Development of Hydrogen Pair-Ion Source on the Basis of Catalytic Ionization

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Abstract. To develop a hydrogen pair-ion source comprising only H^+ and H^- ions, the efficient production of pair ions is required. When discharged hydrogen plasma is used for positive-ion irradiation to a Ni catalyst, the pair ions are produced from the back of the irradiation plane. The number of pair ions produced increases proportionally with the irradiation flux.

Keywords: hydrogen pair ions, catalytic ionization, porous catalyst
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INTRODUCTION

Negative-ion production mechanisms can be classified in terms of the electron source [1]. In the surface effect, an electron at the Fermi level in the conduction band of a metal shifts by tunneling to the electron affinity level of an atom or a molecule approaching the metal surface. The probability of the electron shift is enhanced as the effective work function of the metal surface decreases. The electron shift occurs in particle reflection and sputtering phenomena. Cesium has the lowest work function of all elements. A small admixture of cesium vapor in a hydrogen discharge significantly improves H^- production and decreases the current of coextracted electrons [2-4]. However, the use of cesium complicates ion source operation and requires a careful stabilization of cesium injection and discharge parameters.

There have been many attempts to develop H^- sources with acceptable H^- beam emittance but without a cesium admixture. Plasma electrons are the source of electrons in volume production [5,6]. A highly vibrationally-excited hydrogen molecule effectively captures a low-energy plasma electron to form a H^- ion through dissociative electron attachment. However, there is the problem of a relatively low current density of H^- ions in volume production.

Hydrogen atomic pair ions, H^+ and H^- ions, are the lightest ions and have high response frequencies to electromagnetic fields. To generate a hydrogen pair-ion plasma consisting of only the pair ions [7-9], the production of equal quantities of H^+ and H^- ions and the absence of impurities such as electrons and other ions are required [10]. It is difficult to satisfy these requirements in surface production with a cesium admixture or in volume production. In order to solve this difficulty, we have suggested a catalytic ionization method with plasma assistance [11].

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In our previous work, a Penning ionization gauge (PIG) discharge plasma was used for positive-ion irradiation to a porous catalyst [11]. The both irradiation energy and flux of positive ions depend on the discharge power (anode voltage). The irradiation flux should be controlled independent of the irradiation energy. In this paper, initial results of the operation of an dc arc discharge source with a nickel (Ni) porous catalyst are discussed.

EXPERIMENTAL APPARATUS

To produce the hydrogen pair ions, a hydrogen plasma is generated by a dc arc discharge in a rectangular chamber of 25 cm×25 cm in cross section and 19 cm in length. A schematic diagram of the experimental setup is shown in Fig. 1 (a). The apparatus for producing the pair ions comprises mainly a discharge section for the irradiation and a pair-ion production section. Four horseshoe tungsten filaments of 0.7 mm in diameter and 15 cm in length are installed on the side walls of the chamber. The filaments are the thermal cathode biased at a discharge voltage V_d and the chamber walls are the grounded anode. The plasma discharged is efficiently confined by line-cusp magnetic fields adjacent to the chamber walls which are applied by permanent magnets. A commercially available Ni porous plate (Celmet, Sumitomo Electric Toyama Co., Ltd.) acts a catalyst with a porous body of 56-60 cells/inch, a pore size of 0.45 mm, a thickness of 1.4 mm, a specific surface area of 5,800 m^2/m^3, and a porosity of 96.6 %. The porous catalyst is biased at $V_{pc} = -600$ V here and the irradiation current I_{ir} is measured. The circular irradiation area is 19.6 cm^2 (diameter of 5 cm) and the other electrode is covered with a mica plate. The catalyst is located at $z = 0$ cm, the discharge section is in $z < 0$ cm, and the pair-ion production section is in $z > 0$ cm. Plasma parameters are measured using Langmuir probes at $z = -7$ cm and 3 cm. The operating pressure in the source is about 0.2 Pa.

The surface production mechanism of the pair ions on the porous catalyst is considered to be completely different from the conventional mechanism of surface production with a cesium admixture which is based on the electronic transition of metals with low work functions. Hydrogen atoms, produced by dissociative adsorption, are covalently bound with the surface-metal atoms, and can easily move along the surface, i.e., surface migration occurs. Surface migration on catalysts is a well-known fundamental phenomenon. Hydrogen atoms migrate along the pore surface in the porous catalyst to the back of the irradiation plane. An electronic transition occurs between the surface-metal atoms and hydrogen atoms during desorption from the surface if hydrogen atoms are provided a sufficient energy. Desorption ionization from solid surfaces in the gaseous phase is known to occur in laser desorption/ionization. In our previous experiment, the pair ions are clear not to be produced without the irradiation even though the porous catalyst is additionally heated. The irradiation supplies the energy required for desorption ionization; the thermal energy is too small for desorption ionization to occur. A part of kinetic energy of positive ions irradiated will be transferred to the back of the irradiation plane owing to the migration of hydrogen atoms. If two shared electrons transfer to the surface atoms during desorption, H^+ ions are produced. H^- ions, on the other hand, are produced if the

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electrons transfer to hydrogen atoms. The work function of Ni (5.15 eV) is relatively high, the contact-ionization probability of H^- ions is infinitely close to zero. The catalytic activity for hydrogen is important here, whereas the work function does not affect the ionization. It is known that negative ions are produced from the irradiation plane in the conventional converter-type production, but fast electrons and fast positive ions promote the collisional detachment. It is highly beneficial that the pair ions are produced from the back of the irradiation plane, because the negative-ion production from the back can prevent the collisional detachment. We refer to this process involving dissociative adsorption, surface migration, and desorption ionization as catalytic ionization, which is schematically illustrated in Fig. 1 (b).

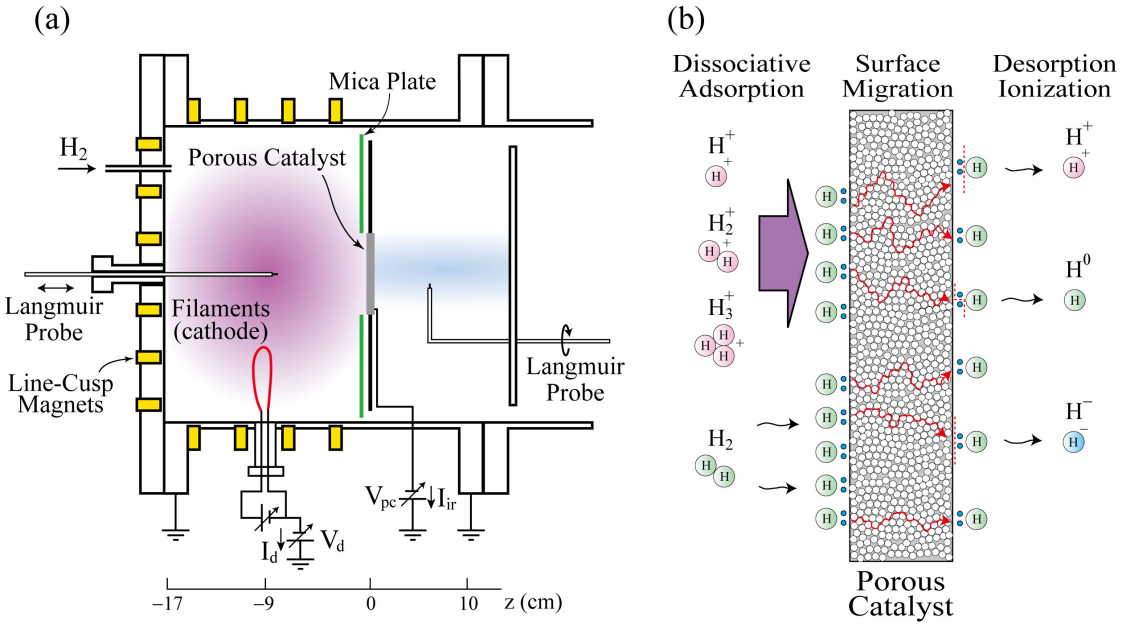

FIGURE 1. (a) Diagram of experimental setup for producing hydrogen pair ions on the basis of catalytic ionization. (b) Process of catalytic ionization under the positive-ion irradiation, which comprises dissociative adsorption, surface migration of hydrogen atoms, and desorption ionization.

RESULTS

The discharged plasma in $z < 0$ cm is used to irradiate positive ions to the Ni porous catalyst. Electron density n_e and plasma potential ϕ_s at $z = -7$ cm are measured. The dependence of n_e and the irradiation energy of positive ions $e(\phi_s - V_{pc})$ on the discharge power of $P_d = V_d \times I_d$ is shown in Fig. 2, where I_d is the discharge current. Positive ions are accelerated up to $e(\phi_s - V_{pc})$ (eV) in the sheath formed in front of the porous plate. V_{pc} is fixed at −600 V here and ϕ_s is varied in +10~20 V with P_d, but the variation width is narrow with respect to V_{pc}. The irradiation energy is almost constant independent of P_d. The irradiation current (flux) of positive ions I_{ir}, not shown here, increases proportionally with n_e. Therefore, the irradiation flux can be controlled by P_d carrying on the constant irradiation energy.

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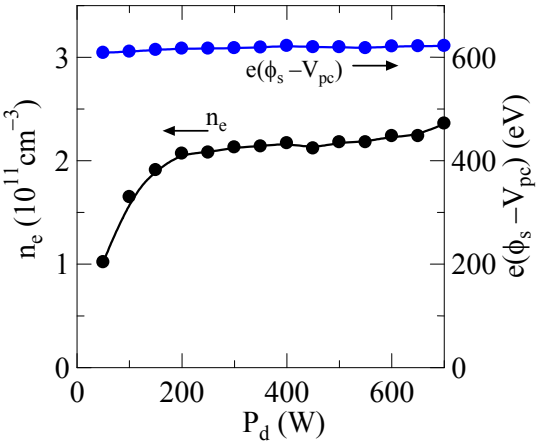

FIGURE 2. Dependence of the electron density and the irradiation energy of positive ions on the discharge power in the discharge region.

Under the irradiation, the pair ions are produced and an ionic plasma is generated in $z > 0$ cm. Typical current (I_p)-voltage (V_p) characteristics of the single probe at $z = 3$ cm are shown in Fig. 3. The I_p-V_p characteristics are symmetrical with respect to the ground voltage and zero current. The plasma potential indicated by a voltage of inflexion point almost coincides with the floating potential. The plasma potential is almost 0 V which is equal to the chamber wall. The positive- and negative-saturation currents of the characteristics are shown to be approximately equal. If electrons measurably exist in the plasma, the negative current would be higher than the positive current. Thus the existence of electrons can be negligible.

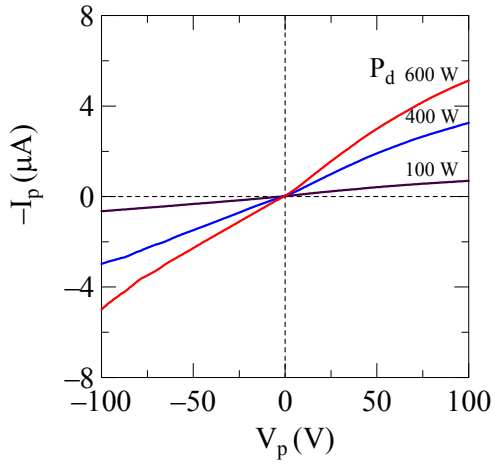

FIGURE 3. Typical I_p-V_p characteristics in an ionic plasma in $z > 0$ cm.

The positive- and negative-saturation currents of the probe, I_+ and I_-, are obtained at probe bias voltages of $V_p = -120$ V and +120 V, respectively. $I_\pm$ and the irradiation current I_{ir} depending on P_d are shown in Fig. 4. Since the porous catalyst and the thermal cathode are biased at $V_{pc} = -600$ V and $V_d = -70$ V, respectively, electrons cannot reach to the porous catalyst and positive ions are only irradiated. I_- appears to be the only negative-ion current. The current ratio I_-/I_+ is close to 1, indicating an

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ionic plasma comprising only H^+ and H^- ions without electrons. Both $I_\pm$ increase proportionally with I_{ir}; that is, the number of pair ions produced increases proportionally with the irradiation flux. The production quantity of H^+ ions from the catalyst surface will be bigger than that of H^- ions in the case of using the Ni porous catalyst, but the obtained $I_\pm$ are almost the same. The flow of H^+ ions toward downstream appears to be restricted to maintain quasi-neutrality in the plasma by the potential structure formed in front of the catalyst. The production balance between H^+ and H^- ions is mainly attributable to the catalyst material. In solid state properties, electronegativity describes the ability of an atom to attract electrons toward itself, which has an influence on desorption ionization. The electronegativities of Ni and H are 1.8 and 2.2 in Pauling units, respectively. In order to produce H^- ions more, the electronegativity of catalyst had better be lower.

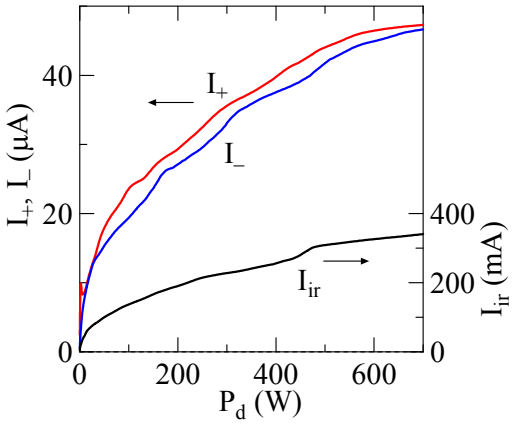

FIGURE 4. Production properties of H^+ and H^- ions and irradiation current of positive ions as function of the discharge power.

SUMMARY

A Ni porous catalyst is used to produce hydrogen atomic pair ions, i.e., H^+ and H^- ions. When hydrogen positive ions produced by a dc arc discharge are irradiated to the porous catalyst, the pair ions are produced from the back of the irradiation plane. We refer to the production mechanism as catalytic ionization. The irradiation flux is controlled by the discharge power maintaining the constant irradiation energy. The number of pair ions produced increases proportionally with the irradiation flux.

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