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ORIGINAL RESEARCH PAPER

Effects of Plasma Discharge Parameters on the Nano-Particles Formation in the PECVD Reactor

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ABSTRACT: In this paper, the effects of plasma discharge parameters on the nano-particles formation process in a plasma enhanced chemical vapor deposition (PECVD) reactor using a model based on equations of ionization kinetics for different active species are studied. A radio frequency applied electric field causes ionization inside the reactor and consequently different reaction schemically active species are formed. The reactions leading to production of nano-particles include negative and positive ion, neutrals and radicals. The dependency of the background gas temperature, frequency and amplitude of applied electric field as the main plasma discharge parameters on the avalanche time constant, nano-particles formation and their growth rate is verified. Silane and hydrogen gases are considered as background species. It was observed that the growth rate at higher frequencies and lower temperatures is higher. It was seen that increase in the applied voltage peak amplitude affects charged and radical particles fairly similar to the applied voltage frequency.

KEYWORDS: *PECVD Reactor; Equations of Kinetics; Silicon Amorphous Thin Film*

INTRODUCTION

Nowadays thin film deposition process has become important, especially for applications that are sensitive toward ion bombardment. One approach is to inject the molecular gas (e.g., SiH_4) into the after section and to place the substrate at the end of the after section [1].

One of the main technologies employed for industrial production of thin films is plasma enhanced chemical vapor deposition (PECVD) [2-3]. In this technique, Electrons will impact with background gases and lead to ionization reactions which are the fundamental processes in this scheme. Various reactions produce positive and negative ions. As the reactions take place through the time; new species will take part in the new reactions and radicals and neutrals being produced after a while. Various particles are deposited on the substrate to form amorphous thin films. Another important factor is the ion bombardment of the substrate surface. Film properties are modified by the ion bombardment. Ion-neutral interactions near the cathode can significantly influence this bombardment process.

For optimizing physical and chemical parameters in the discharge process inside PECVD, reactive chamber geometry and operating regimes of the installation, modeling and simulation of this industrially important technology is essential [4-9]. Moreover, in order to reduce the costs of film production and to provide a better growth rate and film quality in terms of layer composition and homogeneity, an applicable model could be helpful.

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Different types of models have been used to describe and study glow discharges inside PECVD reactor. These self-consistent models are mainly including fluid or kinetic approaches. In these models, the number of physical processes could influence the physical relevance of the modeling results are considered [10-18]. Most of these models have been used for plasma discharges with a relatively simple chemistry and no plasma–wall interaction of the neutrals. Other models include the silane/hydrogen chemistry and the surface interaction, but these models are not self-consistent with respect to the rates of the ionization, dissociation, excitation, and attachment [19]. Models developed for plug-flow silane/hydrogen discharges [13, 17] are self-consistent with respect to the silane/hydrogen chemistry, the electron impact collisions, the plasma–wall interaction, and the transport phenomena.

Krzhizhanovskaya et al. used a model based on the numerical solution of the full Navier-Stokes equations for transient laminar flows of viscous compressible multi-component mixtures of chemically reacting gases in a PECVD reactor [10-11].

Nienhuis et al. used a fluid model for simulation of capacitively coupled plasma discharge [2, 12]. They solved the electron and ion continuity equations consistently with the Poisson equation for the electric field distribution inside PECVD reactor. Moreover, an interesting approach is discussed by Tichibana [20] for RF plasma. They stressed on the importance of controlling the energy and density of the electrons in the first place.

Nomenclature

T	Temperature (K)	Γ_i	Flux of an ith Molecule Towards the Substrate (1/m^{3}s)
V	Average Volume (m^3)	θ	Avalanche Time Constant (s)
K_i	Reaction Rate Constants For i Species (m^{-3}s^{-1})	N_{eo}	Number of Seed Electrons (m^{-3})
V_{rf}	Power Source Frequency (MHz)	V_i, V_A, V_d	Ionization, Attachment and Detachment Frequencies, [MHz]
q_i	Number of Silicon Atoms in Molecule i (1/m^3)	P	Turbulent kinetic energy
		$d(L_Z)$	Boltzmann number (J/K)

In this work, to study the effects of plasma discharge parameters on the charged particles, their avalanche constants, radical formation and nano-particle growth rate process in a PECVD reactor, a model based on equations of ionization kinetics for different active species is considered. Silane and Hydrogen is considered as the background gas. The most essential reactions in the reactor volume are taken into account. The effects of temperature, frequency and potential amplitude on the particle's production inside PECVD reactor are investigated. These parameters and their effects on particle density variations help us to apply the most benefit condition in this process.

SIMULATION MODEL

As shown in Figure 1, the reactor includes two parallel electrodes with substrate on them.

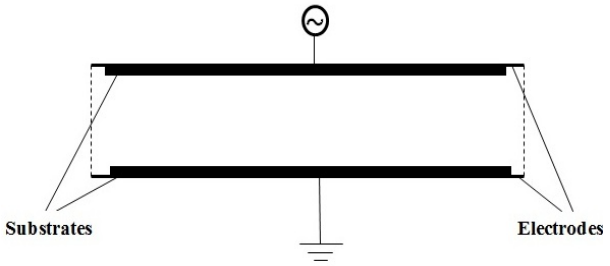

Fig. 1. Schematic representation of the discharge volume in the PECVD reactor

The first electrode is grounded while the upper electrode is connected to a radio frequency (RF) power. Between the electrodes, a glow discharge is produced; during the glow discharge, different reactions will happen and different particles are produced in the reactor volume [12]. Radicals such as SiH_2 and SiH_3 deposited on the substrate, play the most important role in deposition process. However, Si_2H_5 and heavier radicals are rarely produced and can be neglected in this model [13-14]. For the behavior of the electrons in a RF discharge, energy relaxation lengths are important because they contain the information of the energy gain process from the oscillating electric field and energy loss process due to inelastic collisions inside PECVD reactor. Spatial energy relaxation depends on the ratio between the mean free path for energy loss and the relevant length scales in the discharges. The model describes discharge process between two parallel electrodes in the reactor volume. The general balancing equations for each species, i. e. electron, neutrals, ions and radicals can be written as follows:

$$\frac{\partial n_j}{\partial t} - D_a \nabla^2 n_j = S_j \tag{1}$$

The upper electrode is connected to frequency power source in order to change the electric field vector very quickly. The frequency potential applied as follow:

$$V_{(z=L)} = V_{rf} \sin(2\pi t \nu_{rf}) \tag{2}$$

where V_{rf} is the amplitude and set equal to 150-300 Volts and ν_{rf} is power source frequency is set to 13.6-50 MHz. So, a periodic electric field is applied on the anode electrode. Although the charged particles which are produced in the reactor volume can be affected on electric field but here is neglected [2]. While the production of silyl and silylene is based on dissociation of silane, the formation of silylene also occurs along two possible routes. In the first route, hydrogen is produced and in the second route, the molecular hydrogen is produced. Distinction between these two routes is important since atomic hydrogen participates in a secondary hydrogen extraction reaction with silane to form silyl. The equation 1 looks like an ordinary diffusion equation (with ambipolar coefficient) for a problem with bulk sources of charge. For simplification, we assume that the field is spatially homogenous, and hence ionization and attachment coefficients are independent coordinates. Some of the important electron reactions and ionization processes inside in the PECVD reactor are presented in the table 1. Averaging over the volume, we obtain the following equations for mean density in the PECVD discharge volume:

$$\frac{dn_j}{dt} = S_j \tag{3}$$

Reactions are the most important sources which can be written as:

$$A + B \rightarrow C + D$$
$$S_{rec,C,D} = -S_{rec,A,B} = n_A n_B k_{rec} \tag{4}$$

K_{rec} is reaction rate [12].

In this work, essential reactions are taken into account and the creation of nineteen species during discharge process is considered. Radicals are vital particles in the growing rate of amorphous silicon thin film in the PECVD reactor. The deposition of these Nano-sized radicals on the substrate causes the growth of thin film. Table 1 shows the chemical reactions and reactions rate which is used in the model [14, 21,22]. Then, based on these chemical reactions, the equations of kinetics for mean densities of some of important species are as follows:

Table1
Chemical reactions and reaction rates occurred in the reactor.

Chemical reaction	Reaction rate(cm^{-3} S^{-1})
$SiH_4 + e \rightarrow SiH_2^+ + 2H + 2e$	$K_1=1.5 \times 10^{-10}$
$Si_2H_6 + e \rightarrow Si_2H_4^+ + 2H + 2e$	$K_2=3.7 \times 10^{-11}$
$SiH_4 + e \rightarrow SiH_3^- + H$	$K_3=3 \times 10^{-11}$
$H_2 + e \rightarrow H + H + e$	$K_5=4.49 \times 10^{-12}$
$H + SiH_4 \rightarrow SiH_3 + H_2$	$K_4=2.8 \times 10^{-11}$ exp(-1250/T_{gas})
$H + Si_2H_6 \rightarrow Si_2H_5 + H_2$	$K_9=1.6 \times 10^{-10}$ exp(-1250/T_{gas})
$H + Si_nH_{2n+2} \rightarrow Si_nH_{2n+1} + H_2$	$K_{10}=2.4 \times 10^{-10}$ exp(-1250/T_{gas})
$SiH_4 + SiH_2 \rightarrow Si_2H_6$	$K_8=2 \times 10^{-10}$[1- (1+ 0.0032 p_0)$^{-1}$]
$Si_nH_{2n+2} + SiH_2 \rightarrow Si_{n+1}H_{2n+4}$	$K_{11}=4.2 \times 10^{-10}$[1- (1+ 0.0033 p_0)$^{-1}$]
$SiH_3^- + SiH_2^+ \rightarrow SiH_3 + SiH_2$	$K_6=1.2 \times 10^{-7}$
$SiH_3^- + Si_2H_4^+ \rightarrow SiH_3 + 2SiH_2$	$K_7=1 \times 10^{-7}$

$$\frac{dn_e}{dt} = n_e n_{SiH_4} k_1 + n_e n_{Si_2H_6} k_2 - n_e n_{SiH_4} k_3 \quad (5)$$

$$\frac{dn_{SiH_2}}{dt} = n_{SiH_2^+} n_{SiH_3^-} k_1 + n_{SiH_3^-} n_{Si_2H_4^+} k_7 - n_{SiH_2} n_{SiH_4} k_6 \quad (6)$$

$$\frac{dn_{SiH_3}}{dt} = n_{SiH_2^+} n_{SiH_3^-} k_6 + n_{SiH_3^-} n_{Si_2H_4^+} k_7 + n_H n_{SiH_4} k_4 \quad (7)$$

$$\frac{dn_{SiH_2^+}}{dt} = n_e n_{SiH_4} k_1 - n_{SiH_3^-} n_{SiH_2^+} k_6 \quad (8)$$

$$\frac{dn_{Si_2H_5}}{dt} = n_H n_{Si_2H_6} k_9 \quad (9)$$

$$\frac{dn_{Si_2H_4^+}}{dt} = n_e n_{Si_2H_6} k_2 - n_{SiH_3^-} n_{Si_2H_4^+} k_7 \quad (10)$$

$$\frac{dn_{SiH_3^-}}{dt} = n_e n_{SiH_4} k_3 - n_{SiH_3^-} n_{Si_2H_4^+} k_7 - n_{SiH_3^-} n_{SiH_2^+} k_6 \quad (11)$$

$$\frac{dn_H}{dt} = n_e n_{SiH_4} k_1 + n_e n_{Si_2H_6} k_2 + n_e n_{SiH_4} k_3 - n_H n_{SiH_4} k_4 - n_H n_{Si_2H_6} k_9 + n_e n_{H_2} k_5 - n_H n_{Si_4H_{10}} k_{10} - n_H n_{Si_3H_8} k_{10} - n_H n_{Si_5H_{12}} k_{10} - n_H n_{Si_6H_{14}} k_{10} - n_H n_{Si_7H_{16}} k_{10} \quad (12)$$

$$\frac{dn_{Si_2H_6}}{dt} = n_{SiH_2} n_{SiH_4} k_8 - n_e n_{Si_2H_6} k_2 - n_H n_{Si_2H_6} k_9 - n_{SiH_2} n_{Si_2H_6} k_{11} \quad (13)$$

$$\frac{dn_{Si_4H_9}}{dt} = n_H n_{Si_4H_{10}} k_{10} \quad (14)$$

$$\frac{dn_{Si_3H_7}}{dt} = n_H n_{Si_3H_8} k_{10} \quad (15)$$

$$\frac{dn_{Si_5H_{11}}}{dt} = n_H n_{Si_5H_{12}} k_{10} \quad (16)$$

$$\frac{dn_{Si_6H_{13}}}{dt} = n_H n_{Si_6H_{14}} k_{10} \quad (17)$$

$$\frac{dn_{Si_7H_{15}}}{dt} = n_H n_{Si_7H_{16}} k_{10} \quad (18)$$

$$\frac{dn_{Si_3H_8}}{dt} = n_{SiH_2} n_{Si_2H_6} k_{11} - n_H n_{Si_3H_8} k_{10} - n_{SiH_2} n_{Si_3H_8} k_{11} \quad (19)$$

$$\frac{dn_{Si_4H_{10}}}{dt} = n_{SiH_2} n_{Si_3H_8} k_{11} - n_H n_{Si_4H_{10}} k_{10} - n_{SiH_2} n_{Si_4H_{10}} k_{11} \quad (20)$$

$$\frac{dn_{Si_5H_{12}}}{dt} = n_{SiH_2} n_{Si_4H_{10}} k_{11} - n_H n_{Si_5H_{12}} k_{10} - n_{SiH_2} n_{Si_5H_{12}} k_{11} \quad (21)$$

$$\frac{dn_{Si_6H_{14}}}{dt} = n_{SiH_2} n_{Si_5H_{12}} k_{11} - n_H n_{Si_6H_{14}} k_{10} - n_{SiH_2} n_{Si_6H_{14}} k_{11} \quad (22)$$

$$\frac{dn_{Si_7H_{16}}}{dt} = n_{SiH_2} n_{Si_6H_{14}} k_{11} - n_H n_{Si_7H_{16}} k_{10} \quad (23)$$

The growth rate depends on the flux of species including silicon atoms. So, the thin film growth rate R could be obtained as follows:

$$R = \sum_i R_i R_i = V q_i \Gamma_i \quad (24)$$

where V is the average volume of the atom in formation of thin film (for silicon, V=22 A^3), q_i is equal to the number of silicon atoms in molecule i and Γ_i is the flux of an i^{th} molecule toward the substrate [2, 12, 21, 22].

SIMULATION RESULTS

Despite of too much of efforts on the designing and control of PECVD reactors, there still exist the needs for systematic studies to optimize the operation of these systems via investigation of the effects of plasma discharge parameters on the different features of nano-particles formation inside the reactor. The main part of thin films on the substrate is made of SiH_2 and SiH_3. Generally, in the PECVD reactor, the production rate of the greater radicals is rare and consequently they are less important and can be ignored.

In following, the effects of the background gas temperature, frequency, and amplitude and electrodes separation as the main discharge parameters on the avalanche constant time as well as different features of PECVD reactor such as density of species, growth rate and etc. are studied.

Unless otherwise mentioned, all simulations reported in this work are performed for operating conditions specified in table 2.

Table 2
Simulation Parameters

property	symbol	value
Temperature	T	320 K
frequency	υ_{rf}	13.6 MHz
Potential amplitude	V	150 V
Electrodes Separation	L_z	0.27m

Frequency

The simulation results corresponding to the nano-particles formation process in a PECVD reactor are presented here. By using the above explained model which is based on the equations of kinetics for different active species in the PECVD reactor, we have studied the effects of the applied voltage frequency on the nano-particles formation and growth rate. Frequency is an essential parameter affecting formation of charged particles, nano-particles and the reaction occurrence.

The above simulations were performed for the simulation parameters presented in Table 2, with the applied voltage frequency varying from 13.56 to 90MHz.

PECVD reactor is often operated at a very low pressure i.e., from 1 to hundreds of mTorr and at a high frequency i.e., from 10 to 100 MHz.

As shown in Figure 2, at smaller applied RF-frequency, the total number density of the main species in the PECVD reactor (i. e. electron, SiH_2^+ and H) is lower.

In the other hand, at higher frequencies, sooner ionization happens in the reactor volume. Changing the electric field very quickly, charge particles move more quickly and encounter to background gases faster and as a result chemical reactions happen quickly. Also, 90MHz is high frequency but greater silicon radicals still are not produced. Nienhuis et al. [2] have shown that there is a direct relation between partial pressure and frequency of applied voltage.

Increase in RF-frequency makes partial pressure higher in the reactor volume. As a result of higher RF-frequency, the speed of chemical reactions and particle productions increases.

The background gas temperature can influence the ionization processes inside the PECVD chamber dramatically.

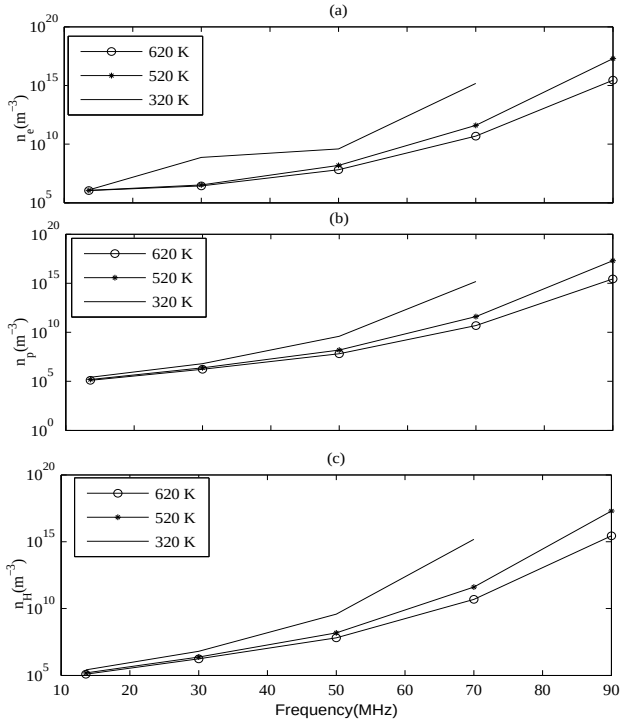

Fig. 2. Density dependence on the frequency (a) electrons, (b) $SiH2^+$, (c) H at different gas temperatures (T_{gas})

As shown in the Figure 2, at lower gas temperatures, the reaction rates are higher, so the ionization rate and production rates of charged particles and radicals are higher.

Figure 3 represents the variations in the density of electron, SiH_2^+, SiH_3 and Si_2H_5 as the ionization process goes on in time.

As it is expected, the density of different species inside PECVD reactor versus frequency increases. Also, the charged particles densities grow up faster than the neutral radicals.

As seen, even in the high frequencies, the heavy particles have no production, which is in agreement with the obtained results by other researchers [2, 12].

The density dependence of different charged particles versus time almost shows an exponential form which is a typical of an avalanche process:

$$N_e = N_{e0}\exp[(\nu_i - \nu_a - \nu_d)] = N_{e0}\exp(t/\Theta) \qquad (25)$$

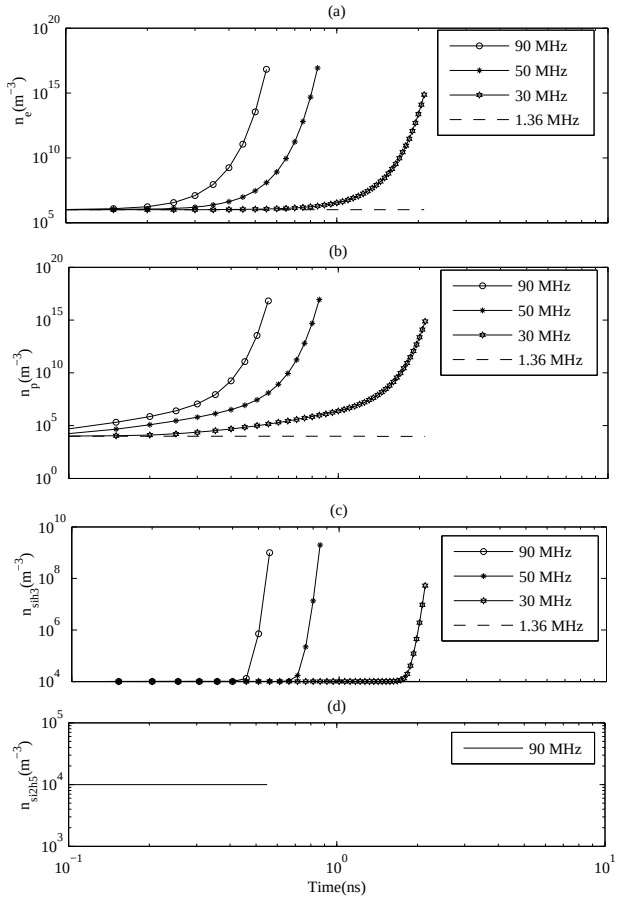

Fig. 3. Temporal variations of density of (a) electrons, (b) SiH_2^+, (c) SiH_3, (d) Si_2H_5

where Θ is the avalanche time constant, N_{e0} is the number of seed electrons that start the avalanche process, and ν_i, ν_a and ν_d are ionization, attachment and detachment frequencies and assumed to be constant values [23].

In this work, the avalanche time constants (Θ) are calculated by an exponential fitting of the obtained data for density dependence on the time of electrons, SiH_2^+ and SiH_3. This parameter is very much helpful to study the speed of production of the different species inside the PECVD reactor.

Avalanche time constant variations with frequency are shown for electrons and SiH_2^+ in Figure 4. As the frequency arises, avalanche time constant becomes shorter. It is obvious that in the higher frequencies, avalanche time constant is smaller and as a result, the production of new species occurs sooner.

It is clear that in higher RF-frequencies, charged particle and radical formation occur in the first moments of time and density of different species increases quickly.

The presented result in the Figure 5 shows an increment in the thin film growth rate as a function of frequency at temperature of 320K. Nienhuis et al showed that the growth rate at higher frequencies is higher [2].

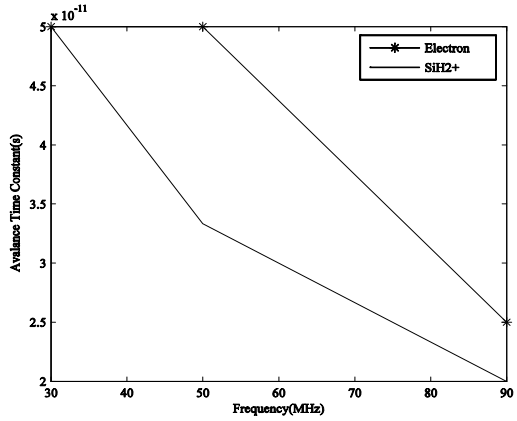

Fig. 4. Variation with frequency of the avalanche time constant (Θ) of electrons and SiH_2^+

On the other hand, at the higher gas temperatures, the ionization rate of Saline gas is higher. This reflects in the higher Silicone atoms generation within the reactor chamber and, their deposition on the substrate will increase. Thus, as seen in Figure 5, at the higher gas temperatures, the growth rate is higher.

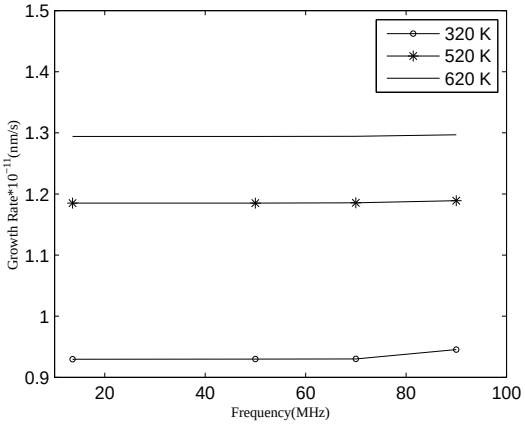

Fig. 5. Thin film growth rate dependence on frequency at the different temperature

Separation of Electrodes

The effect of electrode separation on the discharge properties such as avalanche time constant and consequently on the nano-particles formation process in the PECVD system is considered here. These simulations are performed for the simulation parameters presented in Table II, but the separation of electrodes is varied from 0.03 to 0.27m.

Increase in the electrodes separation has decreasing effects on the electric field in the PECVD chamber. Decrease in the electric field, causes the charged particles move with lesser energy and more slowly and will have lower number of ionizing collisions with the background gas molecules. As a result, it causes reduction in the ionization rate and consequently decreases in production of

charged and radical particles at higher electrodes separation.

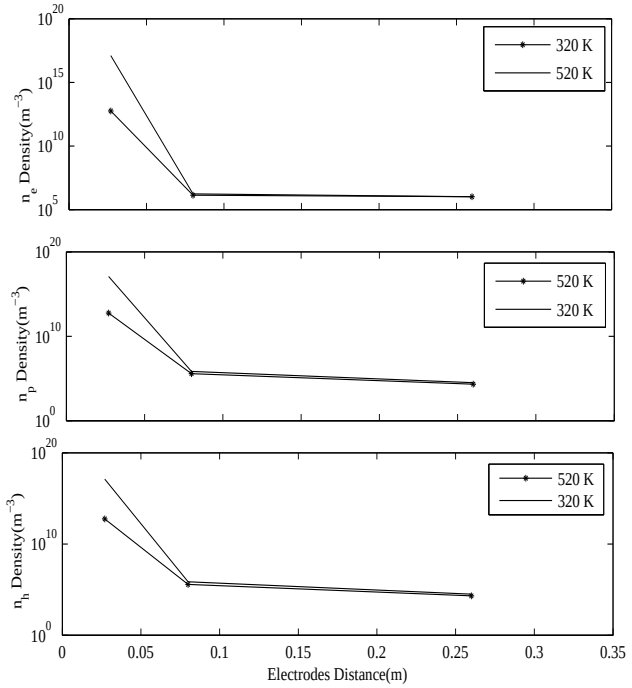

Fig. 6. Density dependence on Electrodes distances, (a) electrons, (b) SiH_2^+, (c) H

Then, as seen in the Figure 6, at smaller electrodes separation, the total number density of the main species in the PECVD reactor (i. e. electron, SiH_2^+ and H) is higher. The strong dependency of nano-particles formation to the applied electric field is shown by Lyka et al [24]. As it is shown in Figure 6, at the same condition, system does not have that much of variations with electrode separations more than 0.8m.

The density growth for particles versus time is represented in Figure 7. As seen, at higher electrode separations, a higher delay in the production of charged species can be observed.

Applied Voltage Amplitude

Increase in V/pd (higher voltage with pd constant) and consequently increase in the mean free path for electrons will lead to an increase in the high energy tail of the electron energy distribution function (EEDF). It causes the production of radicals.

The variation in the amplitude of the applied voltage on the electrodes affects electric field in the discharge medium and consequently effectively the discharge properties and particle growth rate. As shown in the Figure 8, increasing amplitude in order to have greater electric field, contributes ionization thus the creation of different particles with lesser times in the PECVD reactor. When ionization takes place sooner, the production of the charged particles will be more quickly, and then react in order to form radicals.

But, as seen in Figure (7d), Si_2H_5 and greater species still does not grow in the PECVD reactor [2].

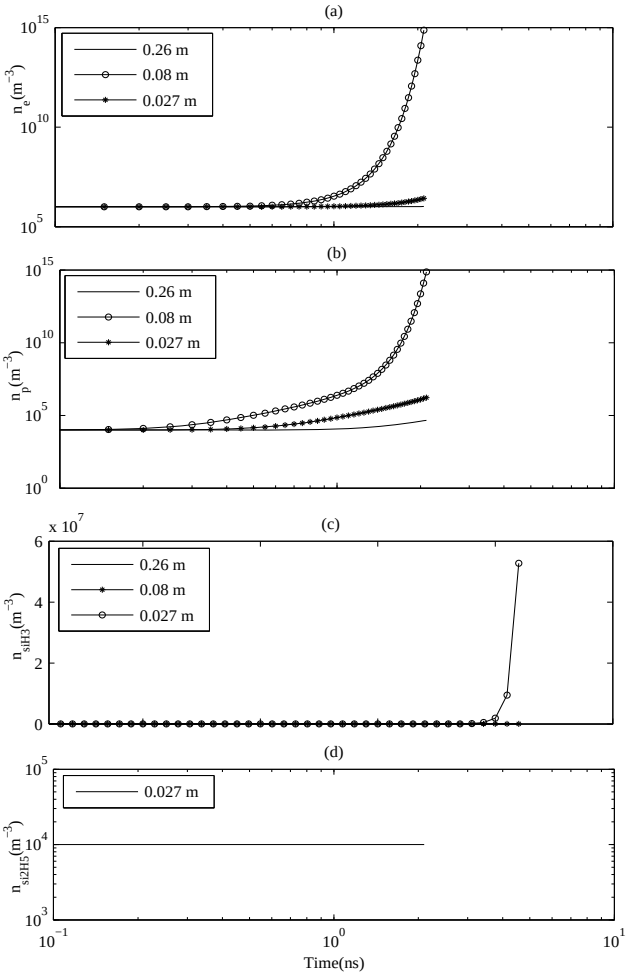

Fig. 7. Temporal variations of density of (a) electrons, (b) SiH_2^+, (c) SiH_3, (d) Si_2H_5

The effects of applied potential amplitude on the electrons, SiH_2^+ and hydrogen atoms number density is depicted in the Figure 9.

It can be seen that increasing the applied voltage peak amplitude has a similar effect on the formation of charged and radical particles as increasing the applied voltage frequency.

Also it is obvious that the particles density at temperature of 520K is more than 620 K which shows the opposite relation between density growing and temperature.

Figure 10 determine avalanche time constant for electron and SiH_2^+ in the various applied potential amplitudes. As it is expected, this parameter decreases with the applied potential amplitude.

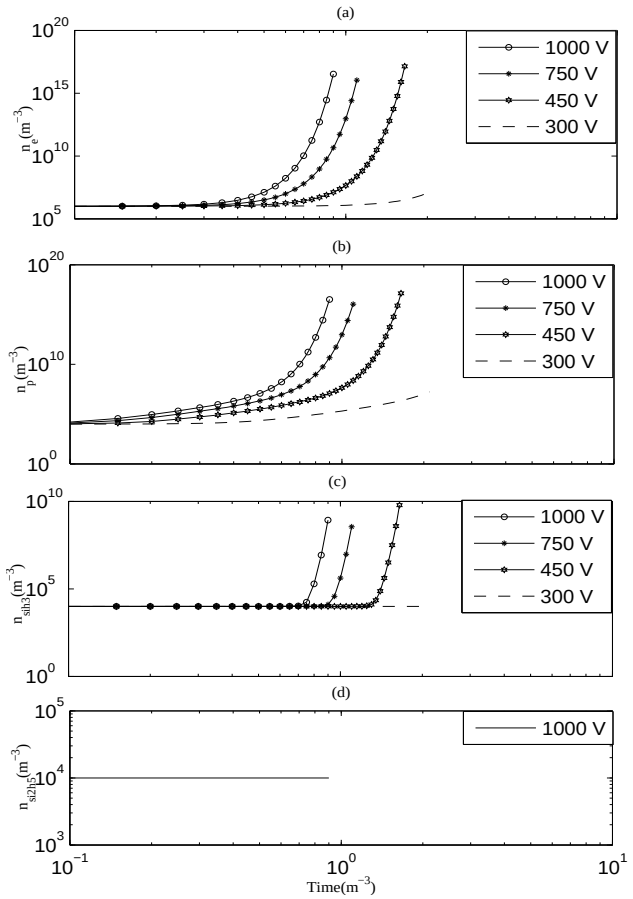

Fig. 8. Temporal variations of (a) electrons, (b) SiH_2^+, (c) SiH_3, (d) Si_2H_5

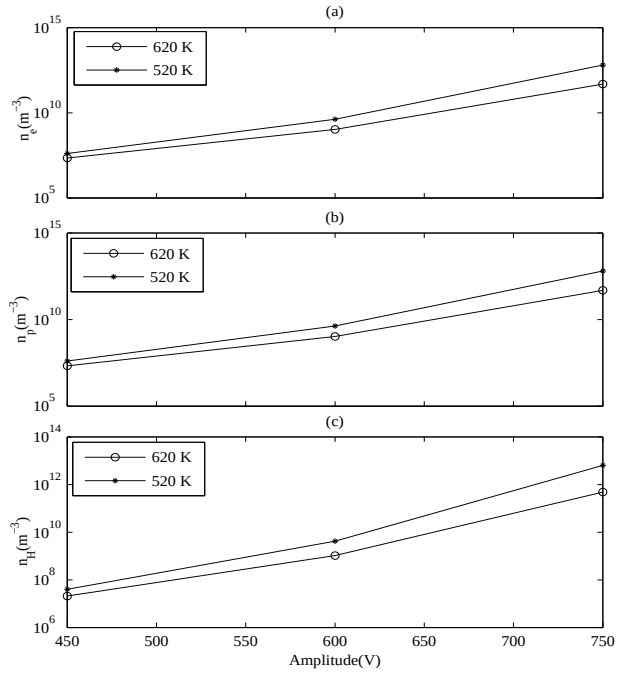

Fig. 9. Density dependence on the amplitude of applied electric potential of (a) electrons, (b) $SiH2^+$, (c) H

It can be concluded that the electric field intensity has a direct influence on the avalanche time constant in the PECVD reactor.

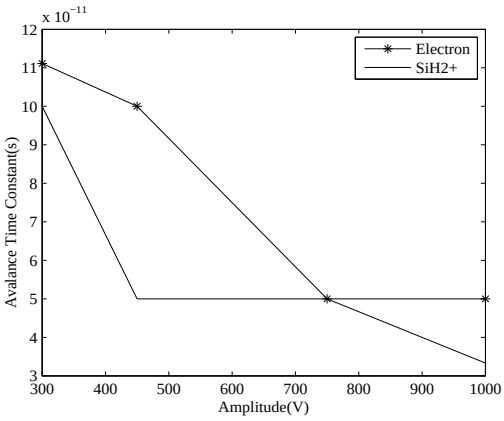

Fig. 10. Variation with amplitude of the avalanche time constant of electrons and SiH_2^+

Huang et al. has shown that the electronic current at higher applied voltages is higher [25]. Their results indicate that as the applied voltage amplitude goes up, the density of different species inside PECVD reactor grows up. Their experiments were performed till applied voltage of 150V. Moreover, Yang et al. showed that as RF power increases, the species concentration grows up [26].

CONCLUSION

In this work, the effects of the main plasma discharge parameters on the avalanche time constant and consequently on the nano-particles formation in the PECVD system using a model based on equations of kinetics are studied. The detailed behavior of ionization process versus time is analyzed. The effects of background gas temperature, applied voltage, applied voltage frequency on the formation of charged and radical particles are studied while keeping the other parameters constant, so that the results are easy to compare and interpret.

The obtained results show that the higher frequency and lower temperature causes more growth density. It was seen that the applied frequency has the most effective impact on growth rate.

It was seen that increase in the applied voltage peak amplitude affects charged and radical particles fairly similar to the applied voltage frequency.

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