Kinetics of electrons and neutral particles in radio-frequency transformer coupled plasma $\text{H}^-$ ion source at Seoul National University

K J Chung, J J Dang, J Y Kim, W H Cho and Y S Hwang

Department of Nuclear Engineering, Seoul National University, Seoul 08826, Korea

E-mail: yhwang@snu.ac.kr

Keywords: negative ion, volume production, transformer coupled plasma, magnetic filter, electron kinetics, neutral transport, degree of dissociation

Abstract

In volume production $\text{H}^-$ ion sources, control of electron temperature is essential due to its close correlation with the generation of vibrationally-excited hydrogen molecules in the driver region as well as the generation of $\text{H}^-$ ions by dissociative attachment in the extraction region. In the ion source group at Seoul National University (SNU) in Korea, a lot of research effort has been made to the development of a volume production $\text{H}^-$ ion source based on radio-frequency (RF) transformer-coupled plasma (TCP) for long lifetime continuous wave (CW) operation. It has a spiral RF antenna located outside the discharge chamber to generate a plasma with high electron temperature in the driver region and employs a magnetic filter field to prevent high energy electrons from being transported to the extraction region. In this paper, we present the recent progress on understanding of the underlying physics of the RF TCP $\text{H}^-$ ion source at SNU. Special attention is paid to the characterization of electron kinetics regime for controlling electron energy distribution and the influence of relaxation of neutral particles during the transport across the magnetic filter region. Effect of the degree of dissociation on the production of $\text{H}^-$ ions is also discussed.

1. Introduction

Among various $\text{H}^-$ generation mechanisms [1, 2], it is well known that volume production of $\text{H}^-$ ions can be accomplished mainly by a two-step process [3]: (1) generation of highly vibrationally-excited molecules and (2) production of $\text{H}^-$ ions by dissociative attachment (DA) reaction. The first step is a generation of highly vibrationally-excited molecules as a precursor of the reaction. They are produced mainly by inelastic collisions of electrons with hydrogen molecules. There are two major channels to generate vibrationally-excited molecules known as e-V and E-V reactions [3]. In e-V reactions, the vibrational states are excited via unstable $\text{H}_2^-$ ($X^2\Sigma_u^-$) resonant state overlapped with $\text{H}_2$ ($X^1\Sigma_u^+$) state. Due to a low threshold energy, the e-V reaction occurs mainly by low energy electrons, and thus it is not suitable for generating highly vibrationally-excited molecules. On the contrary, the E-V reaction has a high threshold energy because the vibrational excitation is induced by a radiative decay of electronically-excited molecules, e.g. $\text{H}_2$ ($B^3\Pi_u^+$). Hence, the E-V reaction is more efficient than the e-V reaction to excite hydrogen molecules to higher vibrational states in spite of its higher threshold energy. For example, the rate of E-V reactions is 4–5 orders of magnitude higher than that of e-V reaction for an excitation reaction from $\nu = 0$ to $\nu = 7$, as depicted in figure 1(a). The second step is a production of $\text{H}^-$ ions by DA reaction of the highly vibrationally-excited molecules generated from the previous step with low energy electrons. In this step, the electron temperature must be kept low because the cross section for DA reaction depends greatly on the energy of electrons interacting with vibrationally-excited molecules (see figure 1(b)). Also, low electron temperature has a benefit in minimizing the destruction of $\text{H}^-$ ions by collisional detachment by electrons.

© 2016 IOP Publishing Ltd and Deutsche Physikalische Gesellschaft
The above mentioned mechanism for volume production of H$^-$ ions demands for two different regions with different electron temperatures. On the one hand, a plasma with high electron temperature is preferred in the driver region (source plasma region) for generating highly vibrationally-excited hydrogen molecules. On the other hand, a plasma in the extraction region (H$^-$ generation region near an extraction aperture) should have low electron temperature in the absence of energetic electrons. These two regions are usually separated by a transverse magnetic field, designated as a magnetic filter field in figure 2, which allows the transfer from the source plasma region to the extraction region of ions, slow electrons, atoms and molecules, but is impermeable to energetic electrons. Therefore, delicate control of electron temperature both in the driver region and extraction region of the plasma source plays a crucial role in volume production H$^-$ ion sources.

In the ion source group at Seoul National University (SNU) in Korea, a lot of research effort has been made to the development of a volume production H$^-$ ion source for long lifetime continuous wave (CW) operation [4]. The ion source developed at SNU is based on transformer-coupled plasma (TCP) operating at radio-frequency (RF) range, typically at 13.56 MHz. It has a spiral RF antenna located outside the discharge chamber to generate a plasma with high electron temperature in the driver region and employs magnetic filter field for preventing high energy electrons from being transported to the extraction region. Since the RF antenna is located outside the discharge chamber, the RF TCP H$^-$ ion source at SNU is free from contamination, making long lifetimes in CW operations possible. However, since there are no primary electrons with high kinetic energy in this plasma source, separate control of the electron temperature in the driver region and the extraction region plays a key role in the performance of H$^-$ ion production. Therefore, understanding the physics of electron heating mechanism and magnetic filtering are of great importance in the development of the SNU H$^-$ ion source.

In our earlier work, we have tried to increase H$^-$ ion production by controlling global plasma parameters such as electron temperature and density. Cusp magnetic field was introduced to increase the electron density in the discharge chamber [4]. Influence of RF wave frequency and surface material on H$^-$ ion production were examined [5, 6]. We also found that the electron temperature could be easily increased by shortening the length of the discharge chamber [7]. However, the overall increase of electron temperature failed to filter high energy electrons near the extraction hole efficiently [8]. Optimization of filter field strength depending on operating conditions were carried out to resolve this problem [9]. The evidence of electron cyclotron heating by filter field was examined [10], and the influence of non-Maxwellian electron energy distribution was taken into consideration for low pressure discharge [11].
Recently, we recognized that fundamental electron heating mechanism for hydrogen discharges needs to be considered in order to enhance $\text{H}^-$ ion production further. Hence, we started the investigation of electron heating kinetics in the absence or in the presence of a magnetic filter field by analyzing electron energy distribution function (EEDF) more rigorously [12]. However, most research carried out so far has been limited to the control of plasma parameters such as electron temperature and density. Considering the fact that $\text{H}^-$ ions are produced in a region far away from the driver region where vibrationally-excited molecules are mostly generated, we notice that transport of vibrationally-excited hydrogen molecules across the magnetic filter region plays an important role in volume production $\text{H}^-$ ion sources. In spite of its importance, the behavior of hydrogen molecules has not yet been taken into account.

In this paper, we present the recent progress of our understanding on the underlying physics of the RF TCP $\text{H}^-$ ion source at SNU. We measured EEDFs along the distance from the extraction hole depending on the operating pressure. The change in the EEDFs is explained by fundamental heating mechanism of hydrogen discharge depending on electron kinetics regime. Furthermore, the influence of relaxation process of vibrationally-excited molecules transporting across the magnetic filter region on $\text{H}^-$ ion production is studied both numerically and experimentally. Effect of the degree of dissociation on the production of $\text{H}^-$ ions is investigated as well.

2. Experimental setup

2.1. RF TCP $\text{H}^-$ ion source at SNU

Cutaway view and schematic diagram of an RF TCP volume production $\text{H}^-$ ion source developed at SNU is depicted in figure 3. The $\text{H}^-$ ion source has a cylindrical structure with inner diameter of 100 mm and the length of 100 mm. A water-cooled 14-row of SmCo magnets (1 cm wide and 10 cm long) surrounds the discharge chamber to enhance the electron confinement and to improve the plasma uniformity [4]. The strength of the cusp field is as strong as 0.1 T near the chamber wall, but it decays rapidly to become almost zero at the axis of the ion source. At the left end of the chamber, a dielectric window made of quartz is located to apply RF power through a 3½-turn external antenna. A 13.56 MHz RF power is fed to the antenna via a standard L-type RF matching network. At the opposite end of the chamber, an extraction hole of 8 mm in diameter is located to extract $\text{H}^-$ ions from the plasma. Near the extraction hole, a pair of NdFeB magnets (1 cm wide and 2 cm long) denoted as ‘virtual filter magnet’ in figure 3(b) is installed to produce transverse magnetic field in order to filter high energy electrons. The magnetic filter field is enhanced by additional transverse magnetic field produced by a pair of SmCo magnets denoted as ‘dipole filter magnet’ in figure 3(b). The strength of magnetic filter field is typically ~0.02 T near the extraction hole, but it decays along the distance from the extraction hole and becomes nearly zero near the opposite end of the chamber, as shown in figure 4. It is noted that the strength of magnetic filter field can be easily adjustable by attaching a pair of virtual filter magnets additionally.

A triode-type ion beam extraction system is adopted to extract $\text{H}^-$ ions from the plasma. Hence, it has a plasma electrode, an extraction electrode, and an acceleration electrode, as depicted in figure 3. In order to control potential structure near the extraction hole, we introduced a bias electrode which is attached in front of the extraction electrode. Since the plasma potential is generally positive, the negative ions as well as the electrons are electrostatically confined so that they are difficult to be extracted from the plasma. However, with the bias electrode, which can be biased at a positive potential slightly exceeding the plasma potential, the potential structure suitable for $\text{H}^-$ ion extraction can be obtained. The level of a positive bias is optimized for each operating condition such that the extracted negative ion beam current is maximized [4].

2.2. Diagnostic system

Most of measurements on plasma properties have been carried out in a diagnostic discharge chamber which is identical to the RF TCP $\text{H}^-$ ion source in shape and dimension, except that it has additional vacuum ports to accommodate diagnostic tools such as a Langmuir probe. For ease of diagnostics, the discharge chamber is installed in the vertical direction, as schematically shown in figure 5. We have considered a Langmuir probe system for measuring plasma properties, a laser photo-detachment system for determining $\text{H}^-$ ion density, and an optical emission spectroscopy system for diagnosing hydrogen molecules.

A Langmuir probe would be the most popular tool to diagnose low temperature plasmas because of its simplicity and high spatial resolution. In this study, an axial scanning Langmuir probe of which a tip made of a tungsten wire of 1 mm in length and 0.3 mm in diameter is installed through the extraction hole to measure the axial profile of plasma parameters. In our experiments, the Larmor radius of an electron with energy of 1.2 eV under magnetic field of 175 G, which corresponds to the maximum magnetic filter field strength in our measurement (5 mm from the extraction hole), is about 0.2 mm. Since the Larmor radius is larger than the radius of the probe tip, the influence of the magnetic field can be neglected. To exclude the noise from RF
fluctuation, the Langmuir probe has a reference ring and an RF choke for 13.56 MHz. The electron density and temperature are determined from the current-voltage characteristic curve measured by the Langmuir probe.

A laser photo-detachment diagnostic technique has been widely used for measuring $H^-$ density in the hydrogen plasma [13]. This method is based on the interaction process between $H^-$ ion and photons. A Nd:YAG
laser of 532 nm in wavelength is used to detach electrons from H$^-$ ions and the photo-detached electrons are collected by a single probe biased higher than the plasma potential [7]. A typical waveform measured by the probe during the laser photo-detachment is shown in figure 6. Note that the waveform shown in figure 6 is obtained by averaging 100 data measured by the probe in which an RF choke filter is employed to minimize RF noises. Before the laser pulse is turned on, the probe collects the electron saturation current, denoted as $I_{es}$. When the laser pulse is turned on, it is clearly observed that the electron saturation current increases rapidly, indicating the collection of electrons detached from H$^-$ ions. Hence, H$^-$ ion density can be easily deduced from the variation of the electron saturation current ($\Delta I$) by the laser pulse and the electron density measured by the probe.

In this study, we used the Fulcher-$\alpha$ band spectrum for investigating the behaviors of hydrogen molecules, especially their vibrational states. The Fulcher-$\alpha$ spectrum is a representative band spectrum accompanied with the radiative transition from the $d^3\Pi_u$ state (upper Fulcher state or briefly u-state) to the $a^3\Sigma_g^+$ state (lower Fulcher state or briefly a-state) of hydrogen molecules. A high resolution spectrograph (Princeton instruments, SP-2500i) is used to measure optical emission spectrum. Its focal length is 0.5 m, a grating is 1800 grooves/mm, and instrumental broadening is $\sim0.07$ nm. Its spectral sensitivity and the wavelength are calibrated with a tungsten lamp (Ocean Optics, LS-1-Cal) and a Hg-Ar lamp (Ocean Optics, HG-1), respectively. A typical Fulcher-$\alpha$ spectrum measured by the calibrated spectrograph is depicted in figure 7. In the analysis, the Q-branches of the diagonal Fulcher-$\alpha$ band spectrum in which the rotational quantum number is conserved during the transition are only utilized, because transition probabilities of P- or R-branches are known to be affected by other electronic state. Using this technique, the rotational and translational temperatures as well as the vibrational distribution function (VDF) of hydrogen molecules and the degree of dissociation are obtained [14].
3. Model

A numerical model suitable for describing the RFTCP H− ion source has been developed to interpret the experimental results comprehensively. The present model extends the previously developed model for hydrogen ion source at SNU [15] by including molecule transport and radiative decay. Considering that vibrationally-excited molecules are produced at the driver region and transported into the extraction region, the model consists of two modules; one module calculates the VDF of hydrogen molecules at the driver region based on the zero-dimensional particle balance model [16]. The other module calculates the transport of the vibrationally-excited molecules based on one-dimensional transport equation. Hence, the combined model can calculate the VDF at the extraction region where the VDF is relaxed by collisions with charged particles or neutrals during the transportation across magnetic filter field. Note that the VDF obtained by the 0D particle balance model is utilized as an initial condition of the 1D molecular transport model. In this model, the electron density is not determined self-consistently because the power balance equation is not employed to prevent from the uncertainty of the absorbed power. Instead, the electron density and temperature measured by the RF-compensated Langmuir probe are used for the 0D particle balance equation, satisfying the quasi-neutrality condition [17]. A coronal equilibrium model is also incorporated to the numerical model in order to check the validity of the Fulcher–α band spectrum intensity ratio.

3.1. Global model

In the zero-dimensional global model, not only each VDF state of \(H_2(0 \leq v \leq 14)\) but also \(H_1^+, H_2^+, H_3^+, \text{ and } H_4^+\) are included. Thus, the model calculates the particle densities of each ion and neutral species, under the conservation of total hydrogen atoms in addition to the charge neutrality as following:

\[
p = k_B T_g \left( \sum_{v=0}^{14} N_v + \frac{1}{2} n_{H_2^+} + \frac{1}{2} n_{H_3^+} + n_{H_4^+} + \frac{3}{2} n_{H_5^+} \right),
\]

(1)

\[
n_e = n_{H_1^+} + n_{H_2^+} + n_{H_3^+}
\]

(2)

where \(T_g\) is the gas temperature determined from the Fulcher–α spectroscopy.

The reaction rates for electron impact processes are obtained by integrating the cross sections for each reaction over the Maxwellian EEDF. Similarly, the reaction rate for collisions between heavy particles is calculated by

\[
K_{\text{heavy}} = 4\pi \left( \frac{\mu}{2\pi k_B T_{\text{eff}}} \right) \int_{0}^{\infty} \sigma(v) \nu \left( -\frac{\mu v^2}{2k_B T_{\text{eff}}} \right) \nu^2 \, dv,
\]

(3)

where \(\mu\) is the reduced mass and \(T_{\text{eff}}\) is the effective temperature defined respectively as

\[
\mu = \frac{m_1 m_2}{m_1 + m_2},
\]

(4)

\[
T_{\text{eff}} = \frac{m_1 T_1 + m_2 T_2}{m_1 + m_2}.
\]

(5)
The temperature of $H$ and $H^+$ is assumed as two folds of $T_g$ [18, 19]. The temperature of $H_2^+$ and $H_3^+$ is set to be the same as $T_g$ [18, 20].

The collision processes included in the 0D model are categorized as two groups. One is the volume process which is the collision between the charged–charged, charged–neutral, and neutral–neutral particles. The other is the wall process, i.e. the collision of the particles to the wall. Both processes are summarized in tables 1 and 2, respectively. The wall recombination factor ($\gamma$) of $H$ atom is set as 0.1 [20].

The characteristic time for ion wall loss, $\tau_{HI}$, is determined by the charged particle balance and the quasi-neutrality [7]. The characteristic wall loss times for molecular ions are weighted by a square root of their masses, because the ion wall loss is closely related to the Bohm velocity. The neutral wall loss is characterized by the characteristic wall loss times, respectively. The wall recombination factor ($\gamma$) of $H$ atom is set as 0.1 [20].

The effective diffusion length and the effective diffusion coefficient. For a cylindrical chamber of radius $R$ and length $L$, The effective diffusion length and the effective diffusion coefficient are given by [30]

$$\Lambda_{eff} = \left[ \left( \frac{2.405}{R} \right)^2 + \left( \frac{\pi}{L} \right)^2 \right]^{-1/2}$$

and

$$\frac{1}{D_{eff}} = \frac{1}{D_{Kn}} + \frac{1}{D_{\text{coll}}},$$

respectively. $D_{Kn}^X$ is the Knudsen diffusion coefficient of particle $X$ which can be approximated as

$$D_{Kn}^X = \frac{\Lambda_{eff}}{3} \sqrt{\frac{8k_b T_X}{\pi M_X}},$$

### Table 1. Volume processes and references of cross section or reaction rate.

| Volume Process | Description | Reference |
|----------------|-------------|-----------|
| $H + e \rightarrow H^+ + 2e$ | Atomic ionization (1x) | [21] |
| $H^+ + e \rightarrow H + h\nu$ | Radiative recombination (Rr) | [21] |
| $H^+ + 2e \rightarrow H + e$ | Three-body recombination (Tr) | [21] |
| $H_2^+ + e \rightarrow H + H^+ + e$ | Dissociative excitation (Dc2) | [21] |
| $H_2^+ + e \rightarrow 2H$ | Dissociative recombination (Dr2) | [21] |
| $H_2^+ + e \rightarrow 2H^+ + 2e$ | Dissociative ionization (DHz) | [21] |
| $H_3^+ + e \rightarrow H_2^+ + H + e$ | Dissociative recombination (Dr3-1, Dr3-2) | [21] |
| $H_3^+ + e \rightarrow 2H^+ + H^+$ | Dissociative excitation (D3c) | [21] |
| $H_2(\nu = 0) + H_2^+ \rightarrow H + H_2^+$ | $H_2^+$ formation (3F) | [21] |
| $H_2(\nu) + e \rightarrow H_2^+(\nu_2) + e$ | Vibrational excitation via $H_2^+$ resonant states (e-V) | [22–24] |
| $H_2(\nu) + e \rightarrow H_2(B^3\Sigma_g^+, \text{SIL}) + e$ | Excitation–radiative decay vibrational excitation/de-excitation (E-V) | [25] |

### Table 2. Wall loss frequency of the wall processes.

| Wall process | Wall loss frequency [s$^{-1}$] | Reference |
|--------------|-------------------------------|-----------|
| $H^+ + \text{wall} \rightarrow H$ | $1/\tau_{HI}$ | [7] |
| $H_2^+ + \text{wall} \rightarrow H_2(\nu = 0)$ | $1/\sqrt{2}\tau_{HI}$ | [7] |
| $H_3^+ + \text{wall} \rightarrow H_2(\nu = 0) + H$ | $1/\sqrt{3}\tau_{HI}$ | [7] |
| $H + \text{wall} \rightarrow 1/2H_2(\nu = 0)$ | $\gamma_{H_2} D_{H_2}^0/\Lambda_{HI}$ | [30] |
| $H_2(\nu) + \text{wall} \rightarrow H_2(\nu)$ | $\gamma_{H_2} D_{H_2}^0/\Lambda_{HI}$ | [30] |
where $T_X$ and $M_X$ are the temperature and the mass of particle $X$. $D_{\text{coll}}^X$ is the collisional diffusion coefficient given by

$$D_{\text{coll}}^X = \left( \sum_Y \frac{n_Y}{n} \frac{1}{D_{\text{coll}}^{X,Y}} \right)^{-1},$$  \hspace{1cm} (9)$$

where $D_{\text{coll}}^{X,Y}$ is the binary diffusion coefficient determined from the Chapman-Enskog theory. $Y$ is every collision partners, i.e. hydrogen atom and molecule in the present model. $n$ is the summation of each $Y$ particle density, $n_Y$. The binary diffusion coefficient is given by

$$D_{\text{coll}}^{X,Y} = 0.0018583 \sqrt{\frac{T_{\text{eff}}^X (1/M_X + 1/M_Y)}{p_Y (\sigma_X/2 + \sigma_Y/2)}},$$  \hspace{1cm} (10)$$

where $M_X$ and $M_Y$ are the mass in amu, $p_Y$ is the partial pressure of particle $Y$ in atm, $\sigma$ is the collision cross section in Angstrom ($\sigma_H = 1.1$ and $\sigma_{H_2} = 2.915$) and $\Omega^{(1,1)}_{X-Y}$ is the collisional integral of first order [32]. The wall relaxation coefficients of the vibrational states, $p_v$, are obtained from the literature [30, 33].

The particle balance model solves eighteen nonlinear equations to find eighteen unknowns. Since the diffusive flux is much larger than the advective flux, the left-hand side of equation (12) can be approximated as follows

$$\frac{d}{dz} \Gamma_Z \approx \frac{d}{dz} \left( -D \frac{d}{dz} N_v + N_v u_{\text{adv}} \right) \approx \frac{d}{dz} \left( -D \frac{d}{dz} N_v \right).$$  \hspace{1cm} (13)$$

To reduce the boundary conditions, equation (13) is further simplified by defining the diffusion flux as following:
- \frac{D}{d z} N_v = N_i u_{\text{diff}}, \quad \text{(14)}

where \( u_{\text{diff}} \) is the diffusive velocity defined as

\[ u_{\text{diff}} = -D \frac{d N_v}{d z} / N_v. \quad \text{(15)} \]

Finally, we obtain one-dimensional transport equation for neutral particles as following:

\[ u_{\text{diff}} \frac{d}{d z} N_v = \left( \sum_{\omega} \nu_{\omega}^{-v, v-1} N_v - \sum_{\omega} \nu_{\omega}^{v, v-1} N_v \right). \quad \text{(16)} \]

Note that equation (16) is only valid when the condition of \( |\nabla N_i| \gg |\nabla u_{\text{diff}}| \) is satisfied. For obtaining numerical solution, we discretized equation (16) as below:

\[ N_{i,k} = N_{i,k-1} + \Delta z / u_{\text{diff}} \left( \sum_{\omega} \nu_{\omega}^{-v, v-1} N_{i,k-1} - \sum_{\omega} \nu_{\omega}^{v, v-1} N_{i,k-1} \right), \quad \text{(17)} \]

where \( \Delta z \) is the infinitesimal thickness of the discretized disk as shown in figure 8. Equation (17) can be easily solved with one boundary condition. This boundary condition is nothing but the VDF of driver region obtained by solving the global particle balance equation described in section 3.1.

The physical meaning of equation (17) is that the hydrogen molecule density in vibrational state \( v \) at position \( z_2 \) is determined by the hydrogen molecule density in vibrational state \( v \) at position \( z_1 \) by the collision during the molecule travels \( \Delta z \) with the velocity \( u_{\text{diff}} \). The collision processes included in equation (17) are the same as those used in section 3.1. In equation (17), the source collision frequency, \( \nu_{\omega}^{v, v-1} \), and the loss collision frequency, \( \nu_{\omega}^{-v, v-1} \), are specifically expressed as following:

\[ \nu_{\omega}^{-v, v-1} = \sum_{\omega} (K_{e^{-v, v}} + K_{E^{-v, v}}) n_e + K_{p^{-v, v}} n_H + K_{V^{-v, v}} n_e + \left( \sum_{\omega} K_{V^{-v, v}}^\text{diff} N_{v, k-1} \right) \]

\[ + \sum_{\omega} (K_{V^{-v, v}}^\text{diff} N_{v, k-1, 1} + K_{V^{-v, v}}^\text{diff} N_{v, 1, k-1}) + \frac{D_{H}}{\text{eff}} \sum_{\omega} b_{\omega}^v, \quad \text{(18)} \]

\[ \nu_{\omega}^{v, v-1} = n_e \left\{ \sum_{\omega} (K_{e^{v, v-1}} + K_{E^{v, v-1}}) \right\} + n_H \left\{ \sum_{\omega} (K_{p^{v, v-1}} + K_{V^{v, v-1}}^\text{TP}) \right\} + K_{V^{-v, v}}^\text{diff} n_H \]

\[ + \sum_{\omega} (K_{V^{-v, v}}^\text{diff} N_{v, k-1}) + \sum_{\omega} (K_{V^{-v, v}}^\text{diff}^\text{L} N_{v, k-1, 1}) \]

\[ \quad + \frac{D_{H}}{\text{eff}} p_v. \quad \text{(19)} \]

The reaction rates of the electron impact processes are determined by the averaged electron temperature and density in the transport region. Here, the transport region is defined as the distance from the center of the driver region to the center of the extraction region. In this region, the fraction of ion species is assumed to be conserved.

### 3.3. Coronal model for Fulcher-\( \alpha \) spectrum

A coronal model in this study assumes that upper Fulcher states are populated by collisional excitation with electrons and the excited upper Fulcher states are depopulated by spontaneous radiative transition. Then, the vibrational distribution of the upper Fulcher states can be expressed as

\[ \frac{d N_{d,v'}}{d t} = n_e \sum_{v=0}^{14} (N_v K_{d,v'}^{0}) - \frac{N_{d,v'}}{\tau_{\text{life},d,v'}} = 0, \quad \text{(20)} \]

or \( N_{d,v'} = \tau_{\text{life},d,v'} n_e \sum_{v=0}^{14} (N_v K_{d,v'}^{0}), \quad \text{(21)} \)

where \( N_{d,v'} \) is the density of \((d, v')\) - state, \( \tau_{\text{life},d,v'} \) is the lifetime of \((d, v')\) -state, and \( K_{d,v'}^{0} \) is the reaction rate for the vibrationally resolved electronic excitation from \((X, v)\) -state to \((d, v')\) -state. According to the Frank–Condon principle and a weighting factor considering the electron temperature effect on the excitation cross section [34], the \( K_{d,v'}^{0} \) can be approximated as

\[ K_{d,v'}^{0} = q_{d,v'}^{0} K_{X,v'}^{0} = q_{d,v'}^{0} K_{X,0}^{0} \exp \left( \frac{\Delta G_X(v)}{k T_e} \right), \quad \text{(22)} \]

where \( q_{d,v'}^{0} \) is the Frank–Condon factor for the corresponding transition and \( \Delta G_X(v) \) is the vibrational energy difference, i.e. \( \Delta G_X(v) = G_X(v) - G_X(0) \). From equations (21) and (22), the VDF of the upper Fulcher state can be obtained as the function of \( N_v \) and \( T_e \) as following:
Table 3. List of negative ion destruction processes.

| Destruction Process | Description | Reference |
|---------------------|-------------|-----------|
| $H^+ + e \rightarrow H + 2e$ | Electron detachment (ED) | [21] |
| $H^+ + H^+ \rightarrow 2H$ | Mutual neutralization (MN-1) | [36] |
| $H^+ + H^+_2 \rightarrow H + H_e$ | Mutual neutralization (MN-2) | [36] |
| $H^+ + H^+_3 \rightarrow 2H + H_t$ | Mutual neutralization (MN-3) | [36] |
| $H^+ + H_2 \rightarrow H + e$ | Associative detachment (AD) | [21] |
| $H^+ + H \rightarrow 2H + e$ | Non-associative detachment (NAD) | [21] |

\[
N_{d,v'} = \sum_{i=0}^{14} \left( \frac{\Delta G_i(v)}{kT_e} \right) \exp \left( \frac{\Delta G_i(v)}{kT_e} \right). 
\]

Since the Fulcher-$\alpha$ intensity is the product of the radiation energy, transition probability and $N_{d,v'}$, the Fulcher-$\alpha$ intensity can be calculated by

\[
I_{v'v} = h \omega_A d_{v'v} A_{v'v} \sum_{i=0}^{14} \left( \frac{\Delta G_i(v)}{kT_e} \right) \exp \left( \frac{\Delta G_i(v)}{kT_e} \right),
\]

where $h \omega_A d_{v'v}$ is the radiation energy defined by the energy difference for the corresponding transition, i.e. $G_i(v') - G_i(v')$. The values for radiative transition probability, lifetime, Franck–Condon factor, and vibrational energy of $X^1Σ_g^+, d^1Σ_g^+$, and $d^1Π_u$ states are reported in the literature [35].

3.4. Determination of $H^-$ ion density

From the global model and the neutral particle transport model described in sections 3.1 and 3.2, respectively, we obtain the VDF of hydrogen molecules in the extraction region where $H^-$ ions are produced by DA reaction. The destruction of $H^-$ ions by the processes of electron detachment, mutual neutralization, associative and non-associative detachment by atom (see table 3) is also considered. Then, $H^-$ ion density in steady-state can be obtained as following:

\[
\frac{\partial n^-}{\partial t} = n_e \sum_{i} K_{DA}^\nu N_i - \left( K_{ED} n_e + \sum_{i=1}^{3} (K_{MN-i} n_{H^+} + (K_{AD} + K_{NAD}) n_H) \right)n^-= 0,
\]

or

\[
n^- = \frac{n_e \sum_{i} K_{DA}^\nu N_i}{K_{ED} n_e + \sum_{i=1}^{3} (K_{MN-i} n_{H^+} + (K_{AD} + K_{NAD}) n_H)}.
\]

It should be noted that the electron density and temperature measured in the extraction region by the Langmuir probe are used in the calculation of $H^-$ ion density.

4. Results and discussion

4.1. Overall characteristics of plasma parameters

Figure 9 shows the measured electron density and temperature with the operating pressure depending on the RF power. The electron temperature shows typical dependency on gas pressure and RF power [7]. It decreases with increasing operating pressure, but it hardly depends on RF power. On the other hand, the electron density depends strongly on both RF power and operating pressure. One can see that the electron temperatures in the extraction region are much lower than those in the driver region due to the magnetic filter field between the two regions. Owing to the filtering of high energy electrons, the electron densities decrease in the extraction region compared to those in the driver region by a factor of 2–3.

The electron density and temperature profiles measured by the Langmuir probe along the distance from the extraction hole are depicted in figure 10. The electron temperature decreases monotonically as the distance from the heating region increases. However, the electron density does not increase monotonically, but it has a peak value in front of the extraction region. This seems to be caused by low energy electron accumulation in the filter region decreasing perpendicular diffusion coefficients across the increasing filter field. More detailed experimental results and analysis of the role of magnetic filter field strength are described in our previous paper [9].

4.2. Electron kinetics regime

As previously reported [11], the EEDF show the bi-Maxwellian for low pressure discharges under $\sim$1 Pa. Such a bi-Maxwellization of the EEPF is known to be a common phenomenon of non-local electron kinetics [37]. Considering that our operating pressure regime covers 0.4–4 Pa, we need to examine the electron kinetics
Figure 9. (a) Electron temperatures and (b) densities versus the operating pressure measured with a Langmuir probe for various RF powers.

Figure 10. The measured electron density and temperature profiles along the distance from the extraction hole for the operating pressure of 1.3 Pa.
regime for deep understanding of our RF TCP H⁻ ion source. In the case of hydrogen gas, various electron-neutral collision processes in elastic or inelastic forms including ro-vibrational collision occur at entire electron energy range. Due to high ro-vibrational collision probability, local property dominates the electron kinetics even at relatively low hydrogen pressure \[12\]. In other words, the heated electrons in the vicinity of the antenna do not conserve their kinetic energy as they escape the skin layer due to various inelastic collisions. Therefore, the EEDFs can be considered as a spatially inhomogeneous function of the total electron energy at relatively lower pressure than those of noble gases such as argon.

To characterize the electron kinetics regime, electron energy relaxation length \( \lambda_e \) needs to be compared to the characteristic plasma scale. The electron energy relaxation length is given by \[38\]

\[
\lambda_e = \frac{\lambda_m \lambda_{\text{loss}}}{3},
\]

where \( \lambda_m \) is the total mean free path for momentum transfer collision and \( \lambda_{\text{loss}} \) is the mean free path accounting for all collisional energy loss processes given respectively as

\[
\lambda_m = \frac{1}{n_e \sigma_m},
\]

\[
\lambda_{\text{loss}} = \frac{1}{n_e \left[ \sigma_m 2m/M + \sigma_{\text{ex}} + \sigma_{\text{iz}} \right]},
\]

where \( n_e \) and \( M \) are the neutral gas density and the mass of hydrogen molecule, respectively. \( \sigma_m \), \( \sigma_{\text{ex}} \), and \( \sigma_{\text{iz}} \) represent the cross sections for momentum transfer, elastic collision, all kinds of excitation, and ionization collisions, respectively.

Figure 11 shows the calculated \( \lambda_e \) for a hydrogen plasma. One can see that \( \lambda_e \) has a minimum value at the electron energy of 2–3 eV. This is because the cross section for the vibrational excitation collision has a maximum value at this electron energy range. In addition, \( \lambda_e \) decreases with increasing the gas pressure due to significant energy loss via dominant collision with hydrogen molecules. It should be noted that \( \lambda_e \) is larger than the discharge chamber length of 10 cm for operating pressure of less than \( \sim 1 \) Pa. This implies that the transition from local to non-local electron kinetics occurs at the operating pressure approximately \( \sim 1 \) Pa, which is well matched with our previous experiments \[11\].

The non-local electron kinetic property for low pressure discharge is clearly observed in the EEPFs measured in the middle of the discharge chamber (5 cm from the extraction hole), as shown in figure 12. In the case of the gas pressure of 0.4 Pa, the minimum \( \lambda_e \) is almost 20 cm, i.e. much larger than the chamber length. Hence, the EEPF has a bi-Maxwellian distribution. When the gas pressure increases, the EEPF becomes the Maxwellian and the population of low energy electrons whose energy is less than 2 eV increases significantly due to the increase in vibrational excitation collision process. The depletion of the electron energy distribution near the electron kinetic energy of \( \sim 3 \) eV is observed. It occurs primarily due to the shortest energy relaxation length at the electron energy of \( \sim 3 \) eV, as shown in figure 11. The low energy depletion disappears when the RF power is increased, i.e. the frequency scale of the electron–electron collision greatly exceeds the inelastic collision between low energy electrons and neutral species.
Recently, we observed that the magnetic filter field changes the bi-Maxwellian EEPF to the low-energy populated Maxwellian \[39\], which is similar to the effect of gas pressure on the EEPF. We are attempting to understand the mechanism of magnetic filter in the context of the enhanced electron-neutral collision by the electron confinement due to the filter field. Our understanding so far is that the enhanced collision probability by magnetic confinement increases the locality of electrons, thus the population of low energy electrons increases, similar to the effect of gas pressure as shown in figure 12.

4.3. Relaxation of vibrationally-excited molecules

Hydrogen molecules which are vibrationally-excited in the driver region are transported to the extraction region across the filter field region in the RFTCP H\(^-\) ion source. Since there are always collisions between neutral–neutral or charged–neutral particles, the VDF of the hydrogen molecules must be changed during the transport. Figure 13 clearly shows the influence of the transport of neutral particles in the analysis of the Fulcher-\(\alpha\) spectrum intensity ratio measured at the extraction region. When the global model is only applied while ignoring neutral particle transport, the calculated Fulcher-\(\alpha\) spectrum intensity ratio cannot reproduce the experimental result. However, the combined model including the 1D neutral particle transport shows a fairly good agreement with the experimental result. According to the numerical model, the higher states of the VDF decay to the lower states during the transport so that the VDF calculated at the extraction region is greatly different from that calculated at the driver region, as shown in figure 14. It should be noted that the vibrational temperature, which is conventionally determined from the 4 lower states \[40\], cannot be a representative of the VDF. Indeed, conventional method gives a much higher vibrational temperature for the extraction region depicted as a dashed line in figure 14. This means that the Fulcher-\(\alpha\) spectrum is not sufficient to diagnose the highly excited vibrational states.

The influence of the relaxation of vibrationally-excited hydrogen molecules during the transport across the magnetic filter region is more clearly demonstrated in the calculation of H\(^-\) ion density at the extraction region. As shown in figure 15, when the transport of neutral particles is ignored, the calculated H\(^-\) ion density does not catch the experimental result at all, especially for high pressure regime. One can see that the agreement with the experiment is significantly improved, when the effect of neutral transport is included. This is mainly due to the consideration of VDF which is greatly changed during the transport especially for high pressure regime. Indeed,
it is observed that higher states which play a key role for the production of H$^-$ ions via DA reaction decay to lower states during the transport, as shown in figure 14.

Figures 16(a) and (b) compare the mean free path of each reaction related to the relaxation of the VDF for two extreme pressures of 0.4 Pa and 4 Pa. In these calculations, we use the gas temperatures ($T_g$) of 1050 K and 1300 K for 0.4 Pa and 4 Pa, respectively, which are determined by spectroscopic method [14]. Most significant 7 reactions among all vibrationally-resolved reactions listed in table 1 are depicted in figure 16. In the high pressure regime, the vibrational-translation (V-T) energy transfer acts as a key relaxation process due to frequent neutral–neutral collisions. For the relaxation of low vibrational states less than 4, e-V process also seems to play an important role, but its influence to the relaxation of high vibrational numbers is not clear due to the lack of cross section data. Note that the scaling law given by Gorse et al [22] and Xiao et al [24] is used in the calculation of e-V reactions for higher states. As the gas pressure decreases, the V-T process decreases as well. At sufficiently low pressure of 0.4 Pa, the dominant relaxation process occurs by the interaction of vibrationally-excited molecules with wall. Meanwhile, the E-V reaction also increases due to a relatively high electron temperature at low pressure. Since the E-V reaction can excite the vibrational state, the vibrational relaxation by the wall can be compensated by the E-V reaction. Thus, the decay of the vibrational state occurs weakly at the low pressure.
However, there is no compensation reaction against the V-T relaxation process at the high pressure, thus vibrational relaxation during the neutral transport is more significant for the operation of $H^-$ ion source at high pressure regime.

Figure 15. Comparison of the calculated $H^-$ ion densities for two different conditions, i.e. whether the neutral transport is considered or not, with the experimental results.

Figure 16. Mean free path of each reaction related to the relaxation of VDF for the operating pressure of (a) 0.4 Pa and (b) 4 Pa.
4.4. Effect of the degree of dissociation

In addition to the VDF relaxation during the neutral transport, the degree of dissociation plays an important role because it determines the number density of hydrogen molecules as a precursor of H\(^-\) ion production. Also, hydrogen atoms contribute the relaxation of VDF via V-Ta process and the destruction of H\(^-\) ions via associative or non-associative detachment reactions. We measured the variation of the degree of dissociation with the gas pressure using optical diagnostic method [14, 40]. As shown in figure 17, the degree of dissociation increases as the gas pressure decreases because the electron temperature increases with decreasing the gas pressure. Due to the wall recombination loss of hydrogen atoms during the downward transport, the degree of dissociation measured at the extraction region is slightly smaller than that measured at the driver region. Its discrepancy becomes larger with increasing the gas pressure because the transport time of neutral particles increases with the gas pressure. Higher degree of dissociation at the low pressure regime partly explains the typical trend of H\(^-\) ion density versus the gas pressure as shown in figure 15. At low pressure, the relaxation of the VDF does not play an important role as discussed in section 4.3. However, the increased degree of dissociation at low pressure greatly reduces the number of hydrogen molecules who participate in H\(^-\) ion production in the extraction region.

**Figure 17.** Variation of the degree of dissociation with gas pressure for RF power of 700 W.

**Figure 18.** Dependence of the electron density, the degree of dissociation, and H\(^-\) ion density on the RF power for a fixed gas pressure of 1.3 Pa.

4.4. Effect of the degree of dissociation

In addition to the VDF relaxation during the neutral transport, the degree of dissociation plays an important role because it determines the number density of hydrogen molecules as a precursor of H\(^-\) ion production. Also, hydrogen atoms contribute the relaxation of VDF via V-Ta process and the destruction of H\(^-\) ions via associative or non-associative detachment reactions. We measured the variation of the degree of dissociation with the gas pressure using optical diagnostic method [14, 40]. As shown in figure 17, the degree of dissociation increases as the gas pressure decreases because the electron temperature increases with decreasing the gas pressure. Due to the wall recombination loss of hydrogen atoms during the downward transport, the degree of dissociation measured at the extraction region is slightly smaller than that measured at the driver region. Its discrepancy becomes larger with increasing the gas pressure because the transport time of neutral particles increases with the gas pressure. Higher degree of dissociation at the low pressure regime partly explains the typical trend of H\(^-\) ion density versus the gas pressure as shown in figure 15. At low pressure, the relaxation of the VDF does not play an important role as discussed in section 4.3. However, the increased degree of dissociation at low pressure greatly reduces the number of hydrogen molecules who participate in H\(^-\) ion production in the extraction region.
Therefore, the reduction of $H^-$ ion density at low pressure regime is affected by the degree of dissociation as well as the reduced electron density and the existence of high energy electrons owing to insufficient filtering.

Dependence of the $H^-$ ion density on the RF power can be explained in the same manner. With increasing the RF power, the electron density increases while keeping the electron temperature unchanged, as shown in figure 9. The higher the electron density, the larger the degree of dissociation and the smaller the amount of hydrogen molecules. This results in the reduced production of $H^-$ ions due to the low population of vibrationally-excited hydrogen molecules and the frequent loss of $H^-$ ions by associative or non-associative detachment with hydrogen atoms. Hence, $H^-$ ion density does not increase with increasing the RF power, but it saturates at a certain level of the RF power, as shown in figure 18. This result reveals that the effort to increase $H^-$ ion density by augmenting the input power might not be a clever solution in terms of power efficiency.

5. Conclusion

In this paper, we present a physics-based approach to interpret the mechanism of $H^-$ ion production in the volume production RF TCP $H^-$ ion source at SNU. Based on the careful EEPF measurements, we characterize the regime of electron kinetics which plays a crucial role for the generation of vibrationally-excited molecules and $H^-$ ions in the driver and the extraction region, respectively. We find that our RF TCP $H^-$ ion source has a transition from nonlocal to local at $\sim 1$ Pa. We hope that this approach provides a new finding on the role of magnetic filter field which remains unsolved up to date. In addition, we consider the influence of the relaxation of vibrationally-excited hydrogen molecules during the transport from the driver region to the extraction region both numerically and experimentally. Analysis with the 0D global model combined with the 1D neutral transport model greatly improves our understanding on the detailed mechanism of $H^-$ ion production in the volume production $H^-$ ion source. It is revealed that VDF relaxation plays an important role at high pressure regimes due to frequent collisions between neutral particles. On the other hand, it is found that the decrease in $H^-$ ion density at low pressure regime occurs mainly due to the decrease in density of hydrogen molecules due to high degree of dissociation. Therefore, the $H^-$ ion source has the optimum operating pressure near $\sim 1.3$ Pa, slightly depending on the RF power. With increasing the RF power, the electron density increases but the degree of dissociation also increases. Hence, $H^-$ ion density increases with increasing the RF power, but it saturates when the RF power is further increased. It means that there is an optimum RF power for obtaining the maximum $H^-$ ion density when the power efficiency is considered. At the same time, it also means that we need to approach in a different way to increase $H^-$ ion beam current further in the present $H^-$ ion source.

Strengthening the magnetic filter field for low pressure operation and control of the electron energy distribution using additional magnetic field we are trying recently might be the solutions. Otherwise, entirely different type of $H^-$ ion source in which the extraction region is separated from the driver region could be tried in the next step. The findings presented here can be utilized as a design tool for such an innovative type of $H^-$ ion source.

Acknowledgments

This research was supported partly by National R&D Program through the National Research Foundation of Korea (NRF) funded by the Ministry of Science, ICT & Future Planning (Nos. 2014M1A7A1A03045367 and 2014M1A7A1A02030165) and partly by Global PhD Fellowship Program through the National Research Foundation of Korea (NRF) funded by the Ministry of Education (2015H1A2A1033741).

References

[1] Hassouni K, Gicquel A and Capitelli M 1998 Chem. Phys. Lett. 290 502–8
[2] Aleleiferis S, Tarvainen O, Svarnas P, Bacal M and Béchu S 2016 J. Phys. D: Appl. Phys. 49 095203
[3] Bacal M and Wada M 2015 Applied Physics Reviews 2 021305
[4] Jeong H S, Kim Y J, Hong I S, Park D H and Hwang Y S 2006 Rev. Sci. Instrum. 77 03A536
[5] An Y, Jung B and Hwang Y S 2010 Rev. Sci. Instrum. 81 02A702
[6] An Y, Cho W H, Chung K J, Lee K, Jang S, Lee S G and Hwang Y S 2012 Rev. Sci. Instrum. 83 02A727
[7] Chung K J, Jung B K, An Y, Dang J J and Hwang Y S 2014 Rev. Sci. Instrum. 85 02B119
[8] Jung B K, Dang J J, An Y H, Chung K J and Hwang Y S 2014 Rev. Sci. Instrum. 85 02B112
[9] Cho W H, Dang J J, Kim Y J, Chung K J and Hwang Y S 2016 Rev. Sci. Instrum. 87 02B136
[10] Kim Y J, Cho W H, Dang J J, Chung K J and Hwang Y S 2016 Rev. Sci. Instrum. 87 02B117
[11] Huh S R, Kim N K, Jung B K, Chung K J, Hwang Y S and Kim G H 2015 Phys. Plasmas 22 033506
[12] Kim Y J, Cho W H, Dang J J, Chung K J and Hwang Y S 2016 Phys. Plasmas 23 023511
[13] Bacal M 2000 Rev. Sci. Instrum. 71 3981–4006
[14] Dang J J, Chung K J and Hwang Y S 2016 Rev. Sci. Instrum. 87 053503
[15] Jung H D, Park J Y, Chung K J and Hwang Y S 2007 IEEE Trans. Plasma Sci. 35 1476–9
[16] Gaboriau F and Boeuf J P 2014 Plasma Sources Sci. Technol. 23 065032
[17] Taccogna F, Minelli P, Diomedè P, Longo S, Capitelli M and Schneider R 2011 Plasma Sources Sci. Technol. 20 024009
[18] Pagano D, Gorse C and Capitelli M 2007 IEEE Trans. Plasma Sci. 35 1247–59
[19] Iordanova S, Paunska T and Pashov A 2015 J. Phys. D: Appl. Phys. 48 485204
[20] Zorat R and Vender D 2000 J. Phys. D: Appl. Phys. 33 1728–35
[21] Janev R K, Keiter D and Samm U 2003 Collision processes in low-temperature hydrogen plasmas Forschungszentrum Jülich Report Jul-4105 (Jülich)
[22] Gorse C, Capitelli M, Bacal M, Bretagne J and Laganà A 1987 Chem. Phys. 117 177–95
[23] Capitelli M, Celiberto R, Esposito F, Laricchiuta A, Hassouni K and Longo S 2002 Plasma Sources Sci. Technol. 11 A7
[24] Xiao B, Kado S, Kajita S, Yamasaki D and Tanaka S 2005 J. Nucl. Mater. 337–339 1082–6
[25] Celiberto R, Janev R K, Laricchiuta A, Capitelli M, Wadehra J M and Atems D E 2001 At. Data Nucl. Data Tables 77 161–213
[26] Celiberto R, Capitelli M and Laricchiuta A 2002 Phys. Scr. T96 32–44
[27] Vibrationally Resolved Excitation and Charge Transfer for Collisions Among Vibrationally Excited H₂ With H⁺ and Vibrationally Excited H₂⁺ With H ORNL-Controlled Fusion Atomic Data Center (http://www-cfadc.phy.ornl.gov/h2mol/homeh.html)
[28] Mandy M E and Martin P G 1999 J. Chem. Phys. 110 7811–20
[29] Matveyev A A and Silakov V P 1995 Plasma Sources Sci. Technol. 4 606–17
[30] Mosbach T 2005 Plasma Sources Sci. Technol. 14 610–22
[31] Bird R B, Stewart W E and Lightfoot E N 2002 Transport Phenomena (New York: Wiley)
[32] Gorse C and Capitelli M 2001 Collision integrals of high temperature hydrogen species Atomic and Plasma-Material Interaction Data for Fusion (APID Series) vol 9 (Vienna: International Atomic Energy Agency)
[33] Hiskes J R and Karo A M 1989 Appl. Phys. Lett. 54 508–10
[34] Fantz U and Heger B 1998 Plasma Phys. Control. Fusion 40 2023–32
[35] Fantz U and Wunderlich D 2006 At. Data Nucl. Data Tables 92 853–973
[36] Mosbach T 2005 Plasma Sources Sci. Technol. 14 351–62
[37] Godyak V A, Piejak R B and Alexandrovich B M 2002 Plasma Sources Sci. Technol. 11 525
[38] Lieberman M A and Lichtenberg A J 2005 Principles of Plasma Discharges and Materials Processing (Hoboken, NJ: Wiley)
[39] Kim J Y, Cho W H, Dang J H, Kim C S, Chung K J and Hwang Y S Enhancement of negative hydrogen ion production at low pressure by controlling the electron kinetics property with transverse magnetic field Plasma Sources Sci. Technol. (accepted)
[40] Fantz U, Falter H, Franzen P, Wunderlich D, Berger M, Lorenz A, Kraus W, McNeely P, Riedl R and Speth E 2006 Nucl. Fusion 46 S297–306