A Kelvin-Helmholtz-Like Instability at the Plasma-Liquid Interface
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Plasma-induced effects in liquids begin at the gas-liquid boundary. Here, chemical reactions are driven by gas phase plasma species as well as physical processes such as diffusive or turbulent mixing, evaporation, and sputtering. The interface acts as the source function for the production of longer-lived species that ultimately diffuse into the bulk liquid. The mass transport of these plasma activated species into the bulk liquid is currently an active area of study. Computational models in particular are being brought to bear on these complex interfacial interactions as in general it is difficult to experimentally interrogate the boundary. In an effort to elucidate processes occurring at the interface, a 2-dimensional trapped bubble apparatus is being used to study not only chemically reacting fronts emerging from the interface but also to understand the role of plasma-induced fluid dynamical effects. These plasma induced processes can play a major role in the transport of radicals far from the interface,[1] Motion in the cell is constrained in 1-dimension so that essentially true 2-d plasma driven physical and chemical flows can be studied. The apparatus enables direct optical interrogation of the interfacial region.

In a recent study, chemical probes were used to assess the transport of reactive oxygen species produced at the interface into the bulk liquid. Large-scale circulation patterns were observed and which originated at the interface. By tracking precipitates formed at the leading edge of the chemical front, a radial velocity gradient at the interface was observed. Precipitates trapped in the circulation patterns traveled faster near the interface than in the far field, suggesting that shear forces are also important. The apparatus was redesigned to enable Schlieren interferometry (which measures changes in index of refraction) to investigate gradients in density at the interface as well as elucidate physical processes that may be driving the flows. Once a streamer discharge is initiated in the bubble, a well-defined interface surrounding the bubble is observed in the Schlieren images. This interface tends to become unstable over time as inferred by observing fingering and rotation in the Schlieren images. Since earlier experiments suggested velocity shear at the interface, the fingering instability is likely due to the onset of a Kelvin-Helmholtz-like instability. Capillary waves initiated at the interface likely seed the instability which then grows over time. A Schlieren image of this fingering instability is shown in Fig. 1. Future work will involve characterizing the growth rate of the instability and obtaining a better understanding its role in producing the previously observed convection patterns. We hope to use these findings to better understand larger scale systems currently under study in the areas of plasma-based water purification and plasma medicine.

References
OBSERVATION OF KELVIN-HELMHOLTZ-LIKE INSTABILITY AT THE PLASMA-LIQUID INTERFACE

- Using a newly constructed 2-D bubble apparatus, Schlieren measurements of plasma induced density gradients at the interface are being studied.
- Previous measurements suggest that plasma induced, liquid flow speeds near the bubble interface are higher than distant points which suggests that shear forces are important at the interface.
- Schlieren images revealed an active boundary layer at the interface that becomes unstable over time giving rise to a Kelvin-Helmholtz-like instability.

- No plasma
- Pulsed plasma in bubble
Plasma-Induced Crystallization of Silicon Nanoparticles

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In a collaboration between Girshick and coworkers at the University of Minnesota (UMN) and Kushner and coworkers at the University of Michigan, we recently developed a two-dimensional numerical model of silicon nanoparticle synthesis in a flowing RF plasma.[1] Algorithms for the kinetics of nanoparticle formation were self-consistently embedded into a plasma hydrodynamics simulation to account for nanoparticle nucleation, surface growth, charging, coagulation and transport. The model was used to conduct numerical simulations of experiments conducted by Aydil, Kortshagen and coworkers at UMN, who hypothesized that nanoparticle crystallization in their system is enabled by plasma heating.[2] In particular, it was estimated in Ref. [2] that heterogeneous recombination reactions on nanoparticle surfaces of positive ions and plasma-generated radicals (particularly H atoms) could heat the nanoparticles to temperatures several hundred degrees above the neutral gas temperature, which otherwise is too low to explain the observed formation of silicon nanocrystals.

Among the simulation outputs are predicted densities of positive ions and H, and of gas temperature, for the conditions of the experiments. These results were then used to calculate nanoparticle heating. The results, shown in Fig. 1 for 2-nm-diameter Si particles, indicate that the nanoparticles can indeed be heated by several hundred degrees above the gas temperature, and to high enough temperatures to enable crystallization. The numerical results also provide new insights on the relation between plasma properties and nanoparticle heating, as well as on how nanoparticle heating scales with parameters such as plasma power.

References

Figure 1 – (left) Predicted density profiles of positive ions (N⁺) and H atoms. (right) Gas temperature and internal temperature of 2-nm-diameter Si nanoparticles.
PLASMA-INDUCED CRYSTALLIZATION OF SILICON NANOPARTICLES

- Hypothesis: Crystallization of Si nanoparticles enabled by plasma heating [1].
- Developed 2D numerical model that self-consistently embeds detailed aerosol dynamics into plasma hydrodynamics simulation [2].
- Conducted simulations for geometry and conditions of experiments [3].

Results from simulations are consistent with hypothesis of plasma-induced crystallization.