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SIMULATION OF THE ELECTRICAL PROPERTIES OF SiH_4/H_2 RF DISCHARGES

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Abstract

The results of a 2D fluid simulator were validated against the experimentally measured electrical properties (power dissipation and discharge current) of SiH_4/H_2 RF discharges used for the deposition of microcrystalline or amorphous silicon thin films. Using the typical values that can be found in the literature for the electron – SiH_4 and H_2 collision cross-sections, a significant underestimation of the calculated power dissipated in the discharge and an overestimation of the current flow was observed. A study of the main parameters that can affect the model results has shown that this deviation is mainly due to the prediction of the H_2 ionization rate. An improved calculation of the power dissipated in the discharge was then possible leading to a significant improvement on the prediction of the deposition rate. This result underlines the importance of H_2 chemistry in SiH_4/H_2 discharges but also indicates that the correct estimation of the discharge electrical properties is a necessary first step in the development of a code capable to simulate the deposition process of silicon films from SiH_4/H_2 discharges.

Keywords: fluid model, PECVD, microcrystalline silicon, amorphous silicon

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1. Introduction

One of the major impediments in understanding and optimizing plasma processes lies in the discrepancies that can be found between the results of different groups for the same topic. One of the main origins of these deviations for the experimental studies is the lack of the electrical control of the discharges[1,2] leading to undefined plasma conditions while for the simulations, there is either no comparison between model and experiments or a bypass of the electrical conditions [3,4].

The present work focuses on the confrontation of this problem through the development of a plasma simulator that is fine-tuned according to experimental measurements of electron, displacement and ion current, discharge impedance and power dissipation. The present results concern SiH_4/H_2 discharges typically used for the growth of either microcrystalline or amorphous silicon thin films[5]. The specific gas mixture presents the advantage that both gases are very well studied and one can find experimental measurements for most of the reaction cross-sections or rate constants required. An accurate experimental method was used for the determination of the discharge current, impedance and power dissipation over a wide range of conditions along with silicon film deposition rate measurements. The SiH_4/H_2 simulator used uses the fluid approximation and includes detailed description of the gas phase chemistry as well as gas-surface interactions[6].

Nevertheless, the use of cross-sections and rate constants that are typically found in the literature[7,8,9] results to strong deviations between calculations and experimental measurements for all plasma properties. A study of the effect of the main parameters that can affect the electrical properties was then carried out to determine the specific weight of these inputs on the model results.

2. Experimental

The measurements were carried out in a capacitively coupled Ultra High Vacuum parallel plate reactor having a base vacuum of 10^{-7} Pa. The rf electrode is of 12 cm in diameter, while the grounded electrode has a diameter of 9.5 cm and consist also the substrate holder. The experiments were performed at total working pressure of 67.7 and 133 Pa and a constant electrode gap (1.5 cm). In these series of experiments, the flow rates were maintained at 100 sccm while silane concentration was 2% and 6 % for the low and high pressure conditions, respectively. Films of constant thickness (~1.6 μm) were deposited on Corning glass 7059 at various power levels and at a substrate temperature of 250 ºC.

The amount of RF power actually fed into the discharge chamber was determined using an accurate method employing the Fourier transform determination of power and impedance phase, from current and voltage measurements[2], while the deposition rate was measured in situ using Laser Reflectance Interferometry[10].

3. Model

The plasma fluid simulator uses the particle, momentum and energy balances obtained from moments of the Boltzmann transport equation, coupled with Poisson's equations for a self consistent calculation of the electric field. The particle balance for electrons, ions and neutrals is described by the continuity equation:

$$\frac{\partial n_j}{\partial t} + \nabla \cdot \vec{\Gamma}_j = S \qquad (1)$$

where, n_j is the density, $\vec{\Gamma}_j$ is the flux of particle j and S is the source of particles consumed or produced in chemical reactions. In the present version, the model includes 24 species and 80 gas phase reactions (neutrals-neutrals, ion-neutrals,

electron-neutrals, ion-ion). The rate constants of these reactions were taken either from the solution of Boltzman equation for the specific mixture in the case of electron-molecule reactions or from[7,11] in the case of radical-molecule and ion-molecule reactions.

The drift diffusion approximation replaces the momentum balance for the electrons:

$$\vec{\Gamma}_j = -D_j \nabla n_j + \mu_j n_j \nabla V \qquad (2)$$

where, μ_j is electron mobility, D_j is the diffusion coefficient and V is the electrostatic potential. Mobility (μ_j) is calculated as a function of electron – molecule momentum transfer collision frequency $\mu_j = \frac{e}{m_e \sqrt{\nu_m^2 + \omega^2}}$, which in turn is calculated as a function of the electron temperature ν_m(Te) and momentum transfer collision cross sections for the specific mixture. The diffusion coefficient is then calculated from Einstein relation $D_j = \mu_j \frac{k \cdot T_e}{e}$.

The energy balance is solved only for electrons, assuming that ions have equal energy with neutrals. The electron temperature T_e is derived from the electron energy balance:

$$\frac{3}{2}\frac{\partial}{\partial t}(n_e T_e) + \nabla \cdot \left(\frac{5}{2} T_e \vec{\Gamma_e} - \frac{5}{2} n_e D_e \cdot \nabla T_e \right) = P - n_e \sum_i N_i K_i \qquad (3)$$

where, the energy transfer is due to convective flux and thermo-diffusion (*e* and *i*, subscripts are used for electron, ion, neutral particle and normal direction respectively), P is the pressure gradient and the last term on the right side is the rate of electron production/consumption by electron impact collisions.

For a self consistent calculation of the electric field/potential, Poisson's equation is solved simultaneously with the fluid equations:

$$\nabla^2 V = -\frac{e}{\varepsilon_0}\left(\sum_{i=ions} q_i n_i - n_e\right); \; E = -\nabla V \tag{4}$$

where ε_0 is the permittivity of free space, q_i is the sign of the charge of ion i and E is the electric field.

The system of equations (1)-(4) is complemented by a set of boundary conditions concerning the species densities and the electric potential. The electric potential at the driven and grounded electrodes is defined as:

$$V_{RF} = V_{dc} + V_0 \sin 2\pi F \;, \quad V_{ground} = 0 \,. \tag{5}$$

The electron flux normal to the electrodes and the reactor walls (assuming no reflection or secondary emission) is given by:

$$\vec{\Gamma}_{e,n} = \frac{1}{4} n_e v_{e,th} ; \qquad v_{e,th} = \left[\frac{8 k_B T_e}{\pi m_e}\right]^{\frac{1}{2}} \tag{6}$$

where, $v_{e,th}$ is the electron thermal velocity, k_B is the Boltzmann constant and m_e is electron mass.

The velocity of ions is determined by the local value of the electric field and their flux to the walls is supposed to be pure drift when the velocity is directed to the powered and grounded electrode

$$\vec{\Gamma}_{i,n} = q_i \mu_i E_n n_i \tag{7}$$

and zero towards the reactor walls. At dielectric surfaces (glass), the net surface charge σ_s is obtained from the particle fluxes:

$$\frac{\partial \sigma_s}{\partial t} = e\left(\sum_i q_i \vec{\Gamma}_{i,n} - \vec{\Gamma}_{e,n}\right), \quad \varepsilon_o E_n = \sigma_s \tag{8}$$

The fluxes of gas-phase species at the surface are modified by surface reactions and their flux balance at the surface yielding:

$$n \cdot \vec{\Gamma}_p = m_p S_p \qquad (9)$$

where, n is a unit vector normal to the surface and S_p is the surface production (consumption rate per unit area). The model includes 25 surface reactions (recombination, abstraction, chemisorption, physisorption, charge loss) taken from[11, 12] while stable molecules are assumed as not interacting with the surfaces (zero flux) at the specific substrate temperature.

3. Results and Discussion

The model and experiments were applied in a parallel plate, capacitively coupled plasma chamber, operating at frequency of 27.12 MHz, low pressure, dust free conditions. The effect of the discharge power on the deposition process at two different gas pressures (67.7 and 133 Pa) was investigated and the model results were compared to experimental measurements performed in identical conditions.

The variation of the total discharge current and the effective power density as calculated by the model as well as the ones measured experimentally for both pressures is presented in fig.1. The model underestimates the power dissipated in the discharge and at the same time overestimates the total current at the driven electrode. It has to be noted that the calculations presented in fig.1 were performed using the most common cross-sections found in literature concerning the collisions of electrons with either SiH_4 or H_2. The discrepancies between model and experimental results are smaller in the lower pressure and the lower power dissipation case. In addition, these differences between the measured and calculated values of the power result to an overestimation of the predicted values of the silicon thin film deposition rate in all conditions. This is illustrated in fig.2, where the model predictions of the film growth rate are presented as a function of the applied voltage together with experimental

measurements. In some cases the model, especially in the low pressure conditions, anticipates values about two times higher than the experimental ones.

An analysis of the contradictory behavior of the discharge current and power (fig.1) together with the overestimation of the deposition rate (fig.2) has indicated that electron-molecule collisions and more precisely dissociation and/or ionization processes in these SiH_4/H_2 discharges are not correctly predicted. The most reasonable explanation is that the calculated electron density is overestimated leading to a subsequent underestimation of the electric field intensity. On the other hand, the higher electron density will lead to the prediction of a higher total current as the current flow scales linearly with electron density. Moreover, the lower electric field in the discharge will seriously affect the power consumption as it scales with the square of the electric field intensity. The lower electric field will also favor dissociation relative to ionization of both SiH_4 and H_2 leading thus to higher values of the deposition rate.

In order to check if the above mentioned rationale is correct a study of the main parameters that may affect both electron density and electric field was carried out. Namely, the mobility of electrons in the SiH_4/H_2 mixtures and the ionization rate of SiH_4 and H_2 were varied by altering the cross sections for momentum transfer and ionization, respectively. The change on these values was always limited to ±10 % of the typical values found in the literature, which is otherwise the typical error in the calculation or the measurement of the cross sections. The results of this parametric study can be summarized as follows: (a) the changes of the mobility and the SiH_4 ionization rate had a negligible effect on the results of either the power dissipation or the discharge current while (b) small variations of the H_2 ionization rate produce enormous changes on either the power dissipation or the current flow.

Figure 4 presents model and experimental results of both power dissipation and current at the same conditions as in fig.1. However, in this case the cross section for H_2 ionization has been increased by 10%. An excellent agreement can be observed between model and experiment for the total current flow, while the difference in power dissipation is for both pressures below 15%. In addition, the variation of the H_2 ionization cross section affects indirectly the calculated values of the deposition rate mainly due to the decrease of the electron density and the subsequent drop of the rate of SiH_4 dissociation. Thus, figure 4 presents the deposition rate calculated with the corrected H_2 ionization cross section together with the same experimental values as a function of the applied voltage. In this case, a much better agreement is observed compared to the results presented in fig. 2.

The correct prediction of electrical properties and the much better estimation of the deposition rate may then allow for a discussion of the role of the most abundant radicals in the discharge which also have a major contribution to the film growth. Ions have been excluded from this discussion as in the present model is deliberately assumed that they do not directly contribute to the film growth. Thus, fig. 5 shows the estimated densities of the main species in SiH_4/H_2 discharges (H, SiH_3, SiH_2, Si_2H_5) as a function of the applied voltage. All the species densities increase with the rise of power consumption, independent of pressure, although they have different behaviors at the different pressures. Silyl radical (SiH_3) has the highest particle density in all the simulated conditions which is about one order of magnitude higher than that of silylene (SiH_2). The highly reactive SiH_2 radical is not favored by the pressure and silane concentration increase, which increase the probability of its consumption in

secondary reactions with mono and poly-silane species. However, taking into account the much higher surface reactivity of SiH_2 compared to SiH_3, it is clear that both species have a significant role in the deposition mechanism. In addition, Si_2H_5 radicals do not show any noticeable variation at these two pressures, having a density of about two orders of magnitude lower than all other species of interest.

Finally, even though H_2 is the main component of the discharge mixture (98% at 67.7 Pa, 94% at 133 Pa respectively), hydrogen atoms density is lower than SiH_3 density, as a result of the much lower cross section for H_2 compared to SiH_4 dissociation. However, as pressure decreases and higher voltage is applied, hydrogen atoms tend to become the most abundant species. This mainly comes from the fact that each H_2 dissociation reaction produces two hydrogen atoms enhancing thus, the fast increase of H atoms density with the H_2 dissociation rate.

4. Conclusions

A 2D self-consistent fluid model was applied for the investigation of the effect of rf power and total SiH_4/H_2 gas pressure and the results of the simulation were compared with experimental electrical and deposition rate measurements.

Using the most common values that can be found in literature concerning electron-molecule collision cross sections and reactions rate constants, the model largely deviates from the experimental measurements. Namely, in all conditions predicts lower power dissipation and significantly higher current flows and silicon deposition rates. The parametric analysis performed has shown that small variations of the hydrogen ionization rate have a tremendous effect on the model results. A reduction of the ionization rate by less than 10% has led to a very good agreement between calculated and measured values of the electrical properties along with a significant

improvement of the prediction of the deposition rate. This result highlights the importance of H_2 chemistry in SiH_4/H_2 discharges and indicates also that, in the development of a reliable model of plasma deposition processes, the correct prediction of the physical processes in the discharge is a necessary first step.

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FIGURE CAPTIONS

Figure 1: Total power and total current of the discharge measured and calculated at the driven electrode as a function of the applied voltage for 67.7 Pa 2% and 133 Pa 6% SiH_4 in H_2 discharges

Figure 2: Displacement, electron, ion and total current (left axis) and applied voltage (right axis) during an rf cycle, for conditions 2 % SiH_4 in H_2, 67.7 Pa and applied voltage $V_{rf}=-68 + 150\sin\omega t$

Figure 3: Deposition rate of silicon films measured and calculated as a function of the applied voltage for 67.7 Pa 2% and 133 Pa 6% SiH_4 in H_2 discharges

Figure 4: (a) Spatial distribution of time-averaged electric field intensity and (b) spatial distribution of electron density for three different values of H_2 ionization cross-section

Figure 5: Total power and total current of the discharge measured and calculated at conditions of fig. 1 and after parametric analysis

Figure 6: Deposition rate of silicon films measured and calculated at conditions of fig. 3 and after parametric analysis

Figure 7: Radical average number densities vs. applied voltage in 67.7 Pa 2% and 133 Pa 6% SiH_4 in H_2 discharges

FIGURES

Figure 1

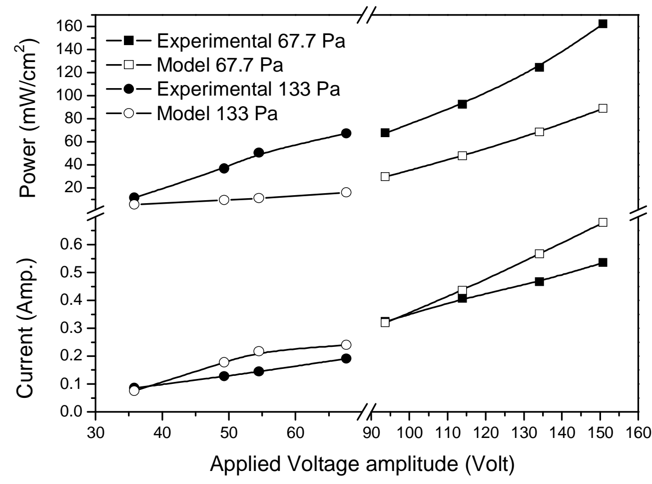

Figure 2

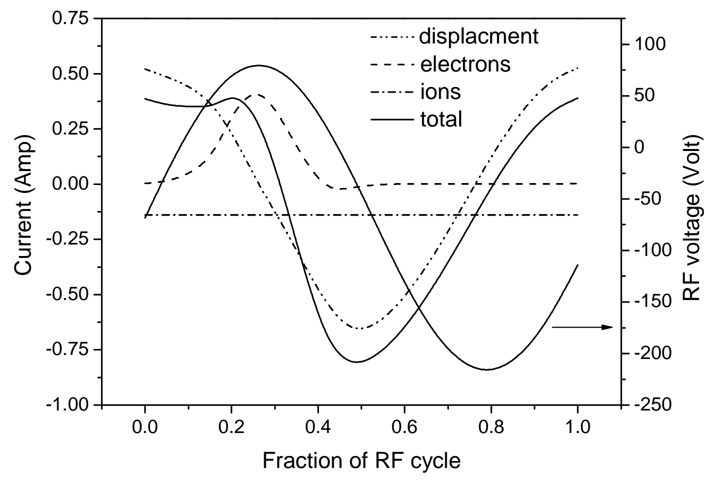

Figure 3

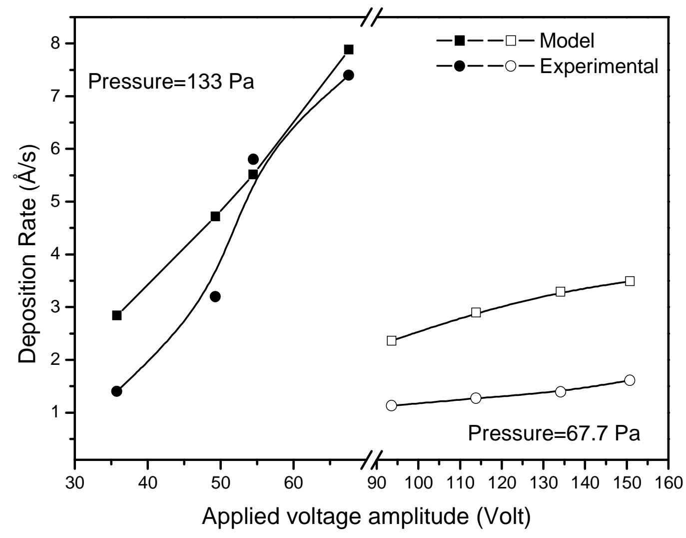

Figure 4

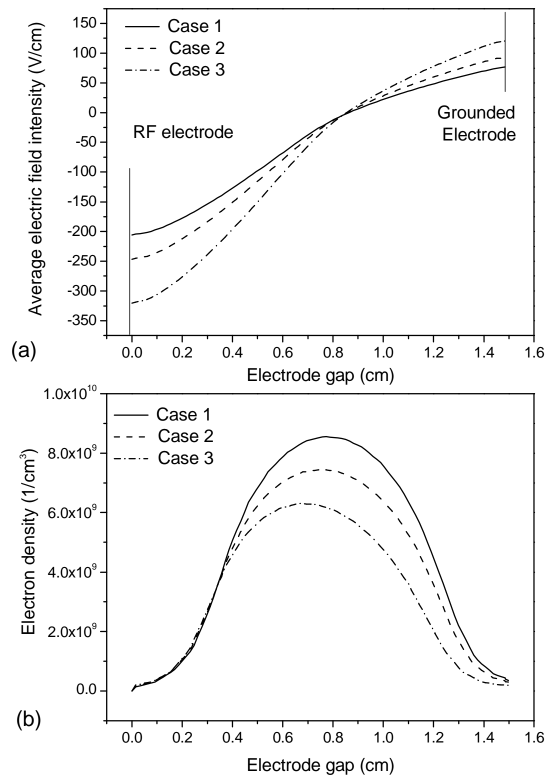

Figure 5

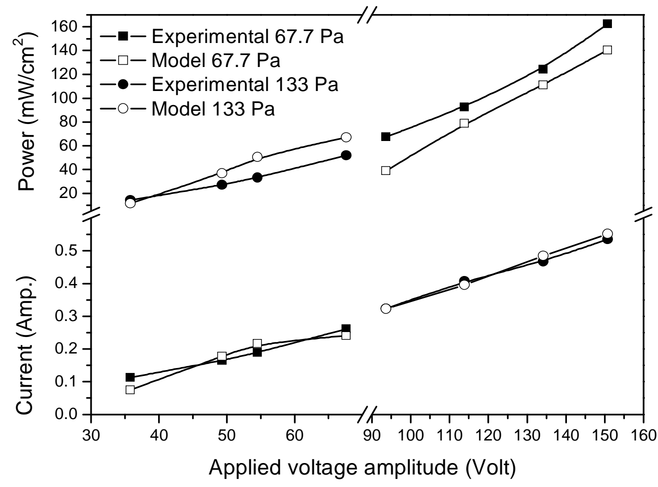

Figure 6

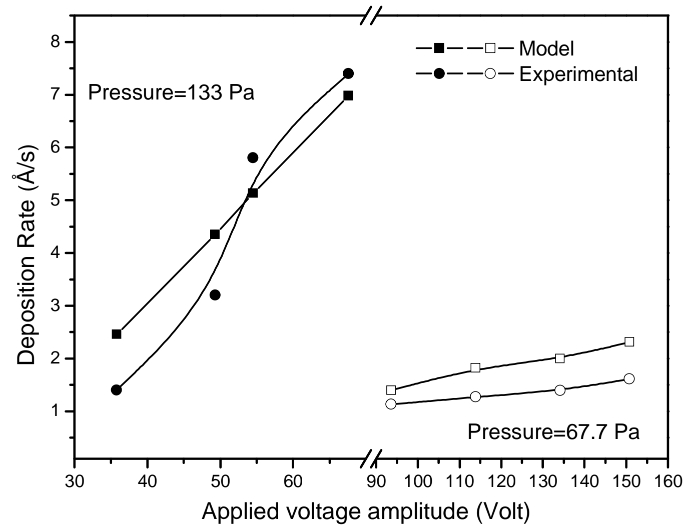

Figure 7