Self-Consistent Modeling of Particle Growth in DC Dusty Discharge

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Nucleation and growth of nanostrusctured dust particules is of particular interest for different fields. In laboratory and industrial chemically active discharges, dust can affect the performance of produced electronic devices or films [1]. In the ITER reactor, for instance, they can pose important safety problems [2].

We report on the modeling of a laboratory DC discharge with a graphite cathode developed at the PIIM laboratory where carbon particles are formed by cathode pulverisation [3]. We previously proposed a particle formation and growth mechanism where particles are formed by nucleation due to negative precursors with high residence time [4]. The molecular growth is facilitated by a trapping effect in the field reversal region of the DC discharge. We first made the assumption that densities of charged precursors and particles were negligible as compared to the electron density, meaning we could consider an electropositive stationary discharge, in pristine argon. However, after a discharge duration of 300 s, negative carbon species could have densities comparable to that of the electrons. Our previous assumption of constant discharge parameter only controlled by electron and argon ion densities then fails. We thus propose here a scheme that couples discharge equilibrium, with a precursors and particles dynamics model. This coupling self-consistently takes into account the evolution of the discharge parameters and their effect on the precursors and particles formation and growth.

1. The dynamic DC discharge model

Results from our previous modeling for the particle formation have shown that precursors and particle growth occurs in the field reversal region and that they are trapped at this position as negatively charged clusters and dust particles ("heavies"). This trapping occurs only in the negative glow (NG) and the Faraday dark space (FDS) regions. Therefore, in order to accommodate long discharge times after which the density of negative heavies becomes significant with respect to the electron density, we modify the discharge model in these two regions, and include these new species in our discharge equilibrium equations. The cathode fall and positive column regions should not be affected and their equations are left unchanged. The cold electron population balance, i.e. the electrons of low enough energy to be trapped by the electric field reversal, and which constitute the bulk of the electron density, is governed by ambipolar transport fluxes, Eq1, which includes production by ionization, losses onto particles and clusters due to sticking and diffusive losses to the reactor walls.

$$\frac{\partial n_e}{\partial t} = -\vec{\nabla}\left(-D_e\vec{\nabla}n_e + \mu_e n_e \vec{E}_{amb}\right) + S_{ioni}(x) - P^e_{dust} - P^e_{attach} - P^e_{rad}\left(\frac{\sum n^-}{n_e}\right) \quad \text{Eq 1}$$

The electric field is obtained from the requirement that the cold electrons and heavies carry no net current, leading to a balance of their ambipolar fluxes, Eq2.

$$\vec{E}_{amb} = \frac{D_+\vec{\nabla}n_+ - D_e\vec{\nabla}n_e - \sum Z_- D_-\vec{\nabla}n_-}{\mu_e n_e + \mu_+ n_+ + \sum Z_-^{2} \mu_- n_-} \quad \text{Eq2}$$

The new discharge model is solved self-consistently with the precursor and particle models.

2. Results

The same case as was presented in [4] is re-run with the new dynamic discharge model. The old results correspond to the 'initial' label on the Figures 1 and 2, corresponding to the situation when the carbonaceous species are only present as traces in the discharge and do not perturb it. We then evolve this initial case, with continuous carbon cluster production and dust growth dynamics, for up to 600 seconds of discharge time.

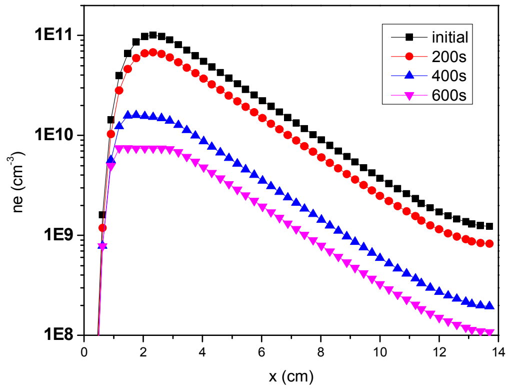

Figure 1. Electron density profiles as a function of time. The cathode is located at the *x=0* position.

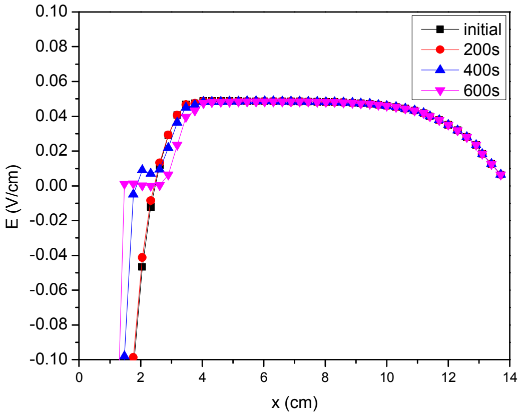

Figure 2. Electric field profiles as a function of time.

The main consequence of the build-up of heavy negative species, as can be seen in Figure 1, is the concomitant decrease in electron density, which is most pronounced between 200 and 400 s of discharge time. Over that interval, the electron density is reduced by roughly an order of magnitude. What we also see from Figure 2 is that the extent of the region where the electric field is positive increases, towards the cathode, by about 1 cm. This means that the large presence of negative charged species pushes back against the static discharge field and that the area where particles are confined and growth can occur also increases.

However, we see that the electron density decrease slows down at late times (after 400 s). We know from our previous work that one of the limiting steps for negative cluster formation is electron attachment on the C_4 clusters, to form C_4^-, which then will undergo molecular growth processes until reaching the threshold nucleation size. As we can see from Figure 3, this attachment rate strongly decreases after a maximum near 200 s, mainly because of the progressive rarefaction of the cold electrons that would be attached. We find a similar behaviour concerning the particle nucleation rate, which drops as a function of time, following the formation rate of negative clusters.

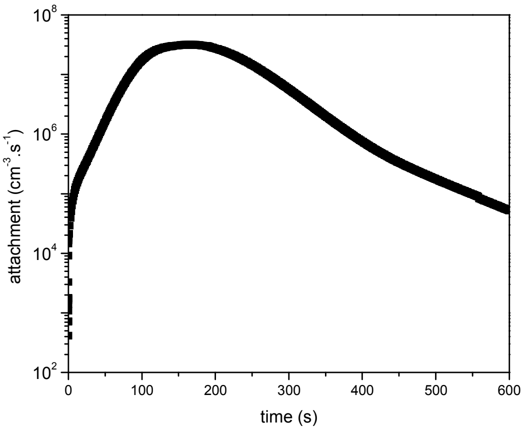

Figure 3. Time evolution of the attachment rate producing C_4^- clusters.

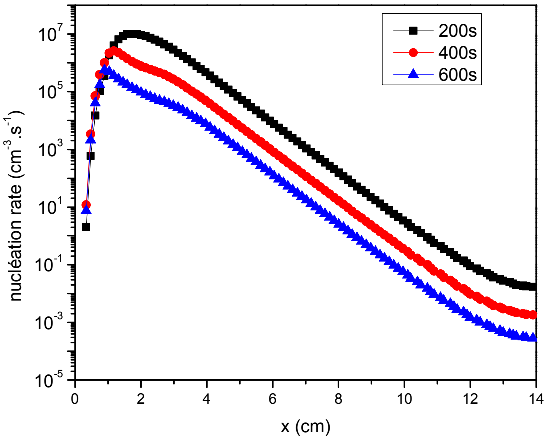

Figure 4. Profiles of the particle nucleation rate along the discharge.

We now consider, Figures 5 and 6, the density and average diameter of the dust particles present in the discharge. We see that the principal increase of dust particle density occurs in the area near the sheath entrance where we also found the extension of the field reversal region. Moreover, we find that the particle size, as well as the average particle charge to which it is correlated (not shown), increases dramatically as a function of discharge duration.

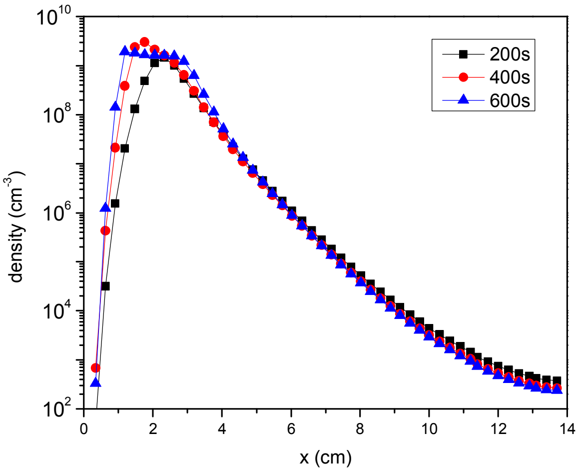

Figure 5. : particle density along the discharge

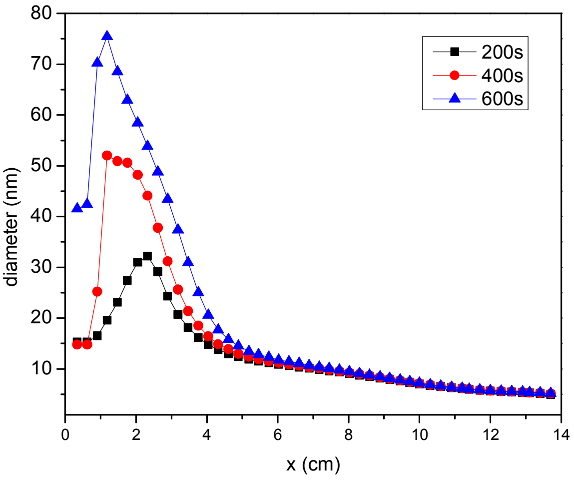

Figure 6. : particle density along the discharge

The time evolution of the various reaction rates and particle properties are consistent with the experimental finding that there is only one generation of dust particles present and that the carbon sputtered at late times is consumed by sticking to the already existing particles, as opposed to participating in the production of new nuclei. The growth rate seen is also in fair agreement with the measurements reported in [3].

3. Conclusions

We have developed a new self-consistent dynamic discharge model for describing a DC Argon discharge containing a large number of carbonaceous dust particles, which are allowed to grow according to a molecular growth and aerosol dynamics model. As the particle density becomes significant and the discharge transitions from electropositive to electronegative, we see an increase of the electric field reversal region towards the cathode, a large growth of a single generation of dust particles, and a strong reduction in the population of cold electrons, reproducing the reported experimental behaviour. The next step in model improvement is a more detailed treatment of the dust particles by treating them not in the aggregate but rather by binning them according to size, which is ongoing work.

Acknowledgements

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