A TUBULAR IONIZER AS AN EFFICIENT NEGATIVE FLUORINE ION SOURCE*

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(Received December 7, 1999)

In order to establish the optimal conditions of F^- ion production by the tubular ionizer extensive studies of SF6 ionization using the mass separator were performed. The SF6, SF5, SF4, SF3 and F negative ions were observed, and the F^- yields as a function of the source temperature, gas pressure and an amount of alkaline metal vapors (K, Na, Ca, Sr, Ba) were measured. The efficiency of F^- production of about 40obtained for the optimal conditions. The delay time and adsorption enthalpy of fluorine on the tantalum surface has been measured for the first time.

PACS numbers: 07.77.Ka, 29.25.Ni, 82.30.Fi

Negative ions are needed for:

— accelerators and RIB (Radioactive Ion Beam) Facilities,

— mass separators (chemical selectivity),

— implantators (for implantation of insulators — no surface charging).

The radioactive fluorine is one of the important isotopes for the RIB (Radioactive Ion Beam) Facilities, because it is needed to study several astrophysical processes like Ne production in CNO stellar cycle, an inhomogeneous Big Bang and explosive hydrogen burning. The tubular ionizer ion source developed in Dubna by Piotrowski *et al.* [1] is one of the most useful sources of positive ions in the ISOL (isotope separation "on-line") facilities, because it has a very high (almost 100%) efficiency and a short holdup time

* Presented at the NATO Advanced Research Workshop, Krzyże, Poland 2–4 September 1999.

for many isotopes. Essentially it is a small tube heated to a very high temperature. An exact mechanism of this very high efficiency is still a subject of discussion. Recently we were able to demonstrate [2] that negative ions are produced by this type of the source. In our source the tubular emitter made of tantalum has an inner diameter of 3 mm, outer of 5mm and the length of 30 mm. The ionizer temperature is measured using an optical pyrometer. The solid material (alkaline metals) is loaded into the Knudsen cell (oven) in order to be evaporated at a temperature controlled by a thermocouple. The gas can be introduced to the ion source by the inlet tube and the gas flow is controlled by a calibrated leak or a flowmeter. In the extraction region the transversal magnetic field suppresses the electron current. Some other details can be found in [2]. The extracted beams are analyzed by a low energy (up to 30 kV) mass separator [3] with the resolving power of 700 (FWHM), and a beam diagnostic system — a mechanical scanner and a Faraday cup with the sensitivity better than 1 pA.

When an atom (or a molecule) hits a hot surface the ionization efficiency

$$\beta = \frac{n_i}{n_0},$$

where n_i is the number of ions, and n_0 is the number of atoms (molecules), is given by a classical Saha–Langmuir formula:

$$\beta = \left[1 + \frac{g_0}{g_i} \exp\left(\frac{\phi - E_A}{kT}\right)\right]^{-1},$$

where g_i and g_0 are statistical weights of the ion and the atom, respectively, E_A is the electron affinity of the incident particle, ϕ is the work function of the surface material, and T is the surface temperature.

The work functions of tungsten (4.54 eV) or tantalum (4.2 eV) are much higher than the affinity for any element, thus the expected surface ionization is very small.

A large increase of the efficiency (in comparison to the Saha–Langmuir formula) in the tubular geometry observed for the positive ions was found by us also for the negative ones.

Here we present some results of systematical studies of negative ion production from the SF_6 gas. Fig.1 shows an example of the temperature dependence of currents of different negative ions produced by decomposition of the SF_6 molecule. This dependence is definitely not described by this formula, which predicts an exponential dependence on the inverse temperature.

It is known that adsorption of electropositive atoms on a surface decreases the work function while electronegative atoms tend to increase this function. We found that the negative ion yield is very strongly dependent on the amount of the impurities present in the source.

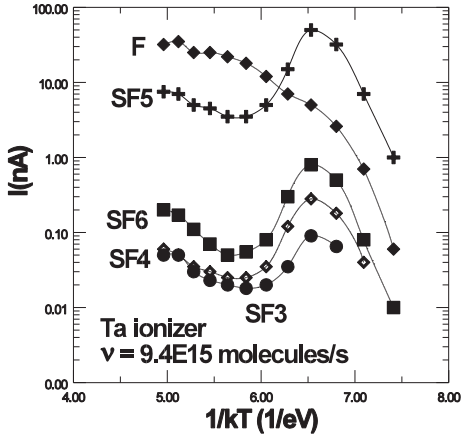

Fig. 1. The dependence of the F, SF_6, SF_5, SF_4 and SF_3 negative ion currents on the Ta ionizer temperature for the gas flux ν of $9.4*10^{15}$ molecules/s.

In our experiment definite fluxes of Na, K, Ca, Sr and Ba metal vapors using the Knudsen cell were introduced into the source, and the cell temperature dependence of F^- currents was measured — some examples are shown on Fig. 2–4. The Na, K and Ba vapors increase these currents very substantially — the strongest enhancement was observed for the K vapor. No effect was observed for Ca and Sr.

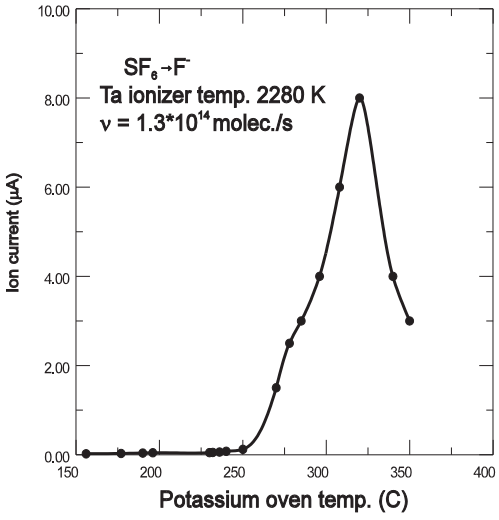

Fig. 2. The F^- current as a function of the temperature of the Knudsen cell (oven) filled with K for the definite ν and T given on the figure.

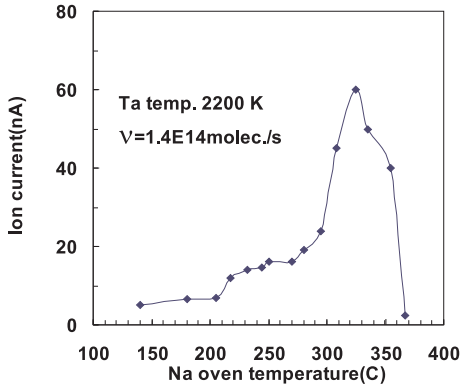

Fig. 3. The F^- current as a function of the temperature of the Knudsen cell (oven) filled with Na for the definite ν and T given on the figure.

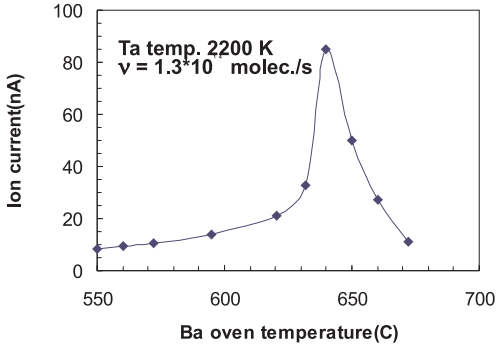

Fig. 4. The F^- current as a function of the temperature of the Knudsen cell (oven) filled with Ba for the definite ν and T given on the figure.

It should be noted that the current of 8 μA of F^- at the maximum corresponds to 40% efficiency per molecule of F^- production.

Such high enhancement of F^- ion formation involves several distinct negative ion formation processes, and the most important ones are:

1. Electropositive adsorbate induces work function changes, and then the value of the surface ionization efficiency.

2. In the hot cavity of the ion source tube the plasma is formed and separated by the plasma sheath potential from the walls. This potential can change the ionization efficiency [4].

3. Negative ions can be formed by a dissociative attachment:

$$e + XY \rightarrow X^- + Y \, , e + XY \rightarrow X + Y^- \, ,$$

or charge transfer collisions:

$$X + Y \rightarrow X^- + Y + \Delta E\,,$$

where ΔE is an energy defect:

$$\Delta E = E_A(X) - E_i(Y)\,.$$

$E_A(X)$ is the affinity of an X atom (molecule) and $E_i(Y)$ is the ionization potential of an Y atom.

The importance of these effects is dependent on the gas and introduced vapor pressure, the ionizer surface and the temperature. Our measurements demonstrate that the increase of the ionization efficiency under the influence of the metal vapor is dependent on the ΔE value, which is the smallest one in the case of K. Thus one can expect that the best impurity to get the highest efficiency should be the Cs vapor.

For the RIB applications any time delays comparable to the isotope half- lives can result in significant losses. We measured the adsorption delay time of the negative fluorine ions produced in dissociation of the SF_6 in the tantalum ionizer at 2395 K.

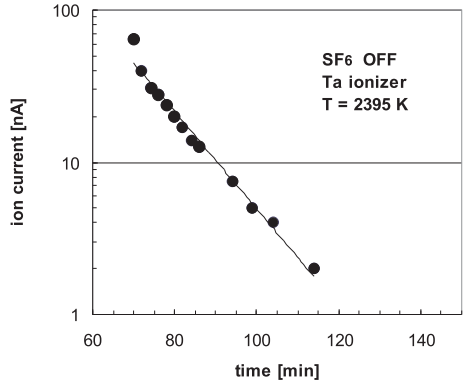

Fig. 5. The time dependence of the F^- ion current after switching off the SF_6 gas flow.

A definite (1.35*10−5 mbar*liter/s) SF_6 flux has been applied to the ionizer from the reference leak and it was found that the F^- current was saturated after 70 min. The time dependence of the current after switching off the gas is shown in Fig. 5. The exponential fit gives the holdup time of 13.6 min. In the tubular geometry this time is affected (multiplied) by the number of collisions before the ion can leave the source volume. In our case this number is equal to 40, and the adsorption delay time on the tantalum

surface is then $\tau_a = 20.4$ s. This number can be compared to the half-life time of the ^{17}F (65 s) and ^{18}F (110 m) radioactive isotopes.

The temperature dependence of the adsorption time is described by the Frenkel equation:

$$\tau_a = \nu^{-1} \exp\left(\frac{\Delta H}{kT}\right),$$

where ν is the attempt frequency with which the adsorbed particle tries to escape from the adsorption surface, ΔH is the enthalpy of adsorption and T is the surface temperature. Assuming the typical value $\nu = 10^{13}$ s^{-1} one obtains $\Delta H = 6.8$ eV. This value has been determined for the first time and can be useful in estimation of the fluorine production efficiency for different ion sources. This definite holdup time shows that the F ions are the result of ionization of the neutral atoms, which are produced in the two step process:

$$e^- + \mathrm{SF}_6 \rightarrow \mathrm{SF}_6^{*-},$$

$$SF_6^{*-} \rightarrow \mathrm{SF}_5^- + F,$$

and then they are adsorbed on the Ta surface. Because in the RIB facilities the neutral atoms are initially produced, the study of F^- production using the SF_6 gas seems to be a good simulation of the ion source properties for these facilities.

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