*35th EPS Conference on Plasma Phys. Hersonissos, 9 - 13 June 2008 ECA Vol.**32D**, P-5.147 (2008)*

Surface production of hydrogen negative ions using catalyst

W. Oohara, O. Fukumasa

Graduate School of Science and Engineering, Yamaguchi University, Ube 755-8611, Japan

Introduction

Pair plasmas consisting of only positive- and negative-charged particles of equal mass have attracted special attention because of their particular physical properties. Instead of electron-positron pair plasmas, a fullerene pair-ion plasma has been generated and its collective phenomena have been investigated [1]. The response frequency of the pair-ion plasma is limited to the narrow frequency range below 50 kHz because of the massive ions. In order to investigate more detailed physical properties of pair plasmas and wave-propagation characteristics up to higher frequency, development of a hydrogen pair-ion plasma source is started [2], because atomic hydrogen ions are the lightest ions and have high response frequencies to electromagnetic fields. Several difficulties exist in the development, and the most crucial problem is an efficient production of hydrogen negative ions H^-. The production method of H^- is classified into a surface production and a volume production, and cesiated multicusp type is the most successful ion source at present. From the possibility of impurity interfusion, alkali metals with low evaporation temperature cannot be used here. In this paper, it is described that H^- can be produced efficiently by using a catalyst.

Apparatus

A schematic diagram of the experimental arrangement is illustrated in Fig. 1. The plasma source mainly consists of an annular PIG (Penning Ionization Gauge)-discharge part and a negative-ion production part. Two annular cathodes are oppositely located in a cylindrical anode

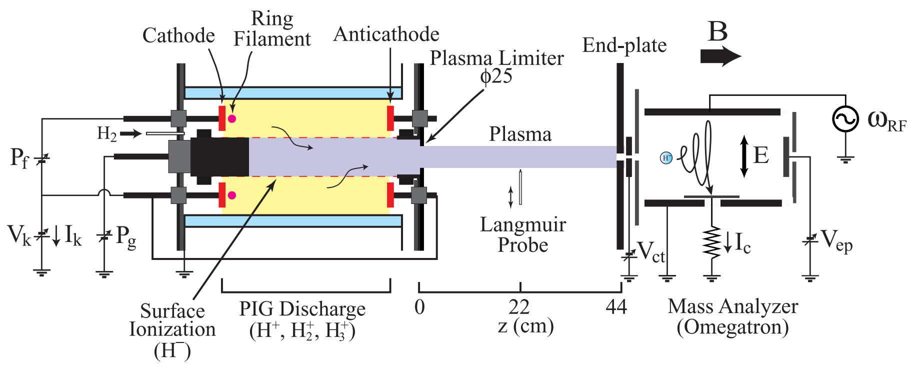

Figure 1: Schematic diagram of experimental setup.

with an inner diameter of 7.5 cm and a length of 15 cm in a uniform magnetic field (B = 65 mT), and a tungsten ring-filament (diameter of ϕ0.5 mm) with the same voltage as the cathodes is set in front of one of the cathodes to supply thermal electrons. The thermal electrons are accelerated in a sheath formed in front of the cathode and injected into the space between the two cathodes, and the electrons are reflected in the sheath in front of the opposite cathode (anticathode) because the same voltage V_k is applied to the cathodes. Since the beam electrons are electrostatically confined between the two cathodes along the B-field lines, neutral particles can be ionized efficiently by electron impact, and positive ions H^+, H_2^+, and H_3^+ are produced. In normal operation, the discharge current I_k is 2A, the cathodes are biased at $V_k \simeq -140$ V with respect to the grounded anode. The back pressure of the vacuum system is 1×10^{-4} Pa and the operating pressure in the source is about 2×10^{-1} Pa. During operation a continuous flow of hydrogen is maintained (20 SCCM).

The surface production on a warm Cs-coated plate with a low work function ($<$ 2.1 eV) is known to be efficient. But the H^- production is attempted on a cylindrical grid of nickel (Ni) expected as a catalyst, which is set in the center of the cylindrical anode. Positive ions and neutral hydrogen interiorly diffuse from the annular discharge region to the central region through the grid. They contact and react with the grid surface in the process of the diffusion. The diffused plasma flows to the downstream and terminated at an end-plate (z = 44 cm) with a center aperture of 1 mm diameter. An omegatron situated behind the end-plate is an rf mass spectrometer [3]. In the omegatron, an rf electric field is applied in the direction perpendicular to the B-field lines, and the ions entering from the aperture are excited at their various cyclotron resonances, causing their orbital radii to increase; the ion current I_c induced when they strike a collector set at an off-axis position of the aperture is measured. A spectrum of the ion current as a function of the rf frequency $\omega_{RF}/2\pi$, i.e., a mass spectrum is obtained by slow sweep of the frequency. The plasma parameters are measured by a Langmuir probe at z = 22 cm from the source.

Results

A steady-state hydrogen plasma is generated by the PIG discharge. Radial profiles of the plasma without and with the Ni grid are shown in Figs. 2 and 3, respectively. I_+ and I_- indicate positive and negative saturation currents of the probe, where the probe collector is biased at $V_p = -80$ and 80 V, respectively. I_+ is proportional to the positive-ion density n_+, i.e., the plasma density. I_- is a total of the electron and negative-ion saturation currents, and its value substantially changes depending on the negative-ion density n_-. ϕ_f indicates a floating potential of the probe. Two dashed lines at $|r|$ = 1.25 cm denote the inner diameter of the plasma limiter

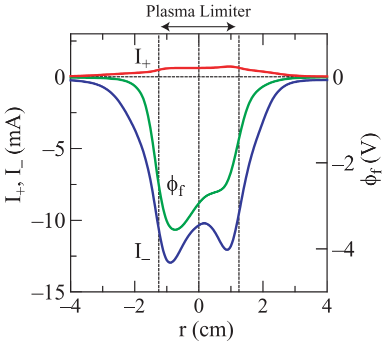

Figure 2: Radial profiles of the plasma without grid.

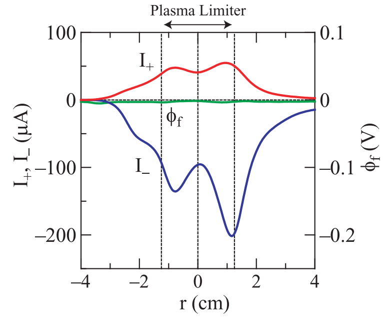

Figure 3: Radial profiles of the plasma with Ni grid.

(ϕ2.5 cm) which is set at the exit of the source (z = 0 cm).

In the case without the grid, the electron temperature and the space potential at r = 0 cm are obtained $T_e \sim 5$ eV and $\phi_s \sim +20$ V from the probe characteristic. Since the temperature is relatively high ($>$ 1 eV), it seems that negative ions are not produced by the volume production and the plasma only consists of electrons and positive ions. Then the electron density of $n_e \sim 3 \times 10^{10}$ cm^{-3} is obtained from the characteristic, and Debye length is about 100 μm here. The saturation current ratio observed is $I_-/I_+ \sim 17$. Ideal current ratios without negative ions are calculated as I_-/I_+ = 28 (H^+–e^- plasma) and 49 (H_3^+–e^- plasma), but the actual measured ratios are known to become lower than the ideal ratios.

The plasma density decreases in the case with the grid. The grid of Ni 200 mesh (wire diameter of ϕ0.05 mm, open area of 37 %) is used and the opening is 77 μm and narrower than the Debye length, thus, the plasma density through the grid decreases substantially. The density is calculated as $n_+ \sim 2 \times 10^9$ cm^{-3} at r = 0 cm. The current ratio observed is $I_-/I_+ \sim 2.4$. The negative saturation current is denoted as $I_- = 0.25en_+S((1-\varepsilon)v_e + \varepsilon v_-)$ from conventional probe theory, where ε is an exchange rate of n_-/n_+, S is the collector area of the probe (3 mm^2), v_e and v_- are thermal velocities of electrons and negative ions, respectively. The electron density cannot be obtained from the probe characteristic when the exchange rate is unclear. The electron temperature seems to be low because visible emission from the plasma is not observed in the downstream ($z > 0$ cm). The exchange rate is calculated ε = 0.58 – 0.81 (supposition with T_- = 0.2 eV) in T_e = 1 – 5 eV. H^- destruction cross sections [4] are $\sigma \sim 10^{-16}$ cm^2 (e^- + H^- $\rightarrow$ H + 2e^-) and 10^{-13} cm^2 (H^+ + H^- $\rightarrow$ 2H), and mean free paths of H^- are in the order of 10^7 cm and 10^3 cm, respectively. Thus, the negative-ion density in $z < 0$ cm seems to be almost the same as it at z = 22 cm. The structure which the plasma diffuses through the grid across the B-field lines is a kind of a magnetic filter, and negative ions produced on the surface are

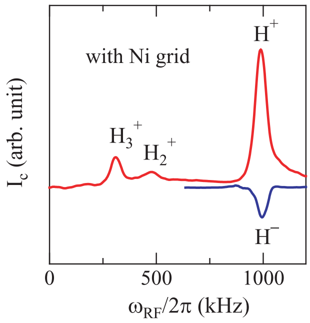

Figure 4: Mass spectra of the omegatron with Ni grid.

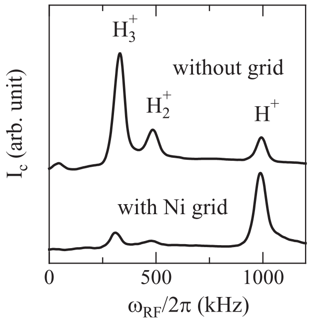

Figure 5: Mass spectra in the case of without and with Ni grid.

spatially separated from high energy electrons in the discharge region. In this point, the source is different from conventional negative-ion source of coverter type [5].

The ion species of the plasma are analyzed by the omegatron at B = 65 mT. Typical mass spectra with the grid are shown in Fig. 4. The ion cyclotron frequency of H^+ is calculated to be $\omega_c/2\pi$ = 987 kHz. Positive and negative peaks at around $\omega_c/2\pi$ in the spectra indicate the existence of H^+ and H^-, respectively. Peaks at the one-half and the one-third frequencies of $\omega_c/2\pi$ indicate the existence of H_2^+ and H_3^+, respectively. The mass spectra of positive ions without and with the grid are shown in Fig. 5. The species ratio without the grid is $H^+ : H_2^+ : H_3^+$ = 1 : 1.7 : 6.3 (the collector current ratio 1 : 1.2 : 3.6), and H_3^+ is the dominat species. The dominant species is well known to be H_3^+ in ordinary hydrogen discharge plasmas, and the tendency without the grid is the same. On the other hand, the species ratio with the grid is $H^+ : H_2^+ : H_3^+$ = 1 : 0.2 : 0.4 (the current ratio 1 : 0.1 : 0.2), and H^+ is the dominant species. Molecular hydrogen including ions dissociates into atomic hydrogen and absorbs on catalyst surface. Resonant electronic transition between the surface metal and the atoms in their desorption, i.e., a desorption ionization is caused, and H^+ and H^- are produced consequently. Therefore the atomic ions are mainly produced by the surface interaction, not by electron impact/attachment.

References

[1] W. Oohara and R. Hatakeyama, Phys. Rev. Lett. **91**, 205005 (2003); Phys. Rev. Lett. **95**, 175003 (2005); Phys. Rev. E **75**, 056403 (2007)

[2] W. Oohara and R. Hatakeyama, Phys. Plasmas **14**, 055704 (2007)

[3] H. Somer, H. A. Thomas and J. A. Hipple, Phys. Rev. **82**, 697 (1951)

[4] K. Prelec and Th. Sluyters, Rev. Sci. Instrum. **44**, 1451 (1973)

[5] K. N. Leung and K. W. Ehlers, J. Appl. Phys. **52**, 3905 (1981)