Status report on ECR ion source operation at GANIL

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Two electron cyclotron resonance ion sources, ECR 4 and ECR 4M, provide high charge state beams to the compact cyclotrons, C01 and C02, which are alternative injectors for the GANIL cyclotrons CSS1 and CSS2. When an injector runs for a long period, the off-line source can be used for beam developments or, together with the off-line injector, deliver a beam to a new beam line, called IRRSUD, for atomic physics experiments. Various ions are requested for beam time for periods of 8 to 11 weeks. Although the majority of the required beams comes from gaseous elements, work on the production of beams of metallic ions is always a main activity. New ovens are being developed to improve the capacity and the performances of the standard micro-oven. The latest results with $^{238}$U beam , using sputtering method and $^{76}$Ge beam using recycling method are reported here.
INTRODUCTION

The SPIRAL facility for radioactive ions beams started operation in November 2001, and was at first used only with various beams from gaseous compounds. The SPIRAL complex was run for two-week periods with intense primary beams of $^{13}\text{C}$, $^{20}\text{Ne}$, $^{36}\text{S}$, $^{36}\text{Ar}$, and $^{78}\text{Kr}$ delivered to the target/ion-source system [1]. For gaseous elements up to argon, ECR ion sources deliver enough intensity to reach the final goal of 6 kW beam power at the target/ion-source system. With sulphur and krypton beams we can reach up to 1.4 kW, and 800 W with calcium, nickel and iron beams [2]. The source and the off-line injector can be connected to the new low-energy beam line, called IRRSUD. This facility is used for irradiation studies, and covers a range of energy from 0.5 MeV/u up to 1 MeV/u. The number and the duration of each primary beam accelerated over a two-year period is shown in figure 1. Compared to previous years, the beam time for metallic ion production and development has decreased. However, the upgrading of the metallic ion beam intensities remains a major objective. Beam developments have been successful with $^{76}\text{Ge}$, and improvements of $^{238}\text{U}$ intensity is under way.

STANDARD OVEN DEVELOPMENTS

The standard micro-oven for metallic ion production used at GANIL since 1985 has allowed us to provide various ion beam with a good reliability. However the operational temperature is limited to below 1500°C. First designed for the 10 GHz Caprice source, its dimensions (external diameter 5mm) were matched to the coaxial tube of that source. So the alumina crucible with an inner diameter of 1.5 mm and a length of 30 mm allows a maximum useful volume of only 45 mm$^3$. This is a drawback for the load capacity and the lifetime of the crucible. Recent off-line observations with germanium oxide and lead have shown the formation of “air” pocket in the crucible leading to the expulsion of the material even after careful filling and outgassing of the container. The filling with metallic $^{48}\text{Ca}$, using the
special technique from JINR Dubna [3], is a delicate operation. In addition, owing to the limited evaporation surface, the tuning of the source at high evaporation rates becomes more difficult, particularly when using liquid compounds.

These facts have led us to study a new oven, better adapted to the 14 GHz ECR 4 ion source and which should avoid the earlier drawbacks. It would have a bigger capacity, an aperture able to conduct a high flux rate of vapours, the possibility of safe working with molten material, an operating temperature close to 1600°C and an improvement in the mechanical reliability. First, we had to test the increase in size of the copper coaxial tube, from 12 to 18 mm external diameter. The \(^{40}\text{Ar}^{9+}\) intensity did not decrease by more than 25% compared to the standard performance of 160 e\(\mu\)A. Secondly, a large-capacity oven, based on the same principle as the micro-oven has been built (Fig. 2). The external diameter is 10 mm. A pure tungsten cylindrical wire of 0.3 mm diameter is used as heating filament. The alumina crucible has an internal diameter of 4 mm, a length of 30 mm and the possibility of adding a half cap over the opening. This reduces the output surface to about 9 mm\(^2\), and this could be a protection against hot electrons when running at low temperature. The bigger useful volume of the crucible, 200 mm\(^3\), will increase the working time, will facilitate the introduction of samples inside the container, and will allow safe use of liquid phases. Another objective is to decrease the operating temperature used for some elements, by using a bigger evaporation surface. Increased tin and magnesium beam intensities are expected with this new oven.

A heating power of 170 W, with a heating current of 6.2 A, is required to reach 1600°C, and a temperature higher than 1500°C has been maintained for 5 days. Off-line evaporation with calcium, lead and tin has been made to measure the evaporation flux rate as a function of the heating power and of the surface of the sample. The consumption rates for a stable operation heating power, i.e. a given temperature, are shown in a vapour pressure table (Fig. 3). For a
given element, the initial weights were the same. In the range of 0.1 to 4 mg/h, the crucible geometry did not disturb the evaporation flux according to the vapour pressure tables. A measurement at the same temperature with an initial weight of lead of 90 mg instead of 30 mg increased the rate of consumption by a factor of about 2, which correlates with the increase of the surface area of the spherical molten lead droplet in the crucible. This surface effect, also roughly verified with a calcium sample, should allow us to work at a lower temperature than with the standard micro-oven. First tests on-line with ECR 4M ion source will be done during the fall of 2003.

HIGH TEMPERATURE OVEN DEVELOPMENTS

In 1996, after preliminary experiments for RIBs with a target/ion-source system called SHyPIE [4], a high-temperature oven able to reach 1900°C was designed and built, but never tested. Last year a new demand arose for the production of an intense uranium beam (10 to 20 eµA of U\(^{25+}\)). We therefore began off-line tests with this prototype in order to validate the technical concept. The crucible, made of a refractory material suitable for the required compound, should be movable and could be changed without breaking the vacuum in the source. Clearly it must not affect the performance of the source and its life must be longer than 15 days. To reach this high operating temperature, the use of any ceramic was prohibited, which is a limitation for the actual micro-oven. The high-temperature oven has a coaxial geometry (Fig.4). Two external coaxial tantalum cylinders used as reflectors and a filament holder are fitted together into a copper tube which is also the RF coaxial line. The copper tube (200 mm long on the offline prototype) is welded to a water-cooling flange. The inner part is made of two coaxial tantalum cylinders used as a filament holder and fitted together into a stainless steel tube which allows the introduction of either a movable crucible or gaseous compounds. The tungsten filament has a conical shape and joins the tantalum
cylinders. Its conical shape gives a better mechanical stability and increases the temperature at the mouth of the oven. The crucible diameter could be adjusted up to 5 mm and the length to 15 mm. A temperature of 1800°C has been measured for an electrical power of 310 W (18.3 V and 17 A), in agreement with simulations with the SYSTUS code [5] used as an aid for our modifications. A temperature higher than 1700°C has been maintained for 50 hours. First off-line evaporation tests with vanadium into a tantalum crucible were successful (1 mg/h at 200 W for 20 hours). However, the next tests with melted metallic uranium or solid uranium oxide were not satisfactory. A chemical reaction with 45 mg of melted uranium destroyed the tantalum crucible at around 310 W. Two other ways of manufacturing the uranium crucible are under investigation: tungsten which should be stable with uranium oxide up to 3000°C [6], and yttrium oxide previously used for target tests for RIB production [7]. Taking into account the experiments done at GSI we plan to use more machinable materials, such as tungsten with 1.5% La₂O₃ as crucible material, and VM tungsten as heating wire [8].

A longer version, with 200 mm added to the copper tube, was built for test on-line with ECR 4M source. Despite increasing the DC heating current up to 18A with the maximum magnetic field we do not observe any constraints on the filament. First tests with lead at low temperatures gave the same performance as those obtained with the standard micro-oven and validate the increase of the diameter of the coaxial tube up to 18 mm. However test with vanadium at high temperature failed. Spectrum analysis reveals copper peaks greater than those of vanadium. We noted a change of the appearance of the copper tube close to the tantalum reflectors. Simulations show that the end of the copper tube could reach up to 840°C, i.e. a vapour pressure of 10⁻⁶ mbar. This large and hot surface could explain the copper peaks seen in the spectra which disturb the tuning of the vanadium beam. Modifications have to be done to increase the cooling of the copper tube. Another way is to
replace the final part with a niobium tube which has also good thermal and RF conductivities. Although the mechanical concept of the heating part has been successfully tested, some improvements are necessary to adapt the oven working to the ion source.

**URANIUM BEAM BY SPUTTERING**

Meanwhile a low-intensity uranium beam with a high charge state, i.e. U$^{31+}$, was requested. The sputtering method could be use for that purpose. Previous runs in 1996 using neon as support gas for producing U$^{25+}$ and U$^{28+}$ gave intensities of 2 eμA and 0.8 eμA respectively, but with a high consumption rate, around 7 mg/h [9]. This time the source was tuned for U$^{31+}$ with oxygen as support gas, and could deliver a beam of 0.4 eμA for 6 days, and then 0.3 eμA for 8 days, with excellent stability (<2%). A low consumption rate of 0.33 mg/h was measured. However the ionization efficiency still remained at a low value of about 1%. The main parameters of the source were an RF power of 210 W, an extracted current of 1.1 mA and 1 kV / 0.76 mA as sputtering values. During the second period the required sputtering voltage for operating conditions decreased drastically down to 350 V / 0.7 mA. After the run some short tests were done with SF$_6$ as support gas instead of O$_2$. The charge state distribution in the spectrum then shifted from U$^{28+}$ (800 enA) to U$^{24+}$ (3.5 eμA) but with only 150 W of RF power, and 150 V / 0.5 mA for sputtering parameters (Fig.5). Possibly another process other than sputtering occurred. It could be the formation of UF$_6$ gas by chemical reaction with uranium and dissociated fluorine. The next day, without a uranium sample, the source was tuned with an SF$_6$ plasma. A low recycling effect, giving some hundred of enA of U$^{24+}$, was observed.
GERMANIUM RECYCLING EFFECT

The standard method for germanium beam production uses germanium dioxide (GeO$_2$) in the micro-oven and helium as support gas. We have observed Ge peaks in the spectrum when running with SF$_6$ with a plasma chamber which had been used before for germanium beam [10]. At JAERI, tests with an SF$_6$ plasma have been successful for the production of metallic ions in the MINI-ECR ion source [11]. Therefore, after running for three weeks with a $^{76}$Ge beam, we investigated the recycling effect of an SF$_6$ plasma [12]. An average $^{76}$Ge$^{14+}$ beam intensity of 3 eµA was obtained with a GeO$_2$ consumption rate around 0.4 mg/h, i.e. an overall ionization efficiency of 3% for $^{76}$Ge. At the end of the run, 120 mg of $^{76}$Ge remained on the walls of the plasma chamber. Then without the oven, and with SF$_6$ gas instead of Helium, a germanium beam with a higher intensity is easily obtained. The $^{76}$Ge$^{13+}$ beam remained very stable during two weeks with an intensity increasing from 25 up to 40 eµA (Fig.6). We note that the RF power and the magnetic field were at the same values as used during the run with the micro-oven. Taking into account the initial quantity of 120 mg and the average number of $^{76}$Ge particles extracted from the source (around 50 pµA for 340 hours), we calculate an overall ionisation efficiency of 40%. A germanium tetrafluoride vapour was directly produced in the plasma chamber leading to a high ionisation efficiency.


CAPTIONS

FIG. 1. ECR 4 & ECR 4M ion sources: Primary beams delivered between September 2001 and August 2003.

FIG. 2. GANIL large-capacity oven: 1) half cap (output surface ~ 9mm²) 2) Al₂O₃ crucible (I.D: 4mm) 3) W wire (Ø:0.3 mm) 4) Al₂O₃ insulator 5) Mo connector 6) Ta tube 7) Mo electrical conductor 8) Al₂O₃ insulator 9) stainless steel oven holder

FIG. 3. Off-line evaporation with the large-capacity oven.

FIG. 4. High-temperature oven prototype: 1) copper coaxial tube (18mm O.D.); 2) stainless steel tube (6mm I.D.); 3) & 4) Ta filament holder; 5) & 6) Ta reflectors and filament holder; 7) 0.5 mm W heating wire; 8) Ta cap (8mm diam. hole).

FIG. 5. Uranium CSD optimised for U^{24+} with SF₆

FIG. 6. Recycling germanium CSD optimised for {⁷⁶Ge}^{13+} at 40 eμA
Fig. 1

![Graph showing beam time (days) for different elements like C, O, Ne, Ar, S, O, S, C, Ca, Ni, Ni, Ge, Kr, Kr, Sn, Xe, and Pb.]

Fig. 2

![Diagram showing a cross-section with labels 1 through 9 and a measurement of 30 mm.]
Fig. 3

Temperature

Vapour pressure (mbar)

1.25 mg/h
0.5 mg/h
0.07 mg/h
1.7 mg/h
0.6 mg/h

Sn
Pb
Ca

Fig. 4

φ 20 mm
Fig. 5

**U sputtering with SF6**

- HV: 16 kV
- Current: 1.1 mA
- V.sput.: 140 V
- I.sput.: 0.5 mA
- RF: 150 W

Fig. 6

**76 Ge (88%)**

- High voltage: 78 kV
- Current: 1.1 mA
- Coils: 1073 A
- RF power: 150 W
- Inject. Pr.: 3 x 10^-7 mbar
- Bias: -150 V

**Ge recycling by SF6**

- 15/10/02