ELECTROHYDRODYNAMIC SPRAYING OF HIGHLY CONDUCTIVE AND VISCOUS LIQUIDS

by

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Abstract

Production of droplets is a very important process in the chemical and liquid-liquid extraction industries. In order to produce monodispersed droplets, high electric fields have been commonly used in liquid spraying, yet the fundamental aspects are not fully understood. In the work reported in this thesis, various fundamental knowledge relating to the applications of DC and pulsed DC electric fields have been advanced. Various spraying modes (such as dripping mode, cone jet mode and smooth jet mode) have been investigated with respect to highly viscous polyvinyl alcohol aqueous solutions. In order to produce monodispersed droplets, the smooth jet should be used together with pulsating DC field. An optimum frequency has been observed where the droplet production frequency follows linearly the applied pulse frequency until a limit, beyond which the liner relationship does not hold. The optimum frequency is a strong function of the liquid viscosity.

An analytical force balance model has been developed for dripping mode. The model allows the estimation of the produced droplet diameter as a function of the applied DC potential. It has been experimentally shown that this model can be applied to a wider range of applied potential compared to other force balance models in the literature. However, as it is a simple model, it does not describe the actual mechanism behind the droplet formation.

A number of liquid spraying geometries have been investigated, where optimum electrode geometry has been developed and tested. In this geometry, an optimum distance between the nozzle tip and the earthed electrode has been used such that satellite formation is inhibited, thus producing monodispersed droplets. This is especially beneficial in the spraying of highly viscous and conductive liquids.

State-of-the-art high-speed video photography and image-processing software have been used for the quantification of the above phenomena. A macro has been specifically written to automatically process a large number of images. The above monodispersed droplet production technique, together with the high-speed video photography and the macro developed for quantification, can be used commercially and/or industrially to generate monodispersed droplets for various functions.
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“Ma sopra tutte le invenzioni stupende, quella eminenza di mente fu quella di colui che s’immaginò di trovar modo di comunicare i suoi più reconditi pensieri a qualsivoglia altra persona, benché distante per lunghissimo intervallo di luogo e di tempo”

Galileo Galilei

Dialogo sopra i due massimi sistemi del mondo: Tolemaico e Copernicano
Firenze 1632 (I giornata VII, 126-131)

“But over all the wonderful inventions, that eminence of mind was that one of whom imagined to find the way to communicate its more recondite thoughts to whatever other person, although distant for longest interval of place and time”

Galileo Galilei

Dialogue Concerning the Two Chief World Systems, Ptolemaic and Copernican
Florence 1632 (First Day VII, 126-131)
To my loved parents: Michelina Mancini and Luigi Speranza
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end of the manuscript.
Nomenclature

$A_c$  Constant
$A_u$  Constant
$A_l$  Constant
$D$  Distance from the tip of the droplet to the earthed electrode
$D_{df}$  Diffusivity
$D_d$  Droplet diameter
$D_{dl}$  Droplet diameter before that the dripping shift to jet
$D_{in}$  Inside nozzle diameter
$D_j$  Jet diameter
$E$  Electric field
$E_n$  Normal electric field component
$E_t$  Tangential electric field component
$F$  Force
$F_e$  Force exert by the electric field
$F_g$  Weight
$F_{st}$  Force exert by the surface tension
$H$  Distance from the tip of the droplet to the tip of the capillary tube
$I$  Current density
$I$  Current
$I_0, I_1$  Bessel function of first kind
$K_0, K_1$  Bessel function of second kind
$M$  Mass flow rate
$M_{curv}$  Point on the droplet profile
$N_e$  Avogadro number
$O$  Axis center
$P_e$  Electrostatic pressure
$P_{e0}$  Electrostatic pressure to the tip of the droplet
$P_i$  Hydrostatic pressure
$P_s$  Surface tension pressure
$Q$  Flow rate
$Q_{total}$  Total charge
$R$  Radius of the wire
\( \mathfrak{R} \) Molar gas constant

\( R_e \) Generatrix cone

\( R_{Es} \) Electric stress to surface tension stress

\( R_1 \) Radius of curvature longitudinal direction

\( R_2 \) Radius of curvature transversal direction

\( S \) Surface area

\( S_e \) Electrode surface area

\( S_d \) Surface droplet

\( T \) Temperature

\( T_n \) Normal electric stress

\( T_t \) Tangential electric stress

\( U_j \) Velocity at which the jet ensues

\( V \) Electric potential

\( V_d \) Volume of the droplet

\( V_p \) Voltage applied between plates

\( V_{sp} \) Potential at which sparking occurs

\( V_{Total} \) Total volume

\( V_0 \) Electric potential applied to the liquid jet

\( V_1 \) Potential applied to the rods

\( V_2 \) Potential applied to the rods

\( W_n \) Electrostatic and free surface energy

\( X \) X axis

\( Y \) Distance from the droplet tip in upward direction

\( a \) \( D_y/2 \) = Jet radius

\( a_1 \) Coefficient

\( a_{11} \) Coefficient

\( a_{22} \) Coefficient

\( b \) Radius of curvature at the tip of the droplet

\( b_1 \) Coefficient

\( c_1 \) Coefficient

\( d_c \) Cylinder diameter

\( d_p \) Distance between electrode plates

\( d_e \) Distance between electrode

\( e^- \) Electron
\( f \)  Frequency

\( f_a \)  Applied frequency

\( f_d \)  Frequency droplet

\( f_{d,\text{opt}} \)  Optimum drop frequency

\( f_{dm} \)  Measured frequency droplet

\( f_{H} \)  Harkins factor

\( g \)  Gravitational constant

\( h \)  Distance between the needle and the earthen electrode

\( l \)  Length

\( l_c \)  Cylinder length

\( l_j \)  Jet length

\( m \)  Wave number

\( m_d \)  Droplet mass

\( n \)  Number of ions

\( n_d \)  Number of droplets

\( n_0 \)  Initial number of ions

\( p \)  Gas pressure

\( q \)  Varicose instability growth factor

\( q_d \)  Droplet charge

\( q_l \)  Lateral instability growth factor

\( r \)  Spherical radius of the cone

\( r_c \)  Generatrix cone co-ordination

\( r_d \)  \( D_d/2 = \) Droplet radius

\( r_{j0} \)  Radius of the jet for at time zero

\( t \)  Time

\( t_h \)  Hydrodynamic time

\( t_l \)  The time taken for the liquid meniscus to reach its limiting length

\( t_{\text{break-up}} \)  Time of break up

\( u \)  Ion mobility

\( u_{SW} \)  Ion mobility Stokes-Walden law

\( u_A \)  Ion mobility Adamczewski law

\( v \)  Velocity

\( v_j \)  Jet velocity

\( v_r \)  Velocity into cone jet
\( \nu_w \) Wave velocity
\( w \) Neck radius for surface tension force evaluation
\( z \) Axial distance
\( \alpha \) Half cone angle
\( \alpha_r \) Coefficient of recombination
\( \beta \) Liquid to vacuum permittivity ratio
\( \gamma \) Surface tension
\( \delta \) Disturbance
\( \delta^* \) Initial amplitude of disturbance
\( \varepsilon \) Dielectric constant
\( \varepsilon_0 \) Dielectric constant for free space
\( \varepsilon_r \) Relative dielectric constant
\( \lambda \) Wave length
\( \mu \) Viscosity
\( \rho \) Density
\( \sigma \) Conductivity
\( \sigma_q \) Surface charge
\( \tau \) Charge relaxation time
\( \tau_\mu \) Viscous relaxation time
\( \tau_Q \) Flow characteristic time
Chapter 1

1.1 Introduction

The spraying of a liquid into droplets in a gaseous atmosphere is of importance in a number of fields such as industrial processing, agriculture, combustion, powder production, spray painting, cooling, etc. and it may be performed in a number of ways such as pressure atomizer, rotary atomizer, air-assist atomizer, airblast atomizer, effervescent atomizer and electrohydrodynamic atomizer. In electrohydrodynamic spraying, the possibility of manipulating the liquid with electric fields has led to several technological applications including spray painting (Hines, 1966), crop spraying (Coffee, 1981), fuel injection (Lehr and Hiller, 1993), particles and powders production (Balachandran et al. 1994; Borra et al, 1997, Brandenberger et al, 1999), spray deposition of ceramic thin films (Chen et al, 1999), polymer films with a specific surface structure (Choy and Bai, 2000), ink-jet printing, space propulsion and emulsification (Bailey, 1988; Masters, 1992).

Nowadays, the sector of chemical industry dealing with paints, adhesive and particle production is looking with increasing interest to water based solution because of the low cost of the solvent and its environmental compatibility. These solutions may be the result of a mix of many compounds hence they may have complex rheological properties and may have viscosities of a few orders of magnitude higher than water. Especially the spraying for high viscous liquid may be a non-easy problem to overcome when the control of the droplet size distribution is a key factor. Depending on the kind of industrial application it is required to produce droplets with a diameter ranging from a few millimeters to a few microns and it is essential to have droplets with a narrow size distribution, this is because properties such as powder handling, powder dispersion, powder deposition, coating uniformity, combustion, etc. are related to droplet diameter and size distribution. Electrohydrodynamic spraying has the characteristic of producing droplets in a wide diameter category with a narrow size distribution and in addition it permits using a flow rate from few hundredth of milliliter to tens of milliliter. These
electrohydrodynamic spraying features make the technique very flexible and easy to set up for new plants as well as to adapt for already existing plants.

The aim of this thesis is to investigate the possibility of producing droplets made from highly viscous and conductive liquids in a controlled size distribution using electrohydrodynamic spraying. There is evidence in the literature (Balachandran et al., 1994; Huneiti et al., 1998) that it is possible to spray in a controlled fashion droplets of highly conductive liquids such as aqueous solutions, however the work done was restricted to liquids having relatively low liquid viscosity (i.e. below 10 mPas). Therefore the emphasis is placed on the effect of the liquid viscosity especially when the viscosity goes above 100 mPas. The droplets produced need to have diameter of about one hundred of microns using flow rates high enough to match industrial needs of few tons a day. Indeed one of the major limitations of electrohydrodynamic spraying used in industrial applications is the very low flow rate that can be achieved.

The objectives of the research are:

- to investigate which are the most suitable electrohydrodynamic functioning modes among smooth jet mode, cone jet mode (converging jet mode) and dripping mode, as well as, determining the most appropriate electrode configuration and applied DC potential for achieving the desired of droplet diameter, size distribution and flow rate using highly viscous and conductive liquids.

- to couple the axisymmetrical wave on the jet surface with a pulsating DC potential in order to control the liquid jet break up regime.

- to investigate how changing the DC potential and its frequency influences the droplet diameter produced and the size distribution with respect to different liquid viscosities.

- to examine whether the liquid viscosity imposes limit for the successful electrohydrodynamic spraying of a highly conductive liquid.
Chapter 2 reviews the relevant literature on the various electrohydrodynamic functioning modes such as dripping, smooth jet and cone jet mode in order to understand which are the fundamental aspects of droplet production in this modes. Chapter 3 reports on the experimental arrangement used to perform the experiments and the technique used for droplet sizing. The liquids used in this work have been tap water, distilled water, and polyvinyl aqueous solutions. Polyvinyl alcohol gives the possibility of preparing solutions whose viscosity can change over wide range while maintaining other physical properties such as surface tension, density and electrical conductivity almost constant.

Chapter 4, 5 and 6 report the experimental results obtained from spraying the above liquids in the dripping mode, cone jet mode and in the smooth jet mode using constant DC potential as well as pulsating DC potential. Also at the end of each chapter the results obtained are discussed. Chapter 7 contains conclusions on the work done and recommendation for future work on this topic.
Chapter 2 - Literature Survey

2.1 Introduction

The application of an electric field to a liquid emerging from a nozzle in air shows a number of electrohydrodynamic spraying functioning modes, leading to the production of drops with diameters between a few millimetres to a few tenths of a micron (Cloupeau and Prunet-Foch, 1994; Rossell-Llompart and De La Mora, 1994). In fact, the application of an electric field and the charge induced by the electric potential generate a force, which acts against the surface tension and may induce surface deformation of the liquid.

Jaworek and Krupa (1999) proposed two criteria based on the geometrical form of the liquid at the outlet of the capillary such as droplet, spindle, cone jet, smooth jet etc. and on the mechanism of the disintegration of the liquid jet into droplets for the classification of the possible electrohydrodynamic spraying modes. They also listed two groups in which the electrohydrodynamic spraying mode may be divided. The first group consists of all the modes in which only fragments are directly emitted from the capillary. These modes are the dripping mode, microdripping mode, spindle mode, multi-spindle mode, and ramified-meniscus mode. The second group consists of all the modes in which the liquid is emitted from the capillary in a form of a jet that breaks up into droplets a few millimeters away from the nozzle. The latter group contains the cone-jet mode, precession mode, oscillating-jet mode, multijet mode and ramified-jet mode. In the present work a new electrohydrodynamic functioning mode has been observed and this is grouped with the well-known electrohydrodynamic functioning modes in a flow chart following the idea originated by Grace and Marijnissen (1994), Figure 2.1.
Figure 2.1 Possible electrohydrodynamic spraying functioning modes; the potential increase with downwards direction.
2.2 Dripping mode

When a liquid emerges from a nozzle in air with a flow rate of 1-5 ml/min, droplets are released individually from the tip of the nozzle: this is dripping mode. Without the application of a potential to the nozzle, the droplet leaving this nozzle has a volume that depends only on the liquid surface tension, density and nozzle diameter. When an electric potential is applied to the nozzle an electric force acts on the liquid droplet forming at the nozzle orifice. The electric force acts in addition to the gravitational force and induces the formation of a droplet with a smaller volume. The increase of the applied potential leads to: (a) the increase of the droplet frequency, (b) further reduction of the droplet volume and (c) a change in the shape of the droplet (Hayati et al. 1987a). Indeed, the increase of the applied potential has the effect of changing the shape of a droplet from an ellipsoidal profile to a near paraboloidal profile when subjected to high electric fields. Borzabadi and Bailey (1978) in the attempt to understand the mechanism behind the above mentioned phenomena, predicted the profile of a pendent droplet by solving the Young-Laplace equation modified for the presence of the pressure due to the electric field, see Equation (2.1).

\[
\frac{2\gamma}{b} - \rho g Y - P_{e0} = \gamma \left( \frac{1}{R_1} + \frac{1}{R_2} \right) - P_e
\]

(2.1)

where \( b \) is the radius of curvature at the tip of the droplet, \( P_{e0} \) is the pressure due to the

![Figure 2.2 A typical pendent drop hanging from a capillary tube (Borzabadi and Bailey, 1978).](image)
electric field at the tip of the droplet, $P_{e}$ is the pressure exerted by the electric field, $Y$ is the distance taken from the tip of the droplet, $\gamma$ is the surface tension, $g$ the gravitational constant, $R_1$ and $R_2$ are the two principal radii of curvature at point $M_{\text{curv}}$ and $\rho$ the density, see Figure 2.2. They found a good match between experimental data and theoretical results only for very high applied potential; 4.3 kV.

The mechanism of dripping consists of a cyclic process in which the pendant droplet is stretched in the downwards direction until it reaches a maximum elongation over which the detachment of the droplet from the nozzle occurs (see Chapter 4). However, the mechanism of dripping with the application of an electric field is difficult to generalise and it may involve very different behaviours depending upon the electrode geometry as well as the physical properties of the liquid used such as viscosity, conductivity and surface tension. Cloupeau and Prunet-Foch (1994) depicted the way in which dripping may occur depending on the liquid viscosity. Droplets of liquid such as cyclohexanol when stretched by an electric field exhibited a very long filament between the forming droplets and the liquid meniscus attached to the nozzle. When this liquid filament reached a maximum elongation, it broke up into a number of little droplets. Liquids such as ethyl alcohol with a lower viscosity than cyclohexanol were characterised by a well-defined break-up of the liquid filament and with the formation of a smaller number of little satellite drops. In addition during each dripping cycle a short jet, was observed, emitting from the tip of the liquid meniscus. Bugarski et al. (1994) observed that when spraying alginate water solutions in dripping mode, the low concentration alginate solutions showed a neck elongation which was less pronounced than the solutions with high concentration in alginate. Therefore, on one hand it may be said that dripping of liquids having a large viscosity is characterised by the presence of a long liquid neck formation and thus a polydispersed spray may be generated. On the other hand, liquids with rather low viscosity but having a high surface tension, e.g. water when sprayed in dripping mode, show a neater and shorter necking process.

As mentioned above, in dripping mode the volume of the droplet and hence the droplet diameter is determined by the liquid surface tension, liquid density and electric field when an electrical potential is applied to the nozzle. Jaworek and Krupa (1992) observed that when the electric field applied was increased above $2\times10^{6}$ V/m, the
The droplet diameter started to decrease considerably. For higher values of the electric field, the droplet diameter decreased in a parabolic manner until a value of about $9.5 \times 10^6$ V/m was achieved. At this value of the electric field, the droplet was accompanied by a much smaller drop called a satellite, and the mechanism of dripping changed. The decrease in the droplet diameter led to an increase in the dripping frequency of one or two orders of magnitude compared to the drop size with no applied potential. Bugarski et al. (1994) reported a similar pattern for the curve of droplet diameter versus applied potential. Above an applied electric field of $4.2 \times 10^6$ V/m, there was a rapid reduction in the droplet diameter until the electric field reached $12.6 \times 10^6$ V/m (this interval was called region I). Over $12.6 \times 10^6$ V/m, the droplet diameter reduced slightly and asymptotically reached a plateau (this was called region II). Indeed, beyond $12.6 \times 10^6$ V/m, once again there was a change in the dripping mechanism with the liquid filament beginning to whip from side-to-side, breaking off droplets to produce a bimodal drop size distribution. Takamatsu et al. (1981, 1983) worked out a theoretical relationship in order to predict the droplet diameter to the electric potential increase. The relationship was derived from a force balance applied to the pendant droplet, see Equation (2.2), and it gave good agreement with their experimental data only if the electrical force was multiplied by a correction factor. However, this kind of approximation gave no details about the dynamics of droplet formation and break up in the presence of an electric field.

\[ F_{\text{total}} = F_g + F_e \]  

(2.2)

A more refined theoretical analysis was recently proposed by Notz and Basaran (1999) who used a finite element analysis to predict the profile of droplets made of an inviscid and perfectly conductive liquid in the presence of an electric field. The characteristic profile of the hanging droplet proposed was in good agreement with what was experimentally observed.
2.3 Cone jet mode

Electrohydrodynamic atomisation in cone jet mode occurs when the liquid is accelerated downstream in an electric field where it assumes a cone-like shape. The acceleration is due to charges which migrate on the liquid surface and accelerate toward the cone apex due to the electric field. Zeleny (1915, 1917) reported the above phenomenon and gave the first known description by reporting photographs of the spraying of alcohol from a capillary tube. Later Vonnegut and Neubauer (1952) "rediscovered" the phenomenon and showed how, using a high voltage, it was possible to create an "electrostatic fountain" of water droplets. At a certain value of the applied potential, it was possible to create a fine smoke, with droplets of the order of one micron. Taylor (1964) gave an explanation of the cone-like shape taken by a liquid meniscus in terms of a pressure balance between the surface tension pressure, $P_s$, and the normal electric pressure, $P_e$; see Equation (2.3) and (2.4) originally due to Taylor (1964) and reviewed more recently by Meesters et al. (1992). A conical liquid surface (calculated from an ideal no-flow equilibrium condition) could exist in equilibrium in an electric field only when the cone had a semi-vertical angle of 49.3°.

\[
P_e = \frac{1}{2} \varepsilon_0 E^2 \quad \text{(2.3)}
\]

\[
P_s = \frac{\gamma}{r_c \tan \alpha} \quad \text{(2.4)}
\]

where $\varepsilon_0$ is the dielectric constant for free space, $E$ is the electric field strength, $\gamma$ is the surface tension, $\alpha$ is the half cone angle and $r_c$ is the generatrix cone co-ordination, see Figure 2.3a. Taylor (1969) observed that once the liquid meniscus formed its conical shape, it was possible to observe drops thrown off from the tip of the liquid meniscus as soon as the applied potential was raised above a critical value. Any further increase in the applied potential led to an increase of the number of droplets emitted per second until the formation of a steady stream took place. Smith (1986) worked out a formula for determining the applied potential needed to achieve a stable cone jet with respect to the electrode geometry used and liquid sprayed, see Equation (2.5).
\[ V = A_i \left[ \frac{2\gamma R_c \cos \alpha}{\varepsilon_0} \right]^{\frac{1}{2}} \cdot \ln \left( \frac{4h}{R_c} \right) \] (2.5)

where \( A_i \) is a constant, \( R_c \) is a generatrix of the cone and \( h \) is the distance between the needle and the earthed electrode.

Equation (2.5) suggests that the electric potential for a stable cone would scale with the square root of the liquid surface tension. Also Smith (1986) indicated that the maximum surface tension for a liquid to be atomised in cone jet mode was circa 0.05 N/m. Liquids with a higher surface tension may require an applied potential strong enough to provoke dielectric break-down of the air. Indeed, the nature of the gas in which the cone may form is fundamental to the process. Hayati et al. (1987a) conducted experiments on droplet deformation due to an electric field using a number of gases. It was observed that when the potential was applied to a hanging water droplet immersed in gaseous He, no droplet deformation was recorded until a value of the applied potential in which sparking occurred. Helium has a much lower break-down potential with respect to that of air, thus gas break down occurred well before any effects on the liquid to be sprayed.

Hayati et al. (1987b) reported that the existence of a stable jet was possible only if there was an electric shear stress on the liquid surface. When an electric field is
applied to the surface of a liquid, the charges relax on the surface in a time, $\tau$, see Equation (2.6). In liquids with considerable electrical conductivity, $\sigma$, such as water solutions ($>10^{-4}$ S/m) the relaxation time can be very short: circa one microsecond, and the surface of the liquid becomes equipotential (Bailey, 1988).

$$\tau = \frac{\varepsilon}{\sigma}$$  \hspace{1cm} (2.6)

Here $\varepsilon=\varepsilon_0\varepsilon_r$, and $\varepsilon_r$ relative dielectric constant of the liquid. On the liquid surface, there is a tangential and normal component of the electric field, which are responsible for the creation of a tangential and normal stress on the drop as given below (Melcher and Taylor, 1969):

$$T_n = \frac{1}{2}\varepsilon_0E_n + \frac{1}{2}(\varepsilon - \varepsilon_0)E_t^2$$  \hspace{1cm} (2.7)

$$T_t = \varepsilon_0E_tE_n$$  \hspace{1cm} (2.8)

In Equations (2.7) and (2.8), $E_n$ is evaluated by solving the integration of Poisson's equation, while $E_t$ is calculated from Equation (2.9).

$$E_t = \frac{I}{2\pi\sigma(1-\cos\alpha)r^3}$$  \hspace{1cm} (2.9)

where $I$ is the current flowing through the nozzle and $r$ is the spherical radius of the cone. Hayati (1987b) evaluated the normal and tangential stresses for a liquid with an electrical conductivity of $1.4 \times 10^{-7}$ S/m, finding that the tangential stress was about 5 to 10 times smaller than the normal stress. Despite the small value of the shear stress, this produced a downward longitudinal force that stabilised the jet by smoothing out any perturbations on the liquid surface. Hence, in a liquid with high electrical conductivity, the shear stresses required to stabilise the jet may be so small that a stable jet cannot be formed.

\cite{Melcher_Taylor_1969}

\footnote{The Equation is dimensionally incorrect and it is reported as in Melcher and Taylor (1969).}
The tangential stress is also responsible for producing eddies inside the liquid cone. Hayati (1992) observed that, when tracer lycopodium particles were inserted in the liquid, an axisymmetric circulation was present within the liquid cone. The particles of lycopodium moved along the surface of the cone and returned upwards along the axis of the cone when the atomised liquid had an electric conductivity of $3.3 \times 10^7$ S/m (semiconductive liquids), see Figure 2.3b. No eddies were observed when the liquid was substituted with water. Hayati (1992), also worked out an equation for evaluating the velocity profile inside the liquid cone as reported below:

$$v_r = \frac{1}{r_c} \frac{\varepsilon_0 E_n I}{4\pi \cdot \sigma (1-\cos \alpha) \mu} \left( \frac{\sin \alpha}{(1-\cos \alpha) \sin \alpha} \right) \cdot \left( \frac{2\cos \alpha}{(1-\cos \alpha) r_c^2} \right) + \frac{Q}{2\pi (1-\cos \alpha) r_c^2}$$  \hspace{1cm} (2.10)

In Equation 2.10, $E_n$ is the normal component of the electric field strength, $\mu$ is the viscosity, $\sigma$ is the electric conductivity and $Q$ is the flow rate and alpha is the cone angle, see Figure 2.3.

When the liquid cone is formed, a liquid filament is emitted from the very end of the cone. The liquid filament moves downstream and breaks up into droplets after some distance (3-5 mm). The effect of the electric field upon liquid spraying is significant when the charge relaxation time is smaller than the hydrodynamic time, $t_h$, which is the time required for the fluid particles with characteristic velocity $v$ to cover a certain distance $l$, see Equation (2.11).

$$t_h = \frac{l}{v}$$  \hspace{1cm} (2.11)

It is known that liquids, such as Toluene (Smith, 1986), with very low electrical conductivity do not exhibit any electrohydrodynamic spraying due to the low amount of charge carriers. While the liquid is flowing downstream the charges are migrating towards the liquid surface and they accumulate into a thin layer beneath the liquid surface, where the thickness of the thin layer is of the order of the Debye length. Two phenomena are present within the jet: charge conduction and charge convection. The former is dominant at the beginning of the jet, while the latter is dominant towards the end of the jet. Between these two regions there is a transition region where convection...
and conduction charges are equally important. However, the charges will eventually leave the jet in the form of charged droplets, while new charges are supplied to the jet from the nozzle. The number of charges that a droplet can hold has been of interest to some researchers. Rayleigh (1882) predicted that the maximum charge-to-mass ratio that a conducting liquid droplet of a given radius could possess, can be given by the following equation:

\[ \frac{q_d}{m_d} = \frac{6}{\rho} \left[ \frac{\gamma e_0}{r_d^3} \right]^{1/2} \]  
(2.12)

where \( q_d \) is the charge of a droplet and \( m_d \) is the mass of a droplet. Vonnegut and Neubauer (1952) established the following relationship for the charge to mass ratio from the energy minimisation principle:

\[ \frac{q_d}{m_d} = \frac{1}{2} \frac{6}{\rho} \left[ \frac{\gamma e_0}{r_d^3} \right]^{1/2} \]  
(2.13)

The value of \( \frac{q_d}{m_d} \) obtained from Equation 2.13 is equal to half the Rayleigh limit (Bailey, 1988). The same result was obtained by Pfeifer and Hendricks (1967) who calculated the ratio \( \frac{q_d}{m_d} \), starting from the minimisation of the electrostatic and free surface energies \( W_n \):

\[ W_n = \frac{3 \gamma V_{\text{total}}}{r_d} + \frac{Q_{\text{total}}^2 r_d^2}{6 \varepsilon_0 V_{\text{total}}} \]  
(2.14)

where \( V_{\text{total}} \) is the total droplets' volume and \( Q_{\text{total}} \) is the total droplets' charge. The usefulness of Equation (2.13), and thus of the evaluation of the charge-to-mass ratio, is that, in the absence of corona discharge, the current \( I \) flowing through the liquid jet is

\[ \text{This equation is the adopted one because of the difficulty of reaching the charge-to-mass ratio suggested by Rayleigh (1882).} \]
equal to the current transported by the drops. In this condition, the ratio \( \frac{I}{M} \) (current-to-mass flow) is equal to the ratio \( \frac{q_d}{m_d} \), so the droplet diameter can be written using Equation (2.14) as:

\[
D_d = \left[ \frac{6(2\varepsilon \sigma \gamma)^{1/3} M}{I \rho} \right]^{2/3}
\]

(2.15)

Experimental evidence shows that this formula permits the prediction of the droplet diameter, \( D_d \), within one order of magnitude difference (Cloupeau and Prunet-Foch, 1994).

Further work has been done in deriving scaling laws for estimating the current needed in a cone jet mode in order to produce droplets with a certain diameter. The usefulness of scaling laws is that of having easy formulae for facilitating the design of electrohydrodynamic systems and droplet diameter prediction. Fernandez de la Mora and Loscertales (1994) proposed a scaling law of the spraying current emitted from an electrified meniscus in the cone jet mode for the case of highly conductive liquids, and found the following relationship:

\[
I = f(\beta) \left( \frac{Q \sigma \gamma}{\beta} \right)^{1/2}
\]

(2.16)

where \( \beta \) is the liquid to vacuum permittivity ratio and \( f(\beta) \) is a dimensionless function that needs to be experimentally determined. Fernandez de la Mora and Loscertales (1994) also propose a scaling law for the jet diameter according to the following relationship:

\[
D_j \approx 0.4(Q \tau)^{1/3}
\]

(2.17)
as reviewed by Hartman (1998). Gañana-Calvo et al. (1997) disagreed with the finding above, and proposed new scaling laws of the spraying current as well as of the droplet diameter emitted from an electrified meniscus in the cone jet mode. The electric current and the droplet diameter were dependent on liquid properties such as conductivity and viscosity rather than the liquid permittivity. Indeed, two sets of equations were proposed based on the following dimensionless number:

$$\left( \frac{\gamma^2 \varepsilon_0^2}{\mu^3 \sigma^2 Q} \right)^{\frac{1}{3}}$$

(2.18)

The dimensionless number in Equation (2.18) can describe two classes of liquids: (1) highly viscous and conductive liquids when the dimensionless number is either smaller than or equal to 1, and (2) low viscous and conductive liquids where the dimensionless number is greater than 1. The flow of high conductive and viscose liquids is characterised by a flat velocity profile due to the way in which the tangential stress is efficiently transmitted, while the velocity profile is not flat in liquids with low conductivity and viscosity. The tangential stress due to the applied electric field accelerates the liquid jet surface more than the liquid bulk and therefore a parabolic velocity profile is formed. These two different velocity profiles of the jet affect the charge convection in a dissimilar way, thus the electric current and the droplet diameter scale differently depending on which above-mentioned category the liquid belongs to. For highly viscous and conductive liquids the current and the droplet diameter scale as in Equation (2.19) and (2.20). For low viscous and conductive liquids, the current and droplet diameter scale as in Equation (2.21) and (2.22).

$$\frac{I}{I_0} = 6.2 \left( \frac{Q}{(\beta - 1)^{\frac{1}{2}} Q_0} \right)^{\frac{1}{2}} - 2.0$$

(2.19)

$$\frac{D_d}{(\beta - 1)^{\frac{1}{3}} d_0} = 1.6 \left( \frac{Q}{(\beta - 1)^{\frac{1}{2}} Q_0} \right)^{\frac{1}{3}} - 1.0$$

(2.20)
Different scaling relationships for the electric current and droplet diameter in the cone jet mode have been worked out by Hartman (1998) who discovered that for the kind of liquid used in his experiments, the current and droplet diameter can be scaled as:

\[ \frac{I}{I_0} = 11.0 \left( \frac{Q}{Q_0} \right)^{\frac{1}{4}} - 5.0 \]  
(2.21)

\[ \frac{D_d}{d_0} = 1.2 \left( \frac{Q}{Q_0} \right)^{\frac{1}{24}} - 0.3 \]  
(2.22)

where \( d_0 = \left( \frac{\gamma \varepsilon_0}{\rho \sigma^2} \right)^{\frac{1}{3}} \), \( I_0 = \left( \frac{\gamma^2 \varepsilon_0}{\rho} \right)^{\frac{1}{2}} \) and \( Q_0 = \frac{\gamma \varepsilon_0}{\rho \sigma} \).

Different scaling relationships for the electric current and droplet diameter in the cone jet mode have been worked out by Hartman (1998) who discovered that for the kind of liquid used in his experiments, the current and droplet diameter can be scaled as:

\[ \frac{I}{I_0} = a_1 \left( \frac{Q}{Q_0} \right)^{b_1} \]  
(2.23)

\[ D_d = c_1 \left( \frac{\rho \varepsilon_0 Q^4}{I^2} \right)^{\frac{1}{6}} \]  
(2.24)

respectively, where the coefficients \( a_1 \) and \( b_1 \) in Equation (2.23) are tabulated by Hartman (1998) in terms of the liquid category, the nozzle diameter used and the range of flow rate, while \( c_1 \) in Equation (2.24) is made equal to 1.76. N.B. Equation 2.16-2.24 are applicable only for the spraying in cone jet mode.
2.4 Liquid jet break up

This section reports on the mechanism of break up of a non-electrified liquid jet as well as an electrified liquid jet by varicose instability (see Section 2.4.1 and Section 2.4.2). Also it reports the effect of an intense liquid jet electrification on the liquid jet break up (see Section 2.4.3) and the mechanism of droplet and satellite formation for varicose instability (see Section 2.4.4.). Finally a number of techniques used in order to control the liquid jet break up by non-static electric field are commented upon (see Section 2.4.5.).

2.4.1. Varicose instability

The break-up of a jet by varicose instability occurs when on the liquid jet surface there are waves that propagate in an axisymmetric mode (i.e. axisymmetric waves). These waves propagate and grow along the jet in a downstream direction, inducing the break-up of the liquid jet into droplets, see Figure 2.4. Varicose instability has been studied by a number of researchers. Plateau as revised by Worthington (1879) proposed that the equilibrium of a free cylinder made of any liquid, under the influence of a surface tension, becomes unstable as soon as the length of the cylinder, $l_c$, exceeded $\pi$ times the diameter, $d_c$, of the cylinder. In other words a liquid cylinder of length $l_c$ breaks up into $n_d$ number of droplets as follows:

$$n_d = \frac{l_c}{\pi d_c}$$  \hspace{1cm} (2.25)

Rayleigh (1878-1879), in disagreement with Plateau, presented a theory based on infinitesimal disturbances, which may be caused by deviation of the nozzle walls, by the compression and expansion of the liquid stream, etc. These disturbances travel along the jet and grow until the liquid jet breaks up into droplets. Thus, the unstable equilibrium depends on the nature of the disturbances. Indeed, Rayleigh supposed that there were a number of disturbances on the liquid jet surface and some of these disturbances were producing their effects much more rapidly than others. Thus, it was necessary to understand the wavelength of the disturbances that grew the quickest.
Rayleigh (1879-1979) proposed an exponential growth factor for liquids in which viscosity may be neglected as follows:

\[ q^2 = \frac{\gamma(1-m^2a^2)maI_1(ma)}{\rho a^3I_0(ma)} \]  

(2.26)

where \( m \) is wave number, \( m=2\pi/\lambda \), \( \gamma \) the surface tension, \( \rho \) the density, \( a \) is the jet radius and \( I_0(), I_1() \) are Bessel functions of the first kind. The growth factor is a maximum for a value of \( ma=0.697 \). The growth rate coefficient \( q \) comes from the mathematical representation given for small disturbances reported by Miesse (1955) as:

\[ \delta = \delta^*e^{\alpha t} \cos(m \cdot z) \]  

(2.27)

where \( \delta^* \) is the initial disturbance and \( t \) is the time.

Figure 2.4 Liquid jet break-up by varicose instability.
Basset (1894) and more recently Bailey (1988) extended the evaluation of the growth factor to liquids with significant viscosity effect, and proposed a growth factor in the following form:

\[
q^2 \left( \frac{\rho a l_0(ma)}{10 ma l_1(ma)} \right) + q \left( \frac{2a ml_0(ma)}{l_1(ma) - 1} \right) \frac{2\mu}{a^210^6} - \frac{\gamma(m^2a^2 - 1)}{10a^2} = 0
\]

(2.28)

Where \( \mu \) is the viscosity. Equation (2.28) shows that, the greater the liquid viscosity, the smaller the wave number for the maximum value of the growth coefficient.

Further theoretical analysis of the break up of a liquid jet for which viscosity cannot be neglected was presented by Weber (1931). Weber suggested a value of the wave number for the most rapidly growing disturbance in terms of viscosity, surface tension, density and liquid jet diameter as follows:

\[
m = \frac{2}{D_j} \left( 2 + \frac{6\mu}{(D_j\gamma p) \frac{1}{2}} \right) \frac{1}{2}
\]

(2.29)

Once more, an increase in the liquid viscosity leads to a smaller value of the wave number for which the growth factor is a maximum, in agreement with Basset’s conclusion, see Equation (2.29) and Figure 2.5.

Observing the way in which the break up of a liquid jet takes place, it is possible to work out a relationship between the wavelength of the fastest growing disturbance and the diameter of the droplet produced. The following mass balance may be written assuming that each section of the cylindrical liquid jet with a length \( \lambda \) decomposes into a spherical droplet having the same volume and a diameter \( D_d \):

\[
\rho \pi \frac{D^2_j}{4} \lambda = \rho \frac{4}{3} \pi \frac{D^3_d}{8}
\]

(2.30)
where \( D_j \) is the jet diameter. Rearranging Equation (2.30) the droplet-diameter-to-jet-diameter ratio may be related to the dimensionless wave number as:

\[
\frac{D_d}{D_j} = \left( \frac{3\pi}{mD_j} \right) \frac{1}{3}
\]  

(2.31)

Thus, from Equation (2.31), the wave number as proposed by Rayleigh for inviscid liquids can be written as:

\[
\frac{D_d}{D_j} = 1.89
\]  

(2.32).

Figure 2.5 Varicose growth rate as a function of the wave number for four values of the liquid viscosity; \( a = 1 \) mPa\(\cdot\)s, \( b = 40 \) mPa\(\cdot\)s, \( c = 140 \) mPa\(\cdot\)s and \( d = 280 \) mPa\(\cdot\)s.
For liquids with larger values of viscosity, the droplet-diameter-to-jet-diameter ratio assumes a more complex form:

\[
\frac{D_d}{D_j} = \left[ \frac{3\pi}{2} \left( 2 + \frac{6\mu}{(D_j\gamma\rho)^1} \right)^{1/2} \right]^{1/3}
\] (2.33)

Equations (2.32) and (2.33) coincide for values of \( \mu \) approaching zero. Therefore, it may be concluded that an increase in the liquid viscosity leads to an increase in the wavelength (i.e. the wave number decreases) as well as to a decrease of the growth factor. Hence when the liquid jet breaks up into droplets, the greater the liquid viscosity, the larger the droplet diameter produced, and moreover the larger the liquid viscosity, the longer the waves have to travel along the jet before gaining an amplitude large enough to induce liquid jet break up.
2.4.2 Electrified liquid jet: varicose instability

The stability of an electrified liquid jet has been of interest to many researchers. Basset (1894) proposed a growth rate factor for an electrified jet as follows:

$$\frac{q^2 I_0(\lambda a)}{\lambda a I_1(\lambda a)} + \frac{2 \mu}{\rho \alpha^2} \left[ \frac{2 \lambda a I_0(\lambda a)}{I_1(\lambda a)} - 1 \right] q - \frac{k^2 \rho \lambda K_0(\lambda a)}{\rho \lambda a K_1(\lambda a)} +$$

$$+ \frac{\gamma (m^2 a^2 - 1)}{\rho \lambda a K_1(\lambda a)} + \frac{\sigma_q^2}{4 \pi \rho a^2} \left[ \frac{1}{\rho \lambda K_1(\lambda a)} \right] = 0$$  \hspace{1cm} (2.34)

where $\sigma_q$ is the charge for unit of length of the undisturbed jet and $K_0(\lambda a)$ and $K_1(\lambda a)$ are Bessel functions of the second kind. Basset, from his comprehensive analysis that also included the effect of the surface tension, viscosity and the surrounding air, drew the following conclusions: (1) the viscosity tended to produce jet stability while the surrounding air tended to produce jet instability: (2) the surface tension tended to produce either jet stability or jet instability depending on whether the wave number was greater or less than unity: (3) the electrical charge tended to produce jet stability or jet instability if the wave number was smaller or larger than 0.6, respectively. More recently, Taylor (1969) found an error within Equation (2.34), and modified it to the following equation for the wave growth rate of an electrified jet:

$$\frac{\rho a q^2 I_0(\lambda a)}{\lambda a I_1(\lambda a)} = \frac{\gamma (m^2 a^2 - 1)}{a^2} - \frac{\sigma_q^2}{\pi a^2} \left[ 1 - \frac{\lambda a K_1(\lambda a)}{\rho \lambda K_1(\lambda a)} \right]$$  \hspace{1cm} (2.35)

Taylor observed that his equation differed from that of Basset’s by a factor of $\frac{1}{4}$, and was thus qualitatively the same. Only for a quantitative comparison, e.g. when comparing experimental data with theoretical calculations, is this factor important, see Equation (2.34) and (2.35).

Schneider et al. (1967) following Rayleigh’s method, investigated how the growth coefficient depends on the applied electric potential, for which they proposed the following equation.
where $V_0$ is the potential applied to the liquid jet. The second term in the above equation illustrates the influence of the electric charge. Schneider et al. (1967) concluded that this second term was negligible when compared with the effect of surface tension on the wavelength, for varicose instability. Equation (2.36) was further modified by Neukermans (1973), but the result was identical to that of Schneider et al. (1967), with the only difference being the second term having a smaller value by a factor of 10.6, making it even more negligible. These researchers concluded that the liquid jet break-up depends only on the viscosity and surface tension.

Moreover, Bailey and Balachandran (1981) investigated the effects of an applied voltage upon the stability of a viscous liquid jet using the following form for the wave growth rate coefficient:

$$q^2 = \frac{\gamma(1-m^2a^2)maI_1(ma)}{\rho \alpha^4 I_0(ma)} - \frac{\varepsilon_0 V_0 maI_1(ma)}{\rho \alpha^4 (\ln \alpha)I_0(ma)} \left[ 1 + \frac{maK_0(ma)}{K_0(ma)} \right]$$

In disagreement with Schneider et al. (1967) and Neukermans’s conclusions, Bailey and Balachandran stated that an applied potential did have an effect on the liquid jet break-up. They observed that the increase of the applied potential lead the maximum growth rate to shift to higher values of wave number. This had the consequence of producing smaller droplets compared with the case when the jet break-up occurred without any potential being applied. The effect of the applied potential was therefore just the opposite of the effect of the viscosity. Hartman et al. (2000) were of the same view, pointing out however that the electric stresses had a stabilizing effect upon long wavelength perturbations, and a destabilizing effect on the short wavelength perturbations. More precisely Hartman et al. (2000) found that, for values of the dimensionless wave number below 0.58, the electrically induced stresses had a stabilising effect.
Turnbull (1992), in his theoretical analysis on the stability of an insulating liquid jet, took into consideration the effect of normal and tangential electric fields. Turnbull concluded that the effect of the charge distribution was that of stabilising the long wavelength perturbations and destabilising the short wavelength perturbations, while the effect of the only normal component of the electric field was that of stabilising the short wavelength perturbations.

It should be observed that in the literature there is a disagreement on the effect of the electric charges on the liquid jet break-up. As reported above, Schneider et al. (1967) and later Neukermans (1973) concluded that the effect of the surface charge had a marginal role in the liquid jet break up. Similar conclusions were found by Cloupeau and Prunet-Foch (1989) who, in their experimental work, recorded the experimental data points of the mean droplet diameter and jet diameter for an electrified liquid jet. By plotting the mean droplet diameter versus the jet diameter Cloupeau and Prunet-Foch (1989) observed that all the data points lie on a line with slope of 1.89 (as predicted by Rayleigh, see Equation (2.32)), regardless of the magnitude of the applied potential to the jet. Hence they concluded that the applied potential had a limited influence on the droplet-to-jet-diameter ratio as long as the charge of the jet was not too high; the experimental data points were obtained for applied potentials of between 3 and 4 kV. More work supporting the hypothesis that the electric charges have a limited effect upon the liquid jet break up can be found in the work of Mutoh et al. (1979). In the work of Mutoh et al. (1979), the measured droplet frequency, for a number of viscous liquids and applied potentials was plotted versus the optimum droplet frequency evaluated by Weber’s work, which considered only viscous and surface tension effects. The formula for the evaluation of the optimum drop frequency is reported below:

\[
f_{d, opt} = M \left( \frac{\pi^2 \rho D_j^3}{4} \left[ 2 \left[ 1 + \frac{3\mu}{(\rho\gamma D_j)^{1/2}} \right]^{1/2} \right] \right)^{-1}
\]

(2.38)

where \( M \) is the mass flow rate. In the plot, all the data points lay close to a line whose slope is 0.97, and thus agrees with Equation (2.38), even though this equation does not
consider the effect of the charges in the liquid. Therefore, Mutoh et al. (1979) concluded that in the liquid jet disintegration process, as well as in the liquid jet formation, the electrostatic field plays a role only in creating finer liquid threads, and does not take part in the disintegration of the liquid jet into droplets. In this thesis the effect of the applied potential on the liquid jet break up is addressed in Chapter 5.
2.4.3 Electrified liquid jet: kink instabilities

Liquid jets, under certain conditions of flow and surface charge, can start to whip from side to side, see Figure 2.6. In the literature, a number of terms have been used to describe this jet behaviour such as whipping, snaking, whip-like motion or kink instability, and a number of hypotheses have been formulated to explain why this phenomenon occurs. Taylor (1969) observed that the lateral oscillation of an electrified jet starts when the jet diameter stops decreasing. This happens when the aerodynamic tangential drag force becomes equal to the electrical and gravity forces. Cloupeau and Prunet-Foch (1988, 1994) stated that kink instabilities for a jet occurred when the electrostatic pressure on the jet surface approached the capillary pressure.

Figure 2.6 Liquid jet break up by kink instability.

Indeed, the presence of surface charges may promote stability when a liquid jet is subjected to varicose instability, depending on the wave number. However when large potentials are applied to the liquid jet, especially for liquids with high electrical conductivity, the jet can shift from varicose to kink instability. Son and Ohba (1998) and much earlier Bailey and Balachandran (1981) compared the growth rate factor for
varicose instability with that for kink instability, i.e. Equation (2.37) and (2.39), respectively.

\[
q_i = \frac{1}{\rho a} \left\{ \frac{ma_l(ma)}{I_1(ma)} - 1 \right\} \left\{ \frac{\sigma_q^2}{\pi a^3} \frac{maK_0(ma)}{K_1(ma)} - m^2 \gamma \right\}
\]

(2.39)

where \( q_i \) is the growth rate factor for lateral instability. They observed that as the electric field strength was increased, the jet surface charge increased, and the growth rate for kink instability was found to grow more rapidly than that for varicose instability. Thus, it is possible to find a critical value of the surface charge for which \( q_i \), the lateral growth rate factor, eventually overcame \( q \), the varicose growth rate factor, resulting in the propagation of jet whipping. When the jet break up occurs by kink instability, it is not possible to find a relationship between the wavelength and the number and size of droplets produced. Indeed kink instability is characterized by the random production of drops to form a polydispersed spray. Finally, it must be observed that kink instability is characteristic only of an electrified jet. In fact, when \( \sigma_q \) is equal to zero, \( q_i \) becomes zero, as in Equation (2.39).
2.4.4 Droplets and Satellites

Droplets are sometimes accompanied by satellites during the break-up of a liquid jet, see Figure (2.3). Satellite formation is a characteristic of both non-electrified liquid jet and electrified liquid jets, and occurs by the rolling up of the liquid neck present between two consecutive bumps (swells) that form between the main droplets. The linear theory proposed by Rayleigh allows a reasonably accurate prediction of the droplet diameter for an inviscid liquid, although it does not predict the actual jet profile close to the break up region, and it cannot be used to explain why, in the jet break-up, droplets and satellites are formed. Yuen (1968) gave a solution for the case of an inviscid liquid jets in the form of an infinite series until the third order term. This non-linear analysis could explain the formation of a ligament between two consecutive bumps. Goeddel and Yuen (1970) observed that when the liquid jet break-up takes place, the bump grows faster than the thinning of the liquid neck, thus revealing the non-linear nature that characterises liquid jet break-up. Despite this, the difference recorded between the bump growth and the neck thinning was linear thus explaining why the linear theory of Rayleigh works for inviscid liquids. However, the non-linear theory may describe the dynamics of a liquid jet break up and the existence of a ligament between two consecutive droplets. As already described, the disturbance on the liquid jet causes the bumps to grow and the neck to thin down until a point where the neck does not thin any more, at which point the bump begins to shorten. The surface between the ligament and the bump starts to locally contract, with the consequent formation of a high pressure gradient between the ligament and the bumps, inducing the break-up of the liquid jet, see Figure 2.7. Once the break-up has occurred, the produced droplet will move in the downstream direction, while the ligament will start to contract back in order to alleviate the high pressure at the pointed end; the pressure at the pointed end is much higher than the pressure at the lateral walls of the ligament. While the free pointed ligament contracts, the opposite end breaks apart from the jet and hence the ligament transforms into a droplet, usually smaller than the main one. This smaller droplet is called a satellite, see Figure 2.8. However, the above-described mechanism can change depending on the liquid properties, wave signal applied and jet electrification.
Rutland and Jameson (1970) studied both experimentally and theoretically, the break-up of a liquid jet. A non-linear theory was used in order to predict the volume of droplets and satellites, and they found that an increase in the wave number leads to a reduction in the diameter of both the droplets and the satellites, until a value of the wave number equal to 0.7, beyond which the theory predicted the total disappearance of satellites.

Figure 2.7 Mechanism of a liquid jet break-up and ligament formation (from Goeddel and Yuen, 1970)

The authors claim a good agreement between the theoretical data and the experimental results, although they observed that satellites were observed even for wave numbers larger than 0.7. The experiments were conducted using water as the liquid and small amplitudes of vibrations were induced on the liquid jet by loudspeakers. A third order analysis was also carried out by Lafrance (1975) who predicted the existence of both droplets and satellites. Although not clearly stated in his paper, Lafrance’s analysis predicted satellites until a wavenumber of 0.8, beyond which no satellites were predicted to occur. In agreement with the results of Rutland and Jameson (1970) a cut-off wave number was predicted for satellite formation. In contrast with Rutland and Jameson’s theoretical analysis, Yuen’s theory predicts the existence of satellites for any wave number (Chaudhary and Redekopp, 1980). These last authors, along with Chaudhary and Maxworthy (1980a, 1980b), investigated theoretically and
experimentally the possibility of determining a favourable configuration of the jet break-up in such a way as to eliminate satellite formation.

![Diagram of ligament roll up](image)

**Figure 2.8 Ligament roll up as described by Goeddel and Yuen, (1970)**

The idea was that of applying an initial perturbation comprising a fundamental and one or more higher harmonics to the liquid jet. Indeed, by applying a third harmonic perturbation to the fundamental perturbation, and varying the phase of the third harmonic, it was possible to give a different velocity to the droplet and satellite in such a way as to make them to collide and to coalesce after their formation. Changing the phase of the third harmonic in the right way caused either a rear-merging or a forward-merging drop. The former happens when the droplet moves faster than the satellite so that the satellite is captured by the droplet that follows it. The alternative case (a forward-merging drop) occurs when the satellite is faster than the droplet and in this case the satellite impacts on the droplet that precedes it. However, using a fundamental perturbation with an appropriate amplitude and wavenumber that was close to the value for which the growth factor is a maximum, and adding an out-of-phase third harmonic perturbation, it was possible to achieve liquid jet break-up without any satellite formation. The droplet was pinched off from the jet with a carrot-like shape with the sharp end relaxing back into the droplet just after the droplet formation. It is worth mentioning that, Chaudhary and Maxworthy (1980b) in their experiments observed that
using a fundamental perturbation alone with a large amplitude could also eliminate the formation of satellites.

2.4.5 Spraying by means of non-static electric field

Both the size and number of drops formed from a liquid jet depend greatly on the nature of the waves that are used to break up the jet. Sato (1984) achieved the production of uniform liquid droplets by the use of an AC potential. The experiment was performed using distilled water and aimed to investigate the effect of the applied voltage with four different wave types on the droplet diameter. The four wave types used were an AC sine wave, a rectified negative AC, a positive sine wave and a positive square pulse wave. Sato (1984) reported that when an applied rectified negative potential of 50 Hz was used, and the peak voltage was gradually increased, producing a reduction in droplet diameter until a point where both the droplet frequency and applied potential frequency were the same. This phenomenon was observed within a certain potential range, which named as the synchronous region. The same phenomenon was observed when using an AC sine wave potential with a frequency of 50 Hz, but in this case the droplet frequency recorded was double that of the applied potential. The synchronous region was observed for each of the wave types used, and at applied potentials beyond the upper limit of the synchronous region, dispersed non-uniform droplets were observed. The limits of the range in which the synchronous region was observed depended on the flow rate and the electrode geometry, where the distance between the tip of the nozzle and the earthed plate were found to be important. Sato also performed a number of experiments by modulating the frequency of the applied potential and he managed to produce uniform droplets with a diameter varying from 2.4 mm to 28 μm.

Balachandran et al. (1994) investigated the effect of an AC potential superimposed on a DC potential. Distilled water containing a zirconia precursor solution was used and the DC potential was varied between 0 and 10 kV whilst the AC potential was varied from 0 to 4 kV peak-to-peak at frequencies between 0.2 and 60 kHz. Two kinds of atomisation were investigated: cone jet atomisation and smooth jet
atomisation. It was found that in cone jet mode, using distilled water, the atomization was persistent only within a few restricted frequency bands, and within these bands synchronization between the applied AC frequency and droplet frequency was recorded. Increasing the flow rate and the DC potential allowed the formation of the smooth jet mode. In this mode it was possible to control the droplet diameter by changing the frequency of the applied AC potential in a drop size range between 650 μm and 350 μm, depending on the liquid flow rate. Good agreement was found between the measured droplet diameter and the value of the droplet diameter predicted by Sato’s formula (1984):

\[
D_d = \left( \frac{6Q}{\pi f_d} \right)^{\frac{1}{3}}
\]  

(2.40)

where \( f_d \) is the droplet frequency, \( Q \) the flow rate. In the smooth jet mode, the applied AC potential allowed not only the control of the droplet diameter but also of stabilizing the jet itself. In fact, by superimposing the DC potential on an AC potential, it was possible to return the jet to a varicose instability from a lateral instability that had been set up earlier by the application of the DC potential alone. More in-depth investigations on the smooth liquid jet break up by a DC potential superimposed with an AC potential were conducted by Huneiti et al. (1996, 1997, 1998). The work showed that, by varying the AC frequency, it was possible to change the wavelength of the jet surface wave within an interval ranging from a minimum wavelength of six times the jet radius to a maximum wavelength of nineteen times the jet radius, thus producing droplets with a diameter between 185 μm and 125 μm. According to them, the droplets were produced in a controlled fashion without secondary atomisation i.e. without satellite. Indeed the AC potential superimposed onto the DC potential had the effect of reducing the surface charge that was responsible for secondary atomization.

Barbet et al. (1997) investigated the break-up of a liquid jet, as well as droplet and satellite formation, by an electrohydrodynamic multimode jet stimulation device. The device consisted of a nozzle from which the liquid issued and had five steel rods positioned below the nozzle, see Figure 2.9. The highest, the lowest and the central rods as well as the nozzle were set at earth potential, whilst a high potential was applied to
the other two rods. On the rod, below the highest rod, a potential with a certain frequency, $f$, was applied, while on the rod below the central rod, a potential with a frequency, $2f$, was applied. All the experiments were performed for wavenumber below 0.5 and the liquid jet break up was driven by the two applied potentials. It was observed that, by changing the phase shift and the strength of the potential $V_2$ with respect to the potential $V_1$, it was possible to find a favourable condition under which the satellite could be eliminated by either forward or rear merging.

Figure 2.9 Schematic representation of the apparatus used in the experimental work of Barbet et al. (1997).
2.5 The effects of operating parameters and liquid physical properties on liquid dispersion

Electrohydrodynamic liquid spraying is greatly influenced by the liquid physical properties such as surface tension, viscosity, electric conductivity, dielectric constant, density and by the electrohydrodynamic liquid spraying parameters such as applied electric potentials and liquid flow rate. Each of these will now be dealt with in turn.

2.5.1 Electrical Conductivity

The electric conductivity of the liquid is one of the most important parameters upon which the electrohydrodynamic spraying of the liquid depends. Liquids are often classified with respect to their electrical conductivity, and labelled as either low, semiconductive or high electrical conducting liquids. Depending on the particular class into which the liquid falls, the specific mechanism of spraying may be predicted (Grace and Marijnissen, 1994). As seen in Equation (2.6), the charge relaxation time is given by the ratio of the dielectric constant to the electrical conductivity, and while the dielectric constant only varies over a small range of values (for water the relative dielectric constant is 80) the electrical conductivity may vary by many orders of magnitude. Thus, liquids with very low conductivity, of less than $10^{-11}$ S/m, such as toluene, hexane, silicones, Vaseline oils, etc. do not exhibit electrohydrodynamic behaviour because there are not enough charge carriers. However, it would appear that no upper limit to the liquid electrical conductivity is present for electrohydrodynamic spraying (Smith, 1986).

The conductivity of a liquid may be evaluated by measuring the current $I$ across a parallel plate capacitor with the liquid occupying the space between the plates. If the distance between the electrodes is $d_p$, the electrode surface area, $S_e$, and the potential applied to the electrode plates, $V_p$, then the electrical conductivity is found from the following expression (Paul and Nasar, 1987).
Hence, liquid conductivity is directly related to the amount of current conducted by the liquid. The mechanism of current conduction through a liquid is complex, though a major factor may be the concentration and position of free electrons and ions, gaseous and solid impurities. The current density, \( J \), is related to the density of ions present in the liquid by some function \( F \) as shown in Equation (2.42), where \( F \) includes parameters such as the ion mobility, \( u \), the ion recombination rate and the ion diffusivity, \( D_f \).

\[
J = F(e^-, n, u, D_f, E)
\]  

(2.42)

where \( n \) is the number of ions per unit of volume and \( e^- \) is the electron (Adamczewski, 1969). Ion mobility, ion recombination and ion diffusion are related to the nature of the ions themselves, as well as to the molecular structure of the liquid, the temperature, and the electric field strength. Indeed the molecular structure and the viscosity of the liquid can heavily influence the conduction of a current through a liquid. Ion mobility is related to the liquid viscosity by Equations (2.43) and (2.44). The former is called Stokes-Walden law, which is better suited to negative ions' mobility, while the latter is called the Adamczewski formula, which works best for positive ions' mobility.

\[
u_{SW} = A_c \mu^1
\]  

(2.43)

\[
u_A = A_c \mu^{3/2}
\]  

(2.44)

where \( A_c \) is a constant depending on the kind of liquid and ion species (Adamczewski, 1969). However, in both equations, as the viscosity increases, the ion mobility decreases, and thus the current conducted by the liquid decreases. For liquids with a viscosity of circa 9 mPa·s, the ion mobility measured is about 30X10^{-6} cm²/volt sec, while for liquid having a viscosity of 100 mPa·s, the ion mobility is of circa 3X10^{-6} cm²/volt sec. Another important parameter is the ion recombination rate. The ion recombination rate is the number of collisions of ions with opposite sign followed by
the neutralisation of their electric charges. The ions become either atoms or molecules that are electrically neutral overall. The coefficient of recombination, $\alpha$, is defined as follows:

$$\frac{dn}{dt} = -\alpha_n n^2$$  \hspace{1cm} (2.45)

where $\alpha_n$ has a dimension of cm$^3$/sec. The number of ions in a liquid after a certain time, $t$, can be evaluated as shown below:

$$n = \frac{n_0}{1 + \alpha_n n_0 t}$$  \hspace{1cm} (2.46)

where $n_0$ is the initial number of ions. The viscosity also influences the ion recombination rate such that an increase in viscosity leads to a decrease in the ion recombination rate. Thus, in viscous liquids, the number of ions is more stable than those in inviscid liquids. Finally, another important factor is the ion diffusivity, $D_f$. This is related to the ion mobility by the Nernst-Einstein formula as

$$\frac{n}{D_f} = \frac{Ne}{9RT}$$  \hspace{1cm} (2.48)

where $Ne$ is the Avogadro number, $R$ is the molar gas constant and $T$ is the temperature of the liquid. The ion diffusivity is viscosity dependent such that an increase in the liquid viscosity produces a reduction in the ion diffusivity. Therefore, there is a close relationship between the electrical conductivity and the viscosity of a liquid.

The liquid conductivity also depends on the applied electric field strength. In fact, the current conducted through a liquid is not proportional to the potential applied, rather it follows a law as depicted in Figure 2.10. An increase in the electrical conductivity is observed when the potential across the liquid is increased, due to the dissociation of liquid molecules into their constituent ions induced by the strength electric field itself (Adamczewski, 1969).
The tangential stresses acting on the liquid jet depend only on the electrical conductivity $\sigma$, see Equation (2.10) and its effect on the cone jet mode and droplet diameter can be summarised as follows: the higher the electrical conductivity of the liquid, (i) the lower the potential required for the cone jet mode formation, (ii) the smaller the mean diameter of the droplet, and (iii) the lower the flow rate required for the cone jet mode formation. In addition, an increase in the electrical conductivity of the liquids leads to a narrower distribution of the droplet diameter produced (Smith, 1986; Bailey and Balachandran, 1981 Hayati et al. 1987b).

![Graph](image)

**Figure 2.10** The qualitatively variation of the current measured through a liquid versus the applied potential (Adamczewski, 1969).

### 2.5.2 Viscosity

The effect of the liquid viscosity on droplet formation is complex. When the viscosity of liquid increases, there is an attenuation of the propagated wave along the liquid jet surface and the classic wave break-up mechanism gradually disappears. This effect can be attributed to the fact that a liquid with high viscosity requires more energy to deform than one with low viscosity. Several authors agree that there exists a relationship between the liquid viscosity and the droplet diameter produced. However, there is a disagreement on the exact relationship between viscosity and droplet diameter.

2.5.3 Dielectric constant

The polarisation of a liquid can result from three different sources: electronic, ionic and orientation. Electronic polarisation occurs when an externally applied electric field induces a shift in the mean position of the positive and negative charges of a neutral atom. Ionic polarisation occurs in molecules composed of both positive and negative charged ions. Once again the application of an electric field results in ions' movement, and leads to the splitting of the molecule if the electric field gradient is high enough. Orientation polarisation occurs in materials that possess permanent molecular dipoles. In the absence of an electric field, the dipoles are randomly orientated and the dipole moment is zero. The presence of an externally applied electric field induces all the permanent dipoles to re-orientate themselves and align with the applied field. These polarisation mechanisms may also be explained in terms of the effects associated with the charges at the liquid surface and in the bulk of the liquid (Paul and Nasar, 1987).

The dielectric constant also depends on the intensity of the applied electric field applied and by its frequency. Huneiti (2000) observed that the dielectric constant of water may be significantly reduced by electric fields of the order of $10^9$ V/m as well as at frequencies of the order of several gigahertz. The dielectric constant is therefore not a true constant at all, and a better term is the permittivity of the liquid.

The dielectric constant may influence the liquid atomization by influencing the amount of current conducted through the liquid jet as reported by Hartman (1998). However, the influence of the dielectric constant on the current conducted through the jet is quite contained if compared with the effect of the applied potential and the ions' species.
2.5.4 Surface tension

The surface tension of a liquid is due to molecular attraction, and it plays an important role in Electrohydrodynamic liquid spraying. A high surface tension would make the liquid more difficult to deform and break up. In particular for cone jet mode formation, a potential high enough to overcome effects of the surface tension is required such that dielectric breakdown of the air occurs (Smith, 1986). The effects of the surface tension on the droplet diameter are not well understood. According to Weber’s formula (Equation (2.33)), an increase in the surface tension leads to a smaller droplet diameter, but Gañana-Calvo et al. (1997) did not report any influence of the surface tension on the droplet diameter produced although the surface tension was still a parameter in their scaling law. Harman (1998) observed that an increase in surface tension must have been responsible for an increase in the current transported through the jet. Otherwise a higher potential than used in his experiments would have been required. This agrees with the conclusion reached from Equation (2.24).

2.4.5 Flow rate

The shift from one electrohydrodynamic liquid spraying mode to another is heavily dependent on the actual liquid flow rate. For cone jet mode formation, a minimum flow rate is required and an increase in flow rate can provoke an increase in jet diameter and thus in droplet diameter produced. In dripping mode, as in jet mode, an increase in flow rate leads to the production of larger droplets (Mutoh et al. 1979; Hayati, 1987a; Jones and Thong, 1971).

2.4.5 Applied electric potential

The applied electric potential is the most important parameter of electrohydrodynamic spraying because it is responsible for producing the electric stresses on the liquid. The electric stress also depends on the electrode geometry, and thus allowing to generate all the well-know electrohydrodynamic spraying functioning modes. In addition, the applied potential, and thus the electric field generated, may
change the physical properties of the liquid sprayed. Andrade and Dodd (1945) extensively investigated the effect of an electric field upon the liquid viscosity. The researchers concluded that if the liquid did not have any dipole moment then its viscosity was not affected by an electric field. But if the liquid was polar, then there were two possible classes: (a) polar liquids with very low conductivity which did not show any viscosity changes and (b) conductive polar liquid, either pure or by the addition of moisture, that would show a viscosity change. In the latter kind of liquid, the viscosity started to change gradually as the potential was increased until a certain magnitude over which there was a phenomenon of saturation and no further viscosity increase was recorded; the electric field used was of the order of 40 kV/cm. The apparatus used by the authors consisted of a number of parallel plates, between which the liquid flowed; a potential difference was applied between two parallel plates. The increase of the liquid viscosity was attributed to the accumulation of ions and molecules into clusters close to the electrode surface. The clusters reduce the space available for the liquid to flow. This was thus interpreted as an increase in viscosity. Indeed, for the researchers to understand whether there was truly a change in the viscosity of the liquid by the effect of an electric field, it was necessary to examine the effect of the same liquid under an alternating electric field. The alternating field prevents cluster formation at the electrodes, allowing the relationship between electric field and liquid viscosity to be examined. Andrade and Dodd (1950) observed that an increase in the frequency of the alternating field leads to the disappearance of the large viscosity effect as soon as the frequency arose above 1000 Hz. However, the authors concluded that the electric field did have an effect on the liquid viscosity of polar liquids, though the effect was quite contained.

The applied potential difference can also change the surface tension, Sato et al. (1998). Sato et al. (1998) used the vibrating jet method to observe that a high enough electric field was able to reduce the liquid surface tension of distilled water by about 50%. The surface tension reduction was proportional to the square of the applied potential, and it was found that the reason for the surface tension reduction was the surface charge density. Huneiti (2000), reviewing the work of Schmitt, stated that the reduction of the water surface tension was due to the orientation of the molecular dipoles induced by the applied electric field. The molecules repelled each other, so reducing the surface energy and hence the surface tension.
Chapter 3 – Experimental Arrangements

3.1 Spraying arrangements

The apparatus used to perform the experiments is a simple configuration consisting of a stainless steel nozzle positioned in vertical direction below which is placed a metallic annular disk. A high voltage supply (EHT) is attached to the nozzle, while the metallic disk is earthed. The liquid is pumped to the nozzle using a syringe pump Harvard Apparatus 22 Model 55-2222, which allows to feed the liquid into the system with a wide range of flow rate. The flow rates used in this work ranged between 1 to 60 ml/min. A number of sets-up have been used, and all the electrode configurations are reported in Table 3.1. The characteristic dimensions used in the experiment such as the nozzle diameter, the disk hole diameter and the distance between the nozzle tip and the disk surface have been chosen by experimental investigations. Two power supplies have been used: a high voltage source Hartley Measurements model 421 and a Nada Electronics model 2300. The former was used to apply a high constant DC voltage, while the latter was used to generate a high pulsating DC voltage. Both devices have positive polarity. The DC voltage was varied between 0 to 20 kV, while the peak of the pulsating DC voltage was varied between 4 to 8 kV with a frequency in the range 30 - 300 Hz. The spraying apparatus was enclosed in a metallic cage with Perspex transparent walls that were covering the left and the right side of the cage, while the front and the rear wall of the cage were left open for observation.

The apparatus dimensions were 40 cm long, 40 cm wide and 85 cm high. Protection from the high voltage was provided by the earthed cage and the Perspex transparent wall, which allowed the process close to the nozzle to be observed, see Figure 3.1. The spraying process was recorded using a high-speed digital video camera model Kodak Ektapro HS 4540 which provided frame rate up to 40500 fps in black-and-white. A Leica Monozoom 7 lens was used. The droplet formation in dripping mode and jet mode may form within a time of the order of milliseconds. The framing rate was therefore adjusted with respect to the characteristic time of the event. The
frames were stored in memory. Each full frame measures 256 x 256 pixels and at 40500 fps the frame size is reduced to 64 x 64 pixels. The required light intensity depends on the frame acquisition rate, however it became significantly dependent only beyond 4500 fps. The camera could store 3072 full frame images, which were transferred to a PC by a Digital Download Interface (DDI). This interface allowed control of the camera remotely. The images were downloaded in Tagged Information File Format (TIFF) via IEEE protocol GPIB-PCII(A) interface card. Halogen lamps were used for lighting the rig, illuminated either the front or the rear for conventional or shadow photography respectively.

Figure 3.1 Experimental set up used in this work.
Each experiment took only a few minutes and hence avoided a long exposure of the jet to the light, which could change the viscosity of the liquid by heating it up. At the end of each experiment the droplets occasionally accumulated on the lateral Perspex transparent wall were wiped off. The recorded images were downloaded into a PC and later processed by software called “Optimas” for the determination of droplet size and formation frequency. The experiments were conducted at room temperature.

Table 3.1 Electrode configurations.

<table>
<thead>
<tr>
<th>Electrode configuration</th>
<th>Nozzle diameter</th>
<th>Distance between nozzle and earthed electrode (mm)</th>
<th>Hole diameter (mm)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>I.D. (mm)</td>
<td>O.D. (mm)</td>
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<tr>
<td>1A</td>
<td>1.5</td>
<td>4</td>
<td>25</td>
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<td></td>
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<td>30</td>
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<tr>
<td>1B</td>
<td>1.5</td>
<td>4</td>
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<td>1D</td>
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<td>2B</td>
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<td>15</td>
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</table>
3.2 Methods for drop sizing

A number of methods have been developed by many researchers over the years to characterize the droplets produced from a spray. These methods can broadly be classified as mechanical, optical and electrical and have their own advantages and disadvantages. The principal problems which can be met when one attempts to characterize a spray may be:

1. the large number of droplets produced;
2. the wide range of droplet size making a spray;
3. the varying droplet velocity; and
4. the change of droplet size due to either evaporation or to droplet-droplet coalescence;

Beside the above problems, there may be others rising from the kinds of technique used, e.g. when using optical methods, the coating of the optics may be a problem. So all the above factors have to be taken into account when choosing a method of characterisation. The characterisation method needs to have some features to be suitable for spray characterization. The characterisation method should not disturb the spraying process, and it should have a wide range of sizing capability. It should provide the measurement of a representative sample in a relatively short time, and finally it should not be too much affected by the variation of the ambient air properties where the spraying is performed. In the following, a number of possible sizing methods which may be used to characterize the droplet size in this work are described.

(a) Collection of drops using slides. In this technique, a slide made of glass is used to collect liquid droplets from a spraying. The droplets are collected on the slide surface which has been previously coated with a material, e.g. soot or magnesium oxide, in order to highlight the droplets. These droplets are subsequently counted and measured. The main disadvantage of this method is the possibility of overlapping between droplets and the collection efficiency for small droplets; small droplets may
evaporate in a very short time. It may also be difficult, to determine a correction factor between the diameter of the droplets sitting on the surface and the diameter of the droplet before impacting on the surface.

(b) Collection of drops in cells. In this technique, the droplets produced by spraying are collected in a cell in which a liquid with suitable properties permits droplets to keep their spherical shape and prevents their evaporation. The main disadvantage of this method is droplet splashing if it is too large, and drop-drop coalescence which is difficult to identify.

(c) Molten wax. This method is based on the spraying of molten wax into air. The wax once sprayed will quickly solidify. A sample is then collected and the solidified droplets are analysed by sieving. The major disadvantage of this technique is that the molten wax used to perform the spraying may have physical properties which are very different from that of the practical spraying liquid commonly used.

(d) Drop freezing. This method is very much similar to the molten wax. In fact, it consists of freezing the produced droplet by cooling with liquid nitrogen, and then collecting the solidified droplets and analysing them by sieving. Although this technique allows the analysis the droplet, it has some disadvantages. The amount of liquid nitrogen required may create safety problems such as room ventilation and handling. Besides, there are others problems such as the increase in size of the frozen droplets, the change of the droplet sphericity and the handling of the frozen droplets. In this case, the main problems are given by droplet swelling, droplet deformation and frozen droplet handling. These phenomena are clearly shown in Figure 3.2 and 3.3 for droplets with a core made of water and a shell made of oil dripped in liquid nitrogen. The droplets are instantly frozen as soon as they touch the liquid nitrogen. Because of the swelling of the water droplets, cracks are propagating just after a few seconds on the shell, this is completely blown away after no more than a minute. This mechanism is depicted in Figure 3.4. Furthermore the frozen droplets are not completely spherical, due to the impact of the droplets with the liquid nitrogen surface.
Figure 3.2 Droplet frozen by liquid nitrogen.

Figure 3.3 Droplet frozen by liquid nitrogen; (a) just after freezing, (b) after a few minutes.

Figure 3.4 (a) Pressure generation due to the increase of volume by freezing; (b) breakage of the frozen oil shell.
In addition, after a few minutes the droplet surface started to be covered by ice crystals due to the crystallization of the moisture present in the air on the cold surfaces of the frozen droplets. This is shown in Figure 3.3a and 3.3b (arrows). Although the technique of drop freezing is quite an elegant method of droplet sizing, it has got some disadvantages, as seen, that are difficult to overcome.

(e) The Malvern particle sizer is a commercial instrument, which permits rapid measurement of the droplet size distribution. The device is based on the Fraunhofer light diffraction produced by a droplet. The diffraction pattern consists of a number of concentric rings, with the gaps between the rings depending on the droplet diameter. Thus, from the shape of the Fraunhofer diffraction, it is possible to calculate the droplet size. Using the same principle, it is possible to determine the size distribution of a number of droplets when the diffraction is due to several droplets. However, a spray may be made of a number of droplets having different diameter, and each droplet or group of droplets having the same size produce a given diffraction pattern. Hence, the resulting Fraunhofer pattern of a light beam throughout a spray is the result of the overlapping pattern produced by each droplet size category. In addition, when the light goes through the spray it may get scattered by more than one droplet. So, all the above-described phenomena may induce errors in determining the size distribution of produced droplets. Therefore, particular algorithms are used to get the right size distribution of the droplets. However, despite the technique being widely used, it has got some disadvantages such as inaccuracy in measuring droplets quite as large as the one produced in this thesis (above 2 mm), and inadequacy in measuring droplets that are not very spherical. Large droplets may deform when travelling along their trajectory after a droplet-satellite collision.

(f) High speed photography. The use of high speed photography in the study of the development and features of water droplets was carried out in the early 1890 by the English Physics Professor A. M. Worthington (1963). In 1894 he gave the first lecture entitled "The Splash of a Drop and Allied Phenomena," which featured drawings of water and mercury splashes. In his lecture, Worthington described various stages in the life of a mercury drop falling against a glass surface, as well as the phases of a water drop on a liquid surface made of milk. The succession of photographs was made by creating time after time droplets of definite sizes and releasing these droplets from
definite heights. To freeze each falling droplet for a time long enough to be seen, lighting by electrical discharges was used. In 1908, Worthington published a book entitled “A Study of Splashes” in which there were about 200 sketches illustrating drops and splashes. Hence, for the first time the different stages of a splashing droplet were observed and investigated. High speed photography techniques consist of recording images of one or more droplets which occur in a short interval of time. The images may be made by using a number of light flashes with an appropriate intensity and life time while holding the camera shutter open (in the dark) before the flash discharge starts. Another way to take high speed photography is by using a camera having a high shutter speed, while holding the light constantly on. Technically, the former technique is easier to implement. However, in both cases, an instantaneous image of the process is recorded on a picture. This kind of technique, used to record a spraying process would give what is called spatial sampling. High speed photography may also be used to make a temporal sampling. In this case, it is needed to record a number of pictures in a given interval of time. It is then possible to convert all the spatial sampling in to temporal sampling.

Since Worthington’s work, high speed photography has been extensively used to study liquid evolution as well as a method of droplet sizing. In this work, high speed photography has been used as a method of droplet sizing for a number of reasons. Firstly, the technique has the advantage of being non-intrusive, and relatively easy and quick to set up. Secondly, for safety reasons, the spraying is performed in a metallic cage with Perspex walls because of the potential danger due to the value of the current used; current above 300 mA can be lethal. Thus, with this kind of system configuration, the high speed photography seems to be more suitable. Thirdly, it is possible with the same set-up to record liquid spraying modes as well as to measure size for the droplets produced.
3.3 Electric field used

In electrohydrodynamic spraying, the choice of the electrode geometry, for example, the gap between the electrodes and the shape of the electrode is a key factor. This is because the interelectrode distance and the electrodes’ shape affect the electric field strength and configuration. If the electrodes are placed in a gaseous atmosphere e.g. air, the electric field present between the electrodes may induce electric discharges. A gas usually contains a small amount of matter, with respect to a solid, and it may be considered as a good insulator. Nevertheless, if the electric potential is increased beyond a certain limit, electric breakdown occurs due to ionisation of the gas. The value of the potential at which dielectric breakdown occurs is given in equation 3.1.

\[ V_{sp} = F(pd_e) \]  

(3.1)

where \( V_{sp} \) is the potential at which sparking starts, \( p \) is the gas pressure and \( d_e \) is the gap distance (Meek and Craggs, 1953). This law is known as Paschen's Law. For uniform field and for gaps the above law in air and has been worked out as follows:

\[ V_{sp} = a_{11}d_e p + a_{22} \sqrt{d_e p} \]  

(3.2)

where \( a_{11} \) and \( a_{22} \) are constants. In a non-uniform electric field as the one used in this thesis, Equation (3.2) may assume a more complex form depending on the shape of the electrode, as well as on other factors, e.g. gas impurities, dust, surface irregularities, relative humidity and radiation. For these reasons, the set-up distances between the electrode have been chosen experimentally. However, Equation (3.2) can help determine the most suitable distance between the electrodes. In a uniform electric field the following relationship among voltage, distance and electric field, \( E \), is valid:
\[ V = E d_e \quad (3.3) \]

Therefore Equation 3.2 can be rearranged as

\[ E = a_{11} p + a_{22} \frac{\sqrt{p}}{d_e} \quad (3.4) \]

which gives the relationship between the electric field, the electrode distance and the gas pressure in an uniform electric field (Roth, 1995). From Equation (3.4) a high electric field may be reached before sparking occurs, when the distance between the electrodes is very close. Hence, if a high electric field is needed then the needle has to be placed close to the earthed plate.

Another aspect, which is important is the effect of the polarity on the dielectric breakdown. Positive discharge needs a higher electric field than that needed for negative discharge (Meek and Craggs, 1953; Roth 1995). In this work, experiments have been performed with highly viscous liquids, and with these liquids, the atomisation can be driven as long as the electric field has an appropriate strength. Therefore, electric potential with positive polarity has been used in order to have the possibility of applying the highest electric field for a given electrode geometry. The electric potentials used are uniform DC and a pulsating DC. For pulsating DC, two characteristic pulse shapes are reported in Figure 3.5 and 3.6. The pulses have been recoded with a digital oscilloscope, Lecroy 400. As it can be observed for each new cycle both the pulse shapes have got a high peak on 1X10^{-4} seconds. The effect of this peak on the liquid jet break up will be commented in Chapter 6.
Figure 3.5 Record of a 100 Hz pulse shape.

Figure 3.6 Record of a 200 Hz pulse shape.
3.4 Spray analysis

There are two possible ways to acquire a droplet size distribution. These are spatial sampling and temporal sampling. The latter has been used in this work to characterise the droplet size distribution. As it has been seen, the temporal sampling gives the size distribution of the droplets produced within an interval of time. The interval of time over which the sampling is performed needs to be specified, and this depends on the number of droplets necessary to give a representative sample of droplets with statistical reliability.

In this work about 500 images of droplets and satellites have been taken to determine size distributions. These images have been downloaded into a PC, and later processed by the software Optimas. Because of the large number of data acquired, a macro is developed in Optimas software to measure the diameter automatically. The macro has been written in ALI language and it is reported in Appendix B. The number of images used to measure the average diameters of droplets and satellites is based on statistical analysis, as detailed in Appendix A. A balance between the data accuracy and the time required to get the data leads to the decision of choosing 500 images. When the spraying experiment is performed, it usually takes 3-5 seconds in which the images of droplets and satellites are recorded. Then, the images are downloaded onto a PC for processing. The time for downloading is about 3 seconds for each image, while the time for processing an image is about 2 seconds. Hence each experiment which determines a data point takes 1 hour. A graph is usually made of a number of data points, varying between 15 to 40 thus requiring a time from about 15 to 40 hr. Therefore the necessity of deciding which is the suitable number of droplets for the measurements of each data point is evident here. 500 droplets assure a reasonable accuracy in the calculation of the droplet and satellite diameter, see Appendix A. In Figure 3.7 to 3.10 the same droplet distribution acquired with a decreasing number of pictures are reported. It can be noticed that the use of 500 images gives a representative droplet size distribution compared to either 1000 or 2000.
Figure 3.7 Droplet size distribution obtained by processing 2000 images.

Figure 3.8 Droplet size distribution obtained by processing 1000 images.
Figure 3.9 Droplet size distribution obtained by processing 500 images.

Figure 3.10 Droplet size distribution obtained by processing 250 images.
3.5 Polyvinyl aqueous solutions

Previous researchers had studied the effects of an applied electric field on the atomisation of many liquids. Grace and Marijnissen (1994) presented a table reporting the physical properties of a large number of liquids that have been studied in this way. A number of the above liquids and their physical properties such as electrical conductivity, viscosity and surface tensions are shown in Figure 3.11. A lot of work has been done with liquids having an electrical conductivity between $10^{-7}$ to $10^{-2}$ S/m and viscosities ranging from a fraction to 10000 of that of water. For electrical conductivities above $10^{-2}$ S/m, as well as below $10^{-7}$ S/m, the published work only considers liquids with viscosities of a few times that of the water. Also, a large majority of the liquids have a surface tensions between 20-40 mN/m. A large amount of work has been done using pure liquids, mixed liquids and doped liquids to explore the effect of physical properties such as electrical conductivity, surface tension and viscosity on liquid atomisation. Many of the liquids are organic, while inorganic liquids such as water, water solutions and slurry represented a minor fraction of those studied.

Recently, the sector of the chemical industry dealing with paints, adhesive and particle production has looked with increasing interest on water based solutions because of the low cost of solvent and its environmental compatibility. Water solutions used in the chemical industry, either as an intermediate or a final product may be the result of a complex mixing component with respect to certain product specifications. Therefore, these solutions may have complex rheological properties and viscosities of a few orders of magnitude higher than water. The high viscosity of a liquid may represent a non-trivial problem to overcome in all those industrial processes where the control of the liquid dispersion is a key factor.

Based on this, it is worth exploring the possibilities and limitations of controlling the atomisation of highly viscous aqueous solutions by the assistance of an applied electrical field. The possibility of effectively atomising a liquid using an applied electric field depends on its physical properties, e.g. electrical conductivity, surface tension, permittivity, viscosity, the electrode geometry of the spraying system, the flow rate condition as well as the type of electrical signal.
Fig 3.11 Viscosity and conductivity for a number of liquids used in the literature.
Table 3.2 Legends used in Figure 3.11

<table>
<thead>
<tr>
<th>No.</th>
<th>Legend Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>Acetone*</td>
</tr>
<tr>
<td>2</td>
<td>Cyclohexanol*</td>
</tr>
<tr>
<td>3</td>
<td>Cyclohexane with MCH Copolymer*</td>
</tr>
<tr>
<td>4</td>
<td>Cyclohexane with MCH Copolymer*</td>
</tr>
<tr>
<td>5</td>
<td>DiButyl phthalate*</td>
</tr>
<tr>
<td>6</td>
<td>Dichloroethane with perchlorovinyl*</td>
</tr>
<tr>
<td>7</td>
<td>Dichloroethane with polystyrene (a)*</td>
</tr>
<tr>
<td>8</td>
<td>Dichloroethane with polystyrene (b)*</td>
</tr>
<tr>
<td>9</td>
<td>Dimethylacetamide with polyacrylonitrile*</td>
</tr>
<tr>
<td>10</td>
<td>Dimethyl phthalate*</td>
</tr>
<tr>
<td>11</td>
<td>Ethanol*</td>
</tr>
<tr>
<td>12</td>
<td>Ethanol with BF-2 glue (b)*</td>
</tr>
<tr>
<td>13</td>
<td>Ethanol with BF-2 glue (a)*</td>
</tr>
<tr>
<td>14</td>
<td>Ethanol with polyvinylpyrolidane (a)*</td>
</tr>
<tr>
<td>15</td>
<td>Ethanol with polyvinylpyrolidane (b)*</td>
</tr>
<tr>
<td>16</td>
<td>Formamide with 1M LiCl*</td>
</tr>
<tr>
<td>17</td>
<td>Freon 113*</td>
</tr>
<tr>
<td>18</td>
<td>Glycerol*</td>
</tr>
<tr>
<td>19</td>
<td>Heptane with stedis 450 (0.3% wt)*</td>
</tr>
<tr>
<td>20</td>
<td>Hexane*</td>
</tr>
<tr>
<td>21</td>
<td>Isopar M</td>
</tr>
<tr>
<td>22</td>
<td>Isopar M with 5% butanol*</td>
</tr>
<tr>
<td>23</td>
<td>Isopar M with 10% butanol*</td>
</tr>
<tr>
<td>24</td>
<td>Isopar M with 15% butanol*</td>
</tr>
<tr>
<td>25</td>
<td>Isopar M with 20% butanol*</td>
</tr>
<tr>
<td>26</td>
<td>Isopar M with 25% butanol*</td>
</tr>
<tr>
<td>27</td>
<td>Isopar M with 30% butanol*</td>
</tr>
<tr>
<td>28</td>
<td>Isopar M with 35% butanol*</td>
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<tr>
<td>29</td>
<td>Isopar M with 40% butanol*</td>
</tr>
<tr>
<td>30</td>
<td>Isopar M with 45% butanol*</td>
</tr>
<tr>
<td>31</td>
<td>Isopar M with 50% butanol*</td>
</tr>
<tr>
<td>32</td>
<td>Methylchloroform with perchlorovinyl (a)*</td>
</tr>
<tr>
<td>33</td>
<td>Methylchloroform with perchlorovinyl (b)*</td>
</tr>
<tr>
<td>34</td>
<td>Propanol*</td>
</tr>
<tr>
<td>35</td>
<td>Propanol with HCl (0.05%)*</td>
</tr>
<tr>
<td>36</td>
<td>Propanol with HCl (0.125%)*</td>
</tr>
<tr>
<td>37</td>
<td>Propanol with HCl (0.25%)*</td>
</tr>
<tr>
<td>38</td>
<td>Propanol with HCl (0.5%)*</td>
</tr>
<tr>
<td>39</td>
<td>Propanol with NaI (0.618 gm/l)*</td>
</tr>
<tr>
<td>40</td>
<td>Toluene*</td>
</tr>
<tr>
<td>41</td>
<td>Distilled water*</td>
</tr>
<tr>
<td>42</td>
<td>Water with 0.5% Hodag*</td>
</tr>
<tr>
<td>43</td>
<td>Water with NaCl (0.01%)*</td>
</tr>
<tr>
<td>44</td>
<td>Water with NaCl (0.05%)*</td>
</tr>
<tr>
<td>45</td>
<td>Water with NaCl (0.1%)*</td>
</tr>
<tr>
<td>46</td>
<td>Water with polyethylene oxide*</td>
</tr>
<tr>
<td>47</td>
<td>Water with polyvinyl alcohol*</td>
</tr>
<tr>
<td>48</td>
<td>Span 20 blend Span 85 (a)**</td>
</tr>
<tr>
<td>49</td>
<td>Span 20 blend Span 85 (b)**</td>
</tr>
<tr>
<td>50</td>
<td>Span 20 blend Span 85 (c)**</td>
</tr>
<tr>
<td>51</td>
<td>Span 20 blend Span 85 (d)**</td>
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<tr>
<td>52</td>
<td>Span 20 blend Span 85 (e)**</td>
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<tr>
<td>53</td>
<td>Span blend ethanol 1%(a)**</td>
</tr>
<tr>
<td>54</td>
<td>Span blend ethanol 1%(b)**</td>
</tr>
<tr>
<td>55</td>
<td>Span blend ethanol 2%(c)**</td>
</tr>
<tr>
<td>56</td>
<td>Span blend ethanol 2%(d)**</td>
</tr>
<tr>
<td>57</td>
<td>Span blend ethanol 2%(e)**</td>
</tr>
<tr>
<td>58</td>
<td>Span blend ethanol 2%(f)**</td>
</tr>
<tr>
<td>59</td>
<td>Span blend ethanol 2%(g)**</td>
</tr>
</tbody>
</table>

(* Grace and Marijnissen, 1994; **Fourches, 1999 and Fry, 1995-1996)
With such a large number of variables involved, the mapping out of all the interactions between the variables is impractical. Therefore, the field of research needs to be narrowed down by fixing some of the variables.

With respect to the physical properties of the liquids, the commercial interest is on the viscosity. Therefore, the ideal model liquid for this study would be a liquid with a constant surface tension and electrical conductivity, but with a variable viscosity. One possible system is a solution of polyvinyl alcohol in water where the viscosity can be changed with the concentration of PVA. This polymer, see Formula (3.1), is soluble in water and its solubility is related to its degree of polymerisation.

\[ -\text{[CH}_2\text{CHOH]}_n- \]  

Formula (3.1)

The great affinity of polyvinyl alcohol for water is due to the presence of the hydroxyl group, see Formula (3.1). However, the hydroxyl group is also responsible for creating a strong hydrogen bonding within other hydroxyl groups belonging to the same polymer chain, as well as to others polymer chains. The presence of these bonds may significantly reduce the solubility of the polyvinyl alcohol in water. Therefore, in order to reduce the strength of this bond in the polyvinyl alcohol and hence to further increase the solubility of the polymer in water, acetate groups are left on the chain, see Formula (3.2).

\[ -\text{[CH}_2\text{CHOH]}_n-\text{CH}_2\text{CHCOO-CH}_3- \]  

Formula (3.2)

The acetate groups are hydrophobic and reduce the strength of the hydrogen bonding, thus increasing the polymer solubility. The greater the number of the acetate groups on the polyvinyl alcohol chain the lower the degree of hydrolysis, with a subsequent increase in the solubility of the polymer in water. The solubility of the polyvinyl alcohol, as a function of the degree of hydrolysis in mol% at several temperatures is shown in Figure 3.12. Water above 80 °C can fully dissolve polyvinyl alcohol with any degree of hydrolysis.

The working liquid solutions used in the experiments have been prepared using polyvinyl alcohol Airvol 325, kindly provided by Air Products Plc. Airvol 325 is a mix
of polyvinyl alcohol with chain lengths between 1000-1500 Daltons. The solutions were prepared then by heating the polyvinyl alcohol powder in distilled water for 30 minutes at 96 °C, while stirring. The solutions once prepared were left for one week to rest and

Figure 3.12 Solubility of the polyvinyl alcohol in relation to its degree of hydrolysis (from Finch, 1992).

<table>
<thead>
<tr>
<th>Liquid</th>
<th>Electrical Conductivity (S/m)</th>
<th>Viscosity (mPa s)</th>
<th>Surface tension (mN/m)</th>
<th>Density (kg/m³)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Distilled water</td>
<td>0.0001</td>
<td>1</td>
<td>72</td>
<td>1000</td>
</tr>
<tr>
<td>Surrey Tap water</td>
<td>0.1</td>
<td>1</td>
<td>72</td>
<td>1000</td>
</tr>
<tr>
<td>PVA 3% wt</td>
<td>0.048</td>
<td>40</td>
<td>62</td>
<td>1002</td>
</tr>
<tr>
<td>PVA 5% wt</td>
<td>0.093</td>
<td>140</td>
<td>58</td>
<td>1009</td>
</tr>
<tr>
<td>PVA 7% wt</td>
<td>0.124</td>
<td>280</td>
<td>57</td>
<td>1016</td>
</tr>
</tbody>
</table>

Table 3.3 Physical properties of the polyvinyl solutions used in the present work.
then filtered. The physical properties of the solutions prepared in this way were measured at room temperature, which was 20 °C, and their values are reported in Table 3.3. N.B. the electrical conductivity has been gathered at low field strength, and the viscosity as well as the surface tension have been measured in absence of electric field.

The electrical conductivity of the solutions was measured using a conductivity meter (4310 Jenway). The surface tension was measured with the method based on the pendant drop, using a contact angle measuring instrument (A G10 Kruss GmbH). The viscosity was measured using a cone-and-plate rheometer (Carri-med 50 rheometer TA instruments). The density was measured using a volumetric flask.
The polyvinyl aqueous solutions have high values of electrical conductivity and viscosity. Their physical properties are reported in Figure 3.11 for comparison with the other liquids used. Polyvinyl aqueous solutions are generally non-Newtonian, and it has been observed that the viscosity can change over time, especially for solutions with a low concentration of PVA. According to Naito et al. (1980) and more recently reviewed by Finch (1992), for dilute aqueous solutions of polyvinyl alcohol (about 5 g/l) and low shear rates (typically 400, 500 s⁻¹) the rheological behaviour can be considered virtually Newtonian, see Figure 3.13. The viscosity of all the solutions used was measured again after four months and after twelve months. The measured changes in the viscosity over these time period were within the error of the viscometer; < 5%.
3.6 Shadow photography

In this work, the objects of interest are droplets, liquid jets and pendant droplets made of either water or PVA aqueous solutions. The shadow photography, as shown in Figure 3.14, is very suitable here because it allows one to distinguish neatly the edge of the liquid jet being sprayed, and so helps to better understand its development and mechanism.

![Figure 3.14 Refraction of the light rays through a water droplet.](image)

A liquid droplet transparent to the light behaves like a lens. When the light rays hit the air-water interface they are refracted and sent as focus shown Figure 3.14. Thus, a cone of shadow is made just behind the droplet. If the droplet image is taken on the plain A-A which is on the local plane, then the droplet image looks like in Figure 3.15.

![Figure 3.15 Shadow image of a droplet of PVA 3%.](image)
The image allows to distinguish neatly the droplet edge and the focus, which is the white spot positioned just at the centre of the droplet. The set up used for the shadow photography is shown schematically in Figure 3.16.

Figure 3.16 Set up used for the recording in shadow photography.
Chapter 4 - Dripping Mode

4.1 Introduction

The dripping mode takes place when a liquid is fed to the nozzle with a small flow rate and drops are released individually from the tip of the nozzle. In this mode, the application of an adequately strong electric potential to the nozzle can increase the dripping frequency with a consequent reduction of the droplet diameter as previously reported by Jaworek and Krupa (1992). As the electric potential is further increased, the dripping frequency increases and the droplet diameter reduces up to a critical value of the electric potential beyond which a transition occurs from the dripping mode to the formation of a stable jet; the latter spraying function mode is termed jet mode (Hayati, 1987a) and is further considered in Chapter 5.

Figure 4.1 Schematic drawing of the electric field lines between the nozzle and the earthed plate in the presence of a pendent drop.
The dripping phenomenon in an electric field has been the subject of several studies by a number of researchers including Borzabadi and Bailey (1978), Takamatsu et al. (1983) and Notz and Basaran (1999); see Chapter 2. The reason why the dripping mode has been the subject of the interest of many researchers is because it is possible to control the droplet diameter by varying the electric potential applied to the nozzle. The electric field generates a pulling force, $F$, as is shown in Figure 4.1. The magnitude of the force $F$ is predicted by Equation (3.1). The force acting on the pendant drop can be modulated by changing the applied potential and may be used to detach the pendant drop when it reaches a certain volume, hence providing a means for controlling the diameter of the drop.

In this work the possibility of controlling the droplet diameter is explored. Several flowrates, i.e. 5, 10, 20, 25 and 30 ml/min have been used. The working liquid is a 3% solution of PVA in water. This is the most viscous liquid that has been possible to test in the dripping regime. More viscous solutions of PVA in water when sprayed in dripping mode create polydispersed droplets and electrospinning. The behaviour of this kind of solution when sprayed in the dripping mode is shown later in this chapter. Three electrode configurations have been used, see Table 3.1 Chapter 3, and an applied DC potential with positive polarity. This potential was varied from 0 to 10 kV for the electrode configurations 1B and 1C and from 0 to 15 kV for the electrode configuration 1D.
4.2 Spraying of PVA 3% in the dripping mode

The liquid is fed with a flow rate ranging from 5 to 25 ml/min (giving a liquid velocity from 0.047 m/s to 0.235 m/s) and for these values it drips drop by drop from the nozzle. The application of the electric potential to the nozzle leads to the reduction of the drop diameter and to an increase of the dripping frequency; see Figure 4.2 and 4.3. The droplet diameter is not strongly affected by the flow rate as shown in Figure 4.2. An increase of the flow rate leads mainly to a higher dripping frequency. This implies that drop formation in this mode is governed essentially by a static force balance, i.e. the momentum contribution is insignificant especially for lower values of the flowrate. These features form the basis of modelling described in this chapter.

Droplet formation for viscous liquids in the dripping mode in the presence of an electric field has previously been described by Cloupeau and Prunet-Foch (1994); the applied electric potential stretches the liquid meniscus, forming a long filament between the growing main droplet and the part of the liquid meniscus attached to the nozzle. The liquid meniscus breaks-up when it reaches a maximum elongation, which has been defined as the limiting length by Notz and Basaran (1999). The time taken for the liquid meniscus to reach its limiting length is defined in this work as $t_f$; Detailed observation of high speed digital video records of Figure 4.4 shows that $t_f$ varies with the applied voltage and flow rate. When the rupture of the liquid meniscus takes place the long filament can evolve in two modes: (a) it is taken up by the part of the meniscus that stays attached to the nozzle, or (b) it becomes a smaller droplet called a satellite. The part of the liquid that does not detach from the nozzle relaxes back and the process starts again. For the electrode configurations used, i.e. 1B, 1C and 1D, the sizes of the main droplets and of the satellites become sensitive to the electric potential changes above about 3 kV as shown in Figure 4.2. An increase of the applied potential leads to narrower size distributions of both droplets and satellites although they remain distinct from each other. However, the main droplet size decreases and the satellite size increases and eventually their distinction becomes impossible, Speranza et al. (2001), see Appendix C. At high electric potentials, above 10 kV, the electric field is sufficiently strong to cause dielectric break down by sparking between the electrodes, and at the same time whipping occurs.
Figure 4.2 Droplet size as function of applied electric potential for electrode 1B at various flowrate. Using a flowrate of 5 ml/min (droplet ●, satellite ○); using a flowrate of 10 ml/min (droplet ■, satellite □); Using a flowrate of 20 ml/min (droplet +, satellite +).
Figure 4.3 Droplet frequency as of applied electric potential for electrode 1B at various flowrate. Using a flow rate of 5 ml/min ■; using a flow rate of 20 ml/min ●.
Figure 4.4 Stages of formation of main droplet for a number of potentials for electrode configuration 1B.
4.3 Dripping to jet mode transition

In the absence of an electric field, if the fluid is pumped through the nozzle with a flow rate of 30 ml/min (velocity 0.283 m/s), it is possible to observe a transition from the dripping mode to the jet mode when using the electrode configuration 1B. The transition velocity is in agreement with the formula suggested by Scheele and Meister (1968b):

\[ U_j = 1.73 \left[ \frac{\gamma}{\rho D_m} \left( 1 - \frac{D_m}{D_{dl}} \right) \right]^{\frac{1}{2}} \]  

(4.1)

where \( U_j \) is the jet transition velocity, \( D_m \) is the inside nozzle diameter, \( D_{dl} \) is the droplet diameter which forms before there is the transition from dripping mode to jet mode, \( \gamma \) is the surface tension and \( \rho \) is the density. Equation (4.1) predicts a jet transition velocity of circa 0.294 m/s; using a \( D_{dl} \) of 5 mm which was measured before that the transition from dripping mode to jet mode occurred.

Within a range of flow rates in the dripping mode, the transition to the jet mode can be also effected by increasing the electric potential, see Figure 4.5. For a flow rate of 25 ml/min, if the electric potential applied to the nozzle exceeds 7 kV there is a transition from the dripping mode to the jet mode. The process of transition from dripping to jet mode is similar to the dripping mode. The main difference between the two modes of operation is in the liquid meniscus, which is attached to the nozzle. In each cycle of dripping, the liquid meniscus relaxes back as a droplet is pinched off. However, once the electric field is increased the meniscus becomes longer and thinner until it suddenly switches into a jet as shown in Figures 4.6. It is worth to observe that the transition from the dripping mode to the jet mode by the increase of the electric potential occurs at flow rates of 25 ml/min (velocity 0.235 m/sec) which is lower than that predicted by Equation (4.1): (0.294 m/s). Therefore, the stresses due to the electric field have an effect that is similar to that of reducing the surface tension of the liquid. The transition from the dripping mode to the jet mode by increasing the applied potential shows hysteresis, see Figure 4.5.
When the jet mode has formed, even if the electric potential is reduced to less than 7 kV, the jet mode still persists, until the electric potential falls below 5 kV. The existence of the hysteresis is discussed in Section 4.8.

![Diagram showing the transition between jet and dripping modes](image)

Figure 4.5 Effect of applied potential on the droplet size for jet and dripping modes for electrode configuration 1B. Using a flowrate of 25 ml/min (— dripping mode, — jet mode); using a flowrate of 30 ml/min (— jet mode).

Referring to the jet mode operation for electrode 1B shown in Figure 4.5 the droplet size seems to be weakly affected by the electric potential increase. The reduction of the droplet diameter, albeit very small can be due to the reduction of either (a) the jet diameter or (b) the droplet diameter to jet diameter ratio, $D_d/D_j$. The former can be caused by the acceleration in the downstream direction of the liquid jet due to the
Figure 4.6 Transition from the dripping mode to the jet mode for electrode 1B, flow rate 25 ml/min of PVA 3% and applied potential of 7 kV.

tangential component of the electric field. While the liquid accelerates the flow rate keeps constant and as consequence the jet becomes thinner.
According to Mutoh et al. (1979), this is the major reason why the droplet diameter reduces as the potential is increased. The droplet reduction can also be due to the effect of the electric field on the ratio $D_d/D_j$. Bailey and Balachandran (1981) suggested that the high charge density on the jet surface promotes the formation of smaller droplets. Thus, an increase of the voltage leads to an increase in the number of surface charges with a consequent production of smaller droplets. In this work, the droplet diameter to jet diameter ratio has been measured for the electrified jet formed by increasing the applied potential as well as for the electrified jet formed just by increasing the flow rate. It has been observed that the value of the measured ratio differs from that predicted by Weber's formula within 5%, see Equation (2.33). Therefore, in this case the reduction of the droplet diameter with an increase in potential can be more attributed to the reduction of the jet diameter rather than a change in the ratio $D_d/D_j$ due to the surface charge. However, this point is addressed in detail in Chapter 5.

The jet break up shifts from varicose to kink instability when the electrical potential is increased above 9 kV. The reason for this transition has been attributed to electrification of the jet surface. In fact, when the surface charge density is increased beyond a certain value the lateral growth factor becomes predominant with respect to the growth rate factor for varicose instability, hence the jet starts whipping (Bailey and Balachandran, 1981).
4.4 The satellite droplets

In the dripping and jet modes, for the same value of the flow rate and of the electric potential, one or more satellite droplets can form along with the main droplet. The presence of the satellite droplets in the dripping mode is due to the rupture of the liquid filament between the main droplet and the liquid meniscus that is left attached to the nozzle as shown in Figure 4.7. The factors that come into play in the formation of the satellite droplet are the liquid surface tension and the applied electric field superimposed upon the space-charge field existing in the drop transport region. The electric potential increase hence the electric field increase leads to an increase of the filament length, and therefore to a bigger satellite droplet (see Figure 4.2). For flow rates of 5, 10 and 20 ml/min the satellite is always present in the range of the operating voltages used here. However, for the flow rate of 25 ml/min the droplet coexists with a satellite only when the electric potential is equal to or more than 5 kV; for applied potentials below 5 kV only droplets are produced. In this condition of flow rate (25 ml/min) and applied potential (below 5 kV) the force balance, due to the liquid surface tension and to the electric field superimposed upon the space-charge field, is such that at the droplet formation the liquid filament is sucked in to the liquid meniscus and no satellite forms. Therefore in the dripping mode it is possible to find an optimum flow rate in which the liquid can be sprayed in a controlled fashion.

![Figure 4.7 Satellite formation with an electrical potential of 8 kV for electrode 1B.](image)

The formation of satellite droplets have been observed in both dripping and jet modes. More detailed information about the satellite formation is given in Chapters 5 and 6.
4.5 **Beyond dripping mode for PVA 3%**

It has been observed that a liquid meniscus can assume a conical shape under the action of an electric field (Taylor, 1964), and this leads to the detachment of droplets from the tip of the liquid meniscus for a certain value of the electric field. Further increases of the electric field bring about an increase in the rate of drop detachment, leading to the formation of a steady stream emerging from the tip of the liquid meniscus (Taylor, 1969).

The interest in spraying a liquid by this electrohydrodynamic spraying functioning mode lies in the possibility of dispersing a liquid into very small droplets with a controlled diameter. Droplets with a narrow size distribution and with diameters of a fraction of millimetres can find application in a number of industrial cases. Consequently, the possibility of spraying PVA aqueous solutions using the cone jet mode has been pursued in this work and the results are reported below. As described above, spraying by the dripping mode is a characteristic feature which prevails only for given flow rates and for a range of applied potentials using a given electrode geometry and a liquid with certain physical properties. Using the type 1B electrode geometry and spraying with PVA 3%, the atomisation occurs by the dripping mode up to applied potentials of 7, 8 and 9 kV for flow rates of 20, 10 and 5 ml/min respectively. Beyond the above potentials, the liquid atomisation occurs by more complex electrohydrodynamic spraying modes, which are described below.

Using a flow rate of 5 ml/min the atomisation occurs by the dripping mode until 9 kV. At a potential of 10 kV the liquid filament, which is present between the main droplet and the meniscus, starts to follow a trajectory determined by the electric field lines set up between the electrode superimposed upon any space-charge field that exists in the drop transport region (see Figure 4.8). The tip of the liquid filament is pointing to the earthed electrode edge and, a brush of fine droplets is emitted from the tip as shown in the record with arrows in Figure 4.8. The liquid filament then relaxes into a droplet. The liquid filament does not take a constant direction, and can point to the left as well as the right edge of the earthed electrode. This phenomenon is still observable until an applied potential of about 11 kV. At 12 kV the liquid filament is drawn further...
downstream until a point is reached at which the entire filament starts to whip from side to side (see Figure 4.9).

Figure 4.8 High speed digital video record of a liquid filament affected by and following the electric field at a flow rate 5 ml/min of PVA 3% and applied potential of 10 kV for electrode configuration 1B.

A part of the filament breaks up into a number of very small droplets (see the frames in Figure 4.9 with arrow) and the rest of the filament becomes one or more droplets. It is
important to observe that at 12 kV the mechanism of atomisation has completely changed in comparison with the dripping mode. Here a thread of liquid is emitted from the liquid meniscus and this breaks up into droplets. It is not possible to distinguish between main drops and satellite drops under these conditions.

time scale: 0.009 sec

Figure 4.9 High speed digital video record of whipping of the liquid filament at high potential (12 kV) for flow rate 5 ml/min of PVA 3% and electrode configuration 1B.
Using a flow rate of 10 ml/min and raising the potential to 9 kV the liquid meniscus elongates to form what is called spindle mode as shown in Figure 4.10 from frame 1 to frame 5. The meniscus tip relaxes back and elongates again to form a liquid thread, which breaks up in two stages. The part indicated with a in Figure 4.10 frame 12 evolves in a way very much similar to the one described in Figure 4.9, while the part indicated with b in Figure 4.10 frame 12 breaks up following a mechanism similar to dripping mode.

Figure 4.10 Flow rate 10 ml/min of PVA 3% and applied potential of 9 kV. The set-up refers to electrode configuration 1B
A further increase of the flow rate leads to more a complex mechanism of liquid atomisation. Feeding the liquid with a flow rate of 20 ml/min and applying a potential of 8 kV, a very elongated meniscus is formed, from the tip of which large and small droplets are released, as illustrated in Figure 4.11. This electrohydrodynamic spraying mode is meta-stable. After some time it can move to a different equilibrium state. The tip of the meniscus can suddenly start to elongate and whip from side to side, following a mechanism which is similar to the one described for the dripping to jet mode transition as previously shown in Figure 4.6, see Figure 4.12. This sudden transition may be due to a change in the space-charge field, which can mutate the system equilibrium. It can be observed that for this condition of flow rate and applied potential the atomisation occurs by a mechanism where there is no dripping and no jet mode, but something in between. However, when the jet mode occurs the applied potential and therefore the jet surface charge induced is sufficiently high to make it whip and thus to short-circuit the system as soon as the jet touches the earthed electrode (see the frame with the arrows in Figure 4.12). Further trials have been performed using a flow rate of 25 ml/min, and once again the liquid atomisation takes place by dripping mode until a potential to about 7 kV. In fact when a potential of 7 kV is applied, the dripping mode switches to the jet mode.

Once the jet has been electrohydrodynamically formed a further increase of the applied potential to a value of 9 kV does not lead to a reduction of the jet diameter, but rather to the whipping motion, see Figure 4.13. N.B. all the above spraying modes shown in this section have been observed for the electrode configuration 1B. But this time from below the earthed electrode. They are a representative of the spraying modes observed using PVA 3% as the working liquid. Many more electrode configurations were set up, several of which are reported in Table 3.1. Several other flow rate regimes were used between 1 ml/min to 30 ml/min. However, it was not possible to produce a stable convergent jet by spraying PVA 3% under these conditions. The difficulty in creating a stable convergent jet seems to be very much related to the kind of liquid that is to be sprayed rather than to an appropriate electrode geometry and applied potential. In fact, a convergent jet was formed when PVA 3% was replaced with propanol, which is well known to be a liquid that can be sprayed by stable cone jet mode. Figure 4.14 shows the convergent jet formed using propanol at an applied potential of 7 kV and the
Figure 4.11 High speed digital video record of filament elongation at a high applied potential, flow rate 20 ml/min of PVA 3% and applied potential of 8 kV for electrode configuration IB.

electrode configuration IB. When propanol is fed with a flow rate of 5 ml/min and the potential is increased over 7 kV, the convergent jet suddenly shifts to what is called a
Figure 4.12 High speed digital video records of meta-stable jet formation and whipping, flow rate 20 ml/min of PVA 3% at applied potential of 8 kV for electrode configuration 1B.
multi jet, see Figure 4.15. In this spraying mode, about six convergent jets are emerging from the liquid, and each jet has a diameter of the order of 100 μm.

Figure 4.13 High speed digital video records of jet whipping, flow rate 25 ml/min of PVA 3% and applied potential of 9 kV for electrode configuration 1B.

Figure 4.14 (a) Convergent jet formed using propanol fed with a flow rate of 5 ml/min and an applied potential of 7 kV; (b) convergent jet formed using propanol fed with a flow rate of 10 ml/min and an applied potential of 7 kV.
Figure 4.15 Multi jet formed using propanol fed with a flow rate of 5 ml/min and an applied potential of 9 kV.
4.6 Spraying of PVA 5% in dripping mode

The process of spraying by the dripping mode may be inappropriate for liquids with high viscosity such as PVA 5%. In fact, when spraying PVA 5% in the dripping mode, it is observed that the top and bottom surfaces of the earthed electrode as well as the earthed electrode hole become full of very thin liquid wires and these wires form what looks like a “spider web”. The mechanism of formation of the above-mentioned liquid wires may be due to two observed phenomena as described below (see Figure 4.16).

Figure 4.16 (a) Wire formation from liquid filaments present between forming droplets and liquid meniscus in dripping mode of high concentration PVA (5%); (b) multi jets departing from the pendant drop: electrospinning.

Firstly, because of the high liquid viscosity, the liquid meniscus reaches a very long limiting length before its break-up takes place. Once the break-up occurs and the droplet forms, then a long filament is produced. This filament does not break up immediately into liquid droplets after its formation, and keeps its wire-like form for a while. Due to electric charges present on the liquid surface and the presence of the electric field, the wire is attracted by the earthed electrode, to which it becomes attached.
Secondly, during the process of droplet formation some wire-like liquid jets may be emitted from the surface of the forming droplet in a way very much similar to what was observed in the multi-jet-spraying mode; this phenomenon is know as electrospinning. On the liquid surface there are features that look like very small “Taylor cones”. They appear suddenly and from the tip of these cones a liquid filament is produced. The liquid filament maintains its wire form until it attaches the earthed electrode (see Figure 4.16 (b) and Figure 4.17). This phenomenon is similar to the multi jet formation seen when spraying propanol, and it may be due to an increase in the liquid viscosity with respect to that of PVA 3%. The increase in viscosity leads to the use of relatively high values of the applied potential with respect to those used in the spraying of PVA 3%, and this may induce the formation of the above mentioned “cones” on the liquid surface. In contrast with what was seen in the spraying of propanol, the cones that form on the surface are not stable, they do not follow any symmetry and they have a relatively short life-time.

Figure 4.17 Wires forming “spider web” when PVA 5% was sprayed in dripping mode.
All the wires formed accumulate on the earthed electrode and form what looks like a "spider web". After a few minutes of atomisation of PVA 5% the earthed electrode is covered in wires, which like a spider web will entrap a number of little droplets formed during the atomisation as shown schematically in Figure 4.17. Therefore, the dripping mode is found be inadequate for spraying liquids with a high viscosity such as PVA 5%.
4.7 A theoretical model for the dripping mode

The process of dripping is essentially quasi-static by nature with negligible inertial and viscous effects of the fluid flow (Scheele and Meister, 1968a; Clift, 1978). Therefore a simple static force balance should apply at all stages of droplet formation including the detachment time, from which the droplet diameter can be expressed as a function of the applied electric force. However, the calculation of the surface tension force requires the specification of the contact area over which the force acts. So far in the literature, this contact area is taken as the nozzle area as it easy to do, but unrealistic because of the necking process where at the time of detachment the drop is hung by a filament of much smaller cross-section area. Furthermore, this approach complicates the calculation of the droplet diameter because a third degree polynomial needs to be solved and the model so evaluated loses accuracy as soon as the potential goes over 3-4 kV reported by Takamatsu et al. (1983).

![Figure 4.18 Force balance on the pendant drop at time 70% of t1.](image)

A close examination of Figure 4.4 shows that the neck size near the detachment time is smaller than the nozzle size. In the model development below the force balance is made...
at a time at which the neck radius is equal to the droplet radius, thus making the force balance and the derivation of detaching droplet size very simple. The characteristic time for this situation is about 0.7 \( t_i \), where \( t_i \) is the detachment time based on the observation of video records of droplet formation.

The forces taken into account in the force balance are \( F_e \) due to the electric field, \( F_g \) due to gravity and \( F_s \) due to the surface tension acting over a circle of radius \( w \) (see Figure 4.18). The electrical force acting on the pendant drop is related to the electrode geometry, which provides a non-uniform electric field.

Jones and Thong (1971) showed that the electric field due to a potential applied to a semi-infinite wire with a rounded end and an infinite plate configuration, has the form:

\[
E = \frac{V}{\sqrt{2R}ln\left(\frac{4h}{R}\right)}
\]

where \( R \) is the radius of the wire end, \( V \) is the electric potential, \( h \) is the distance between the wire end and the earthed electrode. Equation (4.2) is hereafter adopted for the calculation of the non-uniform electric field in the set up used, see Figure 4.18. However, Equation (4.2) is quite an approximate model of the effective non-uniform electric field. It needs to be observed that the same expression for the value of the electric field at the tip end of semi-infinite wire with a rounded end was proposed by Loeb et al. (1941), and later adopted by Smith (1986) for deriving a formula to evaluate the potential required to achieve a stable cone jet (see Equation (2.5)). However, Loeb et al. (1941), and Smith (1986) used values different from \( \sqrt{2} \). In addition, expressions having a similar form to Equation (4.2) can be found in the work of Van Dyke (Taylor, 1969). In particular Van Dyke (Taylor, 1969) derived an expression for a semi-infinite wire with a rounded end which predicted an infinite value of the electric field on the wire end of the nozzle and consequently the expression is inapplicable (Borzabadi and Bailey, 1978). The electrostatic force acting on the pendant droplet can be evaluated from Equation (4.4) in the following way:
where $S_d$ is the surface area of the drop and $\varepsilon_o$ is the permittivity of the air. According to Takamatsu et al. (1983) the electrostatic force acting on the pendant drop can be considered to be equivalent to the electrostatic force acting on the formed drop with a small margin of error. Also, the drop is considered to have a spherical shape ignoring any deformation of the surface induced by the action of the electrostatic force (Byers and Perona, 1988).

The force due to the surface tension can be written as

$$F_s = 2\pi \gamma f_H$$

where $\gamma$ is the surface tension and $f_H$ is the Harkins correction factor. The radius of the neck area $w$ is taken at time 0.7 $t_i$ when it is approximately equal to the radius $r$ of the droplet, (see Figure 4.4). The Harkins correction factor, $f_H$, for the range of the droplet diameters considered here is taken equal to 0.70 (Adamson, 1990). This assumption allows an analytical result to be obtained with an error below 10% in the evaluation of the force due to the surface tension. The force, $F_g$, due to gravity is related to the droplet mass, thus the final force balance can be written as follows:

$$\frac{4}{3} \pi r^3 \rho g - 2\pi \gamma f_H + \frac{1}{2} S_d \varepsilon_o E^2 = 0$$

The result of the solution of Equation (4.5) is shown in Figure (4.19) where a good agreement is observed for an interval 0-8 kV for the electrode configuration 1B and 1C, and between 0-11 kV for the electrode ID. The ability of the model to predict the observed experimental trend deteriorates at higher values of the electrical potential. The model does not take account of the presence of a hole in the earthed disk as well as it does not take account of the space-charge effect and this may explain the difference between the model prediction and the experimental result at high electric fields. Also
for value of the potential above 8 kV the geometry of the pendant droplet at time 0.7t₁ starts to change substantially and the force balance may become inadequate.

Figure 4.19 Droplet size as function of the operating voltage in the dripping mode for PVA 3%, for electrodes 1B, 1C and 1D. Using a flow rate of 5ml/min for electrode 1B (droplet ■, model prediction ———); for electrode configuration 1C (droplet ★, model prediction ———); for electrode configuration 1D (droplet △, model prediction ———).
4.8 Discussion

In the dripping mode condition, it is possible to control the droplet diameter in a wide range by varying the applied potential and by using appropriate electrode geometry. The droplet size distribution is usually bimodal due to the formation of satellite droplets, whose size approaches to the main drops as the potential is increased. However, it is possible to find an appropriate combination of flow rate and applied potential in which only droplets are produced i.e. no satellite. The satellite droplets are a common feature in dripping of viscous liquids, this is due to the formation of a liquid filament between the forming droplet and the liquid meniscus attached to the nozzle. The latter forms into an individual satellite droplet. The satellite does not form when the liquid filament relaxes back in to the liquid meniscus. In the above mechanism the pressure due to the surface tension pulls the liquid filament up when the break up occurs, whilst the liquid viscosity opposes the liquid motion. Therefore, the recovery of the liquid filament into the liquid meniscus is less likely to occur for a more viscous liquid thus the satellite is more likely to form. When the dripping is driven by an electric potential the pressure due to the surface charge is working against the pressure due to the surface tension, hence impeding the liquid filament to recoil and making the satellite formation more likely to occur and with a large size.

Droplet and satellite formation is also affected but only weakly by the flow rate. For the same applied potential it has been observed that an increase in the flow rate leads to an increase in the droplet diameter, though changes in diameter due to the flow rate are small if compared to that due to the applied potential. Indeed, the flow rate and thus the momentum contribution are not significant in the dripping mode mechanism especially for the lower values of the flow rate. The flow rate has also a limited influence on the amount of charge transported in dripping mode in comparison with that transported in the jet mode, (see Figure 4.20 and Figure 5.23). This limited effect of the flow rate has enabled the development of a model for the dripping mode based upon a force balance at a given time. Despite the simplicity of the model it can predict the droplet diameter over a wide range of applied potentials without using any correction factor. The model, however, looses accuracy as the applied potential goes beyond 6-7 kV. This may be due to the approximation used for the non-uniform electric field, to the neglected space-charge effect and to the changes of the mechanism of droplet
formation. The droplet starts to form in such a way that the above-mentioned force balance does not really apply to the new mechanism of droplet formation.

The possibility of a convergent jet has also been investigated. Many trials were done by using a number of electrode configurations, flow rates and nozzles. In all these trials using PVA aqueous solutions it has not been possible to form either a convergent jet or a cone jet as observed for other liquids such as propanol. A collection of high speed video records have been reported to show the electrohydrodynamic functioning mode which instead occurs. The reasons that no convergent jet can form may be due either to the liquid physical properties or to the nozzle diameter used. In fact, as seen in literature the convergent jet occurs if there is a tangential component of the electric field.
large enough to stabilise the liquid surface. This tangential component of the electric field is proportional to the current flowing through the jet. Hartman (1998) observed that smaller nozzles could conduct more current than a large ones. Therefore by using a smaller nozzle than those used in this thesis it would be possible to produce an increase in the conducted current through the jet and so to increase of the tangential component of the electric field acting on the liquid surface, see Equation (2.9), in a way to produce a stable cone jet mode. Hence for spraying highly conductive and viscous liquids the use of a smaller nozzle may be more suitable. Further work supporting this conclusion may be found in Harpur et al. (1996). In this work for liquids having small charge relaxation times small nozzle diameters are recommended. In particular for a liquid with a charge relaxation time of $10^{-3}$ sec a nozzle radius ranging between 0.1 mm to 0.6 mm is recommended. Following the above indications for a liquid with a charge relaxation time smaller than $10^{-7}$ sec as the one used in this work a suitable nozzle should have a diameter of a few tens of microns.

Another problem for the cone jet set up may be the high value of the surface tension. Smith (1986) concluded from his experiment that liquids having a surface tension above a threshold value of 50 mN/m may not be sprayed in cone jet mode. In this work all the PVA aqueous solutions have got a surface tension above the threshold value predicted by Smith. Finally, the high liquid viscosity may be also responsible for the difficulty in setting up the cone jet mode. The high liquid viscosity may reduce the amount of current flowing through the jet and so impeding the creation of a tangential electric stress large enough to stabilise the jet cone. This argument is treated in detail in Section 5.5.

The variation of the applied potential enables the transition from one electrohydrodynamic functioning mode to another keeping constant the flow rate (see Figure 2.1). In this work in the spraying of PVA 3% it has been possible to shift from dripping to jet modes by applying an adequate potential using the electrode configuration 1B. However, this transition shows a hysteresis. The existence of a hysteresis may be due to the way in which charges are transported through the droplets and thus to the current transported through the liquid jet. The amount of charge transported by the droplets for unit mass is equivalent to the ratio between current
conducted through the nozzle and mass flowrate of the liquid issued by the nozzle, hence by Equation 2.12 can be written:

\[ I = 12Q \left[ \frac{2\pi \sigma_0}{D^3} \right]^{\frac{1}{2}} \]  

(4.6)

The above equation predicts an increase in the amount of current conducted when the droplet size is reduced for a constant flowrate. When the potential is increased a shift from the dripping mode to jet mode occurs and this may be attributed to a combined effect of droplet reduction and current increase. In effect, by increasing the potential, the dripping frequency increases and so the current transported through the jet also increases. The increase in current conducted increases the tangential electric stresses until a point at which the tangential electric stresses are large enough to induce the jet to form. When the jet mode is established an increase in the liquid velocity and a reduction of the droplet diameter from that produced in dripping mode is recorded. Thus, for the same flow rate, an abrupt increase in the current conducted through the jet is recorded; the current increases from \(1.5 \times 10^{-8}\) A to \(2.5 \times 10^{-8}\) A. Reducing the applied potential to 6 kV the jet persists despite this value of the potential being lower than the value at which dripping to jet mode shift occurs; in this work 7 kV (see Figure 4.5). This may be due to fact that in the jet mode although a decrease of the potential provokes a reduction of the current conducted, this reduction is smaller than that which would occur in a regime of dripping mode. This is because for the same flowrate in the dripping mode the droplets are larger than those produced in the jet mode and so a lower amount of charge is transported. In addition, the liquid velocity in jet mode is larger than that in dripping mode thus more charges are transported by convection. Furthermore, the jet formation may induce an increase in the capacitance of the electrode-liquid-meniscus system. Therefore, despite the reduction of the potential to 6 kV the system conditions are still favourable to sustain the jet mode. Further reducing the potential a point is eventually reached at which the current is not enough to create a tangential component of the electric stress sufficiently large enough to sustain the jet and so the dripping mode takes place again.
Finally, many other trials have been performed by spraying PVA 5% in the dripping mode. However, many difficulties have been encountered such as the production of long liquid filaments, either from the liquid jet break up or from the liquid meniscus surface and the presence of satellites for any potential and flow rate used. Therefore, it is inappropriate to spray the PVA 5% in dripping mode. However, in Chapter 6, it is shown that water solutions with this concentration in PVA can be easily sprayed in jet mode by using an appropriate electrode geometry and applied potential.
Chapter 5 - Liquid jet break-up by a constant DC potential

5.1 Introduction

Liquids form a stable jet when emitted from a nozzle at a sufficiently high velocity, see Chapter 4. The liquid jet surface may be subjected to interfacial deformation when an electric potential is applied to the nozzle. The electric field configuration depends on the electrode geometry. The interfacial deformation, which is made of peaks and valleys, moves downstream. They are named kink and varicose instabilities as described in Chapter 2. These instabilities, which are affected by the flow rate, liquid physical properties, applied potential, and electrode geometry, are responsible for breaking up the liquid jet into droplets. Hence, by controlling the instabilities it is possible to control the droplets' size distribution.

![Voltage scale; increasing voltage from left to right](image)

Figure 5.1 Effect of the applied potential on the convergent jet (Hayati et al. 1987a).

There is a complex relationship between the droplet diameter and the jet diameter. This relationship depends on a number of variables such as the applied potential, jet diameter and physical properties of the sprayed liquid, i.e. viscosity, density and surface tension (Basset, 1894; Bailey, 1988). However, with respect to the
jet diameter, the thinner the liquid jet the smaller is the droplet diameter when the liquid jet eventually breaks up. An increase of the applied potential on the nozzle from which the liquid is issued may induce the liquid jet to converge, see Figure 5.1. Thus, the droplet diameter can also be varied in this way.

In this chapter the possibility of manipulating the spraying of a liquid jet and hence the size distribution of the droplets is explored by using as working liquids polyvinyl alcohol water solutions and distilled water. There exist two instabilities: the kink instability and the varicose instability. The kink instability is usually typical of high applied potentials and it appears to be a chaotic phenomenon as it is characterized by large and violent lateral displacements that are responsible for producing polydisperse droplets. For this reason the kink instability has not been investigated here as a possible way to spray a liquid, rather it has been seen as a limit over which it is not possible to have an accurate control of the spraying. Therefore, attention is paid primarily to the varicose instability, where the effect of the applied potential on the jet diameter as well as on the ratio of the droplet diameter to jet diameter has been investigated.
5.2 The effect of the applied potential on the distilled water liquid jet break up

The droplet and satellite diameters can be controlled by using a constant DC potential. This is done by applying the constant DC potential on the needle of an appropriate spraying set-up. Ten experimental set-ups have been used to investigate the possibilities and limitations of controlling the droplet and satellite diameters and the results of three types of liquids with different viscosities are described below.

The first series of experiments performed is the spraying of distilled water with the electrode configuration 2A and using a DC voltage. Details of the electrode configuration are given in Table 3.1. The measurement of the droplet diameter as well as the satellite diameter has been performed and the results are presented in Figure 5.2. The droplet diameter as well as the satellite diameter reduce as the potential is increased. The liquid jet breaks up into droplets by varicose instability up to a potential of 3 kV (electric field about 2X10^6 V/m). When the potential is increased to 4 kV (electric field about 2.7X10^6 V/m), the jet starts to oscillate and the kink instability sets in as soon as the potential reaches 5 kV (electric field about 3.5X10^6 V/m). In this instability condition eventually sparking occurs between the nozzle and the earthed electrode. Therefore, this kind of electrode configuration appears to be unsuitable for obtaining a significant diameter reduction of the droplets and satellite.

Based on this result, it is worth exploring the effect of an increase of the distance between the nozzle and the earthed electrode to obtain a longer distance over which the electric field effect prevails. The distance was increased by 10 mm bringing it to 15 mm corresponding to electrode configuration 2B, as shown in Table 3.1. As it can be seen in Figure 5.3 the increased distance allows a larger electrical potential and electric field to be applied and this leads to the production of smaller droplets. The liquid jet flows downstream smoothly and various types of instability occur after the earthed electrode as shown Figures 5.4-5.8. The liquid used is distilled water and the flow rate is 32.2 ml/min. The jet break up in the presence of an applied potential, between 1-5 kV (electric field about 0.5-2.6X10^6 V/m), is shown in Figures 5.5 to 5.8. The break-up is by varicose instability. The jet starts to be subject to an instability that seems to be a combination of varicose and kink as soon as the applied potential reaches
6 kV (electric field about $3.1 \times 10^6$ V/m) as shown in Figure 5.6. Raising the potential further it leads to stronger lateral oscillation, and at 8 kV (electric field about $4.2 \times 10^6$ V/m) the kink instability is fully developed, (see Figure 5.7). With this electrode configuration a potential of 11 kV (electric field about $5.7 \times 10^6$ V/m) can be reached before sparking between the needle and the earthed electrode occurs. At this value of the potential the liquid jet starts to follow the field line approaching what is called in the literature a "ramified jet" (see Figure 5.8).

![Graph](image)

**Figure 5.2** The effect of the applied DC potential on the droplet size for distilled water using the 2A electrode configuration and a flow rate of 32.2 ml/min; ○: droplet; ▪: satellite.

The high speed digital video records shown in Figures 5.4-5.8 help to partially explain the reason why an increase of the applied potential leads to smaller droplets. On the surface of the jet there are electrical charges, which are induced by the electric field.
These charges transported by charge-carriers are accelerated downstream by the tangential component of the electric field acting on the jet. The acceleration of the charge-carriers accelerates the liquid while the flow rate is kept constant. As a result the jet becomes thinner. A second reason why the droplets become smaller is due to the effect of the electric field on the droplet diameter to jet diameter ratio. This is described later in this chapter.

Figure 5.3 The effect of the applied DC potential on the droplet size for distilled water using the 2B electrode configuration and a flow rate of 32.2 ml/min; ⬠: droplet; ⬞: satellite.

The electrified jet breaks up into droplets that are conveying charges. Consequently these droplets start to follow a trajectory determined by the electric field lines set up between the electrodes superimposed upon any space-charge field that exists in the drops transport region, as can be seen in Figure 5.5. Therefore the electric
field underneath the earthed electrode cannot be considered nil despite a very small hole in the earthed electrode (6 mm for electrode configuration 2B).

Figure 5.4 Break up of a jet of distilled water without an electric field due to natural disturbance for electrode configuration 2B.

Figure 5.5 Break up of a jet of distilled water with an applied potential of 4 kV for electrode configuration 2B.
The size distributions of the drops and of the associate satellite droplets are shown in Figure 5.9 to 5.11 for various applied potentials for a constant flow rate of 32.2 ml/min for electrode configuration 2B. As it can be seen an increase in the electric
potential has the effect of increasing the total amount of satellite droplet to the detriment of the amount of the main droplet, and in addition the droplet distribution becomes wider. Further investigations have been performed, in which, the distance between the needle and the earthed plate has been varied to explore its effect on the droplets diameter (see electrode configuration 2C, see Table 3.1). The results are shown in Figure 5.12. Once again the voltage increase leads to a reduction of the main drop diameter, but with little or no change of the satellite size. The maximum potential used here was 5 kV, because about this level an anomalous interaction occurs between the far end of the liquid jet and the orifice edge of the earthed electrode hole (see Figure 5.13). This is referred to as an anomalous interaction because it does not seem to be a whipping of the far end of the jet, but rather due to a disturbance from the close edge of the earthed electrode on the jet break up mechanism. Consequently the drop size distribution becomes very wide as shown in Figure 5.14.

Figure 5.9 Size distribution of droplets of distilled water using the electrode configuration 2B, at an applied potential of 1 kV.
Figure 5.10 Size distribution of droplets of distilled water using the electrode configuration 2B at applied potential of 4 kV.

Figure 5.11 Size distribution of droplets of distilled water using the electrode configuration 2B at applied potential of 8 kV.
Figure 5.12 The effect of the applied DC potential on the droplet size for distilled water using the 2C electrode configuration and a flow rate of 32.2 ml/min; ●: droplet; ○: satellite.

Any further voltage increase would bring about a wider droplet size distribution and so moving away from the original goal of this project, which is to generate monosized droplets. Therefore, it can be concluded that the droplet diameter as well as its size distribution can be manipulated by the use of a DC potential, but the size distributions is bimodal for any distance and potential used. However, a more uniform size distribution is realised when using the electrode configuration 2B.
Figure 5.13 Anomalous behaviour in the jet break-up for distilled water using the 2C electrode configuration and a flow rate of 32.2 ml/min and an applied electrical potential of 5 kV.

Figure 5.14 Size distribution for distilled water using the electrode configuration 2C at applied potential of 5 kV.
5.3 The effect of the applied potential on the liquid jet break up of PVA 3%

The effect of a DC potential on the atomisation of the PVA 3% has been investigated. The experiments have been performed using the electrode configuration 2C (see Table 3.1). The control of the droplet diameter by the applied potential, within the range in which it can be varied, is quite weak and on the satellite it is almost nil. Therefore the use of such a small orifice in the earthed electrode is impracticable for this kind of liquid when a DC potential is used. The droplet diameter as function of applied potential is shown in Figure 5.15 for PVA 3% and the 2C electrode configuration. For this electrode configuration the maximum applied potential is 3 kV because above this value blue positive corona formation was detected. A light was visible between the earthed electrode and the liquid jet as shown in Figure 5.16. This phenomenon suddenly brings about an electric current of the order of few milliamperes and for this value of the current the EHT supply switches automatically off for safety reasons.

Based on these results it has been considered sensible to replace the earthed electrode with one having a larger hole diameter because by increasing the distance between the jet and the edge of the earthed electrode the corona formation is less likely to occur. The new electrode configuration is 3B defined in Table 3.1. The wide hole present in this electrode is 30 mm in diameter and it enables the application of a larger value of the electric potential as compared to the earthed electrode having 6 mm hole in diameter. The maximum value of the applied potential reachable with this electrode configuration is 11 kV (7X10^6 V/m) as compared to 3 kV (2X10^6 V/m) in the previous case. At this value of the applied potential sparking is observed. The results are shown in Figure 5.17. Once more the increase of the applied potential leads to a reduction in the droplet diameter, while the satellite diameter is virtually unaffected by the potential raising. It is worth noticing that the recorded trend for the dependence of the droplet diameter on the potential changes is very much similar to that observed for the experiments performed using the electrode configuration 1B (see Figure 4.5). In contrast with what has been observed in the spraying of distilled water, the jet diameter as well as the droplet diameter are less affected by the potential changes in the spraying of PVA 3% (see Figures 5.18 to 5.20).
Figure 5.15 The effect of the applied DC potential on the droplet for PVA 3% using the 2C electrode configuration and a flow rate of 32.2 ml/min; ●: droplet; ○: satellite.

Figure 5.16 Corona formation at high potential between the liquid jet of PVA 3% and the earthed electrode; a) no electric potential applied; b) 3 kV; c) 4 kV. Electrode configuration 2C.
For an applied potential between 1 to 7 kV the liquid jet is breaking up by varicose instability. When 8 kV potential is reached then the tip of the jet starts to kink, in contrast with what is observed for distilled water, where a potential of 8 kV caused all the jet to kink (see Figure 5.19). Fully-developed kink instability for a jet of PVA 3% is observed for a potential of 10 kV and for 11 kV the kink amplitude becomes so large that the tail may point upstream (see Figure 5.20). It is important to observe that despite the 11 kV applied to the nozzle, the jet diameter does not change appreciably. As observed above in the atomisation of PVA 3%, the jet diameter as well as the droplet diameter are less affected by the applied potential in comparisons with what is observed in the atomisation of distilled water. In order to investigate the difference between the behaviour of PVA 3% and distilled water it was decided to measure the amount of current flowing through the jet. The results of this investigation are described in Section 5.5 of this chapter.

Figure 5.17 The effect of the applied DC potential on the droplet for PVA 3% using the 3B electrode configuration and a flow rate of 32.2 ml/min; ●: droplet; ○: satellite.

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Figure 5.18 Liquid jet of PVA 3% a) no potential applied b) 4 kV applied potential

Figure 5.19 Liquid jet of PVA 3% with 8 kV applied potential

Figure 5.20 Liquid jet of PVA 3% with 11 kV applied potential
5.4 Effect of the applied potential on the droplet to jet diameter ratio

An increase in the applied potential causes a reduction in the droplet diameter as can be seen in Figure 5.2, 5.3 and 5.12. This is achieved by two different processes: (i) a reduction in the jet diameter due to the jet acceleration by the tangential component of the electric field; (ii) a reduction in drop to jet diameter ratio due to the surface charge.

![Figure 5.21 Effect of the applied potential on the droplet to jet diameter ratio for distilled water. The arrow indicates until which potential the jet is subject to varicose instability. ⋆: experimental measurement; ----- Lord Rayleigh's ratio.]

As previously stated, there are charges present on the surface of the jet. These charges are accelerated downstream by the tangential component of the electric field. The acceleration of these charges accelerates the liquid and as a consequence the jet becomes thinner, and the droplets become smaller.
Figure 5.22 Effect of the applied potential on the droplet diameter to jet diameter ratio for PVA 3%. The arrow indicates until which potential the jet is subject to varicose instability. $\ast$: experimental measurement; $-$-$-$-$-$ Weber's ratio.

The applied potential can also influence the droplet diameter to jet diameter ratio. Lord Rayleigh (1878-1879) found that for inviscid liquids the above ratio is 1.89 for a jet break up by varicose instability, see Equation (5.1).

$$\frac{D_d}{D_j} = 1.89 \quad (5.1)$$

Therefore, it is possible to verify if the applied voltage influences the droplet to jet diameter ratio by evaluating how the ratio deviates from Lord Rayleigh's value.
The ratio is weakly influenced for an applied potential below 3 kV, but the trend changes for higher values of the potential. The largest deviation from the Rayleigh ratio is recorded for an applied potential of 8 kV where the value is about 1.5. For applied potentials above 5 kV (see Figure 5.21) the jet does not break up by varicose instability, but by kink instability so any comparison with the value proposed by Rayleigh becomes inappropriate. Therefore, it can be concluded that the jet diameter reduction and the variation of the droplet to jet diameter ratio both come in to play, but the latter only for higher values of the applied potential.

The reduction of the droplet diameter due to an increase of the applied potential is also recorded when PVA 3% is sprayed (see Figures 5.15 and 5.17). To quantify the effect of the applied potential on the droplet diameter reduction, the droplet to jet diameter ratio is also plotted as functions of the potential; see Figure 5.22. The variation of the ratio in this case is compared with the formula given by Weber, because the PVA 3% cannot be considered inviscid. In the Weber formula, Equation 2.33, the ratio depends on the diameter of the jet, which is also affected by the applied potential. Thus for each applied potential the droplet to jet diameter ratio predicted by Weber needs to be calculated. This is plotted in Figure 5.22 as a broken line. However, similar to the case of distilled water a deviation from the ratio proposed by Weber is recorded. The last significant data point is for an applied potential of 7 kV. At 8 kV the tip of the jet starts to whip, thus the Weber ratio, which refers to viscous jet break up by varicose instability, turns out to be unsuitable for comparison with the experimental ratio.

It is observed that for the lower values of the applied potential the droplet to jet diameter ratio is slightly larger than the value predicted by Weber's equation. This result is just the opposite of what was expected and may be explained by die swelling. When a polymer is extruded through a capillary die it becomes stressed from the shear forces at the die entrance and within the die. On leaving the die the polymer is no longer constrained and it relaxes the stresses by swelling. This results in the extrudate having a larger thickness than the die gap. Similarly in our case the liquid jet is "extruded" through by the electric field until the break up occurs. In the break up the PVA 3% relaxes into a droplet, which is larger than it should be. Consequently the droplet to jet diameter ratio becomes larger than the expected. However, for applied potentials over 5 kV the droplet to jet diameter ratio is smaller than the value given by
Weber's relationship. Again in this case the reduction of the droplet diameter at high values of the applied potential can be attributed partially to the influence of the applied potential on the ratio $\frac{D_d}{D_j}$. It can therefore be concluded that the droplet to the jet diameter ratio is affected by the potential for both distilled water as well as PVA 3%. At low values of the potential, droplet diameter reduction is only due to the effect of the decrease of the jet diameter. At higher values the reduction of the droplet diameter is due to the reduction of the $\frac{D_d}{D_j}$ caused by the jet electrification. However the deviation of the $\frac{D_d}{D_j}$ is larger for distilled water than for PVA 3%.
5.5 Discussion

The electrohydrodynamic spraying of a liquid in the smooth jet mode very much depends upon the electrode geometry and the physical properties of the liquid. Using distilled water it is possible to manipulate and vary the diameter of the main droplet as well as the diameter of the satellite droplet when using the 2B electrode configuration. However, with this configuration the jet can behave in a kinking mode and polydispersed drops are produced. The spraying of PVA aqueous solutions, such as PVA 3% is very difficult to perform for two reasons. Firstly, because corona discharge occurs readily as reported earlier. Secondly, the jet diameter and thus the droplet diameter are not that much affected by the potential increase.

The experimental observation that a jet of a highly conductive liquid, such as PVA 3%, is less affected by an electric potential than a jet of distilled water appears surprising at the first glance. It was expected that the higher electrical conductivity the larger are the charge carriers and so the stronger is the electric field effect. In fact, Smith (1986) stated that when liquids with high conductivity were atomised, the liquid jet and thus the droplets were so small that they could be hardly seen. Mutoh et al. (1979) observed that the higher the liquid conductivity the lower was the potential applied for cone jet formation. Hence, to better understand what was happening it was decided to measure by an ammeter the amount of current flowing through the jet of distilled water, PVA 3% and PVA 5% using the same set up. It was found that the current transported by the jet increased as the potential was increased, but more for distilled water than PVA 3% and PVA 5%. In Figure 5.23 it can be noticed that for the same applied potential the jet of PVA 5% is transporting a lower amount of charge in comparison with PVA 3%, and that this one is transporting a lower amount of charge in comparison with the jet of distilled water. Indeed, PVA 3% and PVA 5% have a larger amount of dispersed ions and they should transport more current, than distilled water. Despite that, the trend measured is inverted. This can be due to a number of factors. The liquid viscosity can influence the amount of charges conducted through the jet. Harman (1998) showed that the current conducted through a liquid jet of low viscous heptane was larger than the current conducted through a liquid jet of more viscous sunflower oil or dodecanol. In addition, the liquid viscosity can influence the fluid-dynamics inside the liquid jet which in turn can determine how the charges are
conducted through the jet. In fact, the radial velocity profile inside a liquid jet depends on the liquid viscosity. The radial velocity profile inside a liquid jet made of low viscous liquid (e.g. water) is parabolic, while the radial velocity profile inside a liquid jet made of high viscous liquid (e.g. PVA water solutions) is rather flat, see Figure 5.24.

Figure 5.23 Electric current versus electric potential for a jet of distilled water and a jet of PVA 3%. Distilled water (◇: flow rate 30 ml/min; ◇: flow rate 40 ml/min). PVA 3% (●: flow rate 30 ml/min; ○: flow rate 40 ml/min). PVA 5% (■: flow rate 30 ml/min; □: flow rate 40 ml/min).

This difference between the radial velocity profiles can determine substantial variation in how the charges are conducted through the jet. A developed parabolic velocity
profile promotes more the charge convection towards the liquid jet surface compared to a flat velocity profile, hence liquid jet with low viscosity may conduct more current (Gañana-Calvo et al., 1997 and Hartman, 1998). Furthermore, the amount of current conducted through a liquid jet depends on the diameter of the droplets produced when the jet breaks up into droplets; the larger the droplet diameter the lower the current conducted, see Equation (4.6). As shown, in Paragraph 5.2, 5.3 an increase in the liquid viscosity leads to large droplets produced when applying the same electric potential, hence the higher the liquid viscosity, the larger the droplet produced and the less the amount of current conducted through the liquid jet.

Another factor influencing the amount of charges conducted through the jet is the liquid permittivity. Salt water solutions can have a dielectric constant quite different from that of pure water, depending on the concentration as well as on the kind of salt which constitutes the solute. The relative dielectric constant of an aqueous solution can be by 50%-60% lower than that of pure water (Wang and Anderko, 2001). Therefore, it can be expected that PVA aqueous solutions have a relative dielectric constant lower than that of distilled water. Hartman (1998) observed that the reduction of liquid dielectric constant produced a reduction in the conducted current through a liquid jet, hence more current is predicted to be conducted thought a jet of distilled water than a jet of PVA aqueous solution.

N. B. the spraying behaviour may be qualitatively analysed by the characteristic time constant such as the charge relaxation time, given by Equation (2.6), the viscous
relaxation time, $\tau_{\mu}$, i.e. the time taken for a flowing liquid volume to relax so that its velocity gradients are no longer temporally dependent, see Equation (5.2), and the flow characteristic time, $\tau_f$, that is the time required for sufficient amount of liquid to flow into the system to produce a certain volume, e.g. the volume of the jet as given by Equation (5.3) (Harpur et al. 1996; Woodson, 1968).

\[
\tau_{\mu} = \frac{a^2 \rho}{\mu} \tag{5.2}
\]

\[
\tau_Q = \frac{l_j}{v_j} \tag{5.3}
\]

here $l_j$ is the jet length and $v_j$ is the jet velocity. The calculated values for the above mentioned viscous and charge relaxation times are reported in Table 5.1, while the value for the flow characteristic time, $\tau_Q$, is for all the liquid circa $10^{-1}$ s. The charge relaxation time has been calculated considering for the water as well as for all the PVA water solution a relative dielectric constant of 80. As above stated salt water solutions can have a dielectric constant quite different from that of pure water, however in this case, this approximation would not change the order of magnitude of the charge relaxation time. The very short charge relaxation time of the liquid used with respect to that of the flow characteristic time guarantees the effect of the applied potential on the spraying. Yet, a strong difference is observed when spraying the two liquids.

The fluid-dynamics within the jet can influence the way in which the charges are transported, as stated above. In particular in our experiments the jet fluid-dynamics can be easily observed. The viscous relaxation time for the PVA water solutions is smaller than the flow characteristic time. As a consequence, in the jet of PVA water solution as soon as the jet issues from the needle there is a small and rapid change in the radial velocity profile and then the velocity profile remain the same throughout the whole jet. There is small difference between the liquid velocity at the surface and the liquid velocity in the jet centre for any jet’s radial section; the radial velocity profile is quite flat. Instead, for a liquid jet of water the characteristic viscous time is larger than the flow characteristic time and as result the velocity profile throughout the jet keeps
changing from the needle orifice until the point of jet break up. There is a big difference between the liquid velocity at the surface and the liquid velocity in the jet centre for any jet’s radial section; the radial velocity profile is parabolic (Hartman, 1998). Therefore the is a substantial difference in the fluid-dynamics of the jet of distilled water and that of PVA water solutions, with considerable difference in the way in which charge is conducted through the jet. N.B. another factor influencing the spraying is the relationship between the tangential electric stress and the viscous stress at the gas-liquid interface. The viscous stress for Newtonian liquids is given:

\[ \tau_{z,r} = -\mu \frac{dv_z(z,r)}{dr} \]  
(5.4)

(Bird, 1960) where \( v_z \) is the velocity in the downstream direction. The viscous stress direction is opposite to the velocity direction and proportional to the liquid viscosity.

Table 5.1 Characteristic time constants calculated for water and PVA aqueous solutions.

<table>
<thead>
<tr>
<th>Time constant</th>
<th>Surrey tap water</th>
<th>Distilled water</th>
<th>PVA 3%</th>
<th>PVA 5%</th>
<th>PVA 7%</th>
</tr>
</thead>
<tbody>
<tr>
<td>( \tau ) (ns)</td>
<td>7</td>
<td>700</td>
<td>15</td>
<td>7.6</td>
<td>5.7</td>
</tr>
<tr>
<td>( \tau_{\mu} ) (s)</td>
<td>0.30</td>
<td>0.30</td>
<td>0.0076</td>
<td>0.0022</td>
<td>0.0013</td>
</tr>
</tbody>
</table>

The tangential component of the electric stress is given by:

\[ T_t = \sigma_q E_t \]  
(5.5)

where \( \sigma_q \) is the local surface charge density. The tangential component of the electric field can be evaluated. Ohm’s law gives the relationship between the current density vector, \( J \), and the electric field, \( E \):

\[ J = \sigma E \]  
(5.6)
If the current, $I$, passing through an open surface $S$ is defined as the net charge passing through this surface per unit time, then the current, $I$, and the current density vector $J$ are related as

$$ I = \int_S J \cdot ds $$ \hspace{1cm} (5.7)

or

$$ I = J \cdot S $$ \hspace{1cm} (5.8)

if $J$ is constant with respect to $S$. The relationship between electric field and current is provided by the combination of Equation (5.8) and Equation (5.6), giving

$$ E_t = \frac{I}{S\sigma} $$ \hspace{1cm} (5.9)

(Paul and Nasar, 1987). Using Equation 5.9 it is possible to evaluate the tangential component of the electric field acting on the liquid surface considering a constant section of the liquid jet; the values are reported in Table 5.2. The tangential values of the electric field for tap water are roughly three orders of magnitude larger than that of PVA 3%.

The jet density surface charge may be qualitatively evaluated considering that because the electric relaxation time is very short all the charge instantaneously reaches the surface by Coulombic repulsion, thus the current experimentally measured may be considered all on the jet surface. The assumption that all the current is on the jet surface and that there is only charge convection is quite crude because the current transported by the jet is due to conduction and convection (see Chapter 2). However, the part of the jet in which the conduction is a predominant phenomenon must be quite small compared to the part of the jet in which the major phenomenon is charge convection because of the short charge relaxation time. The surface charge is calculated using the equation below:
\[
\sigma_q = \frac{Ia}{2Q}
\]  
(5.10)


Table 5.2 Tangential component of the electric field at the liquid-gas interface for PVA 3% and distilled water.

<table>
<thead>
<tr>
<th>Applied potential (kV)</th>
<th>(E_t) for distilled water (N/C)</th>
<th>(E_t) for PVA 3% (N/C)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>105</td>
<td>0.2</td>
</tr>
<tr>
<td>2</td>
<td>157</td>
<td>0.25</td>
</tr>
<tr>
<td>3</td>
<td>263</td>
<td>0.3</td>
</tr>
<tr>
<td>4</td>
<td>420</td>
<td>0.37</td>
</tr>
<tr>
<td>5</td>
<td>463</td>
<td>0.4</td>
</tr>
<tr>
<td>6</td>
<td>600</td>
<td>0.5</td>
</tr>
<tr>
<td>7</td>
<td>684</td>
<td>0.55</td>
</tr>
</tbody>
</table>

The values of the surface charge are reported in Table 5.3 for PVA 3% and distilled water and they have been calculated considering the jet diameter is constant. The value of the jet surface charge may be compared for certain values of the applied potential with that calculated by Equation 5.11. Melcher (1963) observed that a liquid jet started whipping when the electric stress was larger than the surface tension stress multiplied by 0.3.

\[
R_{ts} = \frac{\sigma_q^2 a}{2\gamma \varepsilon_0}
\]  
(5.11)

The valued calculated for PVA 3% and distilled water using Equation 5.11 are circa \(2.5 \times 10^{-5}\) C/m² and circa \(2.6 \times 10^{-5}\) C/m² respectively and are comparable with that measured and reported in Table 5.3 when the jet started whipping.

The jet surface charge density for the two jets are of the same order of magnitude. However, the tangential component of the electric field for tap water is about three
times higher than that of PVA 3% thus the force exerted by the electric field on the water jet is of the order of $10^{-4}$-$10^{-3}$ N/m², while for the jet of PVA 3% the force due to the electric field is of the order of $10^{-7}$-$10^{-6}$ N/m². On the other hand the viscous stress opposing the electric tangential stress is proportional to the liquid viscosity and this is higher for PVA 3% than for distilled water. Hence, this clearly explains why the distilled water jet is more affected than the jet of PVA 3% by the electric field.

Table 5.3 Charge density on the jet surface for PVA 3% and distilled water; * for values of the applied potential superior to 7 kV the jet end of PVA 3% start to whip; ** for values of the applied potential superior to 6 kV the jet of distilled water exhibited lateral oscillation mixed with varicose instability.

<table>
<thead>
<tr>
<th>Applied potential (kV)</th>
<th>$\sigma_q$ for distilled water (C/m²)</th>
<th>$\sigma_q$ for PVA 3% (C/m²)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>$5.2 \times 10^{-6}$</td>
<td>$4.1 \times 10^{-6}$</td>
</tr>
<tr>
<td>2</td>
<td>$8.2 \times 10^{-6}$</td>
<td>$6.4 \times 10^{-6}$</td>
</tr>
<tr>
<td>3</td>
<td>$1.3 \times 10^{-5}$</td>
<td>$8.0 \times 10^{-6}$</td>
</tr>
<tr>
<td>4</td>
<td>$2.25 \times 10^{-5}$</td>
<td>$9.16 \times 10^{-6}$</td>
</tr>
<tr>
<td>5</td>
<td>$2.3 \times 10^{-5}$</td>
<td>$1.1 \times 10^{-5}$</td>
</tr>
<tr>
<td>6</td>
<td>$3.0 \times 10^{-5}$**</td>
<td>$1.22 \times 10^{-5}$</td>
</tr>
<tr>
<td>7</td>
<td>$3.5 \times 10^{-5}$ **</td>
<td>$1.35 \times 10^{-5}$ *</td>
</tr>
</tbody>
</table>

Another point which needs to be explained is the effect of the charges upon the liquid jet break up and hence on the droplet diameter to jet diameter ratio. This is quite a controversial issue because as already reported in literature there are a number of researchers such as Cloupeau and Prunet-Foch (1989), Gomez and Tang (1991), Schneider et al. (1967), Neukermans (1973) and Mutoh et al. (1979) who have claimed that the break up of a liquid jet is not influenced by a surface charge, while other groups of researchers, for example Bailey and Balachandran (1981), Hartman (1998) and Huneiti (2000) have observed an influence of the charges on the liquid jet break up. In this work it has been observed that there is a deviation of the droplet diameter to jet diameter ratio as the potential is increased beyond a certain level. In the varicose regime for distilled water with an applied potential of 5 kV the droplet diameter to jet diameter ratio is about 1.5 and not 1.89 as expected by Rayleigh’s theory. A deviation
of the droplet diameter to jet diameter ratio has also been recorded for PVA 3%. However, this deviation is very much constrained with respect to that recorded for distilled water. This may be explained by comparing the two surface charges measured, see Table 5.3. The jet of PVA 3% has a surface charge lower than that of the jet of distilled water and so the droplet diameter to jet diameter ratio is less affected in the jet where the surface charge is lower. As seen the surface charge is due to the current transported by the jet and the amount of current transported depending on the physical properties of the liquid such as viscosity and dielectric constant. Therefore it may be concluded that the charges do have an effect on the droplet diameter to jet diameter ratio and this effect is more evident in a liquid jet where: a) a large current can be transported, b) a large surface tension can hold a large charge surface density before kinking takes place.
Chapter 6 - Liquid jet break-up by pulsating DC potential

6.1 Introduction

Liquid jet break up occurs by the growth of natural disturbances in the absence of other influential factors. As seen in the previous chapters the application of an electric field to the liquid jet generates a tangential stress on the liquid surface, and if a time-varying field is applied then an artificial disturbance is generated. The drop frequency is related to the wave length of the disturbances. Therefore, the idea behind the use of a pulsating DC potential is to control the droplet size distribution by controlling the wavelength of the axisymmetrical waves, which travel and grow along the jet, leading to the disintegration of the liquid jet into droplets. A pulsating DC potential is applied to the nozzle from which the liquid jet is emerging. It has been observed that there is a range of pulsating DC frequencies within which it is possible to control the wavelength of the axisymmetric waves, $\lambda$, which in turn controls the droplet and satellite size distribution, see Figure 6.1. Within the range of pulsating DC frequencies where the control is possible, it is observed that, for each pulsating DC cycle a droplet may be formed. The range of frequencies over which the above phenomenon is observed is termed the synchronous frequency region.

It is well-known that highly conductive liquids such as tap water can be sprayed in a controlled fashion by the use of an AC potential (Sato, 1984) or by an AC potential superimposed on a DC potential (Huneiti et al, 1998); these authors reported the existence of an optimal spraying set-up, applied potential, flow rate and range of frequencies within which it is possible to generate monodispersed droplets. The formation of monodispersed droplets still only occurs within the synchronous region, whose limits depend on a number of variables such as the liquid flow rate, geometrical configuration of the spraying set-up, potential and physical properties of the atomised liquid, e.g. density, viscosity and surface tension.

In this work, the limits of the synchronous region are explored for tap water, PVA 3%, PVA 5% and PVA 7% solutions. Two different electrode types have been
used and a pulsating DC potential of 4 kV or 8 kV has been applied to the needle. Throughout all the experiments, the pulsating DC field has been kept on for a time equal to half of the period. Also, two flow rates have been used: 32.2 and 60 ml/min for the 2A and the 1A electrode configurations, respectively.

Figure 6.1 Water jet wave for applied frequencies of 230 Hz, 190 Hz and 150 Hz and an applied potential of 8 kV using the electrode configuration 1A.
6.2 Effect of frequency on the droplet size distribution

The droplet and satellite size can be controlled by coupling the axisymmetrical waves with a pulsating DC field. This is done by applying a pulsating DC field onto the needle of an appropriate spraying set-up. Two set-ups have been used to investigate the possibility and limitation of controlling the droplet and satellite size and these are the electrode configurations 1A and 2A. The results with respect to four types of liquids with different viscosities are described below. Also in Figures 6.2-6.7 the droplet diameters measured are compared with the numerical data given by Equation 6.3. This equation was given by Sato (1984) and the derivation of Equation 6.3 is reported in paragraph 6.4.

6.2.1 Tap water

The atomisation of tap water can be controlled in the range of frequencies between 90 to 220 Hz using the 1A electrode configuration and between 60 to 240 Hz using the 2A electrode configuration, as shown in Figures 6.2 and 6.3. In both cases, increasing the frequency reduces the droplet diameter, but not substantially i.e. from about 2.5 mm to 1.8 mm. The lower value of the droplet diameter relates to the smaller nozzle used (2A electrode configuration) and an applied frequency of 220 Hz. As well as reducing the size of the droplet, the satellite size is also reduced as the frequency increases. The satellite diameter was reduced from a value of 1.5 mm to a value of 0.5 mm. Once again the lowest value is recorded for an applied frequency of 220 Hz. Satellites are always produced in this case, and near the lower value of the applied frequency the satellite droplets can be accompanied by secondary satellites. The 1A electrode configuration produces larger droplets than the 2A electrode configuration. This is as expected; the larger the jet diameter, the larger the volume of the droplet produced. Thus, the larger the droplet diameter will be when the same frequency is applied.
Figure 6.2 Droplet diameter produced as a function of the applied pulsating DC frequency for tap water using the 1A electrode configuration, an applied pulsating DC potential of 8 kV and a flow rate of 60 ml/min. No secondary satellites are recorded. ●: main droplet; ■ satellite; ————: Sato (1984).

Figure 6.3 Droplet diameter produced as a function of the applied pulsating DC frequency for tap water using the 2A electrode configuration, an applied pulsating DC potential of 4 kV and a flow rate of 32.2 ml/min. ●: main droplet; ■ satellite; + secondary satellite; ————: Sato (1984).
6.2.2 PVA 3%

Increasing the liquid viscosity causes a drastic reduction of the range of frequencies within which the spraying can be controlled. The lower and upper limits of the frequency range for PVA 3% are about 40 Hz and 155 Hz, minimum and maximum respectively. Between the 1A and the 2A electrode configuration only small changes in the minimum frequency limit are recorded (see Figure 6.4 and 6.5). PVA 3% behaves similarly still to tap water in that an increase of the applied frequency leads to a reduction of the droplet and satellite diameters, but in this case the effect on the satellite diameter is significant. In fact, when the atomisation of PVA 3% is performed using the 1A electrode configuration, satellite droplets are always present throughout the applied frequency range, and the satellite diameter changes between 2 mm to 0.8 mm for the minimum and maximum applied frequencies respectively.

![Graph](image)

Figure 6.4 Droplet diameter produced as a function of the applied pulsating DC frequency for PVA 3% using the 1A electrode configuration, an applied pulsating DC potential of 8 kV and a flow rate of 60 ml/min. ●: main drop; ■ satellite; + secondary satellite; —— Sato (1984).
Figure 6.5 Droplet diameter produced as a function of applied pulsating DC frequency for PVA 3% using the 2A electrode configuration, an applied pulsating DC potential of 4 kV and a flow rate of 32.2 ml/min. ●: main drop; ■: satellite; +: secondary satellite; ——— Sato (1984).

If the atomisation of PVA 3% is performed using the 2A electrode configuration the satellite droplets can be totally eliminated. By applying a frequency between 60 Hz to 80 Hz, the diameter of the satellite decreases gradually, but beyond 80 Hz the satellite diameter decreases sharply, until it completely disappears at frequencies over 100 Hz, and a train of monodispersed droplets is produced, as shown in Figure 6.5.

6.2.3 PVA 5%

A further increase in the liquid viscosity leads to a further reduction of the frequency range for which the atomisation can be controlled. For PVA 5% solution, the controlling frequency range is 30 to 70 Hz for 1A electrode configuration, and 30 Hz to 110 Hz for the second electrode configuration. However, the spraying of PVA 5% by the 1A electrode configuration is difficult to control. Although the spraying process has been carried out up to an applied frequency of 105 Hz, the droplet diameter can hardly be controlled above 70 Hz. A consequence of this weak control on the spraying is a
large scattering in the data of the droplet and satellite diameter recorded, see Figure 6.6. Therefore, it can be concluded that the 1A electrode configuration is inadequate to spray liquids with physical properties such as PVA 5%. This because by using the electrode configuration 1A it is not possible to generate an electric force strong enough to drive the axisymmetrical wave on a liquid jet with such viscosity.

![Figure 6.6](image)

Figure 6.6 Droplet diameter produced as a function of the applied pulsating DC frequency for PVA5% using the 1A electrode configuration, an applied pulsating DC potential of 8 kV and a flow rate of 60 ml/min. •: main drop; ■ satellite; + secondary satellite; ——— Sato (1984).

The spraying of PVA 5% can however be successfully performed using the 2A electrode configuration with which the control on the droplet and satellite diameters is straightforward over the entire range of applied frequencies used and, as for PVA 3%, there is a frequency beyond which no satellite is produced. Starting from a frequency of 30 Hz, a small gradual decrease of the satellite diameter can be observed until the applied frequency reaches 60 Hz, over which the satellite is suddenly not produced at all during the spraying, see Figure 6.7.
Figure 6.7 Droplet diameter produced as a function of the applied pulsating DC frequency for PVA 5% using the 2A electrode configuration, an applied pulsating DC potential of 4 kV and a flow rate of 32.2 ml/min. ●: main drop; ■ satellite; + secondary satellite; ———— : Sato (1984).

6.2.4 PVA 7%

The spraying of PVA 7% has not been possible to perform. The electrode configuration 2A has been found not to operate satisfactorily when PVA 7% is used as the spraying liquid. A few seconds after the electric field is turned on the liquid jet is suddenly attracted onto the plate, the jet latches on the plate, and therefore, a short circuit is produced as shown in Figure 6.8. The attraction of the liquid jet on to the earthed plate may be caused by an imbalance of force around the jet in the region close to the hole edge (see Figure 6.9). This is also observed for a liquid jet of PVA 3% and PVA 5%, (see Figures 6.17 and 6.19) The difference, which exists between the other polyvinyl alcohol solutions and PVA 7%, is that in the latter case the jet is deflected until it sticks to the earthed plate. However, because the 2A electrode configuration does not permit a stable spraying for PVA 7%, it has been considered worth modifying the electrode configuration.
Needle of the 2A electrode configuration.

Plate of the Second electrode configuration.

Jet of PVA 7%.

Drop hanging from the plate.

Figure 6.8 Spraying of PVA 7% with the 2A electrode configuration and a pulsating DC potential of 4 kV, showing the control of the liquid jet with the edge of the plate.

Figure 6.9 Qualitative representation of the electric field lines around the diverted liquid jet.

The modification consists of enlarging the hole inside the earthed plate from 0.6 cm to 1 cm in diameter. The idea behind the hole enlargement is to increase the distance between the surface of the jet of PVA 7% and the edge of the hole in such a way as to reduce the attraction between the jet and the earthed plate. When the atomisation of PVA 7% has been performed using the 2A electrode configuration modified as above described, the phenomenon of attraction between the liquid jet and
the earthed plate was still observed. The spraying of PVA 7% lasted a few more seconds but eventually the jet makes contact with the plate.

The very short time within which the liquid jet of PVA 7% was stable and broke up into droplets under the action of the pulsating DC potential was sufficient to allow high speed video records to be taken. These images are just enough to observe that during the short time in which the spraying of PVA 7% is stable, the presence of the satellite has been always seen, thus the 2A electrode configuration with or without the plate modification is inadequate to spray the PVA 7% in a controlled fashion. Therefore, despite the above modification, the problem of attraction between the jet of PVA 7% and the earthed plate remains unsolved.

6.2.5 Frequency effect on size distribution

It can be summarised that inside the synchronous region it is possible to vary the main droplet diameter by 20%-30% depending mainly on the liquid viscosity. The higher control on the droplet diameter refers to the high liquid viscosity. Using an appropriate electrode set-up and applied potential it is possible to control the droplet and satellite size distribution. The size distributions for tap water, PVA 3% and PVA 5% vary for a number of applied frequencies as shown in Figures 6.10 and 6.11 for the 2A electrode configuration. For tap water at low frequencies, e.g. 80 Hz, a trimodal distribution is produced, as shown in Figure 6.10; the three peaks relate, from right to left, to the droplet, the satellite and the secondary satellite. The number percentages of droplets, satellites and secondary satellites produced during the process of jet break up are also reported in Figure 6.10. The number of droplets is almost equal to the number of satellites and secondary satellites produced, since in each break-up process a droplet is usually accompanied by a satellite or a satellite plus a secondary satellite. However, during the break-up process it is possible that only the droplet is produced. This explains why the number of droplets is larger than the other two droplets. The increase in the applied frequency leads to a reduction of the droplet diameter and satellite diameter as well as to a net change in their size distribution. A typical size distribution for high applied frequencies, e.g. 220 Hz, is show in Figure 6.11. The distribution is bimodal since secondary satellites are no longer produced. Also, the number of
satellites is drastically reduced, because (a) in each break up process occurring at high applied frequencies the satellite droplets are less likely to be produced or because (b) as soon as the satellite is produced it may coalesce with the droplet. The number percentage of droplets and satellites produced are presented in Figure 6.11. Under these conditions of applied frequency, almost 90% of the liquid is in the form of droplets (i.e. no satellites). N.B. in Figure 6.12 the size distribution of PVA 3% with an applied frequency of 80 Hz is shown, where a bimodal distribution is observed. The number of droplets recorded is much larger than the number of satellites. This is in contrast with what was observed in the atomisation of water using the same spraying condition. In that case, the number of satellites and secondary satellites produced is close to the number of droplets produced in each break-up process. Therefore, it can be concluded that an increase in the liquid viscosity makes the formation of satellites and secondary satellites less likely to happen for the same applied frequency. In particular, no secondary satellites have been recorded. The percentage of droplets and satellites produced during the process of jet break-up for PVA 3% are reported in Figure 6.12.

![Figure 6.10 Size distribution of tap water droplets using the 2A electrode configuration, with a flow rate of 32.2 ml/min, an applied potential of 4 kV and an applied frequency of 80 Hz.](image)
Figure 6.11 Size distribution of tap water droplets using the 2A electrode configuration, with a flow rate of 32.2 ml/min, an applied potential of 4 kV and an applied frequency of 220 Hz.

An increase of the applied frequency in the spraying of PVA 3% leads to a reduction of the droplet diameter and the satellite diameter. Furthermore, the satellite formation ceases above a frequency, e.g. 150 Hz as given in Figure 6.13. The disappearance of the satellites is due to a phenomenon described in Section 6.3. In each break-up process occurring at a high applied frequency, e.g. 130 Hz, a liquid filament is present between the forming droplet, and this filament recoils into the drop each time the forming droplet is pinched off. Therefore no satellites are produced and more than the 98% of the droplets produced have a diameter between 1.7 and 1.9 mm. A typical distribution recorded for the spraying of PVA 3% at an applied frequency of 150 Hz is reported in Figure 6.13. Measurement of the size distribution has also been performed for the spraying of PVA 5% under two applied frequencies of 50 Hz and 80 Hz. Once more, an increase in the applied frequency leads to a dramatic change in the size distribution. The size distribution at 50 Hz is bimodal, and this becomes monomodal as soon as the applied frequency goes to about 55 Hz. The characteristic distributions for the above applied frequencies, with respect to the spraying of PVA 5%, are reported in Figure 6.14 and 6.15.
Comparing the results shown in Figures 6.3, 6.5 and 6.7, where the droplet and satellite diameters are shown as a function of the applied frequency for the spraying of tap water, PVA 3% and PVA 5% using the same electrode configuration and applied potential e.g. 4 kV, it can be observed that an increase in the viscosity of the liquid leads to a more marked change of the diameter of the droplets and satellites as the frequency is changed. For tap water the change of the droplet and satellite diameters occurs over a range of frequencies between 60 Hz to 180 Hz, while for PVA 3% this occurs over a range of frequency between 60 Hz to 150 Hz, and for PVA 5% the range of frequency shrunk to between 30 Hz to 105 Hz. The viscosity increase also has a strong effect on the size distribution. In Figures 6.10, 6.12 and 6.15 it is reported how the size distribution changes for droplets and satellites for the same electrode configuration used, the same applied potential as well as the same applied frequency and flow rate. It can be seen that for liquids with very low viscosities, e.g. water, droplets, satellites and secondary satellites are produced, while for more viscous liquids, e.g. PVA 3%, only droplets and satellites are produced. In case of PVA 5% only droplets are produced. Therefore, an increase in the liquid viscosity inhibits the formation of satellites and secondary satellites for the same applied frequency.
Figure 6.13 Size distribution of PVA 3% droplets using the 2A electrode configuration, flow rate 32.2 ml/min, an applied potential of 4 kV and an applied frequency of 150 Hz.

Figure 6.14 Size distribution of PVA 5% droplets using the 2A electrode configuration, flow rate 32.2 ml/min, an applied potential of 4 kV and an applied frequency of 50 Hz.
Figure 6.15 Size distribution of PVA 5% droplets using the 2A electrode configuration, flow rate 32.2 ml/min, an applied potential of 4 kV and an applied frequency of 80 Hz.
6.3 Effect of frequency on the liquid jet break-up

The liquid jet break-up is driven by a pulsating DC potential applied to the nozzle from which the liquid is issued. The disruption of the liquid jet into droplets is due to a growing axisymmetrical wave which travels on the jet and induces its break-up. The axisymmetrical waves are affected by the DC potential pulses. In this way, by controlling the frequency it is possible to control the wavelength of the axisymmetrical wave. The mechanism of droplet formation is shown in this section for a number of applied frequencies of the pulsating DC potential for tap water and PVA solutions with different viscosities.

The spraying of tap water is characterized by the production of droplets, satellites and in some cases secondary satellites. For low applied frequencies, e.g. 60 Hz to 100 Hz, the wavelength of the axisymmetrical wave is large enough to make a long liquid filament between two consecutive wave peaks, whose break up creates satellites and secondary satellites. When higher frequencies are applied, e.g. 210 Hz (see arrow Figure 6.16) the break up will create only satellites, which for very high frequencies are likely to be absorbed by either the droplet just formed or by the liquid jet. In this section the bumps and the liquid filament are named using the letters a, b and c for better describing the mechanism of break up observed.

Figure 6.16 Atomisation of tap water, using a pulsating DC potential of 8 kV, a frequency of 210 Hz and flow rate of 60 ml/min.
The increase of the liquid viscosity makes a remarkable change in the satellite and secondary satellite formation. Figure 6.17 and 6.18 show the jet break-up of PVA 3% for an applied frequencies of 130 Hz and 60 Hz, respectively. For a frequency of 130 Hz no satellites are produced. When the applied frequency is reduced to 60 Hz the jet break-up produces droplets and satellites and in addition the liquid jet starts to oscillate. The jet oscillation may be due to an asymmetry of the electric field acting around the liquid jet. Especially when the 2A electrode configuration is used, even a very small asymmetry in the spraying set-up may generate asymmetry in the electric field applied around the jet, (see Figure 6.9). The electric field is responsible for the forces acting on the jet, thus an asymmetry in the electric field will generate an asymmetry in the forces acting on the jet. In Figure 6.17 the PVA 3% liquid jet does not follow the vertical direction, this may be due to the force asymmetry acting on the jet as described above. However, due to the inertial mass of the liquid, there is a time dependence of the fluid deflection as the electric field is pulsed on and off. At high frequencies, e.g. 130 Hz, the jet is not able to return to the vertical position as it passes through the plate, so the liquid jet remains permanently deflected away from the vertical position. For relatively low frequencies, e.g. 60 Hz, the liquid jet has time to return to the vertical position, and so it oscillates, see Figure 6.18, as it occurs in a “kink instability”.

The spraying process of PVA 3% in a controlled fashion is clearly visible in Figure 6.17. For an applied frequency of 130 Hz, the liquid thread (b), which holds the forming droplet (c), relaxes back into the tip of the jet (a) as soon as droplet (c) is pinched off from it, and therefore no satellites are produced. Satellites can be formed when lower frequencies are applied. The process for satellites’ formation is clearly visible in Figure 6.18, whereby the liquid thread (b) which is present between the formed droplet (c) and the end of the jet (a) breaks up into two satellites. The spraying of PVA 5% in a controlled fashion using the 2A electrode configuration is also possible, as shown in Figure 6.19. The formation process is entirely identical to the one seen for PVA 3%. A detailed sequence of the process for PVA 5% is shown in Figure 6.20. While the forming droplet (c) starts to depart from the liquid jet, the liquid thread (b) begins to become thinner (see record number 1 to 7) until it reaches a limiting thickness (see record number 8). At this stage the liquid thread breaks up from the forming droplet, and the thread relaxes back into the liquid jet in a few milliseconds.
(see record numbers 8 to 12). Then the process of the droplet formation starts again. As stated above, the 2A electrode configuration has a limit with respect to the viscosity of the liquid. Figure 6.21 shows the liquid jet break up of PVA 7% using the modified 2A electrode configuration. As it can be seen, a very long filament is present between two consecutive bumps whose break up will produce quite large droplets which will be accompanied by a number of satellite and secondary satellites that are produced from the disintegration of the above liquid filament. This phenomenon is recorded throughout the range of applied frequencies for this concentration of PVA.

Figure 6.17 Atomisation of PVA 3% using a pulsating DC potential of 4 kV, frequency of 130 Hz and a flow rate of 32.2 ml/min.

Figure 6.18 Atomisation of PVA 3% using a pulsating DC potential of 4 kV, frequency of 60 Hz and a flow rate of 32.2 ml/min.
Another effect of the applied pulsating DC potential is that of reducing the length of the liquid jet. A liquid jet made of PVA 3% flows downstream for circa 2.2 cm before starting to break up; the flow rate used is 32 ml/min. When a DC pulsating potential is applied to the nozzle with a frequency of 60 Hz, the jet length is shortened to about 2.0 cm. A further increase in the applied frequency leads to an additional reduction of the jet length. For a frequency of 130 Hz the jet length is about 1.5 cm. Therefore, as a result of the increased frequency the jet length may reduce to almost half of the jet length recorded under no electric field, see Figure 6.18 and Figure 6.22. The jet shortening due to the frequency increase is discussed in Section 6.7.
Figure 6.21 Spraying of PVA 7% using a pulsating DC potential of 4 kV, a frequency of 65 Hz and a flow rate 32.2 ml/min.

Figure 6.22 Spraying of PVA 3% under no electric field applied and a flow rate 32 ml/min.
Theoretical analysis of the effect of frequency on the droplet diameter produced

The effect of the applied frequency on the droplet diameter produced can be determined by a mass balance on the system as shown a schematic diagram in Figure 6.23. If we consider that a liquid with density $\rho$ is fed through the needle with a flow rate $Q$ (IN), and the liquid disintegrates into uniform droplets of volume $V_d$ with a frequency of droplet per second $f_d$ (OUT), then the mass balance can be written as:

$$Q\rho = V_d f_d \rho$$  \hfill (6.1)

Figure 6.23 A sketch of spraying in jet mode to illustrate the conservation of mass.

Within the synchronous region the applied frequency, $f_a$, and the droplet formed per second, $f_d$, are in a one-to-one relationship (see next Section 6.5). Therefore Equation 6.1 can be written in terms of droplet diameter, $D_d$, as follows:
\[ Q \rho = \frac{\pi D_d^3}{6} \rho f_a \]  \hspace{1cm} (6.2)

Rearranging Equation 6.2 with respect to the droplet diameter, it is possible to work out the relationship between droplet diameter and the frequency within the synchronous region, as shown in Equation 6.3, given by Sato (1984).

\[ D_d = \sqrt[3]{\frac{Q \rho}{\pi f_a}} \]  \hspace{1cm} (6.3)

Equation 6.3 plotted as a continuous line in Figures 6.2 and 6.7. Although in Equation 6.3 the effect of the satellite is neglected, the agreement between experimental results and Equation 6.3 is good and the general trend is the same for experimental and calculated results.
6.5 **Effect of the pulsating DC frequency on the droplet formation frequency**

The phenomenon of synchronization between the droplet formation and the applied frequency of the pulsating DC potential occurs when an appropriate spraying set-up is adopted and a suitable value of the potential is used (Sato, 1984). The limits of the synchronization region are strongly related to the physical properties of the liquids atomised as well as to the spraying set-up. In all the experiments performed it has been observed that by increasing the applied frequency a reduction in the wavelength of the axisymmetrical waves is observed until a certain point over which no further wavelength reduction is observed and the wavelength becomes poorly defined. On the other hand reducing the frequency produces larger wavelengths until axisymmetrical instability due to "kink instability" sets in.

The reason why for low frequency applied "kink instability" sets in has already been commented on Section 6.3. To understand the reason why an increase in the applied frequency beyond a certain value led to poorly defined wavelength, it was suggested by Balachandran (2001) to determine how the frequency of the droplet changes with the applied frequency. This was done by performing measurements using the 2A electrode configuration where a good control of droplet formation for tap water, PVA 3% and PVA 5% was possible. Each of these liquids has a characteristic frequency range within which it is observed that the relationship between the applied frequency and the droplet frequency is one-to-one, as shown in Figure 6.24. This relationship is valid for tap water in the frequency range 60 to 220 Hz. Over 220 Hz an increase in the frequency does not linearly produce an increase in the droplet frequency. Rather the droplet frequency seems to reach a plateau. The same phenomenon is observed when PVA 3% and PVA 5% are atomised, but in these cases the deviation of the droplet frequency from the applied frequency occurs at about 155 Hz and 115 Hz for PVA 3% and PVA 5%, respectively, as shown in Figure 6.24.

The reason why an increase in the applied frequency over a certain value does not cause any further linear increase in the droplet frequency can be explained by analysing Figure 6.25. Following the record numbers written on Figure 6.25, it can observed from record number 1 to number 15 that the axisymmetrical wave travels
regularly along the jet with a well-defined wavelength. In number 20 the regular wave pattern suddenly changes, as a bump (b) is observed to travel faster than the one (a) that is preceding it. In record numbers 24-26 the faster bump (b) is merging with the bump (a) well before the jet break-up occurs. Record number 27 shows the jet breaks up and creates a droplet whose shape can be assimilated to a Cassini oval prolate. In record number 28, the oval has become a spheroid and the coalescence is completed. From record numbers 29 to 31, the axisymmetrical wave starts again to regularly travel along the jet with a well defined wavelength.

Figure 6.24 Droplet frequency as a function of the applied pulsating DC frequency. Using the 2A electrode configuration, an applied pulsating DC potential of 4 kV and a flow rate of 32.2 ml/min. ▲: water; □: PVA 3%; ■: PVA 5%; ——: one-to-one relationship.
Therefore, from two bumps (a) and (b) a single droplet is produced. This phenomenon occurs periodically and more frequently as the frequency increases. So, for the higher frequency, this phenomenon is responsible for the mismatch between applied frequency and the droplet frequency. The phenomenon just described for a liquid jet of water is
also observable with the same modality in the case of PVA 3% and PVA 5%. In the case of PVA 5%, a detailed sequence of the coalescence or collision or bunching-up between two bumps within the liquid jet is given in Figure 6.26.

Figure 6.26 Spraying of PVA 5% by a pulsating DC potential with a frequency of 130 Hz, where waves starting later in time catch up with earlier waves.
6.6 **Effect of frequency on the wavelength of the axisymmetrical wave**

The wavelength of the axisymmetrical wave has been measured as a function of the frequency within the synchronous region for tap water, PVA 3%, PVA 5% and PVA 7%. For all the liquids tested and applied frequencies whose value is falling inside the synchronous region, an increase in the applied frequency leads to a monotonic reduction in wavelength until the wavelength seems to reach a plateau. For tap water with 1A electrode configuration, the wavelength of the axisymmetrical waves can be controlled using frequencies ranging between 90 and 220–240 Hz. For these values of frequency the wavelength changes from approximately 15.3 mm to 5.2 mm. Over 240 Hz no further wavelength reduction occurs and rather the wavelength becomes ill-defined. The measurement of wavelength as a function of the frequency has also been made for PVA 3% using the 1A electrode configuration. In this case, the synchronous region ranges from 45 Hz to 155 Hz and between these frequencies the minimum and maximum wavelength recorded is about 6.4 mm and about 24.3 mm respectively. As observed for tap water, there is also a maximum frequency for PVA 3% of 155 Hz, over which the wavelength becomes indefinable.

The liquid viscosity increase leads to a reduction of the frequency limits of the synchronous region. Also the liquid viscosity increase leads to the observation of a longer wavelength for the maximum value of the frequency of the synchronous region. PVA 3% and tap water have two characteristic synchronous regions, which are overlapping each other for frequencies between 90 Hz and 155 Hz. Within the overlapping region, the recorded tap water wavelengths are larger than those measured for PVA 3%. This suggests that the waves travelling along the liquid jet of water are faster than the waves travelling on the jet of PVA 3%. The reason for this is given by Equation 6.4, where the wavelength, $\lambda$, the frequency, $f$ and velocity, $v_w$ of a wave are linked together.

$$\lambda = \frac{v_w}{f} \quad (6.4)$$
Therefore, if we record the wavelength of two waves having the same frequency the faster of the two will also have the longer wavelength. According to Nakoryakov et al. (2000), the velocity of capillary waves moving on a film surface is slowed down by the increased viscosity. In Section 6.5, it has been shown that within the synchronous region the relationship between the applied frequency and the droplet frequency is one-to-one, i.e. the applied frequency of the pulsating DC potential and the frequency of the axisymmetrical wave are equal. In these experiments, at the same applied frequency an increase in liquid viscosity produces a reduction of the wavelength, and this means that there is a reduction in the wave velocity, (see Figure 6.27).

![Graph](image)

Figure 6.27 Liquid jet wavelength as a function of the applied pulsating DC frequency for tap water and PVA 3% using the 1A electrode configuration, an applied pulsating DC potential of 8 kV and a flow rate of 60 ml/min. ○: tap water; ⊕: PVA 3%.

Further measurements of the wavelength as a function of the applied frequency have been carried out using tap water, PVA 3% and PVA 5%, and employing the 2A electrode configuration. It has been observed that, the higher the viscosity of the liquid, the lower the wave velocity, and consequently, the shorter is the wavelength for the same applied frequency. This is valid within a range of frequencies determined by the overlapping of two synchronous regions. The phenomenon is very well observable
when comparing tap water with PVA 3%, see Figure 6.27, but it is less evident when comparing PVA 3% with PVA 5%. The reason for this is that there is a difference of viscosity of 40 times between tap water and PVA 3%, while between PVA 3% and PVA 5% there is a difference of viscosity of 3 times. Therefore any possible effect on the wavelength is not detectable. Also, when using the 2A electrode configuration especially for high values of the frequency, the jet length is about twice the wavelength at which instability occurs making the wavelength measurement less precise. Once more, an increase in the liquid viscosity leads to a reduction of the limits of the synchronous region. The limits of the synchronous region for PVA 5% are 30 Hz and 115 Hz. The corresponding wavelength values are 26.3 mm and 7.3 mm, respectively. For PVA 3% the change of the electrode geometry from 1A to 2A does not seem to significantly affect the limits of the synchronous region. However, the minimum and maximum values of the wavelength change between the 1A and the 2A electrode configuration. This may be due to the difference in nozzle diameter that exists between the 1A and the 2A electrode configuration.

Figure 6.28 Wavelength versus applied pulsating DC frequency for PVA 3% and PVA 5% using the 2A configuration, an applied pulsating DC potential of 4 kV and a flow rate of 32.2 ml/min. +: PVA 3%; ○: PVA 5%.
The minimum and maximum wavelengths with respect to the radius of the jet, $r_j$, are reported in Table 6.1. It can be observed that an increase in the liquid viscosity leads to an increase in the minimum and maximum values of the wavelength. Also, the minimum and the maximum wavelengths shift to lower values when the nozzle diameter used in the spraying is reduced.

Table 6.1 Minimum and maximum wavelengths recorded within the synchronous region for each liquid and electrode configuration. The $r_j$ has been measured on the liquid jet before that any liquid instability has taken place.

<table>
<thead>
<tr>
<th>Liquid</th>
<th>Minimum wavelength</th>
<th>Maximum wavelength</th>
</tr>
</thead>
<tbody>
<tr>
<td>Water *</td>
<td>9.4 $r_j$</td>
<td>28 $r_j$</td>
</tr>
<tr>
<td>Water **</td>
<td>8.2 $r_j$</td>
<td>22 $r_j$</td>
</tr>
<tr>
<td>PVA 3% *</td>
<td>11.8 $r_j$</td>
<td>48 $r_j$</td>
</tr>
<tr>
<td>PVA 3% **</td>
<td>8.8 $r_j$</td>
<td>22.5 $r_j$</td>
</tr>
<tr>
<td>PVA 5% **</td>
<td>14.6 $r_j$</td>
<td>52.6 $r_j$</td>
</tr>
</tbody>
</table>

* 1A electrode configuration used.

** 2A electrode configuration used.
6.7 Discussion

The use of a pulsating DC potential combined with an appropriate electrode geometry enable the spraying of a liquid in a controllable fashion producing monosized droplets. The effect of the applied pulsating DC potential depends on liquid conductivity. The charge relaxation time for the liquid used is of the order of $10^{-9}$-$10^{-7}$ s while the fastest frequency applied is about 250 Hz. At this frequency the electric potential is switched on for a duration of about $10^{-3}$ s. This is long enough for a full effect of the electric field upon the spraying. N.B. The DC pulse has got a high peak just at the beginning of the cycle with a duration time of about $10^{-4}$ s. Hence, because of the very short charge relaxation time of the liquids used, the peak, despite being quite short, has a full effect on the liquid jet break-up regime. However, monosized droplet production is possible only within a window of frequencies located towards the higher values of the frequencies and it is also related to the liquid physical properties; for liquids such as tap water it was not possible to find a window of frequencies in which the satellite formation could be avoided. In particular the liquid viscosity influences the limit of the frequency windows within which no satellites are formed. When the liquid viscosity increases the window of synchronous frequency shrinks. Furthermore an increase in viscosity reduces the frequency at which the satellite is not produced any longer. A number of researchers Yuen (1968), Rutland and Jameson (1970) and Lafrance (1975) have investigated the satellite formation in terms of wave number. However their results differ in the value of the wave number beyond which no satellites are produced; the value of the wave number beyond which no satellite is produced is called the cut-off wave number. In this work it has been observed that the cut-off wave number is dependent on the liquid viscosity. A cut-off wave number of about 0.8 exists for PVA 3%, and a cut-off wave number of about 0.55 exists for PVA 5%. No cut-off wave number has been recorded for tap water. The dependency of the cut-off wave number on the liquid viscosity may by explained as follows. When the wave number is sufficiently small the liquid filament present between the forming droplet and the jet is large enough to detach as a single little droplet (satellite) as depicted in Figure 2.6 and 2.7, (Goeddel and Yuen, 1970). By increasing the wave number the liquid filament becomes shorter until a value of the wave number is reached over which the filament starts to be sucked into the liquid jet; a photographic sequence of the mechanism is reported in Figure 6.20. The liquid filament recoiling is due to the
surface tension which is able to work against the gravity force and overcome the weight of the liquid filament when this is small enough. Once the liquid jet is electrified the surface charge density and the electric field superimposed upon the space-charge field comes into play. The surface charge density and the electric field superimposed upon the space-charge field generates a pressure which is working against the surface tension. Hence, on the surface of the electrified jet it is possible to reach values of surface charge which are able to stop the liquid filament recoiling and thus the liquid filament rolls itself up into a satellite. The amount of charge present on an electrified jet depends on a number of factors including the viscosity. As already seen the increase in viscosity may induce a lower amount of current conducted through the jet, leading to a lower surface charge. Thus, between two liquids having the same surface tension but different viscosities the more viscous liquid has the lower surface charge density and this leads to a lower cut-off wave number. In the same way, it can be explained why no cut-off wave number was recorded for tap water. Being tap water with relatively low viscosity, the current conducted and so the charge surface density cannot be "mitigated" by the viscosity, hence the liquid filament is destabilised and induced to form a satellite for any wave number applied, i.e. no cut-off wave number exist.

The viscosity has also an effect on the droplets' size distribution. In Figure 6.10, 6.12 and 6.15 the droplet size distributions for tap water, PVA 3% and PVA 5% are given respectively, using the same electrode geometry, applied potential and applied frequency. When the viscosity increases the distribution modality changes from trimodal for tap water, to bimodal for PVA 3% and finally become monomodal for PVA 5%. Modality change is related to viscosity in the following way. The velocity of the wave travelling on the liquid jet is inversely proportional to the liquid's viscosity. Hence for the same applied frequency, the lower the viscosity, the larger the wave velocity. According Equation 6.4 the longer the wavelength, the longer the liquid filament between forming droplet and liquid jet. For an applied frequency of 80 Hz the liquid wavelength for tap water is roughly 11 mm, while for PVA 3% and PVA 5% is 7.9 mm and 7.5 mm, respectively. Therefore, when the water droplet forms the liquid filament it is less likely to be absorbed by the jet and it may then break up to form either a satellite or a satellite and a secondary satellite. When a droplet made of PVA water solution forms, the liquid filament becomes shorter than that of tap water and it is more likely to be either absorbed by the liquid jet or to form a single satellite depending
by the liquid viscosity. In this way when the liquid viscosity increases a narrower
distribution is recorded.

The pulsating DC frequency has in addition the effect of stabilising the liquid
jet. It has been experimentally observed that when the electrode configuration 2A is
used in order to spray water and a constant DC potential is applied, the liquid jet starts
to oscillate as soon as 4 kV is reached, see Paragraph 5.2. Instead, when the same
electrode configuration is used to spray water, but a pulsating DC potential is instead
adopted, no lateral oscillation of the jet is recorded, even with an applied pulsating
potential of 4kV, see Paragraph 6.2. This may be explained by comparing the viscous
relaxation time to the charge relaxation time ratio, $\frac{\tau_p}{\tau}$, with the pulsating DC
frequency used. Huneiti (2000) observed that using an AC potential with a frequency
larger than the ratio $\frac{\tau_p}{\tau}$, it was possible to reduce the amount of surface charges on a
liquid jet and so to stabilise the electrified jet. Similarly, in our experimental condition,
the viscous relaxation time to charge relaxation time ratio for the water is below 40,
while the minimum pulsating DC potential frequency used is well above this value.
Therefore, the frequency used is fast enough to avoid an equipotential or near
equipotential jet surface developing and so avoiding the possibility of setting up, on the
jet surface, a radial stress large enough to induce the jet to oscillate. The above
mechanism may be also used with the aim of explaining why the jets of PVA aqueous
solutions are stabilised by the pulsating DC potential. PVA aqueous solutions have a
$\frac{\tau_p}{\tau}$ which is well below 1, while the minimum pulsating DC potential frequency used
for these solutions is above 30 Hz.

Pulsating DC frequency permits control of the droplet diameter. Increasing the
frequency involves the reduction of the wavelength and thus of the droplet diameter.
The droplet diameter control by the frequency change is an advantage in
electrohydrodynamic spraying although there is a limit to the applicable frequency and
therefore, to the droplet diameter control. It has been experimentally observed that
increasing the frequency, a reduction in the wave length occurs until a certain value
over which the wavelength becomes difficult to measure. This is because, over a certain
value of the frequency applied collisions between axisymmetrical waves occur, see Figure 6.25. The mechanism of collision may be qualitatively explained by comparing the break up time and the time between two consecutive bumps. The break up time can be evaluated as follows:

\[ r_j = r_{j0} + \delta^* e^{e^t} \cos \left( \frac{2\pi v_w}{\lambda} \cdot z \right) \]  \tag{6.5}

where \( r_{j0} \) is the radius of the jet for \( t=0 \) and \( \delta^* \) is the initial disturbance. By solving Equation (6.5) for \( r_j=0 \), it is possible to calculate the break up time, \( t_{\text{break-up}} \), as below:

\[ t_{\text{break-up}} = \frac{1}{q} \ln \left( \frac{r_{j0}}{\delta^*} \right) \]  \tag{6.6}

(Lefebvre, 1989). Evaluation of the break up time is quite difficult because it needs an estimate of the value of the initial disturbance which is dependent on a number of parameters such as the physical properties of the liquid sprayed, the nozzle used and the characteristics of the electrode set up. Also, another problem is the evaluation of \( q \) for an electrified liquid by a pulsating DC potential. However, an approximate calculation may be performed, considering the liquid is not electrified and giving an arbitrary initial disturbance equal to 5% of the jet radius. This value is taken from Hartman (1998). The time between two bumps is equal to the inverse of the frequency. Therefore, on one hand, when the frequency increases, the wavelength reduces and thus the time between two consecutive wave bumps also decreases hypothetically to zero for infinite frequency, see Figure 6.29 curve \( \tau \). On the other hand, for a frequency increase, and thus a wave number increase (when the frequency increases the wave length decreases), the wave growth rate factor increases (see Figure 2.4), hence the break up time also decreases, see Equation 6.6. The wave growth rate factor reaches a maximum value for a certain value of wave number (for a certain value of the frequency). When the wave number increases beyond this certain value of the wave number the wave growth rate factor starts to decrease and so the break up time begins to increase. In other words there is a minimum value of the break up time for a certain value of wave number (for a certain value of the frequency), see Figure 6.29 curve \( t_{\text{break-up}} \). Therefore,
there may exist a range of frequency in which the break up time is either equal or larger than the time between two consecutive bumps. In this range of frequency the wave bump reaches the jet end before that the previous bump has been pulled out as droplets collide with it.

Figure 6.29 Time between two consecutive bumps and $\tau_{\text{break-up}}$ versus applied frequency for tap water and electrode configuration 2A. $\bullet$: $\tau_{\text{break-up}}$; - - - - - - - Time between two consecutive bumps.

It has also been observed that for a frequency increase the jet becomes shorter. The jet shortening is due to (a) the increase of the growth factor, (b) the increase of the frequency applied and thus (c) the increase of the wave number as well. In addition, surface charges increase the growth rate amplitude for lower values of the wavelength. An increase in the growth factor leads the waves to reach an amplitude large enough to provoke the jet break up in a shorter time and thus to travel a shorter distance, hence the liquid jet becomes shorter.

Finally, it cannot be stated a priori what is the effect of the viscosity on the droplet diameter. Researchers including Weber (1931), Smith (1986) and Lavernia and Wu (1997) reported that an increase of the viscosity leads to longer axisymmetrical wavelength and therefore to the production of larger droplets. Hartman (1998) observed
that an increase in viscosity would lead to a longer wavelength and so to a larger break up time with the consequence of an increase in the jet length. The increase in the jet length results in a smaller jet radius at the jet break up and so to a smaller droplet size. In this work it has been observed that for the same applied frequency the more viscous liquid has the shorter wavelength and thus the smaller droplet diameter when the jet breaks up.
Chapter 7 - Conclusions and future work

7.1 Conclusions

In this work it has been demonstrated that highly viscous and conductive liquids such as PVA aqueous solutions can be effectively electrohydrodynamically sprayed in a controlled fashion of droplets when an opportune electrode configuration and applied pulsating DC potential are used. Highly conductive and viscous liquids can be very difficult to spray when using a DC potential only. Firstly because the high liquid conductivity determines a very short charge relaxation time so that the liquid jet surface becomes equipotential or near equipotential and in this condition the lateral wave growth rate factor is larger than the varicose wave growth rate factor, hence the whipping mode can set in with the production of polydispersed droplets. Secondly because: a) the high liquid viscosity determines a quite short viscous relaxation time with respect to the flow characteristic time so that the charge convection is reduced; b) the high liquid viscosity increases the droplet diameter such that the amount of current conducted is also reduced; c) the high liquid viscosity can impede the migration of the charge carriers, so the charge conduction also reduces. Hence, the combined effect of the above mentioned mechanisms reduces the total amount of the current conducted throughout the jet with a consequence weakening the tangential component of the stress on the liquid surface and the reduction of the total amount of jet surface charge. Furthermore, the tangential surface stress needs to work against the viscous stress, which rises with the viscosity increase. The overall outcome is that high liquid viscosity reduces the control on the jet diameter as well as on the droplet diameter and their size distribution when only DC potential is used.

As stated above, in order to spray a highly conductive and viscous liquid in a controlled fashion of droplets an opportune electrode geometry is needed. In fact, it has been experimentally observed that if the distance between the needle's tip and the earthed plate is too large, then the electric field may be too weak to control the liquid jet break up when the liquid viscosity becomes considerably high. Conversely, if the distance between the needle's tip and the earthed plate is too short, then dielectric break
down can easily occur well before any effect of the electric field on the liquid jet break up has been recorded. It has been clearly shown that a pulsating DC potential can be effective in controlling the liquid jet break up. The combination of an opportune applied potential and frequency leads to monosized droplet production. The effect of the pulsating DC potential can be explained in terms of the system constants (i.e. charge relaxation time, viscous relaxation time). Using an opportune frequency above the \( \frac{\tau_r}{\tau} \) ratio, it is possible to avoid a fully developed equipotential jet surface and therefore to stay away from kink instability which would promote polydispersed spraying. In addition, there is the experimental evidence that depending on the liquid physical properties (i.e. liquid viscosity) it is possible to find a window of applied frequency within which uniform droplet are produced i.e. no satellites and that in this window of frequency the control over the spraying occurs in such a way that for each DC pulse a droplet is formed.

The lower limit of the window of applied frequency where uniform droplets are produced is characterized from a cut-off frequency for satellite production. In particular, it has been observed that for the frequency increase there is the reduction of droplet and satellite diameter until a certain frequency, beyond which the satellite is not produced. The cut-off frequency for satellite formation has been explained in terms of a force balance between surface tension forces and the electric field superimposed on space-charge field forces. Using an opportune frequency it has been observed that it is possible to generate a certain wavelength that produces a favourable recoiling of the liquid filament, present between the forming droplets and the liquid jet, into the jet itself just after the liquid jet break up has taken place. In addition, there is experimental evidence that the cut-off frequency for satellite production is dependent on the liquid viscosity in a way that as the liquid viscosity increases, the cut-off frequency reduces. This has been justified observing that high liquid viscosity promotes surface charge reduction hence allowing a favourable recoiling of the liquid filament back to the liquid jet for larger values of the wavelength and therefore for lower values of the frequency applied.

The higher limit for the window of applied frequency where uniform droplets are produced is characterized by bunching-up between waves on the liquid jet, just
toward the jet end. Above this limit, there is a linear increase in the frequency at which collisions are likely to occur between waves crests, as the pulse frequency increases such that the number of droplets produced per unit time remains the same. Furthermore, it has been shown that the frequency at which the collisions occur is dependent upon the liquid physical properties (i.e. viscosity). The photographic evidence of the above mentioned collisions has led to analysis of the phenomenon in terms of break up time versus wave period. The mechanism proposed explains why the collision between waves may occur and why the frequency to which the wave collision occurs is dependent on the liquid viscosity. Nevertheless, the proposed mechanism is not exhaustive. In fact, it does not explain why the collisions between waves increases linearly as the pulse frequency increases such that the number of droplets produced per unit time remains identical. However, when frequencies are used such that wave collision occurs, a droplet with double volume is produced which is an undesirable phenomenon when uniform droplets are needed.

This work suggest that the electrohydrodynamic spraying is a suitable technique in order to spray highly viscous and conductive liquid if an opportune electrode configuration and applied pulsating DC potential are used, even so the technique has shown a limit to be used when the liquid viscosity has exceeded 280 mPas. For these values of the liquid viscosity, it has not been possible to efficiently control the spraying for any frequency used (30 Hz-300 Hz) and electrode geometry configuration. No satellite cut-off frequency has ever been observed for all the frequency used.

The electrohydrodynamic spraying by smooth jet mode using a pulsating DC potential has been shown to be the most suitable mode if compared to dripping mode and convergent jet mode with the aim of obtaining monosized droplet of the liquid used with the large flow rate adopted in this work.
7.2 Future work

Broadly speaking, the physical properties of a liquid have a pronounced influence on electrohydrodynamic spraying. Therefore the ideal liquid would be a liquid where it is possible to vary the physical property of interest while keeping the others constant. Hence, in this work PVA aqueous solutions have been used in order to investigate the effect of liquid viscosity, because, as already stated, PVA permitted to create liquids where the viscosity could be varied over several orders of magnitude without influencing the other physical properties. Indeed, this has been partially true for the PVA aqueous solution. In fact, although for PVA aqueous solution the liquid density, electrical conductivity and surface tension are changing within a close range the different amount of PVA used in each solution has doubtlessly created liquid with different value of the dielectric constant, which has beyond a shadow of a doubt a great importance on the mechanism involved in electrohydrodynamic spraying. Hence, the possibility of developing a working liquid where it is possible to vary a physical property while keeping constant all the other physical properties, which are important in electrohydrodynamic spraying, leaves still an open chapter and a non trivial problem to overcome. In addition there is evidence in the literature that an extremely high electric field, (10^6 V/m) as the one used in this work, can change the physical properties of the liquid such as viscosity, electric conductivity and surface tension. Therefore, it would be suitable in the future to set up experiment in which it is possible to measure the variation induced on the physical properties of the liquid by the electric field in order to truly understand their effect on electrohydrodynamic spraying.

Although this work has shown the efficacy of the technique investigated for spraying highly viscous and conductive liquids, it has been shown that an undesirable phenomenon may take place, i.e. wave bunching up on the liquid jet. For this phenomenon a mechanism based on the break up time to wave period time ratio has been proposed, however the phenomenon is not fully understood. The collision between waves on the liquid jet has a tremendous implication from the technological as well as from the scientific point of view. In fact, to understand the reason why the wave collision occurs would enlarge the knowledge on the mechanism of electrified jet break up, which is still far from being fully understood, and it would help to understand which kind of improvements may be made in order to avoid it.
The electrohydrodynamic spraying has been performed by a pulsating DC potential with pulse width of 50%. No experiments have been done changing the pulse width. Indeed, reducing the pulse width could change the amount of current conducted through the jet and so change the amount of surface charge density and space-charge effect at the break up. This could throw new light on the mechanism of cut-off frequency for satellite production as well as on the frequency at which wave bunching occurs.

It has been demonstrated that the control of liquid spraying using a DC potential is strongly influenced by liquid viscosity. The liquid viscosity can heavily influence the mobility of the charge carriers, the fluid dynamics inside the liquid jet and the jet surface instability. How all these phenomena interact in determining the total amount of current conducted through a liquid jet is not clear still. A full understanding of this phenomenon would help in working out a general scaling law for electrohydrodynamic spraying.
Appendix A - Statistical analysis

When the spraying of a liquid is performed and a number of droplets are made, there is a need to characterize the droplet diameter and their size distribution. The characterization requires definition of how many droplets need to be measured in order to have a representative measurement of the droplet size distribution. To understand the kind of distribution which characterise droplet size, four different samples of 250, 500, 1000, 2000 droplets have been recorded, see Figures 3.7- 3.10, Chapter 3.3. Indeed, the sample made of 250 droplets does not allow to neatly distinguish the kind of distribution which characterise the droplet size, as well as the sample made of either 1000 or 2000 droplets does not add any further information to the one which can be obtained from the sample made of 500 droplets. In other words, a sample made of 500 droplets is enough to say that the distribution which characterise the droplet size in our system is bimodal. Each bimodal distribution may be considered as made of two modal and normal distribution and as characteristic diameter for each distribution may be chosen the mean value of droplets diameter and the mean value of satellites diameter, see Figure A.1.

![Figure A.1 Example of bimodal distribution.](image-url)
The former refers to the larger mode and the latter refers to the smaller mode; in a normal distribution the mean diameter, the median diameter and the mode diameter have the same value.

Table A.1 Mean diameter and standard deviation calculated for droplet and satellite with respect to fours samples made of a different numbers.

<table>
<thead>
<tr>
<th>Sample of 250 droplets</th>
<th>Sample of 500 droplets</th>
<th>Sample of 1000 droplets</th>
<th>Sample of 2000 droplets</th>
</tr>
</thead>
<tbody>
<tr>
<td>MDD = 2.033 mm</td>
<td>MDD = 2.049 mm</td>
<td>MDD = 2.059 mm</td>
<td>MDD = 2.057 mm</td>
</tr>
<tr>
<td>s = 0.324 mm</td>
<td>s = 0.328 mm</td>
<td>s = 0.327 mm</td>
<td>s = 0.321 mm</td>
</tr>
<tr>
<td>MDS = 0.413 mm</td>
<td>MDS = 0.425 mm</td>
<td>MDS = 0.442 mm</td>
<td>MDS = 0.44 mm</td>
</tr>
<tr>
<td>s = 0.185 mm</td>
<td>s = 0.181 mm</td>
<td>s = 0.161 mm</td>
<td>s = 0.147 mm</td>
</tr>
</tbody>
</table>

The mean diameter may be calculated with a certain accuracy that is characteristics of the number of droplets present within the sample. In Table A.1 is reported the mean diameter calculated for the droplets (MDD) and satellites (MDS) and their standard deviation $s$ for four sample each of which is made of a different number droplets, see Equation A.1 and A.2. As it can be noticed, the calculated mean values for the droplets are the same within an error of circa 1%, while the calculated mean values for the satellite are the same within an error just below 10%. On this basis, it possible to consider that the sample made of 500 droplets is significant enough to determine the kind of distribution as well as the characteristic diameter with an acceptable error. Once more, using samples of either 1000 or 2000 droplets would not considerably improve the accuracy of the measurement.

\[
\overline{X} = \sum_{i=1}^{N} D_{drop, i}
\]  \hspace{1cm} (A.1)

Where $\overline{X}$ is the either the mean droplet diameter or the mean satellite diameter, $N$ is the number of droplet in the sample and $D_{drop, i}$ is the $i^{th}$ droplet of the sample.
\[ s = \sqrt{\frac{\sum_{i=1}^{N} (D_{drop_i} - \overline{X})^2}{N - 1}} \]  

(A.2)

Where \( s \) is the standard deviation. (Zar, 1984)
Appendix B - Optimas Macro

Hereafter is reported the macro written in ALI language for the size analysis.

```ali
/* Vector whit value, number of droplet */
Real DiaNeedle;
DiaNeedle=1.47; /* diameter needle in millimiter*/
Real Length;
Length=3.148; /* value on the picture*/
integer i,Counter,j,a,b,c,d,e,f,g,h,l, m, n, o, p, q,r, s, t, u, v, z, aa, ab, ac, ad, af, ag,
ah, al, am, an, ao, ap, aq, ar, as, at, au, av, Sat, Ma, Sat2;

i=1;/* start number frame*/
Counter=0;
Boolean Control;
Control= False;
integer number;
number=499;

Real Diameter[],Main[],Satellite[],Satellite2[],
Distribution[];
/*Number of frame, the last that you want see */
a=0;h=0;c=0;d=0;c=0;f=0;g=0;h=0; l=0; m=0; n=0; o=0; p=0; q=0;
r=0; s=0; t=0; u=0; v=0; z=0; aa=0; ab=0; ac=0; ad=0; ae=0; af=0; ag=0; ah=0;
al=0; am=0; an=0; ao=0; ap=0; aq=0; ar=0; as=0; at=0; au=0;
av=0; Sat=0; Sat2=0; Ma=0;

While(i<=number) {
    Char File;
    File="d:/nacho4/ltap/4v235h50/l_.:ToText(i):.tif";
    /* pat of the file that you want analize */
    OpenImage(File,False);
    FillFilter();
    CreateArea(.,TRUE);
    SetExport(TotalTally,1,TRUE);
    Counter=TotalTally+Counter;
    /*RunMacro("C:/optimas6/macros/ellipse.mac");*/
    SetExport(ArAreaEquivDiameter,1,TRUE);
    MultipleExtract();
    ClearScreen();

    } 
    if (Control==False){
        ViewBox(Diameter);
        Control=True;
    }

    j=0;
    While(j<Counter) {
        if (Diameter[j]<=0.2 && Diameter[j]>0.1) {
            a++;
        }
    }
```
{ b++;
}
{ c++;
}
{ d++;
}
{ e++;
}
{ f++;
}
{ g++;
}
{ h++;
}
{ i++;
}
{ m++;
}
{ n++;
}
{ o++;
}
{ p++;
}
{ q++;

else if(Diameter[j]<=0.3 && Diameter[j]>0.2)
{
}
else if(Diameter[j]<=0.4 && Diameter[j]>0.3)
{
}
else if(Diameter[j]<=0.5 && Diameter[j]>0.4)
{
}
else if(Diameter[j]<=0.6 && Diameter[j]>0.5)
{
}
else if(Diameter[j]<=0.7 && Diameter[j]>0.6)
{
}
else if(Diameter[j]<=0.8 && Diameter[j]>0.7)
{
}
else if(Diameter[j]<=0.9 && Diameter[j]>0.8)
{
}
else if(Diameter[j]<=1.0 && Diameter[j]>0.9)
{
}
else if(Diameter[j]<=1.1 && Diameter[j]>1.0)
{
}
else if(Diameter[j]<=1.2 && Diameter[j]>1.1)
{
}
else if(Diameter[j]<=1.3 && Diameter[j]>1.2)
{
}
else if(Diameter[j]<=1.4 && Diameter[j]>1.3)
{
}
else if(Diameter[j]<=1.5 && Diameter[j]>1.4)
{


```c
} else if(Diameter[j]<=1.6 && Diameter[j]>1.5)
{ t++;
}
} else if(Diameter[j]<=1.7 && Diameter[j]>1.6)
{ s++;
}
} else if(Diameter[j]<=1.8 && Diameter[j]>1.7)
{ t++;
}
} else if(Diameter[j]<=1.9 && Diameter[j]>1.8)
{ u++;
}
} else if(Diameter[j]<=2.0 && Diameter[j]>1.9)
{ v++;
}
} else if(Diameter[j]<=2.1 && Diameter[j]>2.0)
{ z++;
}
} else if(Diameter[j]<=2.2 && Diameter[j]>2.1)
{ aa++;
}
} else if(Diameter[j]<=2.3 && Diameter[j]>2.2)
{ ab++;
}
} else if(Diameter[j]<=2.4 && Diameter[j]>2.3)
{ ac++;
}
} else if(Diameter[j]<=2.5 && Diameter[j]>2.4)
{ ad++;
}
} else if(Diameter[j]<=2.6 && Diameter[j]>2.5)
{ ae++;
}
} else if(Diameter[j]<=2.7 && Diameter[j]>2.6)
{ af++;
}
```
else if(Diameter[j]<=2.8 && Diameter[j]>2.7)
{
    ag++;
}
else if(Diameter[j]<=2.9 && Diameter[j]>2.8)
{
    ah++;
}
else if(Diameter[j]<=3.0 && Diameter[j]>2.9)
{
    al++;
}
else if(Diameter[j]<=3.1 && Diameter[j]>3.0)
{
    am++;
}
else if(Diameter[j]<=3.2 && Diameter[j]>3.1)
{
    an++;
}
else if(Diameter[j]<=3.3 && Diameter[j]>3.2)
{
    ao++;
}
else if(Diameter[j]<=3.4 && Diameter[j]>3.3)
{
    ap++;
}
else if(Diameter[j]<=3.5 && Diameter[j]>3.4)
{
    aq++;
}
else if(Diameter[j]<=3.6 && Diameter[j]>3.5)
{
    ar++;
}
else if(Diameter[j]<=3.7 && Diameter[j]>3.6)
{
    as++;
}
else if(Diameter[j]<=3.8 && Diameter[j]>3.7)
{
    at++;
}
else if(Diameter[j]<=3.9 && Diameter[j]>3.8)
{
    au++;
}
j++;  

ViewBox(Distribution);

MacroMessage("Distribution ",Distribution[j]);

*/

j=0;
While(j<Counter)
{
  if (Diameter[j]<=0.31 && Diameter[j]>=0.3)
  {
    Satellite2:=Diameter[j];
    Sat2++;
  }
  else if(Diameter[j]<=1.5 && Diameter[j]>0.31 )
  {
    Satellite:=Diameter[j];
    Sat++;
  }
  else if(Diameter[j]<=3 && Diameter[j]>1.5)
  {
    Main:=Diameter[j];
    Ma++;
  }
  j++;
}

RunMacro("C:/optimas6/dialogs/histogram.mac");
/* ViewBox(Sum(Main)/Ma,Sum(Satellite)/Sat);
MacroMessage("Average Main",(Sum(Main))/Ma);
MacroMessage("Average Satellite ",(Sum(Satellite))/Sat);*/

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Bibliography


**Antonio Speranza**

<table>
<thead>
<tr>
<th>PLACE and DATE of BIRTH</th>
<th>Avellino, 11 July 1970</th>
</tr>
</thead>
<tbody>
<tr>
<td>NATIONALITY</td>
<td>Italian</td>
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<tr>
<td>EDUCATION</td>
<td><strong>Surveyor Diploma</strong> at the Istituto Tecnico Statale Commerciale e per Geometri Gaetano Bruno** at Ariano Irpino (AV), from 1984 to 1989, mark 58/60.</td>
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<td><strong>Degree in Chemical Engineering</strong> at the Università degli Studi di Salerno, from 1989 to 1997, mark 108/110.</td>
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<tr>
<td></td>
<td><strong>Thesis:</strong> SCR process for the NOx reduction of emissions from a thermal power station: study of WOx/TiO2 catalyst. Supervisor: Prof. Paolo Ciambelli.</td>
</tr>
<tr>
<td>WORK EXPERIENCE</td>
<td><strong>Present</strong> Researcher at CNR - Istituto di Biostrutture e Bioimmagini, Napoli</td>
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<tr>
<td></td>
<td>1985-1988 Construction design of housing in architect’s studio.</td>
</tr>
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<td></td>
<td>1992-1993 Military service</td>
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<td></td>
<td><strong>July 1997</strong> Examiner for the Esame di Stato (Italian baccalaureate, equivalent to A-Levels).</td>
</tr>
<tr>
<td></td>
<td>1998-2001 PhD student at the Department of Chemical and Process Engineering, University of Surrey, UK. The research focused on the electro-spraying which is an innovative technology where a high voltage is used as a medium for enhancing the spraying of viscous liquids.</td>
</tr>
</tbody>
</table>
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