Self-organization and Electrolyte Ion Mass Transport Processes in DC Glows

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Self-organization in plasmas is observed in phenomena ranging from plasmoid formation in low pressure, RF plasmas to large-scale, and magnetized structures observed on the surface of the sun. Of recent interest is the formation of self-organization patterns on the surface of liquid anodes in 1 atm DC glows. Shirai [1] documented an array of such patterns over a broad parameter spaced including the variation of gap spacing, current, and sensitivity to feed gas trace oxygen concentration. While these patterns are of interest in regards to understanding collective phenomena, the appearance of the patterns may play an important role in the sub-surface liquid phase chemistry, driving convection and inducing thermal gradients.

In many studies to date of organization resulting from DC glow discharges on liquids, salt water is typically used as the electrolyte. In this work, the effect of a different electrolyte—copper sulfate—was investigated. At similar solution conductivities and applied voltages reported previously with salt water, it was found that the self-organization patterns are markedly different. A side-by-side comparison is shown in Fig. 1 for the self-organization patterns of glow discharges on salt-water pattern and copper sulfate solution. A new, complex, star-shaped structure with round edges was observed with $CuSO_4$. What role does the electrolyte play in determining the pattern shape? This observation suggests that electrolyte ion mass or perhaps ionization state may play a key role in determining overall pattern shape. This dependence has not previously been explored.

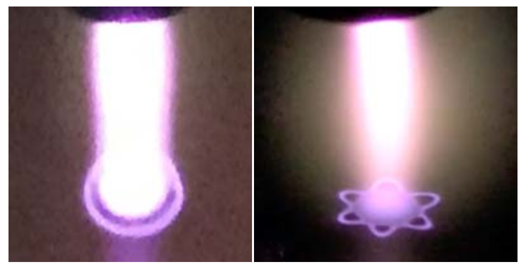


Figure 1 – Different shapes of self-organization anode patterns at 40 mA.

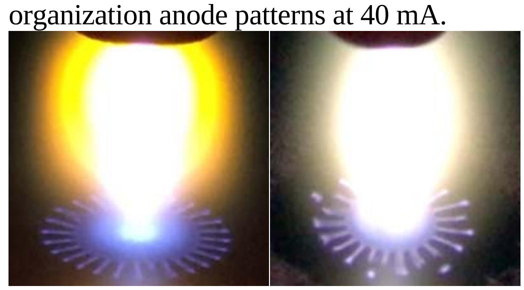


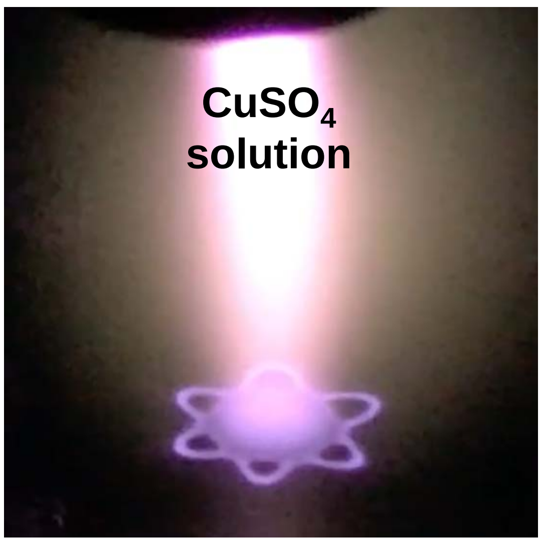
Figure 2 – Plasma emission clouds image with patterns at 2.2kV, 8mm gap length with 200 sccm He flow. *Note: for figures 1 and 2, (L) NaCl solution. (R) $CuSO_4$ solution.*

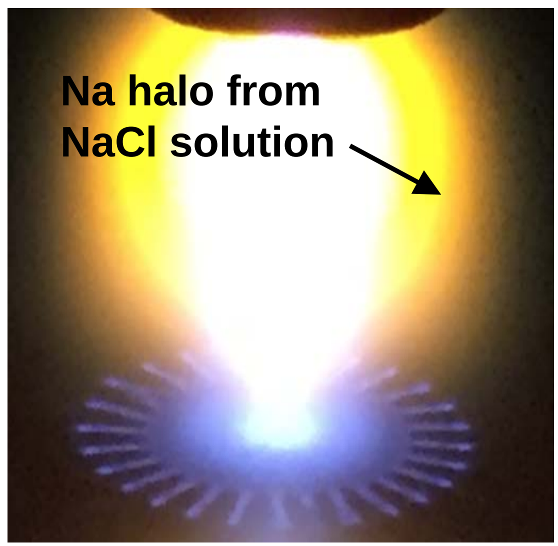
Another interesting comparison between the salt-water solution and a copper sulfate solution for a DC glow is shown in Fig. 2. The transport of ions and electrons as well as the role of electrolytic species to discharge maintenance are not well understood. What is quite apparent, however, is that in both cases a prominent halo surrounds the main plasma column. Spectroscopic analysis of the halo suggests that it consists of sodium in the case of the salt electrolyte and copper in the case of the copper sulfate solution. In these studies, the solution ions play a role not only in electrolytic processes in solution but also apparently in the gas phase plasma. The research issues we are continuing to pursue include: How does the introduction of these low ionization potential species into the plasma column affect ionization? Is penning ionization therefore an important process in discharge maintenance? The relationship between this ionic mass transport from the liquid into the gas phase is being investigated.

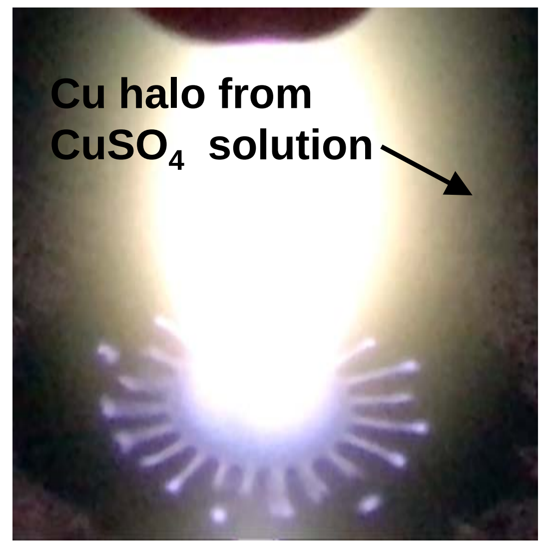
References: [1] N. Shirai, S.Uchida and F. Tochikubo, Plasma Sources Sci. Technol. **23,** 054010 (2014).

A NEW SELF-ORGANIZATION ANODE PATTERN OBSERVED IN ATMOSPHERIC DC GLOWS

- **Copper sulfate solution (DI water) with controlled conductivity of 12 mS/cm was investigated s an anode liquid electrode.**
- **A vector star shaped structure observed on the surface of copper sulfate solution at low current region– a new self-organization anode pattern.**
- **Optical emission spectroscopy revealed a sodium halo from a salt solution and copper cloud from a $CuSO_4$ solution surrounding the positive column from presumably derived from electrolyte evaporation.**







- **A vector star shape**
- **Anode spokes with sodium and copper halo**

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Nanoparticles in Low Temperature Plasmas: Not all Negatively Charged

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Although particles in non-thermal dusty plasmas tend to charge negatively, a population of non-negative particles may still exist. These particles are not electrostatically trapped in the plasma, and thus can diffuse to walls. Additionally, they can facilitate coagulation. In a collaboration between the University of Minnesota and Lam Research Corporation, we conducted Monte Carlo charging simulations to investigate the effects of several parameters on the fraction of particles that are not negatively charged [1]. These simulations accounted for two effects not considered by the orbital motion limited theory of particle charging: single-particle charge limits, which were implemented by calculating electron tunneling currents from particles; and the increase in ion currents to particles caused by charge-exchange collisions that occur within a particle's capture radius. The effects of several parameters were considered, including particle size, in the range 1 to 10 nm; pressure, ranging from 0.1 to 10 Torr; electron temperature, from 1 to 5 eV; positive ion temperature, from 300 to 700 K; plasma electronegativity, characterized in terms of n_+ / n_e ranging from 1 to 1000; and particle material, either SiO_2 or Si.

Within this parameter space, higher non-negative particle fractions are associated with smaller particle size, higher pressure, lower electron temperature, lower positive ion temperature, and higher electronegativity. Materials with lower electron affinities, such as SiO_2 (1.0 eV), have higher non-negative particle fractions than materials with higher electron affinities, such as Si (4.05 eV). Pressure has a strong effect on the non-negative particle fraction, as shown in Fig. 1, because increasing pressure causes incoming ions to be more likely to experience a charge-exchange collision within a particle's capture radius.

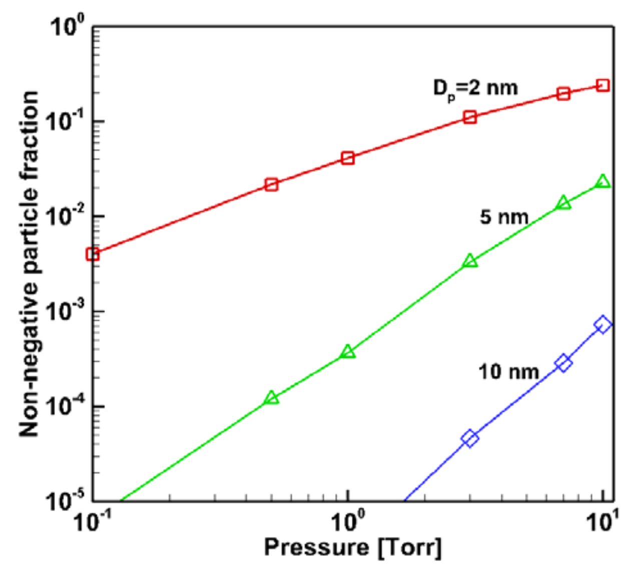


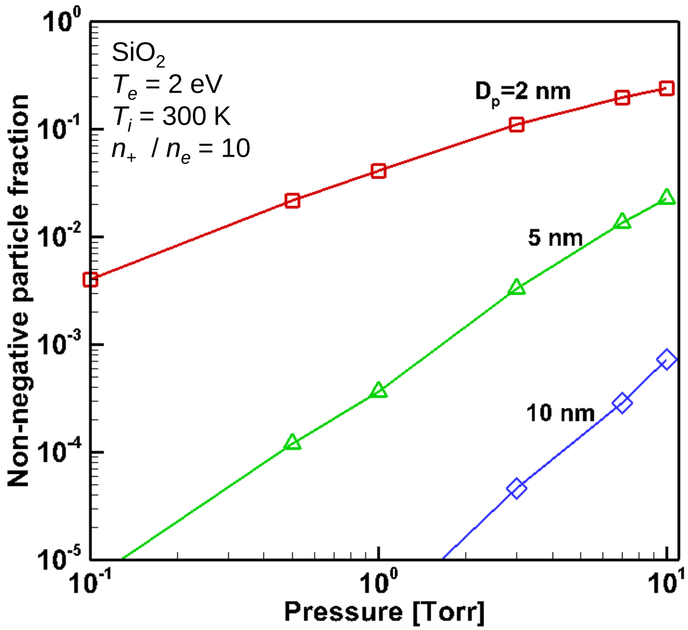
Figure 1 – Effect of pressure and particle diameter D_p on fraction of SiO_2 particles that are non-negative. Conditions: $T_e = 2$ eV, $T_i = 300$ K, $n_+/n_e = 10$.

References

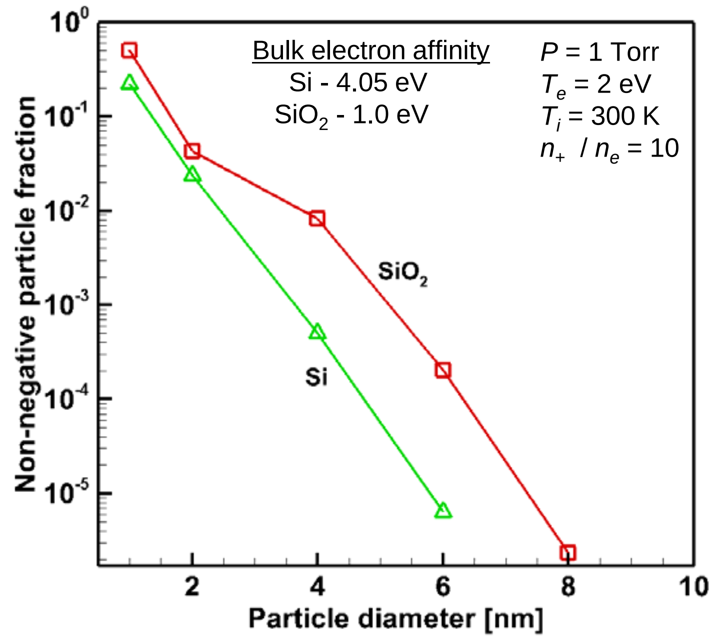
[1] M. Mamunuru, R. Le Picard, Y. Sakiyama and S. L. Girshick, *Plasma Chem. Plasma Process.*, DOI: 10.1007/s11090-017-9798-6, accepted for publication Feb. 13, 2017.

NANOPARTICLES IN LOW-TEMPERATURE PLASMAS ARE *NOT* ALL NEGATIVELY CHARGED

- **Motivation: nanoparticles in plasmas are expected to be negatively charged, trapping them in plasma and suppressing coagulation**
- **Conducted Monte Carlo charging simulations to investigated conditions that affect fraction of nanoparticles that are neutral or positively charged[1]**



- **Increasing pressure increases positive ion current to particles by increasing frequency of charge-exchange collisions within particle's capture radius.**



- **Material's electron affinity affects electron tunneling from particles, resulting in different particle charge limits.**

Collaboration between U. Minnesota & Lam Research Corp.

[1] **M. Mamunuru, R. Le Picard, Y. Sakiyama and S. L. Girshick, *Plasma Chem. Plasma Process.*, accepted for publication Feb. 13, 2017.**

HIGHLIGHT