## Charge and temperature fluctuation of carbon nanoparticles in a DC dusty plasma

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**Abstract:** Statistical nature of particle charge and temperatures of particles in a dusty plasma have been studied. Commonly used models for mean charge and temperature are not accurate for small particles (< 10 nm) and it is necessary to couple charge and temperature fluctuations. Small particles can get heated to very high temperatures compared to large particles. The plasma conditions significantly affect the morphology of the nanoparticles produced.

Keywords: Dusty plasma, nanoparticle, charge, temperature

## 1. Introduction

We consider argon DC glow discharges, generated between two parallel electrodes of 10 cm diameter. The carbon nanoparticles are formed through sputtering of polycrystalline graphite cathode working at a pressure of 60 pa. The detailed description of the discharge is available [1]. Numerical studies have been performed to analyze the nanoparticle formation and the aerosol dynamics in the discharge [2]. These studies have shown that particle charging is a key parameter for the growth of the carbon nanoparticles. Especially the charge of a small particle of radius less than 10 nm can fluctuate resulting in a significant fraction of neutrally charged particles that promote coagulation. Moreover, the charge dynamics of the particle strongly depend on the local plasma conditions as the instantaneous charge is determined by the collisions with electrons and ions.

These collisions further cause intense heating of the particle. Arnas and Mouberi [3] have performed a detailed thermal balance of such a particle based on average charge. The main mode of particle heating was attributed to ion recombination at the surface. It is observed that the small particles can achieve much higher temperature compared to the surrounding gas while the large particles are close to equilibrium. This also plays a role on the morphology of the particles where small particles were observed to be crystalline while the larger particles were amorphous in nature.

Matsoukas et al [4] have shown that the charge fluctuations can be represented by a Gaussian distribution where the mean charge of the distribution can be obtained from OML equilibrium. However, this approach is no more applicable to small particles as discrete nature of charging becomes significant and cannot be ignored. Therefore, the mean temperature determined using the OML equilibrium charging rates is not accurate for small particles and statistical coupling between charge and thermal balance need to be considered. [5]. The present study discusses the statistical dynamics of charging and its effects on thermal balance at different stages of evolution of the DC dusty plasma for small particles.

## 2. Discussion

For this study, we specifically consider three different plasma conditions namely at 100, 200 and 600 s where the ratio of electron density to ion density ( $N_e/N_i$ ) varies from 1, 0.5 and 0.1 respectively [6]. We have performed statistical analysis of charge and temperature for small particles of  $r_p$  1 to 50 nm, as described in Prasanna et al [5].

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r <sub>p</sub> (nm)	<b>q</b> <sub>oml</sub>	<b>q</b> <sub>stat</sub>
1	-0.25	-0.91
2.5	-0.63	-0.99
5	-1.26	-1.66
50	-12.6	12.9

Table 1. Average charge of particles calculated using OML equilibrium and statistical approach for  $N_e=N_i$ 

The average charge of particles of different radius calculated using the traditional OML equilibrium approach and statistical approach have been tabulated in Table 1. It is seen that OML equilibrium value is not accurate for small particles while both models are agreeable for large particles.

The average charge as well as the distribution is strongly influenced by the plasma conditions mainly  $N_e/N_i$ . Figure 1 shows the charge distribution obtained for particle of size 1nm radius at different  $N_e/N_i$  ratios. For the plasma conditions considered, it is seen that significant fraction of particles are neutrally charged even for an isolated particle in a plasma. As the concentration of particles increase, electrons are consumed resulting in a lower  $N_e/N_i$  ratio, which results in lower mean charge of the particle. Consequently, the concentration of neutrally charged particles also increase. It has to be noted that the neutrally charged small particles have significant role on the particle growth by coagulation and hence affects the whole particle size distributions [2]. As a result, the small particles which are continuously produced by nucleation are more quickly consumed by coagulation at 600 s when compared to at 100 s.



Fig. 1 Charge distribution of a particle of  $r_p$  1nm for different N<sub>e</sub>/N<sub>i</sub> ratios at T<sub>e</sub> = 0.1 eV and pressure 60 pa.

However, this observation only depicts the particle size distribution but does not characterize the particle morphology. We perform thermal balance for these particles and the average temperature has been tabulated in Table 2. The mean temperature of the particle decreases with increase in particle size for all plasma conditions. Also, the temperature of the particles decreases as the particle density increases i.e.  $N_e/N_i$  decreases.

Table 2. Average temperatures (K) of particles for various plasma conditions ( $N_e/N_i$ )

		Ne/Ni		
		1	0.5	0.1
rp (nm)	1	710	682	645
	2.5	518	510	562
	5	490	475	478
	50	468	454	444

The morphology of the particle is closely related to the particle temperature. However, the mean temperature alone is not an indicator of the crystallinity of the particle, but it is necessary to consider the statistical temperature history of the particles. Smaller particles have low thermal mass and hence can be instantaneously heated to very high temperatures when compared to larger particles. For example, 1 nm particle in these conditions achieves a maximum temperature of 3000 K which is necessary condition for producing crystalline particles. Further, the

temperature distribution of 1 nm size particles for different plasma conditions is shown in Fig. 2. The fraction of particles having temperatures larger than the phase transition reduces with reduction in  $N_e/N_i$  ratios. This means initial stages of plasma when the particle density is low should produce higher concentration of crystalline nanoparticles. However, after long duration of dusty plasma operation, the particles would be predominantly amorphous in nature.



Fig. 2 Temperature distribution for particle of  $r_p$  1 nm at different N<sub>e</sub>/N<sub>i</sub> ratios and T<sub>e</sub> = 0.1 eV and pressure 60 Pa

## 3. References

- [1] C. Dominique, and C. Arnas, "Cathode sputtering and the resulting formation of carbon nanometer-size dust," *Journal of Applied Physics*, vol. 101, no. 12, pp. 123304-8, 2007.
- [2] A. Michau, C. Arnas, and K. Hassouni, "Aerosol dynamics in a sputtering DC discharge," *Journal* of Applied Physics, vol. 121, no. 16, pp. 163301, 2017.
- [3] C. Arnas, and A. Mouberi, "Thermal balance of carbon nanoparticles in sputtering discharges," *Journal of Applied Physics*, vol. 105, no. 6, pp. 063301, 2009.
- [4] T. Matsoukas, and M. Russell, "Particle charging in low-pressure plasmas," *Journal of Applied Physics*, vol. 77, no. 9, pp. 4285-4292, 1995.
- [5] S. Prasanna, A. Michau, K. Hassouni *et al.*, "Effect of charge fluctuation on nanoparticle heating in dusty plasma," *Plasma Sources Science and Technology*, vol. under review, 2019.
- [6] A. Michau, C. Arnas, G. Lombardi *et al.*, "Nanoparticle formation and dusty plasma effects in DC sputtering discharge with graphite cathode," *Plasma Sources Science and Technology*, vol. 25, no. 1, pp. 015019, 2016.