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Collisional-radiative model of atomic hydrogen in high velocity plasma flow

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Abstract: A collisional-radiative model is applied to investigate the physical kinetic processes of hydrogen plasma arcjet. The results show that in the constrictor region of arcjet, the electron impact excitation and ionization are dominant. In the downstream region of arcjet, it is found that electron impact de-excitation and recombination processes are dominant. For low lying excited states, radiative processes are very important, while for high lying excited states, electron impact processes are dominant.

Keywords: collisional-radiative model, atomic hydrogen, high velocity plasma flow.

1. Introduction

Collisional-radiative models are suitable tools to study chemical reaction processes in plasma system. Their main interest is indeed to access to the source term of the balance equation of a species. In past several decades, these methods are widely used in analyzing argon, helium, nitrogen, air discharges. In most of these applications, the plasma is in stationary conditions for a velocity not too high, and hence the various atoms and mixtures can be calculated without considering the time evolution of the species. While, in high velocity plasma flow, such as highly expanded arc in arcjet nozzle, the situation is more complicated, the characteristic time associated to the convective derivative in species balance equation may decrease sufficiently to give a significant role to the hydrodynamics even for highly excited atoms. The chemical reactions source term may be time dependent. Therefore, the calculation of the temporal evolution of the population densities only due to chemistry is needed [1].

Since the flow velocity within arcjet is large, with fluid residence times in the nozzle of the order of 1 μs, excitation or ionization nonequilibrium occurs because the fluid-dynamic time scales are comparable to the relaxation times for chemical reactions. This study is to focus attention on the evolution of hydrogen excited species in order to provide a better understanding of chemical reaction processes in different regions of arcjet thruster.

2. Collisional Radiative Model

The high velocity plasma flow system studied in this work consists of ground state of atomic hydrogen (H), excited states (H(i)), protons (H^+), and electrons. The levels of hydrogen excited species are distinguished by their principal quantum number up to a maximum value of 20. It is assumed that the plasma is electrical neutral and optically thin and the distributions of energies of particles are assumed Maxwellian. The kinetic processes employed in the modelling study are as follows, (i) the electron impact excitation and de-excitation process; (ii) the electron impact ionization and three-body recombination process; (iii) spontaneous emission; (iv) radiative recombination.

$$H(i) \xrightarrow{A_{ij}} H(j) + h\upsilon \qquad i > j \tag{1}$$

$$H^+ + e \xrightarrow{R_i} H(i) + h\upsilon \tag{2}$$

$$H(i) + e(\varepsilon) \xleftrightarrow{C^e_{ij}} H(j) + e(\varepsilon - \varepsilon_{ij}) \tag{3}$$

$$H(i) + e(\varepsilon) \xleftrightarrow{S^e_i} H^+ + e(\varepsilon - \varepsilon_i) + e \tag{4}$$

where ε_{ij} and ε_i represent, respectively, the threshold energy of the transition $i \to j$ and the ionization energy from the *i*-th level. It should be noted that atom-atom and ion-atom collisions have been neglected assuming that the electron-atom collision frequency dominates the kinetics.

Rate coefficients for electron-atom collisions are calculated from electron energy distribution functions (EEDF) using the equation:

$$C_{ij} = \int_{E_{min}}^{\infty} f(\varepsilon)\sigma(\varepsilon)v(\varepsilon)d\varepsilon \tag{5}$$

where $f(\varepsilon)$ represents the EEDF, $\sigma(\varepsilon)$ the cross section of the transition between atomic levels *i* and *j*, $v(\varepsilon)$ the velocity of the electron of kinetic energy ε and E_{min} the threshold energy of the process. The inelastic cross sections and Einstein coefficients are taken from available databases [2, 3].

Cross-sections for superelastic collisions and three-body recombination are calculated using the detailed balance principle [4]:

$$\sigma_{deexct}(j, E^*) = \frac{g_j}{g_i}\frac{E}{E^*}\sigma_{exct}(j, i, E) \tag{6}$$

$$\sigma_{3b-recomb}(E^*)(n_e)^{-1} = \frac{g_i}{2g_1^+}\frac{E}{E^*}\left(\frac{h^2}{2\pi m_e k T_e}\right)^{3/2}\sigma_{ioniz}(E) \tag{7}$$

where g_i is the statistical weight of the *i*-th level and g_1^+ is the statistical weight of the ion ground state, *h* the Planck constant, m_e the electron mass, *k* the Boltzmann constant and T_e the electron temperature. Eq. (7) is obtained assuming that energy after the ionization of the initially bound electron is zero. Radiative recombination rate coefficients have been calculated according to the work of Johnson [5].

According to the kinetic processes mentioned above, the collisional-radiative model consists of a set of master equations, including ion, electron concentrations and each level of atomic hydrogen:

$$\frac{dn_+}{dt} = \frac{dn_e}{dt} = -n_e n_+ \left(\sum_i R_i + n_e \sum_i Q_i^e \right) + n_e \sum_i S_i^e n_i \qquad (8)$$

$$\frac{dn_i}{dt} = n_e \sum_{i \neq j} n_j C_{ji}^e - n_i n_e \left(S_i^e + \sum_{j \neq i} C_{ij}^e \right) + n_+ n_e^2 Q_i^e$$
$$- n_i \sum_{j<i} A_{ij} + \sum_{j>i} A_{ji} n_j + n_+ n_e R_i \qquad (9)$$

where A_{ij} is the Einstein coefficients of the radiative decay, R_i is the rates of radiative recombination, C_{ij}^e and C_{ji}^e, with $i < j$, are, respectively, the rates of electron impact excitation and deexcitation, S_i^e and Q_i^e are the rates of electron impact ionization and three-body recombination.

Finally, the electrical neutrality of the plasma imposes $n_e = n_+$. The state equation of this plasma system is written as

$$p = \sum_i n_i k T_h + n_e k T_e + n_+ k T_h \qquad (10)$$

The system of differential equations has been integrated numerically using LSODE [6], a routine specifically designed to solve stiff ordinary differential equations. In each set of calculations, T_e and p are kept to be constant.

3. Results and Discussion

The input conditions of the argon collisional radiative model, such as electron number density, atom number density, temperatures and pressures within arcjet thruster, are taken from the numerical modeling results of low power arcjet, reported by Butler [7, 8]. In order to compare the kinetic process features within the arcjet thruster, the plasma parameters are taken along the axis while at the different part inside arcjet thruster (x=-12 mm, x=-8 mm, x=-6 mm in reference [8]), which are shown in Table 1. The initial distribution of all excited states for each case is assumed to follow Boltzmann distribution.

Table 1. Input plasma parameters used in the collision radiative model

Axis position	Electron density (m^{-3})	Atom density (m^{-3})	Temperature (K)	Pressure (Pa)
constrictor	1.0×10^{23}	4.0×10^{22}	40000	131000
downstream	5.0×10^{20}	4.0×10^{21}	8600	2600
downstream	2.0×10^{20}	3.0×10^{21}	4800	1200

Figure 1 shows the temporal evolution of various excited states in the constrictor region of arcjet. It is seen that the number densities of excited states decrease rapidly from the initial distribution to achieve steady state. It is also shown from Fig. 1 that the plasma is in an ionizing state, since the electron number density is larger than those of other species. Figure 2 shows the kinetic processes of low lying excited states and high lying excited states under steady state condition. For the ground state, electron impact excitation process prevails over deexcitation process. It is also shown that the spontaneous radiation process from n=2 to ground state is also very important. For excited states, electron impact excitation, deexcitation processes, electron impact ionization and three body recombination processes achieve equilibrium. Compared with kinetic processes in which electron is involved, radiative processes become less important. Figure 3 shows the populations of excited hydrogen atoms at time t=1.0×10^{-6} s. The results show that in the constrictor region of arcjet thruster, where the most of working gas is heated by plasma arc to a high ionization degree, the population densities of hydrogen excited species follow Boltzmann distribution.

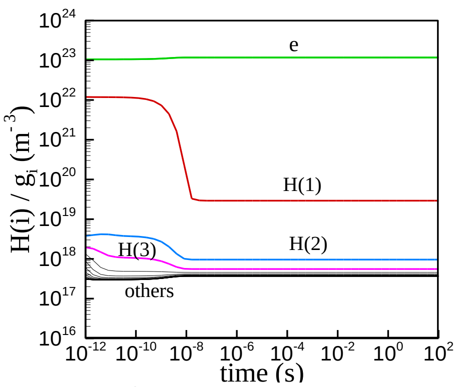

Fig. 1 Time evolution of densities divided by their statistical weight in the constrictor region of arcjet

Fig. 2 Flows of electrons for low lying excited states and high lying excited states ($m^{-3}s^{-1}$). The straight arrows denote the non-radiative processes and the dashed arrows represent the radiative processes. The brackets denote the power of 10 by which the number should be multiplied (t=1.0×10^{-6} s).

Figure 4 shows the temporal evolution of various excited states in the downstream of arcjet nozzle (x=-8 mm). It is shown that the population densities of various excited states achieve quasi-steady-state within 10^{-8} s~10^{-6} s. In this case, a quasi-equilibrium number density of excited system is established almost instantaneously without the number densities of free electrons being appreciably altered [9]. When the density is in a quasi-steady-state, the CR model allows the identification of the main processes responsible for the excited atom concentrations measured as well as the calculation of global rate coefficient, for example, the rate coefficients of ionization, recombination and dissociation.

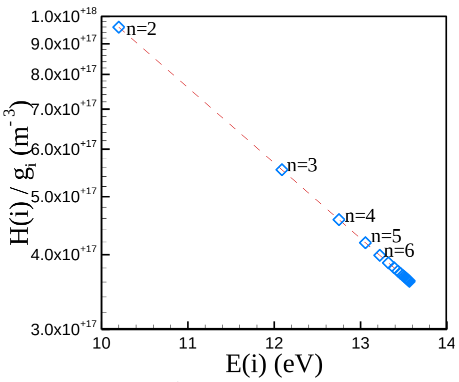

Fig. 3 Transient populations of excited hydrogen atoms in the constrictor region of arcjet (t=1.0×10^{-6} s).

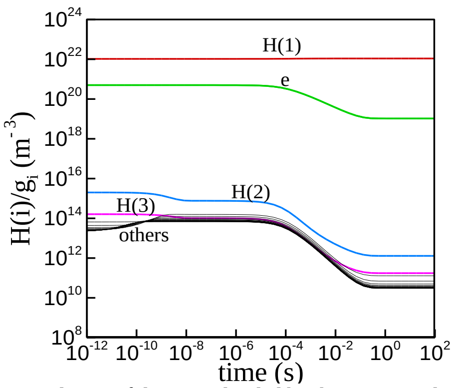

Fig. 4 Time evolution of densities divided by their statistical weight in the downstream region of arcjet

In downstream of arcjet nozzle, the de-excitation processes of excited species and recombination processes are dominant. The corresponding kinetic processes of low lying excited states and high lying excited states in a quasi-steady-state are shown in Fig. 5. It is interesting to find that the dominant depopulating process of ground state is the excitation to the adjacent higher-lying level, while the dominant populating process of ground state is spontaneous emission from excited states. For low lying excited states, the deexcitation processes from the adjacent higher-lying level prevail over the excitation processes. For high lying excited states, the electron impact excitation and deexcitation are dominant processes. It can be seen that the electron impact excitation and deexcitation processes achieve equilibrium, and the electron impact ionization and three-body recombination processes are also in an equilibrium state. The transient populations of excited hydrogen atoms are shown in figure 6. It is seen that for low lying excited states, the densities are lower than the Boltzmann distribution, while high lying excited states are in partial equilibrium which accounts for the major contribution of the electron induced processes for the excitation.

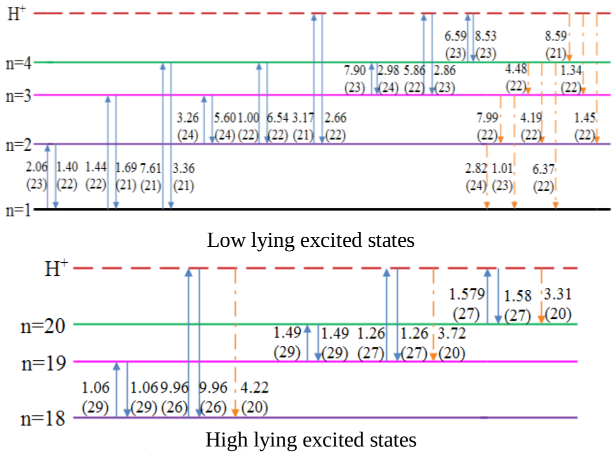

Fig. 5 Flows of electrons for low lying excited states and high lying excited states (m^{-3}s^{-1}). The straight arrows denote the non-radiative processes and the dashed arrows represent the radiative processes. The brackets denote the power of 10 by which the number should be multiplied (t=1.0×10^{-6} s).

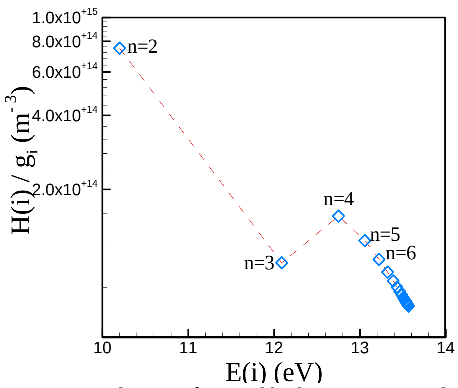

Fig. 6 Transient populations of excited hydrogen atoms in the downstream of arcjet nozzle (t=1.0×10^{-6} s).

The temporal evolution of various excited states in downstream of the arcjet thruster (x=-6 mm) is shown in figure 7, which demonstrate that the plasma is dominated by deexcitation and recombination processes. In early times, the recombination processes play an important role on the evolution of number densities of excited states, so the excited level populations increase and reach a plateau (at time about 10^{-7} s~10^{-5}s), which corresponds to the stationary state of the excited state populations. With the increase of time, the number densities of excited states decrease owing to the electron impact deexcitation processes. The kinetic processes of various excited states in quasi-steady-state are shown in figure 8. It is seen that the kinetic processes of plasma species at the downstream (x=-8 mm) are different from those at position of x=-6 mm. For ground state, the radiative processes dominant, i.e. the spontaneous emission rate is much larger than that of electron impact deexcitation and recombination rate, which lead to overpopulation of the ground state atom. For n=2 state, radiative recombination rate is larger than three body recombination rate. The main populating process is ladder-like de-excitation from n=3 state, the main depopulating process is spontaneous transition to the ground state. For other low lying excited states, the probabilities for collisional de-excitation are larger than the excitation probabilities. Furthermore, the three-body recombination rate is larger than radiative recombination rate.

The transient populations of excited hydrogen atoms in quasi-steady-state are shown in figure 9. The densities of low lying excited states deviate significantly from the Boltzmann distribution due to radiative processes. The high levels are almost in equilibrium owing to the ladder-like excitation and ionization caused by electron impact. It can be seen that radiative processes become very important for low lying excited states due to low pressure at the downstream of arcjet.

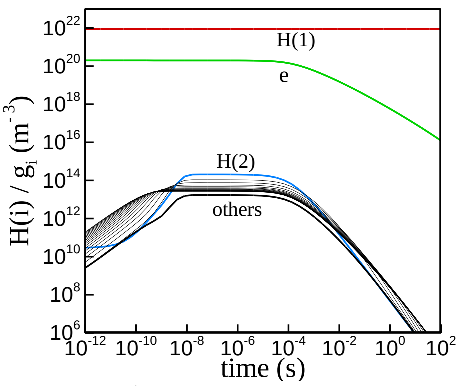

Fig. 7 Time evolution of densities divided by their statistical weight in the downstream region of arcjet

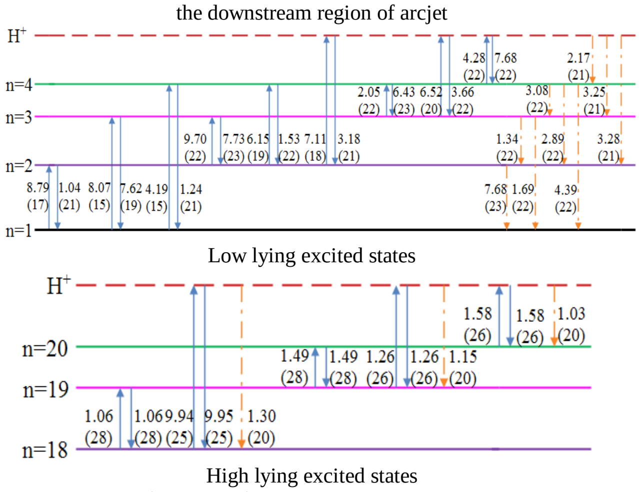

Fig. 8 Flows of electrons for low lying excited states and high lying excited states ($m^{-3}s^{-1}$). The straight arrows denote the non-radiative processes and the dashed arrows represent the radiative processes. The brackets denote the power of 10 by which the number should be multiplied ($t=1.0\times10^{-6}$ s).

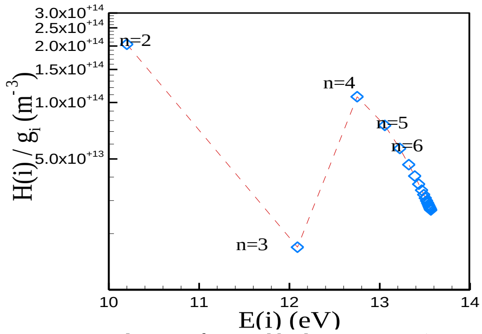

Fig. 9 Transient populations of excited hydrogen atoms ($t=1.0\times10^{-6}$ s).

4. Conclusions

A collisional-radiative model is applicable to hydrogen plasma arcjet thruster to investigate the kinetic processes in different regions of arcjet. It is found that in the constrictor region of arcjet the population densities of hydrogen excited states follow the Boltzmann distribution. The electron impact processes are dominant and radiative processes can be neglected. In the downstream region of arcjet, the electron impact deexcitation and recombination processes are dominant. The calculated population densities suggest low lying excited states deviate from Boltzmann distribution, while high lying excited states follow Boltzmann distribution. It is found that for ground state, spontaneous emission is dominant, while for low lying states, the electron impact deexcitation from adjacent states are dominant processes.

5. References

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6. Acknowledgment

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