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## **Recycling effect of Germanium on ECR Ion Source**

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### ABSTRACT

After running for three weeks with a  $^{76}\text{Ge}$  beam provided by the ECR-4 ion source at GANIL we have investigated the recycling effect of an  $\text{SF}_6$  plasma. The initial beam was produced by the classical method, using germanium dioxide in our micro-oven and helium as support gas. The overall ionization efficiency was measured and found to be around 3%. Without the oven, and using  $\text{SF}_6$  instead of helium, the ECR-4 ion source has been able to produce a very stable beam during a two-week period. The intensity of  $^{76}\text{Ge}^{13+}$  (enrichment 88%) was increased to 40  $\mu\text{A}$ , and the overall ionization efficiency reached 40%.

The oven method of production, the recycling effect and perspectives are described in this paper.

## I. INTRODUCTION

At GANIL most of the ion beams from solid material are provided by the oven method [1]. It takes place on "Caprice" type sources, ECR 4 [2] and ECR 4M [3], with coaxial injection of RF power. They run with a 14 GHz RF transmitter for optimization of medium charge states, typically  $\text{Ca}^{10+}$ ,  $\text{Ge}^{13+}$  or  $\text{Pb}^{25+}$ . The micro-oven is located on the axis of the source inside the copper coaxial tube, also used as the gas feed line. The coaxial tube is biased when running with gases or MIVOC (Metallic Ions from Volatile Compound) compounds, but not yet when using the oven. Our micro-oven works from 300°C to 1500°C to provide the required vapour pressure ( $10^{-3}$  to  $10^{-2}$  mbar) for ease of tuning of the source. However a major part of these vapours is definitively deposited on the water-cooled walls of the plasma chamber. The ionization efficiency with solid materials in an ECRIS is rather low and consequently the intensities are not as high as with gases. A high consumption level can also be a drawback when using expensive isotopes. Constant efforts have therefore been made to increase the ion source efficiency. With gaseous compounds like sulphur hexafluoride, and volatile compounds like nickelocene, or otherwise the micro-oven for solids, our best results are respectively 30% for sulphur, 20% for nickel and 11% for calcium [4], comparable to results obtained in others laboratories [5-6]. Adding a hot screen in the plasma chamber increases significantly the ionization efficiency [7], but only with elements like Ca which do not require more than a few hundred degrees to be evaporated. Nevertheless, most of them have a low vapour pressure and the ionization efficiency stays below 5%.

The method for germanium beam production uses germanium dioxide ( $\text{GeO}_2$ ) in the standard micro-oven. In July 2001 the ECR-4 source ran for three weeks with natural germanium in order to study and improve the reliability. Then, without cleaning the plasma chamber, the source performances were not affected for the production of argon, neon and carbon beams. No peaks containing germanium were observed in spectrum. However, for a following beam the expected intensity (20 eμA of  $^{36}\text{S}^{10+}$ ) using sulphur hexafluoride ( $\text{SF}_6$ ), could not be achieved. The spectrum revealed Ge peaks with an intensity as high as those obtained during the tests with the oven! This was probably a recycling effect due to the  $\text{SF}_6$  gas [1]. The plasma chamber had in fact to be cleaned to produce the sulphur beam at the required intensity. Later a short test was made with  $\text{GeO}_2$  and helium. Fluorine peaks appeared in the spectrum when  $\text{GeO}_2$  began to evaporate and disappeared when the oven heating stopped. A new test with argon did not show any fluorine beam, even with higher RF power. This second observation confirmed the strong affinity between germanium and fluorine in an ECRIS as observed

several years ago in the MINI-ECR ion source at JAERI [8]. These facts led us to study this recycling effect after the germanium run which took place in October 2002.

All tests with germanium have been done on the high-voltage platform, running from 45 kV to 90 kV; it is equipped with the ECR-4 ion source (Fig. 1).

The overall ionization efficiency  $\epsilon_i$  is calculated with the relation below,

$$\epsilon_i = \frac{1}{\epsilon_t} \times \frac{\text{Average ion intensity}}{\text{Ion consumption}}$$

The “average ion intensity” represents the sum of the intensities in the spectrum, corrected with lower charge states, for the ion of interest in particle-microamperes, and taking into account the evolution of intensities during the experiment.

Corrections with lower charge states are calculated from spectra made at lower extraction voltage. At the operational high voltage, 78 kV, the bending magnet does not allow guiding of charge states lower than  $^{76}\text{Ge}^{6+}$  into the Faraday cup. Some spectra have been observed at 55 kV in order to get intensities for lower charge states down to  $^{76}\text{Ge}^{4+}$  or  $^{19}\text{F}^{1+}$ . As the charge state distribution is very similar in both cases, it allows us to correct the particle number of germanium and fluorine at 78 kV and to calculate the transport efficiency more precisely.

“Ion consumption” is the flux of injected particles for the ion of interest, in particle- $\mu\text{A}$ . It is calculated from measured consumption of the compound (e.g.  $\text{GeO}_2$ ) taking into account molecular composition and isotopic enrichment.

$\epsilon_t$  : The transport efficiency of the beam is determined from the sum of intensities for each ion in spectrum, corrected for the lower charge states, divided by the total current delivered by the high-voltage power supply.

In all cases the transport efficiency was better than 75% during the germanium tests.

## II. PRODUCTION OF A GERMANIUM BEAM WITH AN OVEN

The operating temperature with germanium metal (melting point:  $937^\circ\text{C}$ ) in the ECRIS extends from 1250 to  $1400^\circ\text{C}$ . Previous tests have shown that the molten metal reacts with any tantalum or tungsten parts of the oven. We therefore chose to use germanium dioxide, which requires a lower temperature (around  $1000^\circ\text{C}$ ) and diminishes chemical attack by germanium. However some problems occurred during the first runs (melted germanium flowing out of the container, strong instabilities during the starting of the source, failure of the tungsten wire after some days) and this led to poor reliability.

Preparation outside the source can prevent these troubles. The alumina container (internal diameter: 2 mm, length 30 mm) is  $\frac{3}{4}$  filled with about 60mg of white  $\text{GeO}_2$  powder, compressed to avoid air pockets. Then it is heated under vacuum ( $10^{-7}$  mbar) in the micro-oven, increasing step by step (over 2 hours) to  $900^\circ\text{-}1000^\circ\text{C}$  until the recorded pressure shows "spikes" (of up to  $10^{-6}$  mbar). The heating is maintained ( $\sim 2$  hours) until these spikes have disappeared. After breaking the vacuum with argon, the container seems to be empty; the white powder is replaced by a thin vitrified phase along the inner wall of the container. Evaporation measurements under these conditions gives a compound consumption of 0.2, 0.5, and 1 mg/h for temperatures of 1045, 1060, and  $1100^\circ\text{C}$  respectively.

This off-line preparation, turning powder into the vitrified phase, improves the stability and the reproducibility of the beam; it was successfully tested with natural germanium dioxide (producing 8  $\mu\text{A}$  of  $^{74}\text{Ge}^{13+}$ ). The consumption rate reached 1 mg/h and the overall ionization efficiency for  $^{76}\text{Ge}$  was around 10%. Helium was used as the mixing gas.

With this off-line preparation with  $^{76}\text{Ge}$  dioxide, enriched up to 88%, a beam has been provided to physics experiments for a period of three weeks. Optimizing the source required a few hours, and then the oven temperature was progressively increased from  $950^\circ\text{C}$  up to  $1100^\circ\text{C}$  to maintain the required intensity. The lifetime of each container of vitrified germanium dioxide was around 130 hours. The  $\text{GeO}_2$  consumption rates were between 0.3 and 0.5 mg/h for an average  $^{76}\text{Ge}^{14+}$  beam intensity of 3  $\mu\text{A}$  and a background noise around 4%. The overall ionization efficiency for  $^{76}\text{Ge}$ , corrected from transport, was only 3%. These results, lower than those obtained during developments with natural compound, were nevertheless enough for the experiments. A maximum of 300 W beam power, i.e. 70 pA accelerated up to 59 MeV/u, was reached with an intensity of 7.3  $\mu\text{A}$  at the source (Fig. 2). At the end of the run, the total consumption of germanium dioxide was 200 mg. Taking into account the ionization efficiency and the isotopic enrichment, 120mg of  $^{76}\text{Ge}$  remained in the source in either metallic or oxide form.

### III. RECYCLING OF GERMANIUM WITH $\text{SF}_6$ GAS

Without an oven, and even with higher RF power, the  $^{76}\text{Ge}^{13+}$  beam intensity did not exceed some hundreds of enA. Instead of helium, sulphur hexafluoride gas was then injected through the coaxial tube. (The magnetic field was not changed.) With only 45 W RF power, and with a lower quantity of  $\text{SF}_6$  than He, the  $^{76}\text{Ge}^{13+}$  beam immediately began to increase, gradually reaching 1  $\mu\text{A}$  after 30 minutes, and 7  $\mu\text{A}$  after 2 hours. The recycling effect thus seems to be a very promising way to upgrade the intensity. However, how long can it last?

One day later, and without any adjustment, the  $^{76}\text{Ge}^{13+}$  output reached 19  $\mu\text{A}$ . The intensity increases drastically with RF power, but we decided to keep to the same setting, and the intensity remained stable during the next 5 days.

On the 6<sup>th</sup> day the coaxial tube was biased at -150 V, and the  $^{76}\text{Ge}^{13+}$  intensity increased to 31  $\mu\text{A}$ . The source was then very easily tuned to get 40  $\mu\text{A}$  of stable beam for one day by increasing the RF power and the gas flow (Fig. 3).

This first period of tests is indicated in Fig. 4, showing the RF power tuning, the evolution of  $^{76}\text{Ge}^{13+}$  intensity and the total current of germanium particles calculated from some spectra. This current, in  $\mu\text{A}$ , includes corrections with lower charge states and the transport efficiency. The average number of  $^{76}\text{Ge}$  particles extracted from the source is calculated to be around 55  $\mu\text{A}$ . (On the last day, we note that the increase of intensity was mostly due to the biasing of the coaxial tube.)

Usually when we are working with the oven, located just a few millimetres from the end of the coaxial tube, some material can condense onto this tube. In order to know the contribution of the plasma chamber walls in the recycling process, we stopped the source to replace the coaxial tube by a new one. No deposits could be observed on the old tube.

Argon was used for outgassing the source. The RF power, 260 W, and the total extracted current, 1.4 mA, were higher with argon than with an  $\text{SF}_6$  gas feed; however no recycling process occurred. Some hours later we obtained the same beam intensity under exactly the same conditions as before replacing the coaxial tube. The recycling of deposits coming from the coaxial tube is negligible. Then during the ensuing 5 days the intensity decreased gradually down to 26  $\mu\text{A}$ . Increasing the  $\text{SF}_6$  gas flux allowed us to regain a maximum intensity of 40  $\mu\text{A}$  for 3 days (Fig. 5). The spectrum made at 55 kV (Fig. 6) shows the lower charge states and the effective increase of fluorine, even with a lower transport efficiency (62%). Further test is no longer possible because the source was needed for physics experiments. On the last day, no amount of tuning of the source parameters could stop this very slow decrease of intensity. Perhaps this indicated that the recycling effect was nearing its end. In this second period the average number of  $^{76}\text{Ge}$  particles extracted from the source was calculated to be around 45  $\mu\text{A}$  including the transport efficiency.

During all the tests, the increase of the  $\text{SF}_6$  gas flux (from 100 to 300  $\mu\text{A}$  of  $^{19}\text{F}$  measured in the spectrum) seems to be a major parameter for increasing and maintaining the germanium beam production. Magnetic field adjustments allows one to stabilise the beam, while RF power level fine-tunes the  $^{76}\text{Ge}^{13+}$  intensity. The background noise on the  $^{76}\text{Ge}^{13+}$  intensity is lower than 2% and the tuning of the source is as stable as with a gas

beam. Any tuning action definitively disturbs the stability and reliability of the beam. The strong fluorination effect takes place mainly on the walls of the plasma chamber. From the average number of  $^{76}\text{Ge}$  particles extracted from the source around 50 pμA, (i.e. 0.142 mg/h) for 340 hours, we deduce that an amount of 48 mg of  $^{76}\text{Ge}$  was extracted from the source. Since an initial quantity of 120 mg was deposited on the walls, we find an overall ionization efficiency of 40%. This value is a minimum, since the recycling test had to be stopped before its end. Table 1 gives the overall ionization efficiencies from different production methods. The recycling effect with germanium gives results close to efficiencies with noble gases. Note that, in the case of these gases, the pumping in the injection part of the source reduces the efficiency. The injected flux rate of particles may also influence the efficiency, especially with high intensity beams.

#### IV. FLUORINATION PROCESS AND OUTLOOKS

Fluorine coming from dissociated  $\text{SF}_6$  gas in the plasma reacts with the solid Ge or  $\text{GeO}_2$  deposited on the walls. A germanium tetrafluoride ( $\text{GeF}_4$ ) vapour is directly produced in the plasma chamber through this chemical reaction. This compound, which sublimates at  $-36^\circ\text{C}$ , is the only one that could be formed under gaseous conditions at low temperature.  $\text{GeF}_2$  and  $\text{GeS}_2$  have thermal properties which are less favourable than  $\text{GeF}_4$ . Previous experiments using  $\text{GeS}_2$  did not show any significant recycling process. It seems that fluorine reacts gradually with thin layers of germanium or germanium oxide on the walls while the most of the sulphur particles are condensed. The easy tuning of the source, the stability, the very small background noise of the beam, the high intensity and the ionization efficiency confirm that the source behaves as well as with a gas.  $\text{SF}_6$  gas has been used elsewhere for many years for sulphur beam production and no corrosive action occurs.

Another step will be to test the  $\text{GeO}_2$  evaporation from the oven with  $\text{SF}_6$  gas as the mixing gas. An on-line chemical recycling effect could increase the performance and the lifetime of the oven method. However, a fluorination effect with tantalum and tungsten parts of the oven could be a serious drawback.

With germanium, this chemical recycling allows us, to ionize most of the particles previously deposited on the wall of the plasma chamber. It provides higher and more stable intensities than the oven method. It could be very useful when using rare and expensive isotopes that are not easily available in gaseous form. Furthermore, the on-line synthesis avoids the storage and handling of such toxic or corrosive gas as  $\text{SeF}_6$ ,  $\text{GeF}_4$  or  $\text{UF}_6$ .

As fluorine is the most electronegative and reactive of all elements, it reacts with practically all substances. Placing a hot screen inside the plasma chamber could even reinforce the recycling effect. It could allow a recycling effect with other species like Be, Bi, Cd, Cs, Pb, K, U, Rb, Zn, for which fluorides require less than

500°C to supply the right amount of atoms to the ECR plasma. The depositing of thin layers of the element on the screen surface could be done out of the source, using classical evaporation methods. As metallic production with an SF<sub>6</sub> plasma succeeded in the MINI-ECR with B, Mo, Nb, W and Ta [8], it is a promising idea to use a more powerful ECRIS to continue the investigation of this recycling effect for rare isotopes or radioactive ions requiring a very high ionization efficiency. In this last case, since sulphur hexafluoride decomposes at temperatures above 800°C, it could be used as mixing gas to transport condensable elements in gaseous form from the hot target towards the ionization area.



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## FIGURE CAPTIONS

Fig. 1: Cross-sectional view of the ECR-4 ion source installed on the high-voltage platform.

Fig. 2: Germanium spectrum with oven, optimized on  $^{76}\text{Ge}^{14+}$ .

Fig. 3: Recycling germanium spectrum, optimized on  $^{76}\text{Ge}^{13+}$  at 78 kV.

Fig. 4: First period, evolution of the  $^{76}\text{Ge}^{13+}$  beam during 7 days.

Fig. 5: Second period, 8 days, with a new biased coaxial tube.

Fig. 6: Recycling germanium spectrum optimized on  $^{76}\text{Ge}^{13+}$  at 55 kV.

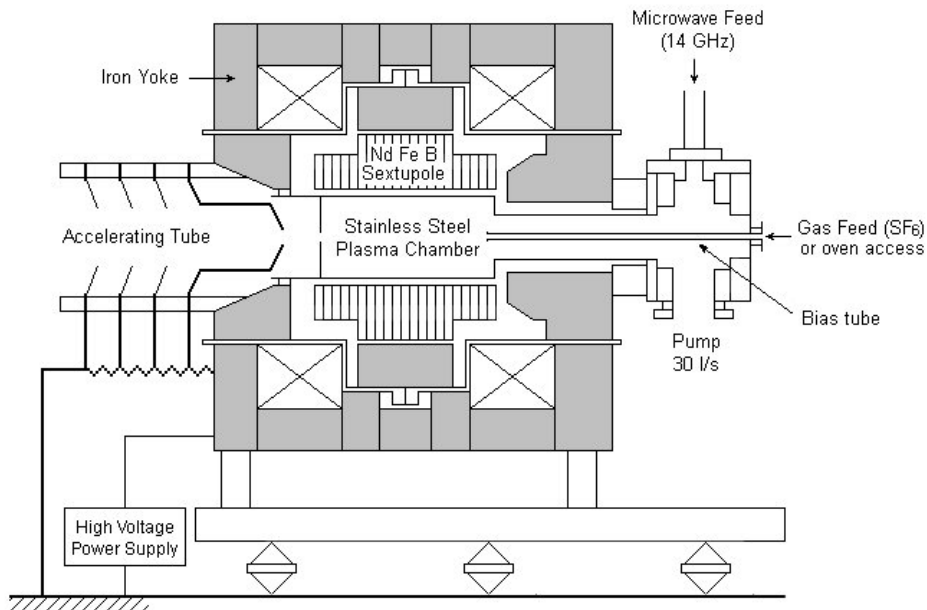


Figure 1

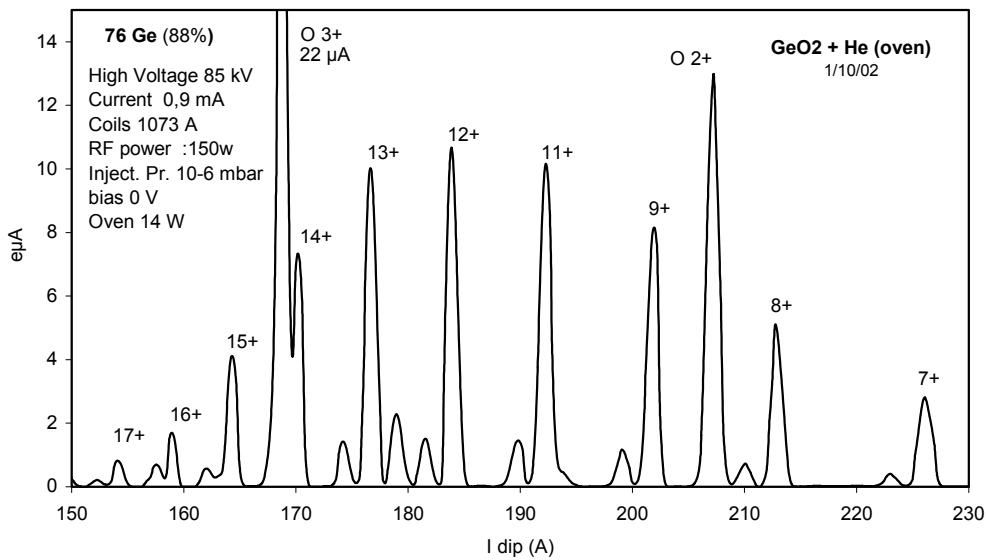


Figure 2

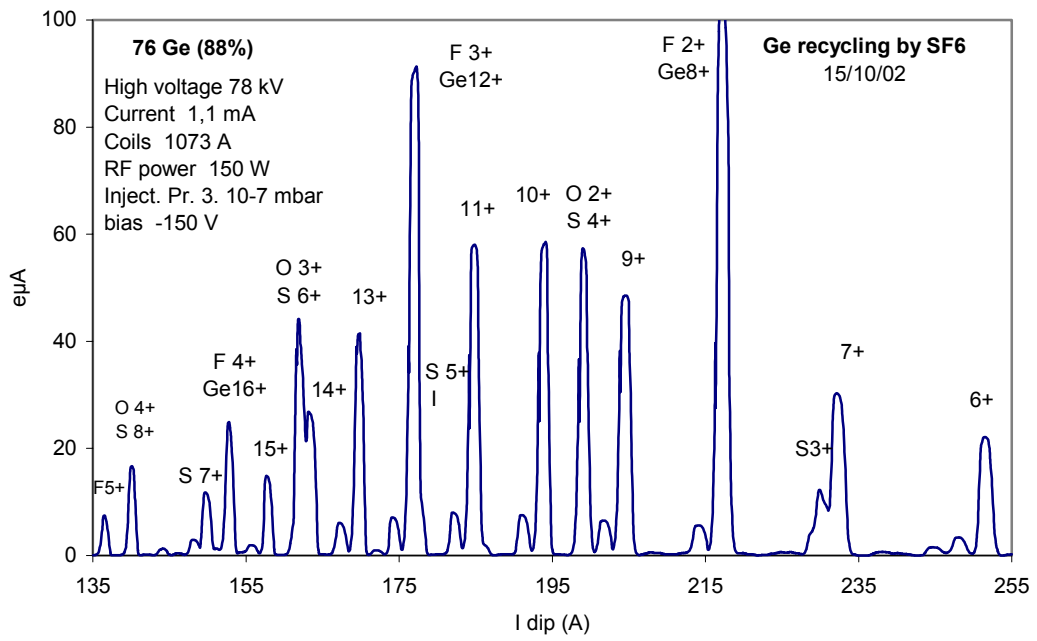


Figure 3

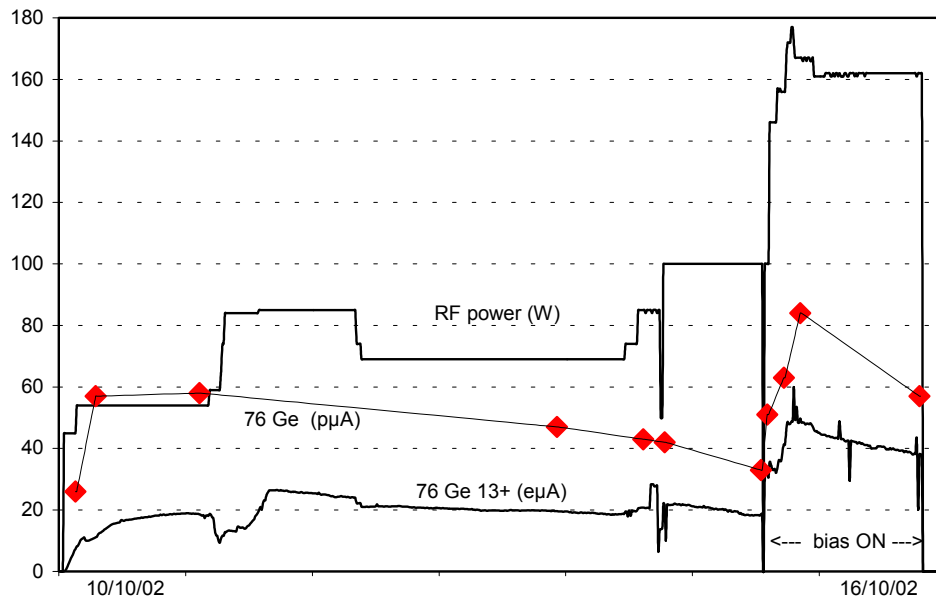


Figure 4

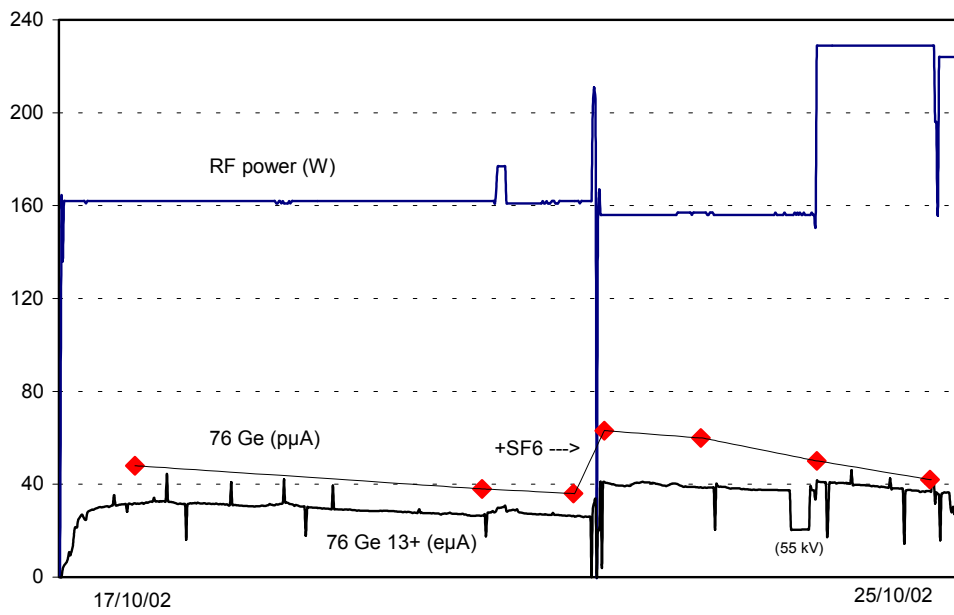


Figure 5

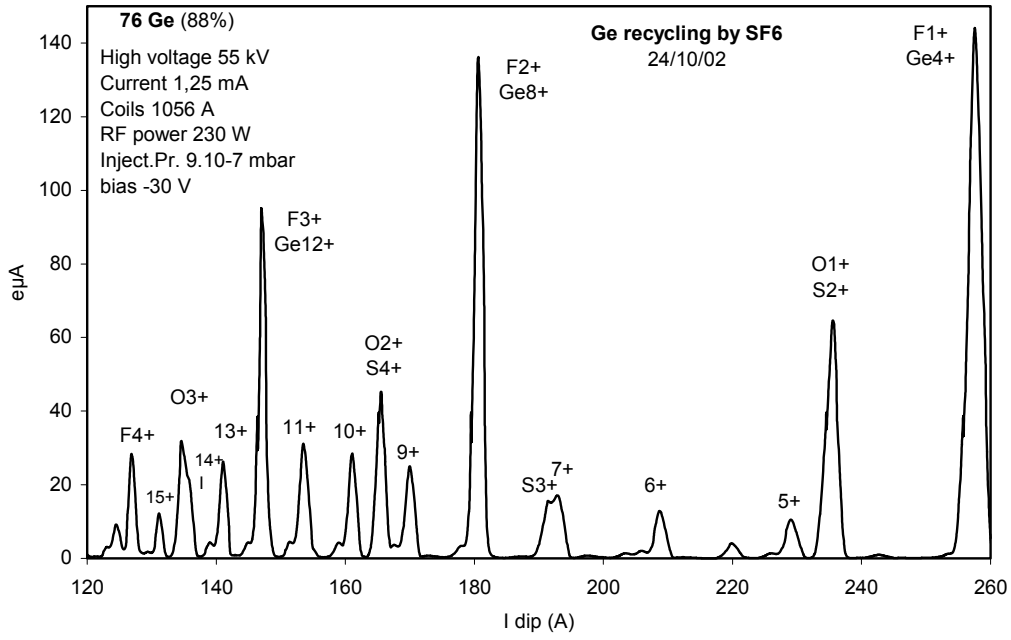


Figure 6

Table 1 : Ionization efficiencies, corrected from transport, and injected flux rate of particles from various production methods measured in operation conditions.  
 \* ECR-4 on high voltage platform      \*\* ECR-4M insulated at 25 kV

Ion beam	% isotope	production method	operation intensity e $\mu$ A	injected flux rate $\mu$ Ap	transport efficiency	overall ionization efficiency
36 Ar 10+	99%	gas	95	360	67% *	50%
36 S 8+	63%	mivoc	30	180	56% **	23%
48 Ca 10+	65%	oven	17	67	69% *	11%
58 Ni 11+	68%	mivoc	24	290	39% **	18%
76 Ge 13+	75%	oven	5	60	43% **	11%
76 Ge 13+	75%	recycling	29	< 124	75% *	> 40%
78 Kr 16+	99%	gas	17,5	86	65% *	61%
78 Kr 16+	99%	gas	29	140	67% *	40%

Table 1