METALLIC ION DEVELOPMENTS AT GANIL

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Radioactive ion beams (RIB) are routinely produced at GANIL by fragmentation of the projectile. A possible way to improve the RIB intensity is to increase the primary beam intensity impinging the target. Although high intensities can be obtained with an ECR ion source for gaseous elements, it is more difficult for metallic elements due to the poor ionization efficiency of the source. This report deals with metallic ion beam production at high intensity. Experimental results for Ca, Ni and Fe are presented. The oven and the MIVOC methods are compared.

I Introduction

The GANIL facility has already been described many times, and details can be found in reference [1]. Two ECR ion sources, called ECR4, are in operation [2]. The stable ion beam produced with the ion source is accelerated up to an energy of 95 MeV/A with a cascade of three cyclotrons. The other source is available for development, but must always be ready for the next run or in case of problem. The accelerated beam is routinely used to produce RIB ion beams by fragmentation of the projectile through a thin target.

In order to increase the RIB intensity, the high energy beam intensity has to be increased [1]. The first step is a 2 kW accelerated beam, i.e. the ECR4 ion source should produce : 40 µA of Mg^{7+}, 70 µA of Ca^{10+}, and 70 µA of Ni^{14+} among others. Let us notice that for the last element, this intensity has never yet been obtained with an ECR ion source !

This report deals with metallic ion beam production at high intensity, with the background motivation to provide a 2 kW accelerated beam. All the tests presented here have been performed with ECR4 and Supershypie [3] ion sources. In view of the nice results obtained at Grenoble for metallic elements with a similar source [4], the ECR4 ion source has been equipped with an aluminum cylinder in the plasma chamber. It was not the case for Supershypie. A schematic drawing of the injection part of the sources is represented in Fig. 1.

The ionization efficiencies given in the following sections do not take into account the transport efficiency of the ions from the source to the faraday cup. Therefore, it must be keep in mind that the ionization efficiencies given in the following sections are in fact the ionization efficiency of the source multiplied by the transport efficiency, i.e. they are underestimated.
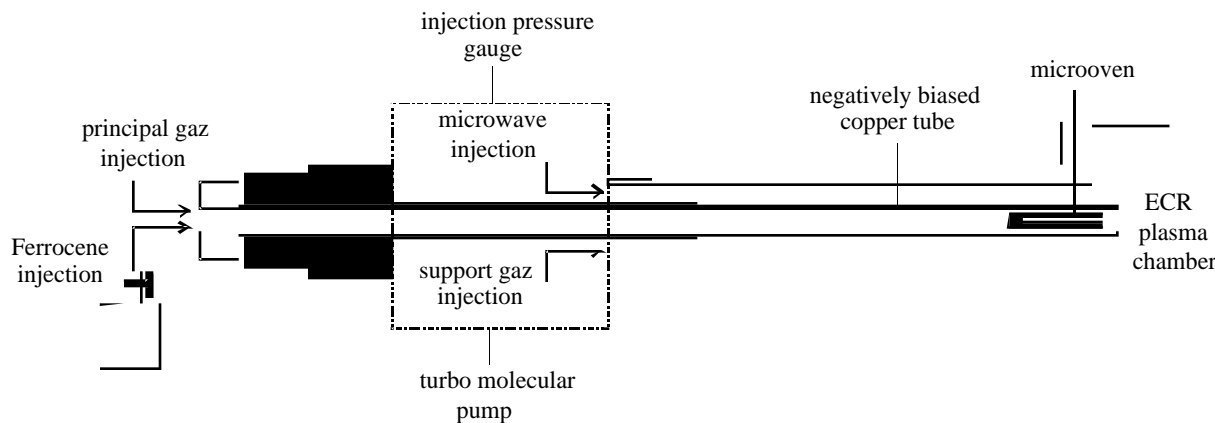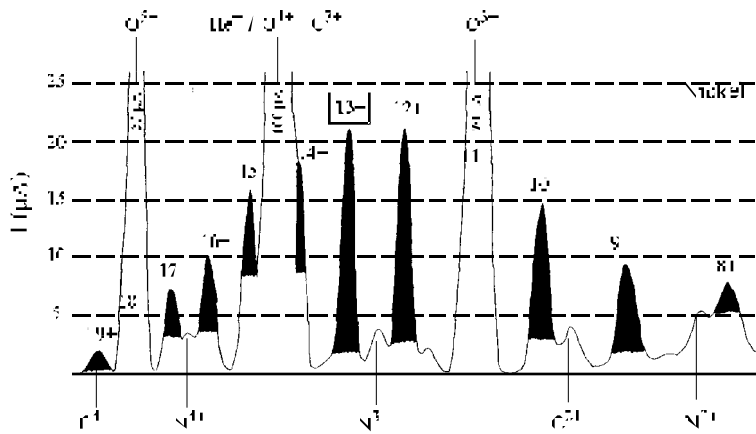

FIG. 1 *Injection part of ECR4 and Supershypie ion sources (microwave frequency 14 GHz).*

II The oven method

Ni^{8+}	7.5 µA	1 pµA
Ni^{9+}	9.5 µA	1.1 pµA
Ni^{10+}	14.5 µA	1.5 pµA
Ni^{11+}	17.5 µA	1.6 pµA
Ni^{12+}	21 µA	1.8 pµA
Ni^{13+}	**21 µA**	**1.6 pµA (13% of total)**
Ni^{14+}	18 µA	1.3 pµA
Ni^{15+}	16 µA	1.0 pµA
Ni^{16+}	10.5 µA	0.7 pµA
Ni^{17+}	7.5 µA	0.4 pµA
Ni^{18+}	4.5 µA	0.2 pµA
Ni^{19+}	1.5 µA	0.1 pµA
total particle current 12 pµA (25 µg/h at the Faraday cup)		

FIG. 2   *The oven method. Best nickel spectrum obtained with ECR4 ion source and optimized on $^{58}Ni^{13+}$. Nickel oxide powder 99% enriched was used. The electrical power of the oven was 30 W (1300°C measured without plasma), oven 1 cm inside the plasma chamber, microwave power 400 W, biased tube −80 V / 0 mA, extraction voltage 15 kV / 1.4 mA, helium support gaz, injection pressure 3 10^{-6} mbar, and extraction pressure 5 10^{-7} mbar.*
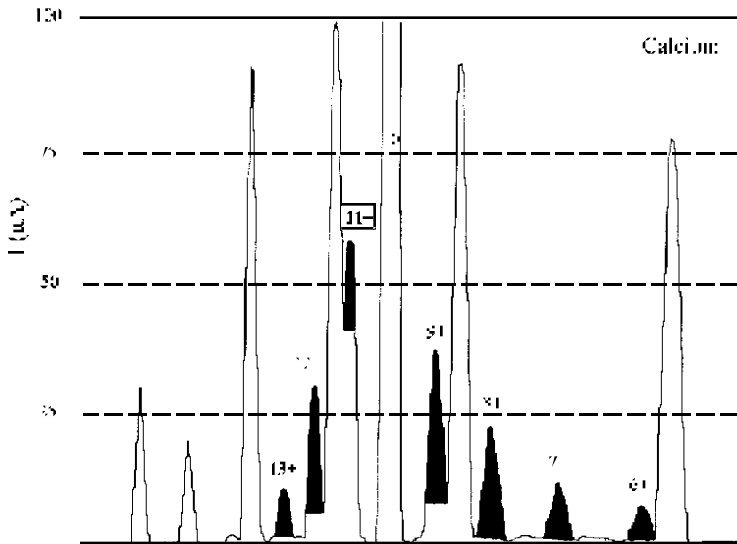
Ca^{6+}	7 µA	1.2 pµA
Ca^{7+}	11 µA	1.6 pµA
Ca^{8+}	22 µA	2.8 pµA
Ca^{9+}	37 µA	4.1 pµA
Ca^{10+}	75 µA	7.5 pµA
Ca^{11+}	**58 µA**	**5.3 pµA (20% of total)**
Ca^{12+}	30 µA	2.5 pµA
Ca^{13+}	10 µA	0.8 pµA
total particle current 26 pµA (39 µg/h at the Faraday cup)		

FIG. 3   *The oven method. Best calcium spectrum obtained with Supershypie ion source and optimized on $^{40}Ca^{11+}$. Natural calcium (97%) was used. The electrical power of the oven was 3 W (350°C measured without plasma), oven 1 cm inside the plasma chamber, microwave power 500 W, biased tube −100 V / 0.76 mA, extraction voltage 18 kV / 2.0 mA, helium and nitrogen support gases, injection pressure 9 10^{-6} mbar, and extraction pressure 5 10^{-7} mbar.*

For metallic elements having a vapor pressure of 10^{-2} mbar at a temperature between 400°C and 1600°C, the oven is usually adopted. The microoven developed at GANIL [5] has an inner volume of $25\ mm^3$, i.e. a capacity load of 220 mg for nickel, 45 mg for magnesium, or 40 mg for calcium....It is routinely used for the production of metallic ion beam at moderate intensity ($I_{q+} < 10\ \mu A$) with a material consumption ranging from 0.1 mg/h to 1 mg/h. When higher intensities are required for low vapor pressure elements, the high temperature must be too much increased, leading to beam instabilities and to definitive damages of the oven. Nickel is a good example. However, surprising results have been obtained with this element.

Nickel oxide has been chosen because it sublimates before melting. Isotopically enriched (99%) powder was used for this test. First, an important contamination of the nickel oxide load by chlorine has been measured in the spectrum. After a few hours, the chlorine completely disappeared and the oven power could be increased. At a power of 31 W, i.e. temperature oven of 1300°C measured off line, a slow increase of the Ni^{13+} intensity up to the surprising value of 21 µA has been observed (c.f. Fig. 2). Then, the intensity slowly decreased to the usual value of 5 µA. The usual consumption rate being 1 mg/h, it gives a very bad ionization efficiency of 0.6% !

The origin of the high nickel beam intensity observed with our microoven, unfortunately during only one hour, has to be understood. It could be due to the physico-chemical characteristics of the nickel oxide powder. Clarifications on this point could lead to significant improvements for high intensity production with the oven method.
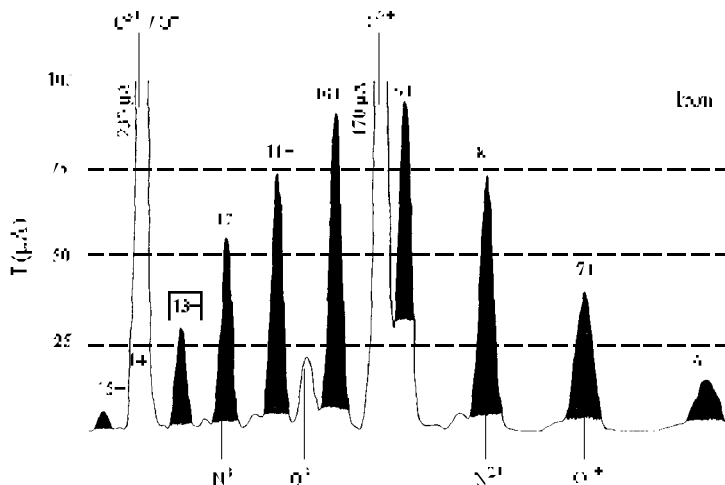
In view of the consumption rates, an oven with a much larger capacity load, like the one developed at GSI [6], is required for ion beam production at high intensity over a long period.

The effect of the oven position on the ionization efficiency has been studied. This short test was performed with calcium on the Supershypie ion source. The best calcium spectrum is shown in Fig. 3.

By changing the position of the oven from z=0 (entrance of the plasma chamber) to z=1cm (1cm inside the plasma chamber), the total ionization efficiency has been improved from 1.6% to 1.9%. The ionization efficiency of the ion source remains poor, and the oven position does not contribute to a radical change of it.

As it was done at Dubna [7], placing a hot cylindrical tantalum sheet inside the plasma chamber can provide a high intensity calcium beam. However, high intensity has been obtained with Supershypie (c.f. Fig. 3) without tantalum sheet. Ionization measurements would give precious information and would validate the real contribution of the tantalum sheet. Tests are planed at GANIL in the next future.

III The MIVOC method

Fe^{6+}	15 µA	2.4 pµA
Fe^{7+}	39 µA	5.6 pµA
Fe^{8+}	72 µA	9.0 pµA
Fe^{9+}	94 µA	10.4 pµA
Fe^{10+}	91 µA	9.1 pµA
Fe^{11+}	73 µA	6.7 pµA
Fe^{12+}	55 µA	4.6 pµA
Fe^{13+}	**29 µA**	**2.2 pµA (4% of total)**
Fe^{14+}	16 µA	1.1 pµA
Fe^{15+}	6 µA	0.4 pµA
total particle current 50 pµA (110 µg/h at the Faraday cup)		

FIG. 4 *The MIVOC method. Best iron spectrum obtained with ECR4 ion source and optimized on $^{56}Fe^{13+}$. The microwave power was 600 W, biased tube −200 V / 0.76 mA, extraction voltage 19 kV / 4.5 mA, no support gaz, injection pressure $2.5\ 10^{-6}$ mbar, and extraction pressure $6\ 10^{-7}$ mbar.*

The advantages of the MIVOC method (Metallic Ions from VOlatile Compounds) have already been highlighted many times by different laboratories [8][9]

some metallic compounds at room temperature : in order of 10^{-3} mbar. A bottle containing the metallic compound is simply connected to the source through a

large conductance regulation valve. Due to the high vapor pressure, a large amount of metallic molecules are pumped by the plasma and therefore, high intensity metallic ion beam can be easily produced. Moreover, for low vapor pressure elements like Mo, preventing the use of an oven, the MIVOC can be used (MoC_5H_5). So why is the oven method still used ?

A major disadvantage of the MIVOC method is the presence of numerous carbon atoms in the molecule : $FeC_{10}H_{10}$, $NiC_{10}H_{10}$, etc...., i.e. one metallic atom for ten carbon atoms ! They contaminate the plasma chamber, resulting in a decrease of the secondary electron emission coefficient [10]. As a consequence, the high charge state production is degraded [11], whereas it is less pronounced in the case of the oven method where pure metallic element can be used. Therefore, the choice of the method is a compromise between the required charge state for acceleration and the desired beam intensity.

The increasing requests for high intensity nickel beam, the impressive results obtained at Riken [9], and the difficulties encountered to produce it with our microoven over a long period, led us to investigate the MIVOC method. Due to the high chemical toxicity of nickelocene preliminary tests have been performed with ferrocene and are now presented.

At atmospheric pressure, ferrocene powder has been placed inside a Pyrex tube and left under a cooker hood for five days. The measured weight after this period was 1 mg (the precision of the balance) lower than the initial weight, i.e. an evaporation rate lower than 8 µg/h. Then, the tube filled with ferrocene has been connected and pumped by the source with the regulation valve full opened. A piranni gauge placed near the ferrocene indicated a pressure of 10^{-2} mbar, and after two hours of pumping an evaporation rate of 18 mg/h has been measured. A dust filter placed at the air evacuation of the primary pump has been weighted after a few hours of functioning : no change in weight has been measured, so that the metallic vapors seem to be condensed in the primary pump, or not trapped by the filter (?). This important point has to be clarified before a routine use of nickelocene.

Despite a rough manual adjustment of the regulation valve, tuning of the ion source was relatively easy and iron has been quickly observed. A maximum of 30 µA Fe^{13+} could be produced after a few days (c.f. Fig. 4).

For a fixed opening of the regulation valve, the total iron current is strongly increased by rising both the microwave power and the biased tube voltage. When the bias voltage was varied from 0 V to –80 V, other parameters being kept constant, the current was increased by more than one order of magnitude for each charge state. This could be explained as a plasma heating contribution leading to a strong re–evaporation of the condensed ferrocene in the biased tube : the vapor pressure of ferrocene is increased by one order of magnitude for a temperature increase of only 20°C.

By increasing the flow of ferrocene with a larger opening of the regulation valve, a saturation of the iron current was observed : the current density in the extraction region becomes too much higher (c.f. Fig. 4), resulting in important beam losses.
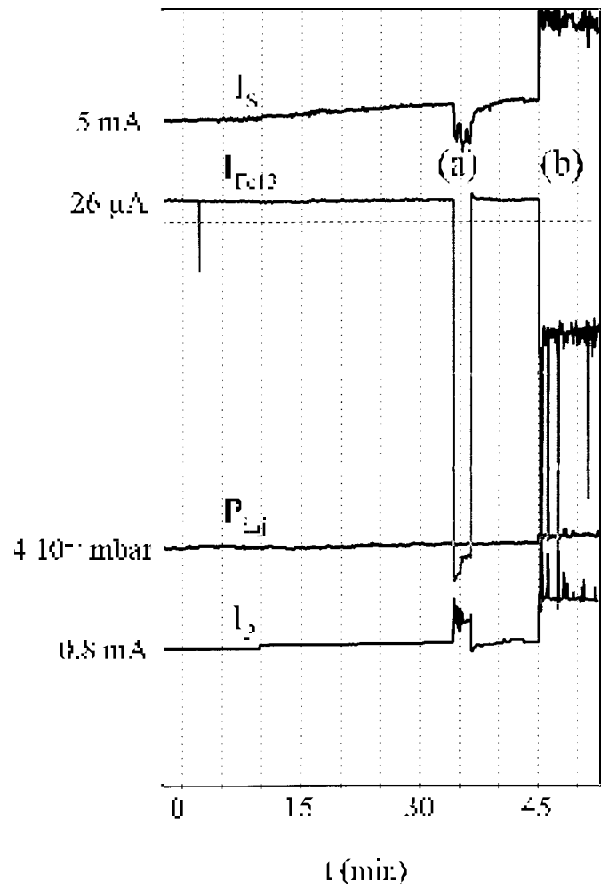

FIG. 5 *Instabilities observed with the MIVOC method without any adjustment of the source parameters. Total extracted current I_S , extracted current I_{Fe13+}, injection pressure p_{inj} , and biased tube current I_p (bias voltage –200 V).*

Unfortunately, we were not able to sustain the high beam intensity more than one hour. These instabilities are represented in Fig. 5.

The beam was very stable for a few tens of minutes, with a notable increase of the total extracted current. Then, the intensity suddenly decreased by a factor 3 ((a) in Fig. 5) : no change of the injection pressure, decrease of the total extracted current, and increase of the biased tube current. After about a few minutes, the parameters were spontaneously restored. An other kind of instability was observed ((b) in Fig. 5) : increase of the injection pressure, increase of the total extracted current, and increase of the biased tube current. Again, spontaneous restoring of the parameters was observed. The origin of these instabilities is not well understood : it could be due to a condensation–evaporation process of the ferrocene in the transfer tube and the plasma chamber (?).

In view of the high intensity and the good charge state produced with this method (almost 100 µA of Fe^{10+}), never obtained with an oven, further investigations are planed in the near future.

IV   Further with metallic ion beam

The nice results obtained at ISN Grenoble with the so-called '' 1+   n+ '' method [12], in particular for metallic elements, could significantly improve the production of some metallic ion beam (Ni) at high intensity. The outstanding result of this method is now briefly summarized.

Let us consider a 1+ ion beam, metallic or not, with an intensity ranging from 0.1 pµA to 1 pµA, and an emittance very closed to 5€ mm.mrad or lower. Injected with the adequate energy into an ECR ion source, Minimafios in the case of this study, the overall ionization efficiency $I(n+) / I(1+)$ reaches 30% for Pb and Rb, 20% for Cr, and 15% for sulphur.

These ionization efficiencies are better than those obtained with the ''neutral '' injection methods like oven, sputtering or metallic gaseous compounds. Let us notice that almost the same ionization efficiency for sulphur is obtained with the neutral method.

Coming back to the results obtained for nickel with the oven method, the very low ionization efficiency of 1% could be significantly improved by the 1+   n+ method. However, preliminary questions must be asked :

(i) what is the ionization efficiency for nickel with the 1+   n+ method ?

If it is better than 1%, the following question is :

(ii) does the MIVOC method for nickel give better ionization efficiency than the 1+   n+ method ?

V   Conclusion

Metallic ion beams at high intensity have been produced with the MIVOC and the oven methods, but it was not possible to maintain the high beam intensity more than one hour : 100 µA of Fe^{10+} and 20 µA of Ni^{13+} respectively. Stability problems have to be solved and stability over a long period has to be studied before a routine use of these beams.

In the case of iron and nickel (similar vapor pressures, q and A), more intensity is obtained with the MIVOC method, but nevertheless on lower charge states compared to the oven method. So that, none of the two methods is the best, except for very low vapor pressure elements where the oven method is not suitable anymore. The choice of the method is a compromise between the required charge state for acceleration and the desired beam intensity.

As the ionization efficiency of the source remains very poor for nickel (1% with the oven method), the use of the 1+   n+ method, characterized by its high ionization efficiency (between 15% and 30%), could be a major improvement for the production of a nickel beam at high intensity. In that way, the ionization efficiency of the MIVOC method for nickel must be measured and compared to the ionization efficiency of the 1+   n+ method for nickel (which has to be measured).

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