Double-Layer Structures in Low-Temperature Atmospheric-Pressure Electronegative RF Microplasmas: Separation of Electrons and Anions

Kirsty McKay, Ding-Xin Liu, Felipe Iza, Member, IEEE, Ming-Zhe Rong, Member, IEEE, and Michael G. Kong, Senior Member, IEEE

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.

Index Terms—Atmospheric-pressure plasma, electronegative discharge, microplasmas, plasma dynamics.

D OW-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 spatiotemporal profiles in these plasmas. A selection of the simulation results is shown in Fig. 1. While the data presented correspond 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 [6], where more than 500 reactions were screened to identify the dominant chemical processes. In this paper, the water concentration is

K. McKay, F. Iza, and M. G. Kong are with the Department of Electronic and Electrical Engineering, Loughborough University, LE11-3TU Leicestershire, U.K. (e-mail: K.McKay@lboro.ac.uk; f.iza@lboro.ac.uk; m.g.kong@lboro.ac.uk).

D.-X. Liu and M.-Z. Rong are with the State Key Laboratory of Electrical Insulation and Power Equipment, Xi'an Jiaotong University, Xi'an 710049, China (e-mail: dxliu@stu.xjtu.edu.cn; mzrong@mail.xjtu.edu.cn).

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fixed at 0.3%, the input power is fixed at 1 W/cm², and the discharge is driven by a voltage source at 13.56 MHz. 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 traveling, 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]–[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 discharge gap [see the spatiotemporal evolution of the space charge profiles in Fig. 1(a)-(d)]. It has been shown that, when this happens in electropositive discharges, the quasi-neutral 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. 1(i)-(1)]. 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 Fig. 1(e)–(h) and (i)–(l).

Of particular interest is the structure found when, at the input power and driven frequency considered in this paper, 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

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Fig. 1. Spatiotemporal profiles in a low-temperature atmospheric-pressure $He + H_2O$ RF microplasma. (a)–(d) Net space charge density. (e)–(h) Electron density. (i)–(l) Total negative ion density $(OH^-, H_2O_2^-, H_3O_2^-, H_5O_3^-, H^-, and O^-)$. (m)–(p) Total positive ion density $(OH^+, H_2O^+, H_3O^+, H_5O_2^+, H_7O_3^+, H_9O_4^+, H_{11}O_5^+, and H_{13}O_6^+)$. White lines are superimposed on the figures to indicate the regions of quasi-neutrality.

beyond the electronegative core [Fig. 1(e)-(h)]. The resulting space charge distribution becomes then strongly nonmonotonous with "islands" of high positive space charge forming between the oscillating electron ensemble and the electronegative central core [Fig. 1(a)-(b)].

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