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Global Model of Inductively coupled CH_4/H_2 Plasmas
誘導性結合型CH_4/H_2プラズマのグロ-バルモデル

Takashi Kimura
木村　高志

Nagoya Institute of Technology, Gokiso-cho, Showa-ku, Nagoya 466-8555, Japan
名古屋工業大学　〒466-8555　名古屋市昭和区御器所町

A global model for electropositive plasma is used in order to understand the plasma chemistry in CH_4/H_2 plasmas assuming the Maxwellian electron energy distribution. Most of CH_4 molecules are easily dissociated, so that the dominant neutral species are hydrogen molecules, CH_4 molecules and hydrogen atoms, followed by the hydrocarbons such as C_2H_m (m=1,2 and 4). The dominant ions are CH_5^+, C_2H^+, $C_2H_3^+$, $C_2H_5^+$, and $C_3H_5^+$ at low hydrogen fractions, whereas the density of H_3^+ becomes dominant ion species as hydrogen fraction increases. Some measurable parameters such as electron density and its temperature are compared with the model results, obtaining reasonably good agreement.

1. Introduction

Theoretical studies of the plasmas in hydrocarbon-based feedstock for the PECVD of diamond-like, graphite-like and amorphous carbon films [1-5] have been reported. The global model is one of the methods that have been used in order to treat the physical and chemical processes in the plasmas. On the other hand, the models, in which the plasma chemistry and surface chemistry are combined, have been developed for thin-film deposition and etching purposes.[4,5] Mantzaris et al [4] combined plasma chemistry and surface chemistry models to study the diamond-like-carbon film deposition process for an rf capacitively coupled CH_4 discharge and showed that the surface reaction can greatly affect on the composition of neutral radicals in the plasma.

The objectives of this research are to investigate the effect of hydrogen addition on plasma parameters in inductively coupled rf (13.56MHz) CH_4/H_2 plasmas and to understand the plasma chemistry of such plasmas using the global model.

2. Description of Model

A global model for electropositive plasma is applied to the CH_4/H_2 plasmas in a cylindrical chamber of radius R (= 80mm) and length L (= 75mm), since the ratio of negative ion to electron density should be assumed to be very low. 27 neutral species (CH_j (j=1-4), C_2H_j(j=1-6), C_3H_j(j=1-8), C_4H_j(j=7-9), C, C_2, H_2, H(1s), H(2s), and H(2p)) and 27 charged species (electrons, CH_j^+(j=1-5),$C_2H_j^+$(j=1-6),$C_3H_j^+$(j=1-8),$C_4H_j^+$(j=7-9), C^+, H^+, H_2^+, and H_3^+) are considered in the model. All neutral species are assumed to be uniform over the chamber. Electrons, which have a Maxwellian energy distribution with electron temperature T_e, are also assumed to be uniform over the chamber. All positive ion species, which are assumed to have the same profile, must satisfy the quasineutrality, $n_e = n_+$, where n_+ is the sum of the positive ion densities. The gas temperature in the discharge space is assumed to be 600 K.

The surface processes for hydrogen atoms, CH_3, and CH_2, are incorporated in the model. The adsorption and desorption processes, and the adsorbed layer processes, in which the adsorbed species can react with the atomic hydrogen from the plasma yielding the gas-phase products, are mainly considered using same parameters [4,5]. The ions are directly incorporated in the growing film. Therefore, the sticking coefficient for ions can be assumed to be unity. The incorporation of ions is considered to occur on the whole surface, whereas the reactions between the adsorbed layer and the ions are not considered in this model.

The equations for each particle and power balance are then solved by the Runge-Kutta numerical method to obtain equilibrium.

3. Results and Discussion

In Figs. 1(a)-1(b), the electron density and its temperature are shown as a function of hydrogen fraction X_{H2}, together with the experimental results. The measured and calculated n_e gradually decrease with increasing X_{H2}. On the other hand, the measured and calculated temperatures slightly increase with the increase in X_{H2}. Since the ionization rates of hydrocarbons by electron impact are much larger than those of hydrogens, the electron temperature increases with the decrease in the density ratio in order to satisfy the balance of the charged species productions and loss to the wall. The increase in electron temperature should contribute to the increase in the collisional energy loss between electrons and neutral species, such as hydrogens and hydrocarbons, resulting in the decrease in electron density with increase in X_{H2} under the same power injected into plasmas.

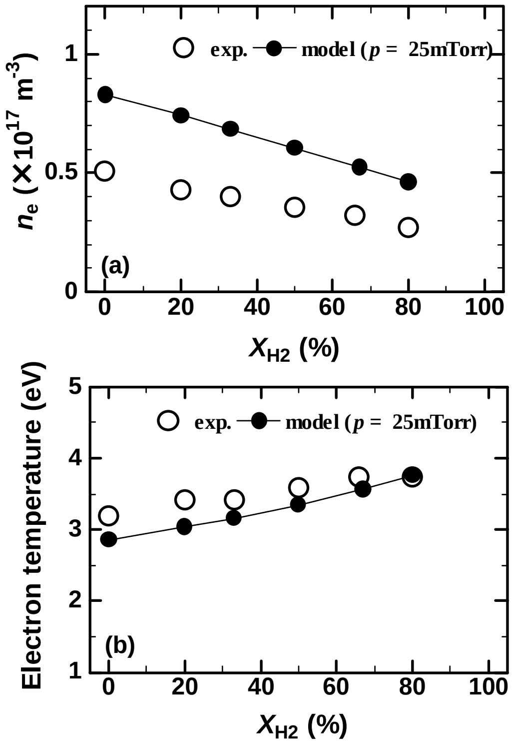

Fig. 1 Electron density (a) and its temperature at 25mTorr, where the power injected into plasma is 140W.

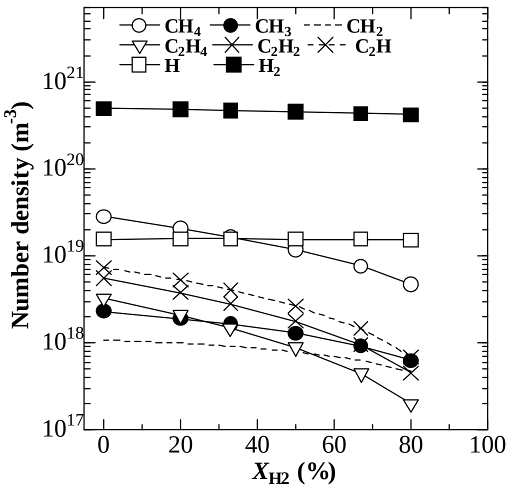

Fig. 2 Densities of main neutral species calculated at p = 25 mTorr, where the power injected into plasma is 140W.

The densities of neutral species calculated at 25 mTorr are shown in Fig. 2. Most of CH_4 molecules are easily dissociated, so that the dominant neutral species are always hydrogen molecules, CH_4 molecules and hydrogen atoms, followed by the hydrocarbons such as C_2H_m (m=1,2 and 4). The increase in net production rate is roughly proportional to the product $[CH_x]$ $[CH_y]$ of the densities of light hydrocarbons, resulting in the increase in densities of C_2H_m larger than the increase in densities of light hydrocarbons.

The positive ion densities calculated at 25mTorr is shown in Fig. 3. The dominant ions at X_{H2} lower than 50% are CH_5^+, C_2H^+, $C_2H_3^+$, $C_2H_5^+$, and $C_3H_5^+$, whereas the density of H_3^+ increases with the increase in X_{H2} and H_3^+ becomes dominant ion species at X_{H2} higher than 60-70%. Most of dominant hydrocarbon ions except C_2H^+ are mainly produced by the reactions between the hydrocarbon ions and CH_4 (or H_2), rather than direct ionization and dissociative ionization processes by the electron impact.

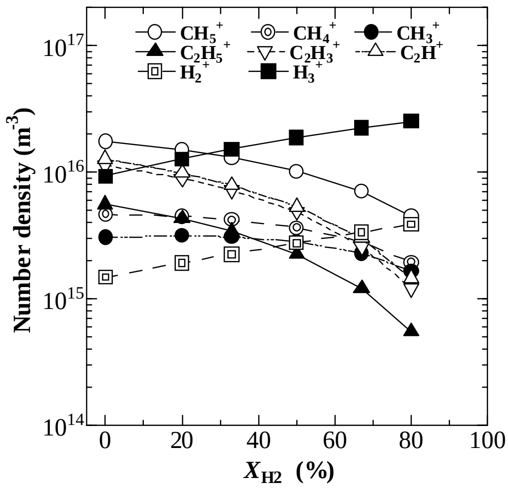

Fig. 3 Densities of main charged species calculated at 25 mTorr, where the power injected into plasma is 140W.

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