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Influence of Penning ionization on ion source efficiency – numerical simulations

Abstract. *The numerical model of ionization in the plasma ion source taking the electron impact and Penning effect into account is presented. The influence of the Penning effect on the ionization efficiency is under investigation - it is shown that the carrier gas could improve ionization efficiency several times compared to the pure electron ionization case. Changes of the yield from the Penning ionization are investigated as a function of carrier gas concentration, ionization degree and concentration of carrier gas atoms in the metastable state*

Streszczenie. *Przedstawiony jest model źródła jonów uwzględniający jonizację Penninga i elektronami. Zastosowanie gazu nośnego skutkujące zachodzeniem jonizacji Penninga może zwiększyć wydajność jonizacji nawet kilkukrotnie. Zbadano zmiany wydajności jonizacji od koncentracji gazu nośnego, atomów w stanie metastabilnym i stopnia jonizacji plazmy* ***(Wpływ jonizacji Penninga na wydajność źródła jonów – symulacje numeryczne)****.*

Keywords: Ion sources, Penning ionisation, numerical simulations.
Słowa kluczowe: Źródła jonów, jonizacja Penninga, symulacje numeryczne.

Introduction

Ion source efficiency is a crucial parameter in mass and nuclear spectrometries as well as in other fields. A variety of ion production mechanisms (including electron impact ionization, surface ionization, photon ionization etc.) is described and employed in ion sources of different designs [1]. In many cases one of these processes can usually be considered as the dominant one, i.e. the one that contributes the most to the ionization yield of the ion source, as e.g. electron impact ionization in the arc discharge ion sources or surface ionization in the hot cavities. There could be, however, additional or concurrent processes that affect the performance of the ion source and could e.g. produce ions that are impossible to be created by the main mechanism, or contribute to the ion source yield to a great extent. It was experimentally shown that the electron impact ionization takes place in the thermoemission ion sources [2] resulting e.g. in multiply charged ion production, which is impossible in the surface process. The fact that the electron impact ionization could be an important (or even dominant) ion production channel in the hot cavity ion source was demonstrated by numerical simulations for both stable [3] and radioactive nuclides [4, 5]. Similarly, free electron capture is the only ionization mechanism that leads to SF6- ion formation in the hot surface ion source [6]. It is also known that two different H^- (or D^-) ion production processes occur in large intensity negative ion sources developed for the ITER plasma heating purposes [7, 8], namely surface and volume ionization channels [9]. The influence of the Penning effect on the efficiency of the plasma ion sources was also intensively studied [10-12]. It was found that using a carrier gas like He or Xe could improve ion source efficiency several times due to the Penning ionization during the collisions of Hg atoms with metastable carrier gas atoms.

The paper describes the studies of the influence of the Penning effect on the ionization in the plasma ion source using computer simulations based on the Monte Carlo method. The brief description of the model, taking into account both electron and Penning ionization, is given. The dependence of the carrier gas (He) ionization efficiency with its concentration is studied and compared with ethe xperimental measurements. Changes of Hg ionization efficiency of both processes with the plasma ionization degree and carrier gas atoms in the ground and metastable state are investigated. Relative efficiency of Hg and He ionization is also calculated and disscussed.

Penning effect

In many cases, especially during ionization of radioactive nuclides, it is necessary to apply an additional carrier gas (besides the sample) to maintain a stable discharge in the ion source chamber. This fact offers the possibility of increasing the ionization efficiency by using not only electron impact ionization of the sample atoms (B):

(1) $$B + e \rightarrow B^+ + e + e$$

but also other kinds of collisions including those of sample atoms with the metastable ones (A^m) of the carrier gas:

(2) $$B + A^m \rightarrow B^+ + A + e\,.$$

The above mentioned process is called the Penning ionization and it takes place when the ionization energy of the sample atom is smaller than that of the metastable state of carrier gas atoms [13]. It should be mentioned here that in the considered case the Penning effect happens as the He metastable levels (2^1S_0 and 2^3S_1) are higher (20.6 eV and 19.8 eV) than the Hg ionization energy (10.4 eV). Atoms in the metastable state are created by collisions with electrons and radiation transitions from the excited states:

(3a) $$A + e \rightarrow A^m + e$$

(3b) $$A^* \rightarrow A^m + \gamma$$

Metastable carrier gas atoms are lost not only in the process (2) but also during the $A^m + A$ collisions. Concentration of He atoms in the metastable state will be considered further as one of the control parameters of simulation.

Numerical model

Simulations were done using the test particle tracking approach similar to that presented in [14-16]. The numerical code follows the trajectories of particles inside the ion source chamber. A schematic drawing of the simulated sytem is shown in Fig. 1. An ionization chamber of the inner diameter of 9 mm is closed with an anode on one side and an endcap with the extraction opening (r_{ext} = 0.5 mm). The simulation area is covered by a 3D (150×100×100 cells) rectangular mesh with $\Delta x = \Delta y = \Delta z = 0.1$ mm. The electrostatic potential is found by solving the Laplace equation using the iterative over-relaxation method, as in [14-18], with the boundary conditions determined by electrode shapes and voltages. The electric field is calculated by numerical derivation of electrostatic potential. Particle trajectories are found by integration of classical equations of motion using the 4th order Runge-Kutta method [19]. Neutral particles are assumed to start their journey inside the circle of 3 mm diameter placed at the distance 3 mm from anode. Ionization is implemented using the Monte Carlo formalism similar to that desribed in [20].

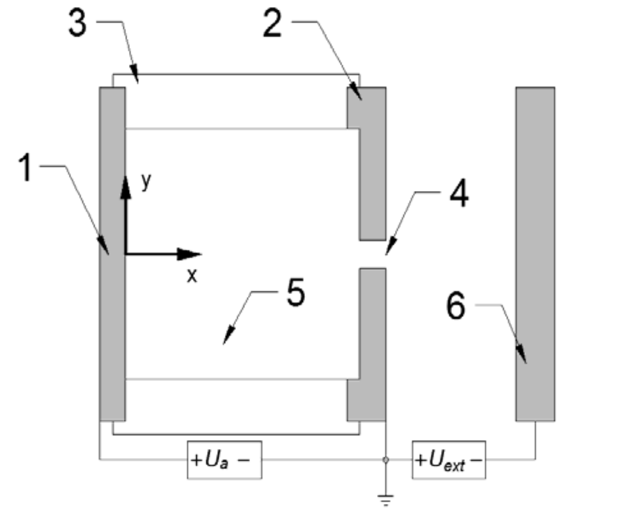

Fig.1. Schematic view of the simulated system: 1 – anode, 2 – cathode, 3 – insulator, 4 – extraction opening, 5 – ionization chamber, 6 – extraction electrode

Let us assume that the neutral particle could be ionized in *n* independent processes. Thus, the total probability of being ionized during a single simulation time step could be estimated as:

(4) $$P_{ion} = 1 - \exp\left(-\sum_{i=1}^{n} \nu_i \Delta t\right)$$

where ν_i is the frequency of *i*-th process, proportional to its total cross-section, density of target particles as well as velocity. In the considered case *n*=2 as both the electron impact ionization (*i*=1) and the Penning ionization (*i*=2) are taken into account.

(5a) $$\nu_1 = \sigma_E v n_e ,$$

where σ_E is the electron impact ionization cross-section, n_e is electron density and v is the estimated average relative velocity (as electrons are much faster than neutrals in plasma). As far as the Penning ionization is concerned, there are the reaction rates $<\sigma_P v>$ rather than the cross-section values given in the literature [21]. Therefore, the assumption that:

(5b) $$\nu_1 = \langle \sigma_P v \rangle n_m ,$$

where n_m is the density of carrier gas atoms in the metastable state. The electron impact ionization cross-sections for Hg are taken from [22]. Ions are neutralised when they hit electrodes. Ions and neutrals are tracked until they pass the extraction opening. The ionization efficiency is calculated as the ratio of the number of extracted ions to the total number of extracted particles (ions and neutrals) of a given kind (metal or carrier gas ions):

(6) $$\beta_s = N_+ / \left(N_+ + N_o\right).$$

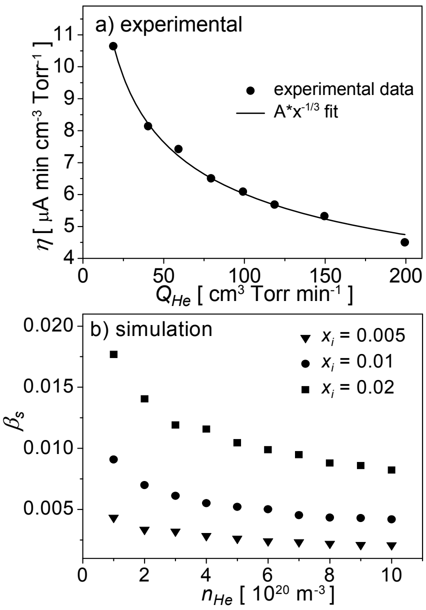

Fig.2. Efficiency of the carrier gas (He) ionization measured in experiments (a) and obtained from simulations (b).

Simulation results

Simulations were done for the Hg/He mixtures, as in the experiments presented in papers [10-12]. The carrier gas density n_{He}varied in the range from $n_{He0}=10^{20}$ m^3 up to 10^{21}m^{-3}. During experiments Hg vapours were added at much smaller leak rate (~0.35 cm^3 Torr min^{-1}) than the carrier gas (20-200 cm^3 Torr min^{-1}). Hence, the influence of electron density depends mostly on the He atom density.

It was assumed that the electron density was scaled with n_{He} as:

(7) $$n_e = x_i n_{He} u(n_{He}),$$

where x_i is the initial ionization degree, considered as a control parameter, and $u(n_{He}) = (n_{He0}/n_{He})^{1/3}$ chosen in order to reproduce the experimental $\beta_s(n_{He})$ dependency [12] (see Fig. 2a).

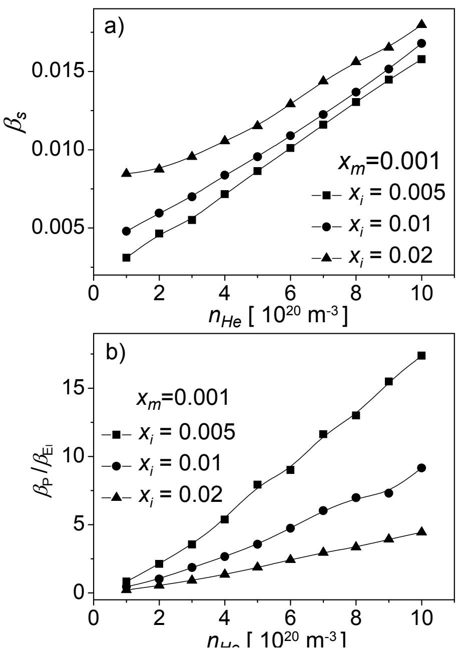

Fig.3. Hg ionization effciency as a function of n_e calculated for different values of ionization degree (a) and relative efficiency of Penning and electron impact processes (b)

Fig.2b shows the ionization efficiency for He atoms obtained in simulations for different x_i values using $3{\cdot}10^7$ of test particles.

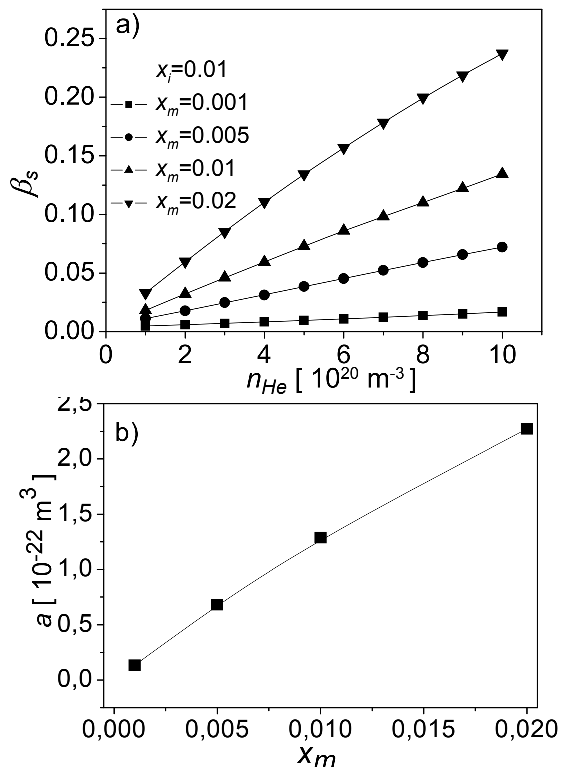

Fig.4. Hg ionization efficiency as a function n_e for different concentrations of the carrier gas atoms in the metastable state (a) and changes of the $\beta_S(n_{He})$ slope value with x_m (b).

A flat extraction electrode on the potential V_{ext}=1 kV was placed at the distance 2.5 mm from the extraction hole. The anode voltage was set to U_a=100 V. The simulation time step was Δt=2*10^{-8} s. One can see a good qualitative agreement with experimentally determined trends – simulation results follow also $n_{He}^{-1/3}$ curves.

As it was already mentioned Hg atoms could be ionized during both the electron impact and the Penning ionization. Probability of the latter process depends on the density of carrier gas atoms in the metastable state:

(8) $$n_m = x_m n_{He}$$

where x_m is another control parameter.

Looking at equations (5), (7) and (8), one may expect that the role played by the Penning ionization increases with the carrier gas density. This is confirmed by the results shown in Fig. 3. Calculations were done for x_m=0.001 using $6{\cdot}10^5$ test particles representing Hg atoms. In Fig. 3a one can see that for larger values of n_{He} ionization efficiency increases almost linearly, particularly in the case of smaller ionization degree (e.g. x_i=0.001). In such cases the Penning ionization is dominant – see also relative contribution of both processes shown in Fig. 3b. The Penning process contributes several times more strongly than the electron impact ionization even for moderate carrier gas concentrations. The effect of Penning ionization becomes more important for smaller values of x_i parameter (ionization degree). Ionisation degree could be related to such working parameters of the ion source as e.g. discharge voltage.

On the other hand, for smaller n_{He} and larger x_i the probability of electron impact ionization is comparable to that of Penning ionization and the discrepancy from the linear trend predicted by equations (5) and (8) is significant, see e.g. $\beta_s(n_{He})$ curve for x_i=0.02. In that case the β_P/β_{EI} ratio is smaller than 1 even for n_{He}= 3*10^{20} m^3.

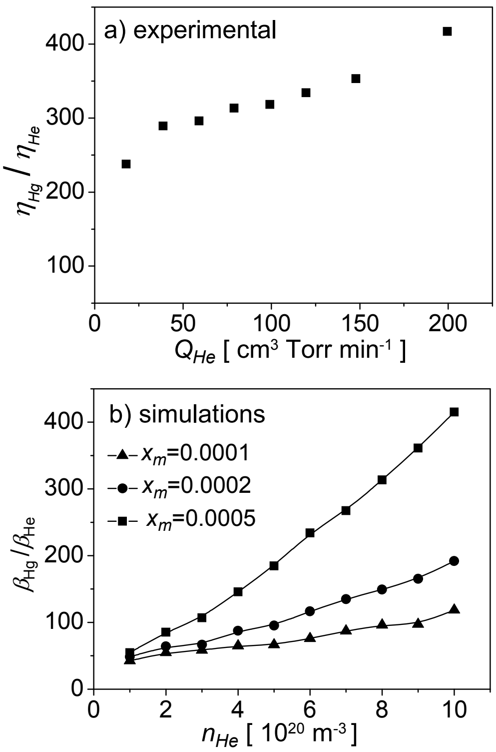

Fig.5. Relative efficiencies of mercury and helium ion production obtained in experiment (a) and in simulations (b).

Changes of the ionization efficiency with the density of the carrier gas atoms in the metastable state were also under investigation. Fig. 4 shows the results obtained for x_i =0.01 and x_m changing from 0.001 up to 0.2. The efficiency increases with x_m as could be expected from (8). The increase is almost perfectly linear in the considered range of parameters: The changes of the slope *a* are presented in Fig. 4b. The dependence of the slope on the concentration of He atoms in the metastable state is also almost linear which means that the Penning ionization is the dominant ionization process in the presented case.

Fig. 5 shows the relative efficiencies of the sample and the carrier gas ions obtained in both experiment and simulations. In both cases one observes a nearly linear increase of relative efficiency with the carrier gas amount. The experimental η_{Hg}/η_{He} values are higher for small Q_{Hg} than that obtained from simulations. The possible reasons for that are (a) larger than assumed in the model contribution from the electron impact ionization (b) more complex dependence of n_m on n_{He} than that assumed in Eq. (8).

Conclusions

A numerical model of ionization in the plasma ion source taking into account both electron impact and Penning ionization is presented in the paper. The shape of the ionization efficiency of carrier gas curves was reproduced by assuming that the electron density scales as ~$n_{He}^{2/3}$. It was also shown that according to the presented numerical model, the importance of Penning effect increases with the carrier gas density. The contribution from the Penning process could be even more than 10 times greater than that from the electron impact ionization (depending on the plasma ionization degree). The relative ioniation efficiency of the sample and thecarrier gas atoms was also studied - nearly a linear increase with the carrier gas atom concentration was observed which is in good agrrement with the experimental results. Using carrier gas and employing the Penning effect could be a very simple and effective way of improving performance of plasma ion sources used for nuclear spectroscopy, ion implantation, isotope separation etc.

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