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Modeling of high frequency atmospheric pressure $Ar/H_2/SiH_4$ glow discharges

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ABSTRACT

In this paper, a one-dimensional self-consistent fluid model is applied to simulate high frequency atmospheric pressure glow discharges. The results show that the plasma density and current density depend strongly on the excitation frequency. When the excitation frequency is below 13.56 MHz, the discharge operates in the α mode, and when the excitation frequency is above 13.56 MHz, the discharge operates in a γ-like mode. The densities of species including SiH_3^+, SiH_3^-, SiH_3, SiH_2, H, Ar^+, Ar^* and electron are enhanced with the frequency increasing from 6.78 to 27.12 MHz. Similar discharge mode transition was observed experimentally in radio frequency atmospheric pressure He glow discharges. The effects of excitation frequency on plasma characteristics and densities of precursors for μc-Si:H film are further discussed. This study reveals that an appropriate excitation frequency is important for the growth of μc-Si:H film.

1. Introduction

Hydrogenated microcrystalline silicon (μc-Si:H) is a multi-phase material with a composition of crystalline granules embedded in an amorphous matrix. The μc-Si:H based thin film solar cell is one of the most promising candidates for the next-generation solar cell due to its high stability. Since they possess many advantages such as large homogeneous plasma volume, good stability, and highly nonequilibrium, high frequency atmospheric pressure glow discharges (APGDs) have drawn much attention recently. A key factor for increasing the deposition rate is to employ high density plasma sources [1–3]; therefore, growing μc-Si:H film with APGDs is a good potential way. The experimental and numerical studies have shown that there exist two different glow modes in APGDs, i.e., α and γ modes [4–7]. The γ mode is usually associated with local high density plasma and high discharge current density [8–11]. Compared to low-pressure discharges, homogeneous APGDs are much easier to evolve into a constricted γ mode when they are driven by the common high frequency range of 3 to 30 MHz [12]. Recently, it was reported that addition of H_2 to the Ar-diluted SiH_4 plasma can optimize the μc-Si:H structure by introducing homogeneity to its network, increasing the crystallinity and improving the electrical properties along with the elimination of adsorbed effects [13,14]. Since the species of SiH_3 and SiH_2 in $Ar/SiH_4/H_2$ plasma are main precursors for crystallization and deposition [15,16], the deposition rate depends strongly on the densities of SiH_3, SiH_2 and H [15]. In the $Ar/SiH_4/H_2$ plasma, the density distributions of individual species and characteristics of the discharge modes under different frequencies are not well understood yet. In this paper, spatial variations of the particle densities of SiH_3, SiH_2 and other reaction species versus the excitation frequency are investigated.

2. Simulation model

The discharge studied in this paper is generated between two parallel steel electrodes under a sinusoidal excitation voltage, as shown in Fig. 1. A one-dimensional (1-D) self-consistent fluid model is used to describe the spatio-temporal characteristics occurring in the direction perpendicular to the electrode surfaces. A total of nine species are included in this model, i.e, electrons *e*, argon ions Ar^+, electronic excited argon atoms (without distinguishing different quantum states) Ar^*, hydrogen atoms H, hydrogen molecule ions H_2^+, molecule ions SiH_3^+, molecule ions SiH_3^-, neutral radicals SiH_3 and SiH_2, and background argon atoms Ar, silane molecules SiH_4, and hydrogen molecules H_2. The reaction coefficient rates used in the simulation are summarized in Table 1. The governing equations are composed of 1D continuity equations, respectively, for electrons, ions, metastable atoms, and molecules, current conservation equation (for replacing the Poisson equation for an electric field) and the electron energy conservation equation . These equations are given as follows:

$$\frac{\partial n_{e,i,*}}{\partial t} + \frac{\partial \Gamma_{e,i,*}}{\partial x} = S_{e,i,*}, \tag{1}$$

$$\frac{\partial E}{\partial t} + e\Gamma_c = e\Gamma_0, \tag{2}$$

$$\Gamma_{e,i} = \mp n_{e,i}\mu_{e,i}E - D_{e,i}\frac{\partial n_{e,i}}{\partial x}, \tag{3}$$

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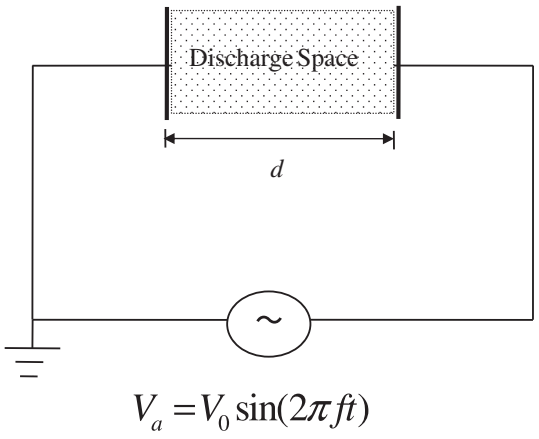

Fig. 1. Discharge configuration.

$$\Gamma_* = -D_* \frac{\partial n_*}{\partial x}, \tag{4}$$

where n and Γ are, respectively, the number density and the flux density of species; S represents sources and losses of particles due to the reactions, and the subscripts e, i, and * denote electrons, ions, and metastable species, respectively. The symbol E represents the electric field, Γ_c represents the conduction current density, and Γ_0 denotes the total current density. The symbols D and μ represent the diffusion coefficient and the mobility, respectively; their values were taken from Refs. [20–22]. The sinusoidal external voltage $V = V_0 \sin(2\pi ft)$ was applied to the right electrode. The electron energy conservation equation then can be expressed as

$$\frac{\partial}{\partial t}\left(\frac{3}{2}kT_e n_e\right) + \frac{\partial q_e}{\partial x} + e\Gamma_e E + p_{coll} = 0. \tag{5}$$

Here k is the Boltzmann constant, T_e is the electron temperature, and p_{coll} is the energy losses resulting from various reactions. The quantity q_e represents the electron thermal flow, and is given by

$$q_e = -K_e \frac{\partial (T_e)}{\partial x} + \frac{5}{2}kT_e\Gamma_e, \tag{6}$$

where

$$K_e = \frac{3}{2}kD_e n_e \tag{7}$$

Table 2
Reduced diffusion coefficients and motilities of species in $Ar/SiH_4/H_2$ mixed gas.

	Species	D_k(cm^2/s)	μ_k(cm^2/vs)	Ref.
1	e	1184.6	394.5	[22]
2	SiH_3^+	0.583	1.667	[20]
3	SiH_3^-	0.583	1.667	[20]
4	Ar^+	0.052	1.973	[22]
5	H_2^+	0.855	12.63	[21]
6	Ar^*	0.988		[22]
7	SiH_2	0.220		[20]
8	SiH_3	0.565		[20]
9	H	0.127		[21]

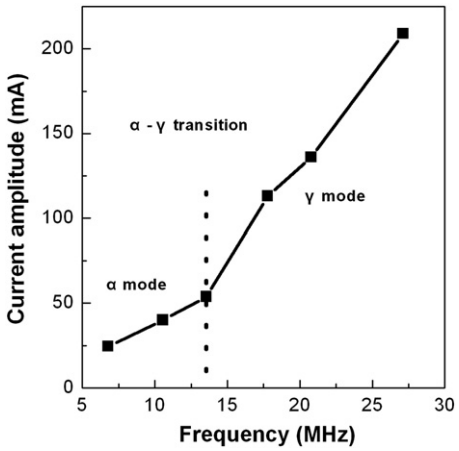

Fig. 2. The current amplitude versus excitation frequency.

The calculation starts from a spatially uniform ion and electron densities, where there exists a small, homogeneous electric field. This set of equations is numerically solved by the semi-implicit Scharfetter–Gummel scheme [23].

3. Results and discussion

The simulation parameters are chosen as follows: The discharge gap is fixed to 2.0 mm and the gas pressure is 760 Torr. The gas temperature is set to 300 K. The initial value of electric field $E = -10$ V/cm, and the total current density $\Gamma_0 = 0$. The values of D and μ are summarized in Table 2. The charged particles are absorbed by the electrodes and the secondary electron emission coefficient is assumed

Table 1
Collision reactions in the simulation.

	Process	Notation	Rate coefficient(cm^3s^{-1})	Reference
1	Direct ionization	$e + H_2 \rightarrow H_2^+ + 2e$	2.32×10^{-11}	[17]
2	Change transfer	$SiH_4 + H_2^+ \rightarrow SiH_3^+ + H + H_2$	6.23×10^{-10}	[18]
3	Direct ionization	$e + SiH_4 \rightarrow SiH_3^+ + H + 2e$	5.64×10^{-12}	[18]
4	Dissociative attachment	$e + SiH_4 \rightarrow SiH_3^- + H$	2.5×10^{-13}	[19]
5	Hydrogen abstraction	$H + SiH_4 \rightarrow SiH_3 + H_2$	2.0×10^{-12}	[19]
6	Recombination	$SiH_3^- + SiH_3^+ \rightarrow 2SiH_3$	2.0×10^{-7}	[19]
7	Reactions of radicals with radicals	$SiH_2 + H_2 \rightarrow SiH_4$	3.0×10^{-12}	[19]
8	Reactions of radicals with radicals	$SiH_3 + H \rightarrow SiH_2 + H_2$	3.3×10^{-10}	[19]
9	Recombination	$e + SiH_3^+ \rightarrow SiH_2 + H$	1.0×10^{-9}	[18]
10	Reactions of radicals with radicals	$SiH_3 + SiH_3 \rightarrow SiH_4 + SiH_2$	7.0×10^{-12}	[19]
11	Resonance ionization	$e + Ar^* \rightarrow Ar^+ + 2e$	$6.8\times10^{-9}Te^{0.67}$	[18]
12	Stepwise ionization	$Ar^* + Ar^* \rightarrow Ar^+ + Ar + e$	5.0×10^{-10}	[18]
13	Excitation	$e + Ar \rightarrow Ar^* + e$	$1.0\times10^{-11}Te^{0.75}e^{-11.6/Te}$	[19]
14	Direct ionization	$e + Ar \rightarrow Ar^+ + 2e$	$4.0\times10^{-12}Te^{0.5}e^{-15.8/Te}$	[19]
15	De-excitation	$Ar^* + SiH_4 \rightarrow SiH_3 + H + Ar$	1.4×10^{-10}	[19]
16	Charge transfer	$Ar^+ + SiH_4 \rightarrow SiH_3^+ + H + Ar$	2.4×10^{-12}	[19]