

# 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.

**L**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 + H<sub>2</sub>O 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/H<sub>2</sub>O 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

fixed at 0.3%, the input power is fixed at 1 W/cm<sup>2</sup>, 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 ( $\alpha$ ), the discharge stratifies into an electronegative core with electropositive edges. As  $\alpha$  increases, the electropositive edges slowly disappear, and at even larger  $\alpha$ , 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)–(l)]. Therefore, an electronegative core plasma forms in the discharge center. For the He/H<sub>2</sub>O 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  $\mu\text{m}$ . There, the amplitude of the electron oscillation becomes larger than half the discharge gap, and the electron ensemble is found to move across and

Manuscript received November 30, 2010; revised May 11, 2011; accepted May 11, 2011. Date of publication June 9, 2011; date of current version November 9, 2011. This work was supported by the Engineering and Physical Sciences Research Council, U.K.

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).

Color versions of one or more of the figures in this paper are available online at <http://ieeexplore.ieee.org>.

Digital Object Identifier 10.1109/TPS.2011.2156815

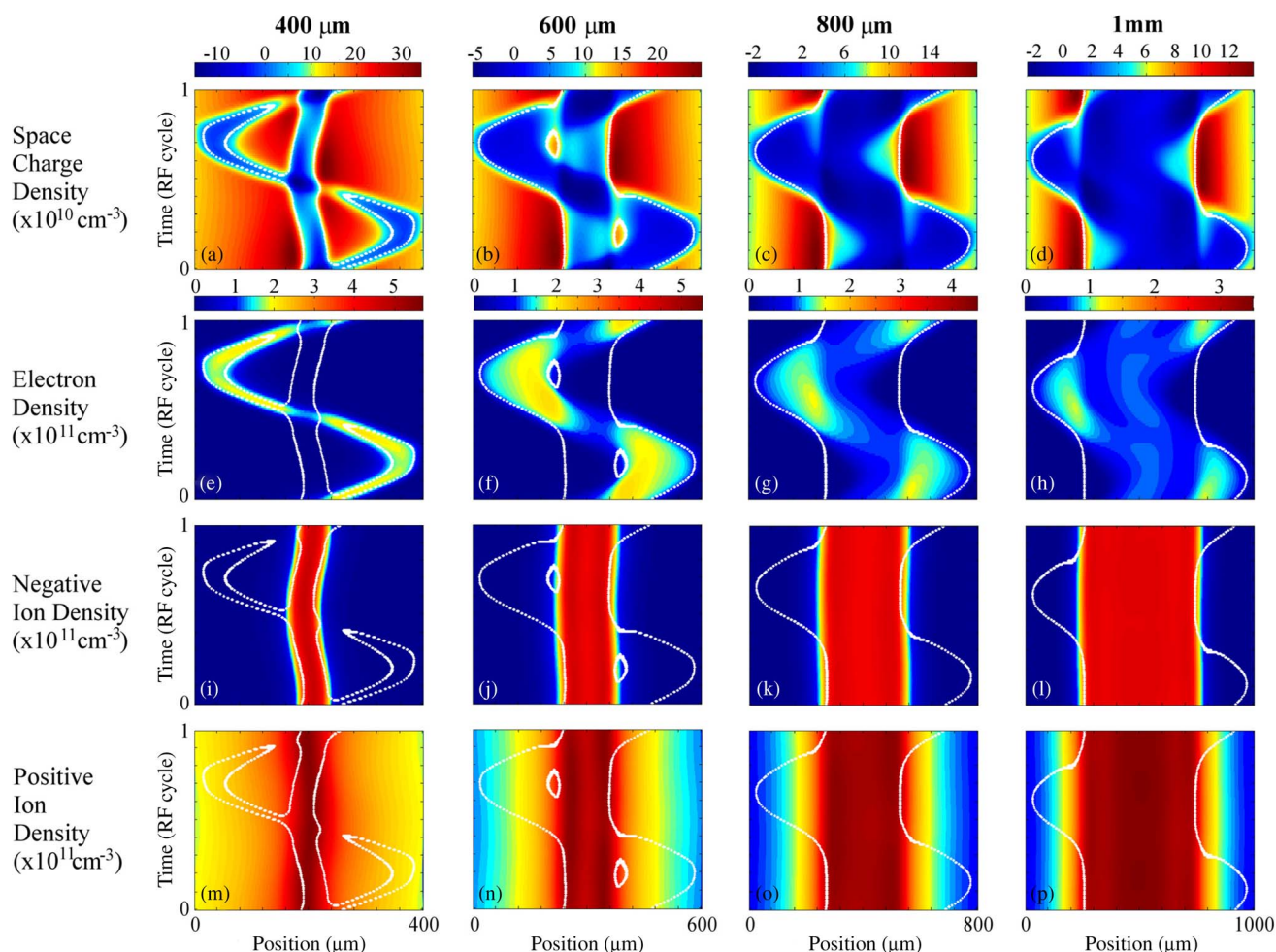


Fig. 1. Spatiotemporal profiles in a low-temperature atmospheric-pressure He + H<sub>2</sub>O RF microplasma. (a)–(d) Net space charge density. (e)–(h) Electron density. (i)–(l) Total negative ion density (OH<sup>-</sup>, H<sub>2</sub>O<sub>2</sub><sup>-</sup>, H<sub>3</sub>O<sub>2</sub><sup>-</sup>, H<sub>5</sub>O<sub>3</sub><sup>-</sup>, H<sup>-</sup>, and O<sup>-</sup>). (m)–(p) Total positive ion density (OH<sup>+</sup>, H<sub>2</sub>O<sup>+</sup>, H<sub>3</sub>O<sup>+</sup>, H<sub>5</sub>O<sub>2</sub><sup>+</sup>, H<sub>7</sub>O<sub>3</sub><sup>+</sup>, H<sub>9</sub>O<sub>4</sub><sup>+</sup>, H<sub>11</sub>O<sub>5</sub><sup>+</sup>, and H<sub>13</sub>O<sub>6</sub><sup>+</sup>). 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 nonmonotonic with “islands” of high positive space charge forming between the oscillating electron ensemble and the electronegative central core [Fig. 1(a)–(b)].

#### REFERENCES

- [1] F. Iza, G. J. Kim, S. M. Lee, J. K. Lee, J. L. Walsh, Y. T. Zhang, and M. G. Kong, “Microplasmas: Sources, particle kinetics, and biomedical applications,” *Plasma Process. Polym.*, vol. 5, no. 4, pp. 322–344, Jun. 2008.
- [2] M. G. Kong, G. Kroesen, G. Morfill, T. Nosenko, T. Shimizu, J. van Dijk, and J. L. Zimmermann, “Plasma medicine: An introductory review,” *N. J. Phys.*, vol. 11, p. 115 012, Nov. 2009.
- [3] G. Fridman, G. Fridman, A. Gutsol, A. B. Shekhter, V. N. Vasilets, and A. Fridman, “Applied plasma medicine,” *Plasma Process. Polym.*, vol. 5, no. 6, pp. 503–533, Aug. 2008.
- [4] D. X. Liu *et al.*, “He/O<sub>2</sub>/H<sub>2</sub>O plasmas as a source of reactive oxygen species,” *Appl. Phys. Lett.*, 2010, to be published.
- [5] D. W. Liu, F. Iza, and M. G. Kong, “Electron avalanches and diffused gamma-mode in radio-frequency capacitively coupled atmospheric-pressure microplasmas,” *Appl. Phys. Lett.*, vol. 95, no. 3, p. 031 501, Jul. 2009.
- [6] D. X. Liu, P. Bruggeman, F. Iza, M. Z. Rong, and M. G. Kong, “Global model of low-temperature atmospheric-pressure He + H<sub>2</sub>O plasmas,” *Plasma Sources Sci. Technol.*, vol. 19, no. 2, p. 025 018, Apr. 2010.
- [7] T. E. Sheridan, “Double layers in a modestly collisional electronegative discharge,” *J. Phys. D, Appl. Phys.*, vol. 32, no. 15, pp. 1761–1767, Aug. 1999.
- [8] S. Kim, M. A. Lieberman, A. J. Lichtenberg, and J. T. Gudmundsson, “Improved volume-averaged model for steady and pulsed-power electronegative discharges,” *J. Vac. Sci. Technol. A*, vol. 24, no. 6, pp. 2025–2040, Nov. 2006.
- [9] D. D. Monahan and M. M. Turner, “Global models of electronegative discharges: Critical evaluation and practical recommendations,” *Plasma Sources Sci. Technol.*, vol. 17, no. 4, p. 045 003, Nov. 2008.
- [10] A. Meige, N. Plihon, G. J. M. Hagelaar, J.-P. Boeuf, P. Chabert, and R. W. Boswell, “Propagating double layers in electronegative plasmas,” *Phys. Plasmas*, vol. 14, pp. 053 508-1-1–053 508-1-11, May 2007.
- [11] “The electro-negative character of He/O<sub>2</sub>,” presented at the Gaseous Electron. Conf., Paris, France, 2010, Paper BT3.00006.
- [12] F. Iza, J. K. Lee, and M. G. Kong, “Electron kinetics in radio-frequency atmospheric-pressure microplasmas,” *Phys. Rev. Lett.*, vol. 99, no. 7, p. 075 004, Aug. 2007.
- [13] J. J. Shi and M. G. Kong, “Evolution of discharge structure in capacitive radio-frequency atmospheric microplasmas,” *Phys. Rev. Lett.*, vol. 96, no. 10, p. 105 009, Mar. 2006.