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METALLIC ION BEAM DEVELOPMENTS

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Abstract

An overview of the production methods mainly used at GANIL is presented in this report. Experimental observations and recent results about germanium beam production at high intensity are detailed.

1 INTRODUCTION

Intense ion beams from gaseous elements are easily and routinely produced by ECR ion sources. It is not so easy for metallic elements for several reasons :

• *evaporation of the sample :*

most of the time, the metallic element to ionize is under solid form. It must be evaporated into the source plasma. Although high intensities can be produced with the oven method (80 μA Ca^{11+}) [1], it becomes more difficult for low vapor pressure elements. Nickel is a good example : increasing the production of nickel by increasing the oven temperature (typically above 1500°C) would prematurely damage the oven.

⇒ *the method must produce enough metallic atoms injected into the plasma in order to reach the intensity required for the experiment.*

• *ionization efficiency :*

since most of the metallic atoms or ions condense on the cold plasma chamber walls, the evaporation rate has to be higher compared to gas operation : the metallic consumption is typically 10 times higher for a given intensity level. This high consumption is not compatible with the use of rare and expensive isotopes, mostly requested by physicists. The ionization efficiency has to be as high as possible.

⇒ *the ionization efficiency has to be maximized in order to produce the required intensity during the total experiment time while consuming no more than the given amount of rare isotopes.*

• *beam stability :*

The chemical reactivity of metallic atoms condensed on the plasma chamber walls influences the source stability. For example, strong chemical getter effects have been observed when using a mixing gaz, leading to uncontrollable intensity flushes (see section 4).

⇒ *the stability must be good or very good depending on the experiments. At least, it must stable enough to tune the accelerator.*

2 CONFIGURATION

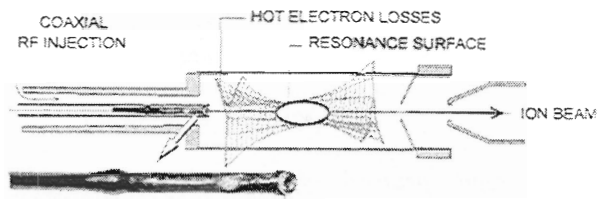


Figure 1 : *Source configuration. The axial RF injection and the biased tube are represented. An axial access is available through the biased tube. The micro-oven developed at GANIL is represented on this drawing.*

Two ECR ion sources are used for the primary beam production at the GANIL facility : the source "ECR4" [2] developed at GANIL, and an improved version "ECR4M" [3] characterized by a stronger magnetic confinement. Both sources work at 14 GHz. The first one is placed on a high voltage plate-form to extract the beam up to 100 kV, and the second one has a 25 kV mono-gap extraction system.

Due to the coaxial RF injection, only one axial access to the plasma is available. This point is of importance for metallic ion beam production : the sample (oven, electrode, etc...) is just located in front of the hot plasma losses and therefore, extra heating of the sample by the plasma itself can not be neglected. Using an oven for example, the source tuning could significantly influence the evaporation rate.

All the results presented in this report have been obtained with this configuration.

3 PRODUCTION METHODS

Only the production methods mainly used at GANIL are presented in this section : the "oven" and the "MIVOC" methods. A complete overview of the methods tested and used at GANIL can be found in reference [4]. An updated list of available beams can be consulted on our web page [5].

3.1 The oven method

This method is well suited to elements having a vapor pressure between 10^{-3} and 10^{-1} mbar for a temperature lower than 1500°C and higher than 300°C. This functioning area is represented in Fig. 2 .

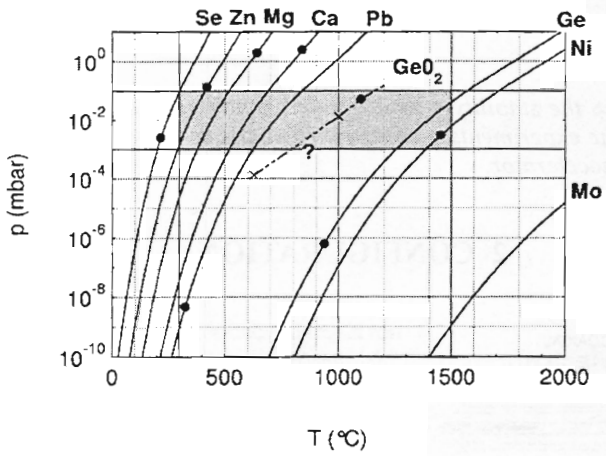
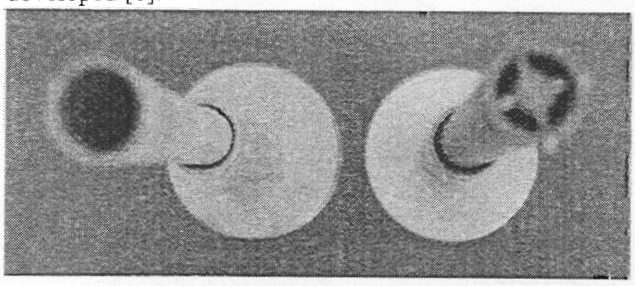


Figure 2 : *Vapor pressure of some elements versus temperature. The melting point is represented by a black dot.*

Above 1500°C the components of the oven (heating wire and crucible) are quickly damaged, decreasing drastically the life time of the oven. As an example, this is the main reason why nickel beam intensity is limited with this technique : high vapor pressure is reached at too high temperatures (see Fig. 2).

Below 300°C the evaporation is dominated by plasma heating, due to the location on axis of the oven. As an example, we were not able to control selenium production in this configuration : selenium immediately evaporates as soon as the plasma is ignited. For selenium, we overcame the difficulty by placing a small disk just in front of the oven (see picture) : it was possible to control a stable intensity of 1.5 μA $^{80}Se^{9+}$ (natural : 50%) during 4 hours and with a consumption around 0.6 mg/h. To increase further the intensity and to eliminate the risk of sur-evaporation, a "low temperature oven" has to be developed [6].



On the left : the coaxial tube , on the right : modified coaxial tube for selenium production (October 2000).

The use of chemical elements melting before reaching the working vapor pressure could be troublesome. The phase change happens within a fraction of a second and is very violent : the liquid instantaneously migrates toward the cold parts of the oven. This has been observed on-line elsewhere using a camera during operation. This violent migration could obstruct the mouth of the oven or eject the metal outside the oven.

It happens sometimes with lead, for no well-known reasons, that the oven empties very quickly. Melting the sample before operation with the oven in vertical position could help to overcome this difficulty.

Oxides can be preferred to pure metals because they melt at higher temperature (GeO_2 for example). Moreover, they sometimes have higher vapor pressure helping evaporation : molybdenum beam has been produced using its oxide form (MoO_3) whereas it would not be possible with pure molybdenum.

Most of the metallic ion beams are produced with the oven technique at GANIL : Mg, Ca, Pb, Sn, etc...[4].

Our micro-oven has been successfully used for the production of calcium 48. The ^{48}Ca beam has been accelerated up to 60 MeV/u with an intensity of 0.13 pμA on target (740 W beam power). Description and results can be found in reference [1].

3.2 The MIVOC method

This method is now routinely used at GANIL for the production of nickel beams at high intensity : a maximum intensity of 70 eμA has been obtained for natural nickel (68% : ^{58}Ni) on charge state 9+ and at 20 kV extraction voltage. The total extracted current from the source was 5 emA.

At this maximum intensity, we observed repetitive sparks, about one spark every second, in the source extraction leading to periodic beam lost during a fraction of a second. Running the source in argon at the same intensity level (5 emA) showed no instability at all. Therefore, these sparks are probably due to the presence of nickelocene molecules in the extraction area. This spark problem must be studied and solved.

The main limitation of the MIVOC method is the high total extracted beam intensity. Thus it seems impossible to further increase the nickel intensity.

More information can be found in references [7][8].

4 RECENT DEVELOPMENTS

4.1 Germanium

This development was initiated by a request for a ^{76}Ge beam at high intensity. First results have been presented at the last ICIS conference at Oakland [1]. They are detailed in this report.

One of the difficulty is the strong chemical reactivity of germanium. We choose the oxide form GeO_2 to reduce the chemical attack of the sensitive parts of the oven (heating wire). The use of pure metal quickly destroys the first turn of the heating wire in direct view of the evaporation. Moreover, the evaporation of GeO_2 form requires a lower temperature (see Fig. 1), and therefore strongly diminishes chemical attack by germanium.

Several tries have been done to find the good procedure and source parameters. The use of helium as mixing gaz improved the stability and the reproducibility of the results.

An intensity of 8.5 eμA $^{74}Ge^{12+}$ natural (36%) has been produced for 48 h. The temperature of the oven had to be progressively increased from 950°C up to 1100°C to maintain this intensity (see Fig. 4). The consumption was 1 mg/h, i.e. 0.25 mg/h ^{74}Ge. The total ionization efficiency was around 5% without transport correction.

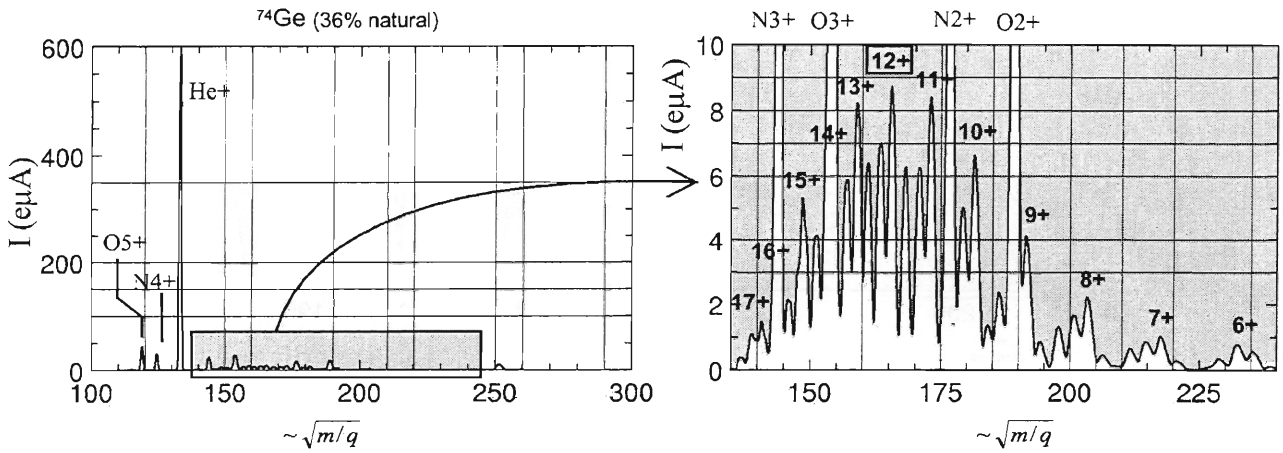


Figure 3 : *Natural germanium spectrum (^{74}Ge : 36%, ^{72}Ge : 28%, ^{70}Ge : 21%) optimized on $^{74}Ge^{12+}$ for stability : an intensity of 8.5 eμA has been maintained for 48 h. Extraction voltage : 71 kV, total extracted current : 1.4 emA, RF power : 115 W, source : ECR4, mixing gaz : He, oven position : +7 mm inside the plasma chamber, electrical power of the oven : 15 W (~1000°C), date : March 13th, 2002.*

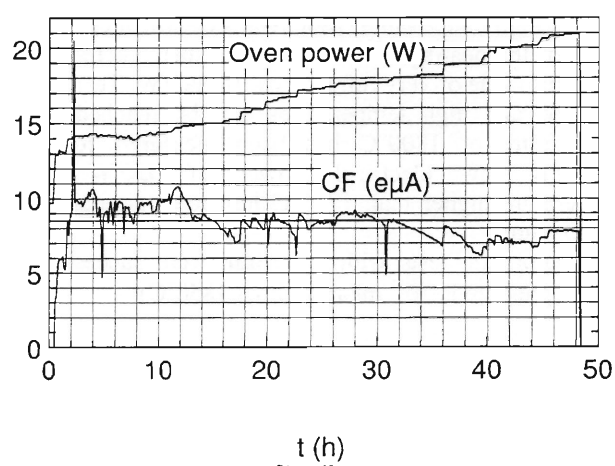


Figure 4 : *Stability of $^{74}Ge^{12+}$ beam for 2 days. The temperature oven was increased from 950°C up to 1100°C to maintain the beam intensity.*

Some interesting behaviours have been observed during and after germanium tests.

• *Chemical getter effect :*

When producing germanium at higher intensity we have observed very strong chemical getter effect although inert mixing gaz (He) was used : the high intensity fall down by one order of magnitude within a few minutes. A spectrum made during this regime has shown that almost no helium was present. By decreasing the evaporation rate of germanium it was possible to come back to a stable situation. No solution has been found to overcome this getter effect at very high intensity.

• *Walls coating effect :*

After the metallic tests we run the source with argon to see the source performance after the germanium contamination. It was a surprise to produce 200 eμA of Ar^{9+} and more over with a good stability ! It has never been obtained before so easily. We concluded that it was probably a wall coating effect with GeO_2.

• *Recycling germanium :*

Since source performance were good in argon after the germanium tests, it was decided to keep the same plasma chamber for the next scheduled beams. It was a big mistake ! First beam for physics (Ne) has been produced without problem. The next planed beam was ^{36}S using $^{36}SF_6$. It was impossible to reach the expected intensity of 20 eμA for $^{36}S^{10+}$: 10 eμA could be obtained with difficulty. Looking at the spectrum (see Fig. 5), we observed that an important amount of natural germanium (12 eμA of $^{74}Ge^{12+}$!) was present in the plasma. Consequently, this presence of germanium degraded so much the charge states distribution of suffer that it was impossible to produce the required intensity on the high charge state 10+. The plasma chamber had to be replaced by a new one. Then, after outgazing the source, it was easy to get 20 eμA of $^{36}S^{10+}$ and we could give the beam back to the physicists. We explained the presence of germanium in the plasma by a chemical reaction between germanium condensed on the wall and fluorine, producing GeF_2 molecules. The compound GeF_2 having a high vapour pressure, it can be re-evaporated into the plasma.

Although this unexpected experiment reduced the beam time for physics, we were satisfied to learn that it is possible to recover germanium condensed on the plasma chamber walls by using SF_6 and moreover with a high intensity : 12 eμA of $^{74}Ge^{12+}$, i.e. about 30 eμA Ge^{12+} with 100% isotopic enrichment ! How long ? recycling

efficiency ? remain open questions. This recycling possibility is of interest for very expensive isotopes like ^{76}Ge and in view of the important amount of germanium remaining in the plasma chamber (95%, i.e. 5% ionization efficiency). It has to be further investigated in terms of stability and efficiency.

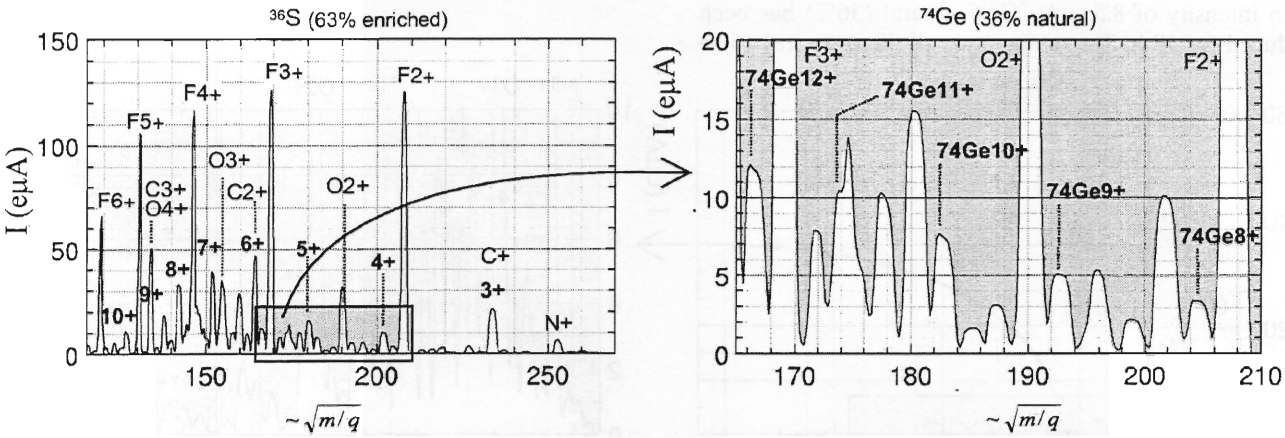


Figure 5 : *Recycling of germanium by using SF_6, RF power : 384 W, extraction voltage : 72 kV, total extracted current : 1.5 emA, source : ECR4, no mixing gaz, date : October 17th 2001.*

4.2 Oven

Two different developments are under way :

• *bigger oven* :

At present, the micro-oven used for metallic ions production is small. The inner diameter of the crucible is 1.5 mm and the length is 30 mm, i.e. about 0.045 cm^3. It allows a maximum load of 70 mg for calcium or 500 mg for lead. With the new oven being developed the crucible volume has been increased to 0.141 cm^3. The maximum load capacity will range from 220 mg up to 1.6 g.

It will increase the functioning time and will facilitate the introduction of samples inside the container.

The evaporation of elements with this oven is being tested off-line.

• *high temperature oven :*

The goal is to increase the temperature up to 2000°C and consequently extend the list of available metallic ions (U for example).

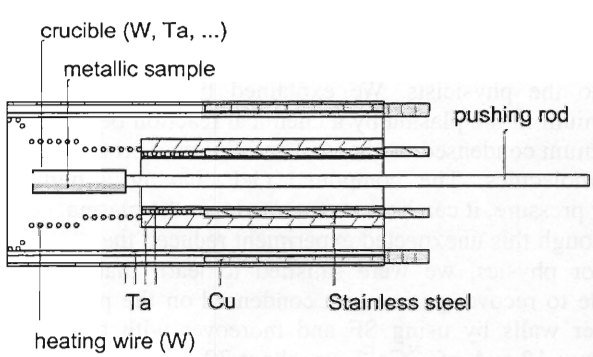


Figure 4 : *Sketch of the high temperature oven.*

The background idea was to remove all Al_2O_3 parts of the oven which are sensitive to very high temperatures. The melting point of Al_2O_3 depends on its quality.

The high temperature oven avoids electrical contact between the heating wire and the crucible. Therefore, crucibles made of very refractory material (W, Ta) can be used and high temperatures can be reached. A temperature of 1800°C has been measured for a relatively low electrical power of 350 W. A temperature higher than 1700°C has been maintained for 50 hours.

Evaporation tests with vanadium were successful (vapor pressure is 10^{-2} mbar at about 1850°C).

5 FURTHER IN METALLIC ION BEAM PRODUCTION ?

The ionization efficiency remains low for metallic ion production : it rarely exceeds 20%. Most of the time, the ionization efficiency is lower than 10%.

It has no importance if the samples are available in abundance, and if it exists a method produce high enough vapor pressure. Nevertheless, for rare and expensive isotopes, the ionization efficiencies have to be improved. Since so many years the metallic ion production methods have been investigated, it seems to show a limitation in terms of ionization efficiency.

The nice results obtained at the ISN Grenoble on the 1+ –> N+ method [9], i.e. 50% ionization efficiency, could be used for the benefit of metallic production. Depending on the existence of a metallic ion source producing mono-charged ions with 100% ionization efficiency, we could inject this 1+ beam into a N+ source, to get an overall ionization efficiency of 50% ! For example : 100 μA of Ni+ will be enough to

reproduce the best result of the MIVOC method. The efficiency of 50% for such 1+ intensity has to be confirmed.

6 NEWS FROM GANIL

• *SPIRAL* :

The safety authorisations to start the SPIRAL installation finally arrived. The first radioactive beam of $^{18}Ne^{4+}$ (1.67 s) has been produced on September 21st 2001 : a 300 W primary beam of ^{20}Ne was sent into the target-source assembly to produce 3 10^6 pps of $^{18}Ne^{4+}$

Until now, $^{8}He^{+}$, $^{8}He^{2+}$, $^{72-77}Kr^{11+}$ have been produced by the SPIRAL facility with a primary beam power up to 1.4 kW.

7 CONCLUSION

Metallic ion production raises new and interesting problems. The chemistry and the ion source configuration are important factors for metallic ion beam production. The development of a new metallic beam requires numerous tries before answering to the feasibility questions : performance, reliability, consumption and stability.

Efforts are still being made to improve the intensity and extend the list of available metallic beams. However, it remains many open questions. The ionization efficiencies are still low and have to be improved : new methods must be investigated.

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