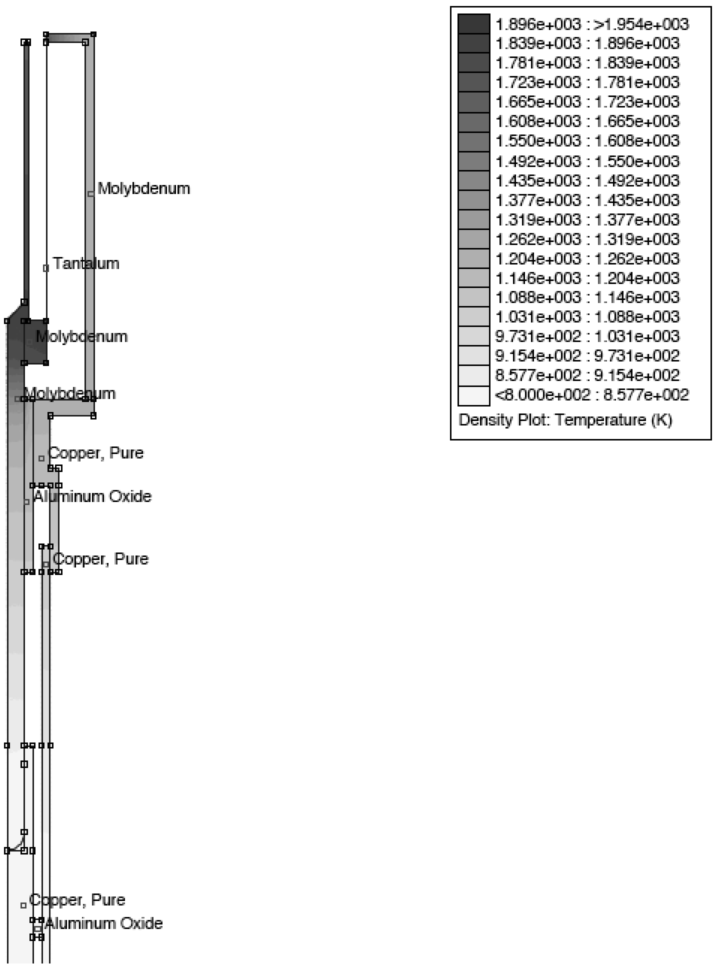

Figure 7: *Temperature distribution of resistively heated foil oven.*

Several modifications to the original structure of the oven were made in order to improve the reliability. The design was fairly complicated, which resulted in different temperature response depending for example on the quality of mechanical connections. Consequently the number of connections was minimized and remaining connections were improved in order to ensure the reproducibility of the operation. The thickness of conducting copper rod (12 in Fig. 6) was optimized to minimize the current and heat losses in the rod (see Fig 8). In simulations both losses, electrical and thermal, were taken into account. Different foil thicknesses have also been studied (18-25 µm). Most of the experiments have been carried out by the thickness of 25 µm, which seems to be a good choice. The main drawback of the oven is that the foil has to be replaced always when the oven is open for the change of the crucible. This certainly can cause temperature variations, and, as a consequence, the temperature of the oven can change even tens of Kelvins because of the foil replacement (see Fig. 9). All temperature calibrations and material evaporation measurements have been performed in the test bench constructed for the oven research and development work.

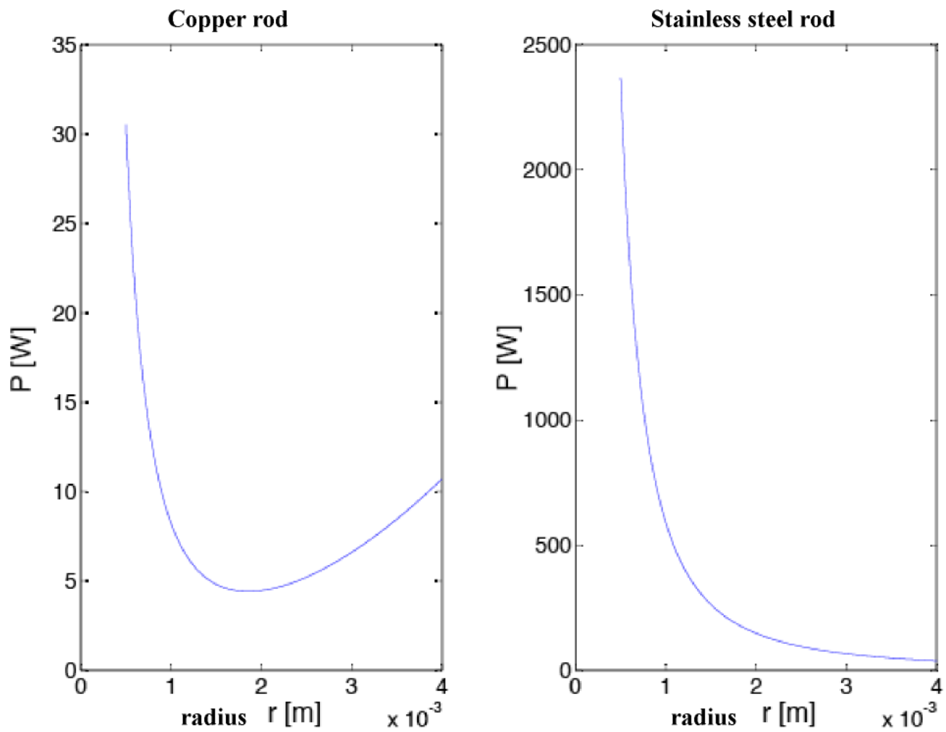

Figure 8: *The effect of current conducting rod on the losses of the oven.*

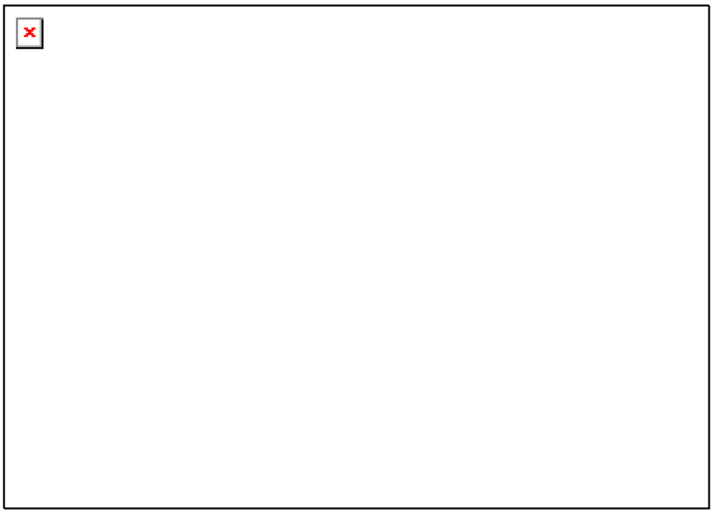

Figure 9: Temperature of oven as a function of power with two different foils (thickness 25 µm).

Figure 10 shows the latest foil oven version. Several durability tests have been performed, where the oven was run close to 2000˚C for about a week without the problems. During one experiment the oven was cooled down couple of times and heated up again. The oven has also been tested up to 2300˚C but only for short period of time (1 - 2 hours). The foil oven has been tested with the ECRIS for the production of Ti, Y and Au ion beams. During the titanium experiment temperature of about 1730˚C was used. The consumption rate of material was 1.9 mg/h, which is enough for the production of intensive ion beams.

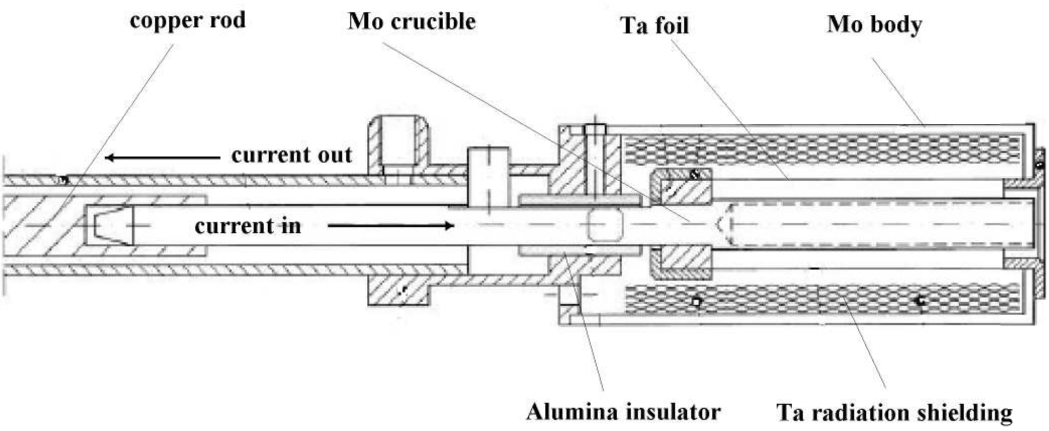

Figure 10: The latest version of the foil oven.

3.2. Inductively heated oven

The induction heating is a very efficient method for the material evaporation. There the energy needed for the heating is transferred by rapidly alternating magnetic field. The oven itself is simple and consists of a water-cooled coil and a Mo crucible as shown in Fig. 11 (please note that the crucible material and its geometry plays an important role together with the coil geometry). Because of that the limiting factor in this structure is the Mo crucible, which melting point is 2620˚C. However, the bottle-neck of the inductively heated oven is a fairly expensive generator, which provides an alternating magnetic field for the heating of the crucible.

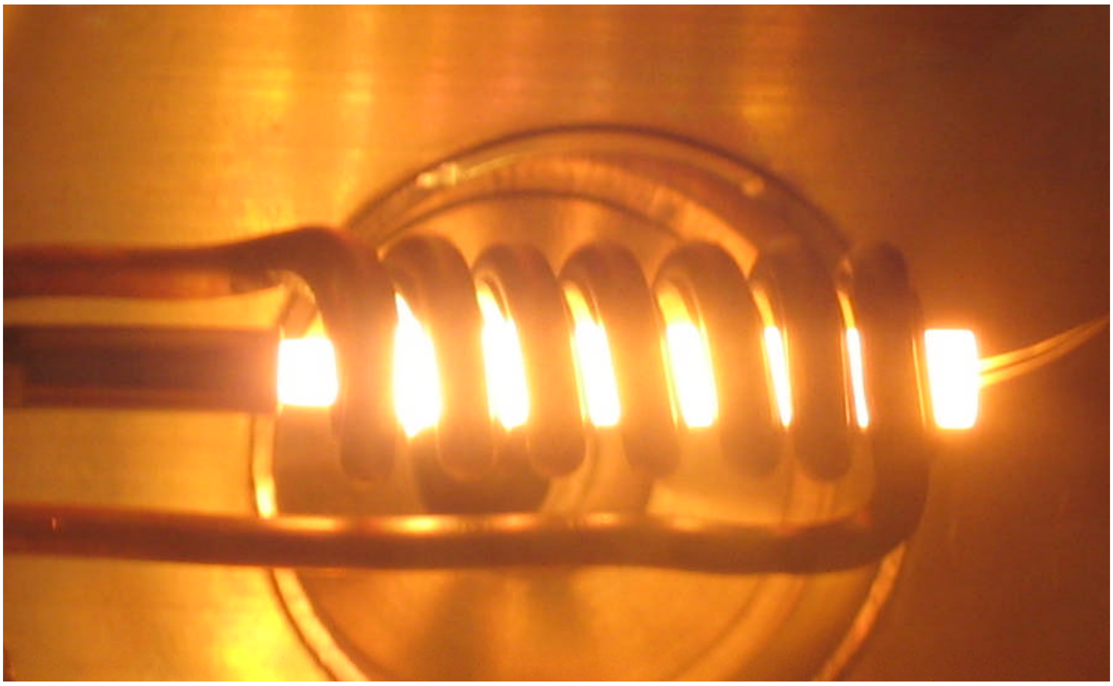

Figure 11: Inductively heated oven at 2000˚C.

In this project a home-made resonant circuit was designed (instead of purchasing a commercial one) and its idea is presented in figure 12. The DC power supply provides the voltage for the resonant circuit (the heater coil shown in Fig. 11 is a part of this circuit), where a transistor plays a crucial role. Originally, when the transistor is closest, the potential difference over the coil is zero. The other side of the coil is connected to the transistor, which is controlled by the signal generator. The signal generator opens and closes the transistor causing the oscillation of current in LC-circuit. The resonance occurs at a frequency of 150-200 kHz. The first version of the resonant circuit gave a pronounced distortion to the signal, which strongly affected the lifetime of the transistor. However, with the version the temperature of 2000˚C was already reached, which encouraged for the further development of the circuit. The life-time problem of transistor was solved with diodes, which unfortunately decreased the heating efficiency.

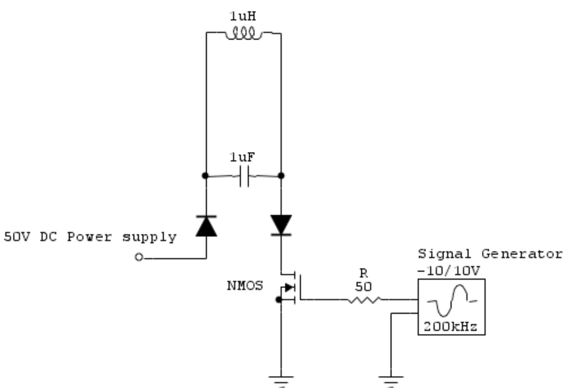

Figure 12: The first version of the resonant circuit of the inductively heated oven.

The second version of the inductively heated oven system was dramatically modified. The transfer line between the coil and capacitor was realized by using coaxial structure. The structure prevented the magnetic field to propagate into the surrounding materials resulting in a remarkably higher heating efficiency. The drawback of the assembly was larger space needed to fit the coaxial line. This problem was solved by changing the polarity of the power supply (negative), which made it possible to ground the outer conductor of the cable. Consequently, no space-consuming insulator was needed. A rectangular control signal is used now instead of sinusoidal in order to minimize the time between the conducting and non-conducting phase of the transistor.

A correct control signal is the main requirement for the proper functioning of the system. The transistor has to be either fully conducting or non-conducting for the optimum operation. Any disturbance in the control signal (which causes a conducting state of the gate at the wrong moment) can cause a destruction of the transistor. Figure 13 shows a control signal (yellow) and gate voltage (blue) of the latest and earlier control unit. The signal of new design (left-side) is good enough to guarantee the reliable operation of the inductively heated oven at 2000˚C. The right-side picture shows a disturbance peak after the switch-off of the control signal. The peak was high enough to open the transistor at the wrong moment resulting in a significant degradation of the transistor life-time. The present layout of the control unit is shown in figure 14.

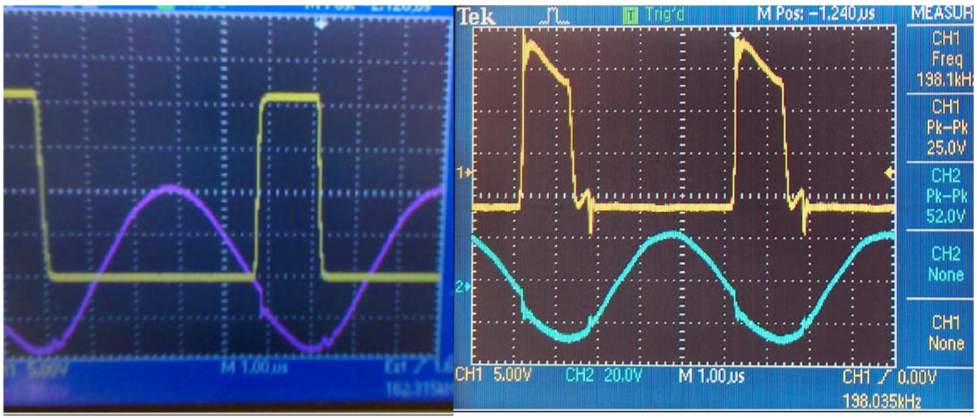

Figure 13: A high quality (left-side) and low quality (right-side) rectangular control signal and collector voltage of the new and the old control unit, respectively.

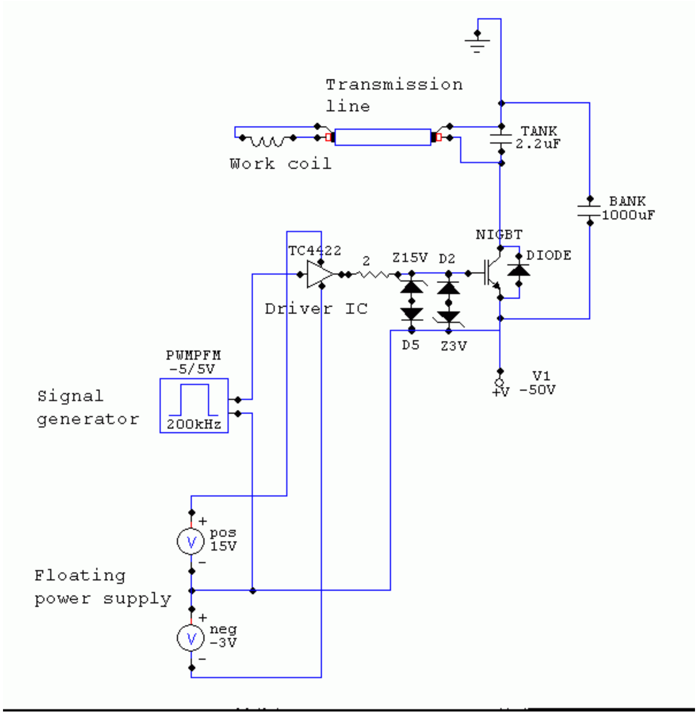

Figure 14: The present layout of the control unit.

The durability tests of the latest version were carried out using the temperature of about 1800˚C. During the period of 9 days no problems were encountered. Temperature slightly increased during the experiment as can be seen from figure 15. However, this change – from 1817˚C to 1831˚C – was only 0.6 %. The result confirmed that no further development for the control unit is needed. Consequently the evaporation experiments were started.

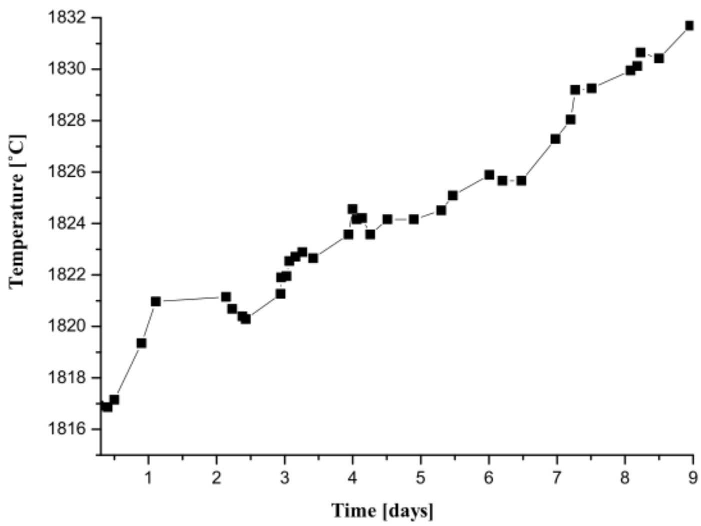

Figure 15: The life-time test with the inductively heated oven. The test was stopped after the run of 9 days without any problems.

Evaporation tests were started with titanium (sample of about 270 mg), which is one of the most requested refractory element for the nuclear physics program of JYFL.

Typical consumption rate for the production of highly charged intensive ion beams is of the order of 1 mg/h or less. As figure 16 shows this consumption rate is reached at temperature of about 1675˚C. According to the measurements the evaporation rate can be dependent on the sample size.

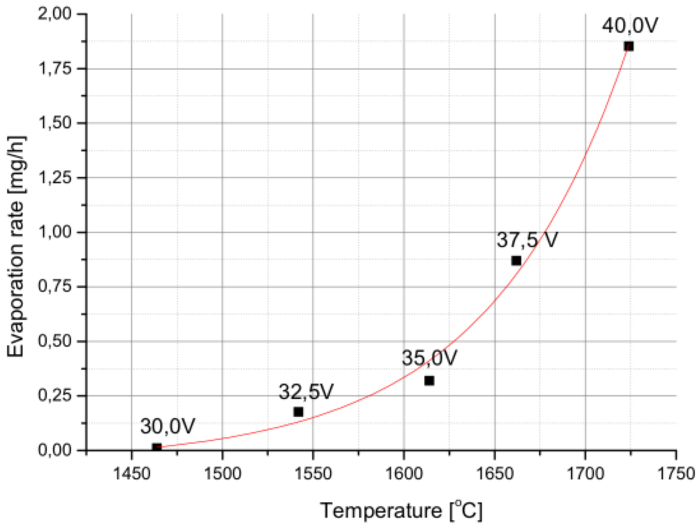

Figure 16: The evaporation rate of titanium as a function of temperature measured with the JYFL induction oven (voltage of the power supply is also shown).

Next the oven was tested with JYFL 14 GHz ECRIS for the production of titanium ion beams. The oven was operated at the temperature of 1700 ˚C, which is needed for the production of Ti ion beam (the oven has been tested up to 2000 ˚C). The operation temperature is remarkably higher than in the case of the miniatyre oven earlier used for the evaporation of metals at JYFL. The experiment confirms that the inductively heated oven makes it possible to produce several new ion beams for the nuclear physics program at JYFL and in other partner laboratories in EURONS. During the experiment the consumption rate was slightly less than 2 mg/h. This was higher than was anticipated from the evaporation experiments indicating that the ionization efficiency in the ECRIS plasma is lower than assumed. Oxygen was used as a buffer gas and clear getter effect was seen during the operation. The intensity level after the ECRIS was about 20 µA for the medium charge states like Ti^{10+}. After the test a clear degradation of ECRIS performance was found. The other experiment was carried out using helium as a buffer gas. In this experiment the same intensities (but more stable) and consumption rates were achieved. In the case of chromium similar intensities were obtained as with titanium. However, the consumption rate was remarkably lower (≈ 0.5 mg/h) and a clear effect in the subsequent performance was not seen.

SUMMARY

During the project a remarkable improvement in the production of metal ion beams was achieved. The parameters affecting the carbon contamination were intensively studied and as a result of the work the contamination level can be decreased by a factor of about 20 (see Fig. 2). With a careful beam time planning (no maximum performance needed in the subsequent experiment) this makes it possible that the ion source can be cleaned with oxygen plasma instead of mechanical cleaning.

In this project two different evaporation oven were developed: 1) resistively heated foil oven and 2) inductively heated oven. Both ovens are capable of operating at about 2000˚C. The induction oven has a home-made resonant circuit, which price is negligible when compared to commercial solutions. These ovens certainly will improve the production of metal ion beams requested by several nuclear physics infrastructures in Europe.