3. Modelling of Dust in a Silane/Hydrogen Plasma

Abstract. A dusty radio-frequency silane/hydrogen discharge is simulated, with the use of a one-dimensional fluid model. In the model, discharge quantities like the fluxes, densities and electric field are calculated self-consistently. A radius and an initial density profile for the spherical dust-particles are given and the charge and the density of the dust are calculated with an iterative method. During the transport of the dust, its charge is kept constant in time. The dust influences the electric field distribution through its charge and the density of the plasma through recombination of positive ions and electrons at its surface. In the model this process gives an extra production of silane radicals, since the growth of dust is not included. Results are presented for situations in which the dust significantly changes the discharge characteristics, both by a strong reduction of the electron density and by altering the electric field by its charge. Simulations for dust with a radius of 2 \( \mu \)m show that the stationary solution of the dust density and the average electric field depend on the total amount of the dust. The presence of dust enhances the deposition rate of amorphous silicon at the electrodes because of the rise in the average electron energy associated with the decrease of the electron density and the constraint of a constant power input.

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3.1 Introduction

A radio-frequency discharge in a mixture of SiH₄ and H₂ is often used to deposit thin films of amorphous hydrogenated silicon (a-Si:H), which is used for instance in solar cells and in thin film transistors. The properties of these films can be influenced by changing the plasma process parameters such as RF power, pressure, RF frequency and gas mixture. Study of the complicated chemistry in a SiH₄/H₂ discharge is of importance to optimize the material properties. For economical reasons a high deposition rate and efficient gas usage is desired.

A higher deposition rate can be achieved by increasing the pressure, RF power or frequency. By increasing these parameters complicated chemical reactions involving different ionic and neutral species can be ignited. An important process is the formation of big (molecular) clusters (dust). These clusters, ranging from a few nm to a few µm in radius, can significantly alter the discharge characteristics, and thus the formation of the deposited material. A discharge enters the so-called γ'-regime when the formation of dust becomes significant. With increasing radius the dust particle becomes more and more negatively charged. When the total charge on the dust is large enough, it alters the electric field locally. Here, we report on an investigation of situations in which the dust influences the discharge, by means of numerical simulations with a one-dimensional (1D) fluid model. An important extension compared to existing models [1, 2, 3] is that the behavior of the dust fluid is modelled self-consistently.

The structure of this paper is as follows. The description of the model is given in section 3.2. In section 3.3 simulation results for the γ'-regime are presented and the dependence of the discharge characteristics on the plasma process parameters (e.g. dust density, frequency) are studied. Conclusions are presented in section 3.4.

3.2 Description of the model

3.2.1 SiH₄/H₂ fluid model

To model the dynamics of a dusty plasma, we have used a self-consistent fluid model. This model is an extension of a previously described one-dimensional model [4]. Only the most important aspects of the model will be summarized here. It consists of particle balance equations for the different species (electrons, different ions and different neutrals) and an energy balance equation for the electrons. Ion-neutral collisions have been included to simulate a possible gas heating mechanism. For this we have used a simple approximation by assuming that the energy taken up from the electric field by the ions is dissipated locally in colli-
sions with the background gas [5]. The electric field is calculated by solving the Poisson equation.

In the model the density balance for each species is replaced by the drift-diffusion approximation. The source terms of the electron impact collisions (e.g. dissociation, ionisation) are derived from the electron energy distribution function (EEDF) and expressed as a function of the average electron energy. The EEDF is calculated by solving the Boltzmann equation in the two-term approximation.

In the model the density balance for each species \( j \) is:

\[
\frac{dn_j}{dt} + \frac{d\Gamma_j}{dx} = S_j, \tag{3.1}
\]

where \( n_j \) is the particle’s density, \( \Gamma_j \) the flux of the species and \( S_j \), the sink or source terms.

The momentum balance is replaced by the drift-diffusion approximation, where the particle flux consist of a diffusive term and a drift term,

\[
\Gamma_j = \mu_j n_j E - D_j \frac{dn_j}{dx}, \tag{3.2}
\]

where \( \mu_j \) and \( D_j \) are the mobility and diffusion coefficient of species \( j \). \( E \) is the electric field.

The drift-diffusion approximation assumes that the charged particles will react instantaneously to a change in the electric field. For the ions this is not a valid assumption, because of the low momentum transfer frequency. Therefore an effective electric field is calculated for the ions, to compensate for inertia effects due to the non-instantaneous reaction to a change in the electric field. An expression for the effective electric field, replacing the instantaneous field in Eq. 3.2 is obtained by neglecting the diffusive transport and inserting the expression \( \Gamma_i = \mu_i n_i E_{eff} \) in the simplified momentum balance

\[
\frac{d\Gamma_i}{dt} = \frac{en_i}{m_i} E - \nu_{m,i} \Gamma_i, \tag{3.3}
\]

where \( \nu_{m,i} \) is the momentum transfer frequency of ions given by:

\[
\nu_{m,i} = \frac{e}{\mu_i m_i}. \tag{3.4}
\]

Here \( e \) is the elementary charge and \( m_i \) the mass of the ion. The effective electric field is then given by:

\[
\frac{dE_{eff,i}}{dt} = \nu_{m,i} (E - E_{eff,i}) \tag{3.5}
\]
The electric field $E$ and potential $V$ are calculated using the Poisson equation:

$$\frac{d^2 V}{dx^2} = -\frac{e}{\epsilon_0} \left( \sum n_i - n_e - Q_d n_d \right), \quad (3.6)$$

$$E = -\frac{dV}{dx}, \quad (3.7)$$

where $\epsilon_0$ is the permittivity of vacuum space, $n_e$ the electron density, $n_i$ the ion density, $Q_d$ the charge on a dust particle and $n_d$ the dust density.

The electron energy density $w_e = n_e \epsilon$ (i.e. the product of the electron density and average electron energy $\epsilon$) is calculated self-consistently from the second moment of the Boltzmann equation:

$$\frac{dw_e}{dt} + \frac{d\Gamma_w}{dx} = -e \Gamma_e \cdot E + S_w, \quad (3.8)$$

where $\Gamma_w$ is the electron energy density flux:

$$\Gamma_w = \frac{5}{3} \mu_e w_e E - \frac{5}{3} D_e \frac{dw_e}{dx}, \quad (3.9)$$

and $\mu_e$ and $D_e$ are the electron mobility and electron diffusion coefficients. The term $S_w$ in the electron energy balance equation is the loss of electron energy due to electron impact collisions, including recombination of electrons on the dust particle’s surface.

The plasma-wall interaction is taken into account by introducing a sticking model, each neutral particle has a certain surface reaction probability when it hits the wall. For the chemistry neutral-neutral, electron-neutral, electron-ion, and ion-ion reactions are taken into account; the chemistry is described in detail in a previous article [4]. The plasma is connected in series to an RLC circuit via the surface charge on the electrodes.

### 3.2.2 Dust in the fluid model

#### Charging of dust

When a dust particle exceeds a certain size it can collect more than one electron and be charged up to the floating potential relative to the surrounding plasma. This potential depends on the local ion and electron density and energy distribution. For a spherical dust particle with a radius $r_d$, the Orbital-Motion-Limited theory (OML) [6] predicts a positive ion and electron current:

$$I_i = 4 \pi r_d^2 e n_i \sqrt{\frac{k_B T_i}{2\pi m_i}} \left( 1 - \frac{e V_{fl}}{k_B T_i} \right), \quad (3.10)$$
\begin{equation}
I_c = 4\pi r_d^2 n_e \sqrt{\frac{k_B T_e}{2\pi m_e}} \exp \left( \frac{eV_{fl}}{k_B T_e} \right). \tag{3.11}
\end{equation}

Here, \( n_i \) the positive ion density, \( k_B \) Boltzmann’s constant, \( T_i \) the positive ion temperature, \( T_e \) the electron temperature, \( m_e \) the electron mass and \( V_{fl} \) the floating potential. All species are assumed to have a Maxwellian energy distribution.

When the ions enter the plasma sheaths, they get a drift velocity \( v_i \) due to the electric field. Therefore, we have replaced \( k_B T_i \) in the expression for the ion current by the mean energy \( \langle E_i \rangle \), which is:

\begin{equation}
\langle E_i \rangle = \frac{4k_B T_{gas}}{\pi} + \frac{1}{2} m_i v_i^2. \tag{3.12}
\end{equation}

In the model the charge \( Q_d = 4\pi \varepsilon_0 r_d V_{fl} \) on the dust is calculated by equating these currents, where of course the ion current has been summed over all positive ionic species. The current of negative ions towards the dust particle’s surface is neglected, the negative ions do not have enough kinetic energy to overcome the negative floating potential of the dust particle.

The floating potential of the dust is assumed to be constant during an RF cycle. This assumption is justified by the fact that the currents towards the dust particle surface are too small to change the charge significantly during an rf cycle.

**Recombination on dust particles**

When a dust particle becomes negatively charged, it will attract positive ions, these will recombine with an electron that has to be replaced again by an electron from the discharge to maintain the floating potential. As a result the equilibrium fluxes of positive ions and electrons arriving at the dust surface will recombine. The electron flux (Eq. 3.11) results in a recombination rate:

\begin{equation}
R = 4\pi r_d^2 n_d n_e \sqrt{\frac{k_B T_e}{2\pi m_e}} \exp \left( \frac{eV_{fl}}{k_B T_e} \right). \tag{3.13}
\end{equation}

**Forces acting on a dust particle**

In a plasma dust particles undergo a wide variety of forces. Assuming that a dust particle is a perfect sphere the gravitational force can be written as:

\begin{equation}
F_g = \frac{4}{3} \pi r_d^3 \rho_d g, \tag{3.14}
\end{equation}

where \( \rho_d \) is the mass density and \( g \) is the gravitational acceleration. For amorphous silicon \( \rho_d \) is approximately \( 2.1 \cdot 10^3 \text{ kg/m}^3 \).
3.2. Description of the model

When a dust particle has a velocity relative to the neutral gas, it will experience a drag force due to momentum transfer from/to the gas. This neutral drag force has been discussed in detail by Graves et al [7]. It can be approximated by,

\[ F_n = -\frac{4}{3}\pi r_d^2 n_n (v_d - v_n) v_{th} m_n, \]  

(3.15)

where \( n_n \) is the density of the neutral with mass \( m_n \), \( v_d \) the drift velocity of the dust particle, \( v_n \) the velocity of the gas and \( v_{th} \) the average thermal velocity of the gas. Because advection of the neutral gas is not included in the model, \( v_n = 0 \), this force will only be present as a damping force on the velocity of the dust particles.

Another force caused by momentum transfer is the ion drag. This force results from the positive ion current that is driven by the electric field. It consists of two components. The collection force represents the momentum transfer of all the ions that are collected by the dust particle and is given by:

\[ F_c^i = \pi b_c^2 n_i v_s m_i v_i, \]  

(3.16)

where \( v_s \) the mean speed of the ions and \( b_c \) the collection impact parameter.

The second component is the orbit force given by:

\[ F_o^i = 4\pi b_{\pi/2}^2 \Gamma n_i v_s m_i v_i, \]  

(3.17)

with \( b_{\pi/2} \) the impact parameter that corresponds to a deflection angle \( \pi/2 \) and \( \Gamma \) the Coulomb logarithm.

\[ \Gamma = \frac{1}{2} \ln \left( \frac{\lambda_L^2 + b_{\pi/2}^2}{b_c^2 + b_{\pi/2}^2} \right), \]  

(3.18)

\( \lambda_L = \left( (1/\lambda_e)^2 + (1/\lambda_i)^2 \right)^{-1/2} \) is the linearized Debye length, which is a combination of the electron Debye length, \( \lambda_e \), and the ion Debye length, \( \lambda_i \). The ion drag is discussed in more detail by Barnes et al [8].

Due to their charge, dust particles will experience an electric force. Daugherty et al [9] derived the following expression:

\[ F_e = Q_d E \left( 1 + \frac{\kappa r_d}{3(1 + \kappa r_d)} \right), \]  

(3.19)

where \( Q_d \) is the charge on the dust particle, \( E \) is the electric field and \( \kappa = 1/\lambda_L \). In a discharge the dust particle radius is much smaller than the linearized Debye length, therefore the term between the bracket is approximately 1 and the electric force is given by:

\[ F_e = Q_d E. \]  

(3.20)
This expression holds for situations where the dust particles are not shielded from
the plasma by positive ions trapped in orbitals around the dust particle [10]. In
that case the particle plus ion cloud will behave as some kind of dipole.

When a temperature gradient is present in a discharge, for instance due to
cooling or heating of the electrodes a third force driven by momentum transfer
will occur. This force is called the thermophoretic force. Atoms impinging from
the hot side have more momentum than their companions of the cold side, this can
result in a force pointing in the direction \(-\frac{dT_{\text{gas}}}{dx}\).

For large Knudsen numbers Talbot et al [11] derived the following expression:

\[
F_{\text{th}} = \frac{-32}{15} \frac{r_d^2}{v_{th}} \left( 1 + \frac{5\pi}{32} (1 - \alpha) \right) \kappa_T \frac{dT_{\text{gas}}}{dx},
\]  

(3.21)

\(\kappa_T\) is the translation part of the thermal conductivity. \(\alpha\), the thermal accommoda-
tion coefficient of the gas is taken equal to 1. In our case, the total thermophoretic
force has been calculated as the summation over the two inlet gasses.

**Implementing dust in SiH\(_4\)/H\(_2\) fluid model**

To obtain a suitable expression for the flux of dust particles, we assume that the
neutral drag force is in equilibrium with the sum of the other forces. This as-
sumption is valid when the final steady state is approached, but should be relaxed,
for instance, when the dust is injected at a high velocity. In that case the inertia
of the dust should not be neglected. With the introduction of a momentum loss
frequency and a mobility and diffusion coefficient for the dust particles given by:

\[
\nu_{md} = \sqrt{\frac{2}{k_B T_{\text{gas}}} \frac{p_{tot}}{m_d} \pi r_d^2} \sqrt{\frac{8k_B T_{\text{gas}}}{\pi m_d}},
\]  

(3.22)

\[
\mu_d = \frac{Q_d}{m_d \nu_{md}},
\]  

(3.23)

\[
D_d = \frac{\mu_d}{k_B T_{\text{gas}}},
\]  

(3.24)

it is possible to define a ”drift-diffusion” expression for the flux of the dust parti-
cles,

\[
\Gamma_d = -\mu_d n_d E_{\text{eff}} - D_d \frac{dn_d}{dx} - \frac{n_d}{\nu_{md}} g
\]

\[+ \sum_{\text{ions}} \frac{n_d m_i v_s}{m_d \nu_{md}} \left( 4\pi b_s^2 / 2 \Gamma + \pi b_c^2 \right) \Gamma_i
\]

\[- \frac{32}{15} \frac{n_d v_d^2}{m_d \nu_{md} v_{th}} \kappa_T \frac{dT_{\text{gas}}}{dx},
\]  

(3.25)
and treat them with the same numerical procedures as the other charged particles in the fluid model. Note that the ion drag force is summed over all the positive ionic species. Because of the low mobility of the dust particles the effective field $E_{\text{eff}}$ is approximated by the time averaged RF field. The diffusion originates from the pressure gradient, $k_B T_d \frac{dn_d}{dx}$. The Einstein relation couples the diffusion and the mobility coefficients, see equation 3.24.

The internal pressure of the crystal due to the inter-particle interaction has been included by means of a density dependence of the diffusion coefficient for the dust. The diffusion coefficient of the dust is increased by a factor $\exp(N_d/N_c)$ where the reference density $N_c$ is chosen such that the dust density saturates at a value $N_{\text{cryst}}$. This models the incompressibility of the crystal. Actually, the (yet unknown) equation of state of the dust crystal should be used to account for the internal pressure. Since we were not primarily interested in the precise structure of the crystallized regions, we have chosen for the simple and computationally robust exponential increase of $D_d$.

The drift velocity and the diffusion coefficient of the dust fluids are much smaller than those of the ions and electrons. Therefore it would require a large computational effort to achieve a steady state solution for the dust when it is followed during an RF cycle. We therefore have developed a method to speed up the convergence toward the steady state solution by introducing a different calculation cycle with a different time step for the dust. Our model thus consists of two calculation cycles. In the first one, the transport equations of the ions, electrons and the Poisson equation are solved during a number of rf cycles, during the RF cycles the dust does not move. After that, the transport equation of the dust is solved with a greater time step, using the time averaged electric field, and electron and positive ion fluxes. During the second calculation, space charge regions are created, because the electron and positive ion densities do not change. These space charge regions will lead to instabilities in the solution of Poisson equation and the electron transport. To solve this problem, we correct the artificially generated space charge by adapting the positive ion density distributions prior to the next series of RF cycles, in which the ion and electron density profiles adapt themselves to the new dust density profile. With this method the required speed-up is established.

## 3.3 Results and discussion

In this section the results, obtained with the 1D numerical code are presented. A comparison is made between two almost identical initial situations. The only parameter that varies is the total amount of dust.

Other plasma process parameters, i.e., the gas temperature (400 K), gas flows (30 sccm SiH$_4$ and 30 sccm H$_2$), RF frequency (50 MHz), RF power (5 W), pres-
sure (40 Pa), electrode spacing (2.7 cm) and the dust particle radius (2 µm) are kept constant. Gravity is neglected. The simulation starts with a fourth order polynomial for the initial dust density profile. Figure 3.1 shows the dust density profile for a line integrated amount of 2.15x10^8 m^-2. It shows that the steady state profile differs significantly from the initial profile. The high peaks formed in the dust density profile are result of the balance between the electric force, ion drag and thermophoretic force at these positions [12, 13].

Figure 3.2 shows the density profile of the dust in case of a low (7.2 \times 10^4 m^{-2}) and high (8.47 \times 10^8 m^{-2}) line integrated amount of dust. This figure shows clearly that the position where the resulting force acting on the dust particles vanishes, depending on the line integrated amount of dust in the reactor. This shows that the dust considerably influences the discharge. Figure 3.3 shows the charge on a dust particle. It varies through the discharge according to the local ion and electron densities and the electron temperature. In the sheaths there are almost no electrons (Fig. 3.4), this results in a strong decrease of the dust particle’s charge. In the pre-sheaths a reasonable number of electrons is available with a high energy. This results in maxima of the grain charge at the pre-sheaths. In the center of the discharge the electron energy is very low, this makes it for the electrons difficult to overcome the repulsive Coulomb force caused by the charge on the dust particle’s surface. This effect represents itself in a minimum of the grain charge at the center. For the case of a high amount of dust (see the dashed line), the dust is also influencing its own charge by enhancing the positive ion.
density and thus increasing the ion flux towards its surface at the positions where the dust density peaks appear, this results in local minima in the charge profile.

Figure 3.2: Dust density in m\(^{-3}\) as function of the position. The solid curve represents a dust density profile with an integrated amount of dust of \(7.2 \cdot 10^4\ m^{-2}\), the dust density is normalized with a factor of \(1.27 \cdot 10^8\). The dashed curve represents a dust density profile with an integrated amount of dust of \(8.47 \cdot 10^8\ m^{-2}\), the dust density is normalized with a factor of \(1.66 \cdot 10^{11}\).
Figure 3.3: Charge on a dust particle in number of electrons as function of the position. The line integrated amount of dust differs between the two curves. The solid curve: $7.2 \cdot 10^4 \text{ m}^{-2}$, dashed curve: $8.47 \cdot 10^8 \text{ m}^{-2}$.

Figure 3.4: The time-averaged electron density in $\text{m}^{-3}$ as function of the position. The line integrated amount of dust differs between the three curves. The solid curve: No dust. Dotted curve: $2.15 \cdot 10^8 \text{ m}^{-2}$. Dashed curve: $8.47 \cdot 10^8 \text{ m}^{-2}$.
3.3. Results and discussion

Figure 3.5: The time-averaged electron energy in eV as function of the position. The line integrated amount of dust differs between the three curves. The solid curve: No dust. Dotted curve: $2.15 \cdot 10^8 \text{ m}^{-2}$. Dashed curve: $8.47 \cdot 10^8 \text{ m}^{-2}$.

According to equation 3.13, the large surface area of the dust particles leads to a significant recombination rate. As result the electron density will drop (Fig. 3.4) and the average electron temperature will increase because the applied power is constant. This is shown in figure 3.5.

When the line integrated amount of dust is low its influence can be neglected, and the electron temperature is low in the quasi-neutral bulk, this is due to the lack of electric field. Only in the plasma-sheath transition zone the electrons can gain energy, as shown by the two peaks in figure 3.5. When the line integrated amount of dust particles is large (dashed curve in fig. 3.5), the heating is more uniform, this is due to the electron density drop which enhances the Debye length and therefore the electric field can penetrate much further into the discharge. Also the oscillating field in the center increases. Figure 3.6 shows the electric potential for different integrated amounts of dust. For a large amount the potential increases. This is a result of the constraint of a constant power, because the electron density decreases (Fig. 3.4) the displacement current also decreases. To compensate for this effect, the RF voltage has to increase, which results in a higher plasma potential.

The higher electron temperature in this case will lead to an increase in the ionization rate (needed to compensate for the recombination on the dust) and dissociation rate. The dissociation rate increases even if the electron density decreases because the rate coefficient almost exponentially increases with the electron energy. Therefore the deposition rate also increases with the dust density. This is
Figure 3.6: The time-averaged electric potential in V as function of the position. The line integrated amount of dust differs between the three curves. The solid curve: No dust. Dotted curve: \(2.15 \cdot 10^8\) m\(^{-2}\). Dashed curve: \(8.47 \cdot 10^8\) m\(^{-2}\).

Figure 3.7: The deposition rate in nm/s as function of the line integrated dust density.
shown in figure 3.7. This increase in the deposition rate can not explain the significant increase which occurs in the experiments [4]. This may be caused by neglecting the contribution of dust in the sticking model. For large amounts of dust, the deposition rate again decreases because the gain in electron temperature can no longer compensate for the loss of electrons, therefore the dissociation of SiH$_4$ and ionization will drop. At still higher amounts of dust, the discharge cannot be maintained any longer and the simulation crashes.

Figure 3.8: The net space charge in elementary charges as function of the position for a line integrated amount of dust of $2.15 \cdot 10^8$ $m^{-2}$, represented by the solid curve and normalized with a factor $1.82 \cdot 10^{14}$. The dotted curve represents the dust density profile in $m^{-3}$ for a line integrated dust density of $2.15 \cdot 10^8$ $m^{-2}$, normalized with a factor $7.07 \cdot 10^{10}$.

Figure 3.8 shows the net space charge for a line integrated amount of dust of $2.15 \cdot 10^8$ $m^{-2}$. As expected, positive space charge regions appear close to the electrodes. At the center between the electrodes a quasi-neutral region can be observed. An interesting phenomenon induced by the dust can be observed. A double space charge layer appears around the sharp boundary of the dust crystal. The positive space charge layer in front of the dust crystal boundary is caused by the recombination on the dust particle’s surfaces of ions and electrons entering the crystal. As in front of an absorbing wall, the difference in mobility between the ions and the electrons results in a net positive space charge that enhances the electric field which in turn accelerates the ions from the center of discharge towards the dust crystal to compensate for the mobility difference. The negative
space charge layer appears very close to the sharp edges inside the dust crystal. It appears due to the fact that the diffusion of the ions prohibits a full compensation of the fast rising negative charge of the dust crystal.

3.4 Conclusions

Simulations with the one-dimensional fluid model show that the deposition rate in SiH$_4$/H$_2$ can be increased by the presence of dust particles with a considerable size (2 $\mu$m). The recombination on these particles alters the radio-frequency SiH$_4$/H$_2$ discharge in such a way, that the electron temperature increases and the electron density decreases. This process leads to an increase in the dissociation and ionization rates. The modelled increase in deposition rate underestimates the increase observed in the experiments [4] during the $\alpha$-$\gamma'$ transition. Depending on the total amount of dust the width of the plasma sheaths may change considerably, thus altering the position where the dust will settle. An other interesting phenomenon is the appearance of a double space charge layer at the edge of the dust crystal which is formed by the difference in mobility between the ions and electrons and the diffusive transport of the ions caused by the steep density gradient imposed by the dust.

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References