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Theoretical Analysis on Effect of Adding Argon to Hydrogen Plasma

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Abstract An increased of H^- ion beam current whenever argon gas is added has been observed. The physics behind argon effect is that in the production of more H atoms, a part of H atoms will be converted to $H_2^*(v>4)$ to increase H^- density. Increase of n_H depends on the rate of added Ar $\eta\%$, primary filling pressure P, electron density n_{eAr} and recombination coefficient γ of H atom. Adding 25% Ar is optimal, although there have been assumed various parameters. Increase of n_H depends on decreased recombination coefficient γ, and increased primary filling pressure P. The increase of n_H is small when the pressure $P < 0.3$ Pa. Optimized volume size $L/R \sim 2$ is optimal for obtaining a maximum Ar effect.

Keywords: H^- ion beam, Ar effect, neutral beam injection

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1 Introduction

Developing negative ion source is getting more important as their applications increase. The negative ion-based neutral beam injector (NNBI) is one of the most promising candidates for the current drive and heating system in a steady-state operation of reactor grade tokamak fusion devices, such as the international thermonuclear experimental reactor (ITER). Toward this end, a large-area RF source for negative ion has been developed in IPP in Garching [1]. With this RF source, we observed that the extracted H^- ion beam current is increased whenever argon gas is added and whenever cesium vapor is injected into the source chamber [2~3].

In this paper, we try to explain the effect of adding argon on the increase of electron density [3.23] and H atom density in drive region, and the conversion of a part of H atom to $H_2^*(v>4)$ [9,14,15] when H atoms are colliding with the wall. The dissociative attachment of $H_2^*(v>4)$ helps in the increase of H^- density. With Cs injection, a part of H atoms directly are converted to H^- when H atom are colliding with the plasma grid whose working function decreases during Cs injection.

2 Theoretical analysis

2.1 Simulating model of negative ion source

To study H^- production in a two tandem system, we used a simulating model of RF negative ion source as shown in Fig. 1, where the plasma is divided in two regions by a magnetic filter (MF): one is drive region and the other is extracting. In the figure, L is the cylindrical discharge cell length, and R is the radius. Many reaction processes will be occurred in the negative ion source, for which the source has been optimized [5~6]. In order to discuss the effect of adding Ar gas to hydrogen plasma, we consider only main reaction processes.

We assume that electron ($T_e < 15$ eV)-produced H atoms give an impact on H_2 and Ar^+ gives a strong

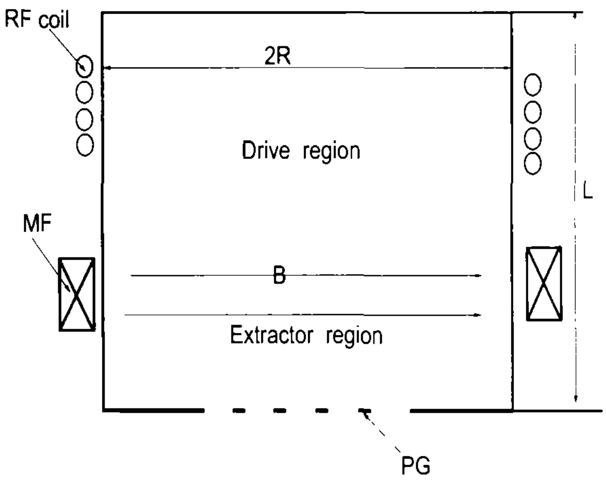

Fig.1 Schematic of RF negative ion source

impact on H_2, and fast electrons e_f (T_e >15 eV)-produced H_2^*(v>4) molecules on highly vibrationally excited states are present only in the drive region. These excited molecules can travel to the extracting region through the magnetic filter, while high-energy electrons are being cooled. On the one hand, a part of H_2^*(v>4) are produced on the wall surface due to the recombination of H and neutralization of positive ions. The cooling electrons ($T_e < 1$ eV) are necessary for the dissociative attachment to the vibrationally excited molecules-produced H^-. On the other hand, H atom produced-H^- collides with the wall surface during Cs injection.

In the drive region:

a. Electrons impact H_2 from the ground state to the excited states, and H_2 is dissociated into two H atoms [11,17],

$$e + H_2 \longrightarrow e + H_2^*(v < 4) \longrightarrow e + H + H. \tag{1}$$

$\sigma_{max} \sim 1.4 \times 10^{-16} cm^2$, T_e around 15 eV.

b. Fast electrons ($T_e > 15$ eV) impact H_2 to the highly excited state [6],

$$e_{fast} + H_2 \longrightarrow H_2^*(v > 4) + e. \tag{2}$$

c. Proton transfer between H_2 and Ar^+[8]

$$Ar^+ + H_2 \longrightarrow ArH^+ + H, \tag{3}$$

$K \sim 10^{-9}\ cm^3 s^{-1}$.

In H^--extracting region [21]**:**

a. Dissociative attachment of an electron on the vibrationally excited state [18]

$$H_2^*(v > 4) + e_{slow}(T_e < 1eV) \longrightarrow H^- + H. \tag{4}$$

b. H_2^*(v) production on the wall surface due to the recombination of H and neutralization of positive ions[9,14,15]

$$H, H_n^+ + wall \longrightarrow H_2^*(v > 4). \tag{5}$$

c. H^- production on the surface caused by Cs injection [22]

$$H, H_n^+ + surface \longrightarrow H^-. \tag{6}$$

2.2 Assumed parameters for main reaction processes

For the above-mentioned reactor processes, there have been assumed as follows:

a. The probability ρ_1 for the conversion of H_2 into H_2^*(v<4), with a subsequent conversion to H atom.

$$\rho_1 = \frac{n_{H_2^*(v<4)}}{n_{H_2}}. \tag{7}$$

Since the H_2^*(v<4) produced by electrons collide with H_2 , the maximum H_2^*(v<4) density $\le n_e$, hence

$$\rho_1 \le \frac{n_e}{n_0}, \tag{8}$$

where, n_0 is the H_2 density which is the primary neutral particles number, n_e is the discharging electron density.

b. The probability ρ_2 of the fraction of fast electron density $n_f(T_e > 15$ eV$)$[19] to full electron density.

$$\rho_2 = \frac{n_f}{n_e} = \frac{(n_{H_2^*(v>4)})_1}{n_0}. \tag{9}$$

c. The probability ρ_3 of the conversion of a part of H atoms to H_2^*(v >4) during the recombination of H atom on the wall.

$$\rho_3 = \frac{(n_{H_2^*(v>4)})_2}{n_H}. \tag{10}$$

d. The probability ρ_4 of the conversion of H atom to H^- on the wall surface during Cs injection only.

$$\rho_4 = \frac{n_{H^-}}{n_H}. \tag{11}$$

In general, the negative ion yield per H impacting on a surface can be predicted [16,22] by

$$\rho_4 = (2/\pi)\exp[-\pi(\Phi - A)/2\alpha v], \qquad (12)$$

where Φ is the surface work function; A is the affinity of the electron (0.75 eV); v is the normal velocity of the incident particle; α is the decay constant.

The effect of Cs is expressed through the value of Φ. For example, Φ is 1.45 eV for the surface covered with half of a monolayer of Cs and 2.1 eV for the surface covered with a monolayer of Cs. If we take the temperature of H atoms to be 0.5 eV, $\Phi = 1.8$ eV [24] and $\alpha = 3.08 \times 10^{-5}$ eVsec m^{-1}[25], we can estimate $\rho_4 = 4.87 \times 10^{-3}$ for H atoms, and if H atoms with an energy of 1 eV, $\rho_4 = 2.05 \times 10^{-2}$.

2.3 Basic theory of balance equation

2.3.1 Particle number conservation equation

Investigation of the negative ion source using a numerical code has been done for pure hydrogen plasma [4,5]. The code is based on the rate equations for particle species H, H_2^*(v>4), H^-, and H_n^+(n = 1,2,3), where $H_2^*(v)$ is the vibrationally excited hydrogen molecule on a high state (v>4). For H_2, a conservation equation of the H atom number density is used. The conservation equation is written as [6,7]

$$2n_{H_2} + 2n_{H_2^*(v)} + n_H + n_{H^-} + n_{H^+} + 2n_{H_2^+} + 3n_{H_3^+} = 2n_0, \qquad (13)$$

where n_0 is the density of hydrogen molecules H_2 before discharge and n_j is the discharged density of the jth species of particle.

$$n_0 = \frac{P}{KT}, \qquad (14)$$

where P is the filling gas pressure.

Adding $\eta\%$ Argon gas to hydrogen plasma,

$$n_0 = n_{0H_2} + n_{0Ar}, \qquad (15)$$

here, n_{0H_2} and n_{0Ar} are the primary neutral particles number for H_2 and Ar respectively.

$$n_{0Ar} = \eta\% n_0, \quad n_{0H_2} = (1 - \eta\%)n_0. \qquad (16)$$

As to the calculation of 'argon effect', only two ion species (H^+ ,Ar^+) are taken into account, (H^+ includes H^+, H_2^+, H_3^+ for a simplified calculation). For the four species of neutral particles (H, H_2, $H_2^*(v)$, Ar) in the drive region of ion source, the particle number conservation equation is written as

$$n_{0Ar} = n_{Ar} + n_{Ar^+} = \eta\% n_0, \qquad (17)$$

$$2n_{H_2} + 2n_{H^*_{2(v)}} + n_H + n_{H^+} = 2(1 - \eta\%)n_0, \qquad (18)$$

here, $n_{H_2^*(v)}$ is the vibrationally excited hydrogen molecule on a high state (v>4), which is contributed by the fast electron colliding with H_2 and a part of H atoms colliding with the wall.

According to the assumed equation (9) and (10), the total $n_{H_2^*(v)}$ is

$$n_{H_2^*(v)} = \rho_2 n_0 + \rho_3 n_H. \qquad (19)$$

2.3.2 Charge conservation equation

Ion source's plasma maintains electrical neutral all the time,

$$n_{eAr} = n_{H^+} + n_{Ar^+}, \qquad (20)$$

where n_{eAr} is the density of electron with discharged Ar and the doubly-ionized species as well as molecular and negative ions are negligible, because it is considered that H^- production is small and takes place mainly in vicinity plasma grid, compared with full electron density.

2.3.3 Fraction of n_{Ar^+} / n_{H^+}

The n_{Ar^+}/n_{H^+} ratio can be calculated using the Boltzmann equation [8]:

$$\frac{n_{Ar^+}}{n_{H^+}} = \frac{n_{Ar}}{n_H}\frac{g_{Ar^+}}{g_{Ar}}\frac{g_H}{g_{H^+}}\exp(\frac{\Phi_H - \Phi_{Ar}}{T_e(eV)}), \qquad (21)$$

where n_p/g_p is the radial profiles of the absolute level population per statistical weight. Φ_H is the H ionization potential (13.6 eV); Φ_{Ar} is the Ar ionization potential (15.8 eV).

It is assumed that H_2 and Ar gas have the same ionization degree, and one molecular H_2 is converted to two atomic H.

For example, if the H_2 gas contains a $\eta\%$ of Ar gas before discharge in ion source, T_e = 15 eV, we get

$$\frac{n_{Ar^+}}{n_{H^+}} = \frac{n_{Ar}}{n_H}\frac{g_{Ar^+}}{g_{Ar}}\frac{g_H}{g_{H^+}}\exp(\frac{\Phi_H - \Phi_{Ar}}{T_e(eV)}) =$$

$$\eta\% \times 0.5 \times 12 \times \exp(\frac{13.6 - 15.8}{15}) =$$

$$5.15\eta\%. \qquad (22)$$

2.3.4 Full electron density with and without Ar

In langmuir probe measurement [3,23], it has been found that the electron density n_{eAr} is higher when adding Ar in drive region, but is a fair value in extracting region. So, we assume that the increase in n_e factor is the same as the added argon factor $\eta\%$ in the drive region, which means that Ar ionization degree is to be roughly the same as that in the pure H_2 case.

$$n_{eAr} = (1 + \eta\%)n_e, \qquad (23)$$

here, n_e is the electron density without Ar.

2.3.5 H atom density balance equation with Ar

Adding argon to hydrogen plasma could increase density of H atom, in the meantime, the H atoms will be lost while colliding with the wall.

The simplified balance equation for the production and loss of H atoms is written as

$$\frac{\mathrm{d}n_H}{\mathrm{d}t} = \rho_1 n_{H_2} \times \int_E F_e(E)\sigma(E)\mathrm{d}E \times 2 + n_{H_2}kn_{Ar^+} - n_H\frac{D_{H-Ar}}{\Lambda^2}\gamma. \qquad (24)$$

The first two terms on the right-hand side correspond to the production of H atoms, by the reaction of (1) and (3), respectively, and the last term represents the loss of H atoms due to diffusion and recombination on the wall.

$F_e(E)\sigma(E)\mathrm{d}E$ is the electron flux energy distribution function;

k is the rate coefficient of Ar^+ produced-H atoms;

σ is the cross section of H_2 produced-H atoms;

D is the H atom diffusion coefficient in argon gas;

γ is the recombination factor, depending on the kind of surface and wall material. (γ = 0.1~ 0.25 where γ is defined as the ratio of atoms striking the surface and recombined molecules to the total number of atoms colliding with the surface $\gamma = 1 - \Gamma_{out}/\Gamma_{in}$ where Γ is the H flux.).

Λ is the characteristic diffusion length;

ρ_1 is the probability of H_2 conversion to $H_2^*(v)$ in volume reaction process.

Since the H atom density is constant under a steady-state condition, the two terms of the production and the loss of H atoms should be equal to each other

$$\rho_1 n_{H_2} \times \int_E F_e(E)\sigma(E)\mathrm{d}E \times 2 + n_{H_2}kn_{Ar^+} = n_H\frac{D_{H-Ar}}{\Lambda^2}\gamma. \qquad (25)$$

Substituting $\nu = \int_E F_e(E)\sigma(E)\mathrm{d}E \approx n_{eAr} \times \sigma \times V$ into equation (25),

$$\frac{n_H}{n_{H_2}} = \frac{[\rho_1 \times \int_E F_e(E)\sigma(E)\mathrm{d}E \times 2 + kn_{Ar^+}]\Lambda^2}{D_{H-Ar}\cdot\gamma} = \frac{\rho_1 n_{eAr}\sigma V \times 2 + kn_{Ar^+})\Lambda^2}{D_{H-Ar}\cdot\gamma}, \qquad (26)$$

here, V is the averaged electron speed,

$$V = \sqrt{\frac{2KT_e}{m_e}}, \qquad (27)$$

taking T_e = 15 eV in the drive region,

$$V = 2.3 \times 10^8 \ \ \mathrm{cm\,s^{-1}}. \qquad (28)$$

D_{H-Ar} can be calculated as follows [12]:

$$D_{H-Ar} = 2.6280 \times 10^{-3}\frac{\sqrt{T^3(M_H + M_{Ar})/2M_HM_{Ar}}}{P(\mathrm{atm})d_{HAr}^2} = 2.65 \times 10^2\frac{\sqrt{T^3(M_H + M_{Ar})/2M_HM_{Ar}}}{P(\mathrm{Pa})d_{HAr}^2}, \qquad (29)$$

where D_{H-Ar} has the unit $\mathrm{cm^2s^{-1}}$, P has the unit Pa and T is the room temperature T = 300 K, d is the molecular diameter in angstrom, d_H = 3 Å, d_{Ar} = 3.64 Å, $d_{HAr} = 1/2(d_H + d_{Ar})$

M is the molecular weight $M_H = 1$, $M_{Ar} = 40$.

So,

$$D_{H-Ar} = 9.0 \times 10^4/P \quad [\mathrm{cm^2s^{-1}, Pa}]. \qquad (30)$$

We apply the diffusion length, which is valid for a cylindrical chamber [13]

$$\Lambda^2 = \frac{1}{(\frac{\pi}{L})^2 + (\frac{2.4}{R})^2}, \qquad (31)$$

where L and R are the length and radius of the (cylindrical) discharge cell, respectively. Taking L = 20 cm, R = 15 cm, all of which are similar to that of the type of IV source,

$$\Lambda^2 = 20 \text{ cm}^2. \quad (32)$$

2.4 Derivation of H atom density n_H with argon

To solve equation (20) and (22),

$$n_{Ar^+} = \frac{5.15\cdot\eta\%}{1+5.15\cdot\eta\%}\cdot n_{eAr} = a\cdot n_{eAr}, \quad (33)$$

here, a is the factor, depending on $\eta\%$ Ar.

Taking $\sigma = 1.4\times10^{-16}$ cm^2, $k = 1\times10^{-9}$ cm^{3}s^{-1}, and substituting equation (28), (30), (32) and (33) to equation (26), we get

$$\frac{n_H}{n_{H_2}} = \frac{1.4\times10^{-16}\times2.3\times10^8\times2\times\rho_1 n_{eAr}\times20}{(9.0\times10^4/P)\times\gamma} + \frac{1\times10^{-9}\times a n_{eAr}\times20}{(9.0\times10^4/P)\times\gamma} = \frac{(1.43\times10^{-11}\cdot\rho_1+2.22\times10^{-13}\cdot a)\cdot n_{eAr}\cdot P}{\gamma} = b, \quad (34)$$

where, n_{eAr} is the electron density with Ar (cm^{-3}), P is the unit of filling gas pressure. Substituting equation (17), (19), (20) and (33) to equation (18), we get

$$2n_{H_2} + (1+2\rho_3)n_H = 2(1-\rho_2-\eta\%)n_0 - (1-a)n_{eAr} = c. \quad (35)$$

To solve the equation (34) and (35), we get

$$n_H = b\cdot\frac{c}{[2+(1+2\rho_3)b]}, \quad (36)$$

in which H atom density depends on the rate of adding Ar $\eta\%$, the primary filling pressure P, the electron density n_{eAr} and the recombination coefficient γ of H atoms.

3 Results

As to the cylindrical discharge cell with a length L = 20 cm, a radius R = 15 cm, and taking average electron density ne without Ar, we have n_e = 4.2 $\times10^{11}$ cm^{-3}.

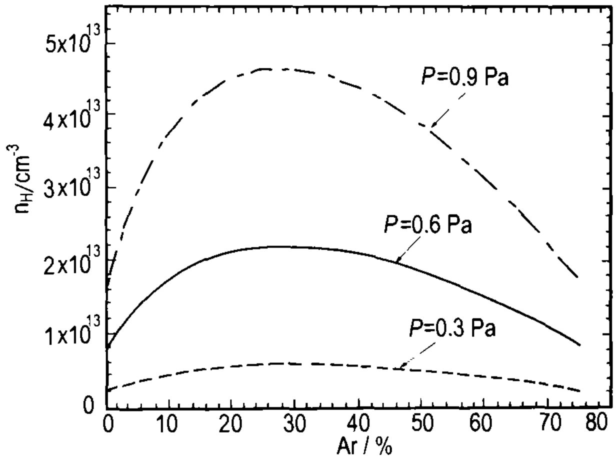

Fig.2 The dependence of the increase of n_H on Ar% under various P. Assumption ρ_1=0.003, ρ_2 = 0.1, ρ_3= 0.005, γ=0.2

3.1 Increase of n_H with various P and γ

When assuming the probability ρ_1= 0.003 for the conversion of H_2 to H atom (7) and (8), the probability ρ_2 = 0.1 for the fraction of fast electrons density to full electron density and the probability ρ_3 = 0.005 for the conversion of a part of H atom to H_2^*(v > 4) keeping wall recombination of H atom constant, H atoms density with Ar depends on the percentage of added Ar (%) under various primary filling pressures P (Fig. 2) while adopting material γ = 0.2. The increase of n_H depends on the increase of P. The increase of n_H is small when the pressure P <0.3 Pa. Also, we obtain that H atoms density with Ar depending on the percentage of added Ar (%) with various material recombination coefficient γ while keeping primary filling pressure P = 0.6 Pa (Fig. 3) constant. The increase of n_H depends on the decrease of γ . The maximum n_H produced at the point of added Ar about 25%, although varying P from 0.3 Pa to 0.9 Pa and varying γ from 0.1 to 0.25 respectively.

3.2 Increase of n_H with assumed various ρ_1, ρ_2 and ρ_3

When applying primary filling pressure P = 0.6 Pa,

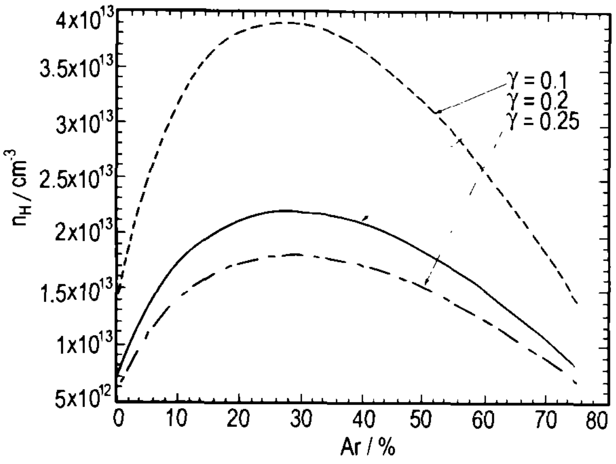

Fig.3 The dependence of the increase of n_H on Ar% under various γ. Assumption $p=0.6$ Pa, $\rho_1=0.003$, $\rho_2=0.1$, $\rho_3=0.005$

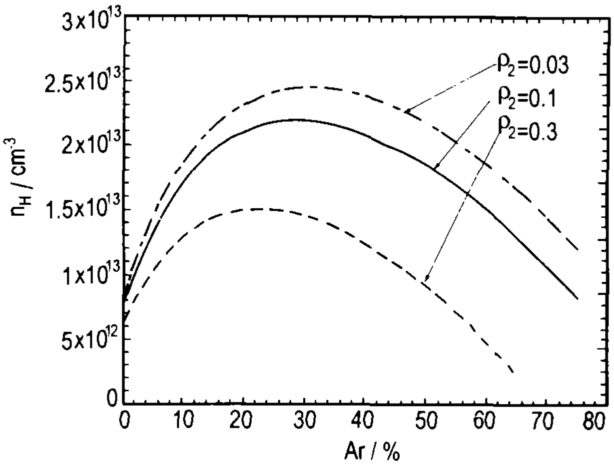

Fig.5 The dependence of the increase of $n_{\rm H}$ on Ar% with various ρ_2. Assumption $P=0.6$ Pa, $\gamma=0.2$, $\rho_1=0.003$, $\rho_3=0.005$

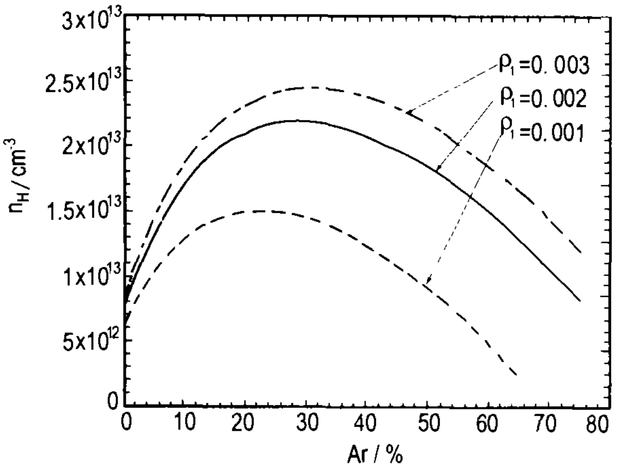

Fig.4 The dependence of the increase of $n_{\rm H}$ on Ar% with various ρ_1. Assumption $p=0.6$ Pa, $\rho_1=0.003$, $\rho_2=0.1$, $\rho_3=0.005$

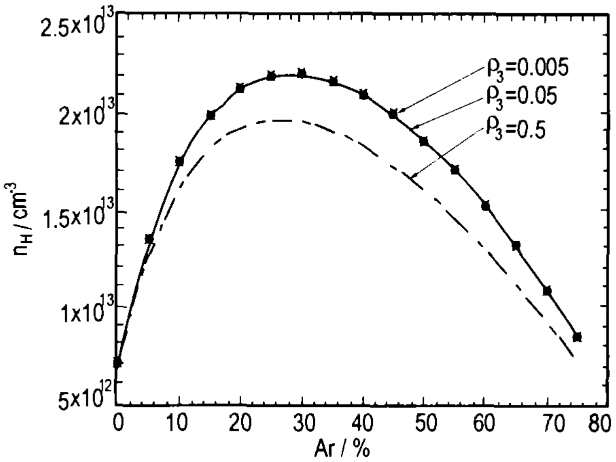

Fig.6 The dependence of the increase of $n_{\rm H}$ on Ar% with various ρ_3. Assumption $P=0.6$ Pa, $\gamma=0.2$, $\rho_1=0.003$, $\rho_2=0.1$

and adopting material $\gamma=0.2$, the H atoms density with Ar depends on added Ar% with assumed various ρ_1 while keeping the assumed $\rho_2=0.1$and $\rho_3=0.005$ constant (Fig. 4), and varying ρ_2 while keeping ρ_1 and ρ_3 constant (Fig. 5), and varying ρ_3 while keeping ρ_1 and ρ_2 constant (Fig. 6). The maximum $n_{\rm H}$ produced at the point of added Ar about 25%, although varying ρ_1 from 0.001 to 0.003, and ρ_2 from 0.03 Pa to 0.3, and ρ_3 from 0.005 to 0.5, respectively.

3.3 Increase factor of $n_{\rm H}$ and $n_{\rm H}$-with Ar

The increased factor A of H denotes the ratio of H atom density with argon to that without argon, without argon,

$$A=\frac{n_{\rm H}\ \text{with Argon}}{n_{\rm H}\ \text{without Argon}},$$

for example, taking $P=0.6$ Pa, and adopting material $\gamma=0.1$, with assumed $\rho_1=0.003$, $\rho_2=0.03$ and varying ρ_3 from 0.03 to 0.3, the factor A of $n_{\rm H}$ increases about 3 while the added Ar is 25% (Fig. 7).

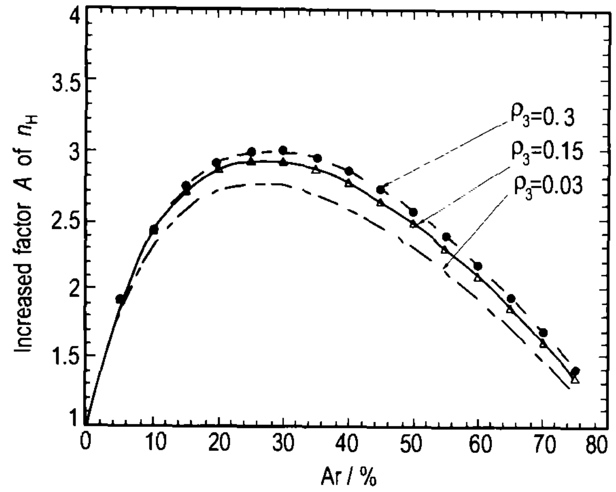

Fig.7 The dependence of the increased factor A of n_H on Ar% with various ρ_3. Assumption $P = 0.6$ Pa, $\gamma = 0.1$, $\rho_1 = 0.003$, $\rho_2 = 0.03$

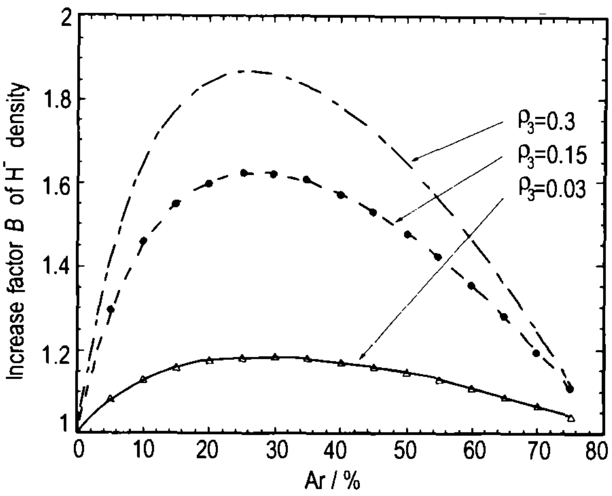

Fig.8 The dependence of the increased factor A of n_{H^-} on Ar% with various ρ_3. Assumption $P = 0.6$ Pa, $\gamma = 0.1$, $\rho_1 = 0.003$, $\rho_2 = 0.03$

The increased factor B of H$^-$ denotes the ratio of H$^-$ density with argon to that without argon

$$B = \frac{n_{H^-}\text{ with Argon}}{n_{H^-}\text{ without Argon}} = \frac{n_{H_2^*(v>4)}\text{ with Argon}}{n_{H_2^*(v>4)}\text{without Argon}}.$$

Considering only the conversion of a part of H atoms to $H_2^*(v>4)$ during wall recombination of H atom, the strongly increased factor B of H$^-$ depends on the probability ρ_3. When varying ρ_3 from 0.03 to 0.3, the factor B varies from 1.2 to 1.85 respectively with added Ar about 25% (Fig. 8).

3.4 Increased factor of n_{H^-} with Ar + Cs

Considering H$^-$ density produced by volume reaction process (4) and surface process (6), H atom can be mainly contributed to the production of H$^-$ during Cs injection (12).

The increased factor C of H$^-$ denotes the ratio of H$^-$ density with argon to that without argon during Cs injection,

$$C = \frac{n_{H^-}\text{ with Argon + Cs}}{n_{H^-}\text{ without Argon + Cs}},$$

hence,

$$C = \frac{n_{H_2^*(v>4)}\text{ with Argon}}{n_{H_2^*(v>4)}\text{ without Argon}} + \frac{\rho_4 n_H\text{ with Argon}}{\rho_4 n_H\text{ without Argon}} = A + B,$$

for example, taking $P = 0.6$ Pa, adopting material $\gamma = 0.1$, and assuming $\rho_1 = 0.003$, $\rho_2 = 0.03$ and varying ρ_3 from 0.03 to 0.3 during Cs injection, the factor C of n_{H^-} increases by 4 to 4.5 with added Ar 25% (Fig. 9). If without Ar, the increased factor is only 2 during Cs injection.

4 Discussions and conclusions

Hydrogen plasma have many reaction processes occurred. It is more difficult to describe exactly, the mechanism of adding argon to the hydrogen plasma, although many authors have researched for many years. According to the simple theoretical analysis above, more H atoms may be increased by the 'argon effect', and then converted to H$^-$, thus, increasing H$^-$ current to be extracted. Argon effects are related with the source's volume, wall material and pressure of operation. Optimizing these parameters may obtain a higher H$^-$ current.

For the ion source's volume (length L and radius R), the effect is expressed through the value of Λ^2 (31). The increase of density n_H depends on the increase of Λ (26), so there exists an optimized parameter L/R for a constant volume. $L/R \sim 2$ is optimal for obtaining a maximum Ar effect (Fig. 10).

Argon effect exists all the time. Although those assumed values are not exact enough, the factor value of the argon effect could be changed by the assumed

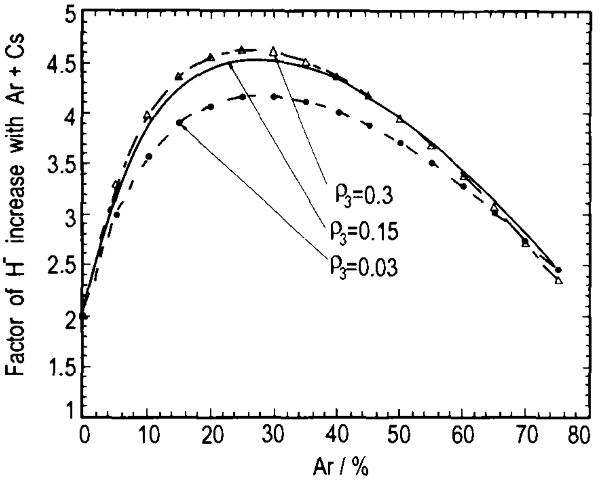

Fig.9 The dependence of the increased factor C of n_{H^-} on Ar% with various ρ_3 during Cs injection. Assumption. $P=0.6$ Pa, $\gamma=0.1$, $\rho_1=0.003$, $\rho_2=0.03$

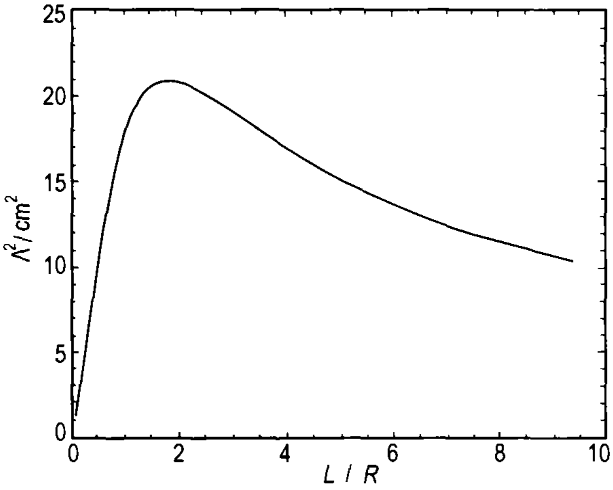

Fig.10 Λ^2 characteristic diffusion length vs L/R

various parameters. If the H^- produced by fast electron is higher than converted from H atom, the argon effect can not play a dominant role.

The argon effect would be different from that of other noble gases compared, but the difference in mass is not important on the assumption that other parameters of the other noble gases are the same as that of the argon except the mass .

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