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Interaction of Nonthermal Plasmas and Organic Liquids

Felipe Iza

¹ Wolfson School of Mechanical, Electrical and Manufacturing Engineering Loughborough University, LE11-3TU, UK e-mail: f.iza@lboro.ac.uk

Advancements in non-thermal plasmas operating at atmospheric pressure have made possible novel plasma processing of liquids that were not conceivable in conventional vacuum systems due to vapor pressure limitations.¹ In addition to their rapidly growing use in plasma medicine,² plasma agriculture and food industries,^{3,4} nanoparticle synthesis,⁵ catalysis⁶ and water treatment,⁷ the interaction of plasmas with organic liquids opens additional avenues of research, including the development of novel organic compounds for the characterization of plasmas^{8,9} and the use of gas plasma in novel chemical synthesis processes.^{10,11}

Of special interest are novel chemical synthesis processes in which one has the potential to eliminate waste streams. For example, existing oxidation processes often generate large waste streams (e.g. the widely used oxidant Oxone produces ~25 kg of waste per kg of oxygen transferred), whilst plasma-driven oxidation has the potential to completely eliminate the oxidant waste stream.¹⁰

In all these emerging applications, plasmas are brought in contact with liquids (e.g. body fluids, culture media, water, organic solvents, etc) but our current understanding of the plasma-liquid interaction and the transport of reactive species from the gas to the liquid phase remain limited. Furthermore, the lack of readily accessible detection methods to quantify reactive species in plasma-treated liquids further hinders progress. As any other gas-liquid interface, plasma-liquid interfaces are inherently asymmetric, display steep gradients and discontinuities, and involve a rich variety of physicochemical processes.

It turns out that fluid dynamics play a very important role in most plasma-liquid systems, even in those without imposed flows or active mixing. In fact, minor changes in gas/liquid composition can significantly affect the plasma liquid interaction, the induced flow patterns and thereby the process efficacy.

In addition to the complex fluid dynamics, chemical transport and thermodynamic processes encountered in conventional gas-liquid interfaces, plasma-liquid interfaces also involve nonthermal chemistry, nonlocal particle kinetics, and electromagnetic forces, which introduce new challenges both for theoretical and experimental studies. From electrohydrodynamic effects not seen in normal gas-liquid systems¹² to the generation

of Marangoni flows due to the non-equilibrium nature of the plasma chemistry,¹³ there is little doubt that plasmaliquid interfaces warrant further fundamental investigation before we can harness their full potential.

Although operation at atmospheric pressure limits the ability to generate high-energy ions, nonthermal atmospheric pressure plasmas create reactive environments at low temperature that initiate and catalyze useful chemical reactions. As a result, most plasma-liquid applications exploit this non-equilibrium chemistry. For example, in plasma medicine oxidative atmospheres are created in oxygen-containing plasmas for the inactivation of cells and microorganisms. Similarly, oxidative species are used for the oxidation of pollutants in water treatment.

Here we explore the possibility of exploiting the plasma chemistry to drive environmentally-friendly organic synthesis processes. For example, reactive oxygen species created in nonthermal atmospheric pressure plasmas can be used to drive epoxidation processes with much better atom economy than conventional chemical methods; and plasma generated electrons can be used to reduce alkynes and alkenes, as well as to drive carboxylation reactions that incorporate waste CO_2 into valuable organic compounds.

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