Borra,J.P.,Ehouarn,P.,&Amouroux,J.(1999). EHD pulverization of charged water droplets for the reduction of particulate plasma by -products. Paper presented at HTTP1998 StPetersburg, published in Progress in Plasma Processing of Materials, Ed. Begell House, New-York, USA, pp.803–808. (http://www.lpgp.u-psud.fr/~jp/HTTP_1998.pdf).

ELECTRO-HYDRODYNAMIC PULVERISATION OF CHARGED WATER DROPLETS FOR THE REDUCTION OF PARTICULATE PLASMA BY-PRODUCTS

BORRA J-P. EHOUARN P., et AMOUROUX J.*

Laboratoire de Physique des Gaz et des Plasmas, CNRS Equipe Décharges Electriques et Environnement, Ecole Supérieure d'Electricité, Plateau du Moulon, 91192 Gif-sur-Yvette Cedex, France

* Laboratoire de Génie des Procédés Plasmas et Traitement de Surface Université Pierre et Marie Curie ENSCP - 11, rue Pierre et Marie Curie, 75231 Paris, France

ABSTRACT

In order to develop a filtration device for sub-micron particles, we need to produce electrically charged water droplets which role is to collect negative sub-micron pollutant particles by electric coagulation. The droplets are produced by Electro-HydroDynamic Pulverisation (EHDP) in the cone-jet-glow mode, in air, at atmospheric pressure. We describe a number of electric and granulometric experiments to define some of the different modes of the water sprays according to the liquid properties (conductivity and flow rate) and according to the voltage, i.e. to the electrical field near the liquid related to electrical discharge phenomena (continuous or impulse). We found that :

- a continuous corona discharge current (glow) allows the EHDP of water in, what we decided to call the "cone-jet-glow" mode, leading to the production of unimodal sprays.

- a strong impulse discharge current (streamer) disturbs the continuous pulverisation in the "cone-jet-glow" mode to, what we decided to call, the "electric-dripping" mode. The disturbance is due to the impulse electric discharges (streamer) in the gas around the liquid mode, leading to the production of polymodal sprays.

1) INTRODUCTION

Plasma devices are known to produce particles in the sub-micron size range either by plasma/electrode interactions or by gas-to-particle conversion (nucleation) of condensable species produced by the plasma activity in the gas phase [BOR 98 & PAR 98]. The aim of our research is to develop a filtration device for sub-micron pollutant particles. To do so, the sub-micron particles will be charged negatively and will be collected by positive water droplets by coulomb interaction. The aim of this work is to produce electrically charged water droplets which will play the role of collectors in the final process.

In this respect, we chose the ElectroHydroDynamic Pulverisation (EHDP) of liquid droplets with higher charge levels than obtainable by any other production and charging process. EHDP requires an electric field to induce surface charges in a droplet. These charges are influenced by the applied electric field, and the droplet shape transforms into a cone. At the cone apex, a liquid jet emerges which breaks up into a number of main droplets with a narrow size distribution, and a number of satellites [TAY 64].

By increasing the nozzle potential through which a liquid is flowing, different modes of jet fragmentation were defined by Cloupeau and Prunet-Foch [CLO 94] :

- the "dripping mode" leads to millimeter diameter droplets,

- the " cone-jet mode " leads to micron diameter droplets,

- the "multiple cone-jet mode" leads to micron diameter droplets,

Some liquids, like water, are difficult to disperse in air. Because of the high surface tension of water, the liquid potential has to be increased which may induce electrical discharges in the gas around the liquid. The establishment of stable water sprays in air is generally prevented by the occurrence of electric breakdown in the gaseous environment surrounding the spray [CLO 94 & JAW 97 & HAY 86].

However, some solutions to stabilize the spray of high surface tension liquids, without any impulse discharge have already been found :

- a reduction of surface tension by surfactants [SMI 86],

- a polarized ring with the same polarity than the nozzle [MES 92],

- an increased dielectric strength of the gas, either by increasing the pressure or by using more insulating gases (SF₆, CO₂ [TAN 95]).

The first electrical regime characterization related to granulometric consequences due to either spray stabilisation by glow (continuous current) [TAN 95] or destabilisation by streamers (impulse current) [BOR 96], have already been investigated.

The aim of the present work is to study the role of the discharges (continuous and impulses), in air at atmospheric pressure, in the establishment of a stable water electrospray.

2) EXPERIMENTAL SET UP AND MEASUREMENTS

2-1) EXPERIMENTAL SET UP

The experimental configuration is shown in Fig.1. It consists of a nozzle (diameter either 1.8 or 0.5 mm) supplied with positive DC voltage, from 0 to 30 kV, and a ground electrode (metallic plate) perpendicular to the nozzle positioned 7.5 cm away. The liquid was fed into the nozzle from a liquid pump. Liquid flow rate is controlled and varied between 0.1 and 200 ml/h.

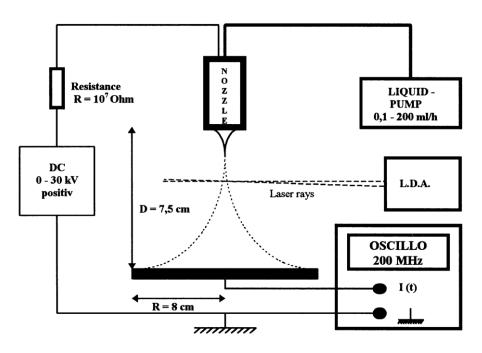


Fig.1 : Schematic of the experimental system

2-2) MEASUREMENTS

Two diagnostic techniques were used to monitor the spraying mode:

granulometric measurements : the diameter of the water droplets was measured by the intensity of the (back-)scattering of interference fringes arising from the crossing of two flat laser beams on particles i.e. by a technique based on Laser Doppler effect. The L.D. system was used to determine the droplets size from 1 to 262 μm. Figure 2 shows a size distribution in cone-jet-glow mode, measured in the center of the spray, 0.5 cm from the tip of the jet, at a conductivity of 5 μS/m and at a liquid flow rate of 200 ml/h.

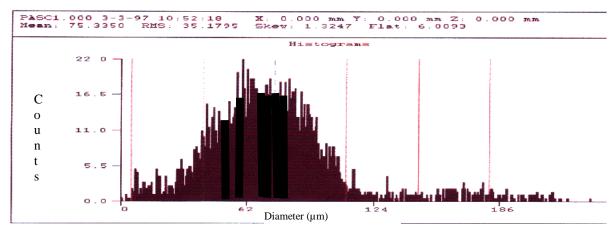


Fig. 2 : size histogram in " cone-jet-glow " mode

• *electric measurements* : The electric current collected by the grounded plate electrode (cf. figure 3) was measured by an oscilloscope (200 MHz), enabling the differentiation of fast electric pulses (200 ns), of millimeter droplets transported within the gap (5-10 ms), and of the stable continuous cone-jet-glow mode current (because of the high number concentration in the gap and because of the fast and constant flux of charged droplets, the current of each droplet is no more distinguishable from the others and leads to a continuous current : continuous current + cone-jet mode current).

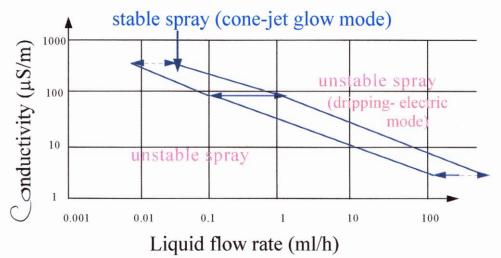


Fig. 3 : currents as function of the time (a) "electric-dripping" ; (b) "cone-jet-glow"

3) RESULTS AND DISCUSSIONS

3-1) relation between conductivity and liquid flow rate for stable ehdp of water

Within the domain defined on figure 4 versus electric conductivity and liquid flow rate for different potentials, the electrospray works in a stable mode, that we decided to call the "cone-jet-glow" mode because this mode produces a unimodal electrospray established with a continuous discharge : "glow" (cf. § 3-3 Granulometric consequences of discharges ; figure 6).

Outside this domain, two types of phenomena were observed: the electrospray works in an unstable mode with an emission of bigger droplets either in dripping mode, or in, what we decided to call, "electric dripping" mode. This electric dripping mode creates a polymodal electrosprays (cf. § 3-3, figure 6) due to the succession of the stable spray (stabilized by a glow leading to a unimodal spray) disturbed by impulse discharges (streamer) leading to bigger droplets (cf. § 3-3). From the electric point of view, the electric dripping mode represents a superposition of three different linked to the production of space charges with very different mobilities (drops of different sizes and gaseous ions). Thus, the electric field is permanently changing, preventing the establishment of the stable cone-jet-glow mode.

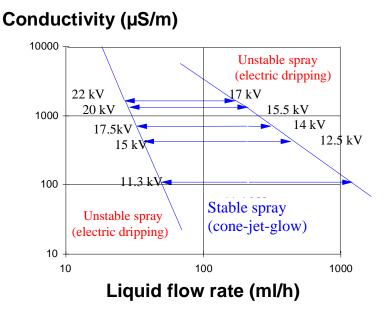


Fig.4 : stability diagram in the electrical conductivity-liquid flow rate plane for different potentials

Figure 4 indicates that when the conductivity increases the liquid flow rate has to be decreased. Actually, when the conductivity and voltage increase, the intensity of the electric field at the liquid surface increase, which implies that the surface tension has to be increased to balance the increased electric forces. In this respect, the hydrostatic pressure in the liquid (e.g. the liquid flow-rate) has to be decreased in order to create a smaller cone.

It has to be underlined that the jet diameter and particle diameter decrease with increased conductivity and reduced flow-rates, have already been mentioned by Ganan-Calvo [GAN 97]. Moreover, from the electric discharge point of view, the reduction of the jet diameter (flow rate reduction) induces a higher field divergence favorable to the glow [HAR 63] and its stabilizing consequences [BOR 98].

3-2) VALIDATION OF ELECTRIC CURRENT MEASUREMENTS

In order to validate the experimental measurements, we have compared our measured currents with those predicted from the theoretical Ganan-Calvo scaling laws. Ganan-Calvo has established scaling laws which give the current and the particle diameter in stable cone-jet mode as function of liquid properties (surface tension, viscosity, conductivity, permitivity, density) and experimental parameters (liquid flow rate, nozzle voltage) [GAN 97].

In order to separate the two continuous current components (glow discharge and droplet currents), a ground ring has been placed around the spray, 1.7 cm from the tip of the nozzle. In the gap, the charged species have very different mobilities: the gaseous ions were collected on the ring (glow current), and the micron size droplets on the ground metallic plate (droplet current).

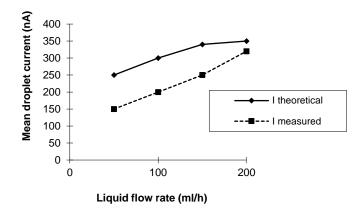


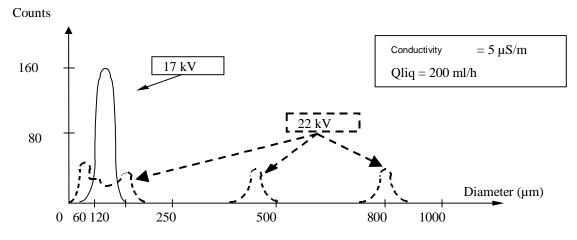
Fig.5 : mean droplet currents collected on the plate as function of liquid flow rate (5 μ S/m)

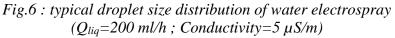
Figure 5 shows the current of the stable mode (experimental and theoretical) versus the flow rate. The concordance of the theoretical and measured currents is only true at 200 ml/h with the 10% of error related to the measurement technique. Actually, only high flow rates leads to a stable cone-jet-glow mode for the used conductivity of 5 μ S/m water. If the flow rate decreases, the experimental current is lower than the theoretical current. The mean experimental droplet current is lower while working in the electric dripping mode obtained for low flow-rates, than while working in the cone-jet-glow mode for high flow-rates (at 200 ml/h). This is due to the succession of electric discharge regime characteristic of the electric dripping mode (e.g. the droplet current close to the theoretical current for the cone-jet glow period) and of the dripping current which is intermittent and much lower than the spray current. So, electrical measurements are proven to be representative.

3-3) GRANULOMETRIC CONSEQUENCES OF DISCHARGES

It has been shown that with a continuous current, the cone-jet-glow mode is stable, whereas, with strong impulse current, the spray is unstable and works in the electric dripping mode. To define the influence of impulse discharges on the granulometry, we performed experiments with water spray (flow rate of 200 ml/h, conductivity of 5 μ S/m). Figure 6 shows a typical size distribution of water with different voltages of 17 kV (---) and 22 kV (---).

The main droplet diameter is in the micrometer range with continuous discharges (glow) in the cone-jet-glow mode of EHDP and about micrometer and millimeter with impulse discharges (streamer) in the electric dripping mod of EHDP.





For the same counting duration, the same conductivity and liquid flow rate, the granulometry confirms that the threshold of impulse discharges implies a granulometric distortion described in figure 6. It corresponds with that change of cone-jet-glow mode producing an unimodal size distribution to electric dripping mode with the characteristic multimodal size distribution.

4) CONCLUSION

Electric and granulometric measurements enable us to differentiate stable continuous mode from unstable pulsative mode of water Electro-HydroDynamique Pulverisation related to electrical discharges phenomena, in air, at atmospheric pressure. The cone-jet-glow stability domains are given versus the liquid conductivity, the flow rate and nozzle voltage.

This study has shown that :

- a continuous corona discharge current (glow) allows the EHDP of water in, what we decided to call the "cone-jet-glow" mode, leading to the production of unimodal sprays.

- a strong impulse discharge current (streamer) disturbs the continuous pulverisation in the "cone-jet-glow" mode to, what we decided to call, the "electric-dripping" mode. The disturbance is due to the impulse electric discharges (streamer) in the gas around the liquid mode, leading to the production of polymodal sprays.

In the future, we plan to test the collection efficiency of negative sub-micron pollutant particles with the different droplets sizes and mobilities, produced by these different modes.

ACKNOWLEDGEMENTS

We gratefully acknowledge the ECODEV program for their financial support of this study. The granulometric measurements were done in the "Génie des Procédés Plasmas et Traitement de Surface" laboratory.

REFERENCES

- [BOR 96] Borra J-P., Hartmann R., Marijnissen J., Scarlett B. (1996), "Destabilisation of sprays in the cone-jet mode by electrical discharges on the jet " *J. Aerosol Sci.*, **Vol.27**, pp.203-204.
- [BOR 98] Borra J-P., Tombette Y. and Ehouarn P. (1998), "Influence of Electric Profile and Polarity on the Mode of EHDA related to Electric Discharge Regimes " *J. Aerosol Sci.*, to be published.
- [CLO 94] Cloupeau M. and Prunet-Foch (1994), "Electrohydrodynamic Spraying Functioning Modes : a Critical Review " *J. Aerosol Sci.* Vol. 25, No. 6, pp. 1021-1036.
- [GAN 97] Ganan-Calvo A.M., Davia J. and Barrero (1997), "Current and droplet size in the Electrospraying of liquids. Scaling Laws " J. Aerosol Sci., Vol.28, No 2, pp249-275.
- [HAR 63] Hartmann G, Goldman A., Buchet G. (1963), "Contribution à l'étude de la décharge pré-disruptive entre pointe et plan ", Conf. Int.sur les Phénomènes d'ionisation dans les Gaz, Paris, **Vol.2**, pp301-303.
- [HAY 86] Hayati I., Bailey A. I., Tadros TH. F. (1986), "Investigations into the Mechanisms of Electrohydrodynamic Spraying of Liquids " J. of Colloid and Interface Science, Vol.117, No1, pp 205-221.
- [JAW 97] Jaworek A., Krupa A. (1997), "Studies of the corona discharge in end spraying "*J. of Electrostatics* **40&41** pp 173-178. [MES 92] Meesters G. (1992), "Mechanisms of droplet formation "*Ph-D thesis of Delft University.* [PAR 98] Parissi L., Odic M., Goldman M., Goldman A., Borra J-P. (1998), "Formation of Particulate by-products by nucleation
- [PAR 98] Parissi L., Odic M., Goldman M., Goldman A., Borra J-P. (1998), "Formation of Particulate by-products by nucleation mechanisms in VOC combustion processes monitored by a dielectric barrier discharge "Proc. Int. Symp. on High Pressure, low Temperature Plasma Chemistry, Cork, Ireland, to be published.
- [SMI 86] Smith D.P.H. (1986), "The EHD atomisation of liquids", IEEE Trans., 1A 28, pp 1432.
- [TAN 95] Tang K. and Gomez A. (1995), "Generation of Monodisperse Water Droplets from Electrosprays in a Corona-Assisted Cone-Jet Mode " *Journal of Colloid and Interface Science*, **175**, 326-332.
- [TAY 64] Taylor G.I. (1964), "Disintegration of water drops in an electric field" *Proceeding of the Royal Society of London , Mathematical and Physical Sciences*, **A280**, 383-397.