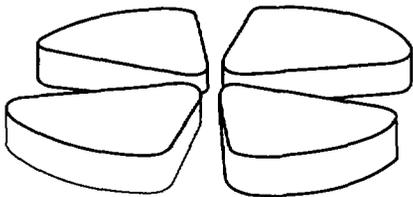




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HIGH INTENSITY METALLIC ION BEAM FROM AN ECR ION SOURCE USING THE MIVOC METHOD

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The MIVOC method has been successfully used at GANIL to produce a high intensity nickel beam with the ECR4 ion source : $20 \mu\text{A } ^{58}\text{Ni}^{11+}$ at 24 kV extraction voltage. This beam has been maintained for 8 days and accelerated up to 74.5 MeV/u by our cyclotrons with a mean intensity of $0.13 \mu\text{A}$ on target. This high intensity, required for experiment, led to the discovery of the doubly magic ^{48}Ni isotope.

Experimental setup, handling and off-line preparation using a residual gas analyzer are described in this report. The ion source behavior, performances and limitations are presented in the case of nickel and iron. The ionization efficiencies have been measured and compared to the oven method usually used at GANIL.

I. Principle and advantages of the MIVOC method

The MIVOC (Metallic Ions from Volatile Compound) method [1,2,3] is based on the use of high vapor pressure metallic compounds at room temperature : as an example, nickelocene $\text{Ni}(\text{C}_5\text{H}_5)_2$ having a vapor pressure of $3.5 \cdot 10^{-3}$ mbar at 20°C . Usually under powder form, the compound is placed in an hermetic box called MIVOC chamber, and is connected to the source via a rough regulation valve. The molecule flow injected into the source is easily increased by a larger opening of the regulation valve. As the vapor pressure is high at room temperature, the molecules condensed on the cold walls of the source can be recovered, and therefore higher ionization efficiencies can be obtained.

This is an undeniable advantage when using rare and expensive isotopes. It is also an advantage for metals with low vapor pressure. Nickel is a good example : to get the same vapor pressure as the MIVOC compound with the oven method, a temperature of about 1500°C must be reached. An increase of the evaporation rate would require higher temperature not suitable for the oven, whereas it is simply obtained by a larger opening of the regulation valve with the MIVOC method.

II. Experimental setup

As a complex condensation-evaporation process occurs along the way the MIVOC molecules take, the conductance could not be calculated by classical formula. Several tries were required before finding the best position of the MIVOC chamber and the adequate regulation valve. This optimum configuration is represented in Fig. 1. The chamber is axially positioned, about 60 cm from the ECR plasma, and a mechanical valve with a maximum conductance of about 1 L/s for air is used to adjust the input flow. The valve opening is remotely controlled by using a mechanical gear.

In order to measure the consumption, the first MIVOC chamber was equipped with a removable cup containing the MIVOC powder ((a) in Fig. 1). Since the vacuum has to be broken and perilous handling has to be done, this way of weighting was not suited

respectively for air-sensitive and toxic compounds. Therefore, a new balance measuring up to 1 kg with a precision of 1 mg has been bought, allowing the whole chamber to be weighted without breaking the vacuum. The new chamber ((b) in Fig. 1) has been designed for safety handling : no container and easy filling.

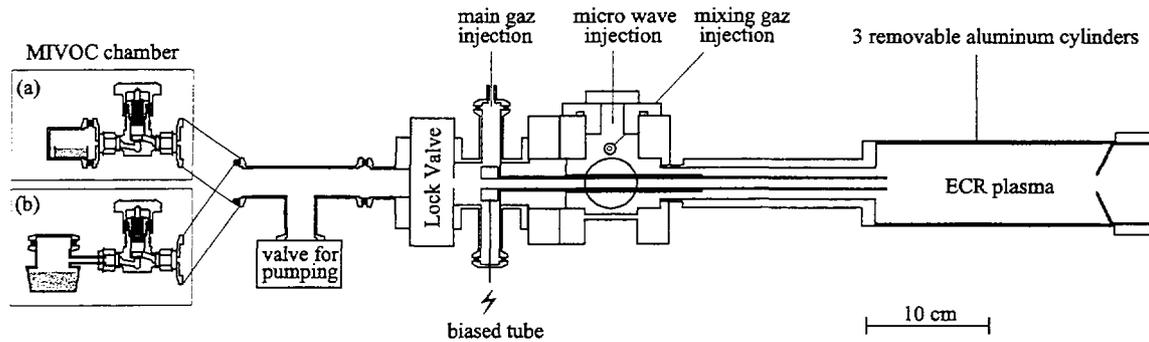


FIG. 1. The MIVOC chambre and the 14 GHz Electron Cyclotron Resonance (ECR) ion source. The aluminum plasma electrode has a hole of 12 mm diameter. Aluminum cylinders have been placed inside the plasma chamber in order to make easier the later cleaning.

III. Experimental results

All the results presented in this report have been obtained with the ECR4 or ECR4M ion source [4]. First tries with iron have already been presented in Ref. [5].

Further investigations started in January 1999. The MIVOC chambre was a simple bottle usually used for isotopically enriched gases, and the valve of the bottle was not remotely controlled. A few days only have been needed to condition the source and find the right opening of the manual valve. An intensity of $92 \mu\text{A}$ for Fe^{9+} at 19 kV extraction voltage has been produced. Although this high intensity was easily obtained, we have observed some instabilities : the beam intensity suddenly decreased for a few tens of second and then came back to the nominal value. The reason of these instabilities has not been yet understood.

–we have observed that the negative voltage applied to the biased tube (see Fig. 1) has a major importance : by doing a short-circuit between the biased tube and the plasma chamber, the intensity for each charge state is strongly lowered. This could be explained in this way : the negative voltage repels the low energy electrons (energy $< -eV_{\text{bias}}$) escaping from the plasma, and therefore reduces the number of broken or ionized molecules by electron impact inside the biased tube. The number of molecules injected into the plasma is then strongly enhanced.

–Although there are numerous carbon atoms in the ferrocene molecule (1 iron atom for ten carbon atoms) , i.e. important carbon deposition on the plasma chamber walls, the ion charge-state distribution is not too much degraded : the spectrum is still shifted on $9+$.

–Due to the presence of numerous and useless atoms in the molecule, the total extracted current is high : 4.5 mA for $92 \mu\text{A}$ Fe^{9+} ! It seems to be the major limitation factor for increasing further the iron intensity.

More details are given in the proceeding of the last ECR workshop [6].

In view of this nice results obtained with iron and the increasing requests for a nickel beam at high intensity, we initiated tests with nickel in May 1999. A MIVOC chambre has

been designed in order to allow a measurement of the consumption ((a) in Fig. 1). As for iron, a high intensity nickel beam has been easily obtained (see Fig. 2).

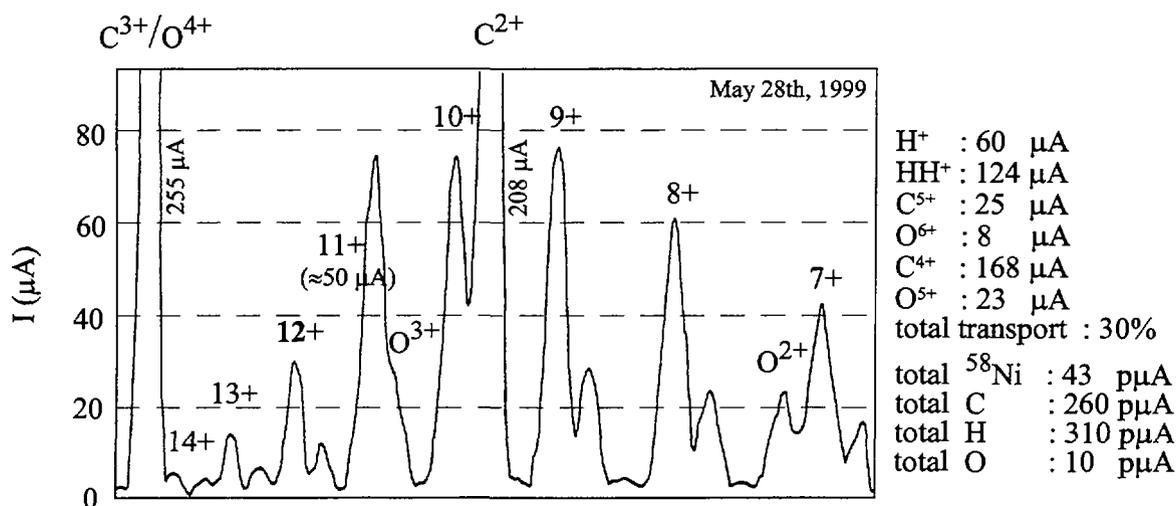


FIG. 2. Best charge state distribution of natural nickel (68%) produced by the ECR4 ion source. Optimized on Ni¹²⁺. Extraction voltage 23.6 kV, total extracted current 4.45 mA, RF power 400 W, reflected RF power 50 W, biased tube -157 V/0.51 mA, no mixing gas, injection pressure $3.5 \cdot 10^{-6}$ mbar, and extraction pressure $5 \cdot 10^{-7}$ mbar.

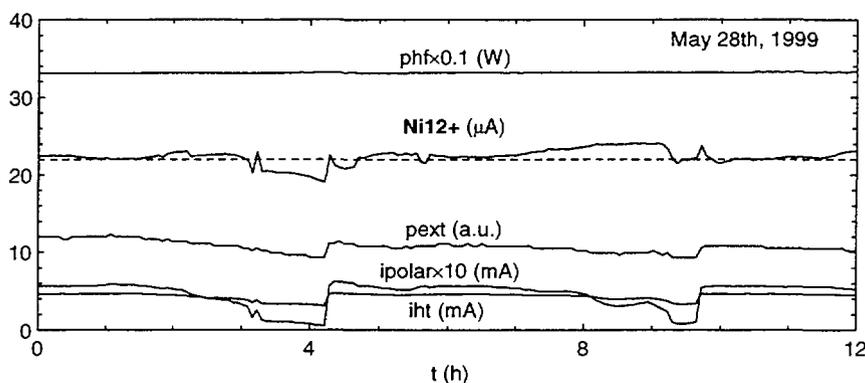


FIG. 3. Stability of the Ni¹²⁺ beam.

A stable Ni¹²⁺ beam with an intensity of 22 μA was accelerated by the first cyclotron C02 at 1 MeV/u for test. An acceleration efficiency of 20% has been measured. This value was slightly lower than the usual value, meaning that the beam emittance was not too much degraded although the high intensity of 4.5 mA extracted from the ion source.

A nickelocene consumption of 345 mg has been measured after 80h, i.e. 4.3 mg/h. The consumption is then 2.9 mg/h for ⁵⁸Ni(C₃H₅)₂ and 0.9 mg/h for ⁵⁸Ni (see Tab. 1). This consumption can be converted into neutral atoms injected in the source : 415 pμA for ⁵⁸Ni (1 pμA=6 · 10¹² s⁻¹). As the total number of nickel ions extracted from the source has been measured to 35 pμA, it gives an ionization efficiency of 8% for nickel. Let us notice that it supposes a transport efficiency of 100%. As a comparison, we get an ionization efficiency 8 times lower with the oven method.

	neutral injected		ions extracted	ionization efficiency
$^{nat}\text{Ni}(\text{C}_5\text{H}_5)_2$	4.3 mg/h	610 μA		
$^{58}\text{Ni}(\text{C}_5\text{H}_5)_2$	2.9 mg/h	415 μA		
^{58}Ni	0.90 mg/h	415 μA	35 μA	8%
Hydrogen		6100 μA	250 μA	4%
Carbon		6100 μA	210 μA	3%

TAB. I. Ionization efficiencies measured in May 1999.

A first high intensity nickel beam was scheduled in September 1999. Unfortunately, we were not able during this run to reproduce the impressive results obtained in May 1999. Abnormal amount of oxygen was present in the spectrum. At that time, we noticed that the nickelocene was brown whereas it was black during the tests of May. However an mean intensity of 20 μA Ni^{11+} has been produced for 8 days. This nickel beam was accelerated to 74 Mev/u with a mean intensity of 0.13 μA on target. It led to the discovery of the doubly magic ^{48}Ni isotope [7].

At the beginning of the year 2000, the source ECR4 used for metallic ion developments has been replaced by an ECR4M. This change was justified by a detachment of some permanent magnets of the hexapole. This last one has been replaced by a Halbach type hexapole, mechanically stable (no glue), and with a stronger magnetic field. The axial magnetic field has also been reinforced with a new set of coils.

In order to better understand the problems of oxygen contamination during the run of September, we bought a residual gas analyzer. It enables to diagnose off-line the MIVOC chambre and to scan up to mass 200. The MIVOC chamber were first analyzed off-line ((a) in Fig. 4) : much more water than nickel was observed. Then, the chamber was connected to the ECR4M ion source. After a few days running the source, we obtained the spectrum (b) shown in Fig. 4 : abnormal amount of oxygen was present (as seen in September), and the nickelocene was brown again. The different observed-colors for nickelocene were in close correlation with the different manufacturers, and therefore with the chemical process used for the nickelocene synthesis. With time, a decrease of the oxygen level together with a rise of the nickel have been observed ((c) in Fig. 4). Then the chamber was removed and analyzed off-line ((d) in Fig. 4) : the water level was much lower than those initially measured. As a conclusion, the chemical purity of nickelocene is a crucial requirement to get good source performance : a preliminary off-line analysis of the compound must be done.

This study has been beneficent for the run of June 2000, and the best performances obtained in May 1999 have been reproduced. We have noticed that the nickelocene color was black again as it was in May 1999. Awkward instabilities were present during this run : about every 10 s the beam collapsed for 1 s. They were clearly in close correlation with a shut down of the high voltage power supply. A so high repetition rate was not previously observed with the ECR4 ion source. Let us notice that these instabilities were not observed when running the source ECR4M with pure argon at the same total intensity level. A higher sensitivity in the extraction area with ECR4M, and therefore a more difficult tuning of the source, could explain this behavior. Investigations are planed in the near future.

Last June, our chemist colleague P.A. Jaffres from ISMRA laboratory has successfully realized the synthesis of ferrocene from enriched ^{58}Fe metal, following the procedure described by A.F. Reid [8]. The efficiency was 100% ! Let us notice that the dissolution phase in hydrochloric acid was 10 times longer compared to the use of natural iron. The ion source performances were reproduced, and a total ionization efficiency of 7% has been measured.

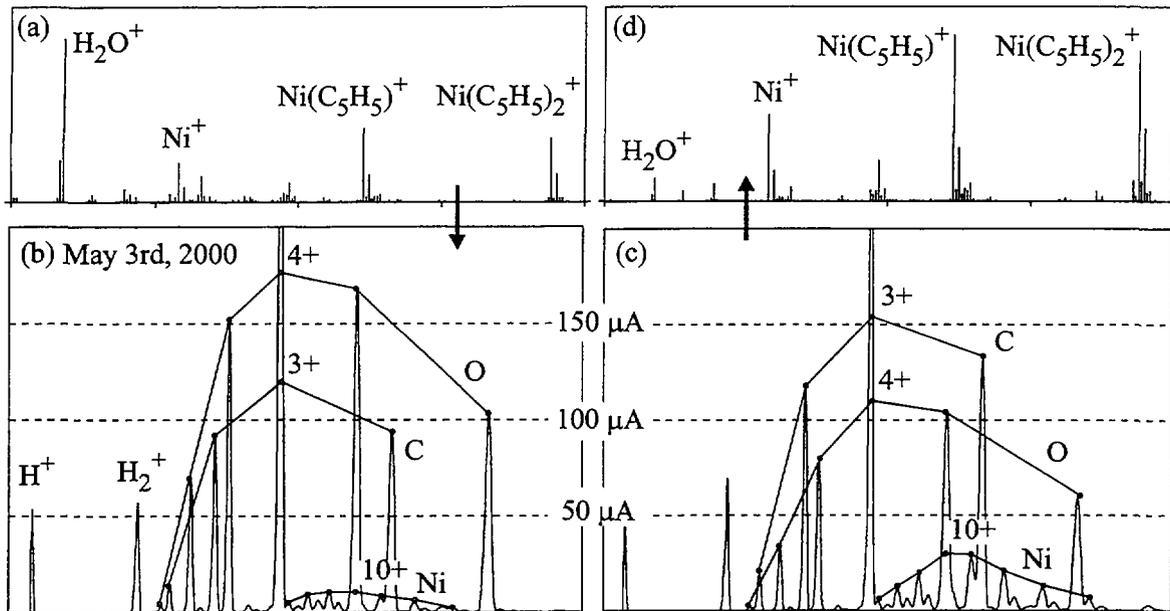


FIG. 4. On top : spectra obtained with the residual gas analyzer. Below : spectra obtained with the ECR4M ion source.

IV. Conclusion

Reproducibility of the source performances for nickel has been significantly improved by an off-line analysis of the MIVOC compound. The high intensities produced (never reached with an oven), as well as the successful synthesis of ^{58}Fe , offer new perspectives for physics experiments. However, severe instability problems have to be investigated and solved before it becomes a routine method of production. Although high ionization efficiencies have been measured with nickelocene and ferrocene, it has to be investigated for other organometallic compounds.

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