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Opinion

Plasma and Aerosols: Challenges, Opportunities and Perspectives

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Received: 2 August 2019; Accepted: 10 September 2019; Published: 14 September 2019

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Featured Application: Plasma-aerosol systems open new potential opportunities in a wide range of applications including agriculture, combustion control, nanotechnology, medicine and cosmetics.

Abstract: The interaction of plasmas and liquid aerosols offers special advantages and opens new perspectives for plasma–liquid applications. The paper focuses on the key research challenges and potential of plasma-aerosol interaction at atmospheric pressure in several fields, outlining opportunities and benefits in terms of process tuning and throughputs. After a short overview of the recent achievements in plasma–liquid field, the possible application benefits from aerosol injection in combination with plasma discharge are listed and discussed. Since the nature of the chemicophysical plasma-droplet interactions is still unclear, a multidisciplinary approach is recommended to overcome the current lack of knowledge and to open the plasma communities to scientists from other fields, already active in biphasic systems diagnostic. In this perspective, a better understanding of the high chemical reactivity of gas–liquid reactions will bring new opportunities for plasma assisted in-situ and on-demand reactive species production and material processing.

Keywords: plasma–liquid interaction; aerosol; droplet; plasma diagnostic; plasma medicine; material processing; plasma agriculture; plasma cosmetics; nanotechnology

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1. Plasma-Liquid as a Bench of Emerging Applications from 2010

Plasmas in/above/with liquids [1] science and technology at atmospheric pressure have been reawakened these last few years, partly due to the very active research activity in the field of plasma medicine and the requirement to account for liquid layers covering in vitro cell culture or organs during in vivo or clinical studies. Grown from the pioneering works of Gubkin [2] and Cavendish [3], and accounted as the origin of life on Earth [4,5], the investigation of plasma–liquid interaction at atmospheric pressure became a fruitful and multidisciplinary research field [6,7]. Focus was drawn, in particular, to the crucial role of so-called reactive oxygen and nitrogen species (RONS) [8] in various biomedical applications, ranging from biomaterials to therapeutics. The use of plasma-treated liquids has also been proposed where a direct contact between target and plasma needs to be avoided or is not possible. The combination of extensive efforts devoted to both plasma action on animal cells and the chemical analysis of plasma-treated solutions, together with the development of dedicated plasma sources for such applications, were between the key reasons why new emerging fields have been recently introduced, such as plasma agriculture, plasma catalysis, plasma nanomaterial synthesis/functionalization, plasma cancer, and plasma for skin treatment and cosmetics [9–15]. In this framework, many scientific works and reviews [1,16] provided insights into the non-equilibrium chemistry created at the plasma–liquid interface and addressed the role of liquid and gas reaction pathways. The most common experimental setups, involving the direct contact of a narrow gas discharge with a batch solution, limit the production of reactive species and, thus, process throughput. While direct contact with a flowing liquid film has been proposed as an alternative to increase the plasma–liquid interface [17,18], some recent works have proposed the use of plasma-droplet interactions with the aim of overcoming the limit of batch process enabling in in-situ and on-demand dispensing [19–21]. The large number of reaction pathways and available plasma parameters, already challenging for batch processes, bring even more complexity to the analysis and investigation of plasma–liquid interaction but open new possibilities for technological applications.

2. Plasma and Aerosols: Opportunities, Economical and Societal Benefits

All these recent advances should, in our opinion and as has already been documented in the literature [19,20,22], result in a strong effort and a specific interest in the development, optimization and applications of plasma in interaction with liquid droplets and aerosols in various fields of technology over the next decade. The topic of this paper is to summarize the opportunities for generating and delivering plasmas into aerosols and sprays, as a specific branch of plasma–liquid science and technology.

The term "plasma-aerosol", where "aerosol" means a dynamic suspension of liquid droplets dispersed in a gas, encompasses a wide range of scenarios that can involve a range of options, from single microscopic droplets up to dense sprays and jets, while atmospheric pressure plasmas may vary from the low temperature and non-equilibrium family of devices to extremely hot plasmas.

The plasma–aerosol configuration offers special beneficial advantages, not only in furthering the development of plasma–liquid applications, but also enabling greater scientific insights into what is an extremely complex problem involving potentially thousands of transient and non-equilibrium chemical reactions. Moreover, the incorporation of large surface-to-volume ratio media into cold plasma creates new opportunities, correlated to highly demanding societal and technological needs, in:

- *Enhancing the transfer of activation energy from the plasma to the liquid*. Batch processes are often limited by the transport of species through the liquid surface. This limit is overcome in the plasma–aerosol configuration thanks to a large surface-to-volume ratio and the production of species in close proximity to the droplet surface.
- *Controlling reactivity in the liquid*. The droplets can act as individual microreactors, enabling a range of conditions that cannot otherwise be achieved in batch processes.
- *In-flight production and on-demand delivery of designed micro/nanomaterials* associated with the generation of clusters and/or liquid evaporation. Aerosol droplets could be employed as micro-carriers, able to deliver particles and molecules in the discharge region opening, for example, for the use of low-volatility liquid compounds.
- *Delivery of short-lived species*. Plasma-aerosol driven by high velocity sprays may represent a new and unique way to generate and deliver significant amounts of short-lived species (sub-second lifetime) together with solvated electrons away from the plasma itself on millisecond timescales.

Dealing with economic benefits, in the synthesis of high value chemicals, drugs and nanomaterials, there is an increasing trend to move away from batch processing, which is expensive, difficult to control, and has an unwelcome environmental impact. Efficient green process research is looking to develop micro reaction technology. Plasma and aerosols offer many further advantages with regard to cross-contamination, throughput, chemical recovery and waste generation. In addition, the ability to generate high value chemicals, locally and on-demand, offers tremendous potential. This includes, for example, pristine or drug-coated nanoparticle generation and delivery to patients for nanomedicine applications (e.g., wound healing and cancer treatment) and size-controlled nanoparticles for volume catalytic processes. Alternatively, the encapsulation of biomolecules, drugs, and nanocarriers in liquid droplets seems to prevent possible plasma-induced damage during the deposition of bioactive, nanostructured and functional coatings, allowing the retention in the deposited coating of important chemical functionalities [23–26].

On the side of energy saving, the valuable work of Locke and Shih [27] on energy yields for H_2O_2 generation by plasma-water processes reported how the highest efficiency is achieved by adopting liquid droplets that sequester H_2O_2 and hinder its decomposition by radicals in the gas phase. One would expect that optimizing the energy transfer from plasma to liquid as an aerosol will also broaden opportunities for on-demand, in-situ plasma activated aerosols, likely to be used in developing countries where electricity availability is still a challenge, together with the specific design of battery operated and solar energy powered devices. Such development would be of key importance and highly beneficial for new agriculture, medicine and decontamination solutions [28]. The use of plasma aerosols will also consist in a water/high value solution-saving technology, matched for applications like indoor cultivation and large surface material deposition, thus keeping plasma as an almost dry technology, preventing from huge effluent volumes to be treated as byproducts. As detailed further in the following sections, plasma and aerosols could also help to prove our understanding of the climate impact and significance of streamer discharges in the atmosphere [29]. Moreover, introducing plasma and aerosols as a new alternative in 3D or ink printing technologies has been very recently discussed [30] and should be an objective for large-scale dissemination of plasma processes, even in today's unexplored applications.

3. Plasma and Aerosols: Challenges

Compared to plasma discharges in and over liquids, biphasic aerosol–plasmas have been far less investigated, and many aspects of their nature are still unknown. The generation of plasma in such finely dispersed biphasic media is very challenging for physical and chemical diagnostics and modeling studies, where very local, transient and fast dynamic behaviors, and long-distance effects should be accounted for. Nowadays, the significant lack of basic understanding concerning the mechanisms governing these plasmas hinders the optimization of their applications. The scientific questions that need to be addressed to improve our knowledge on plasma–aerosol interactions can be ideally sorted into three main categories (see Figure 1):

- Modification of the plasma due to liquid droplets
- Modification of the liquid droplets due to plasma
- Multiphase species transport between plasma and liquid droplets

In the first category all the phenomena governing the plasma discharge propagation and intersection with the droplets in a biphasic environment can be found. Based on this, some pioneering studies [31,32] highlighted how the propagation of streamers can be significantly disturbed by the presence of droplets due to the local modification of the electric field that results from the droplets polarization and charging [33]. This local modulation of the electric field in turn modifies the local rate of ionization.

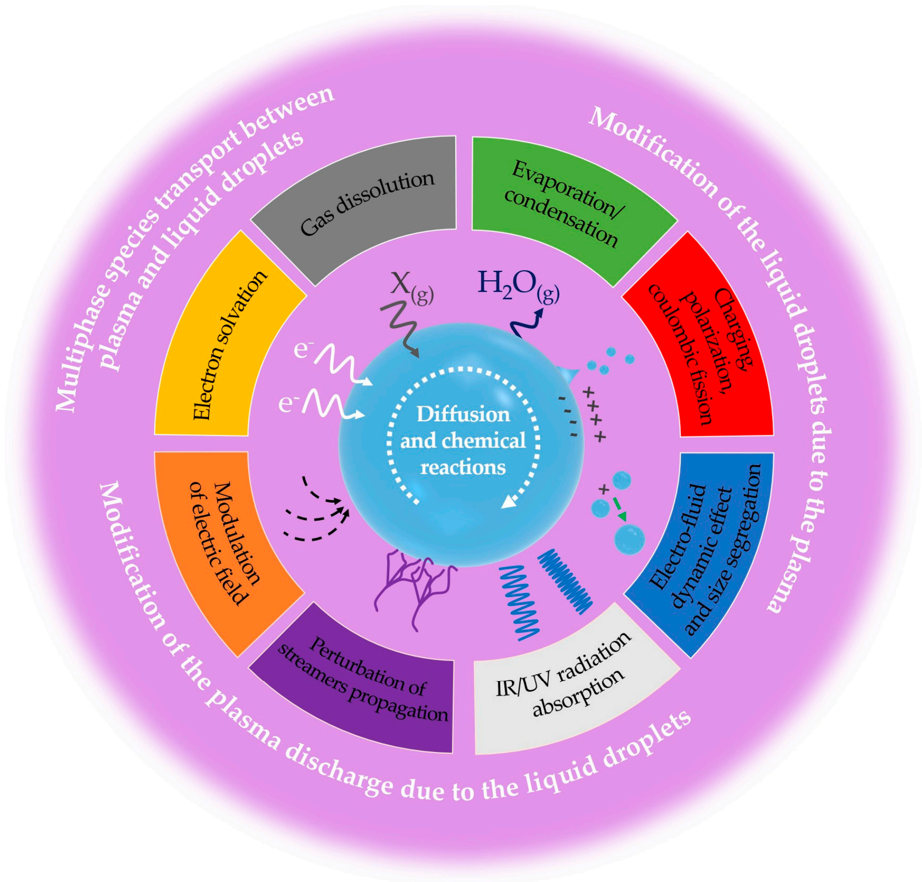

Figure 1. Schematic representation of the interactions between the plasma and an aerosol droplet.

Other possible effects include charge removal from the streamer and shadowing of the photoionization behind the droplets; both mechanisms strongly depend on liquid characteristics (e.g., permittivity) and droplet size. Small droplets (tens of μm or less) of moderate permittivity are expected to be enveloped by the streamer and do not significantly perturb its propagation, while larger drops, with a big enough capacitance, can intercept and reinitiate the streamer or stall its further propagation. Droplets may thus be considered as controlling and tracing the path where the discharge energy is deposited [31]. Other mechanisms that can possibly play a role and that deserve further investigation include: droplets induction of streamer branching; extraction of electrons from the droplet surface owing to photons emitted from the streamer head; alteration of the streamer propagation velocity. Besides, arising from surface evaporation, the interfacial layer in proximity of the droplet surface is suspected to contain a cocktail of positively and negatively charged clusters with a large size range, along with a very high localized vapor density, possibly up to tens of times that possible in a normal plasma before extinction. We can speculate that this may lead to amplified non-equilibrium gas-phase chemistry, the products of which may be sequestered by the droplet boundary. Even if the analysis of transient phenomena (streamer propagation, streamer droplet interaction) at the microscale requires a great deal of effort, a combined simulative and experimental approach to the subject greatly benefits the community in addressing the role of different phenomena in the plasma–droplet interaction. In particular, a better understanding of the effect of liquid (conductivity, droplet dimension, colloidal state) and gas (electron density, electron temperature, electric field) properties will help researchers to optimize the design of plasma processes and sources.

Concerning droplets modifications due to the interaction with plasma, a droplet entering a plasma region is subjected to charging and subsequently experiencing deformation and possibly splitting due to Coulombic fission as a result of exposure to a plasma-related electric field [20]. In addition, the rate of evaporation of such droplets could be enhanced by additional factors other than those encountered in non-reactive gases, such as irradiation by ultra-low energy electrons [34]. In fact, these electrons can have energies much lower than is achievable by any other technique, such as radiolysis. Irradiation, however, may also lead to other processes, such as electron–ion or anion–ion recombination, Auger and UV emission, which have received very little attention to date in this context. Moreover, droplets introduced in atmospheric plasma are also susceptible of being accelerated by the combined effect of the drag forces, ionic wind and electrostatic forces and this could result in various effects including size segregation. While it is generally believed that the weak magnetic field produced by atmospheric pressure plasma discharge [35] cannot be accounted for, for the direct modification of a droplet's properties (Moses effect [36], evaporation and modification of surface energy), the application of an external field can be employed to modify the plasma discharge characteristics and droplet properties [37–40], inducing breakdown [41] or colloid segregation [42]. Precise control over these mechanisms by means of plasma technology would open new perspectives from the point of view of droplet manipulation, from millimetric to nanometric scales, from a single droplet to an aerosol. From this perspective, a deeper investigation of droplet evaporation, taking into account both thermal and physical effects, can strongly influence the use of plasma for on-demand and in-situ synthesis of chemicals and reactive species. Further efforts should be devoted to the fundamental study of droplet charging and electron impaction, with an important focus on the roles of electron density, temperature and droplet size.

Concerning multiphase species transport, in the last ten years, considerable advancements have improved our knowledge on the subject [1]. It is considered that plasma–aerosol droplets act as efficient microreactors where reaction rates, mixing and surface/volume ratio are considerably enhanced, posing new unique challenges in the understanding of transport of a species in this configuration [27,43]. Computational studies [43–46] showed the importance of the synergy between the plasma and the liquid, including evaporation and the solvation of ions, electrons and neutral particles. They also investigated the plasma treated water chemical and transport processes. They highlighted the importance of the water microdroplet size (or thin water film thickness) on the transport processes of plasma reactive species. While extremely challenging, devoting additional efforts to the analysis of convection and reaction pathways inside a droplet microreactor would be valuable, as well as comparing results with scientific works already published for batch processes. The achievement of better knowledge on this issue will result in the possibility to tune the chemical and physical characteristics of an effluent. Future applications will also take advantage of a better understanding of droplet transportation for the in-situ delivery of chemicals.

The understanding of the aforementioned mechanisms is still at an early stage and certainly deserves more attention from the scientific community. The operative ranges for biphasic plasma generation based on aerosol density and granulometry are widely uncharted; the influence of various and numerous process parameters (e.g., pulse repetition frequency, aerosol density) and most of the proposed hypotheses still need to be validated for a wider number of conditions and plasma source architectures. Future studies will have to face the challenges of identifying and validating diagnostic methods able to allow in-situ characterization of this multiphase environment, despite the intrinsic inhomogeneity and transient nature. The measurement of a microdroplet thermal, electrical and chemical characteristics presents a significant challenge that has, to date, seriously limited our understanding of this important system. Recent advances in spectroscopic techniques, from UV to infra-red, and with high spatial and temporal resolution now offer the possibility for accurate study of this system, aided by recent developments in plasma control and in precision microdroplet generation [34,47,48]. Considering, for example, the range of available gases, liquids or colloids and the choice of plasma parameters, droplet sizes and exposure time, this experimental and simulation environment offers very fertile ground for future discoveries.

Nevertheless, the most important challenge in the study of complex multiphase plasmas probably comes from accounting for the strong coupling that links the above mentioned phenomena. For example, a droplet entering a plasma region locally modifies the electric fields, but may in turn be deformed by the electrostatic forces up to the point of splitting into smaller droplets, characterized by a higher rate of evaporation that will modify the liquid vapor content in the gas phase and therefore the plasma discharge. The acknowledgment of this coupling will demand a considerable effort and the adoption a multi-diagnostic approach combining techniques for the characterization of the plasma, the aerosol, the fluid dynamic mixing and the liquid properties together with advanced numerical models able to account for droplet surface modification. The design of new experiments aimed to "decouple" these phenomena, for example considering the interaction between a single drop with a single pulse discharge, will also be essential to further improve our knowledge on these mechanisms.

New plasma source architectures, best coupled with aerosol nebulizers, will need to be developed and customized to ensure a precise control of droplet dimension distribution and residence time. The optimization of existing applications based on the interaction of plasma-droplets (e.g., plasma-assisted coating deposition [23], wastewater treatment and decontamination [49]) will promptly benefit from research effort on biphasic plasmas while numerous new applications (e.g., plasma aerosol catalysis, plasma assisted printing [50]) are expected to rapidly develop and gain interest. With a precise control of the plasma and microdroplet parameters we could engineer a unique non-equilibrium multiphase structure comprised of gas, plasma, liquid and, importantly, a dynamic interfacial layer. This enhanced transport, combined with a fine control of the droplet trajectories and residence time in the plasma region, opens new opportunities in the control of the treatment time and potentially in the use of solvated electrons and short-lived reactive species. With this control, not possible with any other steady-state system, we may gain insight into the transport and mass accommodation of radical species before the onset of Henry's Law [51]. This is of critical importance in the study of atmospheric chemistry, pollution and climate change [29]. With the introduction of solid and polymer materials, or their precursors, into a liquid droplet, the scope for new materials and new diagnostics appears immense.

4. Bridges to Other Communities

Great help in tackling the presented new challenges may come from closely related research fields and communities that have already been addressing effects of interest for biphasic plasmas. As an example, there are multiple studies, some of them over 100 years old, on electrospraying (electrohydrodynamic atomization) of liquids by applying strong electric fields, i.e., high voltages on the nozzle [52,53]. The electrospray community has produced a huge amount of valuable literature on the mechanisms implied in droplets charging, deformation and fissioning in the presence of strong electric fields and eventually in the presence of plasma discharges when operating with liquids with high surface tension (e.g., water) that require the use of high voltages [54]. However, in most of electrospray applications, the occurrence of plasma discharge is undesired, as the discharge typically perturbs the stability and homogeneity of the spray (see Figure 2).

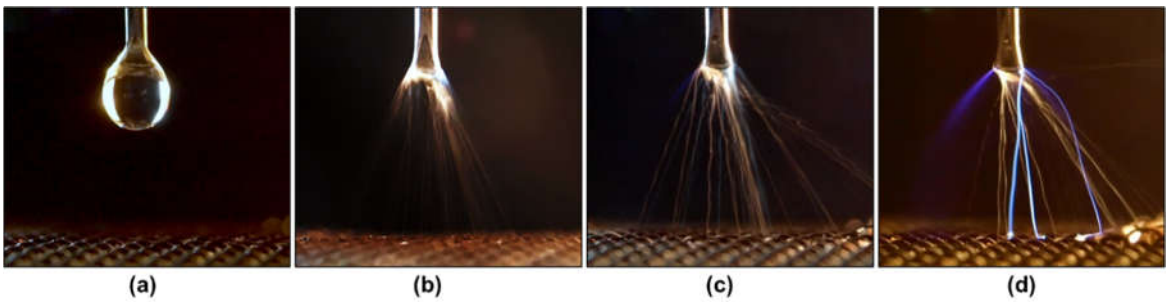

Figure 2. Photographs of the electrospraying of water in an 8-mm gap, water flow rate 0.5 mL min^{-1}: (**a**) droplet without a high voltage, (**b**) electrospray with a high voltage applied, 5.5 kV, (**c**) electrospray combined with streamer corona, 6.5 kV, (**d**) electrospray with transition streamer corona-transient spark, 7.8 kV. Reproduced from Machala et al. [55].

There were a few studies, which investigated the interactions of the sprayed charged aerosol droplets and the discharge, especially with respect to the space charges and ion mobility effects [56–58], or even studies that intentionally employed these complex interactions, e.g., for water treatment or surface decontamination [59–61]. Valuable information, methods, techniques and models can be extracted from these studies or reviews [54,62] and used for a deeper investigations of plasma-aerosol interaction. Other fields that probably deserve attention from the plasma community are fuel injection for combustion engines [62], low pressure dusty plasmas [63], aerodynamic [64], thermal plasma spray [65], space plasmas and once again atmospheric plasma phenomena [29]. Far from being trivial, the scouting of these domains' literature and the instauration of efficient collaborations with the relative researchers will be a key factor to support the advancement of the research in biphasic plasmas. Certainly, the benefit will be mutual and the collaboration will provide new perspectives and tools to the cited domains (e.g., enhanced evaporation and introduction of reactive species in fuel droplets to increase flame control and reduce pollutant production).

Author Contributions: A.S., T.G., M.G., Z.M., P.M., V.C., J.-M.P. and E.R. contributed to the writing, reviewing and editing of the manuscript.

Funding: This research received no external funding.

Conflicts of Interest: The authors declare no conflicts of interest.

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