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Double-layer structures in low-temperature atmospheric-pressure electronegative rf microplasmas: separation of electrons and anions

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Abstract – Stratification of negatively charged species in electronegative discharges is a well-known phenomenon that can lead to various double-layer structures. Here we report on the separation of electrons and anions in atmospheric pressure electronegative microdischarges. In these discharges, electrons oscillate between the electrodes moving across and beyond an electronegative core. As a result of this motion, positively charged regions form between the oscillating electron ensemble and the central electronegative discharge (Fig. 1a).

Low-temperature atmospheric-pressure microplasmas have received growing attention in recent years for their potential use in many technological applications, including plasma medicine. [1-3] In this emerging field, helium is often used as a buffer gas due to its excellent thermal properties while water is inevitably present due to the moist nature of biological targets. In addition, water can also be introduced as a precursor in the feed gas to generate reactive oxygen species of biological relevance.[4] Motivated by the need of better understanding the dynamics and chemistry of He+H₂O discharges, we have performed computer simulations that reveal intricate spatio-temporal profiles in these plasmas. A selection of the simulation results is presented in Figure 1. While the data presented corresponds to a He/H₂O admixture, similar behavior is expected in other electronegative plasmas.

The model used to simulate a parallel plate reactor is a conventional 1-D fluid model.[5] The 27 species and 58 reactions used in the model are taken from ref. [6], where more than 500 reactions were screened to identify the dominant chemical processes. In this study, the water concentration is fixed at 0.3%, the input power at 1W/cm² and the discharge is driven by a voltage source at 13.56MHz. Under these conditions the discharge displays a clear electronegative character.

Due to the electronegativity of the discharge, double layers that confine the colder negatively charged species (anions) to the center of the discharge appear in these plasmas. Double layers, standing or travelling, have been

reported in a variety of systems including magnetized and unmagnetized plasmas, collisionless and collisional regimes and electropositive and electronegative discharges. [7-11] For electronegative discharges confined in a single cavity (as it is the case under study here), three different regimes are typically observed.[7,8,9] At low electronegativity (α), the discharge stratifies into an electronegative core with electropositive edges. As α increases, the electropositive edges slowly disappear, and at even larger α , the plasma density profile in the discharge center flattens. Transitions between these regimes depend among other things on plasma density and pressure. Here, however, we report on a different double layer structure that is observed in microplasmas when the gap size is reduced.

As the discharge gap reduces, the width of the bulk plasma decreases and the sheaths progressively occupy a larger portion of the gap (see spatiotemporal evolution of the space charge profiles in Fig.1a-d). It has been shown that when this happens in electropositive discharges the quasineutral bulk plasma is not longer stationary and it oscillates between the two electrodes following the motion of the electron ensemble.[5,12,13] In the case of an electronegative discharge, however, negative ions remain confined in the discharge center and due to their large inertia their spatial oscillation is negligible (Fig.1i-l). Therefore, an electronegative core plasma forms in the discharge center. For the He/H₂O admixture considered here, the electronegativity is high and electropositive edges are not observed in time averaged profiles of any of the discharges (data not shown explicitly). The stratification of electrons and negative ions, however, is readily visible when comparing figure 1e-h and 1i-l.

Of particular interest is the structure found when, at the input power and driven frequency considered in the study, the discharge gap is reduced below 600 μ m. There the amplitude of the electron oscillation becomes larger than half the discharge gap and the electron ensemble is found to move across and beyond the electronegative core (Fig. 1e-h). The resulting space charge distribution becomes then strongly non-monotonous with “islands” of high positive space charge forming between the oscillating electron ensemble and the electronegative central core (Fig. 1a-b).

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Fig. 1. Spatio-temporal profiles in a low-temperature atmospheric pressure He+H₂O rf microplasma. a-d) net space charge density; e-h) electron density; i-l) total negative ion density (OH⁻, H₂O₂⁻, H₃O₂⁻, H₅O₃⁻, H⁻, O⁻); m-p) total positive ion density (OH⁺, H₂O⁺, H₃O⁺, H₅O₂⁺, H₇O₃⁺, H₉O₄⁺, H₁₁O₅⁺, H₁₃O₆⁺). White lines are superimposed on the figures to indicate the regions of quasi-neutrality.

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