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Ionization of short-lived isotopes in a hot cavity – Numerical simulations

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ABSTRACT

An extended Monte Carlo method based numerical model of the hot cavity ion source is presented. Not only the radioactive decay and delays due to the sticking of atoms to hot walls are taken into account, but also (i) delays due to the diffusion of nuclides, (ii) contributions from electron impact ionization, (iii) calculations of ion release curves and (iv) the case of hemispherical cavities are implemented. The code enables calculations of ionization efficiency for a broader range of parameters including ionizer material, size, geometry and temperature; extraction voltage; timescales characterizing radioactive decay, particle sticking, out-diffusion; and electron impact ionization cross-sections. The dependences of ion source efficiency on decay half-life, sticking time and diffusion timescale are shown and discussed. Two different schemes of radioactive nuclide generation are introduced and compared. Influence of ionizer length and extraction voltage is extensively studied. The importance of the electron ionization for short-lived isotope ion production is for the first time demonstrated. Two new analytical models of ionization in the case short-lived nuclides are introduced and compared to simulation results. Good agreement of experimental and simulated data (efficiency vs. temperature) is shown. New features enabling simulations of ion release curves are demonstrated.

1. Introduction

An efficient and reliable ion source is a key part of a variety of facilities that employ ion beams, including ion implanters; devices used in nuclear spectroscopy and medicine (particle therapy); systems for ion beam etching and sputtering; analytical tools like secondary ion mass (SIMS) or Auger electron (AES) spectrometers and many others. A multitude of ion source designs, characterized by different principles of operation and size, were invented over tens of years in order to fulfill particular requirements of scientists and engineers [1]. The hot cavity surface ionization source is one of those devices, especially useful for purposes of nuclear spectroscopy, experimental astrophysics and electromagnetic isotope separation. It was invented in 1970s and successfully used e.g. for finding new isotopes [2–5]. High efficiency of hot cavity ion sources and the fact that they require very small amounts of ionized substance in order to obtain a good quality ion beam made them useful for on-line separation of short-lived isotopes, created during irradiation of targets with high-energy particle beams [6–9]. The short time spent inside the ionizer by atoms could also be a very important factor in the case of studies short-lived isotopes. The other advantages to be mentioned are: compact design and reliability; very low energy spread of the ion beam and its high purity.

Thus, hot cavity ion sources still attract attention of e.g. scientists involved in different ISOL (Isotope Separation On-Line) activities [10–14]. Over forty years of design and optimization resulted in a variety of forms. The crucial part is, however, a semi-opened ionizer. It has usually the form of a tube, but the spherical ones were also used [15,16]. In the ion sources used in the ISOL facilities, the ionizer could be connected to a target by a transfer line [15,17], the target could be placed near the ionizer [13,18], or the ionizer itself is the irradiated target that releases newly created isotopes directly into the hot cavity [9,19]. Either positive or negative [15,20–23] ions are effectively produced, depending on the ionizer material work function. It should be mentioned here that a great effort was made in order to estimate usefulness of different classes of materials (carbides, oxides, nitrides, sulfides etc.) for the ISOL purposes [24]. A variety of low-work function materials, that would be suitable for negative ion production were also tested [13,15,21,25].

The classic surface ionization hot cavity ion sources evolved into the resonant ionization laser ion sources (RILIS) [26–31]. They base on a stepwise excitation of the valence electron to the continuum states using tunable lasers. As the excited state

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energies are specific for each element, RILIS ion sources are characterized by an excellent elemental selectivity. The hot cavity is used to store the atoms that are to be ionized by the laser beam from the system with a high repetition rate. It should be stressed here that surface ionization might be considered as the unwanted mechanism of ion production. The suppression of the background contaminants could be achieved e.g. by the proper choice of the cavity material [21] or bunching of the laser radiation produced ions by fast gates [27]. The coatings that reduce the surface trapping of desired atoms are developed [32] This paper, however, focuses on modeling of hot cavity ion sources with surface ionization.

Numerous explanations and theoretical models of ionization in a hot cavity were discussed over the years [33–38]. Most of them are based on the assumption that the atoms closed in the cavity may undergo multiple collisions with the hot wall, which results in ionization efficiencies much higher than that predicted by the Saha–Langmuir equation describing the single act of surface ionization [1]. Additionally, it is assumed that the intense electron emission from hot surfaces may lead to the creation of a potential trap inside the ionizer that prevents the recombination of ions during the collisions with walls. On the other hand, numerical simulations were rather rarely used to describe the processes occurring in ion sources like vapor transport from the irradiated target to the ionizer [39,40] or release of radioactive isotopes from the ionizer [41,42]. Up to our knowledge the ionization of short-lived isotopes in an ionizer being a target has not been studied yet using numerical simulations. Such approach enables optimization of ion source design as well as supports interpretation of experimental results [43,44].

In the previous papers a numerical model of hot cavity ion source was developed [45,46]. The model was based on the above mentioned assumption that multiple collisions of atoms with hot ionizer walls lead to high ion source ionization efficiency. Electron impact ionization could also bring an important contribution for high ionizer temperatures and in the case of hard-to-ionize elements [47]. The model enabled studies of ion source efficiency as a function of many factors, ionizer geometry (including the spherical one [48]), the properties of extraction system like the electrode geometry and voltage, ionizer temperature, ionizer/sample combination etc. However, the model was adequate mostly for stable or long-lived isotopes, as it did not take into account effects of radioactive decay of nuclides during their stay in the ion source, which restricted severely its usefulness in the field of nuclear spectroscopy. The early attempt to overcome that restriction was presented in the paper [49,58]: particles were assumed to undergo radioactive decay and the model of that process is implemented using the Monte Carlo method. The code took into account not only the time of flight of particles but also the time they spend on the surface of the ionizer (the sticking time). The numerical code presented in the current paper is a widely upgraded version of the previous one. It includes also the model of diffusion of nuclides out of the thin target inside the ionizer, which is of great importance, as the out-diffusion time in many cases could be greater than the nuclide half-life. Another important new feature is ion release curve simulation subroutines, giving the possibility of fitting hold-up experiment results with that of numerical calculations. The upgraded numerical model enables also taking into account electron impact ionization due to the thermionic electrons accelerated by the extraction voltage. It should be mentioned that another upgrade, which allows simulations of the ion sources with the hemispherical ionizer, was made. Hence, the code enables studies of the changes of efficiency with the ionizer and extraction system geometry, temperature and the other above mentioned parameters also for short-lived nuclides.

The paper includes the description of the numerical model used for simulations as well as its physical background. As the first, the influence of the half-life on the efficiency of the ionization in the ion source was under investigation. The effect of the average sticking time length was also checked out. Simulations were also done for different lengths of the ionizer in order to determine the influence of that factor on the ion source efficiency of short-lived isotopes in a much more comprehensive way than in Ref. [49] – two different schemes of the distribution of new nuclides in the ionizer/target are considered. Current–voltage characteristics were determined both for the case of short (compared to the intrinsic delay time) and moderate values of the half-life time. Another important factor that has great impact on the ion source efficiency is the characteristic timescale of the diffusion of the freshly produced nuclides out of the target/ionizer. Some experimental results, as the changes of ion source efficiency with the temperature of the ionizer were compared with the data obtained from the numerical simulation. Two new analytical models enabling estimation of the hot ion source efficiency in the case of radioactive elements, were introduced and their predictions were discussed. The models are based on that presented in Ref. [38] but take into account losses due to the radioactive decay. The predictions of the analytical models are compared the simulation results in the case of pure intrinsic delay process as well in the case taking into account the sticking of atoms to the hot surface. The influence of electron impact ionization on the ion source efficiency was also under investigation, dependences of relative contribution from this process on the ionizer temperature as well as the nuclide half-life were considered. The presented numerical code can also provide release curves – the examples of calculated release profiles for different values of the half-life time, the average sticking time as well as the characteristic diffusion time are included.

2. Ionization, effusion and diffusion in a hot cavity

The hot cavity ion source is a relatively simple device. Its main part is a tube (sometimes semi-opened) made most often of refractory metal (those having high work function are especially suitable for the production of positive ions). As it was already mentioned, a variety of low work function materials e.g. LaB_6, GdB_6, Ir_5Ce, as well as oven-dosed vapors was applied for efficient generation of negative ions [15,20,23]. The ionizer is heated to a high working temperature T (~2500–3000 K) either ohmically or by electron beams. In the on-line facilities, the ionizer is either connected to the target by a transfer line, or even the ionizer wall could serve as the target.

Diffusion in the irradiated target, effusion and ionization on the hot surface of the ionizer are the crucial physical processes occurring in a hot cavity ion source [50]. After the irradiation of the target with a proton or heavy ions new nuclides (including radioactive ones) are created. The product particles start to diffuse out of the target. The process of diffusion is described by the second Fick's law:

$$\frac{dc}{dt} = D\frac{d^2c}{dx^2}, \tag{1}$$

where c is the concentration of the dopant (sometimes referred to as the tracer) and D is the diffusion coefficient. The function describing the fraction of tracer leaving the target after some time t could be found in e.g. Ref. [51]. The rate of diffusion depends on the geometry of the target and on the values of the diffusion coefficient, varying in the range from 10^{-7} up to 10^{-12} cm^2/s for the most common tracer/host combinations [52]. It should be mentioned