

Efficient production of negative hydrogen ions in RF plasma by using a self-biased grid electrode

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ABSTRACT:

Volume production of negative hydrogen ions is established efficiently in a pure hydrogen RF discharge plasma by using a self-biased grid electrode for production of low electron-temperature and high density plasma. Using this electrode both high and low electron temperature plasmas are produced in the regions separated by the grid electrode in the chamber, in which the electron temperature in the downstream region is controlled by the mesh size and plasma production parameters. The production rate of negative ions depends strongly on the electron temperature varied by the RF input power and hydrogen pressure. In the case of the grid electrode with the 5 mesh/in., the negative hydrogen ions are produced effectively in the downstream region in the hydrogen pressure range of 0.9 – 2.7 Pa. In addition, the production rate of the negative ion H^- raises from 62 % to 87 % at 0.9 Pa by changing the RF power from 20 W to 80 W.

Keywords: negative hydrogen ion, self-biased grid electrode, capacitively-coupled RF discharge, low electron temperature.

I. INTRODUCTION

In reactive plasmas widely used for material processing, electron energy distribution would give a big effect on the products obtained, because excitation and desociation are determined by the electron energy. When electrons distribute appoximately like Maxwellian, the average electron energy is expressed by the electron temperature T_e . Therefore, the control of T_e is of crucial importance for finding the best conditions of the chemical reactions necessary for the material processing [1-4]. In general, however, it is difficult to control T_e in weakly-ionized discharge plasmas, although several methods have been reported for the T_e control [1-9].

In our method using a mesh-grid [5], T_e is decreased by almost two orders of magnitude, being accompanied by an increase in electron density n_e . The grid-bias method, applied to RF silane plasma, revielded a good reproducible yield of hydrogenated amorphous silicon (a-Si:H) films with high drift mobility [10]. We also applied this method to negatve-ion production in RF hydrogen plasma, where the negative hydrogen ion production was enhanced by the decrease in T_e [11]. In the case of RF discharge plasma, the grid bias method was found to be quite suitable for optimizing the plasma conditions for negative ion production, compared with the magnetic filter method [12, 13].

It is pointed out that the grid-bias method is inappropriate for reactive plasmas, in which grid is often covered by thin insulating films like

hydrogenated amorphous and diamond-like carbon films. However, we have already demonstrated that the grid bias method can still control T_e over a wide range [9]. In fact, high-quality diamond was produced in RF discharge CH_4/H_2 plasmas [14], where T_e was decreased by varying mechanically the length of slits in plasmas [8]. In the case of Argon plasma, T_e was widely controlled by almost one order of magnitude by changing the mesh size of the grid electrode.

It is well known that negative hydrogen ions H^- are generated by the next two stages. In the first stage, high energy electrons e_h with T_e more than 20 eV excite H_2 molecules to generate vibrationally excited molecules $H_2^*(v)$ via the reaction $H_2 + e_h \rightarrow H_2^*(v) + e$. In the second stage, low energy electrons e_l with $T_e \sim 1$ eV collide with $H_2^*(v)$ for the production of H^- via dissociative attachment collisions reaction $H_2^*(v) + e_l \rightarrow H^- + H$ [15]. In the second stage, however, very low energy electrons with $T_e < 0.5$ eV have too small energy to cause the above reaction effectively [16]. Note that negative ions are easily destroyed by the collisions with warm electrons. Therefore, it is necessary to generate e_h and e_l in separate regions, and keep them isolated each other. Our device using the self-biased electrode is very suitable to generate negative ions efficiently because e_h and e_l are isolated in two regions by the self-biased grid electrode.

In this paper, we propose a new method using a self-biased grid electrode for a production of the RF discharge hydrogen plasma. Here,

negative hydrogen ions are produced by controlling T_e in a steady-state capacitively-coupled RF plasma [17]. By using a grid electrode with mesh size of 5 mesh/inch, we have succeeded in generating H^- ions efficiently in the RF plasma under the H_2 pressure range of 0.9 – 2.7 Pa.

II. EVALUATION OF NEGATIVE ION DENSITY

First, let us estimate negative ion density n^- by measuring negative and positive saturation currents I^- and I^+ of a Langmuir probe in hydrogen plasmas. Here, we assume that the ions of positives and negatives are monovalent ions, H^+ and H^- , respectively, as confirmed experimentally below. In addition, by supposing that M_+ , M_- , T_- and T_e are mass of positive ion, mass of negative ion, temperature of negative ion, and electron temperature, respectively, the saturation currents I^- and I^+ are expressed by the next equations [18-20].

$$I^- = eS \left[n_e \sqrt{\frac{kT_e}{2\pi m_e}} + n^- \sqrt{\frac{kT_-}{2\pi M_-}} \right] \quad (1)$$

$$I^+ = eSn^+ \sqrt{\frac{kT_e}{2.72M_+}} \quad (2)$$

Here, the factor 2.72 is the Napier's constant, coming from a density difference between the sheath edge and the plasma region. From Eqs. (1), (2) and $n^+ = n_e + n^-$ (Condition of the electric charge neutrality) one can obtain the next equation.

$$\frac{I^-}{I^+} = \sqrt{\frac{2.72M_+}{2\pi m_e}} \left[1 - \frac{n^-}{n^+} \left(1 - \sqrt{\frac{m_e T_-}{M_- T_e}} \right) \right] \quad (3)$$

Here, by using $T_- \ll T_e$ and $m_e \ll M_-$, the last term in Eq. (3) can be neglected. Therefore, the density ratio of negative to positive ions n^-/n^+ is given by the next expression.

$$\frac{n^-}{n^+} \cong 1 - \frac{I^-}{I^+} \sqrt{\frac{2\pi m_e}{2.72M_+}}$$

By using positive hydrogen-atom ion mass M_+ , one can obtain the next expression

$$\frac{n^-}{n^+} = 1 - 0.0348 \times \frac{I^-}{I^+} \quad (4)$$

In the case that different positive hydrogen ion species (H^+ , H_2^+ and H_3^+) are produced, M_+ must be replaced by weighted average mass M_{av} [21]. However, in the case of H_2 plasma as in our experiment, the majority of positive and negative ions are H^+ and H^- , respectively [See Fig. 13 of Ref. 21]. From Eq. (2) positive ion density $n_i = n^+$ is expressed as follows with a use of $I^+ = I_{is}$.

$$\begin{aligned} n_i = n^+ &= \frac{I^+}{eS} \sqrt{\frac{2.72M_+}{kT_e}} \\ &= 1.50 \times 10^{11} \frac{I_{is}}{\sqrt{T_e}} \text{ [cm}^{-3}\text{]} \end{aligned} \quad (5)$$

where the units of I_{is} and T_e are mA and eV, respectively. Probe surface area is $S = 0.07 \text{ cm}^2$.

III. EXPERIMENTAL SETUP

Figure 1 shows a schematic of the experimental apparatus. A rectangular electrode with a mesh grid shown in Fig. 2 is placed on a side wall of a grounded cubic chamber of $15 \times 15 \times 15 \text{ cm}^3$ in volume. The cubic chamber is installed in a stainless steel vacuum chamber of 60 cm in diameter and 100 cm in length, which is evacuated to a pressure of 0.3 Pa by using a rotary pump. Back and side surfaces of this electrode are made of aluminum plates with thickness of 0.5 mm, which are covered with aluminum plates for an electric shielding. Space of 3 mm exists between the aluminum electrode and the shielding plates. The back area and height of this electrode are $5 \times 5 \text{ cm}^2$ and 2 cm, respectively. The mesh electrode of 5 mesh/inch is made of stainless steel wires of 0.5-mm-diam. The area of the mesh grid is 25 cm^2 . Hydrogen plasma is generated by a 40 MHz power source in pressure range of 0.7 ~ 3.7 Pa. RF power P_{RF} is supplied to the electrode directly through a blocking capacitor (BC) in a range of 20 – 80 W. As a result, self-bias voltage is induced on the electrode [22, 23]. If necessary, Argon gas is used for obtaining basic parameters of plasma.

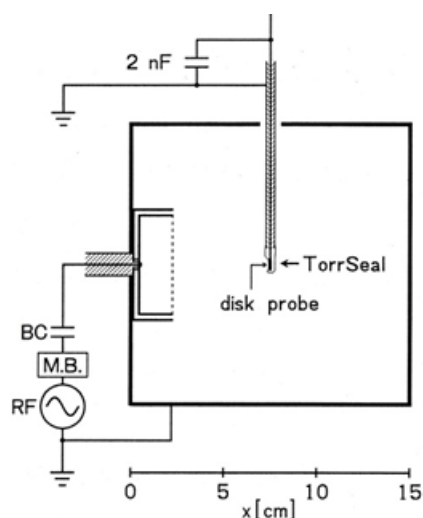


Figure 1. Schematic of the experimental apparatus for an efficient production of negative hydrogen ions in RF (= 40 MHz) discharge plasma.

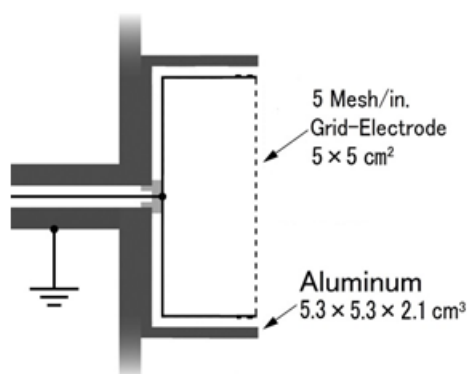


Figure 2. Details of the rectangular self-biased RF electrode with a mesh grid.

Plasma parameters are measured by a disc probe of 3 mm in diameter. One side of the probe is used as a collector of the probe current, and the other surface is covered with Torr Seal which is a sealant made of low vapor pressure resin. Therefore, the surface area of the probe is equal to 0.07 cm². The values of the dc probe current I_p and voltage V_p collected through Source Meter-2400 manufactured by Keithley Instruments are stored on a personal computer, and at the same time, a probe characteristic curve is drawn automatically

on a display of the computer using the software that we have created using Microsoft Visual C++. The measurement method of plasma parameters mentioned above is explained by Ref. [24] in detail. The origin of x -axis is set at the wall ($x = 0$ cm) of the cubic-box chamber, as shown in Fig. 1. The probe is fixed at the center of the cubic chamber ($x = 7.5$ cm). In the case of Argon gas, plasma parameters generated by using electrode shown in Fig. 2 are almost the same as those produced by a grid-cage electrode [16].

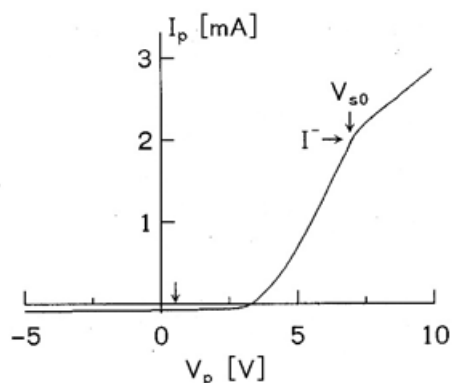


Figure 3. Time-averaged $I_p - V_p$ probe characteristic curve in H₂ plasma. P_{RF} and P_{H_2} are 20 W and 3.6 Pa, respectively. $x = 7.5$ cm.

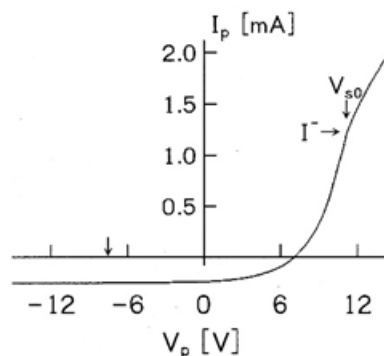


Figure 4. Time-averaged $I_p - V_p$ probe characteristic curve in H₂ plasma. P_{RF} and P_{H_2} are 60 W and 0.9 Pa, respectively. $x = 7.5$ cm.

IV. EXPERIMENTAL RESULTS

Time averaged probe $I_p - V_p$ characteristic curve for pure hydrogen plasma is shown in Fig. 3 in the case of RF power $P_{RF} = 20$ W and pressure $P_{H_2} = 3.6$ Pa. The mesh size and

wire diameter of the electrode are 5 mesh/inch and 0.5 mm, respectively. From a semi-log plot of this electron current I_e , the space potential $V_{s0} = 7.0$ V, electron saturation current $I_{es} = 2.05$ mA, electron temperature $T_e = 0.31$ eV, and electron density

$n_e = 2.1 \times 10^{10} \text{ cm}^{-3}$ are derived. The method for obtaining these parameters has been already reported in Ref. [24]. The electron temperature $T_e = 0.31 \text{ eV}$ is too low to evolve the process of $\text{H}_2^*(\text{v}) + e_l \rightarrow \text{H}^- + \text{H}$ [15]. Therefore, negative hydrogen ions are supposed not to be produced. In this case, negative component of the probe current $I^- = 2.02 \text{ mA}$ seems to express the electron saturation current I_{es} . With a use of the ion current

$I^+ = 0.07 \text{ mA}$ at 0.52 V , indicated by a downward arrow in Fig. 3, one can obtain $I^-/I^+ = 2.02/0.07 = 28.9$. Here, I^+ is the positive ion saturation current I_{is} . We obtain $n^-/n^+ \approx 0$ from eq. (4). This means that almost no negative hydrogen ions are generated in such a very low electron temperature of 0.31 eV in Fig. 3.

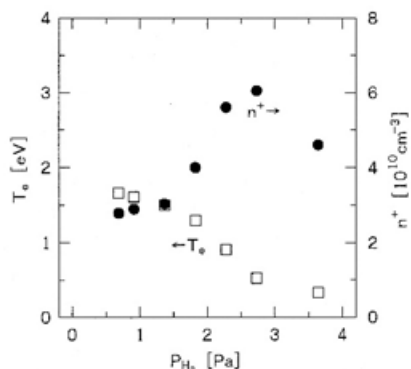


Figure 5. Variations of T_e and n^+ as a function of hydrogen pressure P_{H_2} at $P_{\text{RF}} = 60 \text{ W}$. $x = 7.5 \text{ cm}$.

Time averaged probe $I_p - V_p$ characteristic for pure hydrogen plasma in the case of $P_{\text{RF}} = 60 \text{ W}$ and $P_{\text{H}_2} = 0.9 \text{ Pa}$ is shown in Fig. 4. From a semi-log plot of this electron current I_e one can obtain the space potential $V_{s0} = 11.6 \text{ V}$, electron temperature $T_e = 1.59 \text{ eV}$ and electron density $n_e = 6.27 \times 10^9 \text{ cm}^{-3}$. Here, $I^-/I^+ = 5.0$ (< 28.5) from $I^- = 1.25 \text{ mA}$ and $I^+ = 0.25 \text{ mA}$. This result suggests a generation of negative ions. Therefore, the above n_e does not express accurate electron density. By substituting $I_{is} = I^+ = 0.25 \text{ mA}$ and $T_e = 1.59 \text{ eV}$ into Eq. (5), $n_i = n^+ = 3.0 \times 10^{10} \text{ cm}^{-3}$ is obtained. Further, by substituting $I^-/I^+ = 5.0$ into Eq. (4), $n^-/n^+ =$

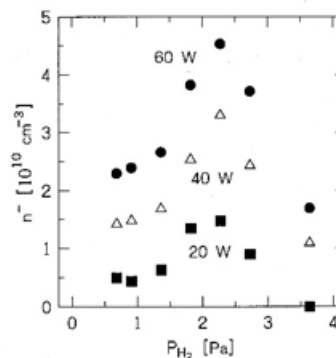


Figure 6. Dependence of n^- on P_{H_2} for three different RF powers. $x = 7.5 \text{ cm}$.

0.84 is obtained. The result suggests an efficient production of negative hydrogen ions.

Figure 5 shows the dependences of T_e and n^+ on hydrogen pressure P_{H_2} at the center of the device. RF power P_{RF} is 60 W . Here, n^+ is obtained from Eq. (5). As shown clearly, n^+ increases first with an increase in P_{H_2} , then reaches the maximum $6.1 \times 10^{10} \text{ cm}^{-3}$, then decreases drastically. We can notice an efficient generation of positive hydrogen ions at $P_{\text{H}_2} = 2.7 \text{ Pa}$. It is also noticed that T_e is lower than that of usual RF discharge plasmas [25]. The electron temperature monotonously decreases from 1.7 eV to 0.3 eV with an increase in P_{H_2} from 0.7 Pa to 3.6 Pa .

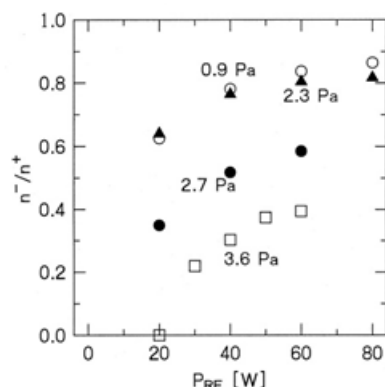


Figure 7. Variation of n^-/n^+ as a function of RF power P_{RF} with gas pressure P_{H_2} as a parameter. $x = 7.5 \text{ cm}$.

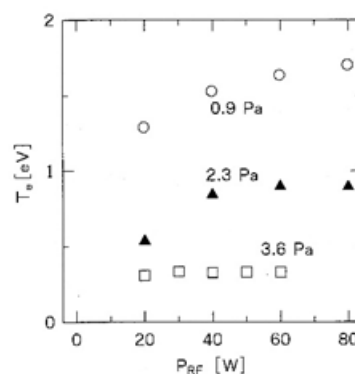


Figure 8. Variation of T_e as a function of RF power P_{RF} with gas pressure P_{H_2} as a parameter. $x = 7.5 \text{ cm}$.

The dependence of n^- on P_{H_2} for three different RF powers at the center of the device is shown in Fig. 6. n^- reaches the maximum $4.6 \times 10^{10} \text{ cm}^{-3}$ at $P_{H_2} = 2.3 \text{ Pa}$, being almost independent of the RF power, where an efficient generation of negative ions occurs. This n^- is at the same level as the density obtained by using DC discharge with magnetic filter at 4 kW [2]. This result suggests that much higher density n^- plasma can be produced if permanent magnets are employed [11]. In this case, the generation of high-density hydrogen plasma ($n^+ > 1 \times 10^{11} \text{ cm}^{-3}$) could be possible around $T_e \approx 1 \text{ eV}$ in the pressure range 0.9 Pa - 2.3 Pa by performing a low power ($\approx 100\text{W}$) RF discharge [17]. The dependences of n^-/n^+ and T_e on RF power P_{RF} are shown in Figs. 7 and 8, respectively, with hydrogen pressure P_{H_2} as a parameter. With an increase in the RF power, the ratio n^-/n^+ increases first, then it is saturated finally. This might be due to an increase in $H_2^*(v)$ by an increase in the high energy electron e_h in the source region. Further, as the hydrogen pressure decreases, T_e raises to an appropriate range of 1.3 – 1.5 eV for an effective formation of H^- ions. From Fig. 7 and Fig. 8 we can confirm that when T_e varies from 1.3 eV to 1.74 eV, n^-/n^+ increases from 0.79 to 0.87. This temperature range almost agrees with the results derived in the numerical study [16]. The reason why generation efficiency of negative ions is so low at $P_{H_2} = 3.6 \text{ Pa}$ might be due to that vibrationally excited molecules are not

enough supplied in such a low T_e plasma in the high pressure discharge.

V. DISCUSSION

In our method high T_e plasma is produced inside the grid electrode, where the reaction, $H_2 + e_h \rightarrow H_2^*(v) + e$, takes place by the high electron temperature electrons. On the other hand, in the region outside the grid electrode T_e decreases to a suitable level for the negative H^- ion production through the reaction $H_2^*(v) + e_l \rightarrow H^- + H$. The value of T_e can be basically varied by changing the mesh size under keeping the hydrogen pressure and input power. However, in this experiment, T_e is varied by a change of P_{RF} and/or P_{H_2} under the condition of fixed mesh size. The results are summarized by plotting all data on a $n^-/n^+ - T_e$ diagram, as shown in Fig. 9, with a use of the data at $P_{RF} = 60 \text{ W}$ in Figs. 5 - 8. The dotted curve is derived from a second order polynomial approximation using least-squares method. One can see that most suitable electron temperature for maximizing the ratio $n^-/n^+ (\approx 0.9)$ is about 1.3 eV, being consistent with a numerical simulation [16]. The maximum value of $n^- = 4.6 \times 10^{10} \text{ cm}^{-3}$ is also established at $P_{H_2} = 2.3 \text{ Pa}$ under 60 W, as shown in Fig. 6, which is higher than the other result of $n^- = 4.5 \times 10^{10} \text{ cm}^{-3}$ at $P_{H_2} = 0.9 \text{ Pa}$ under $P_{RF} = 4 \text{ kW}$ [2].

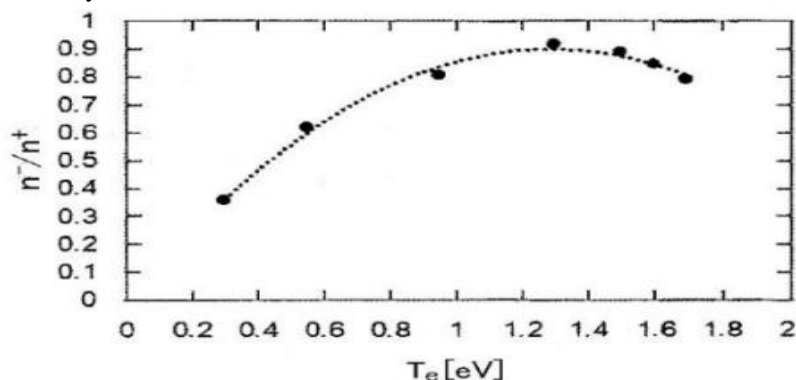


Figure 9. Diagram of n^-/n^+ and T_e , obtained from Figs. 5 – 8 at $P_{RF} = 60 \text{ W}$. Dotted curve is derived from a second order polynomial approximation using least-squares method.

Pulse-plasma operation [1] and magnetic filter method [3] are known as conventional production methods of negative hydrogen ions. However, in the former case the electron density fairly drops with a decrease in T_e , accompanied by its temporal variation. Therefore, it is difficult to keep T_e fixed at most suitable electron temperature. In the latter case, it is difficult to suppress RF electric-field penetration into the downstream low T_e region, which disturbs negative ion production [13]. To overcome these faults, cesium is needed for seeding additionally to the discharge chamber to raise the ratio n^-/n^+ [27]. Our method (i.e. self-biased method) with a grid electrode is quite suitable for negative hydrogen ion production in RF plasmas, because the high and low electron temperature regions are separated by the grid electrode. The electron temperature outside the grid electrode can be controlled by the mesh size and discharge parameters. This method works even when the grid electrode is covered by resistive films such as hydrogenated amorphous films, diamond-like carbon, and so on. In Ar plasma the electron temperature can be further lowered by employing a fine mesh electrode, accompanied by an increase in the electron density.

VI. CONCLUSION

We have demonstrated a new method with a use of a self-biased grid electrode for lowering electron temperature in an RF discharge plasma. High electron temperature plasma is produced inside the grid electrode for $H_2^+(v)$ production. In the region outside the grid electrode the electron temperature decreases to a suitable value for a negative H^- ion production through the dissociative attachment of $H_2^+(v)$. In the case of mesh electrode of 5 mesh per inch, n^-/n^+ reached to 0.87 at $P_{H_2} = 0.9$ Pa and $P_{RF} = 80$ W at the center of the discharge chamber. The maximum value of $n^- = 4.6 \times 10^{10} \text{ cm}^{-3}$ is established at $P_{H_2} = 2.3$ Pa when the RF power is 60 W. It is confirmed that negative hydrogen ions are produced efficiently in the range $T_e = 1.0 - 1.6$ eV, which is consistent with a simulation result. The self-biased method with a grid electrode is quite suitable to improve negative ion volum generation in capacitively-coupled RF discharges.

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