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REVIEW PAPER

Review of electrospray observations and theory

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Abstract
While a liquid is dripping out of a capillary tube, there is a possibility to affect the characteristics of the exiting flow in certain ways. One of the ways already used is by introducing an electric field that can be used to change the average droplet diameter and the droplet size distribution. This process is called electrospraying, while the theory behind it is sometimes called electrohydrodynamics (EHD). This phenomenon has been investigated for more than a hundred years both empirically and theoretically. In this paper, a review of the available literature and the empirical and theoretical findings is presented. A new classification of the electrospray modes had to be given to include all the different modes mentioned by different authors. The necessary pieces of the equipment and their different types are also given.

1. INTRODUCTION

The two main ways that are used for changing the properties of a liquid flow exiting a capillary tube are based on introducing either electric forces or new friction forces by adding an immiscible fluid.
William Gilbert is the first known author (in the year 1600) that observed and documented that introduced electric field has an effect on a liquid surface in "The Magnete", where he wrote about the change in a droplet's shape from spherical to conical (Ondimu, Ganesan, Gatari, Marijnissen, & Agostinho, 2017). When applied to the dripping liquid, the electric field causes a change in the average droplet diameter and the droplet distribution, while the droplets become charged (Yeo, Gagnon, & Chang, 2005). Depending on the characteristics of the electric field, the droplets fall down in different ways (i.e. different electrospray modes) and repel each other because of a Coulombic force, which causes a widening of the spray (Pongrác, Kim, Negishi, & Machala 2014). This process is called electrospraying, while the theory behind it is sometimes called electrohydrodynamics (EHD). Investigations in the last century led to many equations which have proved their usability in certain cases.
The other way is called flow focusing and is based on introducing a fluid that flows with a high speed around the capillary. This fluid is usually an inert gas (usually nitrogen) and does not mix with the liquid. Similar effects of these two methods and a similarity between the resulting microfluidic structure prompted Gañán-Calvo and Montanero (2009) to make a successful analogy between them that includes appropriate equations. Another phenomenon that deals with droplets is called coalescence. In the theory regarding this phenomenon, the problem of calculating the droplet distribution is dealt with by using population balance equations and empirically determined parameters within them (Kamp, 2017). Because the same problem exists in electrospraying, similar equations could probably prove their usability in this phenomenon, too.
A process that also uses electric field and that is used today is the electrocoalescence, but it cannot be very helpful in understanding the electrospray because it is not well understood since there are

different hypotheses about its mechanism (Shin, Yiacoumi, &Tsouris, 2004). If fluids are switched so that the fluid that has a low conductivity is sprayed into the fluid that has a high one, the process is called the inverse electrospray and is much less investigated and understood than electrospray, but it has potential applications in many fields (Shin, et al., 2004).

2. TYPES OF CURRENT

In electrospraying, either direct current (DC) or alternating current (AC) can be used for generating electric field. Both types of electric current have their advantages so the choice between them should be governed by the application.

DC electrospraying produces smaller, charged particles, uses higher voltages (electrical potentials), is used more frequently and was the first type to be examined (Yeo, et al., 2005). For applications concerning portable devices for patient treatment, AC electrospraying is the better option because it produces electroneutral particles that are less prone to surface adsorption and potential drug destabilization and because it eliminates the need for charge neutralization and thus helps in the equipment miniaturization, while the most important advantage is the elimination of patient safety issues associated with high voltages used in DC electrospraying (Yeo, et al., 2005). For laboratory or industrial use, the mentioned disadvantages of the DC electrospraying are not so important, so it is used much more often and the following theories were derived for it.

3. ELECTROSPRAY MODES

Depending on the liquid's flow rate and the applied voltage, a liquid flowing out of a capillary tube can be electrosprayed in different ways (i.e. modes). The modes can be classified in different ways. Generally, the modes, in the order of the increasing voltage, are (cf. H.-H. Kim, Kim, & Ogata, 2011):

- dripping mode – two droplets are produced in this mode (the first one is bigger while the second one is smaller and formed from a liquid neck connecting the bigger droplet with the liquid in the tube) while droplet deformation can be noticed after the voltage has reached a certain value (H.-H. Kim, et al., 2011);
- micro-dripping mode – the two mentioned drops are present in this mode, too, but the difference is that the bigger drop is deformed into a cone while still being connected to the liquid inside the tube and while still growing before the detachment and that it emits even smaller droplets than the droplet that forms from the neck, both while it is still being connected and while it is falling down (cf. H.-H. Kim, et al., 2011);
- spindle mode – as the voltage increases, a long liquid filament, that is straight for lower voltages and that bends and oscillates for higher ones, forms and emits drops before detaching and breaking up into many small droplets (cf. H.-H. Kim, et al., 2011);
- rotating jet – the mentioned long filament stops detaching and starts rotating apparently always clockwise (H.-H. Kim, et al., 2011), what should be connected to the Ampere's right-hand grip rule, if true for every system;
- stable jet – in this mode, the long filament stops both rotating and oscillating (i.e. it becomes stable), but some movement can still be seen near its tip, while the droplet size distribution remains bimodal, which still stems from the first mentioned mode (H.-H. Kim, et al., 2011);
- unstable jet – irregularities start in some way (Cloupeau, 1994), either the filament has an irregular shape and size and it moves unpredictably while emitting droplets of a random diameter and hence a wide size distribution (H.-H. Kim, et al., 2011) or a ramified jet (Hartman, Brunner, Camelot, Marijnissen, & Scarlett, 2000) or multiple jets (two or more) are formed (Jaworek and Krupa, 1999).

For low flow rates, the electric force is much greater than the inertia, so it creates a big decrease in the jet diameter (in the modes with a noticeable filament) and hence a sharp meniscus (or the Taylor cone, i.e. the cone with the defined angle) can be seen at the capillary tube exit, while it is barely visible for high flow rates because of the small reduction of the jet diameter (I. Park, Hong, Kim, & Kim, 2017).

Based on this, the jet modes can be in one of the three regimes depending on the used flow rate (cf. I. Park, et al., 2017):

- cone-jet – the jet that forms for low flow rates that has a noticeable meniscus (I. Park, et al. 2017);
- micro-simple jet – a transitional regime (I. Park, et al. 2017);
- simple jet – the wider jet that has a barely noticeable meniscus that forms for high flow rates (I. Park, et al. 2017).

Therefore, the most important mode, the stable jet mode, can be (cf. I. Park, et al., 2017):

- stable cone-jet – the stable jet that has a droplet size distribution closer to monodispersity than any other mentioned mode or regime (I. Park, et al. 2017);
- stable micro-simple jet – the transitional regime that does not produce monodisperse droplets (I. Park, et al. 2017);
- stable simple jet.

According to Morad, Rajabi, Razavi, and Pejman Sereshkeh (2016), the stable jet mode can be maintained in a greater voltage range for lower flow rates than for the higher ones because the stability of a stable jet decreases as the flow rate increases. Ku and Kim (2002) found that the standard deviation of the droplet diameter for a highly viscous liquid has a minimal value (i.e. the droplet size distribution is the narrowest) for a certain flow rate, which is somewhat greater than the minimal flow rate needed for the stable cone-jet mode. Ku and Kim (2002) also indicated that other authors had not found such behavior for liquids with lower viscosities, for which they had reported that the standard deviation had decreased or stayed almost constant when the flow rate had been increased. It should be noted that the reported changes in the standard deviation could be connected to the transition from the cone-jet mode into the micro-simple or simple jet mode, which was not mentioned but that could had occurred in the first case and that could had not yet been reached in the other cases. Even though the standard deviation has an optimal value of the flow rate in certain cases, a similar phenomenon was not found for the mean droplet diameter, which is the smallest for the mentioned minimal flow rate (Ku & Kim, 2002).

As the voltage increases, a usually undesired phenomenon is going to occur at one point, namely a corona discharge, which causes irregularities in the electrospray and which can prevent the formation of a stable jet if it occurs before it (Cloupeau, 1994). This can happen for liquids that have a high surface tension, as opposed to low surface tension liquids that are usually able to form a stable or unstable jet before the corona discharge occurs happens (Cloupeau, 1994). At first, corona discharges do not affect the droplet size distribution very much and mild corona discharges can even improve the stability of a jet, but after the critical corona intensity has been reached, the jet becomes unstable (Ku & Kim, 2002). Another undesired phenomenon that can happen as the voltage increases is a beginning of needle oscillations when needles that have a small diameter are used because of their lighter weight (Bugarski, Li, et al., 1994). In Bugarski, Li, et al. (1994) it had bad effects on the droplet size distribution because it became bimodal, but one of those two fractions was the smallest when compared to the droplets emitted in other modes of operation that they were able to achieve. It was reported by some authors that a stable cone-jet cannot be formed when pure water is used if it is surrounded by air, so CO_2 or SF_6 sheaths were used, but other authors did not have this problem (I. Park, et al., 2017). Corona discharges were eliminated because those gases have a higher electrical breakdown threshold than the substituted surrounding air (López-Herrera, Barrero, Boucard, Loscertales, &Márquez, 2004). The reported problem might be connected to the corona discharge or to the fact that there exists a minimum flow rate that must be reached for a stable jet to form, which was confirmed experimentally and predicted theoretically (I. Park, et al., 2017).

In experiments, the determination of the mode in which a liquid is being electrosprayed is usually done just by an observation, so the problem is that this can’t be done in industrial or many other applications, so Verdoold, Agostinho, Yurteri, and Marijnissen (2014) measured the electric current flowing through the system and were able to make a relatively good distinction between the electrospray modes based on certain values.

After analyzing the current measurements made by Verdoold, et al. (2014), the differences between the modes can be described:

- dripping – the current slightly oscillates around the zero value;
- micro-dripping – two joined big peaks can be seen when the two mentioned droplets fall down after what the current returns to zero;
- spindle mode – the current oscillates near the zero, but when the filament falls down an even larger peak appears;
- stable jet – the current slightly oscillates around a big value, so now the current does not come close to the zero.

The currents measured in Verdoold, et al. (2014) were ranging from few tenths to few hundreds of nanoamperes, but only one capillary tube was used.

4. THEORIES

When a liquid is dripping downwards through a capillary tube, a pendant droplet detaches from the bulk of the liquid when the gravitational force becomes equal to the surface tension force (Poncelet, Babak, Neufeld, Goosen, &Bugarski,

1999) if the system is stationary, if it is surrounded by stationary air and if there is no electric field. Other forces exist too, but their influence is normally very small so they can be neglected. So, the equation from which the pendant droplet diameter can be calculated is (Poncelet, Babak, et al., 1999):

$$m_d g = \pi d_d \gamma \tag{1}$$

where m is the mass, g is the gravitational acceleration, d is the diameter, γ is the surface tension and the subscript d denotes the droplet.

In the electrospraying process, the electrostatic force, the fact that the flow rate is too big for letting the droplet reach the equilibrium and the diffusion (adsorption) of the ions caused by the electric field need to be accounted for (Poncelet, Babak, et al., 1999). Viscous shear stress could be accounted for too, but it can be neglected for polymer solutions that have usual viscosities when usual flow rates are used (Poncelet, Babak, et al., 1999). Magnetic force can also be neglected (Gañán-Calvo & Montanero, 2009). Fernandez de la Mora, et al. (2000) discussed the role of electrochemistry in this process and apparently concluded that electrochemical reactions are often present and important when analyzing the atomization process from a chemical point of view, but rarely much affect ions and can be overlooked when analyzing just from a physical point of view, even though they have an effect on the fluid dynamics. According to López-Herrera, et al. (2004), there exists a maximal normal electric field in the cone-jet mode, which is approximately equal to (as cited in López-Herrera, et al., 2004; Pongrác, et al., 2014):

$$E_{n,cj} \sim \sqrt{\frac{\gamma}{\varepsilon_0 r_j}} \tag{2}$$

where ε0 is the vacuum permittivity and r_j is the jet radius. So if the value of the normal electric field is greater than the value needed for the electrical discharge, which regularly happens when the surface tension is high, the cone-jet mode can't be reached (Pongrác, et al., 2014). Because the r_j increases as the flow rate increases and as the electric conductivity decreases, they could be manipulated when a corona discharge is encountered (López-Herrera, et al., 2004). The problem with high conductivity liquids is that they act almost as conductors and hence corona discharges happen at lower applied voltages (Pongrác, et al., 2014).

The tangential component of the electric field in the cone-jet mode is approximately (Pongrác, et al., 2014):

$$E_{t,cj} \sim \frac{1}{\pi} \frac{I}{K r_j^2} \tag{3}$$

where I is the emitted electric current and K is the liquid's electrical conductivity.

The electrical charge of a droplet in the equilibrium state depends just on the droplet capacitance and on the applied voltage, while its value will be reduced in a non-equilibrium state (Poncelet, Babak, et al., 1999). The maximal value of the electrical charge that a droplet can withstand, before breaking up, is called the Rayleigh limit (Ondimu, et al., 2017). The Rayleigh limit can be calculated and is equal to (Ondimu, et al., 2017):

$$q_R = \pi \sqrt{8 \varepsilon_0 \gamma d_d^3} \tag{4}$$

Fernandez de la Mora (2000) showed that some large and not very compact ions, such as those made from proteins or a linear polyethylene glycol, can have greater charge than the Rayleigh charge calculated by assuming that they are spherical, but the reason for such values could be in the fact that he stated that the droplets become cylindrical or similarly shaped (i.e. not spherical). Poncelet, Babak, et al. (1999) started from the Lippman's theory, which states that the liquid surface tension changes in the electric field (Poncelet, Babak, et al., 1999):

$$\mathrm{d}\gamma = -\sigma \mathrm{d}U \tag{5}$$

where σ is the effective surface charge density and U is the voltage, and derived the equations for the surface tension reduction and then for the droplet diameter that can be applied from zero voltage to the voltage needed for starting a filament formation (i.e. for entering the spindle mode) and the corresponding electrical potential for polyelectrolyte solutions (Poncelet, Babak, et al., 1999):

$$U_{cr} = \sqrt{\frac{l \cdot \gamma_0}{k \cdot \varepsilon_0}} \tag{6}$$

where $l=d_0 \wedge k=0.28$ when one needle without a plate is used and $l=h \wedge k=3$ when a multi-needle system is used, d_0 is the needle diameter, h is the distance between a plate and a receiving solution, γ_0 is the unaltered surface tension and U_{cr} is the voltage at which a filament formation starts. Poncelet, Babak, et al. (1999) also observed a big reduction in the droplet diameter when the spindle mode is entered, but it seems that they were not able to enter the stable jet mode, which might be connected to a corona discharge happening before it when high surface tension liquids are used, as mentioned before. A big difference can be seen when theories derived from different authors are compared – while the previously mentioned theory regarded the surface tension as a variable, theories that will be mentioned next use it as a constant. Even though the approaches are fundamentally different, both approaches led to equations that proved their usability in the investigated cases. The complexity of the electrospray process and the fact that the best mode of operation is the stable jet mode led many authors to derive theories mainly for the stable cone-jet, while some were derived for the stable simple jet, too.Gañán-Calvo (1997) (see also Gañán-Calvo, 2000) gave an analytical solution for the stable cone-jet by making a model for an almost perfectly or perfectly conducting liquid that consists of a Taylor cone directly under the needle and a thin and infinite jet that starts from its end. Starting from that model, Gañán-Calvo (1997) derived equations for electric current and for the droplet diameter that was correlated because it can't be obtained from an infinite jet, which are equal to (Gañán-Calvo, 1997; Gañán-Calvo, 2000):

$$I = 4.25\sqrt{2}\left(QK\gamma/\ln\frac{\rho KQ}{\gamma\varepsilon_0}\right)^{1/2} \tag{7}$$

$$d_d = 3.78\pi^{-2/3}Q^{1/2}\left(\frac{\rho\varepsilon_0}{\gamma K}\right)^{1/6} f_b \tag{8}$$

where ρ is the density, Q is the flow rate and f_b is the nondimensional jet radius at the break-up point that can be estimated as being equal to 0.6, while the Rayleigh most probable jet to droplet diameter ratio, which is equal to 1.89, was incorporated in 2·1.89=3.78 in the equation for the d_d (Gañán-Calvo, 1997). Hartman, Brunner, Camelot, Marijnissen, and Scarlett (1999) started from the same equation as Gañán-Calvo (1997), also assumed that a liquid is a perfect conductor, and derived an equation for the shape of the stable cone-jet that they later used in Hartman, et al. (2000) for deriving an equation for the droplet diameter. Hartman, et al. (2000) explained that a jet breaks up (i.e. emits droplets) due to axisymmetric (varicose) or lateral (azimuthal or kink) instabilities, while the most important instability is the fastest growing one because the jet breaks up through instabilities that have a wavelength close to the value of its wavelength (called the dominant wavelength). This fact was used by Hartman, et al. (2000) for calculating the jet radius at the break-up position, which they assumed is approximately equal to the droplet diameter that then can be calculated (Hartman, et al., 2000):

$$d_d = \left(\frac{\rho\varepsilon_0 Q^3}{\gamma K}\right)^{1/6} \tag{9}$$

Hartman, et al. (2000) pointed out that this model needs a starting position of the jet and a starting amplitude of the perturbation to be assumed, which influence the final result, and that a problem with this approach is that it cannot distinguish between primary (main, big) droplets and secondary (small) and satellite (even smaller) droplets, so the calculated droplet volume is equal to the sum of volumes of one primary droplet and its two other types of emitted droplets. The last problem is caused by the fact that no droplet size distribution is incorporated in the model, so this problem exists in every similar model. An important fact that was observed by Hartman, et al. (2000) is that the droplet diameter scales as $d_d \propto Q^{0.33}$ when the filament is whipping (i.e. when the unstable jet mode is reached), so it becomes evident that these equations become unusable in other modes. The droplet diameter calculated from the equation derived by Gañán-Calvo (1997) when a value of 0.6 is used for f_b is just 1.06 times greater than the one calculated from the equation derived by Hartman, et al. (2000). Gañán-Calvo, Dávila, and Barrero (1997) started from the scaling based on the experimental data presented by other authors for a liquid that has a low viscosity and a low conductivity and obtained the equation (9) and the equation that is different from the equation (7) (Gañán-Calvo, et al., 1997):

$$I \sim \left(\frac{\varepsilon_0 QK\gamma^3}{\rho}\right)^{1/4} \tag{10}$$

For a liquid that has a high viscosity and a high conductivity, Gañán-Calvo, et al. (1997) started from a one-dimensional momentum equation and showed that the final equations are different because they obtained (Gañán-Calvo, et al., 1997):

$$I \sim (\varepsilon_r - 1)^{-1/4}(QK\gamma)^{1/2} \tag{11}$$

$$d_d \sim (\varepsilon_r - 1)^{1/6}\left(\frac{\varepsilon_0 Q}{K}\right)^{1/3} \tag{12}$$

where ε_r is the relative permittivity. So if a liquid has a low viscosity and a low conductivity $d_d \propto Q^{1/2} \wedge I \propto Q^{1/4}$ and if a liquid has a high viscosity and a high conductivity $d_d \propto Q^{1/3} \wedge I \propto Q^{1/2}$ (Gañán-Calvo, et al., 1997). Y. Zhao and Yao (2017) experimentally obtained the scaling for the current that corresponds to the former case for organic liquids and to the latter case for inorganic liquids. Rosell-Llompart and Fernández de la Mora (1994) said that it seems that the viscosity does not affect the jet diameter but that it affects the droplet diameter, while its effect can be connected to the viscous parameter, which was derived earlier by a dimensional analysis (as cited in Rosell-Llompart and Fernández de la Mora, 1994):

$$\Pi_\mu = \frac{1}{\mu}\left(\frac{\rho\varepsilon_r\varepsilon_0\gamma^2}{K}\right)^{1/3} \quad (13)$$

where μ is the viscosity. This parameter (i.e. Π_μ) is important when it is less than 0.15 according to Rosell-Llompart and Fernández de la Mora (1994). Other authors use a greater value and state that the viscosity does not affect the droplet diameter when $\Pi_\mu \gg 1$, while an increase in the viscosity causes an increase in the droplet diameter for lower values of the Π_μ (Lai, et al., 2017). Ku and Kim (2002) investigated electrospraying of highly conducting and highly viscous liquids and showed that representative analytical and empirical equations predicted droplet diameters that were two to six times smaller than the measured ones probably because of the combination of the high viscosity with the high conductivity. Ku and Kim (2002) did not investigate the applicability of the equation (12), but, the equation (12) should predict droplet diameters that are around two times greater than those obtained by the other equations, while it is almost identical to the experimental scaling that they obtained. A study published by Pongrác, et al. (2014) investigated in which way the water conductivity influences the electrospraying process by deliberately changing it, and showed that the lower conductivity liquid had prolonged and pointy filaments, while the higher conductivity liquid had shorter and broader ones. This fact contradicts many published theories concerning the stable cone-jet mode because as the conductivity increases, the droplet diameter and the jet radius should decrease, but that was not the case (Pongrác, et al., 2014). Gañán-Calvo, et al. (1997) also showed that there exists a minimum flow rate that must be reached for entering the stable cone-jet mode, gave a theoretical explanation for it and stated that its magnitude and the magnitude of the corresponding droplet diameter are of the order of (Gañán-Calvo, et al., 1997):

$$Q_{min} \sim (\varepsilon_r - 1)^{1/2}\frac{\gamma\varepsilon_0}{\rho K} \quad (14)$$

$$d_{min} \sim \left[(\varepsilon_r - 1)\frac{\gamma\varepsilon_0^2}{\rho K^2}\right]^{1/3} \quad (15)$$

Chen and Pui (1997) investigated an earlier version of Q_{min} published in Gañán-Calvo (1994) that had just εr1/2 instead of (ε_r-1)1/2 and stated that the calculated value was six times higher than the measured value, but that could be expected because it was stated that it calculates the order of the value. Equations derived by other authors, who had used a dimensional analysis, were also investigated by Chen and Pui (1997) and showed similar results. Another important process that could be analyzed when equations regarding electrospraying are investigated is electrospinning. Electrospinning is a process used for producing fibers and textiles that is very similar to electrospraying because the same equipment is used in both cases, so it can be said that it is an electrospray variant (Deitzel, Kleinmeyer, Harris, & Beck Tan, 2001). In the electrospinning theory, different equations are given for the jet diameter near the nozzle, far from the nozzle and at the collector (Cramariuc, et al., 2013). There is a possibility that these equations (especially the equation for the jet diameter near the nozzle) could also be used in electrospraying, but the problem with this approach is that electrospinning is adapted for producing fibers (i.e. not spheres) and that it uses solutions that are very different from fluids typically used in electrospraying, so the applicability of the mentioned equations is questionable. Eggers (2005) explained that dropping of a liquid is a very complex phenomenon (even without the electric field), so the Navier-Stokes equations in a changing domain must be solved. Eggers (2005) also stated that the determining factors of the drop size distribution are still largely unknown, while the fact that many primary droplets are followed by secondary smaller droplets does not help, but he was able to come to the solution for the shape of a liquid jet when there is no electric field. The mentioned complexity of the electrospray process recently led some authors to start using computational fluid dynamics for describing these systems because the number of the needed assumptions could be reduced and because the system's geometry could be better described. It can be expected that future theoretical researches will focus primarily in this direction. Notz and Basaran (1999) also assumed that a liquid is a perfect conductor and solved the Laplace's equations for the velocity potential and for the electric potential, and the augmented surface Bernoulli equation for the drop shape, while the initial drop shape was either a sphere section or was calculated from the Young-Laplace equation. Notz and Basaran (1999) used both cylindrical and spherical coordinates, divided space into eight subdomains and used Galerkin finite element method for space discretization and an adaptive finite difference method for time discretization, which were difficult because they had to incorporate a remeshing that moves the horizontal plane and that adds new elements. The algorithm presented by Notz and Basaran (1999) was able to predict different electrospray modes and the resulting droplet diameters, but they stated that a steady jetting from a Taylor cone can't be predicted because of the perfectly conducting liquid assumption, so the Navier-Stokes equations should be used instead of the Laplace's equations for the velocity potential. Ondimu, et al. (2017) first calculated the electric field in the system without a liquid and then used it for calculating the electric force that, together with other forces, acts on the droplets and investigated what could be the cause of the spray dispersion. Ondimu, et al. (2017) pointed out that other authors had attributed the spray dispersion to the droplets' initial displacement or to their random initial velocity vector, but these approaches could not provide good results. Instead of that, Ondimu, et al. (2017) moved the droplet's center of the charge from its center of the mass, because deformations can be seen on every droplet, and confirmed that this was the cause of the spray dispersion by obtaining a dispersion that was in a very good agreement with the experimental observations. The model made by Ondimu, et al. (2017) can be used for calculating the spray shape and the droplet trajectories, but their approach is somewhat problematic because it needs many variables that must be determined experimentally (Ondimu, et al., 2017): a droplet diameter, a droplet diameter distribution, an initial droplet velocity, a jet break-up length, a time interval between two droplets, an interval of the droplet charge as a percentage of their Rayleigh limit, a maximal vertical and a maximal horizontal displacement of the centers and a deformation frequency.

5. VARIABLE PHYSICAL PARAMETERS

Since this process is concerned with small scales and fast moving separate liquid particles, some precautions are needed regarding physical parameters usually considered constant.

Surface tension is one of those parameters. Even when its possible dependence on the electric potential (which exists according to some theories and which does not exist according to other theories) is overlooked, it can still change its value. If there is enough time for the system to reach its equilibrium state, the surface tension value will be constant, but if the time tends toward the zero (as it does in electrospraying), its value will still be constant just for a pure liquid (Rulison, n.d.). If the time tends toward the zero and if a liquid is a surfactant solution, the surface tension value will be equal to the solvent's surface tension value (Rulison, n.d.). The reason for the change when surfactant solutions are used is the fact that the surfactant molecules won't have enough time to diffuse from the center of a drop to its outer edges (Poncelet, Neufeld, Goosen, Bugarski, &Babak, 1999). The presence of the electrical field accelerates the movement of the ions found in the solutions so times that are closer to the zero are needed for the surface tension to reach the value equal to the solvent's surface tension (Poncelet, Babak, et al., 1999). If the measured values are plotted against the time, the line will have an S-shape between the two mentioned values so non-equilibrium methods, such as pendant drop or bubble pressure method, should be used for the measurements (Rulison, n.d.). It was proposed that the liquid's electrical conductivity changes because of the existence of charge rarefaction fronts, but it was disputed in Gañán-Calvo and Montanero (2009) both theoretically and experimentally by providing a good agreement of the opposing theory with the experiments. A phenomenon that can cause problems in the experiments if overlooked, but that can be used positively, is hysteresis. Hysteresis can be encountered in fluid systems even in old long household faucets. When a flow is started and increased very slowly, at one point, the liquid stream flowing out of the faucet will become narrower with almost every drop going directly downwards. After that point has been reached, if the flow is reduced for a certain value, the stream will still be narrow and it will still look uniform. López-Herrera, et al. (2004) saw that when the flow rate of an already stable cone-jet is reduced, while the voltage is kept constant, the resulting emitted current is higher than the emitted current of a stable cone-jet formed usually (without decreasing neither the flow nor the voltage), which is important because they associated the increase in the emitted current with the increase in the cone length. López-Herrera, et al. (2004) also investigated what would happen if the flow rate is kept constant, while the voltage is reduced from higher values to the ones needed for the formation of a stable cone-jet, and saw the opposite effect – the emitted current was lower and hence the cone was shorter, when compared to the usually obtained stable cone-jet. López-Herrera, et al. (2004) also noted that hysteresis phenomena did not exist when the applied voltage was too high or when water that has a conductivity lower than 0.9 S/m was used. The existence of the hysteresis phenomena might be connected to the fact that the ratios of the normal and the tangential electric forces are different in the mentioned cases (Morad, et al., 2016).

6. EXPERIMENTAL SET-UP

All the experimental set-ups that were used in the referenced studies had similar pieces of the equipment, but there were some differences caused either by the convenience or by the needs of the study.

The force needed for the dripping of a liquid is usually created by a syringe pump, but a peristaltic pump (Ondimu, et al., 2017) and no pump (H.-H. Kim, et al., 2011) were also used. Generally, when a pump is used, the flow should be constant enough not to cause any problems, but when a pump is not used, some care could be taken to prevent changes in the flow rate as the liquid level decreases. The next piece of the equipment that can be customized is the needle, or the nozzle, that is usually metallic, but a ceramic one (I. Park, et al., 2017) or a silica one (López-Herrera, et al., 2004) can be used, too. The imperfections in metallic needles (nozzles) can be the cause of the electrical discharges, so a nonmetallic needle (nozzle) has an advantage when there is a need for controlling the electric field (López-Herrera, et al., 2004). Maybe the most important characteristic of a needle (nozzle) is its diameter because it has a very big effect on the diameter of the resulting droplets (Poncelet, et al., 1994). Usually, only one needle is used for electrospraying and it was usually used for checking the mentioned theories. Other systems that were used in Poncelet, et al. (1994) were a system with one needle and a flat metallic plate around it and a multi-needle system, which are good for a process scale-up because of the similarity of their electric field with the electric field of the systems that should be used for industrial applications. As explained by Poncelet, et al. (1994), for the same applied voltage, the surface charge is the highest for the case when just one needle is used, so much higher potential is needed in other cases for the same effects. H. Park, Kim and Kim (2004) showed that when a set-up that has a metallic plate is compared to the set-up without it, although higher voltages are needed for reaching the stable cone-jet mode, the current is almost the same, the jet is stable in a wider voltage range and it is expected that the produced droplets have almost the same diameter, while the effects of the metallic plate increase as the distance of the plate from the tube exit decreases. A slight reduction of the average droplet diameter that can be noticed H. Park, et al. (2004), when the set-up with a metallic plate is compared to the set-up with a nonmetallic plate, could be caused by a slight increase of the measured current that could be associated with a possible increase in the jet length (as mentioned in the paragraph about hysteresis) and an increase in the stability. Morad, et al. (2016) investigated whether putting a nonconducting hemispherical cap 0.2 mm above a capillary tube exit would change the stability of the cone-jet and, according to them, the jet was more stable and larger flow rates could be used for a needle of the same diameter because the liquid would rise up the cap and hence the jet diameter would increase. The problem with the study of Morad, et al. (2016) that the hemispherical cap case was not compared with the nozzle that would have the same flow rate or produce a jet of the same diameter, so some questions remain unanswered. The stability of the cone-jet was increased by some authors by using a ballpoint pen, a carbon fiber or externally wetted emitters because of a modification of the electric field . (Morad, et al., 2016). As reported by H. Park, et al. (2004), when a multi-nozzle system is used, the cone-jets at the system's edges can be curved to the outside, so some authors had put additional capillaries at the outer edges that decrease this distortion. Below the needle(s), usually a metallic plate is placed, but a metallic dish (H.-H. Kim, et al., 2011), a metallic ring (Verdoold, et al., 2014), a metallic tube (Yeo, et al., 2005), a liquid solution (Poncelet, et al., 1994), or a metallic ring followed by a liquid solution (Lai, et al., 2017) were also used. A plate is used because of its simplicity and because the created electric field has the simplest shape, but it has problems with the disposal of the liquid, so that needs to be addressed by placing a proper drainage, as Verdoold, et al. (2014), or by using a porous plate, as Cloupeau (1994).). A wide dish is an easier option for liquid disposal, but some deformation of the electric field (because of the edges of the dish) and some changes in the field (because of the increasing layer of the liquid) exist. A ring can be used when there is no need to collect the liquid in one place, for example in applications concerning the mass spectrometry, while a tube is similar to the ring, but the problem of disposing the liquid is eliminated. A liquid solution is usually used when the electrospray is used for producing microbeads, as in Poncelet, et al. (1994). For producing the electric field, a voltage is maintained between a needle or a plate connected to the needle(s) and an object located below by connecting a power supply to them. A positive potential should be at the needle or the plate, as in Poncelet, et al. (1994). A negatively charged needle can also be used and it was investigated in Poncelet, Neufeld, et al. (1999) for checking the effect of the reversal of the ion migration in the solution before the droplet detaches, where it did reduce the droplet size, but proved to be worse than a positively charged needle because the negative ions were much bigger and hence slower. Another variable that can be used for manipulating the electric field is the distance between the needle(s) and the piece of the equipment below it because as the distance decreases, the current increases (Y. Zhao & Yao, 2017). This variable had the biggest influence on the droplet diameter in Bugarski, Amsden, Goosen, Neufeld, and Poncelet (1994) when a system that had a plate (called a parallel plate set-up) was used. For measuring the emitted current in the system, a nanoampere meter should be used for systems with one needle, as in H. Park, et al. (2004). The current in multi-needle systems might be too high for so sensitive devices so an ampere meter that can measure higher currents should be used.

A need for obtaining droplets that do not possess a positive charge exists in cases when (as cited by Cloupeau, 1994): a reduced electrical mobility is preferred, evaporation is used for producing smaller droplets, or free aerosols are desired. For neutralization purposes, negative ions can be created by (Cloupeau, 1994): a negative corona discharge, a negative electrospraying of volatile liquids, a thermoelectronic emission or flames; so some additional pieces of the equipment are needed that should produce negative ions near the electrospray (Cloupeau, 1994). Full and perfect neutralization shouldn't be expected even if the absolute values of the positive and the negative currents are equal because larger droplets become neutralized before smaller ones, but a free aerosol that follows the ambient air can be easily obtained by reducing the positive charge in this way (Cloupeau, 1994). The droplet diameter is measured in many ways, primarily depending on the used liquid and on the electrospray application. One possibility is to photograph the falling droplets and then measure their diameters, but the problem is that they are falling quickly in a very short time frame, so complex solutions are used, as in Ondimu, et al. (2017) where a high-speed camera, a microscopic lens and a backlight illumination were used for generating a huge number of images which were processed by a computer program. If measurements are done in this way, some small image distortions (that can influence the results because of the small droplets) are created because a perfect focusing is not possible, as pointed out Hartman, et al. (2000). If electrospraying is used for producing microbeads, their size is the most important parameter so a microscope can be used to measure the resulting microbeads, as was done by Y. Zhao and Yao (2017) who used a scanning electron microscope and by Lai, et al. (2017) who used an inverted microscope. It should be noted that the microbead size differs from the droplet size because of the contraction or swelling during their formation (Poncelet, Babak, et al., 1999). The diameter of alginate microbeads is usually around 50% of the droplet diameter, while microcapsules made from nylon are usually 1.3 times larger than the droplets (Poncelet, Poncelet De Smet, Beaulieu, & Neufeld, 1993). For low volatility liquids an aerodynamic size spectrometer can also be used (Ku & Kim, 2002). As it can be seen in Y. Zhao and Yao (2017), where measurements of the electric field in three dimensions are given, the effect of the flowing air on the electric field can be important because of a displacement of the liquid droplets. Other factors that can influence the resulting droplets and that should be controlled are the room temperature, the room humidity, and their changes. By choosing the most suitable experimental set-up, the droplet diameter can be adjusted because as the spray current increases or as the flow rate decreases or as the polymer solution conductivity increases, the droplet diameter decreases (Y. Zhao & Yao, 2017).

7. APPLICATIONS

Some of the applications of the electrospraying are in (H.-H. Kim, et al., 2011): inkjet printing, mass spectrometry, surface coating, paint spraying, fuel atomization and agriculture. One of the most important applications of the electrospraying is for the mass spectrometry, since the mass spectrometry that uses the electrospray ionization is one of the most widely used techniques in the present (bio)chemistry (Fernandez de la Mora, et al., 2000). Production of polymer microbeads by electrospraying was investigated by Poncelet, et al. (1994)) and it showed promising results as small microbeads could be produced and as their size could be changed by changing some of the mentioned experimental set-up characteristics that can easily be changed, namely the flow rate, the needle diameter, the applied voltage and the distance between the needle and the solution below it, as well as the polymer concentration.

Immobilization of cells on microbeads created by electrospraying can be done and good results (concerning the growth and the attachment of the cells) were obtained for insect cells in Bugarski, Smith, Wu, and Goosen (1993). The applied voltage had an impact on the insect cell viability in Bugarski, Li, et al. (1994), where it was reduced by 7% immediately after the electrospraying, while they stated that the prolonged cultivation had not shown any reduction of neither the cell density nor the cell viability. Poncelet, et al. (1994) concluded that electrospraying can be used for cell immobilization and encapsulation, based on the results published in that paper and on the results in a referenced paper. It should be noted that in studies concerning encapsulation, electrospray is sometimes called electrostatic extrusion (Đorđević, et al., 2015). Electrospraying was also used for encapsulation of probiotics (J. U. Kim, et al., 2016) and proteins (Xie & Wang, 2007). According to Xie and Wang (2007), electrospray should be a good option for encapsulation of antibiotics, enzymes and DNA fragments, too. Lai, et al. (2017) showed that electrospraying can be used for producing multi-core hydrogel microspheres, which were made from carboxymethylcellulose or alginic acid or both and which could be used for drug delivery because of good encapsulation efficiencies, good drug release profiles (that were manipulated by changing the microspheres' composition) and a potential negligible toxicity. Electrospray could be used for solar cells because it already showed promising results when it was applied for organic solar cell enhancement (X.-Y. Zhao, et al., 2014) and for one-step nanosheet deposition on perovskite solar cells (Mahmood, Khalid, Nawaz, & Mehran, 2018).

Xu, Zhu, Han, Luo and Wang (2014) obtained good results when they used electrospraying in combination with electrospinning to produce fiber paper electrodes for Li-ion batteries, so potential applications in this field exist, too. A combination of electrospraying and flow focusing can also be used, as it was done by Moghaddam, Mortazavi and Khayamian (2015), who also added preheating of the electrosprayed fluid and called the resulting method – the melt coaxial electrospray method. Moghaddam, et al. (2015) used two immiscible fluids, which were flowing through two coaxial needles, to produce a phase change material that can be used for thermal energy storage. Since n-nonadecane was flowing through the inner needle and calcium alginate was flowing through the outer needle in the experiment done by Moghaddam, et al. (2015), they produced a material that has an *n*-nonadecane core and a calcium alginate shell.

Lim and Paul Chen (2007) used electrospraying for synthesizing a sorbent that has an iron oxide core and a calcium alginate shell. The sorbent produced by Lim and Paul Chen (2007) showed good results for copper and arsenic(V) ions' adsorption because both the core and the shell have good adsorption properties, while the possibility to easily separate the sorbent by using a magnetic force is also important.

CONCLUSIONS

Based on the reviewed literature, it can be concluded that electrospraying can be used in certain applications. The mentioned applications mainly need to be further developed, while new applications could be developed, too. The theoretical knowledge about this phenomenon exists, but it could also be further developed and used for drawing analogies between similar phenomena. It can be said that the empirical knowledge needed for a scale-up of this process has the same problem. The mentioned problems associated with electrohydrodynamics and the electrospray process are expected to be solved in the future.

NOMENCLATURE

d_d	droplet diameter
d_{min}	minimal droplet diameter
d_0	needle diameter
$E_{n,cj}$	maximal normal electric field in the cone-jet mode
$E_{t,cj}$	tangential electric field in the cone-jet mode
f_b	nondimensional jet radius at the break-up point
g	gravitational acceleration
h	distance between the plate and the receiving solution
I	emitted electric current
K	liquid's electrical conductivity
k	constant
l	characteristic length
m_d	mass of the droplet
Q	flow rate

Q_{min}	minimal flow rate for the stable cone-jet mode
q_R	Rayleigh limit
r_j	jet radius
U	electric potential
U_{cr}	critical electric potential
γ	surface tension
γ_0	unaltered surface tension
ε_0	vacuum permittivity
ε_r	relative permittivity
μ	viscosity
Π_μ	viscous parameter
ρ	density
σ	effective surface charge density

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