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STUDIES OF THE ION SOURCE WITH SURFACE-VOLUME IONIZATION

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Introduction

In papers [1,2] a design has been described of the ion source where ionization is realized in the cylindrical cavity of the tungsten rod ($\varnothing$ ~ 5 mm) heated to temperature $T \approx 3000°K$. Ions are extracted from the orifice about 0.2 mm in diameter in the wall of the cavity. In refs. [1-4] it has been shown that the ionization efficiency in such a source, with a microgram amount of the feed material, is much higher (in some cases more than one order of magnitude) than that calculated by the Saha-Langmuir formula:

$$\alpha_{S-L} = \exp(\phi - V_i^*/kT), \qquad (1)$$

where $V_i^* = V_i - kT \ln A$ is the corrected value of the first ionization potential V_i involving the ratio A of the statistical weights of ionic and atomic states of particles, T is the temperature, ϕ is the work function, k is the Boltzmann constant. By the ionization efficiency we shall call here the following quantity:

$$\beta_{S-L} = \alpha_{S-L}/(1+\alpha_{S-L}). \qquad (2)$$

In calculating the ratio A we used the data on electronic levels from paper [5] and values of V_i from ref. [6]. For all rare earth elements (REE) as well as for rubidium, barium, strontium, yttrium, zirconium, niobium and hafnium the value of A varies from 0.25 to 2.0, that at $T = 3000°K$ changes the tabulated values of the first ionization potentials by a value from -0.2 to + 0.4 eV.

The agreement between theoretical and experimental values of the ionization efficiency is observed if it is assumed that the work function of our ionizer is $\phi \approx 6$ eV, for instance, due to oxidation of tungsten. However, as has been shown clearly in ref. [7], at $T \geq 2500°K$ the effect of this oxidation can be neglected. As the estimates and results of modelling with the use of an electrolyzer indicate, such a large discrepancy between the calculated by the Saha-Langmuir formula and experimental values of β, for the ion source geometry under consideration, cannot completely be explained by the increase of the effective extraction orifice for ions due to the penetration of electric field inside the cavity.

When working with an ion source of the type [1] it was observed that its efficiency does not depend in practice on the ionizer material (tungsten, rhenium, tantalum), however, it depends strongly on the value of the extracted ion current density and on the ionizer temperature. Since here ionization takes place in the closed volume it is necessary to take account of the fields of space charges, produced in the volume by ion and electron flows from walls [8] when considering the mechanism of work of the source.

In what follows we will present a qualitative description of the mechanism of work of the ion source with surface-volume ionization as well as the experimental results obtained.

Mechanism of the Surface-Volume Ionization

For sufficiently large concentrations of neutrals, electrons and ions in a cavity, i.e., for cavity dimensions much larger than the Debye shielding distance ($R \gg r_D$), in the cavity volume the thermodynamically equilibrium plasma is generated with the following potential drop in the sheath [9]:

$$\Delta U = (kT/2)\ln\left[A_0 T^2/j^{\circ +}\beta_{S-L}(M/m)^{1/2}\right] - (\phi/2), \qquad (3)$$

where $A_0 = 120$ acm^{-2}degr^{-2} is the Richardson-Dushman constant, $j^{\circ +}$ is the flux of neutrals and ions per unit cavity surface, m and M are the electron and ion masses, respectively. The magnitude and sign of this potential barrier depend both on the thermoelectron flux, defined by the temperature T, and on the ion flux ($j^{\circ +} \cdot \beta_{S-L}$), produced in the surface ionization on the cavity walls, into the volume. Under the accepted conditions of equality of the particle flux from the walls into the volume and that from the volume to the walls we write expression (3) for the equilibrium ion concentration n^+ in the cavity volume:

$$\Delta U = kT\ln\left[A_0 T^2/en^+(kT/2\pi m)^{1/2}\right] - \phi \qquad (4)$$

(e is the electron charge). For the Boltzmann distribution of charged particles one can obtain the following expression:

$$\alpha^* = n^+/n^\circ = \alpha_{S-L}\exp(\Delta U/kT), \qquad (5)$$

where n° is the neutral concentration in the cavity, for the ionization degree in the cavity. By substituting formulae (1) and (4) into (5) and changing the ion concentration in the cavity by the density of the ion current extracted from the isothermal plasma according to ref. [10]: $j^+ = 1.47 e n^+ (kT/2\pi M)^{1/2}$, the final result is as follows:

$$\alpha^* = \left[1{,}47 A_0 T^2/j^+(M/m)^{1/2}\right]\exp(-V_i^*/kT),$$
$$\beta^* = \alpha^*/(1+\alpha^*). \qquad (6)$$

Strictly speaking, such an expression for the ionization degree is obtained only for the values of the work function in formula (1) close to the Richardson ones. In this approximation, the ionization degree in the cavity volume should not depend on the work function of the ionizer surface. Analogous expressions can be found by using the Saha formula for the equilibrium ionization.

The ratio of the ionization degree α^* to that calcu-

lated by the Saha-Langmuir formula is as follows:

$$N = \alpha^* / \alpha_{S-L} = [1{,}47\, A_0 T^2 / j^+ (M/m)^{1/2}] \exp(-\phi/kT). \quad (7)$$

In Fig. 1 we have shown the values of the ratio calculated for different temperatures as a function of the extracted ion current density. From expression (7) it follows that the value of N grows with decreasing work function, however, since at the same time the ionization degree α_{S-L} falls, the value of α^*, roughly speaking, does not change. At $\phi = 5$ eV the value of N should not be larger than $\sim 10^3$ as at high temperatures it is difficult to obtain the ion current density smaller than 10^{-5} a/cm^2 (see below). At the same time the volume ionization efficiency β^*, as compared with that on the surface β_{S-L}, for instance, for REE ($5.4 \leq V_i^* \leq 6.2$ eV) should be 5 - 100 times large. On the other hand, at definite values of the ion current density and ionizer temperature, also a decrease can be observed for β^* as compared with β_{S-L}, that is due to the change of sign of the barrier ΔU.

Experimental Procedure

Experiments were carried out on the electromagnetic mass-separator of the YASNAPP facility [11]. The ion source [1] was employed with a modified ionizer drawn in Fig. 3. With this device, i.e., with an ionizer and vaporizer separated from each other, a temperature almost steady along the ionizer length is achieved and it is possible to keep constant the extracted ion current density j^+ in the course of experiment. To obtain the required ion current density we used the following elements: barium, ytterbium, dysprosium, lutecium and hafnium.

The ionization efficiency was measured for the radioactive isotopes ^{83}Rb, ^{85}Sr, ^{88}Y, ^{88}Zr, ^{131}Ba, ^{140}Nd, ^{146}Eu, ^{149}Gd, ^{160}Tb, ^{168}Tm, ^{169}Yb, ^{172}Lu and ^{175}Hf, obtained in quantities of 10^8 - 10^{10} atoms from the tantalum and molybdenium targers irradiated on the

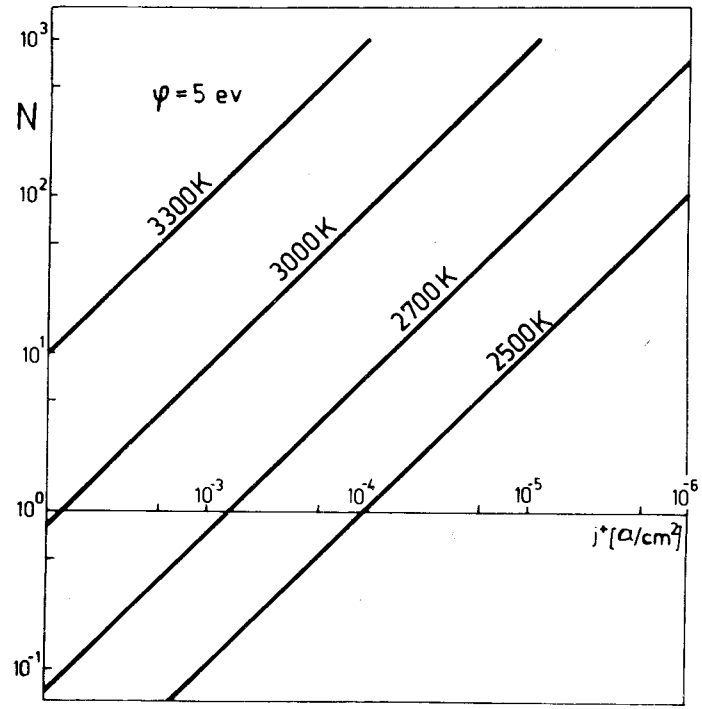

Fig. 1. The ratio of the volume ionization degree α^* to the surface one α_{S-L} as a function of the ion current density extracted from the volume. The calculation has been performed for $\phi = 5$ eV and various temperatures of the metal surface.

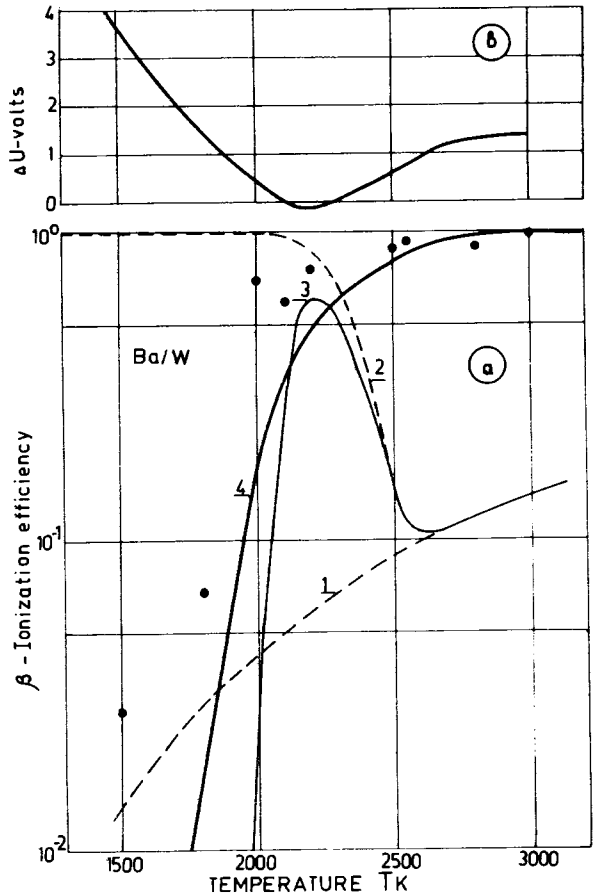

Fig. 2. a) The ionization efficiency of barium ($V_i^* = 5$ eV) on tungsten as a function of the metal surface temperature. Curves 1-3 are taken from ref. [7]. Curve 1 is the ionization on the clean tungsten surface, curve 2 the same on the oxidated surface, curve 3 the ionization on the oxidated surface with the barium coverage taken into account. Curve 4 represents the ionization efficiency β^*. Points are the experimental results for barium ionization in the ion source [1]. b) The dependence of the potential drop ΔU on the ionizer temperature under the conditions given by curve 3.

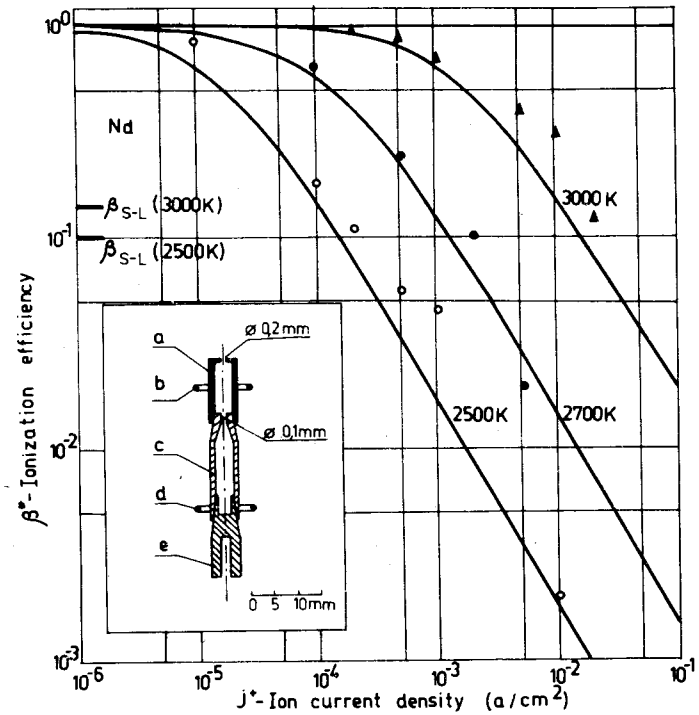

Fig. 3. The ionization efficiency β^* of neodimium ($V_i^* = 5.5$ eV) as a function of the ion current density. Curves represent the values calculated for different ionizer temperatures. Experimental points are obtained at the following temperatures: ▲ – T = 3000°K, ● – T = 2700 °K, o – T = 2500°K. Here also the scheme of the ion source is drawn: a is the tungsten ionizer, b the cathode for heating the ionizer by electron bombardment, c the tungsten vaporizer, d the cathode for heating the vaporizer, e the tantalum plug of the vaporizer.

JINR synchrocyclotron. The ionization efficiency was defined by the formula:

$$\beta^* = J_c / (J_t - J_i)\eta_t \quad ,$$

where J_t is the intensity of characteristic gamma-transition in nuclei of one of the isotopes loaded into the ion source, J_i is the same in the nuclei remaining in the ion source after the mass-separation, J_c is the intensity of this gamma-transition measured on the collector of the mass-separator, η_t is the coefficient of the mass-separator transmission equal to about 0.9. The gamma-spectra were measured by the 50 cm^3 Ge(Li) -detector and processed on the "Hewlett-Packard" computer.

The ion current density $j^+ = I^+/S$ was determined by measuring, in the Faraday cylinder, the total ion current I^+ from the source with the orifice area $S = 3.10^{-4} cm^2$. The ionizer temperature was determined by using the curve found in paper [4] which gives T as a function of the electric power applied to the ionizer. The measurement accuracy for the ionization efficiency, allowing for the errors of the ionizer temperature and ion current density, was about 30%. The statistical error in measuring the intensities of gamma-transitions is significant only for values of $\beta^* < 1\%$ where it can reach 100%.

Results

It is known that the tungsten work function and surface ionization efficiency depend very much on the surface impurity. This is illustrated for barium on tungsten in Fig. 2 by curves 1 - 3 obtained in paper [7]. Curve 1 gives the values of the ionization efficiency for the clean tungsten surface (ϕ = 4.5 eV) as a function of temperature. For the partial pressure of oxygen in the volume 10^{-5} mm Hg and T < 2500°K there occurs oxidation of the tungsten surface that increases its work function and, consequently, the value of β_{S-L} (curve 2). At the same time, with decreasing T the barium coverage of the tungsten surface grows in magnitude that decreases the value of

ϕ and ionization efficiency. Curve 3 illustrates an influence of the sum of oxidation and coverage on β_{S-L}, for the constant flux of the barium atoms onto the tungsten surface $j^+ \approx 10^{14}$ cm^{-2} sec^{-1}. By using the values of β_{S-L} and ϕ of the latter curve and expression (3) it is possible to evaluate the potential drop $\Delta U(T)$ in the sheath for ionization in the volume of a closed cavity (Fig. 2b). Curve 4 on Fig. 2a gives the ionization efficiency β^* calculated by (5), with the obtained ΔU taken into account. At T ≈ 2200°K the potential barrier ΔU becomes negative, and the volume ionization efficiency is lower than the surface ionization efficiency. Experimental points in Fig. 2a are found under conditions close to those indicated above. From these results it follows that the agreement of the data and the discrepancy between values of β_{S-L} and β^* are most pronounced for T≥2500°K. Therefore, we will present the results found for the temperatures when the surface effects can be neglected.

In Figures 3 and 4 the experimental values of β^* for terbium and lutecium are plotted versus the ion current density which are in good agreement with those calculated by formula (6). For comparison, in the same figures the values are presented for β_{S-L} at T = 2500°K and T = 3000° K for ϕ = 5 eV. As can be seen, the ionization efficiency β^* strongly depends on the magnitude of the ion current density extracted. At low temperatures the high ionization efficiency can be obtained only for extremely small values of j^+. Under real experimental conditions, even the ion current due to contaminations of the ion source or due to vaporization of a material of the target inside the ionizer (see ref. [4]) can limit the efficiency. For instance, in the mass-separation of radioactive isotopes of ribudium, strontium, yttrium and zirconium diffusing from the irradiated molybdenum target, which is inside the ionizer, it was observed that the ion current density of molybdenum from the source grows exponentially with the ionizer temperature and at T = 3000°K equals 10^{-3} a/cm^2 (Fig. 5). In this case the calculations of expected values of β^* for the above indicated isotopes, as well as the experimental results for T from 2600°K

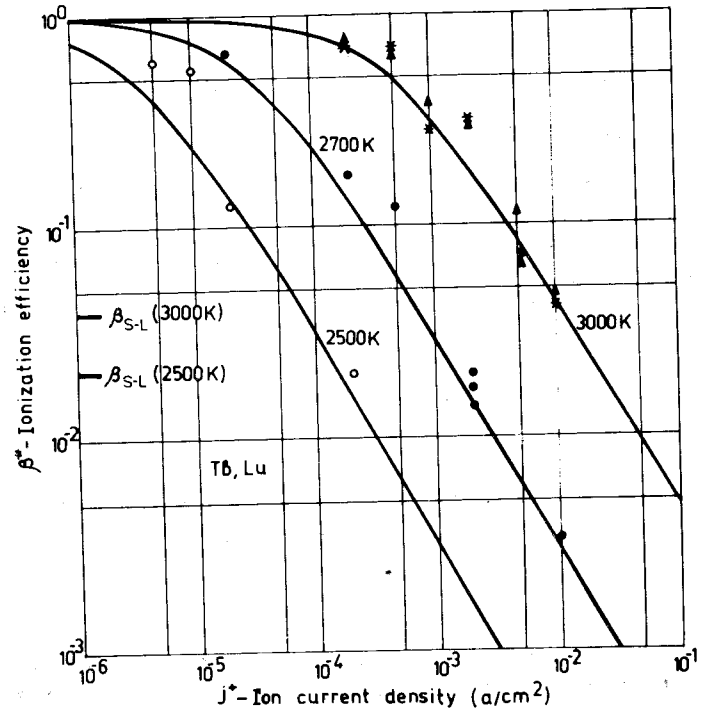

Fig. 4. The ionization efficiency β^* of terbium ($V_i^* = 5.85$ eV) and lutecium ($V_i^* = 5.8$ eV) as a function of the ion current density. Experimental points are obtained at the following temperatures: ▲ (Tb) and * (Lu) – –T=3000°K, ●(Tb) - T = 2700°K, o(Tb) - T = 2500°K.

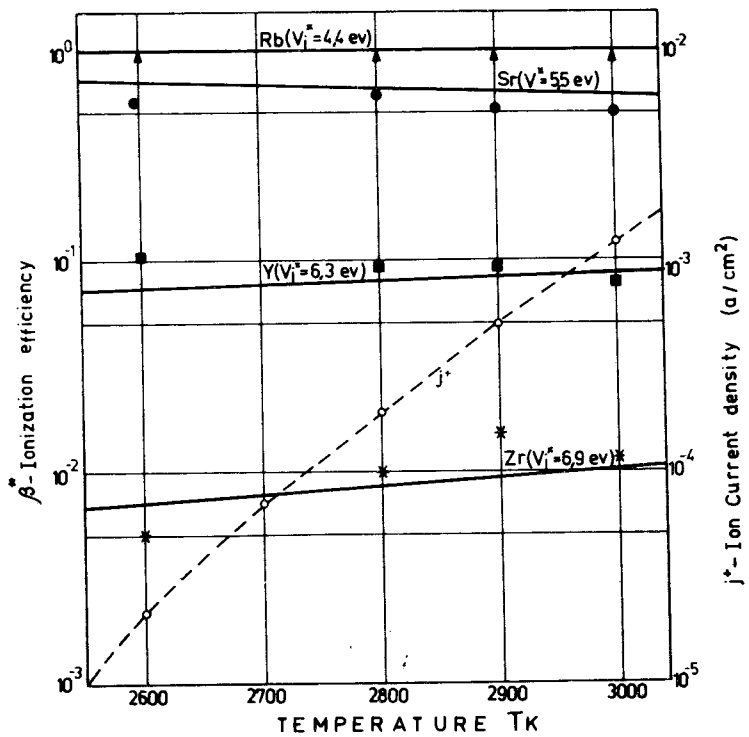

Fig. 5. The calculated and experimental values of β^* for the ionization of rubidium, strontium, yttrium and zirconium from the irradiated molybdenum as a function of the ionizer temperature. Experimental points: ▲ - rubidium, ● - strontium, ■ - yttrium, * - zirconium. The dotted line gives the dependence of the ion current density of molybdenum on the ionizer temperature.

to 3000°K show that the ionization efficiency here is, in practice, independent of T since, according to (6), the increase of ionizer temperature is compensated by the growth of the ion current density of molybdenum.

In Fig. 6 some experimental results are presented for the ionization efficiency at T = 2500°K and T = 3000°K for $j^+ = 10^{-3}$ a/cm^2, illustrating the dependence of β^* on the corrected values of the ionization potential. For the known values of the ion current, also the measured values of the ionization efficiency listed in refs. /1,3,4/ are in good agreement with the presented description. When the ion current from the source is small enough, e.g., $\sim 10^{-5}$ a/cm^2, and the ionizer temperature is 3000°K, for more than 2/3 elements of the periodic table the ionization efficiency β^* should be about 100% for $V_i^* \leq 6$ eV and should fall to 1% at $V_i^* = 8$ eV. It was observed on experiment that the value of $j^+ \approx 10^{-5}$ a/cm^2 is, obviously, a minimal possible one for the tungsten ionizer because this is the tungsten ion current due to the ionizer vaporization.

A characteristic example of possibilities of the source with surface-volume ionization is the results obtained for hafnium. The ionization efficiency of this, difficult for ionization, element ($V_i^* = 7.05$ eV) has increased by almost a factor of 100 in comparison with the conventional method of surface ionization. Figure 7 shows the part of gamma-spectrum of the irradiated tantalum target before mass-separation and the part of gamma-spectrum of ^{175}Hf measured on the collector of mass-separator at the position of mass M = 175 (the bottom Figure).

The results obtained indicate that an ion source of this type is especially suitable when using the ultrasmall quantities of the feed material. Hence, it can also be successfully applied in the mass-spectrometry and in the "on-line" mass-separators for obtaining the short-lived radioactive isotopes, especially when using the process of thermodiffusion from targets of the radioactive atoms without a carrier /12/.

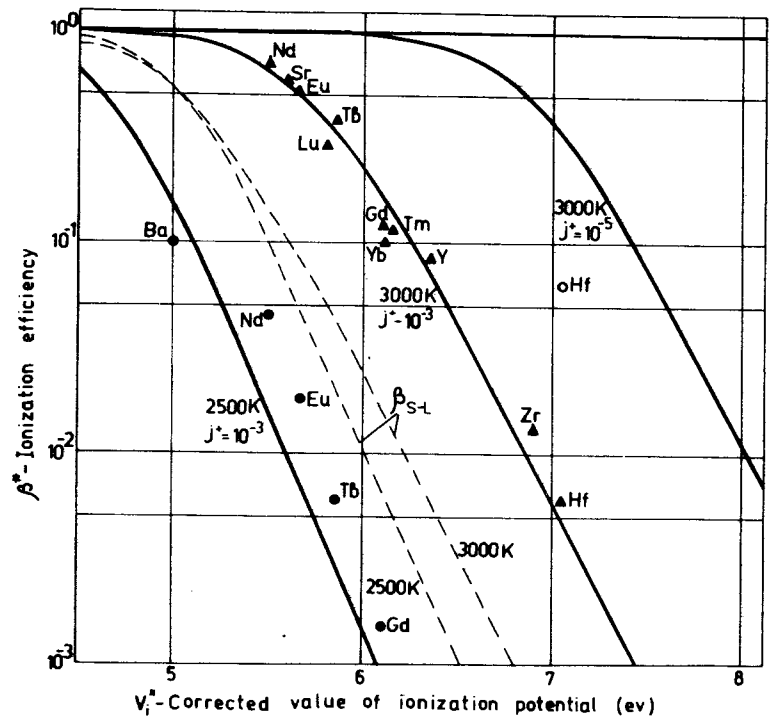

Fig. 6. The calculated and experimental values of the ionization efficiency β^* for various elements as functions of the corrected values of the ionization potential. Experimental points are obtained at the following ionizer temperatures: ▲ – T = 3000°K, ● – T = 2500°K, o – T = 3000°K ($j^+ \approx 10^{-4}$ a/cm^2). The dotted lines represent the ionization efficiency by the Saha-Langmuir formula for T = 2500°K and T = 3000°K.

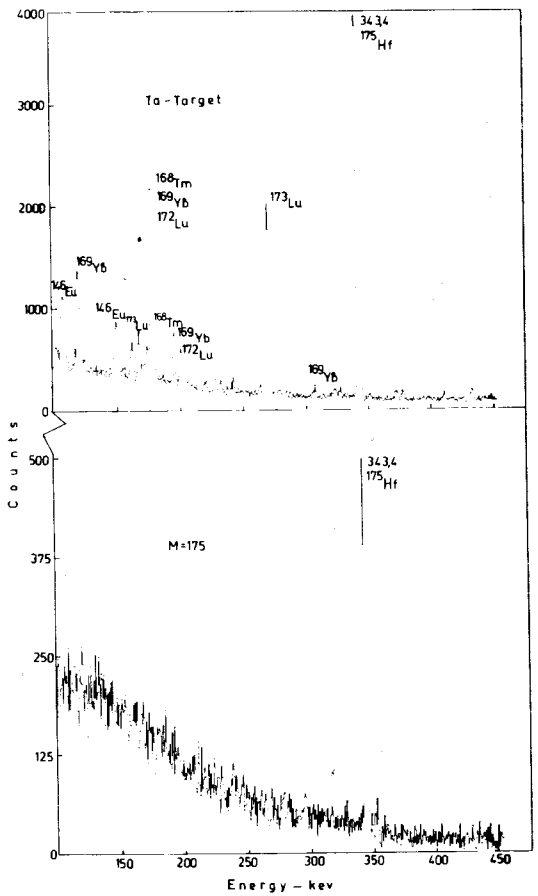

Fig. 7. The part of gamma-spectrum of the irradiated tantalum target in the ion source before mass-separation (the top figure) (the measurement time is 5 min) and the part of gamma-spectrum of ^{175}Hf measured on the collector of the mass-separator (the bottom figure) (the measurement time is 10 min).

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