Stellingen

behorend bij het proefschrift
"Electrohydrodynamic Atomization in the Cone-Jet Mode
From Physical Modeling to Powder Production"
van Rob Hartman

1. Electrohydrodynamisch Verstuiven in de Cone-Jet mode kan gemodelleerd worden met behulp van fysische modellen zonder dat er parameters geft moeten worden aan experimentele resultaten.

2. De betrouwbaarheid van vergelijkingen die verkregen zijn door het fitten aan experimentele resultaten hangt volledig af van het fysische begrip van het achterliggende verschijnsel. Er is altijd wel een wiskundige vergelijking te bedenken die toevallig voldoet aan de experimentele waarden.

3. De validatie en verbetering van een wetenschappelijke theorie gaat het snelst als er tijdens het onderzoek zowel computersimulaties als experimenten gedaan worden. Echter, onderzoekers die beide goed kunnen en beide ook leuk vinden zijn zeldzaam.

4. Opschaling en optimalisatie van een produktieproces bieden vooral een uitdaging indien het proces ook commercieel mogelijk zou kunnen zijn.

5. Het continue reorganiseren, lees afslanken, van grote bedrijven ten behoeve van grotere winsten op de korte termijn, leidt op langere termijn tot verdere afslanking, omdat er niet voldoende nieuwe dingen worden ontwikkeld.

6. De technische en medische vooruitgang hebben er voor gezorgd dat ook genetisch minder gezonde mensen veel meer kans hebben om zichzelf voort te planten. Daardoor zullen we er in de verre toekomst niet gezonder op worden. De op dit moment mogelijke maatregelen om dit proces tegen te gaan zijn echter nog minder ethisch verantwoord dan gewoon niets doen.

7. Als het autoverkeer zich zou gedragen als een stromende vloeistof dan zou het fileprobleem in één klap zijn opgelost.

8. De steeds kortere levensloop van produkten in de huishoudelectronica heeft weinig meer te maken met de technische ontwikkeling maar alles met mode. Dit in tegenstelling tot de computerindustrie waar elk jaar weer snellere en meer geheugenruimte bevattende chips worden geproduceerd.


10. Er zal altijd een dilemma zijn tussen voldoening in het werk en voldoening in het privé leven, omdat er te weinig uren in een dag zitten.
ELECTROHYDRODYNAMIC ATOMIZATION
IN THE CONE-JET MODE

FROM PHYSICAL MODELING
TO POWDER PRODUCTION

PROEFSCHRIJT

ter verkrijging van de graad van doctor
aan de Technische Universiteit Delft,
op gezag van de Rector Magnificus prof. ir. K.F. Wakker,
in het openbaar te verdedigen ten overstaan van een commissie,
door het College voor Promoties aangewezen,
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CURRICULUM VITAE
ELECTROHYDRODYNAMIC ATOMIZATION
IN THE CONE-JET MODE

FROM PHYSICAL MODELING
TO POWDER PRODUCTION

Rob Hartman

The objective of this thesis is to investigate the parameters that influence the droplet size and current produced in Electrohydrodynamic Atomization in the Cone-Jet mode. This thesis shows that Electrohydrodynamic Atomization in the Cone-Jet mode can be modeled by means of several physical models. These models are developed, and the results are compared to experimental results. The acquired insight into the phenomenon of Electrohydrodynamic Atomization in the Cone-Jet mode is used to improve the droplet size and current scaling laws. The scaling laws predict the droplet size and the current as functions of the liquid properties, and the liquid flow rate. The simulations showed that the influence of the dielectric permittivity of the liquid is much smaller than previously assumed. This lead to the conclusion that the relative dielectric constant of the liquid should be removed from the current scaling law. The influence of the radial profile of the axial liquid velocity on the current was estimated. When this estimation is used, then the scaling law for high conductivity liquids, meaning a flat velocity profile in the jet, and the scaling law for low conductivity and low viscosity liquids, meaning a much steeper velocity profile, can be merged into one scaling law. The comparison between experimental results and calculations also revealed that the produced droplet size depends on the jet break-up mechanism. At relatively low electric stresses in the liquid jet compared to the surface tension stress of the jet, the jet breaks up due to axisymmetric perturbations of the jet diameter, the varicose jet break-up regime. Above a certain threshold value of the electric stress, the jet starts to show a whipping movement, the whipping jet break-up regime. This whipping movement has an influence on the droplets size. Two relations are derived to calculate the droplet size for each of these two jet break-up regimes. Electrohydrodynamic Atomization in the Cone-Jet mode has been used to produce a monodisperse polymer powder.

Chapters 1 and 2 give an introduction to electrohydrodynamic atomization in general, and to the Cone-Jet mode in particular. Chapter 2 describes the various spraying modes of electrohydrodynamic atomization. The Rim-Emission and the Multiple-Jet mode are the most promising spraying modes for monodisperse aerosol production at higher production rates. However, they are more difficult to control than the Cone-Jet mode. Since, the Cone-jet mode is the basis of the Rim-Emission and the Multiple-Jet mode, the Cone-Jet mode was chosen as
subject of research. Chapter 2 also describes the characteristics of the Cone-Jet mode.

In chapter 3, the Cone-Shape model is presented. This physical numerical model describes the phenomena that occur inside the liquid cone. The model is able to calculate the shape of the liquid cone and jet, the electric fields in and outside the cone, and the surface charge density on the cone and jet surface. For liquids with a flat radial profile of the axial velocity in the jet, the calculated cone-jet shape agrees very well with the experimental shape. The calculated current through the liquid cone agrees within 10 percent with the measured values. In cases, where the radial profile of the axial liquid velocity in the jet was not flat, the model deviates more from the measured current, but still predicts the right trends.

Chapter 4 describes the effects of electrical discharges in the air on the droplet production process in the Cone-Jet mode. Electrical discharges occur when the electric field strength near the liquid cone exceeds the onset field strength. When the total current of these discharges is of the same order of magnitude or larger than the current through the liquid cone without discharges present, then the droplet production is influenced. When the discharge current consists of pulses of charge, then the droplet production is completely disturbed. When the discharge is in the glow mode, then it is sometimes possible to produce narrow size distributions. However, the produced droplet size and charge will be different from the Cone-jet mode without discharges. The highest discharge probability can be found near the cone apex, where the jet emerges from the cone. In order to reduce the discharge probability, a liquid should be sprayed just above the minimum flow rate.

Chapter 5 describes the evolution of a spray just after production. The Cone-Jet mode produces highly charged droplets. Due to the electric forces between these droplets, and due to differences in inertia, a size segregation effect occurs in the spray just after production. A model is presented, which is able to simulate the size segregation, the droplet velocity, and the droplet concentration in the spray. The model underestimates the radial dispersion of the secondary droplets, but is able to predict the radial dispersion of the main droplets. Comparison between measurements and simulations shows that air velocities of several meters per second can occur in the first few centimeters of the spray. When in the spray an region of low droplet concentration occurs, then the aerosol has a bimodal size distribution, and the amplitude of the whipping motion of the jet is still small.

Chapter 6 describes the process of the jet breaking up into droplets. With increasing flow rate the following five jet break-up regimes have been distinguished. At low flow rates, the jet breaks up into main droplets due to axisymmetric varicose instabilities. Secondary droplets and satellites are not produced, because the filament between the main droplets has been observed to flow back into one of the main droplets. The second regime occurs when the filament breaks a second time before it can flow back into the main droplet. Secondary droplets are produced. The third regime occurs when the filament between a main droplet and a secondary droplet breaks two times. In this case, main droplets, secondary droplets, and satellites are produced. The fourth regime occurs when lateral or kink instabilities become more important. Secondary droplets, and the filaments between the main droplets, start to leave the jet axis. This regime will be called the whipping filament regime. The fifth regime occurs when the whole jet started to
Summary

make a spiraling whipping motion. In the last two regimes, the width of the main droplet size
distribution starts to increase. The stress ratio of the electric normal stress over the surface
tension stress has been found to be the dominant parameter. This ratio mainly depends on the
current through the liquid cone. A mathematical physical model is presented, which is able to
predict for varicose instabilities the main droplet size, and the droplet velocity at jet break-up.
A relation is presented which enables to calculate the droplet size in the whipping jet break-up
regime from the current and the liquid flow rate. In the varicose jet break-up regimes, the main
droplet size has been found to scale with the flow rate to the power 0.48.

Chapter 7 presents a new scaling of the current through the liquid cone. A new derivation
is presented, and the scaling is extended to low viscosity and low conductivity liquids. These
liquids have a higher axial velocity at the jet surface than in the jet center. When accounted for
the electrode configuration, and the ion species used to enhance the conductivity of a liquid, then
this scaling deviates less than 10 percent from the measured current for Newtonian liquids. HCl
has been added to n-butanol and ethylene glycol. HCl as additive yields up to 50 percent lower
currents, compared to the same liquids, with the same conductivity, but with LiCl as additive.
A relation is presented which enables the calculation of the droplet size in the varicose jet break-
up regime from the current and the liquid flow rate. The accuracy of this relation is better than
15 percent.

In Chapter 8 a first design of a reactor for the production of monodisperse polymer
powder is presented. The influence of neighboring nozzles can be reduced by bringing a plate
with holes close to the nozzles. It is shown that it is possible, to produce a polymer powder with
a narrow size distribution. Discharging of the droplets does not have an influence on the droplet
production, as long as only a few ions used for the discharging of the droplets can reach the liquid
cone. High discharging efficiencies seem to be possible. The initial polymer concentration in the
solvent has a strong influence on the shape of the produced particles. At high concentrations,
strings of polymer are produced instead of droplets. At low polymer concentrations, the particles
are elongated or look like bursted balloons. At a polymer concentration of about 20 percent,
spherical particles are produced.

In Chapter 9 a summary of all the conclusions of this thesis is presented.
ELECTROHYDRODYNAMISCH VERSTUIVEN VAN VLOEISTOFFEN IN DE CONE-JET MODE

VAN FYSISCHE MODELLERING TOT DE PRODUKTIE VAN POEDERS

Rob Hartman

Het doel van dit proefschrift is te laten zien wat de invloed is van de verschillende parameters op de druppelgrootte en de stroom, die geproduceerd wordt tijdens het electrohydrodynamisch verstuiven van vloeistoffen in de Cone-Jet mode. Dit proefschrift bewijst dat electrohydrodynamisch verstuiven van vloeistoffen in de Cone-Jet mode kan worden gemodelleerd met behulp van een aantal fysische modellen. Deze modellen zijn ontwikkeld en de resultaten zijn vergeleken met experimentele resultaten. Het uit deze vergelijking verkregen inzicht in het verschijnsel heeft geleid tot nieuwe schalingsregels voor de druppelgrootte en de stroom. Het is gebleken dat de invloed van de dielectrische permittiviteit veel kleiner is, dan tot nu toe is aangenomen. De schalingsregels voor de stroom moeten daarom op een andere manier worden afgeleid. Daarnaast zijn de schalingsregels voor de stroom uitgebreid met een schatting van de invloed van het radiale profiel van de axiale vloeistofsnellheid. Uit de vergelijking van de resultaten van de simulaties en de experimenten is tevens gebleken dat de geproduceerde druppelgrootte afhangt van het mechanisme waarmee de straal op breekt in druppels. Bij een relatief lage elektrische spanning in het oppervlak van de straal, vergelijken met de oppervlakte spanning van de straal, breekt de straal op in druppels door axisymmetrische verstoringen van de straal diameter. Als de elektrische oppervlakte spanning boven een bepaalde grenswaarde komt dan begint de straal een slingerende beweging te maken. Deze slingerende beweging beïnvloedt de geproduceerde druppelgrootte. De invloed van deze mechanismes zijn verdisconteert in de afleiding van de schalingsregels voor de druppelgrootte door de oude schalingsregel te vervangen door twee andere vergelijkingen voor de druppelgrootte. Tenslotte, Electrohydrodynamisch verstuiven van vloeistoffen in de Cone-Jet mode is gebruikt voor de produktie van een poeder met een monodisperse deeltjesgrootteverdeling.

de Cone-Jet mode. De Cone-Jet mode is de basis voor de Multiple-Jet en de Rim-Emission mode, daarom, is de Cone-Jet mode gekozen als onderwerp van onderzoek. In hoofdstuk 2 wordt de Cone-Jet mode verder beschreven.

In hoofdstuk 3 wordt het Cone-Shape model beschreven. Dit numerieke fysische model beschrijft de verschijnselen die in de vloeistof kegel plaats vinden. Het model is in staat om de vorm van de kegel en de straal, de elektrische velden binnen en buiten de vloeistof en de oppervlakte ladingdichtheid op de kegel en de straal te voorspellen. Afhankelijk van de straal diameter en de viscositeit van de vloeistof kan de axiale vloeistofsnellheid aan het oppervlak groter zijn dan de snelheid op de axiale as van de straal. Voor vloeistoffen met een vlak radiaal profiel van de axiale snelheid in de straal komt het berekenende vorm van de kegel en de straal zeer goed overeen met de gemeten vorm. De berekende stroom verschilt minder dan 10 procent met de gemeten stroom. In die gevallen waar het radiaal profiel van de axiale vloeistofsnellheid niet vlak is, is het verschil tussen de gemeten en de berekende stroom groter. Echter, het model voorspelt nog steeds de juiste trend.

Elektrische ontladingen in de lucht rond de vloeistof kegel hebben effect op het druppelproductieproces in de Cone-Jet mode. Dit effect wordt beschreven in hoofdstuk 4. Elektrische ontladingen ontstaan als het elektrisch veld sterker wordt dan een bepaalde grenswaarde. Als de totale stroom van deze ontladingen van dezelfde orde van grootte of groter is dan de stroom door de kegel zonder de aanwezigheid van ontladingen, dan wordt het druppelproductieproces beïnvloed. In het geval dat de stroom tengevolge van de elektrische ontladingen een pulserend karakter heeft, dan wordt het druppelproductieproces volledig verstoord. Als de elektrische ontladingen vooral corona ontladingen zijn dan is de stroom tengevolge van deze ontladingen veel gelijkmatiger en kan in sommige gevallen de kegelvorm gehandhaafd blijven. De geproduceerde druppelgrootte en de druppellading verschilt echter van de lading en grootte die geproduceerd zou zijn als er geen elektrische ontladingen aanwezig zouden zijn. De plaats met de grootste waarschijnlijkheid voor elektrische ontladingen bevindt zich op de plaats waar de stroom uit de kegel te voorschijn komt. De kans op elektrische ontladingen kan worden verminderd door een vloeistof net boven zijn minimale debiet te verstuiven.

Hoofdstuk 5 beschrijft het gedrag van de druppels vlak na de vorming. De Cone-Jet mode produceert druppels met een zeer hoge lading. Door deze lading oefenen de druppels onderling een elektrische kracht op elkaar uit. Tengevolge van deze elektrische krachten, en tengevolge van verschillen in traagheid vindt er in de nevel een scheidingseffect plaats. Grote druppels blijven in het centrum van de nevel, terwijl de kleinere vooral naar de rand worden gedreven. In dit hoofdstuk wordt een model beschreven dat in staat is om dit scheidingsproces te simuleren. Dit model voorspelt de druppelsnelheid, de druppelconcentratie en de radiale dispersie van de primaire druppels. De radiale dispersie van de secundaire druppels en satellieten wordt enigszins onderschat. Vergelijking van de resultaten van experimenten en simulaties leverde op dat de luchtsnelheid in de nevel op een afstand van 1 cm van het uiteinde van de straal kan oplopen tot 8 meter per seconde. In die gevallen dat er een gebied met een lage druppelconcentratie wordt
waargenomen tussen het gebied met de primaire druppels en het gebied met de secundaire druppels, dan heeft de nevel een bimodale druppelgrootteverdeling en de slingerende beweging van de straal is nog gering.

Hoofdstuk 6 beschrijft het opbreken van de straal in druppels. Met toenemend vloeistof debiet zijn er vijf opbrekeregimes gevonden. Bij een debiet vlak boven het minimale debiet van de vloeistof breekt de vloeistof op in primaire druppels met een monodisperse druppelgrootteverdeling. Secundaire druppels en satellieten worden niet gevormd. Er is waargenomen dat de vloeistofdraad tussen twee primaire druppels vlakbij de eerst geproduceerde druppel breekt, waarna de vloeistofdraad terugstroomt in de andere primaire druppel. Het tweede regime wordt bij een iets hoger debiet waargenomen. De vloeistofdraad breekt een tweede keer, ditmaal vlakbij de tweede primaire druppel. De vloeistofdraad vormt nu een secundaire druppel. Het derde regime verschijnt als na de eerste breuk van de vloeistofdraad een secundaire druppel wordt gevormd. Deze secundaire druppel is met een nog smallere vloeistofdraad verbonden met een primaire druppel. Als nu deze dunne vloeistofdraad twee keer breekt dan wordt er een secundaire druppel en een satelliet gevormd. Het vierde regime ontstaat als de vloeistofdraad tussen de primaire druppels ten gevolge van de oppervlakte lading dichtheid de axiale as verlaat en zijaarts worden weggeslingerd. Het vijfde regime verschijnt als ook de primaire druppels de as verlaten en de hele straal begint te slingeren. Bij de laatste twee regimes neemt niet alleen het aantal pieken in de druppelgrootteverdeling toe met toenemend debiet, maar wordt ook de druppelgrootteverdeling van de primaire druppels breder. De verhouding tussen de elektrische spanning in het oppervlak en de vloeistof oppervlakte spanning is de belangrijkste parameter voor deze opbrekeregimes. Deze verhouding hangt voornamelijk af van de stroom door de vloeistof kegel. Een wiskundig fysisch model wordt gepresenteerd dat in staat is om voor de eerste drie opbrekeregimes de primaire druppelgrootte, en de druppelsnelheid bij opbreking te voorspellen. In deze regimes schaalt de druppelgrootte met het vloeistofdebiet tot de macht 0.48. Een vergelijking wordt gepresenteerd, die in staat is om aan de hand van de elektrische stroom en het vloeistofdebiet de druppelgrootte in de laatste twee opbrekeregimes te voorspellen.

In hoofdstuk 7 worden nieuwe schalingsregels voor de stroom door de vloeistofkegel gepresenteerd. Deze schalingsregels zijn uitgebreid met een schattingsmethode voor de invloed van het radiaal profiel van de axiale vloeistofsnellheid op de stroom. Hierdoor zijn de schalingsregels geldig voor alle Newtonse vloeistoffen. Indien er rekening wordt gehouden met de invloed van het type ionen dat zorgt voor het ladingstransport in de vloeistof en indien er rekening wordt gehouden met de electrodenconfiguratie dan is het verschil tussen de gemeten stroom en de berekende stroom kleiner dan 10 procent. HCl dat toegevoegd wordt aan n-butanol en ethyleen glycol geeft een tot 50 procent lagere stroom dan dezelfde vloeistoffen met dezelfde geleidbaarheid maar waaran LiCl is toegevoegd om deze geleidbaarheid te bereiken. Er wordt een vergelijking gepresenteerd waarmee de druppelgrootte in de varicose opbrekeregime berekend kan worden aan de hand van de stroom en het vloeistofdebiet. De nauwkeurigheid van deze vergelijking is beter dan 15 procent.

Hoofdstuk 8 beschrijft een eerste ontwerp voor een reactor voor de produktie van een
monodispers polymeerpoeber. De invloed van de naburige sproeipunten kan worden vermindert door een plaat met gaten dichtbij de sproeipunten te brengen. Dit hoofdstuk laat zien dat een monodispers polymeerpoeber kan worden geproduceerd. Ontlading van de druppels heeft geen invloed op de geproduceerde druppels zolang relatief weinig ionen, die voor de ontlading worden gebruikt, de vloeistofkegel en de straal kunnen bereiken. Een hoge ontladingsefficiëntie lijkt mogelijk. De polymeer concentratie in de oplossing die versproeid wordt is van groot belang voor de vorm van de geproduceerde deeltjes. Bij een hoge concentratie worden lange polymeer draden geproduceerd. Bij lage concentraties zijn de deeltjes langwerpig of ze lijken op gebarsten ballonnen. Bij een polymeer concentratie van ongeveer 20 procent worden er ronde deeltjes gevormd.

In hoofdstuk 9 worden de conclusies van dit proefschrift samengevat.
<table>
<thead>
<tr>
<th>Symbol</th>
<th>Definition</th>
<th>SI Unit</th>
</tr>
</thead>
<tbody>
<tr>
<td>C_D</td>
<td>drag coefficient</td>
<td>-</td>
</tr>
<tr>
<td>C_profile</td>
<td>correction factor for the kinetic energy of a velocity profile</td>
<td>-</td>
</tr>
<tr>
<td>d_d</td>
<td>droplet diameter</td>
<td>m</td>
</tr>
<tr>
<td>d_g</td>
<td>gap distance between two electrodes</td>
<td>m</td>
</tr>
<tr>
<td>d_i</td>
<td>jet diameter</td>
<td>m</td>
</tr>
<tr>
<td>d_nozzle</td>
<td>nozzle diameter</td>
<td>m</td>
</tr>
<tr>
<td>d_p</td>
<td>characteristic liquid property diameter</td>
<td>m</td>
</tr>
<tr>
<td>E</td>
<td>electric field strength</td>
<td>V m⁻¹</td>
</tr>
<tr>
<td>E_b</td>
<td>breakdown electric field strength</td>
<td>V m⁻¹</td>
</tr>
<tr>
<td>E_ext</td>
<td>external electric field strength</td>
<td>V m⁻¹</td>
</tr>
<tr>
<td>E_n</td>
<td>electric field strength normal to the liquid surface</td>
<td>V m⁻¹</td>
</tr>
<tr>
<td>E_n.ins</td>
<td>normal electric field strength just inside the liquid cone</td>
<td>V m⁻¹</td>
</tr>
<tr>
<td>E_n.out</td>
<td>normal electric field strength just outside the liquid cone</td>
<td>V m⁻¹</td>
</tr>
<tr>
<td>E_t</td>
<td>electric field strength tangential to the liquid surface</td>
<td>V m⁻¹</td>
</tr>
<tr>
<td>f</td>
<td>force per unit of volume</td>
<td>N m⁻³</td>
</tr>
<tr>
<td>g</td>
<td>gravitational constant</td>
<td>m s⁻²</td>
</tr>
<tr>
<td>K</td>
<td>conductivity</td>
<td>S m⁻¹</td>
</tr>
<tr>
<td>I</td>
<td>current through the liquid cone</td>
<td>A</td>
</tr>
<tr>
<td>Iₘ₈</td>
<td>current through the liquid cone for a flat velocity profile in the jet</td>
<td>A</td>
</tr>
<tr>
<td>I_k</td>
<td>conduction current</td>
<td>A</td>
</tr>
<tr>
<td>I_p</td>
<td>characteristic liquid property current</td>
<td>A</td>
</tr>
<tr>
<td>I_c</td>
<td>charge convection current</td>
<td>A</td>
</tr>
<tr>
<td>L</td>
<td>nozzle length</td>
<td>m</td>
</tr>
<tr>
<td>k</td>
<td>wave number</td>
<td>m⁻¹</td>
</tr>
<tr>
<td>m</td>
<td>droplet mass</td>
<td>kg</td>
</tr>
<tr>
<td>m</td>
<td>in Chapter 6: constant describing the perturbation type</td>
<td>-</td>
</tr>
<tr>
<td>n</td>
<td>in Chapter 5: constant</td>
<td>-</td>
</tr>
<tr>
<td>n</td>
<td>in Chapter 6: vector normal to a surface</td>
<td>-</td>
</tr>
</tbody>
</table>
\( n \) in Chapter 6: distance in number of grid points
\( \text{Oh} \) Ohnesorge number
\( p \) pressure
\( p_{\text{kin}} \) velocity pressure
\( p_{\text{En}} \) pressure due to normal electric surface stress
\( p_{g} \) gravitational pressure
\( p_{\text{liq}} \) liquid pressure at the liquid surface
\( p_{\text{At}} \) pressure due to viscous normal surface stress
\( p_{\text{sur}} \) pressure of the surrounding air
\( p_{s} \) pressure due to surface tension stress
\( p_{\text{surf}} \) pressure due to stresses in the liquid surface
\( p_{0} \) reference pressure
\( Q \) liquid flow rate
\( Q_{\text{min}} \) minimum flow rate Cone-Jet mode
\( Q_{0} \) characteristic liquid property flow rate
\( q \) charge
\( q_{c} \) encapsulated charge
\( q_{R} \) Rayleigh Charge Limit
\( r \) radial coordinate
\( r_{d} \) droplet radius
\( r_{j} \) jet radius at a certain position along the cone
\( r_{j_{b}} \) jet radius at jet break-up
\( r_{\text{jet}} \) jet radius
\( r_{s} \) radius of the liquid cone surface
\( r_{e_{v}} \) characteristic radius related to the jet acceleration
\( r_{e_{s}} \) characteristic radius related to the current conduction
\( r_{w} \) wire radius
\( \text{Re} \) Reynolds number
\( R_{E_{s}} \) ratio of electric surface stress over surface tension stress
\( S \) surface
\( T \) temperature
\( T_{0} \) reference temperature
\( t \) time
\( u \) velocity
\( u_{e_{s}} \) axial liquid velocity at the liquid surface
\( V \) potential
\( V_{0} \) reference potential
\( \text{We} \) Weber number
\( z \) axial coordinate
\( z_{E} \) distance between top and bottom electrode plate
\( m^{3} s^{-1} \) m$^3$s$^{-1}$
\( \text{C} \)
\( \text{m} \)
\( \text{K} \)
\( \text{s} \)
\( \text{m} s^{-1} \)
\( \text{V} \)
### Nomenclature

#### Greek symbols

<table>
<thead>
<tr>
<th>Symbol</th>
<th>Description</th>
<th>Unit</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\alpha$</td>
<td>amplitude of the perturbation</td>
<td>m</td>
</tr>
<tr>
<td>$\gamma$</td>
<td>surface tension</td>
<td>N m$^{-1}$</td>
</tr>
<tr>
<td>$\delta$</td>
<td>relative density</td>
<td>-</td>
</tr>
<tr>
<td>$\varepsilon_0$</td>
<td>dielectric constant of vacuum</td>
<td>C$^2$ N$^{-1}$ m$^{-2}$</td>
</tr>
<tr>
<td>$\varepsilon_r$</td>
<td>relative dielectric constant of the liquid.</td>
<td>-</td>
</tr>
<tr>
<td>$\theta$</td>
<td>angular coordinate</td>
<td>rad</td>
</tr>
<tr>
<td>$\lambda$</td>
<td>wave length</td>
<td>m</td>
</tr>
<tr>
<td>$\mu$</td>
<td>absolute viscosity</td>
<td>Pa s</td>
</tr>
<tr>
<td>$\mu'$</td>
<td>effective viscosity</td>
<td>Pa s</td>
</tr>
<tr>
<td>$\rho$</td>
<td>density</td>
<td>kg m$^{-3}$</td>
</tr>
<tr>
<td>$\rho_{\text{air}}$</td>
<td>density of the air</td>
<td>kg m$^{-3}$</td>
</tr>
<tr>
<td>$\sigma$</td>
<td>surface charge density</td>
<td>C m$^{-2}$</td>
</tr>
<tr>
<td>$\sigma_e$</td>
<td>electric polarization normal stress</td>
<td>Pa</td>
</tr>
<tr>
<td>$\sigma_n$</td>
<td>viscous normal stress</td>
<td>Pa</td>
</tr>
<tr>
<td>$\tau$</td>
<td>charge relaxation time</td>
<td>s</td>
</tr>
<tr>
<td>$\tau_{\theta}$</td>
<td>in Chapter 7: shear stress</td>
<td>Pa</td>
</tr>
<tr>
<td>$\tau_{\theta}$</td>
<td>tangential electric shear stress</td>
<td>Pa</td>
</tr>
<tr>
<td>$\tau_d$</td>
<td>time constant for varicose jet break-up</td>
<td>s</td>
</tr>
<tr>
<td>$\tau_p$</td>
<td>viscous shear stress</td>
<td>Pa</td>
</tr>
<tr>
<td>$\omega$</td>
<td>growth rate of the perturbation</td>
<td>s$^{-1}$</td>
</tr>
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</table>

#### Subscripts

<table>
<thead>
<tr>
<th>Subscript</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>cyl</td>
<td>cylinder around the jet</td>
</tr>
<tr>
<td>g</td>
<td>gas</td>
</tr>
<tr>
<td>i</td>
<td>droplet number</td>
</tr>
<tr>
<td>j</td>
<td>in Chapter 5: droplet number</td>
</tr>
<tr>
<td>j</td>
<td>in Chapter 6: grid point number</td>
</tr>
<tr>
<td>f</td>
<td>fluid</td>
</tr>
<tr>
<td>main</td>
<td>main droplet</td>
</tr>
<tr>
<td>r</td>
<td>in the radial direction</td>
</tr>
<tr>
<td>ref</td>
<td>reference</td>
</tr>
<tr>
<td>sat</td>
<td>secondary-droplets and satellites</td>
</tr>
<tr>
<td>z</td>
<td>in the axial direction</td>
</tr>
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#### Superscripts

<table>
<thead>
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<th>Description</th>
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<tr>
<td>'$'$</td>
<td>dimensionless</td>
</tr>
<tr>
<td>''</td>
<td>perturbation</td>
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CHAPTER 1

INTRODUCTION

This chapter gives an introduction to electrohydrodynamic atomization in the Cone-Jet mode, possible applications of the process, the objective of this thesis, and an introduction to the contents.

1.1 ELECTROHYDRODYNAMIC ATOMIZATION IN THE CONE-JET MODE

Electrohydrodynamic atomization, also called electrospraying, refers to a process where a liquid jet breaks up into droplets under influence of electrical forces. However, a liquid jet will also break up into droplets without any electric force present. Depending on the strength of the electric stresses in the liquid surface relative to the surface tension stress, and the kinetic energy of liquid jet leaving the nozzle, different spraying modes will be obtained.

One of these spraying modes is the Cone-Jet mode, sometimes referred to as the Taylor Cone. In this mode, a liquid is pumped through a nozzle at a low flow rate. An electric field is applied over the nozzle and some counter electrode. This electric field induces a surface charge in the growing droplet at the nozzle. Due to this surface charge, and due to the electric field, an electric stress is created in the liquid surface. If the electric field, and the liquid flow rate are in the appropriate range, then this electric stress will overcome the surface tension stress. In that case, the electric stresses transform the droplet into a conical shape. The tangential electric field accelerates the charge carriers at the liquid surface toward the cone apex. In a liquid, the charge carriers are mainly ions. These ions collide with the surrounding liquid molecules. This results in an acceleration of the surrounding liquid. As a result, a thin liquid jet emerges at the cone apex. This jet can break up into a number of main droplets with a narrow size distribution, and a number of smaller secondary droplets and satellites, Cloupeau and Prunet Foch (1989), Gomez and Tang (1994), Chen et al. (1995). The number of secondary droplets can be of the same order of magnitude as the number of main droplets. However, the total volume of these secondary droplets is much smaller than the volume of main droplets. Due to the excess of surface charge in the liquid cone and jet, the droplets are highly charged.

The droplet size and droplet charge depend mainly on the liquid flow rate and on liquid properties like density, viscosity, conductivity, electrical permittivity, and surface tension.
Depending on the liquid properties, the main droplet size produced ranges from nanometers with production frequencies in the order of $10^5$ Hz to hundreds of micrometers with production frequencies of about $10^4$ Hz.

Also of interest, when studying the Cone-Jet mode is the evolution of the spray shortly after the droplet production. Electrical interaction between highly charged droplets and differences in inertia are the main cause for a size segregation effect. Small droplets are found at the edge of the spray, while large droplets are found in the spray center, among others Gañán-Calvo et al. (1994). This makes separation of the main droplets from the smaller secondary droplets possible, and a really monodisperse spray can be obtained. If the highly charged droplets evaporate, then the Rayleigh limit for droplet charge can be reached. In that case droplet fission can take place. This effect changes the droplet size distribution.

1.1.1 Possible Applications

Electrohydrodynamic Atomization in the Cone-Jet mode produces an aerosol of droplets. These droplets have a charge close to the Rayleigh Charge Limit. In some possible applications, the objective is to produce particles from the droplets by means of evaporation. In these applications the liquid consists of a solute in a solvent. Depending on the application, Electrohydrodynamic Atomization in the Cone-Jet mode offers advantages and disadvantages. For instance, the droplet charge prevents the coalescence of droplets, and it enhances collection of the droplets or dried particles. On the other hand, an evaporating charged droplet can exceed the Rayleigh Charge Limit. In that case, the monodisperse size distribution disappears. Also, if the particles are collected too soon after the production, then the droplets still contain too much solvent, and they will stick to the surface on which they are collected. So, applications in which the droplet charge is a problem also require an aerosol discharge technique.

Big advantages of Electrohydrodynamic Atomization in the Cone-Jet Mode are the possible monodispersity of the aerosol, and the sphericity of the particles. The big disadvantage of all production processes that use jet break-up to produce droplets, including Electrohydrodynamic Atomization in the Cone-Jet mode, is the low production rate per spraying point. So, possible applications should require a narrow size distribution, a high droplet charge, or a spherical particle shape. Otherwise, more traditional production processes are likely economically superior to Electrohydrodynamic Atomization in the Cone-Jet mode. In the following paragraphs possible applications of electrohydrodynamic atomization are discussed.

Some medicines require a controlled release of the active substances to the body. The surface of a particle determines the exchange rate between a particle and its surrounding. So, when the particle size can be controlled, and when the particles are spherical, then this release of the active substances can be controlled more easily. Electrohydrodynamic atomization in the Cone-jet mode is able to produce spherical particles of the required size.

Some medicines have to be inhaled into the lungs. When particles are too big, they cannot reach the lower areas of the lungs, because they are collected in the airways. When the particles are too small, a large percentage of the droplets are not collected. They just leave the
Introduction

lungs again. It is also difficult to produce an aerosol of particles in the order of micrometers, because particles of this size stick to each other due to the van der Waals force.

The production of toner powder requires a narrow particle size distribution. If the particles are too big, then the quality of the printing is too low. If the particles are too small, then it is impossible to print them. Also, the toner powder should form a firm layer of particles. This is only possible if the size distribution is sufficiently narrow.

In combustion processes a smooth burning process requires droplets of equal size. However, in this case the low flow rate per spraying point is often a large drawback.

The high droplet charge offers for paint spraying the possibility to guide the droplets to the target. In this way only a small amount of paint is lost.

Also, for crop spraying, the high droplet charge helps to enhance the collection of the droplets on the plants. So less toxic chemicals have to be used, and less chemicals come into the environment.

Electrohydrodynamic Atomization can also be used to produce thin layers of polymorf material on a surface. The high droplet charge guides the droplet to the surface. The controllable size makes it possible to vary the thickness of the layer, and to vary the pore size of the layer. The distance between the spraying point and the surface of the counter electrode is an extra parameter that can be used to control the amount of evaporation of the droplets. A monodisperse spray produced in the Cone-Jet mode has also a very narrow charge distribution. So in this mode, the residence time in the air is almost equal for all droplets.

A completely new application is Bipolar Coagulation, Borra et al. (1996a, 1998a). In this process both positively charged as well as negatively charged sprays are produced. The coalescence of oppositely charged droplets brings a discharged droplet, and offers the possibility to induce chemical reactions inside the droplets at the moment of coalescence.

1.2 INTRODUCTION TO THE CONTENTS OF THIS THESIS

The objective of this thesis is to investigate the parameters that have an influence on the droplet size, the droplet size distribution, and the current produced in Electrohydrodynamic Atomization in the Cone-Jet mode. When this influence is known, then it should be possible to completely control the production of a truly monodisperse spray. The thesis will show that Electrohydrodynamic Atomization in the Cone-Jet mode can be modeled by means of several physical models. These models are developed and the results are compared to experimental results. The acquired insight into the phenomenon of Electrohydrodynamic Atomization in the Cone-Jet mode will be used to improve the scaling laws. The scaling laws predict the droplet size and the current as functions of the liquid properties, and the liquid flow rate. The simulations will show that the influence of the dielectric permittivity of the liquid is much smaller than previously assumed. The influence of the radial profile of the axial liquid velocity on the current can be estimated, and this estimation can be used to derive one current scaling law for all Newtonian liquids that can be sprayed in the Cone-Jet mode. The comparison between experimental results and calculations will also reveal that the produced droplet size depends on the jet break-up
mechanism. So the jet break-up mechanism has to be incorporated into the derivation of the scaling law for the droplet size. Two relations will be derived one for the varicose jet break-up regime and one for the whipping jet break-up regime. This insight and these scaling laws can be used for the production of powders with a narrow size distribution.

Chapter 2 describes electrohydrodynamic atomization in general and the Cone-Jet mode in particular. This chapter describes the various spraying modes of electrohydrodynamic atomization. It answers the question why the Cone-Jet mode was chosen as subject of this research, and it describes the history and characteristics of the Cone-Jet mode.

Electrohydrodynamic atomization in the Cone-Jet mode has to be described by at least three different processes. The first process is the acceleration of the liquid in the liquid cone and jet. This acceleration process and the shape of the liquid cone are a result of the balance of the liquid pressure, liquid surface tension, gravity and electric stresses in the liquid surface, and of the inertia and viscosity of the liquid. Chapter 3 will describe the physical numerical Cone-Shape model. This model is the first model able to calculate the shape of the liquid cone and jet, the electric fields in and outside the cone, the surface charge density on the cone and jet surface, and the influence of the electrode geometry on the current. The calculated shape is compared to pictures of the experimental shape. The simulation current is compared to the experimental current, and to the present current scaling laws.

The second process is the break-up of the jet into droplets. The jet emerging at the cone apex breaks up into droplets. Chapter 6 describes this jet break-up process. This process is investigated with a high definition camera with a long distance microscopic lens. Five different jet break-up regimes have been distinguished, which all produce different size distributions. The parameters influencing the occurrence of these jet break-up regimes, and their influence on the average particle size are investigated. A mathematical physical model is presented, which is able to predict the main droplet size, and the droplet velocity at jet break-up for axisymmetric or varicose instabilities. A relation is derived, which can be used to calculate the droplet size in the whipping jet break-up regime from the current and the liquid flow rate.

The third process is the evolution of the spray. Chapter 5 describes this evolution of the spray just after production. The Cone-Jet mode produces highly charged droplets. Due to the electric forces between these droplets, and due to differences in inertia, a size segregation effect occurs in the spray just after production. A model is presented, which is able to simulate the size segregation, the droplet velocity, and the droplet concentration in the spray. The simulation results are compared to droplet size and droplet velocity measurements. The air velocity inside the spray is investigated.

Sometimes, a fourth process can be of importance, namely the occurrence of electrical discharges in the gas surrounding the liquid cone. Chapter 4 describes the effects of electrical discharges in the air on the droplet production process in the Cone-Jet mode. When the electric field strength near the liquid cone exceeds the onset field strength for electrical discharges, then these discharges will occur. The electrical discharges will disturb the droplet production process at the cone apex. Results from literature are discussed, and simulation results of the Cone-Shape model concerning electrical discharges are presented. The influence of the liquid flow rate, the
surface tension, and the conductivity on the discharge probability is shown. The position on the jet with the largest discharge probability is located.

Chapter 7 presents a new scaling of the current through the liquid cone. A new derivation is presented, and the scaling is extended to low viscosity and low conductivity liquids. The influence of the ion species used to enhance the conductivity is investigated. A relation is derived, which can be used to calculate the droplet size in the varicose jet break-up regime from the current and the liquid flow rate.

The final aim of this research is to produce powders with a narrow size distribution with a required average particle size. Disadvantages of Electrohydrodynamic Atomization in the Cone-Jet mode can be the high droplet charge and the low production rates per spraying point. Chapter 8 presents a first design of a reactor for the production of monodisperse polymer powder. The influence of neighboring nozzles, and ions produced for discharging of the droplets is investigated. A polymer powder with a narrow size distribution is produced. This chapter will show that the initial polymer concentration in the solvent influences the particle size and shape.

Chapter 9 presents a summary of the conclusions of this thesis.
CHAPTER 2

ELECTROHYDRODYNAMIC ATOMIZATION
IN THE CONE-JET MODE

This chapter describes electrohydrodynamic atomization in general, and the Cone-Jet mode in particular. It briefly discusses the various spraying modes that can be distinguished in electrohydrodynamic atomization. It will answer the question, why the Cone-Jet mode is chosen as subject of investigation, and not one of the other spraying modes. The third section discusses the history and some characteristics of the Cone-Jet mode.

2.1 ELECTROHYDRODYNAMIC ATOMIZATION
AND ITS VARIOUS SPRAYING MODES

A lot of confusion existed, and sometimes still exists between researchers in the field of Electrohydrodynamic Atomization. The reason of this confusion is that depending on the liquid, the liquid flow rate, the potential difference, the electrode configuration, and the capillary or nozzle used, various spraying modes can be obtained. Bailey (1988), Cloupeau and Prunet-Foch (1994), and Grace and Marijnissen (1994) gave reviews of these spraying modes.

Electrohydrodynamic atomization refers to a process, where a liquid jet breaks up into droplets under influence of electrical forces. However, a liquid jet will also break up into droplets without any electric force present. Depending on the strength of the electric stresses in the liquid surface relative to the surface tension stress, and depending on the kinetic energy of the liquid leaving the nozzle, different spraying modes will be obtained. Section 1.2 already introduced the Cone-Jet mode. The other Electrohydrodynamic Atomization modes are discussed in the following sections.

2.1.1 Microdripping, Spindle Mode and Intermittent Cone-Jet Mode

A liquid is pumped through a capillary at a constant flow rate. In case of a low liquid flow rate, without an electric field applied over the droplet hanging down from the capillary, droplets are produced at frequencies lower than 1 Hz. When an electric field is applied, which slowly increases, then the frequency of droplet production increases. Depending on the electric field, and
on the liquid flow rate, at first three different spraying modes can occur, Microdripping, Spindle mode and Intermittent Cone-Jet mode.

Microdripping occurs at the lowest liquid flow rates, and produces a narrow size distribution at a production frequency of a few kHz. The drop hanging down from the capillary is transformed due to the applied electric field. The electric field generates a surface charge in the droplet. This results in an electric stress, which reduces the effective surface tension stress in the droplet surface. The size of the droplet at the capillary increases, until the electric stresses and the gravitational pressure overcome the surface tension stress. In that case, a small droplet will be ejected. The large droplet at the capillary relaxes to its original shape. It will start to grow until a new droplet is ejected.

The Spindle and the Intermittent Cone-Jet mode occur at a higher liquid flow rate and at higher electric fields than Microdripping. They produce broader droplet size distributions. The droplet shape at the capillary is transformed by the electric stresses into a conical shape. At the apex of this cone, a jet emerges, which can break up into a large number of small and highly charged droplets. This is similar to the Cone-Jet mode. The highly charged aerosol reduces the electric field at the liquid cone. This field reduction makes it possible for the surface tension stress to overcome the electric stresses. The liquid cone relaxes to a normal droplet shape. The way the cone relaxes to a normal droplet shape, determines the spraying mode. The Spindle mode produces a bimodal size distribution. After having produced a spray of small droplets, the jet detaches from the relaxing cone, and forms one big droplet. In the Intermittent Cone-Jet mode, the relaxation of the liquid cone produces a wide range of droplet sizes.

2.1.2 Multiple-Jet Mode and the Rim-Emission Mode

When coming from the modes of the previous section, the applied potential difference can be increased. As a result, the electric field strength around the liquid cone will increase. Suddenly, the liquid cone does not relax to a droplet shape anymore. The droplet production process becomes stable in time. This is the Cone-Jet mode. If the applied potential difference is increased even further, then the spraying mode goes from the Cone-Jet mode into the Multiple-Jet mode or into the Rim-Emission mode.

With increasing potential difference the liquid cone of the Cone-Jet mode becomes smaller and smaller. At a certain moment, the liquid cone is too small for the capillary. The cone moves from the center of the capillary toward the edge. When the potential difference is increased further, then a second cone will occur. With increasing potential difference, more and more cones will be formed. For a Multiple-Jet mode, the bases of these cones are still attached. Together, these cones can be considered to be one droplet with multiple spraying points. In the Rim-Emission mode, a large number of very small cones are formed in a thin layer of liquid on the edge of the capillary. Which mode will occur depends mainly on the shape of the capillary, but also on the applied potential difference.
2.1.3 Simple-Jet Mode and Ramified-Jet Mode

If the liquid flow rate is relatively high, then the kinetic energy of the liquid leaving the capillary can be too large to be compensated by the surface tension. In this case, a free jet is formed, which breaks up into droplets. This is the Simple-Jet mode. The electric forces are mainly used to control the break-up of the jet into droplets. Frequency modulation can enhance the monodispersity of the produced droplets, Balachandran et al. (1992). If the electric field around this free jet is increased, then the radial electric stresses in the jet can overcome the surface tension stress. In that case, small jets are emerging from the surface of the main jet. These jets break up into a polydisperse spray. This mode is called the Ramified-Jet mode. Figure 6.1 shows a Ramified-Jet as an example of the m=2 mode for the instabilities that play a role in the jet break-up.

2.2 WHY IS THE CONE-JET MODE OF INTEREST?

Why is the Cone-Jet mode chosen as subject of this research? As already partly discussed in the previous sections, the Spindle mode, and the Intermittent Cone-Jet, produce polydisperse sprays. All kinds of very simple nebulizers can produce polydisperse sprays at much higher flow rates than these two modes of electrohydrodynamic atomization. So little benefit can be gained by using electrohydrodynamic atomization in one of these modes. The Ramified-Jet mode produces also a polydisperse spray, but the liquid flow rates are much higher. So, in cases where polydispersity is not a problem, and high droplet charges are a benefit, it can be interesting to use this mode. However, the aim of this study is to produce a monodisperse spray.

Microdripping produces a monodisperse spray, but the production frequency is very low. The Simple-Jet mode is interesting for certain applications, but has the same disadvantage as the vibrating orifice. The inner diameter of the capillary determines the produced droplet size. The smaller the required droplets, the smaller the inner diameter of the capillary, the more problems with clogging of the capillary. The lower limit is a droplet diameter of about 20 µm.

Probably the most interesting modes for the production of powders at an industrial scale are the Multiple-Jet and Rim-Emission mode. It is theoretically possible to produce a monodisperse spray. The liquid flow rate per spraying point is the same as in the Cone-Jet mode, but the number of spraying points per nozzle is much higher than for the Cone-Jet mode. However, these spraying modes are more difficult to control than the Cone-Jet mode. The challenge is to design a nozzle, and an electrode configuration that yields the required droplet size distribution.

The Cone-Jet mode is the basis of the Rim-Emission, and the Multiple-Jet mode. In order to be able to understand, and to control the Rim Emission and the Multiple-Jet mode, first the Cone-Jet has to be understood. That is why this research is concentrated on the Cone-Jet mode.
Figure 2.1 Schematic representation of the processes inside the liquid cone.
2.3 HISTORY AND CHARACTERISTICS OF THE CONE-JET MODE

Electrohydrodynamic atomization in the Cone-Jet mode has been studied for many years now. The phenomenon was first mentioned in 'De Magnete' by William Gilbert in 1600. He observed that a piece of amber, held at a suitable distance, attracts spherical droplets lying on a dry surface, and draws them up into cones. However, it was Zeleny (1914, 1915, 1917), who gave the first solid scientific description of the process. After Vonnegut and Neubauer (1952) rediscovered this phenomenon, many people have contributed to the understanding of Electrohydrodynamic Atomization in the Cone-Jet mode.

Electrohydrodynamic atomization in the Cone-Jet mode has to be described by at least three different processes. The first process is the acceleration of the liquid in the liquid cone. Based on the work of Gañán-Calvo et al. (1997), Fernández de la Mora and Loscertales (1994), and many other people, the following description of this acceleration can be given. The electric field induces a free surface charge in the cone surface. In a liquid, charge is mainly transported by ions. So, the free charge at the liquid surface mainly consists of ions. Due to this surface charge, the normal electric field inside the liquid is small compared to the normal field outside the liquid. The tangential electric field accelerates the ions at the liquid surface toward the cone apex. These ions accelerate the surrounding liquid. As a result, a thin jet emerges at the cone apex. This acceleration process, and the shape of the liquid cone, are a result of the balance of the liquid pressure, liquid surface tension, gravity and electric stresses in the liquid surface, and of the inertia and viscosity of the liquid. Figure 2.1 shows the processes that play a role in the liquid cone. It also shows the cylindrical coordinate system used in this thesis. Taylor (1964) was the first to describe the balance between the surface tension stress and the normal electrical stress in a liquid cone. However, Taylor's solution is only valid in one extreme case. In his solution, there are no jet, and no tangential stress. In reality the process is more complicated. Inside a liquid, two mechanisms of charge transport exist. Firstly, due to the electric fields inside the liquid, charge is transported through conduction. Secondly, due to the tangential electric field, the ions of the free surface charge are accelerated toward the cone apex. Their velocity is mainly determined by the flow properties of the liquid. This mechanism is called charge convection. The shape of the cone is mainly determined by a balance between surface tension, normal and tangential electrical stress, and gravity. The liquid cone can be considered as an almost perfect conductor with low electric field strengths inside the liquid. The transport of charge mainly occurs through conduction. However, when going from the cone base toward the cone apex, then the electric fields, the surface charge, and the axial liquid velocity, will increase. This results in a higher charge transport due to charge convection, and a larger contribution of the kinetic energy to the cone shape. In the jet, the charge transport occurs mainly through charge convection.

The second process is the break-up of the jet into droplets. The jet emerging at the cone apex, breaks often up into a bimodal size distribution. This bimodal size distribution occurs because the jet breaks up into main droplets with a narrow size distribution, and a number of smaller secondary droplets. In some cases, the filament between the secondary droplet, and a main droplet forms an even smaller satellite droplet. Figure 2.2 shows a schematic jet breaking
Electrohydrodynamic Atomization in the Cone-Jet Mode

up into droplets due to axisymmetric varicose instabilities. The number of secondary droplets is of the same order of magnitude as the number of main droplets. However, the volume fraction of the secondary droplets is much smaller. Due to the free surface charge, the produced droplets are highly charged.

The break-up of a jet has been studied by many people. For instance, Rayleigh (1878), and Weber (1931), have presented theories to predict the growth rate of varicose instabilities on a liquid jet. In their calculations, there was no electric field, and the velocity of the jet was constant. This theory is able to predict the wavelength of the fastest growing varicose instability, and the influence of viscosity on this wavelength. The higher the viscosity, the longer the dominant wavelength. The fastest growing wavelength is often called the dominant wavelength, because the jet always breaks up through instabilities with a wavelength close to the fastest growing wavelength. Although a longer wavelength means a relatively larger volume of secondary droplets, this theory is not able to predict the secondary droplets. Yuen (1968) included nonlinear effects in the jet break-up. From this theory, the total volume of the secondary droplets could be estimated.

Electric stresses play an important role in electrohydrodynamic atomization. Among others, Melcher (1963), and Parkin (1973), have studied the influence of charge on the jet break-up. With increasing electric stresses, a larger perturbation growth rate, and a shorter dominant wavelength, were found. The last effect that has to be taken into account is the acceleration of the jet due to the electric forces. In the above-mentioned theories, the calculated dominant wavelength always depends on the radius of the jet. The jet velocity, and the jet radius, are constant. For an accelerating jet, the jet radius decreases, and the wavelength of a perturbation on the jet surface increases. A certain perturbation on the jet surface does not have a fixed wavelength. Therefore, the above-mentioned theories have to be adjusted for Electrohydrodynamic Atomization in the Cone-Jet mode.

The third process in Electrohydrodynamic Atomization in the Cone-Jet mode is the evolution of the spray after droplet production. Electrical interaction between highly charged droplets causes a size segregation effect. The smaller droplets will be found at the edge of the spray, and the larger droplets will be found in the spray center, Gañán-Calvo et al.(1994). If the highly charged droplets evaporate, then the Rayleigh limit for droplet charge can be reached. In that case, droplet fission can take place, which changes the produced size distribution. This happens, if the electric stress, due to the droplet charge, becomes larger than the surface tension stress. The
droplet becomes unstable, and will expel a number of smaller droplets. Rayleigh derived the following equation,

$$ q_R = 8\pi \left( \gamma \epsilon_0 r_d^3 \right)^{\frac{1}{2}} $$

(2.1)

where, $q_R$ is the Rayleigh Charge Limit [C], $\gamma$ is surface tension [N m$^{-1}$], $r_d$ is droplet radius [m], and $\epsilon_0$ is dielectric constant of a vacuum [$C^2 N^{-1} m^{-2}$]. Schweizer and Hanson (1971) confirmed experimentally this equation. Tang and Gomez (1994) showed that droplet charge for electrohydrodynamic atomization in the Cone-Jet mode is lower than the Rayleigh Charge Limit. However, at higher flow rates the droplets are closer to the Rayleigh Charge Limit than at lower flow rates. Law (1989) showed that the evaporation rate of a liquid is not influenced by the charge of the droplet, as long as the particles are larger than 0.1 μm. He also showed, that charge is not dissipated by evaporation, but solely by the impact of ions and electrons from the surrounding gas.

Sometimes, a fourth process can be of importance. Namely, the occurrence of electrical discharges in the surrounding gas, Hayati et al.(1987), Cloupeau and Prunet-Foch (1989). These discharges can completely destabilize the droplet production process. Whether this phenomenon will occur, depends on the electric field, on the shape of the liquid jet, and on the properties of the surrounding gas. The electric field, and the shape of the jet, depend in their turn on the electrode configuration, and on the liquid properties, like surface tension.

Many parameters influence the finally produced droplet size, and droplet charge. These parameters will be discussed in the following sections.

2.3.1 Scaling of Droplet Size and Current

Various equations have been derived to estimate the produced average droplet size and the required electric current for a liquid sprayed in the Cone-Jet mode. Based on experimental results of Kim and Turnbull(1976), Gomez and Tang (1990), and Fernández de la Mora et al.(1990) came with the following dimensionless number,

$$ \frac{\gamma r_{jet}}{\rho Q^2/2\pi^2 r_{jet}^4} $$

(2.2)

where, $Q$ is liquid flow rate [m$^3$ s$^{-1}$], $\rho$ is liquid density [kg m$^{-3}$], and $r_{jet}$ is jet radius [m]. This dimensionless number represents the ratio of the surface tension stress in the jet over the kinetic energy of the jet. Combining dimensional analysis with experimental results, Fernández de la Mora and Loscertales (1994) arrived at the following relationships for liquids with relatively high conductivities.
Electrohydrodynamic Atomization in the Cone-Jet Mode

\[ I = f (\varepsilon_r) \left( \frac{\gamma Q K}{\varepsilon_r} \right)^{\frac{1}{2}} \]  

(2.3)

\[ d_{jet} = 0.4 \left( \frac{Q}{\varepsilon_0 \varepsilon_r K} \right)^{\frac{1}{3}} \]  

(2.4)

where, K is conductivity \([\text{S m}^{-1}]\), \(\varepsilon_r\) is relative dielectric constant of the liquid, \(I\) is current \([\text{A}]\), and \(d_{jet}\) is jet diameter \([\text{m}]\).

Also based on dimensional analysis and extensive experimental results, Gañán-Calvo et al. (1997) presented two different relations for the droplet size and current as functions of the liquid properties, and the liquid flow rate. They found that highly conductive liquids, which have a flat radial profile of the axial liquid velocity, behaved differently from low conductivity and low viscosity liquids. This last category of liquids do not have a flat velocity profile in the jet. The fluid velocity at the jet surface is higher than at the jet center. In order to judge which relation is valid, they introduced the following dimensionless number.

\[ \left( \frac{\gamma^2 \varepsilon_0^2}{\mu^2 K^2 Q} \right)^{\frac{1}{3}} \]  

(2.5)

Where, \(\mu\) is absolute viscosity of the liquid \([\text{Pa s}]\). This dimensionless number represents the ratio of the change in kinetic energy of the liquid jet in the axial direction over the change in viscous stress in the radial direction. If the viscosity is high or the jet diameter is small, then the viscous stresses are relatively high. The size of the jet diameter is mainly determined by the liquid flow rate and the liquid conductivity. A higher conductivity requires a lower liquid flow rate. This results in a smaller jet radius. The higher the viscosity, and the smaller the jet radius, the less difference between the axial liquid velocity in the center of the jet and this velocity at the liquid surface. The radial profile of the axial liquid velocity, see Figure 2.1, will be almost flat. In that case, dimensionless number (2.5) is smaller than one, and the current and droplet size relations are equal to

\[ \frac{d_j}{d_o} = 1.6 (\varepsilon_r - 1)^{\frac{1}{6}} \left( \frac{Q}{Q_o} \right)^{\frac{1}{3}} - 1.0 (\varepsilon_r - 1)^{\frac{1}{3}} \]  

\[ \frac{I}{I_o} = \frac{6.2}{(\varepsilon_r - 1)^{1/4}} \left( \frac{Q}{Q_o} \right)^{\frac{1}{2}} - 2.0 \]  

(2.6)

\[ I_o = \left( \frac{\varepsilon_0 \gamma^2}{\rho} \right)^{\frac{1}{2}} \]  

\[ d_o = \left( \frac{\gamma \varepsilon_0^2}{\rho K^2} \right)^{\frac{1}{3}} \]  

\[ Q_o = \frac{\varepsilon_r \gamma}{K \rho} \]  

(2.7)
Based on their dimensional analysis and on their experimental results, Gañán-Calvo et al. (1997) concluded that the minimum flow rate $Q_{\text{min}}$ [m$^3$ s$^{-1}$] is related to $Q_o$ in the following way

$$ Q_{\text{min}} \sim Q_o \left( \varepsilon_r - 1 \right)^{\frac{1}{2}} $$

(2.8)

$I_o$ represents the current at the minimum flow rate [A], and $d_o$ is the droplet diameter at the minimum flow rate. The minimum flow rate is the lowest flow rate for a certain liquid at which it can be sprayed in the Cone-Jet mode. Below this flow rate the liquid cone relaxes every now and then to a normal droplet shape. Rosell-Llombart and Fernandez de la Mora (1994) concluded from their experimental results that the minimum flow rate was related to $Q_o$ in the following way

$$ Q_{\text{min}} \sim \varepsilon_r Q_o $$

(2.9)

Rosell-Llombart and Fernandez de la Mora (1994) also used the relations for $I_o$ and $d_o$ at the minimum flow rate. Chen and Pui (1997) compared both relations and arrived at the conclusion that equation (2.8) was closer to their experimental results. However, a real fit between experiments and equation (2.8) was not found. Also, a solid theoretical explanation of the minimum flow rate was not given.

In case of low viscosity and low conductivity liquids, the radial viscous stresses are relatively low compared to the change in kinetic energy in the axial direction. The dimensionless number (2.5) will be larger than 1. In that case the liquid velocity at the jet surface will be much larger than the velocity in the jet center. In this case, the current and droplet size can be estimated using the relations

$$ \frac{d_d}{d_o} = 1.2 \left( \frac{Q}{Q_o} \right)^{\frac{1}{2}} - 0.3 \quad \frac{I}{I_o} = 11.0 \left( \frac{Q}{Q_o} \right)^{\frac{1}{4}} - 5.0 $$

(2.10)

The validity of the relations presented in this section will be discussed in Chapters 3, 6, and 7.

2.3.2 Parameters Influencing the Droplet Production Process

Looking at the equations given in the previous section, it can be concluded that liquid properties like the dielectric constant, conductivity, viscosity, and surface tension, play an important role in the atomization process in the Cone-Jet mode. Other factors that influence this process are the liquid flow rate, the electrode configuration, the shape of the capillary in combination with the
wettability of the material, and the direction of gravity.

**Conductivity**

The Electrical conductivity of a liquid is one of the main parameters in Electrohydrodynamic Atomization. If the conductivity is too low, about $10^{-8} - 10^{-10}$ S m$^{-1}$, the liquid cannot be sprayed in the Cone-Jet mode. Except, when extra free charge is produced in the liquid drop at the capillary. Kelly (1990, 1994) created this free charge by creating an electrical discharge inside the liquid. The produced free charge will move toward the liquid surface. Due to this free charge, and due to the electric field, the required tangential electric stress will occur in the liquid surface. For liquids with a higher conductivity, the creation of extra free charge is not necessary. The conductivity of the liquid is sufficient to conduct a current from the nozzle electrode to the liquid surface. Whether such a liquid can be sprayed in the Cone-Jet mode, depends especially on the applied liquid flow rate, and on the occurrence of electrical discharges in the air surrounding the jet.

**Dielectric Permittivity**

The dielectric constant of a material determines the polarization of this material in an electric field. The polarization reduces the electric field inside the liquid. The electric field, and the liquid conductivity determine the time constant for the charge transport toward the cone surface. According to Velenga and Klinkenberg (1965), the following relaxation time can be used at the cone apex.

$$\tau = \frac{\varepsilon_0 \varepsilon_r}{K} \quad (2.11)$$

Fernández de la Mora and Loscertales (1994) used this time constant to derive equations (2.3), and (2.4).

**Surface Tension**

In order to form a stable Cone-Jet, the surface tension has to be overcome by the electric stresses. The higher the surface tension, the stronger the required electric field strength. The stronger the required electric field, the more chance that electrical discharges will occur in the air surrounding the cone. Stronger electric fields will also result in a higher free surface charge at the cone surface. This results in a higher electric current through the cone and jet. A higher current results in a higher droplet charge. The droplet size is often considered to be independent of the surface tension. Gañán-Calvo et al. (1997) considered in their analysis for highly conducting liquids the droplet size to be independent of the surface tension. However a small dependence came back
due to fitting parameters, which were used to fit the theoretical relations to experimental results.

**Liquid Flow Rate**

The liquid flow rate has a strong influence on the spraying process. If the liquid flow rate is too low, the spraying process is not stable in time, because the liquid cone relaxes every now and then to a droplet shape. Above the minimum flow rate, the droplet production process is stable, and the produced size distribution of the main droplets is narrow. However, if the flow rate is increased above a certain value, then the produced droplet size distribution becomes more polydisperse. With increasing flow rate, the produced droplet size distribution becomes more and more polydisperse. At even higher flow rates, the droplet production process will be unstable in time. Equations (2.7), (2.8), and (2.9), show that the minimum flow rate depends on the liquid properties, like conductivity and surface tension.

**Density**

The liquid density is of some importance since the density plays a role in the acceleration process of the liquid near the cone apex. Gravity is not relevant in that area, because the electric stresses and the surface tension stress are much larger than the hydrostatic pressure. At the cone base, the acceleration of the liquid is very small, and does not play an important role. However, the liquid density is of some importance due to the influence of gravity on the cone shape. For large capillaries, diameters larger than 1 mm, gravity influences strongly the shape of the liquid cone. As a result, large nozzles must spray downwards. When the nozzle is turned upside down, then the electric forces at the cone base are not strong enough to maintain a stable cone. For capillaries smaller than 1 mm, the electric stresses and the surface tension stress are dominant everywhere in the cone. These small nozzles can be turned upside down.

**Viscosity**

Viscosity has its influence on the jet break-up process, and the size of the produced main and secondary droplets. According to Weber (1931), an increase in viscosity will lead to lower dominant wave numbers. Lower wave numbers will lead to a small increase in the droplet size, and to a larger increase of the secondary droplet size. However, in most cases, viscosity is reported to have little influence on the produced droplet size. The only reported influence, Gañán-Calvo et al. (1997), was already discussed in the previous section, and has nothing to do with the jet break-up process. This paradox will be discussed in Chapter 6.

**The Applied Potential Difference**

The applied potential difference is not found in the equations describing the Cone-Jet Mode. One of the reasons is that Electrohydrodynamic Atomization is a field driven process. The electric
field strength depends not only on the applied potential difference but also on the electrode configuration, and on the shape of the liquid cone. The shape of the liquid cone depends in its turn on the electric field strength. This makes it difficult to include the potential difference into analytical equations and makes it even more difficult to include the electric field strength in these equations. Another reason why the potential difference is not found in the equations is that if the applied potential difference is too low, then the spraying process is in the Microdripping mode, the Spindle mode or the Intermittent Cone-Jet mode. If the potential difference is too high, then the spraying process is in the Rim-Emission mode or in the Multiple-Jet mode. In the range of the potential differences of the Cone-Jet mode, the droplet size and current varied less than 7 percent, Gañán-Calvo et al. (1997).

If the potential difference over the electrodes increases and the supply of liquid to the cone remains the same, then the liquid flow rate into the jet will increase, and the liquid cone will become smaller. As a result, the increased electric field strength will be reduced again to values just above its original value, and the flow rate toward the jet will decrease again until a stable jet with the appropriate flow rate into the jet is formed. This adjustment of the cone size reduces the effect of the increase in potential difference. Consequently the influence of the potential difference on the current and the droplet size is small.

Capillary Shape and Material Wettability

The shape of the capillary and the wettability of the material has a large influence on the shape of the cone, the stability of the jet, and the spraying mode obtained. Cloupeau and Prunet-Foch (1994) state that very small changes in capillary shape or wettability can cause the occurrence of a different spraying mode. Cloupeau and Prunet-Foch (1989) also stated that a change in wettability can cause the following effect. 'Liquid rises on the capillary and accumulates near the outlet section, forming a ridge over the entire periphery or on a small part only. This ridge sporadically emits a jet of fine droplets, or finally detaches into a large drop and the cycle begins again.' At the base of the cone, near the capillary, the shape of the cone strongly depends on the shape of the capillary, so an unstable behavior at the liquid-capillary interface will give rise to an unstable jet.
CHAPTER 3

PHYSICAL MODELING OF THE LIQUID CONE AND JET

A physical model has been developed which is able to calculate the shape of the liquid cone and jet, the electric fields in and outside the cone, and the surface charge density at the liquid surface. The model also estimates the liquid velocity at the cone surface. From these results the current through the liquid cone can be calculated. The results of this Cone-Shape model fit well with experimental values with respect to the cone shape, and the current as functions of the flow rate, the conductivity, the potential difference, and the electrode configuration. The nozzle diameter gives rise to deviations from the present scaling laws. The calculations showed that a spraying nozzle with a diameter of 1 mm yields a 15% higher current than an 8 mm nozzle. Thus, current scaling laws that do not take the nozzle diameter into account can never be very accurate. For the 8 mm nozzle the current is almost independent of the potential difference, where for the 1 mm nozzle a stronger relation was found between current and potential. The model neglects the space charge due to the droplet charge. Neglecting the space charge has an influence on the electric field around the cone. This influence can be compensated by reducing the potential in the model by 5 to 7 percent. So, the space charge has only a small influence on the spraying process. The jet diameter was calculated and measured as function of the liquid flow rate.

3.1 INTRODUCTION

In electrohydrodynamic atomization in the Cone-Jet mode a liquid is supplied to a nozzle at a low flow rate. A droplet is formed at this nozzle. When a strong electric field is applied over this droplet, then the electric field induces free charge in the liquid surface. As a result, electric stresses occur in this surface. These stresses transform the droplet shape into a conical shape. At the cone apex a liquid jet with a high charge density occurs. The shape of the liquid cone is a result of the force balance of surface tension, gravity, electric stresses in the liquid surface, inertia and viscous stresses. Taylor (1964) was the first to calculate analytically a conical shape. For this cone shape the surface tension stress is balanced with the electrical normal stress. Taylor assumed a cone without liquid jet. Consequently, there was no electrical current and liquid flow
through his liquid cone. Joffre and Cloupeau (1986) approached the problem numerically, which allowed a more flexible cone shape. In Taylor’s case the cone had to be a perfect mathematical cone. Only the angle at the cone apex could be used as a parameter. However, also in Joffre’s calculations there was no jet. The cone apex was just rounded off.

In section 2.3.1, the scaling laws for the droplet size and current were presented. The following relations can be derived from these scaling laws

\[ d_d = b_1(\varepsilon_r) \left( \frac{Q\varepsilon_0\varepsilon_r}{K} \right)^{1/3} + c_1 \]
\[ I = b_2(\varepsilon_r) \left( \frac{\gamma Q K}{\varepsilon_r} \right)^{1/2} + c_2 \]  

(3.1)

where, \( d_d \) is droplet diameter [m], \( Q \) is liquid flow rate \([\text{m}^3 \text{s}^{-1}]\), \( K \) is conductivity \([\text{S m}^{-1}]\), \( \gamma \) is surface tension \([\text{N m}^{-1}]\), \( \varepsilon_r \) is relative permittivity of the liquid, \( \varepsilon_0 \) is permittivity of a vacuum \([\text{C}^2 \text{V}^{-1} \text{m}^{-1}]\), \( I \) is current [A], \( b_1 \) and \( b_2 \) are functions of the liquid permittivity, and \( c_1 \) and \( c_2 \) are constants. These relations are often called the scaling laws for electrohydrodynamic atomization in the Cone-Jet mode.

In this chapter a model is presented, which is the first model able to calculate the shape of the liquid cone and jet, and which makes it possible to investigate the influence of the electrode geometry. The model also calculates the electric field strength, the charge density, and the current through the liquid cone. This model is of interest, firstly, because the present scaling laws can be evaluated and improved. In experiments, there is always the problem that when one liquid property is changed, also other properties will change. Secondly, for liquids with a surface tension larger than 0.05 N m\(^{-1}\) electrical discharges in the surrounding air will occur, Smith (1986). These discharges can disturb the droplet production process. The model calculates the electric fields in and around the liquid cone and jet. This allows further investigation of electrical discharges in the air surrounding the cone. Thirdly, knowing the shape of the liquid jet and knowing the surface charge on this jet, allows the investigation of the jet breaking up into droplets. Fourthly, the model calculates also the electric field in the gap between the jet and some counter electrode. This improves the calculation of the size segregation effect that occurs in the spray just after the droplet production. Finally, the influence of electrode configurations on the current can be investigated much more easily.

In the following sections a description of the model, will be given. Calculations will be compared to new experimental results and results presented by Gañán-Calvo et al. (1997). In most cases, the deviation between experiment and simulation is less than 10 percent. The simulations will also show the influence of the conductivity, liquid flow rate, viscosity, the potential difference, and the electrode configuration on the current, and on the jet diameter.

### 3.2 NUMERICAL CONE-SHAPE MODEL

The droplet size produced in the Cone-Jet mode mainly depends on the liquid flow rate and liquid properties like density, viscosity, conductivity, electrical permittivity and surface tension.
Also, the electrode configuration and the potential difference have some influence. A physical numerical model has been developed in order to investigate the influence of these parameters on the cone and jet radius. The model has also been used to calculate the current through the liquid cone. Figure 3.1 shows the forces that have to be taken into account.

In the liquid cone, the charge is transported in two ways. The first way is through conduction in the liquid due to the electric fields. The second way is by charge convection. The electric field induces free charge at the liquid surface. This free charge consists of ions. The electric field at the liquid surface accelerates the ions towards the cone apex. The ions in their turn accelerate the surrounding liquid. It is assumed that the ion velocity cannot be much larger than the liquid velocity, since the ions lose their velocity due to collisions with the liquid molecules. So, the liquid velocity and the ion velocity depend on the local electric field strength, the amount of free charge, and on the flow properties of the liquid. In the present model it is assumed that the liquid is a Newtonian liquid, and the flow properties can be described with the viscosity.

Figure 3.2 shows a schematic representation of the cone shape calculation method. First a certain cone shape is assumed. Then, the liquid velocity in the axial direction at the liquid-air interface is estimated. In the second part a current balance is solved. The balance between the conduction
towards the liquid surface and the charge convection at this surface determines the surface charge at the liquid surface. In the third part of the simulation, the electric fields are calculated using Gauss’ Law. The electric field calculation and the calculation of the current balance are repeated until the change in surface charge is very small. After convergence of the calculations of the electric field and the surface charge, the first three steps of the model are repeated two times. In the fourth part, a new cone shape is calculated using the results of the first three parts of the model. If the new shape is equal to the input shape, then the iteration process is stopped. Otherwise, the new shape is used as input for a new cycle of calculations.

In order to calculate the shape of the liquid cone, the Navier-Stokes equation is solved in one dimension, assuming an axisymmetric, steady state situation. In this equation, the change in potential energy (pressure $p_{liq}$, gravity $p_g$), and the kinetic energy $p_{Ekin}$ (velocity pressure) is balanced with the energy input due to the tangential electric stress $\tau_{Ei}$, the change in polarization stress $\sigma_p$, and the energy dissipation due to the viscous stresses in the liquid $\sigma_v$, $\tau_v$. This balance is calculated over short distances in the axial direction of the cone ($z$ direction).

\[
\frac{\partial}{\partial z} (p_{Ekin} + p_{liq} - \sigma_v - \sigma_p - p_g) = \frac{2}{r_s} (\tau_v + \tau_{Ei})
\]  

(3.2)

where, all stresses and pressures are in N m$^{-2}$, $r_s$ is the radial distance of the liquid-air interface from the axis [m], $z$ is the axial coordinate [m], and where

\[
p_{liq} = p_{out} + \Delta p_{n,liq} + \Delta p_{Ei} + \Delta p_s
\]  

(3.3)

where, $p_{out}$ is the air pressure, $\Delta p_{n,liq}$ is the stress in the liquid surface due to viscous stress, $\Delta p_{Ei}$ is the normal electric stress in the liquid surface, and $\Delta p_s$ is the surface tension stress in the liquid surface.

### 3.2.1 Pressure

The pressure inside the liquid cone depends on the pressure of the surrounding air and the stresses that occur in the liquid-air surface. Since, the air pressure is independent of the axial coordinate $z$, this pressure can be neglected, and only the stresses in the liquid surface are taken into account. The stresses that occur in this surface are the surface tension stress, the normal electric stress, and the viscous surface stress.

#### Surface Tension Stress

The pressure difference due to surface tension between a liquid and the surrounding air can be calculated using
\[ \Delta p_s = \gamma \left( \frac{1}{r_{s1}} + \frac{1}{r_{s2}} \right) \]  

(3.4)

Where, \( \Delta p_s \) is the pressure difference between the liquid and surrounding air due to the surface tension stress \([N \, m^{-2}]\), \( r_{s1} \) and \( r_{s2} \) are radii of curvature of the surface \([m]\), and \( \gamma \) is the surface tension of the liquid \([N \, m^{-1}]\). For a cylindrical coordinate system this equation can according to Kingham and Swanson (1984) be rewritten into,

\[ \Delta p_s = \gamma \left( \frac{1}{r_s \left( 1 + \left( \frac{d r}{d z} \right)^2 \right)^{1/2}} - \frac{d^2 r / d z^2}{\left( 1 + \left( \frac{d r}{d z} \right)^2 \right)^{3/2}} \right) \]  

(3.5)

where, \( \Delta p_s \) is the pressure inside the liquid minus the pressure of the surrounding air \([N \, m^{-2}]\).

**Normal Electric Surface Stress**

The normal electric field induces a free electric charge and a polarization charge in the liquid surface. This electric field and these charges create a normal electric stress in the liquid surface. Following Landau and Lifshitz (1975), the stress in the liquid-air interface due to the normal electric field is equal to

\[ \Delta p_{E_n} = -\frac{1}{2} \epsilon_0 \left( E_{n,\text{out}}^2 - 2 \epsilon_r E_{n,\text{ins}}^2 + E_{n,\text{ins}}^2 \right) \]  

(3.6)

where, \( E_{n,\text{ins}} \) is the electric field strength normal to the liquid-air surface inside the liquid \([V \, m^{-1}]\), and \( E_{n,\text{out}} \) is the electric field strength normal to the liquid-air surface outside the liquid \([V \, m^{-1}]\).

**Viscous Surface Stress**

An accelerating liquid experiences viscous stresses inside the liquid. These stresses can be described by a second order tensor. For an axially symmetric flow the normal stress components are \( 2\mu \partial u_r / \partial z \) and \( 2\mu \partial u_r / \partial r \), Batchelor (1967). For a non compressible flow, with cylindrical symmetry, the continuity equation yields

\[ \frac{\partial u_r}{\partial r} + \frac{1}{r} \frac{\partial u_z}{\partial z} = 0 \]  

(3.7)

where, \( u_r \) is the liquid velocity in the radial direction \([m \, s^{-1}]\), and \( u_z \) is the liquid velocity in the axial direction \([m \, s^{-1}]\). Figure 3.3 represents the balance of normal stresses at the liquid surface.
These normal stresses have to be in balance with each other. This results in the following relation

$$\Delta p_{n, \mu} = \frac{2(\partial r / \partial z)^2 - 1}{(\partial r / \partial z)^2 + 1} \mu \frac{\partial u_z}{\partial z}$$

(3.8)

where $\mu$ is the absolute viscosity of the liquid [Pa s].

In order to calculate the viscous stress in the liquid surface, a flat radial profile of the liquid velocity in the axial direction is assumed. This is a reasonable assumption because, when the influence of the viscosity is relatively large, then this velocity profile in the cone will be almost flat. If the viscosity is low, then the radial profile will not be flat, but the contribution of the viscous stress will be small. When assuming a flat radial liquid velocity profile, then the liquid velocity is equal to

$$-u_z = \frac{Q}{\pi r_s^2}$$

(3.9)

This assumption yields

$$\frac{\partial u_z}{\partial z} = \frac{2Q}{(\pi r_s^3)} \frac{dr_s}{dz}$$

(3.10)

### 3.2.2 Kinetic Energy

The contribution of the kinetic energy is equal to

$$p_{\text{kin}} = C_{\text{profile}} \frac{1}{2} \rho (u_z)^2$$

(3.11)

where, $C_{\text{profile}}$ is a correction factor for the radial profile of the axial velocity inside the liquid. The total kinetic energy of a flow with a flat radial profile deviates from the kinetic energy of a flow with the same flow rate, the same average axial velocity, but with a radial profile of the axial velocity, which is not flat.
3.2.3 Viscous Stress

Above the previously discussed effect of the viscous surface stress, there are two other effects of viscosity that have to be taken into account. Figure 3.3 shows that in order to reach a balance of forces at the liquid-air interface, a tangential stress $\tau_\mu$ is needed. This tangential stress is equal to

$$\tau_\mu = \frac{3\mu (\partial u_z/\partial z) (dr/\partial z)}{1 + (dr/\partial z)^2}$$  \hspace{1cm} (3.12)

The second effect due to viscosity occurs in the bulk of the liquid. As already stated, the normal stress in the axial direction is equal to

$$\sigma_\mu = 2\mu \frac{\partial u_z}{\partial z}$$  \hspace{1cm} (3.13)

The assumption is made that the viscous shear stress $\mu \partial u_z/\partial r + \mu \partial u_z/\partial z$ in the bulk of the liquid does not play a role in the average acceleration of the liquid. The reason for this assumption is that this shear stress decelerates the liquid at the liquid surface but accelerates the liquid at the axial axis of the liquid cone. Thus, the shear stress transfers the tangential stress from the liquid surface into the bulk of the liquid. So, it is only of importance for the radial velocity profile in the liquid, but not for the average value of the velocity at that axial position. That is why this term can be removed from the Navier-Stokes equation for a one dimensional jet.

3.2.4 Polarization Stress

A material, which can be polarized in an electric field, experiences inside the material, due to that electric field, a normal stress. Following Landau and Lifshitz (1975) the force per unit volume in a material is equal to

$$\vec{f} = \vec{E} \nabla \epsilon_0 \epsilon_r \vec{E} + \frac{1}{2} (\epsilon_r - 1) \epsilon_0 \nabla \vec{E}^2$$  \hspace{1cm} (3.14)

where, $\vec{f}$ is a force per unit volume [N m$^{-1}$], $\vec{E}$ is the electric field strength [V m$^{-1}$]. The assumption is made that all free charge in the liquid is at the liquid-air interface. This means that in the bulk of the liquid there is no free charge, which yields

$$\nabla \epsilon_0 \epsilon_r \vec{E} = 0$$  \hspace{1cm} (3.15)
Using equations (3.14) and (3.15) in the liquid cone, results in a stress equal to

\[
\sigma_e = \frac{1}{2} ( \varepsilon_r - 1 ) \varepsilon_0 ( E_n^2 + E_t^2 )
\]  

(3.16)

where, \( E_t \) is the tangential electric field strength at the liquid surface [V m\(^{-1}\)].

3.2.5 Gravitation

The contribution of gravitation is equal to 

\[
p_n = \rho g z
\]  

(3.17)

3.2.6 Tangential Electric Stress

Free charge in an electric field experiences a force. Using the assumption that all free charge can be found at the liquid-air interface, a tangential electric stress is created in the liquid surface which is equal to

\[
\tau_{Et} = \sigma E_t
\]  

(3.18)

where, \( \sigma \) is surface charge [C m\(^{-2}\)].

3.2.7 Electric Field Calculations

In order to be able to solve equations (3.2) and (3.3), the electric field strengths, the surface charge and the radial velocity profile inside the liquid have to be known. A certain conical shape with a jet is used as a starting point. Figure 3.4 shows the electrode configuration and the boundary conditions used in the simulations. The electric fields inside and outside the liquid are numerically calculated using Gauss’ law

\[
\varepsilon_0 \varepsilon_r \int E_r dS = q_e
\]  

(3.19)

where, \( S \) is a surface [m\(^2\)], and \( q_e \) is the encapsulated charge [C]. The calculation method is an iterative finite volume method which balances the electric fluxes around each grid point until convergence.
has been reached.

3.2.8 The Current Balance at the Liquid Surface

The free charge at the liquid-air interface is calculated in the following way, Fernández de la Mora and Loscertales (1994). Charge transport can occur in two ways. Firstly, there is the conduction of charge because of the electric field inside the liquid. Secondly, there is the free charge convection at the liquid surface. When using the assumption that all free charge can be found at the liquid surface, a current balance can be made at the liquid-air interface. Figure 3.5 shows this current balance. The current due to charge convection is equal to

\[ I_v = 2 \pi r_s u_z \sigma \] (3.20)

where, \( u_z \) is the liquid velocity at the liquid-air interface in the axial direction. The current toward the liquid-air interface due to conduction is equal to

\[ I_k = K E_{n,\text{int}} S = K E_{n,\text{int}} 2\pi r_s ( 1 + (dr/dz)^2 )^{1/2} \Delta z \] (3.21)

3.2.9 The Axial Liquid Velocity at the Liquid Surface

In order to be able to calculate this current balance, the liquid velocity at the liquid-air interface has to be known. The following approximation has been used. At the liquid-air interface, the shear stress due to viscosity must be equal to the viscous shear stress that occurs when balancing the pressures at the liquid-air interface and the viscous normal stresses, equation (3.12), and the tangential electric shear stress, equation (3.18). This yields the following equation

\[ \mu \frac{\partial u_z}{\partial r} + \mu \frac{\partial u_r}{\partial z} = E_z \sigma + \frac{3 \mu (\partial u_z/\partial z) (dr/dz)}{1 + (dr/dz)^2} \] (3.22)
using equations (3.9) and (3.10) and using

\[ u_r = u_z \frac{dr}{dz} \]  
(3.23)

the following relation can be derived

\[ \mu \frac{\partial u_r}{\partial r} = E_i \sigma + \frac{\mu}{r_s} \frac{u_z (dr/dz)^2}{1 + (dr/dz)^2} - \frac{2(dr/dz)^2 - 4}{1 + (dr/dz)^2} \mu \frac{d^2r}{dz^2} \]  
(3.24)

Assuming that the radial pressure profile is flat, assuming that in the bulk of the liquid the influence of the \( \mu \partial u_r / \partial z \) can be neglected because of the continuity equation, and taking care that the average axial velocity meets the liquid flow rate supplied by the syringe pump, the radial axial-velocity profile can be estimated using

\[ \frac{\mu}{r} \frac{\partial \left( r \partial u_r / \partial r \right)}{\partial r} = \frac{\partial (\frac{1}{2} \rho u^2)}{\partial z} \]  
(3.25)

This equation is solved in the radial direction by means of an iterative numerical method. The boundary condition at the cone axis is equal to

\[ \frac{\partial u_r}{\partial r} = 0 \]  
(3.26)

The right-hand side of the equation for the present axial position is solved by using the profile obtained from the previous axial position.

The surface charge, the axial liquid velocity, and electric field strengths interact with each other, so these three calculations have to be solved simultaneously. After convergence of the iterations, the results are used to calculate a new cone shape that fulfills equation (3.2). This shape calculation is quite unstable, and requires an error minimization method. If the appropriate derivative \( dr_z/\partial z \) is chosen for the liquid-air interface at the nozzle, then the error can be reduced to zero. For the electric field calculations the jet was ended with a spherical shape. The radius of this sphere is equal to the jet radius for the last grid point on the jet. For this last grid point on the jet, the influence of \( dr_z/\partial z \) and \( d^2r_z/\partial z^2 \) on the jet radius is small. That is why this jet radius in the last grid point on the jet can be calculated using the values of \( dr_z/\partial z \) and \( d^2r_z/\partial z^2 \) of the neighboring jet grid point. The new cone shape will be the input for new electric field strength, surface charge, and liquid velocity profile calculations.
3.3 EXPERIMENTAL

Experiments and model calculations were conducted for 7 electrode configurations similar to the one shown in figure 3.4. Table 3.1 gives an overview of the electrode configurations used in the experiments and in the numerical model. The nozzles in these electrode configurations were cylindrically shaped. At the bottom side, the nozzle was closed with a flat surface. In the middle of this surface, there was a small hole of a few hundred micrometers. For nozzles with a small diameter, this nozzle shape might give experimental problems with the attachment of the liquid to the nozzle surface. This could result into destabilisation of the liquid cone. In the simulations, these problems cannot occur. The model requires a fixed position, where the liquid is attached to the nozzle. This cylindrical nozzle shape with a flat end defines clearly this position. So, the model can be used to investigate the influence of the nozzle shape outside the liquid, including the nozzle diameter. However, the influence of the nozzle shape on the liquid attachment to the nozzle, and the influence of this attachment on the cone shape, cannot be investigated.

<table>
<thead>
<tr>
<th>Electrode Configuration Number</th>
<th>d_{nozzle}</th>
<th>L</th>
<th>z_E</th>
<th>r_E Top Electrode</th>
<th>r_E Bottom Electrode</th>
</tr>
</thead>
<tbody>
<tr>
<td>I model</td>
<td>8</td>
<td>6</td>
<td>40</td>
<td>100</td>
<td>100</td>
</tr>
<tr>
<td>I experiment</td>
<td>8</td>
<td>6</td>
<td>40</td>
<td>100</td>
<td>100</td>
</tr>
<tr>
<td>II model</td>
<td>2</td>
<td>1.5</td>
<td>10</td>
<td>25</td>
<td>25</td>
</tr>
<tr>
<td>III model</td>
<td>2</td>
<td>6</td>
<td>14.5</td>
<td>25</td>
<td>25</td>
</tr>
<tr>
<td>IV model</td>
<td>1</td>
<td>6</td>
<td>14.5</td>
<td>25</td>
<td>25</td>
</tr>
<tr>
<td>IV experiment (G.C. 1997)</td>
<td>1</td>
<td>??</td>
<td>??</td>
<td>??</td>
<td>??</td>
</tr>
<tr>
<td>V model</td>
<td>8</td>
<td>6</td>
<td>127</td>
<td>0</td>
<td>200</td>
</tr>
<tr>
<td>V experiment</td>
<td>8</td>
<td>6</td>
<td>127</td>
<td>0</td>
<td>200</td>
</tr>
<tr>
<td>VI model</td>
<td>8</td>
<td>5.1</td>
<td>40</td>
<td>25</td>
<td>100</td>
</tr>
<tr>
<td>VI experiment</td>
<td>8</td>
<td>5.1</td>
<td>40</td>
<td>25</td>
<td>50</td>
</tr>
<tr>
<td>VII model</td>
<td>8</td>
<td>5.1</td>
<td>30</td>
<td>25</td>
<td>100</td>
</tr>
<tr>
<td>VII experiment</td>
<td>8</td>
<td>5.1</td>
<td>30</td>
<td>25</td>
<td>50</td>
</tr>
</tbody>
</table>

The measurements were performed at room temperature, which was between 20 and 25 °C. The liquids used were ethylene glycol technical grade, ethylene glycol chemical grade 99% purity.
and n-butanol chemical grade 99% purity. LiCl was sometimes added to enhance the conductivity. The Liquid conductivity was measured with a WTW Microprocessor conductivity meter LF96. The other liquid properties were taken from literature. Simulation results were also compared to experimental results presented by Gañán-Calvo et al. (1997). (G.C. 1997) refers in some tables and figures to this publication. Table 3.2 shows the liquid properties of the liquids used in the simulations, and the experiments.

**Table 3.2 Liquid properties.**

<table>
<thead>
<tr>
<th>Liquid</th>
<th>$\rho$ kg m$^{-3}$</th>
<th>$\mu$ Pas</th>
<th>$\gamma$ N m$^{-1}$</th>
<th>$\varepsilon_r$</th>
<th>$K$ $\mu$S m$^{-1}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ethylene Glycol Technical grade</td>
<td>1109</td>
<td>0.02</td>
<td>0.048</td>
<td>37</td>
<td>76</td>
</tr>
<tr>
<td>Ethylene Glycol 99% + LiCl</td>
<td>1109</td>
<td>0.0199</td>
<td>0.0477</td>
<td>38</td>
<td>21</td>
</tr>
<tr>
<td>Ethylene Glycol 99% + LiCl</td>
<td>1109</td>
<td>0.02</td>
<td>0.048</td>
<td>37</td>
<td>72</td>
</tr>
<tr>
<td>Ethylene Glycol 99% + LiCl</td>
<td>1109</td>
<td>0.02</td>
<td>0.048</td>
<td>37</td>
<td>186</td>
</tr>
<tr>
<td>n-Butanol 99%</td>
<td>810</td>
<td>0.00294</td>
<td>0.0246</td>
<td>17.8</td>
<td>15</td>
</tr>
<tr>
<td>n-Butanol 99%+ LiCl</td>
<td>810</td>
<td>0.00276</td>
<td>0.0252</td>
<td>17.3</td>
<td>101</td>
</tr>
<tr>
<td>n-Butanol 99% + LiCl</td>
<td>810</td>
<td>0.00276</td>
<td>0.0252</td>
<td>17.3</td>
<td>216</td>
</tr>
<tr>
<td>Methanol (G.C. 1997)</td>
<td>795</td>
<td>$0.59 \times 10^3$</td>
<td>0.021</td>
<td>33.6</td>
<td>85</td>
</tr>
<tr>
<td>Heptane +0.1% Stadis (G.C. 1997)</td>
<td>684</td>
<td>$0.39 \times 10^3$</td>
<td>0.021</td>
<td>1.9</td>
<td>0.77</td>
</tr>
<tr>
<td>Heptane +0.4% Stadis (G.C. 1997)</td>
<td>684</td>
<td>$0.39 \times 10^3$</td>
<td>0.021</td>
<td>1.9</td>
<td>1.9</td>
</tr>
<tr>
<td>Heptane +1.0% Stadis (G.C. 1997)</td>
<td>684</td>
<td>$0.39 \times 10^3$</td>
<td>0.021</td>
<td>1.9</td>
<td>4.5</td>
</tr>
</tbody>
</table>

In the experiments, the liquid was supplied by a syringe pump at a constant flow rate. The potential was applied by connecting a high voltage supply to the nozzle and top plate. The counter electrode was grounded through an oscilloscope. The accuracy of the current measurement was checked by comparing measurements with a Keithley 610B electrometer. The Current measured with the Keithley was about 3 percent lower than the current measured by the oscilloscope. The measured current fluctuated by about 5 nA in time due to instabilities in the liquid supply rate and due to air movement. After starting the experiment, the relatively large liquid cone needed a few minutes in order to reach steady state. Each measurement point consists of 400 measurements of the average current. This average current was measured in a period of 0.2 seconds. This method allows to average out the instabilities in the measurement signal, and allows to check the long time stability of the current. Before measuring the current coming from the liquid cone, the background current was measured. This background current is the current measured without a liquid cone present, but with the potential applied over the electrodes. This background current was about 8 nA for a potential difference of 20 kV, and depended on the potential difference. The fluctuations in this current were about 0.3 nA. The current coming from the liquid cone was determined by subtracting this background current from the measured current.
3.4. COMPARISON OF MODELING AND EXPERIMENTAL RESULTS

Figure 3.6 shows the shape calculated for a ethylene glycol cone. This result was calculated using the liquid properties of table 3.2, a conductivity of $69 \mu S \text{ m}^{-1}$, a liquid flow rate of $1.4 \times 10^9 \text{ m}^3\text{s}^{-1}$, a potential difference of $20 \text{ kV}$, and electrode configuration I. Figure 3.7 shows a picture of this ethylene glycol cone. The conductivity of this liquid cone was $73 \mu S \text{ m}^{-1}$. Simulations showed that this difference in conductivity has little influence on the cone shape. The cone shape obtained from the simulations and the real shape are very similar.

![Figure 3.6 The calculated cone shape.](image)

![Figure 3.7 The experimental cone shape](image)

The simulations yielded a cone length of 3.95 mm, where a cone length of 4.25 +/- 0.2 mm was measured. This cone length is defined as the distance between the nozzle tip and the position where the jet emerges from the cone. The position where the jet emerges, the cone apex, is defined as the position where the second derivative of the cone radius $d^2r_c/dz^2$ reaches a maximum. The same was done for a potential difference of 18 KV. In this case the simulated cone length was 4.57 mm and the measured cone length was 4.9 +/- 0.2 mm. The model underestimates the cone length by about 7%. Except a possible deviation of the surface tension from the literature value, the most likely explanation for this underestimation of the cone length is the neglect of space charge.

Normally, the model does not take into account the presence of the highly charged droplets in the gap between the cone and the counter electrode. However, the presence of these droplets reduces the electric field around the cone compared to the situation without charged droplets present. When the current through the liquid cone is known, together with the droplet velocity, and the radius of the spray as function of the z coordinate, then the model can calculate
the influence of space charge. The Particle Analysis System (Dantec) was used to measure the droplet size and velocity in electrode configuration V. This system is a single particle measurement system that uses the principle of Laser-Doppler to measure the droplet velocity. The intensity of the scattered light is used to measure the droplet size. In electrode configuration V, there was no plate above the nozzle. Ethylene glycol with a conductivity of 69 μS m\(^{-1}\) was sprayed at a flow rate of 1.4 \(10^9\) m\(^3\) s\(^{-1}\). The potential difference was 16 kV. From the results of these measurements, the influence of space charge could be calculated. It was found that the influence of space charge was almost the same as neglecting the space charge, and reducing the potential difference by 7%. Simulations showed, that the most important reduction in field occurs due to the droplets less than 10 mm from the cone apex. This means that the distance between the nozzle and counter electrode is only of importance, if the counter electrode is close to the nozzle. In electrode configurations I and V these distances are relatively large namely 34 and 121 mm. So the influence of space charge should be in both cases in the order of 7% of the potential difference. The measured cone length was compared to the cone length

**Figure 3.8** The current as function of flow rate for ethylene glycol.

**Figure 3.9** The current as function of flow rate for n-butanol.
simulated for electrode configuration I. It was found, that a cone simulated for a potential difference of 19 kV would give the same cone length as the experimental length for 20 kV. If this is due to space charge, then the space charge has an influence of 5% on the potential difference. This is in agreement with the 7% in electrode configuration V.

Figure 3.8 shows a comparison between the measured currents coming from a liquid cone of ethylene glycol in electrode configuration I, and the currents calculated for a potential difference of 20 kV. The small differences in conductivity can be accounted for by using the scaling laws of Gañán-Calvo et al. (1997). After this correction, the calculated currents were 5 percent larger than the values measured for technical grade ethylene glycol. If we compare the simulations to the measurements with 99% pure ethylene glycol + LiCl with a conductivity of 72 $\mu$S m$^{-1}$, the model predicted a 10 percent higher current than measured.

Figure 3.9 shows a comparison between the measured currents coming from a n-butanol cone in electrode configuration I, and the currents calculated for an potential difference of 15 kV.

Figures 3.10 and 3.11 show a comparison between the currents measured by Gañán-Calvo et al. (1997) coming from methanol and heptane cones in electrode configuration IV, and the calculated currents. In the simulation the potential difference was 4.5 kV, except for most of the methanol simulations. The electrode configuration in the experiments was not described.
completely. Only the nozzle diameter was given. So, the electrode configuration IV was assumed for the model. This is why, the comparison of the absolute values of the current between experiment and simulation is not of real interest. What is of interest are the relative results, when comparing different flow rates, different potentials, and different liquids with each other. Figure 3.10 shows that the current in the methanol experiments increase with the liquid flow rate in the same way as in the simulations. Table 3.3 shows a comparison between the model and experimental values for the other simulations.

**Table 3.3 A comparison of the current obtained in the simulations and experimental values for a potential difference of 4.5 kV.**

<table>
<thead>
<tr>
<th>Liquid</th>
<th>K</th>
<th>Q</th>
<th>I&lt;sub&gt;model&lt;/sub&gt;</th>
<th>I&lt;sub&gt;experiment&lt;/sub&gt;</th>
<th>I&lt;sub&gt;model&lt;/sub&gt; - I&lt;sub&gt;experiment&lt;/sub&gt;</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>µS m&lt;sup&gt;-1&lt;/sup&gt;</td>
<td>m&lt;sup&gt;3&lt;/sup&gt; s&lt;sup&gt;-1&lt;/sup&gt;</td>
<td>nA</td>
<td>nA</td>
<td></td>
</tr>
<tr>
<td>Methanol</td>
<td>85</td>
<td>0.7 × 10&lt;sup&gt;9&lt;/sup&gt;</td>
<td>82</td>
<td>97</td>
<td>-0.15</td>
</tr>
<tr>
<td>Heptane</td>
<td>4.5</td>
<td>1.2 × 10&lt;sup&gt;9&lt;/sup&gt;</td>
<td>42.3</td>
<td>42.3</td>
<td>0</td>
</tr>
<tr>
<td>Heptane</td>
<td>1.9</td>
<td>1.2 × 10&lt;sup&gt;9&lt;/sup&gt;</td>
<td>34.6</td>
<td>29.6</td>
<td>0.17</td>
</tr>
<tr>
<td>Heptane</td>
<td>0.77</td>
<td>1.2 × 10&lt;sup&gt;9&lt;/sup&gt;</td>
<td>28.9</td>
<td>23.3</td>
<td>0.24</td>
</tr>
<tr>
<td>Heptane</td>
<td>0.77</td>
<td>2.4 × 10&lt;sup&gt;9&lt;/sup&gt;</td>
<td>32.5</td>
<td>30.1</td>
<td>0.08</td>
</tr>
</tbody>
</table>

For methanol at a potential difference of 5.5 kV, the results of the model fit well to the current measured as function of flow rate. For a potential difference of 4.5 kV, the simulated current was below the measured current. For heptane at a potential difference of 4.5 kV, the results do not fit as well as before. The current simulated for this potential difference was expected to be 15% below the measured current. The main difference between methanol and heptane is the conductivity.

The ratio of the charge convection current over the conduction current plus the charge convection current increases from zero to one, when going from the cone base towards the tip of the jet, see also figure 3.28. When, considering the cone radius at a position where this ratio is equal to 0.5, then this cone radius is larger for a lower conducting liquid, than for a higher conducting liquid. A larger cone radius in this area yields, for the same viscous stresses, a larger ratio of the axial velocity at the liquid surface over the average axial liquid velocity. A larger ratio makes a larger error in the estimation of the liquid velocity at the liquid-air interface possible. This would explain the increasing error with decreasing conductivity of heptane. When considering the same position on the cone, then an increasing flow rate will also result in an increase in the jet diameter. This could yield a larger ratio of the velocity at the liquid-air interface over the average axial velocity. However, when the liquid properties are kept constant, then the simulations showed that the electrical tangential stress decreases with increasing flow rate. This results in a lower velocity ratio. Secondly, with increasing flow rate, the influence of the velocity pressure increases, equation (3.25). The last
result of table 3.3 indicates that these two effects are stronger than the increase in the velocity ratio due to an increased jet radius. The conclusion is that the deviations in table 3.3 are most likely caused by the estimation procedure of the axial velocity near the liquid surface. So, this procedure still needs some improvement.

3.5 EXPERIMENTAL VERIFICATION OF THE JET SHAPE.

The shape of the jet was checked by means of a High Speed Spray Imaging System (HSSIS) purchased from Oxford Laser. This system consists of a digital camera (KODAK) connected to a computer, which is equipped with a frame grabber. A long distance microscopic lens is fixed to the camera. The illumination of the object is done by an infra-red laser. A control box synchronizes the camera and the laser. Pictures can be made with an illumination time down to 0.5 μs. The separation between two following pictures can be down to 15 μs. The optical system allows to see particles of a few micrometers. However, perfect focusing on a moving object proved to be difficult. Experiments with polymer particles on a glass slide, proved that a picture, which was not perfectly focussed resulted in an overestimation of the droplet size by 4 μm. On the pictures, one pixel was 1 μm.

The measurements with ethylene glycol + LiCl with a conductivity of 21 μS m⁻¹ were done in electrode configuration VI. n-Butanol 99% with a conductivity of 15 μS m⁻¹ yielded better pictures in electrode configuration VII. In electrode configuration VI the n-butanol cone apex was not fixed at one position, but moved slowly around. That is why electrode configuration VII was used.

Figures 3.12 and 3.13 show the comparison between the measured jet shape and the calculated jet. The black shape is from the camera picture of the cone for ethylene glycol at flow rates of 2.78 \times 10^{-4} and 5.55 \times 10^{-3} m³ s⁻¹, and a potential difference of 17 kV. The conductivity was 21 μS m⁻¹. The white lines on the picture is the cone shape calculated by the model. As can be seen the model agrees very well with the measurements.

The jet diameter and the current were measured for both ethylene glycol and n-butanol. The current was again measured with the same oscilloscope as before, but the measurement time was a much shorter. The current was checked to be stable in time. Figure 3.14 shows the current as function of the liquid flow rate. The agreement between these calculations and the measurements is even better than the ones presented in the previous section. Two possible reasons could be found. First another conductivity meter was used, namely a YSI model 32, and secondly, the deviation between the results of the model and the measurements grows with increasing flow rate. Gañán-Calvo et al. (1997) introduced a dimensionless number, that can be used to estimate the minimum flow rate of a liquid sprayed in the Cone-Jet mode.

\[
\frac{QKp}{\varepsilon_0\gamma}
\]  

(3.27)
Figure 3.12 Liquid flow rate $2.78 \times 10^2 \text{ m}^3 \text{ s}^{-1}$. Figure 3.13 Liquid flow rate $5.55 \times 10^4 \text{ m}^3 \text{ s}^{-1}$.

Figure 3.14 The current as function of the flow rate.
The higher the conductivity the lower the minimum flow rate. In the previous section the conductivity of the liquids that were used was several factors higher than the ones used in this section. However, the used flow rates were not much lower, because of the lower flow rate limit of the syringe pump. As a result, the measurements in this section were considerably closer to the minimum flow rate than in the previous section. This could explain the smaller deviation found in this section.

The jet diameter was measured 0.6 mm from the cone apex. The cone apex was again defined as the position where \( d^2 r / dz^2 \) reaches its maximum. Figure 3.15 shows the jet diameter measured and calculated as function of the liquid flow rate. The measurements showed the same tendency as the simulations. However, the measured jet diameter was in the whole measurement range approximately 4 \( \mu \)m larger than the calculated value. Two reasons could be found for this. Firstly, the air friction is neglected in the model. When air friction is taken into account, then it would result in a slightly thicker jet. Secondly, the jet is moving in space. So it is very hard to get perfectly focused pictures. The size of the jet makes it also impossible to focus on the front and the back of the jet at the same time. The procedure of focussing on the jet was kept constant during all measurements. However, the experiment with the polymer droplets proved that the chosen way of focusing gave rise to an overestimation of the jet size by 4 \( \mu \)m. So, the jet diameters measured and calculated agree with each other within the measurement accuracy of the system.

6. COMPARISON TO THE PRESENT SCALING LAWS

The current as function of the flow rate for liquids with flat radial profiles of the axial velocity in the jet are often fitted using the following relation, see equation (3.1)

\[
I = b_i Q^{a_i}
\]  

(3.28)

where, \( a_i \) , and \( b_i \) are constants. In literature the value of \( a_i \), based on dimension analysis, is taken equal to 0.5. Table 3.4 shows the values of \( a_i \) calculated from the experiments, and calculated from the simulations.
Table 3.4 Comparison of the value of $a_i$ between experimental values and the value calculated by the model.

<table>
<thead>
<tr>
<th>Liquid</th>
<th>K  $\mu S \cdot m^{-1}$</th>
<th>Electrode Configuration</th>
<th>$a_i$ Experiment</th>
<th>$a_i$ Model</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ethylene Glycol Technical grade</td>
<td>76</td>
<td>I</td>
<td>0.490</td>
<td>0.49</td>
</tr>
<tr>
<td>Ethylene Glycol 99% +LiCl</td>
<td>21</td>
<td>VI</td>
<td>0.531</td>
<td>0.58</td>
</tr>
<tr>
<td>Ethylene Glycol 99% +LiCl</td>
<td>72</td>
<td>I</td>
<td>0.492</td>
<td>0.49</td>
</tr>
<tr>
<td>Ethylene Glycol 99% +LiCl</td>
<td>186</td>
<td>I</td>
<td>0.479</td>
<td></td>
</tr>
<tr>
<td>n-Butanol 99%</td>
<td>15</td>
<td>VII</td>
<td>0.489</td>
<td>0.534</td>
</tr>
<tr>
<td>n-Butanol 99% +LiCl</td>
<td>101</td>
<td>I</td>
<td>0.425</td>
<td>0.462</td>
</tr>
<tr>
<td>n-Butanol 99% + LiCl</td>
<td>216</td>
<td>I</td>
<td>0.40</td>
<td>0.428</td>
</tr>
<tr>
<td>Methanol</td>
<td>85</td>
<td>IV</td>
<td>0.545</td>
<td>0.576</td>
</tr>
</tbody>
</table>

The results in this table show that the model predicts the value of $a_i$ well. However, the value of $a_i$ is not constant 0.5. It was found that the value of $a_i$ depends on the average value of the dimensionless number given by Gañán-Calvo et al. (1997), equation (3.27). The liquid flow rates were fixed by the syringe pump, so a higher conductivity means a higher value of this dimensionless number. Both model and experiments show a lower value of $a_i$ for a higher value of this dimensionless number.

The higher value of $a_i$ for methanol is maybe also a result of the nozzle diameter. In this experiment the nozzle diameter was only 1 mm compared to the 8 mm nozzle for the other experiments. A smaller nozzle influences the electric field, and consequently can have some influence on the current through the liquid cone.

Figure 3.16 shows the calculated influence of the conductivity on the jet radius, and the current in electrode configuration I for ethylene glycol at a liquid flow rate of $1.4 \times 10^{-9}$ m$^3$ s$^{-1}$ at a potential difference of 20 kV. The jet radius was again determined by taking a fixed distance from the cone apex, see section 3.5. In order to check the relations of equation (3.1), the following equations can be fitted to the simulated values

\[ I = b_{t_k} K^{a_{t_k}} \]
\[ d_{jet} = b_{jet,k} K^{a_{jet,k}} \]  

(3.29)
where, \( d_{\text{jet}} \) is jet diameter [m], \( a_{iK}, b_{iK}, a_{\text{jet},K}, \) and \( b_{\text{jet},K} \) are constants. This yielded \( a_{\text{jet},K} = -0.136 \) and \( a_{iK} = 0.538 \).

Table 3.5 shows the results of fitting the following relation to values calculated with the model and to experimental values

\[
d_{\text{jet}} = b_{\text{jet}}Q^{a_{\text{jet}}} + c_i
\]  

(3.30)

where, \( b_{\text{jet}} \) and \( a_{\text{jet}} \) are constants. In those cases where \( c_i \) is equal to zero, the fit was done keeping this value equal to zero.

**Table 3.5 Comparison of the value of \( a_{\text{jet}} \) and \( c_i \) between experimental values and the values calculated by the model.**

<table>
<thead>
<tr>
<th>Liquid</th>
<th>electrode configuration</th>
<th>distance from cone apex [mm]</th>
<th>( a_{\text{jet}} ) experiment</th>
<th>( c_i ) experiment</th>
<th>( a_{\text{jet}} ) model</th>
<th>( c_i ) model</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ethylene Glycol K=69 ( \mu )S m(^{-1})</td>
<td>I</td>
<td>1.0</td>
<td></td>
<td>0.71</td>
<td>0</td>
<td></td>
</tr>
<tr>
<td>Ethylene Glycol K=69 ( \mu )S m(^{-1})</td>
<td>I</td>
<td>2.3</td>
<td></td>
<td>0.64</td>
<td>0</td>
<td></td>
</tr>
<tr>
<td>Ethylene Glycol K=21 ( \mu )S m(^{-1})</td>
<td>VI</td>
<td>0.6</td>
<td>0.51</td>
<td>0</td>
<td>0.72</td>
<td>0</td>
</tr>
<tr>
<td>Ethylene Glycol K=21 ( \mu )S m(^{-1})</td>
<td>VI</td>
<td>0.6</td>
<td>1.10</td>
<td>8.0</td>
<td>0.91</td>
<td>3.5</td>
</tr>
<tr>
<td>n-Butanol K=15 ( \mu )S m(^{-1})</td>
<td>VII</td>
<td>0.6</td>
<td>0.59</td>
<td>0</td>
<td>0.71</td>
<td>0</td>
</tr>
<tr>
<td>n-Butanol K=15 ( \mu )S m(^{-1})</td>
<td>VII</td>
<td>0.6</td>
<td>0.94</td>
<td>5.6</td>
<td>0.86</td>
<td>2.5</td>
</tr>
</tbody>
</table>

The results in Table 3.5 show that, when fitting equation (3.30) using the constant \( c_i \), then \( c_i \) is 5.5 and 3 \( \mu \)m larger than the value calculated for the model. This difference is likely the result of the focussing problem of the camera, as discussed before.

The values obtained of \( a_i \) and \( a_{iK} \) in this section agree quite well with the present current scaling laws of equation (3.1). However, the values obtained of \( a_{\text{jet}} \) and \( a_{\text{jet},K} \) differ strongly from the droplet size scaling laws of equation (3.1). Two reasons could be found for this deviation, which are both related to the fact that the scaling law is developed for the droplet size, where \( a_{\text{jet}} \) is calculated for the liquid jet. Firstly, the jet is measured at a fixed distance from the cone apex. The length of a jet, before it breaks-up into droplets, depends on the liquid flow rate, and on the charge on the jet. The higher the conductivity, the higher the charge, the shorter the jet. The
higher the flow rate, the bigger jet, the longer the jet. The jet radius decreases with increasing distance from the cone apex. So, the longer the jet, the smaller the jet radius. If this effect is taken into account, then it would result in a lower value of \( a_{jet} \) and \( a_{pl,k} \). This would bring them closer to the scaling laws for droplets. However, this cannot explain the large difference between the results presented here and the scaling laws. First experimental results, also showed that the ratio of the droplet diameter over the jet diameter slightly depends on the liquid flow rate. Whether this effect could explain the remaining difference between the scaling laws and these results, will be discussed in chapters 6 and 7.

### 3.7 SIMULATION RESULTS

Figure 3.17 shows the influence of the potential on the cone shape for ethylene glycol under the same conditions as for figures 3.6 and 3.7. Figure 3.18 shows the influence of the potential on the current for four different electrode configurations. The value of \( V_0 \) is the potential that gives in the simulations for the electrode configuration a current of 150 nA for ethylene glycol with a conductivity of 69 \( \mu \)Sm\(^{-1}\), and at a flow rate of \( 1.4 \times 10^9 \) m\(^3\) s\(^{-1}\). The figure shows that the nozzle diameter has a strong influence on dependency of the current on the applied potential. This was confirmed by experiments. In case of the 8 mm nozzle, the influence of the potential on the current was really small, because in experiments it could not be detected. The influence was smaller than the measurement accuracy, 5 percent. The relation between current and potential can be described by

\[
\frac{I}{I_0} = (V/V_o)^n
\]

\( \text{(3.31)} \)

**Figure 3.17** The liquid cone shape as function of the potential.

**Figure 3.18** The current as function of the normalized potential.
Gañán-Calvo et al. (1997) showed a figure for electrode configuration IV, where the current depended on the potential. From this figure a value of $a_v$ could be estimated which ranged from 0.4 to 0.7, while in the simulations this value is equal to 0.6. Figure 3.18 also shows that the length of the nozzle has some influence on the way the current depends on the potential.

Since, the cone length depends on the potential, the current can be plotted as function of the cone length. Experiments with the 8 mm nozzle showed that a cone length equal to the nozzle radius yields a cone which seems to give a stable spray. In pictures of other nozzles, the cone length is often in the order of the nozzle radius. That is why the cone length is normalized by dividing through the nozzle radius. Figure 3.19 shows the current as function of this normalized cone length. The figure shows that the nozzle length L does not have an influence on the current. So, this figure indicates that the nozzle length L influences the dependency of the current on the potential. However, this effect is compensated by its influence on the cone length. So, for equal liquid cone lengths, the nozzle length does not have much influence on the current. For a normalized cone length equal to one, the current for the 1 mm nozzle is 15% higher than for the 8 mm nozzle. This means that scaling laws that do not take the nozzle diameter into account can never be very accurate.

**Figure 3.19** The current as function of the normalized cone length.

**Figure 3.20** The normalized axial velocity at the liquid surface as function of the cone radius for different values of the viscosity.

As already concluded before, the estimation of the axial velocity in the model needs some improvement. In spite of this, the model is already able to show the trends with respect to the influence of the radial profile of the axial velocity in the jet. For a liquid with the properties of ethylene glycol, electrode configuration I, a conductivity of 69 $\mu$S m$^{-1}$, at a liquid flow rate of $1.4 \times 10^9$ m$^3$ s$^{-1}$, and a potential difference of 20 kV, the viscosity was varied. Figure 3.20 shows the axial velocity at the liquid-air interface, which is normalized by the average velocity at that
axial position. This normalized velocity is given as function of the cone radius. When going down from the cone base towards the jet, the radius of the cone decreases. The smaller this radius, the larger the viscous stresses. That is why, the axial liquid velocity at the liquid-air interface for a large cone radius can be much larger than the average velocity. While in the jet the profile is almost flat. The normalized axial velocity is sometimes lower than one, because in those areas the tangential viscous stress is larger than the tangential electric stress. The results in section 3.4 indicated that the model overestimates the ratio of the axial liquid velocity at the liquid surface over the average axial liquid velocity. This should be kept in mind when looking at Figure 3.21, which shows the influence of the viscosity on the calculated current.

The following figures are all showing results for ethylene glycol in electrode configuration I, at a liquid flow rate of $1.4 \times 10^9 \text{ m}^3 \text{s}^{-1}$, a conductivity of $69 \mu \text{S m}^{-1}$ and a potential difference of $20 \text{ kV}$. Figure 3.22 shows that near the base of the cone the normal electric stress, and the surface tension stress are the dominant factors in the one dimensional Navier-Stokes equation, equation (3.2). The figure also shows that the tangential electric stress term and the kinetic energy term become dominant when going from the cone into the jet. In the figure, the tangential stress is plotted as the result of a derivation to the jet radius, and an integration over the axial distance from the nozzle, or cone base, see equation (3.2). This change in dominant factors must lead to a decrease in the value of $dr/\text{dz}$, because the surface tension stress depends on the cone radius to the power -1, equation (3.4), while the kinetic energy depends on the cone radius to the power -4, see equations (3.9) and (3.11).

![Figure 3.21 The current as function of the viscosity.](image1)

![Figure 3.22 The kinetic energy, surface tension, normal electric stress and tangential electric stress as functions of the axial distance from the nozzle.](image2)
Figure 3.23 shows the static liquid pressure in the cone, equation (3.3), minus the pressure of the surrounding air. The figure shows that the pressure in most of the cone is higher than the surrounding air. There are two exceptions namely near the nozzle, which is not shown, and near the position where the jet is formed.

Figure 3.24 shows the normalized stresses which occur in the liquid-air interface. The normalization was done by dividing the stresses by $\gamma / r_c$. The strange bump in the curves near the cone apex are a result of the normalization. The curves, which are not normalized are smooth.

![Figure 3.23 The pressure difference between liquid and surrounding air.](image1)

![Figure 3.24 The stresses in the liquid surface.](image2)

Figure 3.25 shows the normal electric field strength as function of the axial distance from the nozzle. The maximum field strength is reached where the jet emerges.

Figure 3.26 shows the tangential electric field strength as function of the axial distance from the nozzle.

Figure 3.27 shows that the ratio of the free charge at the air-liquid interface over the free charge plus the polarization charge is in most of the cone equal to one. This means that the normal electric field inside the liquid is much lower than outside the liquid. The only exception is again the area where the jet emerges.

Figure 3.28 shows the charge convection current and the total current. At the base of the liquid cone, the conduction current is dominant. In the jet, the charge convection current is dominant. Only at the position where the jet emerges from the cone, both currents are of the same order of magnitude. Figures 3.26 to 3.28 clearly show that the liquid cone can be considered as a conductor until the radius of the cone becomes small, and the limited conductivity of the liquid starts to play an important role.
Figure 3.25 The normal electric field strength in the air at the liquid-air interface.

Figure 3.26 The tangential electric field strength at the liquid-air interface.

Figure 3.27 The ratio of the free surface charge over the free surface charge + the polarization charge.

Figure 3.28 The charge convection at the liquid-air interface.

3.8 CONCLUSIONS

Electrohydrodynamic atomization in the Cone Jet mode can be modeled by means of a physical model without any fitting parameters. Such a model should consist of at least three parts. The
Cone-Shape model is the first part, and is able to calculate the shape of the liquid cone and jet, the electric fields in and outside the cone, and the surface charge density on the cone and jet. The model also estimates the liquid velocity at the liquid surface. From these results the current through the liquid cone can be calculated. The model allows further investigation of electrical discharges of the air surrounding the cone, it gives more insight in the phenomena itself, which makes it possible to come to better current scaling laws, and the influence of electrode configurations can be investigated much more easily. The second part of the physical model describing the Cone-Jet mode should describe the jet breaking up into droplets. The results of the Cone-Shape model make it possible to do this, because the shape of the jet, the velocity of the jet and the surface charge on the jet can be calculated. The third part of the physical model describing the Cone-Jet mode should describe the movement of droplets in the electric field after production. The Cone-Shape model allows the calculation of the electric field in which the highly charged droplets are moving.

The following main conclusions from the Cone-Shape model and the comparison to experimental results are here summarized:

1. The results of the Cone-Shape model fit well with experimental values.
   a. The calculated liquid cone shape is very similar to the experimental shape.
   b. The calculated cone length is about 7 percent shorter than the experimental result. This difference can be explained by the neglect of the space charge in the model.
   c. The currents calculated with the Cone-Shape model differ for liquids with a flat radial profile of the axial liquid velocity in the jet, less than 10 percent from the experimental values. The closer the liquid flow rate to the minimum flow rate, the smaller the deviations between experiment and model.
   d. The dependence of the current on the potential in the simulations agrees with experimental results.
   e. The dependence of the jet diameter on the liquid flow rate in the simulations agrees with the experiments within the measurement accuracy of 4 μm.
   f. The dependence of the current on the liquid flow rate in the simulations agrees with the experimental results.

2. The model needs some improvement with respect to the calculation of the liquid velocity near the liquid surface for liquids where the radial profile of the axial liquid velocity is not flat.

3. With respect to current as function of the liquid flow rate, the results of the Cone-Shape model are in good agreement with relations obtained by fitting to experimental results. However the value of the power αl is not constant but depends on the liquid flow rate, and most likely on the nozzle diameter. With respect to current as function of the conductivity, the results of the Cone-Shape model are in good agreement with the present scaling laws.

4. With respect to the jet radius as function of the liquid flow rate and as function of the conductivity, the results of the Cone-Shape model seem to show a different result compared to the scaling laws. However, the scaling laws are obtained by fitting to experimental droplet sizes. Two reasons could be found to explain this difference. Both reasons are linked to the fact that the
new results consider the jet diameter, where the scaling laws consider the droplet size. The first reason had to do with the fact that the jet length is not constant but depends on the flow rate, and the conductivity of the liquid. In the comparison, the jet length was taken constant. However, the real jet length depends on the liquid flow rate and the conductivity. The other reason had to do with the observation that the ratio of the droplet radius over the jet radius slightly depends on the flow rate. The validity of the droplet size scaling laws will be discussed further in chapter 6 and 7.

5. The current for a liquid cone with a cone length equal to the nozzle radius depends on the nozzle diameter. The simulations showed that a nozzle with a diameter of 1 mm yields a 15% higher current than a 8 mm nozzle. Thus, current scaling laws that do not take the nozzle diameter into account can never be very accurate. For the 8 mm nozzle the current is almost independent of the potential difference, where for the 1 mm nozzle there is a much stronger relation.

6. The model neglects the space charge due to the produced droplets. This neglect has an influence on the electric field around the cone. This influence can be compensated by reducing the potential in the model by 5 to 7 percent. This means that the space charge has only a small influence on the spraying process.
CHAPTER 4

ELECTRICAL DISCHARGES IN THE AIR SURROUNDING THE LIQUID CONE

This chapter describes the influence of electrical discharges in the air on the spraying process in the Cone-Jet mode. The occurrence of electrical discharges in the air prevents some liquids from being sprayed in a stable Cone-Jet mode. Background theory will be given about electrical discharges. Results presented in literature will be discussed, and simulation results of the Cone-Shape model are presented. The model has been used to investigate the position on the cone, where discharges are likely to occur. It has also been used to investigate the influence of the liquid flow rate, the liquid conductivity, and the surface tension, on the electrical discharge probability.

4.1 BACKGROUND THEORY

In this section a qualitative description of electrical discharges in gases will be given. The basis of this description is taken from White (1963), Loeb and Meek (1941), and Kuffel and Zaenogl (1984). A gas contains a very small amount of ions and free electrons, and can be considered as a good insulator. However, when the electric field strength in the gas is increased above a certain limit, suddenly large currents can start to flow, and the air becomes conducting. This transition point is called the electric breakdown of the air.

Free electrons are rare in air but can be produced by collisions between electrons and molecules, by collisions between ions and molecules, by photo-ionization of gas molecules, by photo electric emission of electrons from the cathode, and by ionization of metastable gas atoms. In an electric field, free electrons are accelerated until they collide with a gas molecule. During such a collision several things can happen. If the electron energy is very low at the moment of collision, then an elastic collision can occur, see Figure 4.1.a. Depending on the electron affinity of the molecule, it is also possible that the electron is caught by the molecule, and a negative ion is formed. The energy state of this new ion is too high to be stable. However, if the ion collides soon with another molecule, then it can transfer its excess energy to this molecule, see Figure 4.1.b. When no collision with another molecule takes place, then the ion will dissociate again into a neutral molecule and an electron, see Figure 4.1.c. If the electron energy at the moment of
collision is higher than in the previous cases, then the gas molecule is excited to a higher energy state, or the electron ionizes the molecule. When a molecule is ionized, then an extra electron, and a positive ion, are formed, see Figure 4.1.d. When the molecule is excited to a higher energy state, then the molecule can return to its normal state under expulsion of a quantum of photon energy, which can be often detected as a blue light, see Figure 4.1.e. An excited molecule can be ionized more easily than a molecule with a normal energy state. So, if this higher energy state is metastable, then there is sufficient time for a photon or a second collision with an electron to ionize the molecule, see Figures 4.1.f, and 4.1.g. It is also possible that an electron collides with a positive ion. In that case, recombination to a neutral molecule can occur under expulsion of a photon, see Figure 4.1.h.

What will happen during a collision depends on the electron energy, the ionization energy, and the electron affinity of the molecule, and on the way the electron collides with the molecule. At low electron energies and high electron affinities, negative ions will be formed. However, the probability of exciting or ionizing the molecule rises with increasing electron energy. With increasing average of the electron energy at the moment of collision, the number of electrons after a number of collisions can become larger than the number of electrons before these collisions. In that case, an avalanche of electrons is formed and an electrical discharge occurs.

The energy of an electron colliding with a molecule does not only depend on the electric field strength, but also on the mean free path between the molecules. The longer the mean free path, the lower the required electric field strength to reach an electron energy that is required to induce an electrical discharge. However, also fewer collisions will take place when an electron travels a certain distance. A larger mean free path means a lower relative density of the gas. The relative density of the gas is defined as

\[ \text{Relative Density} = \frac{\text{Number of Molecules}}{\text{Volume}} \]
\[ \delta = \frac{T_0 \ p}{T \ p_0} \]  

(4.1)

where, \( p_0 \) is the reference pressure [Pa], and \( T_0 \) is the reference temperature [K].

![Diagram of positive ions, photons, and electric field](image)

**Figure 4.2 Schematic representation of the formation of a streamer.**

In an homogeneous electric field, the electron avalanches or electrical discharge can easily grow into a spark. The avalanche produces a large amount of charge in the air. The high concentration of electrons at the front of the avalanche enhances the electric field strength. Combined with an exponentially growing number of electrons, this effect makes that the avalanche will grow faster and faster. When the avalanche reaches the electrode, the current density has increased enormously. Since, the mobility of the electrons can be a factor \( 10^6 \) larger, than the mobility of the positive ions, the electrons will soon be collected on the electrodes. This leaves an excess of positive charge in the gap between the electrodes. Especially, the extremely high space charge near the anode enhances the electric field between the cathode, and this positive space charge. The photons emitted by the avalanche trigger new avalanches, and the enhanced field strength makes it more easy for new avalanches to grow. The incoming avalanches are attracted by the positive space charge, and a plasma is formed of positive ions and incoming electrons. This process is called a positive streamer. Figure 4.2 shows a schematic representation of the formation of a streamer. Due to the continuously produced avalanches, the streamer grows toward the cathode. When the streamer connects the two electrodes, the spark is formed. The current in this spark is in most cases too high to be maintained by the system. As a result, the electric field between the electrodes can not be maintained. Due to this lower electric field strength, new discharges cannot be formed until the system has recovered from the spark. For a uniform electrode gap, the breakdown electric field strength at which sparking occurs, can be calculated with Paschen’s law.
\[ E_b = k_1\delta + k_2\left(\frac{\delta}{d_g}\right)^{1/2} \] (4.2)

where, \( E_b \) is the breakdown electric field strength [V m\(^{-1}\)], \( d_g \) is the gap distance [m]. \( \delta \) can be calculated with equation (4.1) with \( T_0 \) is 293 K, and \( p_0 \) is 101325 Pa. The constants \( k_1 \) and \( k_2 \) are for air equal to \( 24.22 \times 10^5 \) V m\(^{-1}\) and \( 6.08 \times 10^5 \) V m\(^{1/2}\). These values are taken from Bruce (1953). The first term of this equation represents the electric field strength that gives on average one electron after each collision between an electron and a molecule. If the gap distance is limited, then the avalanche has less distance to grow. So the number of collisions in the avalanche is small before it reaches the electrode, and only low current densities will be reached. Lower current densities generate less photons and less positive ions. These photons are necessary for the generation of free electrons, which can trigger new avalanches. The lower the current density, the lower the probability of the creation of a new free electron. Also, the produced positive space charge is much lower. Thus, the electric field is also less enhanced, and no streamer will be formed. In order to get a breakdown of the air, a higher current density in the avalanches is needed. This can be reached by increasing the electric field strength. This is represented in the second term of Paschen’s law.

![Diagram of Electrical Discharges in the Air Surrounding the Liquid Cone](image)

**Figure 4.3 Schematic representation of a negative discharge in a divergent field.**

In case of a divergent electric field, the avalanches grow only near the electrode with the highest electric field strength. Apart from the local distortion of the electric field strength very close to the avalanche, also another distortion of this field occurs. If the electric field strength near the discharge electrode is just above the breakdown electric field strength, then the avalanches will start to grow. In a case with the lowest potential on the discharge electrode, called a negative discharge, the electrons are moving away from this electrode. They will reach the area with a lower electric field strength. When the gas has an electron affinity, then the electrons will form negative ions in this area. Figure 4.3 shows a schematic representation of a negative discharge. Because, it takes more time for the negative ions to reach the grounded
counter electrode, than it takes the positive ions to reach the discharge electrode, a space charge occurs. This space charge will reduce the electric field strength near the discharge electrode to values below the breakdown field strength. This prevents new avalanches to be formed. When the ions are collected, or have moved far away from the discharge electrode, then the electric field increases again, and new avalanches can be formed. In case of the highest positive potential on the discharge electrode, called a positive discharge, the production of charge stops when the electron avalanche reaches the discharge electrode. In that case only the positive ions are left in the gap. Again their charge will reduce the electric field near the discharge electrode, and will prohibit the formation of new avalanches until they have moved far away from this electrode. Figure 4.4 shows a schematic representation of a positive discharge.

![Schematic representation of a positive discharge in a divergent field.](image)

**Figure 4.4** Schematic representation of a positive discharge in a divergent field.

In order to trigger new avalanches, electrons are needed. In case of a negative discharge, the free electrons come mainly from the surface of the discharge electrode. The electrode molecules are ionized by impacting positive ions and by photo-ionization. The photons are emitted by the excited gas molecules returning to their basic energy state. Also, the photo-ionization of gas molecules can contribute to this electron production. In case of a positive discharge, the free electrons can only come from the surrounding gas. They are mainly produced by photo-ionization of the gas molecules. In general, the ionization of the electrode molecules is more effective than the photo-ionisation of the gas. As a result, a positive discharge needs a higher current density in the avalanche to generate a new free electron. A higher current density requires a higher electric field strength. That is why, the onset electric field strength for negative discharges is lower than for positive discharges.

The production process of the triggering electron is much less effective, than the production process where electrons ionize molecules. However, for each avalanche only one triggering electron is needed. So, even these low effective processes can supply a sufficient number of free electrons. When on average each avalanche generates one new triggering electron, then a pseudo steady state is reached.

Because of the coincidence of the production of new free electrons that trigger
avalanches, because of the short period of charge production in the avalanche, and because of
the reduction of the electric field directly after the occurrence of an avalanche, the resulting
current pulses are very irregular in time and in intensity. If the electric field strength is increased,
than the number and the intensity of the current pulses increase.

Above a certain field strength, the current production, and the emitted blue glow, become
more regular. This electrical discharge is called a corona discharge or a glow mode discharge.
The current is more regular, because the number of avalanches is quite large, and the intensity
is related to the previously produced space charge. The higher the space charge, the lower the
electric field, the lower the intensity of new avalanches. Since the number of avalanches is large,
a relatively large avalanche can be compensated by a number of smaller ones. Also, the
avalanches are able to generate a sufficient number of new triggering electrons. This results in
a more regular current.

If the electric field strength is increased even further, then in case of a negative discharge,
it becomes possible for avalanches to cross the area of low electric field strength. The electric
field strength created near the front of the avalanche is strong enough to keep the avalanche
growing in the area of low electric field strength. Whether an avalanche can cross the area of low
electric field strength, depends on the concentration of charge in the avalanche. This
concentration depends on stochastic processes like, where the avalanche started, and whether in
the beginning many new electrons are formed or only a few. So, only a few avalanches can cross
the low field area. As a result, the current becomes pulsating again. The production of sparks is
not very easy, because the electric field at the anode is too low to generate easily new avalanches
in the area close to the anode. As discussed before these new avalanches are needed for the
formation of streamers. The electric field has to be increased further in order to get sparks. In
a positive discharge, sparking is more easy. The avalanche reaches the discharge electrode. The
electric field strength in this region is large. So, it is more easy to generate new avalanches in this
region, which are necessary for the formation of streamers. At a certain point, the streamer can
cross the area of low electric field strength, and a spark occurs.

The required strength of the electric field to create an electrical discharge depends on the
size of the area in which the avalanche can be formed. Due to the mean free path in the gas, an
electron can have on average only a fixed number of collisions while traveling through the area
of high electric field strength. The shorter this length, the lower the number of collisions. So,
only high currents can be reached, if the probability of electron production is high. That is why,
the required electric field strength increases with a decreasing size of the area of high electric
field strength. In a divergent electric field, the extend of this area depends on the curvature of the
electrodes. For wires, the radius is one of the most important parameters. The best known
formula to calculate the discharge onset electric field strength for a wire is given by Peek (1929),

\[ E_b = 3.1 \times 10^6 \delta \left( 1 + \frac{0.0308}{(r_w \delta)^{1/2}} \right) \]  (4.3)
where, \( r_w \) is the wire radius [m], \( \delta \) is calculated according to equation (4.1) with \( T_0 \) is 298 K, and \( p_0 \) is 101325 Pa. The constants are for a negative discharge in air. Rousse (1947) investigated thinner wires and found the following relations.

\[
E_b = 3 \times 10^6 \left( 1 + \frac{0.03}{r_w^{1/2}} \right) \quad \text{for} \quad r_w > 100 \mu m \quad (4.4)
\]

\[
E_b = 6.27 \times 10^6 + \frac{5502}{r_w^{0.75}} \quad \text{for} \quad 15 \mu m < r_w < 100 \mu m \quad (4.5)
\]

### 4.2 Electrical Discharges and the Liquid Cone

Whether electrical discharges occur around the liquid cone, depends on the radius of the jet, and on the electric field strength needed to create this jet. Especially for liquids with a relatively high surface tension, electrical discharges can easily occur. Smith (1986) found experimentally that a liquid with a surface tension higher than 0.05 N m\(^{-1}\) could not be sprayed in air.

If the liquid droplet is transformed into a conical shape, the electric field around the cone and jet increases. If the electric field in the air at the liquid surface is above the breakdown field strength for such a configuration, then an electrical discharge will occur. What will happen with the liquid cone depends on the intensity of the discharge. If the intensity is much smaller than the current through the cone, then the discharge might slightly influence the cone, but the droplet production process remains largely undisturbed. If the intensity of the discharge current is equal or higher than the current through the cone then three effects can take place. First, the extra produced space charge reduces the electric field near the cone. As a result, the surface tension can overcome the electric stress, and the liquid cone relaxes to a droplet shape. Increasing the potential has not much use, since higher electric fields will result in higher discharge currents, Hayati et al. (1987). These currents produce a higher

![Liquid Cone Diagram](image)

**Figure 4.5** Electrical discharges around the liquid cone.
space charge. This increase in space charge reduces the electric field near the liquid surface to values close to the initial value. Again surface tension can overcome the electric stresses. Only in a case with a constant discharge current, like in a corona discharge, it is possible to have a stable Cone-Jet mode, and a corona discharge at the same time. Tang and Gomez (1995), and Borra et al.(1998). However, due to the currents caused by the corona, the behavior of such a Cone-Jet mode will be different, from the Cone-Jet mode without electrical discharges.

The second mechanism of destabilisation of the cone is the pulsating character of many currents produced by electrical discharges. A pulsating current also means pulsating electric fields. This results in pulsating stresses, and destabilisation of the cone, Hayati et al.(1987), Cloupeau and Prunet-Foch (1989), Cloupeau (1994a), Borra et al.(1996).

The third mechanism is the discharge current reaching the liquid cone. As a result of this current, the charge distribution at the liquid surface is disturbed. A lower surface charge means lower electrical stresses, and again surface tension can become dominant over the electric stresses.

Electrical discharges can be prevented in at least five ways. As discussed in the previous section, the intensity of the discharge depends on the polarity of the discharge. In general, negative discharges require a lower electric field strength, so in a few cases it is better to switch from negative to positive polarity. The most drastic way to reduce discharges is to spray in a vacuum. If the mean free path becomes extremely long, then the number of collisions between an electron and a molecule is too low to create a detectable discharge current. The third method is to increase the pressure of the gas. In that way the mean free path is reduced. This means that the electron cannot acquire a large amount of energy between two collisions. A lower electron energy means a lower probability for the formation of a positive ion and an extra electron. The fourth way is to change the gas. Molecules with a high electron affinity form more easily negative ions. With increasing electron affinities, the electric field strength required to cause electrical discharges increases. In air, the oxygen has a high ion affinity. If pure nitrogen is used, a lower electric field strength is required to create discharges. SF₆ has an extremely high electron affinity, and much higher electric fields are necessary to create discharges. Tang and Gomez (1994) used a gas sheath of CO₂ around a water cone. They showed, that the suppression of discharges made atomization of water in the Cone-Jet mode possible. Tang and Gomez (1995) also used SF₆, to control the discharges around a water jet. The spraying process was stabilized by bringing the discharges around the jet in the glow mode. The fifth way is changing the electrode configuration. Borra et al.(1996) included an extra ring close to the nozzle, and applied a potential to this ring. In this way, electrical discharges in the air around an ethylene glycol cone could be suppressed. Borra et al.(1998) shows that a larger nozzle diameter results in a lower divergence of the electric field. Since, the electric fields at the liquid surface are almost independent of the nozzle diameter, a larger nozzle diameter will yield an increase in the probability of electrical discharges. Borra et al.(1998) also showed that the discharge probability increased with increasing liquid flow rate. This is also an effect of the lower divergence of the electric field near the jet.
4.3 SIMULATION RESULTS CONCERNING ELECTRICAL DISCHARGES IN THE AIR

As discussed in the previous sections, the electric field strength in the gas near the cone can become higher than the onset field strength for electrical discharges in the surrounding air. This results in a discharge probability larger than zero. Consequently, some discharges will be formed. Figures 3.25 and 3.26 showed the normal and tangential electric field strength in the air at the liquid surface. These figures show that the highest probability for the occurrence of discharges can be found in the region near the cone apex where the jet emerges from the cone.

As Experimentally found by Smith (1986), liquids with a surface tension larger than 0.05 N m\(^{-1}\) could not be sprayed in a stable Cone-Jet mode. Smith concluded that it was most probably due to electrical discharges in the air. However, as shown by Borra et al. (1998), the reality is more complicated. The electrode configuration, liquid flow rate, and conductivity, also play a role in the occurrence of discharges.

Table 4.1 Simulation results of the Cone-Jet Shape Model.

<table>
<thead>
<tr>
<th></th>
<th>(Q) (10^{-10}) m(^3) s(^{-1})</th>
<th>(\gamma) N m(^{-1})</th>
<th>(K) (\mu)S m(^{-1})</th>
<th>(I) nA</th>
<th>(r_0) (\mu)m</th>
<th>(E_0) (10^6) V m(^{-1})</th>
<th>(E_\chi) (10^6) V m(^{-1})</th>
</tr>
</thead>
<tbody>
<tr>
<td>A</td>
<td>7</td>
<td>0.048</td>
<td>69</td>
<td>104.2</td>
<td>9.25</td>
<td>29.9</td>
<td>3.48</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>5.65</td>
<td>34.2</td>
<td>4.50</td>
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<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>4.57</td>
<td>33.7</td>
<td>4.29</td>
</tr>
<tr>
<td>B</td>
<td>21</td>
<td>0.048</td>
<td>69</td>
<td>181.1</td>
<td>9.25</td>
<td>33.6</td>
<td>2.92</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>5.65</td>
<td>26.2</td>
<td>1.15</td>
</tr>
<tr>
<td>C</td>
<td>7</td>
<td>0.048</td>
<td>186</td>
<td>175.0</td>
<td>9.25</td>
<td>33.9</td>
<td>2.44</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>5.65</td>
<td>45.9</td>
<td>4.40</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>4.57</td>
<td>48.1</td>
<td>4.47</td>
</tr>
<tr>
<td>D</td>
<td>14</td>
<td>0.048</td>
<td>69</td>
<td>149.0</td>
<td>7.87</td>
<td>34.6</td>
<td>3.70</td>
</tr>
<tr>
<td>E</td>
<td>14</td>
<td>0.024</td>
<td>69</td>
<td>120.2</td>
<td>7.87</td>
<td>29.7</td>
<td>2.40</td>
</tr>
</tbody>
</table>

The Cone-Jet Shape model presented in Chapter 3 was used to investigate the influence of the surface tension, the liquid flow rate, and the conductivity, on the probability of electrical discharges in the air. Table 4.1 shows some simulation results for the electrode configuration shown in Figure 3.4. The dimensions were equal to the electrode configuration I in table 3.1. The potential difference over the electrodes was 20 kV, the dielectric constant was equal to 37, the density was equal to 1100 kg m\(^{-3}\), and the viscosity equal to 0.02 Pa s. The simulations confirmed
that the higher the flow rate, the higher the probability of electrical discharges. Simulation A is compared to simulation B at a position with a jet radius of 9.25 μm. At that position, the electric field strength at the liquid surface is higher for the high flow rate (B) than for the lower flow rate (A). At the position with a jet radius of 5.65 μm, it is the other way round. As already discussed in section 4.1, not only the electric field strength, but also the radius of the electrode is an important parameter for the discharge probability. The electric field at the position with a jet radius of 5.65 μm for case A can be compared with the field at the position with a radius of 9.25 μm for case B. This comparison shows that the electric field strength is only slightly higher at the position with a jet radius equal to 5.65 μm. So, the probability of discharges in the 5.65 μm radius area of case A is most likely lower than this probability for the 9.25 μm area of case B. This means that the total probability of electrical discharges in the air increases with increasing flow rate.

When simulation A is compared to simulation C then it can be unambiguously concluded that the probability of discharges increases with increasing conductivity. However, with increasing conductivity, the minimum and maximum liquid flow rates, between which the liquid can be sprayed in the Cone-Jet mode, decreases. Reducing the flow rate, reduces the discharge probability. Whether the influence of an increase in conductivity on the discharge probability is completely compensated by the influence of a decrease in the liquid flow rate can’t be concluded from these results. It can be concluded, that the lowest electrical discharge probability can be found near the minimum flow rate for a certain liquid.

When comparing simulations D and E, it can be concluded that the discharge probability increases with increasing liquid surface tension. Although, a relatively large change in surface tension, only yields a small change in the electric field strength.

4.4 CONCLUSIONS

Electrical discharges can completely disturb the droplet production process in the Cone-jet mode. As soon as, the current produced by electrical discharges becomes of the same order of magnitude as the current without discharges present, then the droplet production process will be disturbed. If the discharges produce an irregular pulsating current, then the droplet production process is completely disturbed, and a very wide size distribution is produced. When, the electrical discharges produce a steady current in the glow mode, then a spray with a narrow size distribution can sometimes be produced. However, the produced droplet size and charge can be expected to be different from the droplet size and charge produced in electrohydrodynamic atomization in the Cone-Jet mode without discharges present.

When electrical discharges start to occur, then it is most likely that they will first occur around the cone apex at the position where the jet emerges from the liquid cone.

The electrical discharge probability can be reduced by switching to a positive potential applied to the nozzle, by changing the type of gas around the cone, by spraying in a vacuum, by increasing the gas pressure, or by reducing the surface tension of the liquid.

The lowest electrical discharge probability can be found near the minimum flow rate for
a certain liquid.

A nozzle with a small diameter yields a lower electrical discharge probability than a nozzle with a larger diameter. This is caused by the fact that the electric field strength at the liquid surface is almost not influenced by the nozzle diameter, but the divergence of the electric field around the cone is larger for a smaller nozzle. A larger divergence of the electric field means a lower discharge probability.
CHAPTER 5

SPRAY EVOLUTION

This study concerns the effects that occur in a spray produced by Electrohydrodynamic Atomization in the Cone-Jet mode. Droplet sizes and velocities were measured. A physical model has been developed, which takes the drag force, electric forces, and the droplet inertia into account. The results show the electrical interactions between the charged droplets. They also show an acceleration of the gas in the center of the spray. A relation between droplet size and droplet charge in a spray could be established. The results of the model confirm that the mutual electric interaction of charged droplets, and differences in inertia, are the reason for the size segregation effect in the spray.

5.1 INTRODUCTION

In Electrohydrodynamic Atomization in the Cone-Jet mode, an electric field induces surface charge in a droplet. The electric field accelerates this surface charge. As a result, the droplet shape transforms into a conical shape. At the cone apex, a jet with a high charge density occurs, which can break up into a number of primary main droplets with a narrow size distribution, and a number of secondary droplets and satellites, Cloupeau and Prunet Foch(1989), Gomez and Tang (1994), Chen et al.(1995). Each liquid has a minimum flow rate, below which a stable Cone-Jet mode cannot exist. At this minimum flow rate the jet breaks up due to axisymmetric instabilities. These instabilities are also called varicose instabilities. At higher flow rates, the current through the liquid cone increases. With increasing current, the surface charge on the jet increases. Above a certain surface charge the jet break-up will also be influenced by lateral instabilities of the jet. These instabilities are also called kink instabilities. When the influence of these kink instabilities increases, then the produced size distribution of the main droplets becomes wider, Cloupeau and Prunet Foch(1989). These kink instabilities are sometimes also described as a whipping motion of the jet, Gomez and Tang (1991). Above a certain maximum flow rate, no stable Cone-Jet mode can exist. The process of jet break-up will be discussed in
more detail in Chapter 6.

This chapter concerns the effects that occur in the first few centimeters of a spray produced by Electrohydrodynamic Atomization in the Cone-Jet mode. In the spray, electrical interaction between highly charged droplets causes a size segregation effect. Small droplets will be found at the edge of the spray and large droplets will be found in the spray center, Gañán-Calvo et al. (1994), Tang and Gomez (1994). The evolution of the spray is of importance, since most droplet size measurements can only be performed several millimeters away from the jet break-up point. Another reason is, if we are able to control the size segregation effect taking place in the spray shortly after production, then the main droplets can be separated from the smaller secondary droplets, Tang and Gomez (1994a).

The following sections will show measurement results of droplet sizes and velocities at short distances from the droplet production zone. It will show some pictures of the jet breaking up into droplets. Results will be evaluated with a Lagrangian model, similar to the model presented by Gañán-Calvo et al. (1994). From the comparison between simulations and measurements, conclusions will be drawn regarding the size segregation mechanism in the spray, the balance between electric force and drag force, the acceleration of the surrounding gas, the influence of electrostatic interaction of charged droplets on the axial velocity, and the relation between droplet size and droplet charge in a spray.

5.2 EXPERIMENTAL METHOD

A spray was produced by Electrohydrodynamic Atomization in the Cone-Jet mode. A nozzle-to-plate spraying configuration was used. The nozzle diameter was 5 millimeters, it was a cylindrically shaped nozzle with a flat tip, and the inner diameter was 0.25 millimeters. The nozzle to plate distance was 12.1 centimeters. Figure 5.1 shows the experimental set-up. This figure also shows the cylindrical coordinate system used in this chapter. Ethylene glycol was sprayed at various flow rates. The conductivity of the liquid was 70 μS m⁻¹. The potential difference between nozzle and plate was in most cases equal to 16 kV, the measurement position ranged from 0.7 to 2.6 cm below the visible tip of the jet.

The droplet size and droplet velocity were measured with the Particle Analysis System (PAS Dantec). Two crossing laser sheets create an interference pattern. The optics of the system are focused on the measurement volume which is only the center of the interference pattern. The measurement volume was 1 millimeter long and 0.2 millimeters wide. The velocity was measured with the principle of Laser-Doppler. The droplet size was evaluated from the intensity of the forward scattered light. Every measurement period, the system was calibrated with a reticle with a 10-μm pin hole. Since the system is focused on the center of the interference pattern, the light intensity

![Figure 5.1 Experimental set-up.](image-url)
profile in the measurement volume is reasonably flat. This profile was tested with the following procedure. First, the system was calibrated with the pin hole in the center of the measurement volume. Then, the size of the pinhole was measured at various places in the measurement volume. Table 5.1 shows the results of these measurements.

**Table 5.1** The results of the size measurement of a 10-μm pin hole at various places in the measurement volume.

<table>
<thead>
<tr>
<th>Measured Size of Pin Hole [μm]</th>
<th>Fraction of Total Number of Measurements</th>
</tr>
</thead>
<tbody>
<tr>
<td>10</td>
<td>0.18</td>
</tr>
<tr>
<td>9</td>
<td>0.48</td>
</tr>
<tr>
<td>8</td>
<td>0.28</td>
</tr>
<tr>
<td>7</td>
<td>0.06</td>
</tr>
</tbody>
</table>

This size measurement yielded a narrow size distribution with an average which is 12 percent smaller than the real size of the pin hole. The alignment of the system was reasonable, but not 100 percent. If the alignment had been better, then a slightly more narrow size distribution could have been obtained. The alignment was not improved, because the system could only be used for a short period of time.

The measurement system is also susceptible to an effect that overestimates the droplet size. If one droplet is in the measurement volume and another droplet is in the interference pattern just outside the measurement volume, then the recorded intensity will be slightly too high. This effect was reduced by moving the measurement volume away from the droplet production zone with its extremely high droplet concentrations. However, the effect cannot be avoided completely.

Both effects together mean that the system measures a wider size distribution, than the real size distribution. Especially for sprays with narrow size distributions, this effect is of importance. Due to these effects, the relation between droplet velocity and droplet size is disturbed. If a truly monodisperse aerosol is produced with a constant droplet velocity, then the system will measure a size distribution with a certain width. Since the velocity in this spray is constant, it seems that a range of droplet sizes occurs in the spray, and the droplet velocity seems to be independent of the droplet size in this distribution. In fact there is only one size with only one velocity.

Electrohydrodynamic Atomization in the Cone-Jet mode, produces a spray in which a size segregation effect occurs. This effect makes the size distribution in one position of the spray very narrow. That is why, the profile, where the droplet velocity is plotted as function of the droplet size for one measurement position deviates from the same profile that is obtained at different positions in the spray. The velocity-size profile obtained in one measurement position is not reliable. However, the average velocity obtained for the modal value of the size distribution at that position is only affected by a change in the average size and not by a change in the width of the size distribution. This means that the velocities of the modal sizes measured at different
positions in the spray can be compared with each other. That is why, only the average velocity of the modal droplet size is accepted as a result. The only exceptions were cases where a bimodal size distribution was measured in one measurement position. In that case, the two average velocities of the modal size of both modes were accepted. Since the modal size depends on the measurement position in the spray, a velocity-size profile can still be obtained.

The total measurement accuracy of the droplet size was estimated to be 20 percent. The repeatability in one measurement period without a new calibration was in the order of a few percent. The lower limit of the system was 3 μm due to a limited laser power.

The velocity measurement seems to be reasonably accurate, but an evaluation of the real accuracy was not made. However, for the modal droplet size at a certain measurement position in the spray, the measured droplet velocities were in agreement with previous measurements. Also, the repeatability was excellent.

5.3 LAGRANGIAN MODEL

Droplet dynamics can be described by a Lagrangian model, Gañán-Calvo et al. (1994), Tang and Gomez (1994). Gañán-Calvo derived his model from the momentum equation given by Tchen (1947), and Maxey and Riley (1983). This physical model describes the movement of highly charged droplets in a strong electric field. It takes the drag force, the electric force due to the applied electric field, the electric forces due to electrical interaction between charged droplets, and the droplet inertia into account. Since electric forces are much stronger than gravity, and since the density of the surrounding air is much lower than the density of the droplets, gravity and virtual mass effects can be neglected. Droplet evaporation was neglected, because ethylene glycol is a low volatile liquid, and because droplets need less than 1 millisecond to travel the first centimeter from the droplet production area. Although, not valid in the area within one millimeter from the droplet production area, it is assumed that the drag of the surrounding gas is not influenced by the presence of neighboring droplets. Using these assumptions, droplet dynamics can be described by the following equation

\[
m_i \frac{\text{d} \bar{u}_i}{\text{d}t} = q_i E_{\text{ext}} + C_D \frac{\pi d_{i}^2}{4} \frac{\rho(\bar{u}_f - \bar{u}_i)}{2} + \sum_{i \neq j}^{N} \frac{q_i q_j}{4 \pi \varepsilon_0 r_{ij}^3} \bar{F}_{ij}
\]

where, \(C_D\) is drag coefficient, \(E_{\text{ext}}\) is external electric field strength [V m\(^{-1}\)], \(m\) is droplet mass [kg], \(q\) is droplet charge [C], \(d_i\) is droplet diameter [m], \(r_{ij}\) is distance between two droplets [m], \(t\) is time [s], \(u_i\) is droplet velocity [m s\(^{-1}\)], \(u_f\) is fluid flow velocity [m s\(^{-1}\)], which is assumed to be equal to zero, \(\varepsilon_0\) is permittivity of a vacuum [C\(^2\) N\(^{-1}\) m\(^{-2}\)], \(\rho_{\text{air}}\) is air density [kg m\(^{-3}\)], indices \(i\) and \(j\) refer to the droplet number. The droplet acceleration depends on the mass of the droplet and three forces. The first force is the electric force exerted on a charged droplet due to the background electric field. This background electric field is created by the potential difference
over nozzle and plate. The second force is the drag force of the surrounding gas. The third force is the summation of the electric forces exerted by the surrounding charged droplets.

The drag coefficient can be calculated following Klyachko,

\[
C_D = \frac{24}{Re} \left( 1 + \frac{Re^{2/3}}{6} \right) \quad 2 < Re < 800
\] (5.2)

where, Re is droplet Reynolds number.

5.3.1 The External Electric Field

In Chapter 3 a Cone-Shape model was presented that is able to calculate the shape of the cone and jet, the surface charge on the jet, the electric fields in and outside the liquid cone, and the current through the liquid cone. The electric fields were calculated with Gauss' law. For the electric field calculation the presence of the charged droplets is of importance. Simulations with the Cone-Shape model showed that the introduction of the droplet space charge into the model resulted into a change in the liquid cone shape. This change could be compensated by an increase in the potential difference of about 7 percent. Although this effect is not very large, it was taken into account. The droplet space charge as function of the position in the spray was calculated from the current, the droplet velocities, and the droplet concentrations. These values were obtained from the experiments. This space charge was included into the Cone-Shape model. The shape of the liquid cone and jet was calculated. Then, the droplet space charge, which was within a one centimeter distance from the tip of the jet, was removed. Without changing the liquid cone shape, and without changing the surface charge on the liquid surface, the background electric field strength was calculated. This background electric field strength can be found in the first force term of equation (5.1). The electric field, due to the droplets within a one centimeter distance from the tip of the jet, is represented by the third force term of equation (5.1).

In the Lagrangian spray model, the electric field is simulated with the electric field of four point charges. Figure 5.2 gives a schematic representation of the position of these charges. Table 5.2 gives the charges used to simulate the radial and the axial electric field. Since four point charges are a huge simplification of the charge distribution on the liquid electric field.

\[\text{Figure 5.2 Schematic representation of the position of the point charges, which are used in the simulation of the external electric field.}\]
cone, and on the electrodes, it proved to be impossible to choose four point charges that create the correct radial and axial electric field at the same time. That is why, in the simulation different charges were used to simulate the radial and the axial electric field in the gap between the tip of the jet and the measurement position.

Figure 5.3 shows the comparison between the axial and radial electric field, calculated by the Cone-Shape model using Gauss’ law, and the fields calculated with the point charges. This figure shows that it is possible to simulate the electric field in this way. The advantage of the point charge method is the very short computation time of the electric field at any place in the spray within the first centimeter of the tip of the jet. The field strengths of Figure 5.3 were calculated as functions of the axial position and in various radial positions within the spray. The deviations are less than 10 percent. The influence of these deviations is even much less than 10 percent, because they are averaged out along the axial axis. This figure is calculated for the electrode configuration of section 5.2 and for an ethylene glycol spray at a liquid flow rate of $1.39 \times 10^{-9}$ m$^3$s$^{-1}$.

Table 5.2 Point charges used to simulate the background electric field in the first centimeter from the tip of the jet, see Figure 5.3.

<table>
<thead>
<tr>
<th>Point Charge Number</th>
<th>Axial Field Charge</th>
<th>Axial Field Position z Coordinate</th>
<th>Radial Field Charge</th>
<th>Radial Field Position z Coordinate</th>
</tr>
</thead>
<tbody>
<tr>
<td>q1</td>
<td>$5.16 \times 10^{-13}$</td>
<td>-0.0000191</td>
<td>$3.15 \times 10^{-13}$</td>
<td>-0.0000238</td>
</tr>
<tr>
<td>q2</td>
<td>$4.27 \times 10^{-12}$</td>
<td>-0.000392</td>
<td>$2.00 \times 10^{-12}$</td>
<td>-0.000168</td>
</tr>
<tr>
<td>q3</td>
<td>$1.48 \times 10^{-9}$</td>
<td>-0.00574</td>
<td>$3.24 \times 10^{-10}$</td>
<td>-0.00328</td>
</tr>
<tr>
<td>q4</td>
<td>$1.40 \times 10^{-8}$</td>
<td>-0.0191</td>
<td>$1.98 \times 10^{-7}$</td>
<td>-0.0502</td>
</tr>
</tbody>
</table>

5.3.2 Droplet Charge Modeling

The average droplet charge can be calculated from the droplet size and the current. However, small droplets will have a lower charge than large droplets. That is why a relation between droplet charge and droplet size is needed. Normally this relation is assumed to be...
\[
\frac{q_{\text{sat}}}{q_{\text{main}}} = \left( \frac{d_{\text{sat}}}{d_{\text{main}}} \right)^n
\]  

(5.3)

where, \( n \) is a constant, \( q_{\text{main}} \) refers to the primary or main droplets, and \( q_{\text{sat}} \) refers to the secondary and satellite droplets. Tang and Gomez (1994) measured the average size of the primary droplets, and the secondary droplets for a spray of heptane. They also measured the total current, and the current transported by the secondary droplets. This resulted into a value of \( n \) equal to 1.5.

In the presented simulations, the charge of the droplets was calculated in the following way. The current and the droplet diameter were measured. These measurements proved that the size ratio of the primary droplets over the secondary droplets was about equal to two, see Figure 5.8. Since the number of secondary droplets was not much larger than the number of the main droplets, it was assumed that these numbers were equal. In that case, the total volume of one primary main droplet plus one secondary droplet is 1.125 times the volume of a main droplet. With this volume, and with the liquid flow rate, the frequency of the droplet production can be calculated. This frequency is then used to calculate the total charge of the primary plus the secondary droplet. With this result, and with equation (5.3), the charge of the main droplet can be calculated. Equation (5.3) can then be used to calculate the charge for every particle size.

5.3.3 Droplet Seeding, Initial Velocity, Initial Position, and Size Distribution

Now being able to calculate the forces on the droplet, the calculation of the droplet velocity and position still requires the initial droplet position, and the initial droplet velocity. In another experiment, the break-up of a liquid jet was investigated by means of a High Speed Spray Imaging System (HSSIS) purchased from Oxford Laser. This system consists of a digital camera (KODAK) connected to a computer, which is equipped with a frame grabber. A long distance microscopic lens is fixed to the camera. The illumination of the object is done by an infrared laser. A control box synchronizes the camera and the laser. Pictures can be made with an illumination time down to 0.5 \( \mu \)s. The separation between two following pictures can be down to 15 \( \mu \)s. The optical system allows to see particles of a few micrometers. However, perfect focusing on a moving object proved to be difficult. Due to this effect, the measurement accuracy was about 4 \( \mu \)m. Figure 5.4.a shows a camera picture of the break-up of a jet induced by axisymmetric or varicose instabilities. Figure 5.4.b shows the jet break-up induced by both varicose and kink instabilities at the same time. Figure 5.4.a shows that the radial component of the initial velocity can be assumed to be equal to zero. The camera system was used to estimate the droplet velocity at jet break-up. From measurements with ethylene glycol with a conductivity of 21 and 52 \( \mu \)S m\(^{-1}\) at a flow rate of 1.4 \( 10^9 \) m\(^3\) s\(^{-1}\) the initial velocity for ethylene glycol with a conductivity of 70 \( \mu \)S m\(^{-1}\) was estimated to be 25 m s\(^{-1}\).

With respect to the initial droplet position, the axial position \( z \) is equal to zero, and with respect of the radial position two extremes can be given. If the initial droplet position is fixed at
the jet axis \((r=0)\), combined with an initial radial velocity equal to zero, then the radial dispersion of droplets in the spray would be equal to zero. In the whipping jet break-up regime, the fluctuation in initial radial position can be very large. However, measurements with the camera system proved that below a certain maximum flow rate, the jet breaks up due to varicose instabilities. When the jet breaks up due to varicose instabilities, then the fluctuation in the initial radial position of the droplets is smaller than the jet radius. Above this maximum flow rate, the fluctuations in the radial position increase.

In the whipping jet break-up regime the width of the size distribution increases. So, the measured size distribution already gives an indication of the jet break-up regime. In the simulations the initial radial position of each droplet was randomly generated. The maximum value of this radial position was mostly taken equal to 1 \(\mu m\).

This choice of 1 \(\mu m\) will be discussed in section 5.4.7.

The simulation of the spray evolution requires a size distribution in order to generate randomly droplets with a certain charge and size. The droplet size distribution was measured, see Figure 5.8. From this size distribution the chance on a certain droplet size can be deduced. A random generator was used to generate a number between 0 and 1. The size distribution was used to determine which droplet size corresponds with the value of this number. Further, it was assumed that the bimodal size distribution comes from the production of primary droplets with secondary droplets and satellites in between. So, first a droplet larger than the median value was produced, followed by a droplet smaller than this value.

5.3.4 The Evolution in Time

The evolution in time of the spray was modeled by taking constant time steps. The time step was equal to 1/5 of the time between the production of two droplets. For ethylene glycol at a flow rate of 1.39 \(10^9\) m\(^3\) s\(^{-1}\), this time step was equal to 2 \(10^7\) s. The distance, these droplets will travel in one time step, can be estimated using the velocity acquired in the previous time step. In order to calculate the forces working on a droplet during this time step, all droplets were moved half the estimated distance. Using these positions, the forces acting on the droplets are calculated. This results in an acceleration in three directions. These accelerations are then used to calculate the
new droplet position and the new droplet velocity taking the position and velocities acquired in the previous time step as a starting point.

5.3.5 Electrical Interaction between Droplets

In order to save computation time not all droplet-droplet interactions were taken into account. Simulations proved that the interaction with only the 120 closest droplets had to be taken into account.

5.3.6 Differences between this Model and other Models

Four main differences can be distinguished between the above presented model and the models presented by Gañán-Calvo et al. (1994), and Tang and Gomez (1994). The first difference is the method of calculating the external electric field. The external electric field was numerically calculated using Gauss' Law in the Cone-Shape Model, Chapter 3. Where, Gañán-Calvo used an estimation of the electric field, and Tang calculated the electric field from the droplet velocities that he measured. The second difference is the droplet seeding procedure. Gañán-Calvo assumed a larger variation in initial radial position of the droplets in the droplet production area. This assumption will be discussed in section 5.4.7. In all models it was assumed that no acceleration of the surrounding gas takes place. However, in the presented simulations, the possibility of this effect to occur in the real spray was kept open. The fourth difference was that Gañán-Calvo assumed a log-normal size distribution with a standard deviation coming from measurements. In the presented model the bimodal size distribution obtained in the measurements was used as an input into the model.

5.4. COMPARISON OF MEASUREMENTS AND SIMULATIONS

5.4.1 Measurement Results

When liquids are sprayed in the Cone-Jet mode, then the size distribution of the spray can be monodisperse, bimodal or polymodal. In the sprays of ethanol, acetone and ethylene glycol, the size segregation effect and the acceleration of the surrounding gas inside the spray were observed. However, the volatility of ethanol and acetone makes investigation of these effects difficult. However, when ethylene glycol is sprayed, even the smaller secondary droplets do not evaporate. For instance at a flow rate of $1.39 \times 10^{-9} \text{ m}^3 \text{s}^{-1}$, the average secondary droplet size is equal to $8 \mu\text{m}$. The evaporation time of these droplets is still in the order of seconds. This low volatility of ethylene glycol allows the investigation of the size segregation effect in the spray, the interaction between charged droplets, the relation between the droplet charge and droplet size, and the acceleration of the surrounding gas inside the spray. That is why, the results concerning ethylene glycol will be discussed.
Ethylene glycol was sprayed at a flow rate of $1.39 \times 10^9$ m$^3$ s$^{-1}$. The droplet size was measured at different radial positions in the spray with the PAS system described in section 5.2. The potential difference was 16 kV, and the measurement position was 1 cm below the droplet production area. The droplet size distribution depended on the radial coordinate of the measurement position. Figure 5.5 shows the size distribution measured in the spray center. Figure 5.6 shows the size distribution at the boundary between the area with main droplets and the area with the smaller secondary droplets and satellites. Figure 5.7 shows the size distribution at the

**Figure 5.5** Droplet size distribution measured in the spray center.

**Figure 5.6** Droplet size distribution measured at the boundary of the spray area with main droplets and the area with secondary droplets.

**Figure 5.7** Droplet size distribution measured at the spray edge.

**Figure 5.8** Droplet size distribution integrated over the total spray area.
spray edge. The size distribution of all the produced droplets was obtained by integrating the particle flux over the surface. Figure 5.8 shows this integrated size distribution. Due to the lower limit of the measurement system, it is not known whether also satellites smaller than 3 μm are produced. The small mode of the droplet size distribution in Figure 5.8 can't be a result of droplet evaporation at the edge of the spray. The droplets need less than 1 millisecond to travel to the measurement position, while the total evaporation of these ethylene glycol droplets takes in the order of seconds. Meesters et al. (1992) reported electrical discharges in the air around the tip of an ethylene glycol cone. These discharges can disturb the droplet production process, and change the charge distribution on the droplets. That is why, the occurrence of electrical discharges was checked. It was found that the current due to these electrical discharges was orders of magnitude smaller than the current through the liquid cone without discharges present. So, these electrical discharges can be neglected for ethylene glycol sprayed in the described electrode configuration at the liquid flow rates presented in this chapter.

5.4.2 Comparison Simulations and Measurements

During the measurements, the electric current received by the grounded plate was measured. At a flow rate of 1.39 $10^9$ m$^3$ s$^{-1}$, the current was equal to 1.4 $10^7$ A. When it is assumed that the constant $n$ of equation (5.3) is equal to 1.4 or 1.6, together with the size distribution of Figure 5.8, and the obtained current, then the procedure described in section 5.3 can be used to simulate the evolution of the spray. Figure 5.9 shows the average droplet size measured and calculated with the model as function of the radial position. Figure 5.10 shows the normalized droplet

![Graph showing average droplet size as function of radial distance from spray center.](image1)

![Graph showing normalized droplet concentration as function of radial distance.](image2)

**Figure 5.9** Average droplet size as function of the radial position in the spray.  
**Figure 5.10** Normalized droplet concentration as function of the radial position in the spray.
number concentrations as function of the radial position. The concentration is normalized with the concentration in the center of the spray. The number density per cubic meter measured in the spray center was equal to $2.3 \times 10^9$. On the other hand, the model predicts a number density per cubic meter of $4.6 \times 10^9$. Three reasons could be found to explain this difference.

Firstly, the measurement system only recorded 45 percent of the droplets that should have been produced. This comes from the fact that not every recorded signal of a droplet is accepted and counted as a droplet, and a small size measurement error will have a large effect on the volume of the droplet, and consequently on the calculated number of droplets that should have been produced. However, when it was assumed that the average size of the size distribution was 11 percent larger than the measured value, then the simulations showed that the concentration in the center of the spray was even $4.9 \times 10^9 \text{ # m}^3$. In that case the radial dispersion of the spray was lower than the dispersion shown in Figures 5.9 and 5.10. For the size distribution of Figure 5.8 and Figure 5.10, the area of low droplet concentration was found at a radial position of 1.8 mm. For the size distribution with an 11 percent larger average size, this radial position was equal to 1.5 mm.

Secondly, due to the assumption that the air velocity is equal to zero, the droplet velocity in the spray center is underestimated by the model. This will be shown later. A lower velocity results in a higher droplet concentration.

Thirdly, A measurement slightly out of the center of the spray results in a lower particle concentration. The measurement volume is 1 mm long and 0.2 mm wide. So, the edges of the measurement volume are not in the spray center. This results in a lower average concentration compared to the real spray center.

The difference in the radial dispersion of the secondary droplets, between the calculations and the measurements, can possibly be explained from the following effect. The surrounding gas in the spray center is accelerated by the droplets as will be discussed later. This will cause a gas flow, which might reduce the spray width close to the cone apex. However, this gas flow might enhance the dispersion of the droplets as soon as the droplets start to decelerate.

Looking at Figures 5.9 and 5.10 it can be concluded that, in spite of the underestimation of the radial dispersion of the secondary droplets, the model predicts well the droplet size segregation effect in the spray, and the normalized concentration profile as function of the radial position. It even predicts the low particle concentration inside the spray at the boundary between the large main droplets and the smaller secondary droplets and satellites. We will call this area the Dark-Zone. When laser light falls through the spray, the area with a low droplet concentration at the boundary between main droplets and secondary droplets appears to be dark compared to the other areas of the spray. This area was also observed by Tang and Gomez (1994).

5.4.3 The Size Segregation Mechanism

Tang and Gomez (1994) gave the following explanation of the size segregation effect in the spray. This explanation was confirmed by the model. At the moment of production, the distance between main droplets and secondary droplets is small, approximately half the distance between
two main droplets. For ethylene glycol at a flow rate of $1.39 \times 10^9 \text{ m}^3 \text{ s}^{-1}$ the distance between main droplets is about 60 $\mu$m. The camera picture in Figure 5.4.a, showed that the radial velocities are much lower than the axial velocities. This picture also showed, that the initial radial positions, and the axial velocities for main droplets and secondary droplets are almost equal. Due to a difference in droplet charge, droplet diameter, droplet mass, the electric force, the drag force, and the inertia of a large droplet will differ from those of a small droplet. This will soon result in a difference in the axial velocity. As a result the distance between a main droplet and a secondary droplet will be reduced. The electrical interaction between the two charged droplets increases. Both droplets experience an equal force with a radial component. The small droplet has a lower inertia than the large droplet. This results in a larger acceleration for the small droplet. Due to the larger acceleration in the radial direction and a lower drag force, the small droplet will attain a higher radial velocity, and a segregation in droplet size occurs in the spray. As soon as the secondary droplets segregate from the main droplets, the radial velocity will be reduced due to the drag force of the gas. However, the radial electric field, which is stronger at the edge of the spray than in the spray center, will keep the secondary droplets at the outside of the spray as long as the spray is not disturbed by any external force.

Simulations showed that the Dark-Zone, the low droplet concentration at the boundary between main droplets and secondary droplets, only occurs when a bimodal size distribution is produced. The occurrence of a Dark-Zone is an indication that the produced spray is bimodal. Figure 5.11 shows the size segregation effect just after the jet has broken up into droplets.

5.4.4 The Axial Droplet Velocity

The electric interaction between two charged droplets of different droplet mass causes the size segregation effect as discussed in the previous section. This electrical interaction between two charged droplets has also an influence on the axial velocity of the droplets. When a fast droplet approaches a slower droplet, then the slower droplet is accelerated, and the faster droplet is slowed down. When the fast droplet overtakes the slow droplet, then the fast droplet is accelerated, and the slow droplet is slowed down. Due to the increase in the radial distance between these droplets during this process, on average the fast droplets are slowed down and the slow droplets are accelerated. Especially close to the droplet production area this effect can be
considerable. Figure 5.12 shows the result of calculations with and without electrical interaction of charged droplets. The results were obtained at a position at a distance of 1 cm from the droplet production area. The solid line shows the axial velocity as function of the droplet diameter without droplet-droplet interaction. In this calculation the axial electric field of the spray center was used. The points are the axial droplet velocity as function of the droplet diameter. These points are calculated at different radial positions in the model spray. The droplet-droplet interaction has been taken into account. When comparing the results of one position in the spray, then Figure 5.12 shows a small reduction of the velocity of the larger droplets and a small acceleration of the smaller sizes. However, the absolute effect on the axial velocity is limited at this distance from the droplet production area. It is also seen that the average velocity at the edge of the spray is lower than the velocity calculated without droplet interaction using the external electric field of the spray center. This result confirms the conclusion that the axial electric field is a bit lower at the spray edge, Gañán-Calvo et al. (1994).

5.4.5 Acceleration of the Surrounding Gas inside the Spray

A more important influence on the velocity profiles was found from the velocity measurements in the spray center. Figure 5.13 shows the average droplet diameter and the average droplet velocity as function of the radial position in the spray for ethylene glycol at a flow rate of 1.39 $10^3$ m$^3$ s$^{-1}$, for a measurement distance of 2.6 cm from the droplet production area. The potential difference was equal to 16 kV. The average droplet size was constant in the first 3 mm from the spray center. However, the axial velocity decreased by 40 percent. This effect was found in all the sprays tested, although the absolute values differed from spray to spray. Simulations showed that a difference in droplet charge could not explain this result. The only other possible explanation is an acceleration of the surrounding gas. In the first centimeter from the droplet production area, the width of the spray is in the order of a few hundreds of micrometers, and the electric field is still very high. So, the acceleration of the air will be confined to an area with a radius of several hundreds of micrometers. Further away from the droplet production zone, the air flow spreads out, but remains confined to a relatively small area in the spray center.
Figure 5.13 The average droplet size and droplet velocity as functions of the radial position measured for ethylene glycol.

Figure 5.14 The average droplet velocity as function of the radial position in the spray.

Figure 5.14 shows the average droplet velocities as functions of the radial distance from the spray center for ethylene glycol at a liquid flow rate of $1.39 \times 10^9 \text{ m}^3 \text{ s}^{-1}$. The measurement position was 1 cm from the tip of the jet. The figure clearly shows that the model underestimates the droplet velocity in the spray center.

The following procedure can be used to estimate the air velocity, Gañán-Calvo et al. (1994). Since, the velocity of the main droplets in the spray center was equal to the initial droplet velocity, we can assume that the electric forces exerted on the droplets are transferred to the gas. As a result, the electric power generated by charged droplets traveling in the electric field should be equal to the kinetic power of the gas flow. This results in the following equation

$$\Delta V I = \frac{1}{2} \rho_{\text{air}} v_{\text{air}}^2 Q_{\text{air}}$$

(5.4)

where, $\Delta V$ is potential difference between the tip of the jet and the measurement position [V], $I$ is current through the liquid cone [A], $v_{\text{air}}$ is air velocity [m s$^{-1}$], and $Q_{\text{air}}$ is the flow rate of the air flow [m$^3$ s$^{-1}$]. The air flow rate can be estimated from the following equation

$$Q_{\text{air}} = \pi r_{\text{air}}^2 v_{\text{air}}$$

(5.5)

where, $r_{\text{air}}$ is the radius of the air flow [m]. Figure 5.14 shows, that a radius of the air flow at 1 cm from the cone tip, could be taken equal to 1 mm. The potential drop $\Delta V$ was calculated using
the Cone-Shape model presented in Chapter 3. The potential difference was 4 kV. Together with a current of 1.4 $10^7$ A, this results in an air velocity of 6.6 m s$^{-1}$. When the radius of the gas flow is taken equal to 2 mm, then the velocity is equal to 4.2 m s$^{-1}$. From Figure 5.14 it can be deduced that the air velocity in the spray center is larger than 4.2 m s$^{-1}$. At a radial position of 2 mm, this velocity is lower than 4.2 m s$^{-1}$. So, in the following calculations, the air velocity in the center of the jet is estimated with the assumption of an air flow radius of 1 mm.

For ethylene glycol with a conductivity of 21 $\mu$S m$^{-1}$ the influence of the liquid flow rate on the air velocity was investigated. The electrode configuration was different. The diameter of the nozzle was 8 mm. The length was 6 mm. Above this nozzle there was a plate with a radius of 25 mm. The potential of this plate was the same as the potential of the nozzle. The counter electrode plate was only 40 mm below the top plate. The radius of this plate was about 50 mm.

The potential drop between the tip of the jet, and the position at 1 cm distance from this tip, was calculated with the Cone-Shape model. The potential drop in this electrode configuration was 6.1 kV. This was a higher potential drop than for the electrode configuration used for the measurements described in the previous sections. The potential drop appeared to be almost independent of the liquid flow rate. It increased from 6.0 to 6.2 kV. This can be explained from the fact, that the jet size and the surface charge on the jet have only a local influence on the electric field. The simulations showed that the electric field depended on the liquid flow rate. However, this dependence was only noticeable in the first millimeter from the tip of the jet. In the next 9 mm it was independent of the liquid flow rate. In this area the field mainly depends on the surface charge on the liquid cone, and on the electrodes. The surface charge distribution on the cone is almost not influenced by the liquid flow rate.

Figure 5.15 shows the calculated current through the liquid cone, and the estimated air velocity. The figure shows that the air velocity goes up with increasing liquid flow rate. This increase in the air velocity is caused by the increase in current.

In other simulations, the potential drop appeared to be strongly depending on the length of the nozzle. The lowest air velocities in the spray can be obtained in electrode configurations with long nozzles with a small diameter. The electric field in the gap mainly depends on the electrode configuration that is used. A long nozzle with a small diameter will create an electric field with a strong divergence. As a result, the potential drop in the gap is lower, while the electric field strength at the jet surface is of the same order of magnitude as for other nozzles. Also, the air

![Figure 5.15 The current through the liquid cone calculated by the Cone-Shape model and the estimated air velocity as functions of the liquid flow rate.](image-url)
velocities can be reduced by spraying a liquid at a flow rate just above the minimum flow rate, because in that case the current is at its lowest value.

5.4.6 The Influence of the Power $n$ of the Charge to Diameter Relation

Figure 5.14 showed that the model underestimated the droplet velocity in the spray center by several meters per second. This was explained in the previous part as an effect caused by the movement of the air. In order to investigate the influence of the droplet charge, the value of the constant $n$ of equation (5.3) was varied. This can only be done, if the air flow inside the spray is taken into account. The average axial velocity of the modal size at a certain radial position was measured. The modal droplet size depends on the measurement position in the spray. So, by measuring at different radial positions in the spray a plot can be made of the axial velocity as function of the modal droplet size. These results were compared to simulation results. In the section about the axial droplet velocity, it was shown that the influence of the electrical interaction between charged droplets has little influence on the axial droplet velocity at a distance of one centimeter from the droplet production area. In order to save computation time, reduce processing time, and because the influence of droplet interactions is relatively small, the droplet interaction was neglected. The axial droplet velocity was now only calculated with the external axial electric field of the spray center. Figure 5.16 shows the velocities as functions of the modal size for ethylene glycol at a flow rate of $1.39 \times 10^9$ m$^3$s$^{-1}$. The measurement distance was 1 cm from the droplet production area. The size segregation effect implies that the small droplets will be found at the edge of the spray. So, the modal size at the spray edge will be small. At the edge of the spray the air velocity should be low. So, at the edge of the spray measurement and simulation velocities should come close. The velocity of the large modal sizes should be underestimated by the model because they can be found in the spray center. The calculated velocities seemed to fit best to the measurement results, when $n=1.5$. A value of $n=1.0$ does not seem to be possible because in that case the model would predict faster droplets than the measurements. A value of $n=2.0$ also does not seem to be possible, because that would mean that the air velocities at the edge of the spray are larger than at a position closer to the spray center were the modal droplet size is equal to 15 $\mu$m.

![Figure 5.16 The axial droplet velocities as functions of the modal droplet size.](image_url)
However, more experimental conditions had to be tested. This was done for flow rates of 0.278, 0.695, 1.39, 2.78 $10^5$ m$^3$s$^{-1}$, at potential differences of 16, 17.5 and 19 kV, and at 0.7, 1 and 2.6 centimeter distance from the droplet production area. All these measurements showed the same trend. A value of $n=1.5$ yielded the best results.

Figure 5.17 shows the same results as Figure 5.16. The only difference is that this time the complete velocity profile obtained at each measurement position is shown. Three things can be seen in this figure. Firstly, the velocity profile measured at one radial position deviates in most cases strongly from the simulated profile. In the section 5.2, it was already explained, that the droplet size obtained from the measurements depends on the concentration, and on the position where the droplet goes through the measurement volume. This also affects the velocity profiles. Secondly, the velocity in the spray center is underestimated, because the acceleration of the air in the spray center is neglected. Thirdly, for one measurement position, the simulated results fit surprisingly well for $n=1.5$. This measurement position is located in the Dark-Zone. At this boundary between main droplets and secondary droplets, both main droplets and secondary droplets can come into the measurement volume. In this case the difference in droplet size between main droplets and secondary droplets is much larger than the deviations that occur due to the measurement tool, see Figure 5.6. In this position, the measurement of the axial velocity profile as function of the droplet size is reliable. The same effect occurred in all investigated sprays where a measurement was made at the boundary between main droplets and secondary droplets. The results all fitted best to $n=1.5$.

![Figure 5.17 The axial droplet velocities as functions of the droplet size.](image1)

![Figure 5.18 The measured axial droplet velocity of the modal size minus the simulated axial velocity as functions of the radial position, liquid flow rate $0.7010^5$ m$^3$s$^{-1}$.](image2)

The air velocity can be made visible when, the difference in average axial velocity of the
modal size minus the simulated velocity is plotted as function of the radial position. Figures 5.18 to 5.20 show the profiles obtained at 1 cm from the tip of the jet. These figures can be used to check three things. The airflow velocity in the spray center can be estimated. The air flow for a liquid flow rate of $2.78 \times 10^9 \text{ m}^3 \text{s}^{-1}$ was lower than expected. At this flow rate the Dark zone had disappeared and the size distribution was much wider. Especially the number of secondary droplets and satellites was much larger. This indicates that the jet break-up was in the whipping jet break-up regime. The average droplet size was even smaller than for a flow rate of $1.39 \times 10^9 \text{ m}^3 \text{s}^{-1}$. This resulted in a larger radial dispersion of the droplets, and the velocity of the droplets was also lower. All these results could lead to a lower air velocity in the spray.

**Figure 5.19** The measured axial droplet velocity of the modal size minus the axial velocity simulated as function of the radial position, liquid flow rate $1.39 \times 10^9 \text{ m}^3 \text{s}^{-1}$.

**Figure 5.20** The measured axial droplet velocity of the modal size minus the axial velocity simulated as function of the radial position, liquid flow rate $2.78 \times 10^9 \text{ m}^3 \text{s}^{-1}$.

Secondly, the measured velocities minus the modeled velocities should for every measurement in a certain measurement position give the same value of the air velocity. As already said in the previous paragraph, a value of $n=1.5$ yields the best results.

Thirdly, the calculated air velocity should not become negative at the edge of the spray. This is a reasonable assumption, because the measurement position is far from the counter electrode. When an airflow meets an obstacle, then an area of high pressure occurs close to that obstacle. This could cause negative air velocities at the edge of the spray. In this case, this does not seem likely, because the counter electrode is quite far, 10 cm, from the measurement position.
5.4.7 The Influence of the Initial Droplet Seeding on the Radial Droplet Dispersion

A maximum radial droplet position of 1 μm, and an initial radial droplet velocity equal to zero were used to calculate the above presented simulation results. García-Calvo claimed that simulation results are not sensitive to the initial random perturbation seeding position, when the displacements are small in comparison with the jet length. Figure 5.4a showed a picture of a jet breaking up due to varicose instabilities. This figure clearly showed that the radial displacement at the moment of break-up was smaller than the jet radius. Above a certain flow rate, kink instabilities become more important in the jet break-up. In that case the radial displacement for the droplet seeding should be taken larger than the jet radius. Ethylene glycol, at a liquid flow rate of 1.39 $10^9$ m$^3$ s$^{-1}$ and a conductivity of 70 μS m$^{-1}$, is close to the maximum flow rate that still yields a jet break-up due to varicose instabilities. So, some simulations were done to investigate the influence of the assumptions of the initial droplet seeding.

If the maximum initial radial position of the droplets was put equal to 100 μm, then the size segregation effect occurred in the spray. However, the Dark-Zone, the area with low droplet concentrations at the boundary between main droplets and secondary droplets did not occur. Also, the size segregation effect was much less pronounced than the effect that occurred in the real spray. The Dark-Zone was only observed when the maximum initial radial position of the droplets in the production area was of the same order of magnitude as the jet radius. When, a 5-μm radial displacement was used, then the spray profile was identical to the spray profile obtained for a radial displacement of 1 μm. The Dark-Zone still occurred. The radial position of the Dark-Zone at 1 cm from the tip of the jet was unchanged. The radial position of the Dark-Zone was again found at 1.8 mm from the center of the spray.

The measurements of a spray for ethylene glycol at a flow rate 2.78 $10^9$ m$^3$ s$^{-1}$ did not show a Dark-Zone, and the size distribution was much wider than at a flow rate of 1.39 $10^9$ m$^3$ s$^{-1}$. This flow rate was clearly above the maximum flow rate for jet break-up in the varicose jet break-up regime.

The simulations also proved that the Dark-Zone only occurred when the initial radial velocities were small compared to the axial velocity. When the initial axial velocity was taken equal to 22 m s$^{-1}$ instead of 25 m s$^{-1}$, then the radial dispersion of the droplets increased by a few percent. The velocity of the main droplets in the measurement position became 1 m s$^{-1}$ less, and the velocity of the secondary droplets and satellites remained unchanged. This results in a flatter velocity profile as function of the droplet size. This can be compensated by increasing the value of n of equation (5.3) to 1.6 or 1.7.

During simulations it was assumed that the small mode of the bimodal size distribution for ethylene glycol came from secondary droplet production. In that case a main droplet was followed by a secondary droplet. This assumption did not have much influence on the results, because the number of secondary droplets was approximately the same as the number of main droplets. This means that even when droplets are generated randomly from a bimodal size distribution, only a small number of secondary droplets will be generated before a main droplet.
will be produced.

5.4.8 The Charge to Diameter Relation in a Spray

Taking all the mentioned effects in account, the value of \( n \) of equation (5.3) could be established. The value that fitted best to the measurements was \( n=1.5 \). Due to the discussed effects like acceleration of the air in the spray center, the approximated position of the droplet production area, the approximation of the initial velocity of the droplets, and the possible inaccuracy of the size measurements, the value of the power of this relation is estimated. However, taking extreme assumptions showed that the absolute accuracy of the power, 1.5, is lower than 0.4.

This result confirms the value measured by Tang and Gomez (1994). This also confirms that the presented model is able to predict the droplet charge at the edge of the spray, the electric field inside the spray, and the droplet velocity at the edge of the spray. Neglecting the air velocity is not valid inside the spray. The air velocity has to be taken into account to be able to predict the droplet velocity in the spray center.

The value of \( n=1.5 \) shows that the surface charge density on the small droplets is larger than on the large droplets. It also means that for all droplets, the ratio of the electric normal surface stress over the surface tension stress is constant. This ratio is equal to

\[
\frac{q^2}{2\varepsilon_0(\pi d_d^2)^2} \frac{d_d}{4\gamma}
\]

(5.6)

where, \( \gamma \) is the surface tension of the liquid [N m\(^{-1}\)].

5.5 CONCLUSIONS

Profiles, where the droplet velocity is given as function of the droplet diameter in one measurement position, require highly accurate size measurements for aerosols with narrow size distributions. When a perfect monodisperse spray is measured, the measurement tool will measure a size distribution with a certain standard deviation. In order to obtain reliable velocity-size profiles, this standard deviation should be much smaller than the standard deviation of the size distribution of the aerosol at the measurement position. If this is not the case, then the velocity profile measured as function of the droplet diameter will be flatter than the real profile, and only the average velocity of the modal size will be a reliable result.

Due to the strong gradient of the electric field in an area within a few centimeters from the droplet production area, electric force and drag force are not in balance, so the droplet inertia has to be taken into account when calculating the droplet velocity or the droplet charge.

Comparison between measurements and simulations shows the already known droplet size segregation effect in the spray. The model predicts well the droplet size as function of the radial position, the concentration profile in the spray, and the dispersion of the main droplets.
However, it underestimated the radial dispersion of the secondary droplets and satellites in the spray. Neglecting the dispersion due to flow of the surrounding air is thought to be the main reason for this underestimation.

The maximum fluctuation in the radial droplet seeding position in the droplet production area does not have an effect on the fact that the size segregation effect occurs in the just produced spray. However, it has its influence on the droplet concentration and average droplet size profiles obtained as functions of the radial position. When, the maximum initial radial displacement was smaller than the jet radius, then this maximum radial displacement did not have any influence on the spray profiles. The low droplet concentration at the boundary between the areas with main droplets and secondary droplets, which we called the Dark-Zone, indicates that the change in initial radial position between two droplets is smaller than the jet radius. This was confirmed by pictures of a liquid jet in another experiment. The initial radial velocity of the droplets has to be small compared to the axial velocity.

The model confirmed that the mutual electric interaction of charged droplets, and differences in inertia are found to be the main reason for the size segregation effect. Charged droplets exert equal, but oppositely directed, electric forces on each other. The droplet mass determines the change in droplet velocity due to this electric force, and the radial component of this force, make that small droplets will be found at the edge of the spray and the large droplets will be found in the spray center.

The Dark-Zone, the low droplet concentration at the boundary between main droplets and secondary droplets, only occurs when a bimodal size distribution is produced. The occurrence of a Dark-Zone is an indication that a bimodal spray is produced.

Calculations showed that the electrical interaction between charged droplets influences also the axial velocity of these droplets. Fast highly charged droplets are slowed down by slower lower charged droplets. However, the effect is limited.

When all the mentioned effects are taken into account, a relation between droplet size and droplet charge could be established. This relation was found to be equal to

\[ q \sim d_d^{1.5} \]

This result confirms the value measured by Tang and Gomez (1994). This value of 1.5 means that the surface charge density is higher on the smaller droplets than on the larger droplets. The ratio of the normal electric surface stress due to the droplet charge over the surface tension stress in the droplets is constant. Whether this relation is valid for all liquids that are sprayed in the Cone-Jet mode has to be investigated. These measurements were performed close to the maximum flow rate for the varicose jet break-up. So, more research at lower flow rates has to be carried out.

The agreement between this result and the value of Tang confirms that the presented model is able to predict the droplet charge, the electric field inside the spray, and the droplet velocity at the edge of the spray. Neglecting the air velocity is not valid inside the spray. The air velocity has to be taken into account to be able to predict the droplet velocity in the spray center.
Comparison between measurements and calculations showed that fast ethylene glycol droplets, 25 m s\(^{-1}\), can accelerate the surrounding gas. In the spray center, at one centimeter from the droplet production area, gas velocities can reach up to approximately 8 m s\(^{-1}\).

The lowest air velocities in the spray can be obtained in electrode configurations with long nozzles with a small diameter. The electric field in the gap mainly depends on the electrode configuration that is used. A long nozzle with a small diameter will create an electric field with a strong divergence. As a result, the potential drop in the gap is lower, while the electric field strength at the jet surface is of the same order of magnitude as for other nozzles.

These air velocities can also be reduced by reducing the current through the liquid cone. Liquids, which are sprayed at flow rates close to their minimum flow rate, will produce a lower current than liquids, which are sprayed at higher flow rates.
CHAPTER 6

JET BREAK-UP

The jet break-up mechanism has been investigated with a high resolution camera. A model is presented, which is able to predict the droplet size, the velocity at jet break-up, and the wavelength at jet break-up.

It was found that the jet break-up mechanism depends on the ratio of the electric normal stress over the surface tension stress. At a low value of this ratio, the jet breaks up due to varicose instabilities. The number of secondary droplets is much lower than the number of main droplets. With increasing flow rate, the current increases, the stress ratio increases, and the number of secondary droplets and satellites increases. A threshold value of the stress ratio on the jet was found, above which the jet starts to whip. In order to reduce the number of secondary droplets, the current through the liquid cone should be reduced. It is shown, that viscosity, surface charge, and the acceleration of the jet, have to be taken into account in the jet break-up process. The main droplet diameter for varicose jet break-up scales with the flow rate as \( d_f \sim Q^{0.48} \). When, the jet breaks up in the whipping regime, then the main droplet size scales as \( d_f \sim Q^{0.33} \). In the whipping jet break-up regime, the stress ratio on the main droplets is between 0.7 and 0.9.

6.1 INTRODUCTION

In the Cone-Jet mode, the tangential electric field accelerates the surface charge toward the cone apex. Due to this acceleration, a jet with a high charge density occurs at the cone apex. This jet breaks up into a number of primary or main droplets and a number of secondary droplets and satellites, Cloupeau and Prunet Foch (1989), Gomez and Tang (1994), Chen et al.(1995). In order to be able to simulate the jet break-up processes in the Cone-Jet mode, the acceleration of the jet, the shape of the jet, and the surface charge density on the jet surface have to be known. The Cone-Shape Model described in Chapter 3 is able to calculate these values.

The size distribution produced in Electrohydrodynamic Atomization in the Cone-Jet mode depends on the diameter of the jet, and on the break-up of this jet into droplets. Every liquid has a minimum flow rate, below which a stable Cone-Jet mode cannot exist. At this minimum flow rate the jet breaks up due to axisymmetric instabilities. These instabilities are also
called varicose instabilities. At higher flow rates, the current through the liquid cone increases. With increasing current, the surface charge on the jet increases. Above a certain surface charge the jet break-up will also be influenced by lateral or azimuthal instabilities of the jet. These instabilities are also called kink instabilities. When the influence of these kink instabilities increases, then the size distribution of main droplets becomes wider, Cloupeau and Prunet Foch (1989). These kink instabilities are sometimes also described as a whipping motion of the jet, Gomez and Tang (1991). Gomez noticed that the occurrence of the whipping motion depended on the applied potential difference.

The break-up of a jet has been studied by many people. For instance, Rayleigh (1878) has presented a theory for small varicose instabilities on a liquid jet. In his calculations, the electric stresses were equal to zero, and the velocity of the jet was constant. This theory is able to predict the wavelength of the fastest growing varicose instability. Weber (1931) included the liquid viscosity into the theory. The higher the viscosity, the longer the dominant wavelength. The fastest growing wavelength is often called the dominant wavelength, because the jet always breaks up through instabilities with a wavelength close to the fastest growing wavelength. Yuen (1968) included nonlinear effects in the jet break-up theory. His theory showed that a longer dominant wavelength means a relatively larger volume of secondary droplets and satellites. Rutland Jameson (1970) used Yuen’s theory to estimate the size of these droplets as function of the wavelength. In Electrohydrodynamic Atomization, also electric stresses play an important role. Among others, Melcher (1963), Parkin (1973), and Turnbull (1992) have studied the influence of charge on the jet break-up. Turnbull (1992) assumed an insulating liquid, where Melcher assumed the jet to be a perfect conductor. Parkin (1973) measured the growth rate of perturbations on a water jet, which could be assumed to be a perfect conductor. His results showed that the results of Melcher were not far from reality. However, due to scattering in the data, due to low electric stresses, and due to relatively long wavelengths, only a qualitative confirmation could be given of Melcher’s results.

Cloupeau and Prunet Foch (1989), and Gomez and Tang (1991) both claimed that the break-up of the jet in the Cone-Jet mode was not influenced by surface charge. Gomez found theoretical confirmation in the results of Schneider et al. (1967) and Neukermans (1973). Cloupeau and Gomez both measured the ratio of the droplet diameter over the jet diameter. This ratio was for low viscosity liquids within the measurement accuracy equal to 1.89. This value of 1.89 follows from Rayleigh’s theory of an uncharged jet with a constant liquid velocity.

This chapter concerns the jet break-up in the Cone-Jet mode. Jet break-up mechanisms were investigated with a high resolution camera with a long distance microscopic lens. A description will be given of the regimes of the jet break-up mechanism, and their influence on the droplet size distribution was investigated. By choosing the right experimental conditions, the number of secondary droplets and satellites could be reduced considerably. This enhances a lot the monodispersity of the spray. Another, interesting transition is the transition between the jet break-up due to axisymmetric or varicose instabilities into the jet break-up due to the lateral or kink instabilities. This transition brings about an increase in the width of the size distribution of the main droplets. A relation will be given, which makes it possible to calculate at which liquid
flow rate the transition between varicose break-up and kink break-up occurs.

Measurement results will be compared to the simulation results of a model, which consists of two parts. The first part is the already presented Cone-Shape model, Chapter 3. The second part is a mathematical physical model, describing the jet break-up due to axisymmetric or varicose instabilities. The results clearly show that the jet break-up in the Cone-Jet mode cannot be described with the simple assumptions of Rayleigh’s theory. The influence of the flow rate on the main droplet size distribution is investigated. Also, another numerical model has been used to calculate the diameter and charge of the secondary droplets as functions of the dimensionless wave number at jet break-up, and as functions of the liquid viscosity.

6.2 JET BREAK-UP THEORY AND MODELING

In Electrohydrodynamic Atomization in the Cone-Jet mode, a highly charged jet emerges at the apex of the liquid cone. Due to its charge, the liquid is still accelerating, while breaking up into droplets. This jet break-up produces often a bimodal size distribution. In order to model the jet break-up process with mathematical relations, a number of simplifications are made. When, in a cylindrical coordinate system, a jet is moving at a constant velocity, and when only harmonic perturbations are present on the jet surface, then this surface can be described by

\[ r_s = r_{jet} + \alpha_0 e^{(\omega t + jm\theta - jkz)} \]  \hspace{1cm} (6.1)

where, \( r \) is the radial component [m], \( \theta \) is the angular component [rad], \( z \) is the axial component [m], \( r_s \) is the radius of the surface [m], \( r_{jet} \) is the radius of the unperturbed jet, \( \omega \) is the growth rate of the perturbation [s\(^{-1}\)], \( t \) is time [s], \( \alpha_0 \) is the amplitude of the perturbation at \( t=0 \) [m], \( m \) is a constant, and \( k \) is the wave number of the perturbation [m\(^{-1}\)] equal to

\[ k = \frac{2\pi}{\lambda} \]  \hspace{1cm} (6.2)

where, \( \lambda \) is the wavelength of the perturbation [m]. Depending on the value of \( m \), three modes of jet break-up can be distinguished. Figure 6.1 shows these three jet break-up axisymmetric varicose break-up, the lateral modes. If the value of \( m \) is equal to zero, then kink break-up, and the ramified jet break-up.

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the jet break-up is independent of the angular component $\theta$. This kind of axisymmetric instabilities are also called varicose instabilities.

If the value of $m$ is equal to 1, then the radius of the jet depends on angle $\theta$. For half of the values of $0$, $r_e$ is larger than $r_{jet}$, and for the other half $r_e$ is smaller than $r_{jet}$. This represents the whipping motion of the jet. This kind of lateral instability is also called a kink instability. This mode can only occur, if the jet is slowed down by external forces like the drag force of the surrounding air, or when the jet is charged.

If the value of $m$ is equal to 2, then the jet is no longer circular. This mode can only occur if the jet is highly charged. If the surface charge is above a certain threshold value, then the electric stresses can overcome the surface tension. These electric stresses will transform the shape of the jet. The ramified jet is an extreme case of this kind of instability.

Weber (1931) included viscosity in Rayleigh's theory. This resulted in the following relation

$$\omega' = \frac{\mu k^2 r_{jet}^{1.5} (24 + k^2 r_{jet}^{-2})}{\gamma^{0.5} \rho^{0.5} (8 + k^2 r_{jet}^{-2})} \omega = \frac{4 k^2 r_{jet}^{-2}}{(8 + k^2 r_{jet}^{-2})} (1 - k^2 r_{jet}^{-2}) \tag{6.3}$$

where, $\mu$ is absolute liquid viscosity [Pa s], $\rho$ is liquid density [kg m$^{-3}$], and where, the normalized growth rate is equal to

$$\omega' = \frac{\omega r_{jet}^{1.5} \rho^{0.5}}{\gamma^{0.5}} \tag{6.4}$$

Melcher (1963) derived an equation, for non viscous liquids, that takes the influence of surface charge into account. This resulted in

$$\omega' = kr_{jet} I_m'(kr_{jet}) I_m(2kr_{jet}) \left[ \frac{I_m^2(2kr_{jet}) - I_{m-2}^2(2kr_{jet})}{I_m(2kr_{jet}) - I_{m-2}(2kr_{jet})} \right] (1 + 2R_{E}) \tag{6.5}$$

where, $I_m$ are Bessel functions.

When the theories of Weber, and Melcher are combined then the following derivation can be made. The flow pattern inside a free jet of incompressible liquid can be described by the Navier-Stokes equation.
\[ \rho \frac{\partial \vec{u}}{\partial t} + \rho (\vec{u} \cdot \nabla) \vec{u} = -\nabla p + \nabla 2\mu e_{ij} + \vec{f} \quad (6.6) \]

where, \( \rho \) is density \([\text{kg} \, \text{m}^{-3}]\), \( p \) is pressure \([\text{Pa}]\), \( \vec{u} \) is liquid velocity vector, \( \mu \) is absolute viscosity \([\text{Pa} \, \text{s}]\), \( \vec{f} \) is the summation of the external forces per unit of volume \([\text{N} \, \text{m}^{-3}]\), and

\[ e_{ij} = \frac{1}{2} \left( \frac{\partial u_i}{\partial x_j} + \frac{\partial u_j}{\partial x_i} \right) \quad (6.7) \]

When considering a coordinate system moving with the jet, and when assuming small velocity disturbances, then the term \( \rho (\vec{u} \cdot \nabla) \vec{u} \) can be neglected. For thin jets with velocities of about 20 \( \text{m s}^{-1} \) the contribution of gravity can also be neglected. This makes, together with the assumption of the jet being a perfect conductor, that no external forces are present inside the liquid. Also it is assumed that the shear stress at the free liquid surface is equal to zero, Batchelor (1967). This means that for \( i \neq j \) at the liquid surface \( e_{ij}=0 \). As a result, it can be concluded that the radial integration of shear forces in the axial direction is equal to zero. So, these shear forces do not contribute to the average axial velocity. Thus, the shear stress in the axial direction can be assumed to be zero everywhere in the liquid jet. When, we only consider the varicose instabilities then the value of \( m \) is equal to zero and all relations become independent of \( \theta \). When, it is assumed that \( u_r \) depends linearly on \( r \), and \( u_z \) is independent of \( r \), and with equation (6.7), then equation (6.3) becomes

\[ \rho \frac{\partial u_r}{\partial t} = -\frac{\partial p}{\partial r} + \mu \left( \frac{\partial^2 u_r}{\partial z^2} \right) \quad (6.8) \]

\[ \rho \frac{\partial u_z}{\partial t} = -\frac{\partial p}{\partial z} + 2\mu \left( \frac{\partial^2 u_z}{\partial z^2} \right) \quad (6.9) \]

and, the pressure disturbances and velocity disturbances inside the liquid can be described by

\[ p = p^0 e^{(\omega t - k_z)}, \quad u_r = u_r^0 e^{(\omega t - k_z)} \quad \text{with} \quad u_r^0 = \frac{\alpha_0 \omega r}{r_{\text{jet}}}, \quad u_z = u_z^0 e^{(\omega t - k_z)} \quad (6.10) \]

Using the assumption that \( u_r \) depends linearly on \( r \), then the continuity equation, \( \nabla \cdot \vec{u} = 0 \) yields
\[ \frac{\partial u_r}{\partial r} + \frac{1}{2} \frac{\partial u_z}{\partial z} = 0 \]  

(6.11)

and

\[ u_z'' = \frac{-j2\omega \alpha_0}{kr_{jet}} \]  

(6.12)

When we consider a grounded cylinder, then the electric field of an undisturbed jet in the center of this cylinder can be described with

\[ E_r = \frac{\Delta V}{r_{jet} \ln \left( \frac{r_{cyl}}{r_{jet}} \right)} \]  

(6.13)

where, \( r_{cyl} \) is the radius of the cylinder, \( E \) is the electric field strength \([\text{V m}^{-1}]\), and \( \Delta V \) is the potential difference \([\text{V}]\). Since an electric field is always rotation free, \( \nabla \times \vec{E} = 0 \), and when it is assumed that the disturbance of the electric field strength is equal to

\[ \vec{E} = \vec{E}''(r) \ e^{(rot \cdot j\omega \cdot \cdot \cdot k\theta)} \]  

(6.14)

then

\[ E_r'' = \frac{j}{k} \frac{dE_z''}{dr} \]  

(6.15)

\[ E_\theta'' = -\frac{m}{k} \frac{E_z''}{r} \]  

(6.16)

If outside the jet, there is no space charge, then

\[ \nabla \cdot \vec{E} = 0 \]  

(6.17)

When equations (6.15) and (6.16) are substituted, then equation (6.17) yields
\[
\frac{r^2}{dr^2} \frac{dE_z^{\prime\prime}}{dr} + r \frac{dE_z^{\prime\prime}}{dr} - (m^2 + k^2 r^2)E_z^{\prime\prime} = 0
\]
(6.18)

The normal vector of a surface can be calculated using

\[
\vec{n} = \frac{\nabla r_i}{(\nabla r_i \cdot \nabla r_i)^{1/2}}
\]
(6.19)

With \( u_r = \partial \phi / \partial r \), this yields for the jet

\[
n_r = 1 \quad n_\theta = \frac{jm}{\omega_{jet}} u_r^{\prime\prime} \quad n_z = -\frac{jk}{\omega} u_r^{\prime\prime}
\]
(6.20)

and for the cylinder

\[
n_r = 1 \quad n_\theta = 0 \quad n_z = 0
\]
(6.21)

At the surface just outside a perfect conductor the electric field must be perpendicular to the surface of the conductor. This yields

\[
\vec{n} \times \vec{E} = 0
\]
(6.22)

When using again a first order approximation, these normal vectors give the following boundary conditions of the electric field

\[
E_z^{\prime\prime}(r=r_{cyl}) = 0 \quad E_z^{\prime\prime}(r=r_{jet}) = n_z^{\prime\prime} \frac{\nabla}{r_{jet}}
\]
(6.23)

where, \( \nabla \Delta \phi / \ln(r_{cyl}/r_{jet}) \). The solution of equation (6.18) with these boundary conditions is equal to

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\[ E''_z = \frac{j\kappa u''_r}{\omega r_{jet}} \left( \lim_{n \to m} \frac{I_n(kr_{cyl})}{I_n(kr_{jet})} - \frac{I_n(kr_{cyl})^2}{I_n(kr_{jet})^2} \right) \]

(6.24)

Where, \( I_n \) are Bessel functions. If the cylinder radius \( r_{cyl} \to \infty \) then the influence of this electrode should disappear and the electric field can be approximated with

\[ E''_z = \frac{j\kappa u''_r}{\omega r_{jet}} \left( \lim_{n \to m} \frac{I_n(kr) - I_n(kr_{jet})}{I_n(kr_{jet})^2} \right) \]

(6.25)

The pressure inside the liquid is determined by the stresses in the liquid surface and by the flow profile inside the liquid. As already discussed in section 3.2.1, the viscous normal stress in the liquid surface has its influence on the pressure. In case of a liquid jet with small disturbances with wavelengths normally about ten times larger than the jet radius, the normal stress at the liquid surface is almost equal to \( 2\mu \partial u / \partial r \). Using the assumption that \( u \), depends linearly on \( r \) and using equation (6.11), then the viscous stress becomes \( -\mu \partial u / \partial z \).

The surface tension as given in equations (3.4) and (3.5), can be approximated by a first order approximation. This results in the following relation

\[ \frac{1}{r_1} + \frac{1}{r_2} = \frac{1}{r_{jet}} - \frac{\alpha_0 e^{(\alpha_0 + jkz)}}{r_{jet}^2} - \frac{\partial^2 \alpha_0 e^{(\alpha_0 + jkz)}}{\partial z^2} \]

(6.26)

For a small disturbance, the electric field can be approximated with

\[ E_r = u(\frac{1}{r_{jet}} - \frac{\alpha_0 e^{(\alpha_0 + jkz)}}{r_{jet}^2}) + E''_r \]

(6.27)

With the electric stress in the liquid surface being equal to \( 0.5\varepsilon_0 E_r^2 \), The pressure disturbance [Pa] at the liquid surface is then equal to

\[ p''_{surf} = \gamma(k^2 - \frac{1}{r_{jet}^2})\alpha_0 + \frac{\varepsilon_0 u^2}{r_{jet}^3}a_0 - \frac{\varepsilon_0 u}{r_{jet}}E''_r - \mu \frac{\partial u''}{\partial z} \]

(6.28)
Using equations (6.8), (6.10), (6.12), and averaging over the radial coordinate yields

$$
\frac{\partial p}{\partial l} = \frac{1}{4} \rho a_0 \omega^2 r_{jet}^2 + \frac{1}{4} \mu k^2 a_0 \omega r_{jet}^2 + p_{surf}''
$$

(6.29)

Substituting this result with m=0 into equation (6.9) using equations (6.10), (6.12), (6.15), and (6.25) yields

$$
\frac{\omega^2 \rho r_{jet}^3}{\gamma} + \frac{\mu k^2 \omega r_{jet}^3}{\gamma} \left( \frac{24 + k^2 r_{jet}^2}{8 + k^2 r_{jet}^2} \right)
$$

(6.30)

$$
\frac{4k^2 r_{jet}^2}{\left(8 + k^2 r_{jet}^2\right)} \left( 1 - k^2 r_{jet}^2 - \frac{\varepsilon_0 c_0^2}{\gamma r_{jet}} \right) \left( 1 + k r_{jet} \left( \lim_{m \to 0} \frac{I''_m(k r_{jet}) - I''_{-m}(k r_{jet})}{I''_m(k r_{jet}) - I''_{-m}(k r_{jet})} \right) \right)
$$

This equation can be made dimensionless yielding

$$
\frac{24 + k^2 r_{jet}^2}{\left(\gamma \rho r_{jet}\right)^{1/2}} \frac{\omega'}{\omega} = \frac{4k^2 r_{jet}^2}{\left(8 + k^2 r_{jet}^2\right)} \left( 1 - k^2 r_{jet}^2 - 2R_{Es} \left( 1 + k r_{jet} \left( \lim_{m \to 0} \frac{I''_m(k r_{jet}) - I''_{-m}(k r_{jet})}{I''_m(k r_{jet}) - I''_{-m}(k r_{jet})} \right) \right) \right)
$$

(6.31)

The value of this normalized growth rate depends on three dimensionless numbers

$$
Oh = \frac{h}{\left(\gamma \rho d_{jet}\right)^{1/2}} \frac{k r_{jet}}{2 \gamma \varepsilon_0}, \quad R_{Es} = \frac{\sigma^2 r_{jet}^4}{2 \gamma \varepsilon_0}
$$

(6.32)

The first dimensionless number is the Ohnesorge number (Oh = We^{1/2}/Re). This number describes a ratio of the loss of energy due to viscosity over the potential energy of the jet due to surface tension. The thinner the jet, or the higher the viscosity, the larger the influence of viscous forces. The second dimensionless number, the dimensionless wave number, describes the ratio of the jet radius over the wavelength times 2π. The third number R_{Es} describes the ratio of the
electric stress over the surface tension stress, where, \( \sigma \) is surface charge on the jet \([\text{C} \text{m}^{-2}]\). The value of the Bessel functions can be calculated with the following relations

\[
I_n(x) = \sum_{k=0}^{\infty} \frac{1}{k! \Gamma(n+k+1)} \frac{x^{n+2k}}{2^n}
\]

(6.33)

where, \( \Gamma \) is the gamma function equal to

\[
\Gamma(x+1) = \lim_{k \to \infty} \frac{k! x! k^x}{(k+x)!}
\]

(6.34)

For larger values of \( x \) the gamma function can be approximated with Stirling’s asymptotic series

\[
\Gamma(x) = x^x e^{-x} \left( \frac{2\pi}{x} \right)^{1/2} \left(1 + \frac{1}{12x} + \frac{1}{288x^2} - \frac{139}{51840x^3} - \frac{571}{2488320x^4} \right) + \ldots
\]

(6.35)

The derivative of these Bessel functions can be calculated using the relation

\[
I_n'(x) = \frac{1}{2} \left( I_{n+1}(x) + I_{n-1}(x) \right)
\]

(6.36)

Using equations (6.31), the normalized growth rate \( \omega' \) can be calculated as function of the dimensionless numbers of equation (6.32). Figures 6.2 to 6.7 show the results of these calculations. From these figures it can be concluded, that the dominant wavelength of the axisymmetric \( m=0 \) mode shifts toward the short wavelengths with increasing electric stress. The dominant wavelength shifts toward the long wavelengths with increasing viscosity. Below a dimensionless wave number of 0.58, the growth rate of the varicose \( m=0 \) mode decreases with increasing electric stress. For perturbations with wave numbers larger than 0.58, the growth rate increases with increasing electric stress. Long waves are stabilized by the electric stress. The asymmetric \( m=1 \) and \( m=2 \) modes have negative growth rates, if \( \Re_s = 0 \). This means that the jet is stable for these instabilities. The growth rate of the \( m=2 \) mode only becomes positive, if the electric stress is larger than about 1.5 times the surface tension. The growth rate of the kink \( m=1 \) mode becomes positive when the electric stress is larger than about 0.3 times the surface tension.

Viscosity does not change the range of wave numbers where the jet breaks-up into droplets. With increasing viscosity or with a decreasing jet diameter, the Ohnesorge number increases. With increasing Ohnesorge number, the normalized growth rates of all waves decrease. However, the longer waves are less affected than the shorter ones.
Figure 6.2 The normalized growth rate for varicose jet break-up as function of the dimensionless wave number and the stress ratio $R_{Es}$.

Figure 6.3 The normalized growth rate for kink jet break-up as function of the dimensionless wave number and the stress ratio $R_{Es}$.

Figure 6.4 The normalized growth rate for the ramified jet break-up as function of the dimensionless wave number and the stress ratio $R_{Es}$.

Figure 6.5 The normalized growth rate for varicose jet break-up as function of the dimensionless wave number and the stress ratio $R_{Es}$, with Oh=0.18.
6.2.1 The Influence of the Surrounding Air

At relatively high jet velocities, the influence of the surrounding gas on the jet break-up can become important. Among others, Levich (1962) derived an equation to estimate this influence. He performed a stability analysis similar to the one of Melcher. The difference is that he considered a case without electric stresses and included the influence of the surrounding gas. He solved the Navier-Stokes equation for both the liquid jet and the surrounding air. As boundary conditions, he assumed at the liquid-air interface, that the velocity of the liquid, in the radial as well as in the axial direction, is equal to the velocity in the gas. Far away from the jet, he assumed the radial velocity to be equal to zero, and the axial velocity of the gas to be equal to some constant value. For a coordinate system moving with the jet, the axial velocity at the jet surface is equal to zero. Far away from the jet, the axial gas velocity is taken equal to minus the jet velocity.

For short wavelengths and low viscosities simplifications lead to the following relation

$$\omega = \left( \frac{\rho k^2 u_k^2 - \gamma k^3}{\rho} \right)^{\frac{1}{2}}$$

(6.37)

The instabilities will grow, if the value of $\omega$ is real. This yields
\[ k \leq \frac{\rho_g u_g^2}{\gamma} \]  

(6.38)

For typical values of a jet in the Cone-Jet mode \( r_{jet} = 5 \times 10^{-6} \text{ m}, \ u_{jet}=20 \text{ m s}^{-1}, \ \gamma=0.03 \text{ N m}^{-1}, \) and \( \rho_g = 1.2 \text{ kg m}^{-3}, \) the fastest growing perturbation has a normalized wave number equal to 0.08. Such a low normalized wave number means a long wave length instead of a short wave as was assumed. Consequently, this jet is stable for short waves induced by the surrounding gas.

For long wave lengths and low viscosities, the equation for the growth rate of the perturbation becomes

\[ \omega^2 + \frac{3\mu k^2 - \omega}{\rho} = \frac{\gamma k^2 r_{jet}^2}{2 \rho r_{jet}^3} (1 - k^2 r_{jet}^2) - \frac{\rho_g k^4 r_{jet}^2 u_g^2 \ln(\frac{k r_{jet}}{2})}{2 \rho} \]  

(6.39)

In this case, the jet is not stable. For the same case as above, and for a normalized wavelength of 0.71, the influence of the air on the growth rate is about 4 percent. Only for liquid jets with higher liquid velocities or for low conductive liquids with their relatively large jet radii, the influence of the air has to be taken into account. However, for the liquid jets in this thesis, the influence of the gas can be neglected.

6.2.2 Modeling of the Jet Break-Up due to Varicose m=0 Instabilities in the Cone-Jet Mode

Two models have been developed to calculate the droplet size and charge produced by Electrohydrodynamic Atomization in the Cone-jet mode. The first model is a mathematical model based on equation (6.31). This model calculates the wave number and velocity at jet break-up, and it calculates a droplet size. This model is not able to calculate the size of secondary droplets and satellites. The second model, a numerical model, is able to calculate the size and charge of main and secondary droplets. This model does not take the acceleration of the liquid into account but assumes a jet with a constant velocity.

The Mathematical Jet Break-Up Model

The Cone-Shape model presented in Chapter 3 is used to calculate the cone and jet shape, the electric fields along the jet, and the surface charge on the jet. In the numerical physical Cone-Shape model a liquid cone shape is assumed. Then, the electric fields in and outside the liquid cone are calculated using Gauss' law. A liquid velocity profile inside the liquid is estimated using a simplified Navier-Stokes equation. The surface charge as function of the position on the surface
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is calculated from the current balance at this surface. Since, the velocity profile, and the surface charge have an influence on the electric fields, this process is repeated until the surface charge does not change anymore. Then, a one dimensional Navier-Stokes equation is solved to calculate a new cone shape. In order to be able to calculate the new shape, the cone and jet are discretized along the axial axis. This new cone shape is used as the input for the model. This is repeated until the output shape is equal to the input shape.

In the Mathematical Jet Break-Up model, an initial perturbation was assumed. The initial amplitude and wave number of this perturbation, and the position on the jet were fixed. Then, the local jet radius, and the local surface charge, were used to calculate the growth rate with equation (6.31). A flat liquid velocity profile in the jet was assumed. When the radius of the jet is known, then the velocity can be calculated. From the distance between the grid points on the cone calculated by the Cone-Shape model, a time can be calculated. This time can be used to calculate a new amplitude of the perturbation. When, the increase of the wavelength with increasing liquid velocity is taken into account, then a new growth rate can be calculated at the new grid position. This can be repeated until the amplitude of the perturbation is equal to the jet radius. The starting wavelength that gives the shortest jet was chosen as the dominant wavelength.

The Numerical Jet Break-Up Model

In this model a one dimensional Navier-Stokes equation is used to calculate the velocity and the change in shape of the jet. It is assumed that the jet is not accelerated. In this model a number of simplifications are made. The radial liquid velocity profile is assumed to be flat in the jet. This makes the calculation of the currents inside the jet much more easy. The results of the Cone-Shape model proved that this assumption was valid for the experimental conditions presented in this chapter. It was assumed that the charge is relaxed to the surface of the jet. The electrical conduction in the liquid is limited in the axial direction, due to the small jet radius. However, the surface in the radial direction is much larger than the surface in the axial direction. The current to the jet surface is in the same order of magnitude as the current in the axial direction. So, the current density toward the jet surface is much lower than the current density in the axial direction. If this is the case, then the electric field strength in the radial direction is also much smaller than the field in the axial direction. This situation can only be reached, if most of the free charge is relaxed to the liquid surface. That is why, the pressure difference between air and liquid, caused by the normal electric stress, can be approximated with

$$\Delta p_{En} = -\frac{1}{2} \frac{\sigma^2}{\epsilon_0}$$

(6.40)

The influence of gravity and the polarization stress are much smaller than the influence of the kinetic energy of the jet, and much smaller than the influence of the surface tension, and the

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electric stresses in the jet surface. That is why, the influence of gravity and the polarization stress can be neglected.

Using these assumptions and taking the viscous stress in the liquid surface into account, the one dimensional Navier-Stokes equation becomes

$$\rho \frac{\partial u_z}{\partial t} = \frac{\partial \left( \frac{\sigma^2}{2\epsilon_0} - \Delta P_s + 3\mu \frac{\partial u_z}{\partial z} - \frac{1}{2} \rho u_z^2 \right)}{\partial z} + \frac{2}{r} \sigma E_r \quad (6.41)$$

where, $E_r$ is tangential electric field strength [V m$^{-1}$], and the surface tension stress is according to Kingham (1984) equal to

$$\Delta P_s = \gamma \left( \frac{1}{r \left( 1 + (dr/dz)^2 \right)^{1/2}} - \frac{d^2r/dz^2}{(1 + (dr/dz)^2)^{3/2}} \right) \quad (6.42)$$

However, most of the time, the value of $dr/dz$ in the jet is very small. Thus, the tangential electric field strength can be replaced by the axial electric field strength. The right-hand side of this equation can be solved numerically by discretization of the jet along the $z$ axis. The left-hand side of this equation can be calculated using an explicit method of taking small time steps, where the present situation is used to calculate a new velocity. The new surface charge $\sigma_j$ can be calculated from the following relations

$$\frac{\Delta q_j}{\Delta t} = - \frac{\Delta (q_j u_z + \pi r_{sj}^2 K E_{sj})}{\Delta z} \quad q_j = 2\pi r_{sj} \sigma_j \Delta z (1 + (dr_{sj}/dz)^2)^{1/2} \quad (6.43)$$

where, index $j$ refers to a discretization point, $q$ to charge [C], $u_z$ is liquid velocity [m s$^{-1}$], $\Delta z$ is discretization distance [m], $E_{zj}$ is the electric field strength in the axial direction [V m$^{-1}$]. The laws of conservation of charge and mass were fulfilled at all times.

In order to solve equation (6.41), the boundary conditions at the beginning and at the end of the jet, and the situation at the beginning of the simulations have to be known. At the beginning of the calculations, the radius, and surface charge of the jet were fixed to a certain value. Half way down the jet, a small perturbation was applied to the jet-radius. At both ends of the considered jet, a zero flux condition was created for the liquid flow as well as for the electric field.
In this model, the axial electric field strength is on average equal to zero. However, the local axial electric field strongly depends on the shape of the jet, and on the conductivity of the liquid. This local electric field of the charged jet can be simulated with a number of charged rings. The radius of such a ring is equal to the jet radius at that position. The charge on the ring can be calculated with equation (6.43). Figure 6.8 shows a schematic representation of the calculation of the axial electric field strength. The field is calculated half way between two rings. The total local axial electric field strength requires integration over angular component \( \theta \), and summation over a number of rings on both sides of the jet position \( j+0.5 \). Even in a perturbed jet, most of the time, the value of \( dr_{\text{jet}}/dz \) is very small. Thus, the tangential electric field is almost equal to the axial electric field. The tangential electric field is not affected by polarization of the liquid. So, the polarization can also be neglected for the calculation of the axial electric field strength. The local axial electric field in the center of the jet in position \( j+0.5 \) is now equal to

\[
E_{z, \text{local}}(0) = \sum_{n=0}^{N} \frac{q_{j,n}(n+0.5)\Delta z}{4\pi \varepsilon_0 ((n-0.5)^2 \Delta z^2 + r_{j,n}^2)^{3/2}} - \frac{q_{j,n+1}(n+0.5)\Delta z}{4\pi \varepsilon_0 ((n+0.5)^2 \Delta z^2 + r_{j,n+1}^2)^{3/2}}
\]  

(6.44)

where, \( n \) is the number of grid points between the charged ring and the jet position \( j+0.5 \). However, the total electric flux through the jet at position \( j+0.5 \) is not equal to \( \pi r_{j+0.5}^2 E_{z, \text{local}}(0) \). The axial electric field strength also depends on the radial position. If \( (n+0.5)\Delta z \gg r_{j,n} \), then this dependance can be neglected. However, at low values of \( k \) this dependance can’t be neglected. In order to compensate for this effect a correction factor is introduced. The axial electric field, caused by one charged ring, is now calculated as function of the radial coordinate in position \( j+0.5 \). This is done by a numerical integration over the angular component \( \theta \). The correction factor \( C \) is then equal to.
\[
C = \frac{\int_0^{r_{j-0.5}} 2\pi r E_{z,\text{local}}(r)dr}{\pi r^{2-j-0.5} E_{z,\text{local}}(0)}
\]  

(6.45)

where, the integral is calculated numerically. Calculations and geometrical symmetry prove that the value of \( C \) only depends on the value of two ratios, which are equal to

\[
\frac{r_{j-n}}{(n-0.5)\Delta z} \quad \frac{r_{j-n}}{r_{j-0.5}}
\]

(6.46)

The first ratio is the radius of the charged ring over the distance between the charged ring and jet position \( j+0.5 \). The second ratio is the radius of the charged ring over the jet radius in position \( j+0.5 \). In order to save computation time, a table of values of \( C \) as function of these two ratios has been calculated. For each charged ring close to jet position \( j+0.5 \), the value of \( C \) can be calculated by interpolation from this table.

6.3 EXPERIMENTAL METHOD

The frequency of droplet production in the Cone-Jet mode is very high (up to a few MHZ). The velocity of the droplets at the moment of production is in the order of tens of meters per second. Thus, in order to see the jet break up, one has to make pictures with an exposure time below 1 microsecond. This requires a powerful illumination source. Moreover, the size of the subject is in the order of tens of microns. This means that an appropriate optical system is needed. The High Speed Spray Imaging System (HSSIS) from Oxford Laser is responding to these constraints.

Figure 6.9 shows a schematic representation of the experimental setup. The HSSIS system is composed of three parts. The first part is the high definition digital camera (Kodak; megaplus camera model ES 1.0), to which is attached a lens (Infinity; Model K2 long-distance microscope), equipped for video mode. The lens allows to focus on objects down to five microns from a distance of approximately 5 centimeters. The second part of the set up is a laser (Oxford laser; HSI 1000 fast illumination system). It is a semiconductor laser, with an 805 nm emission
wavelength. The peak power is 150 watts. The laser and camera are controlled by a control box. This control box sets parameters such as the initial delay, the burst repetition rate, the burst duration, and the separation time between bursts. The third part of the set up is a computer, equipped with software for control of the camera, and a frame grabber. In order to minimize interfering optical effects, the light has to come from the back of the object. For safety reasons a mirror was used to obtain this. The illumination time was below 0.5 µs. The separation between two following pictures was 15 µs. This separation time allows to measure the droplet velocity.

The optical system makes it possible to see particles of a few micrometers. However, perfect focussing on a moving object proved to be difficult. Experiments with polymer particles on a glass slide, proved that a picture, which was not perfectly focussed, resulted in an overestimation of the droplet size with 4 µm. This overestimation was independent of the droplet size. One pixel on the pictures was equal to 1 µm.

The experiments were done, using a classical spraying setup. The nozzle diameter was 8 mm. The nozzle length was 5.1 mm. The radius of the top plate was 25 mm. The distance between the plates was 40 mm for the ethylene glycol experiments, and 30 mm for the other liquids. The potential difference was 17 kV for the ethylene glycol experiment, and 10.8 kV for the other experiments. The distance between the plates proved to have some influence on the behavior of the jet. A n-butanol jet moved slowly around for a 40 mm plate distance, where it was fixed to one position for a plate distance of 30 mm. The liquid was supplied by a syringe pump (Welmed P1000). The minimum flow rate of this pump is 0.1 ml/h. However, at this flow rate, the liquid flow was not steady. This was only the case above a flow rate of 1.0 ml/h.
Four different liquids were used in this study, namely, n-butanol, iso-butanol, 2-butanone and ethylene glycol. The liquid properties, taken from literature, are listed in Table 6.1. The liquid conductivity was increased by adding some LiCl salt to the solutions. Considering the very low quantities added (< 1 mg/100 g), it is assumed that the salt does not alter the other properties of the liquid, Gómez and Tang (1994), Chen et al. (1995). The conductivity was measured with a YSI model 32 conductance meter.

Table 6.1 Liquid properties.

<table>
<thead>
<tr>
<th></th>
<th>Density kg m⁻³</th>
<th>Relative Permittivity</th>
<th>Surface Tension N m⁻¹</th>
<th>Viscosity 10⁻³ Pa s</th>
<th>Conductivity μS m⁻¹</th>
</tr>
</thead>
<tbody>
<tr>
<td>n-Butanol (nB15)</td>
<td>809.8</td>
<td>17.8</td>
<td>0.0246</td>
<td>2.948</td>
<td>15</td>
</tr>
<tr>
<td>n-Butanol + LiCl (nB29)</td>
<td>809.8</td>
<td>17.8</td>
<td>0.0246</td>
<td>2.948</td>
<td>29</td>
</tr>
<tr>
<td>iso-Butanol (iB13)</td>
<td>801.8</td>
<td>18.5</td>
<td>0.0230</td>
<td>4.703</td>
<td>13</td>
</tr>
<tr>
<td>iso-Butanol + LiCl (iB30)</td>
<td>801.8</td>
<td>18.5</td>
<td>0.0230</td>
<td>4.703</td>
<td>30</td>
</tr>
<tr>
<td>2-Butanone (2B45)</td>
<td>805.4</td>
<td>18.5</td>
<td>0.0246</td>
<td>0.417</td>
<td>45</td>
</tr>
<tr>
<td>Ethylene Glycol+LiCl (Eg21)</td>
<td>1108.8</td>
<td>38.0</td>
<td>0.0477</td>
<td>19.9</td>
<td>21</td>
</tr>
<tr>
<td>Ethylene Glycol +LiCl (Eg52)</td>
<td>1108.8</td>
<td>38.0</td>
<td>0.0477</td>
<td>19.9</td>
<td>52</td>
</tr>
</tbody>
</table>

The experimental procedure was as follows. First, a stable cone-jet is established for the liquid sprayed. This stability is checked by measuring the current with an oscilloscope (LeCroy 9310 M Dual, 300MHz). When the current reaches a stable value, the camera is focused on the tip of the cone where the jet emerges. Then, the camera was focused on the area where the jet breaks up into droplets. Several pictures were taken. The best pictures were used for measurements of the perturbation wavelength. Then, the camera was focused on the area just below the droplet production zone. Sets of two pictures were taken. The time interval between the two pictures was 15 μs. The best sets of pictures were used to measure the droplet size and the droplet velocity.

6.4 EXPERIMENTAL RESULTS

The camera system described in the previous section was used to investigate the jet break-up mechanism, as function of the flow rate. For all seven liquids, the same trend was observed. The jet break-up mechanism went through five different regimes. Figure 6.10 shows the first three regimes. The pictures are taken for different liquids. These pictures were chosen, because they were found to be the best in illustrating the observations. All other pictures showed the same effects. The scale is the same for all pictures shown.

At a relatively low flow rate for a certain liquid, an almost monodisperse spray was produced. Only every now and then, satellites or secondary droplets are produced in between primary main droplets. The most likely explanation for this observation can be seen in figure 6.10.a. The small filament between main droplets a and b breaks at main droplet b. As a result the surface tension in the tip of the broken filament creates an area of high pressure. Due to this

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high pressure, the liquid in the filament starts to flow back into main droplet a. This can be concluded from the position of the filament between droplets a, b, c, and d. Between droplets a and b, the filament is still intact. Between droplets b and c, the filament is broken near droplet c and is flowing back into droplet b. Between droplets c and d, the liquid of the filament has almost completely flowed back into droplet c.

If the flow rate increases, then the filament cannot flow back into the main droplet anymore, but breaks at the main droplet. The filament forms a secondary droplet. Between every pair of main droplets, one secondary droplet is formed. Figure 6.10.b shows an example of this jet break-up regime. Figure 6.10.c shows what happens at an even higher flow rate. The filament between main droplets a and c breaks near droplet c, and forms a secondary droplet b. When the filament between droplets a and b breaks, then a satellite is formed. This satellite is even smaller than the secondary droplet.

Figure 6.10 showed that the number of the secondary droplets and satellites depends on the liquid flow rate. The reason for this dependence on the flow rate is directly related to the increase in current. Figures 6.11.a and 6.11.b show the jet break-up for iso-butanol with conductivities of 13 and 30 μS m⁻¹, at a liquid flow rate of 1.11 × 10⁻⁹ m³ s⁻¹. The current measured for a conductivity of 13 μS m⁻¹ was equal to 31 nA, and for a conductivity of 30 μS m⁻¹, it was equal to 57 nA. Figure 6.11.a shows a low number of secondary droplets and satellites, where figure 6.11.b showed a lot more of them. These pictures clearly show that the system is able to see these secondary droplets. They also show that these secondary droplets are almost absent in the low current case, and that they are present in the high current case.
When the liquid flow rate or current was increased even more, then the jet did not only break up due to axisymmetric or varicose instabilities. The lateral or kink instabilities became more and more important. In the jet break-up of charged jets, the repulsion force due to the electric charge competes with the surface tension of the liquid. If no charge is present, then the jet can only break-up due to varicose instabilities. With increasing charge, the growth rate of the short varicose waves is increased. At a low ratio of the normal electric stress over the surface tension stress, the surface tension can suppress the kink instabilities. However, with increasing surface charge, the repulsion force of the charge becomes stronger, and the kink instabilities can start to grow. Figures 6.12 a, b, and c show the transition from a varicose jet break-up to the whipping jet break-up, as function of the flow rate. Figure 6.12.a shows the jet with varicose instabilities. Figure 6.12.b shows the first signs of kink instabilities. The liquid filament, and the secondary droplets start to leave the straight line. This fourth jet break-up regime will be referred to as the whipping filament regime. If the current or flow rate increases even more, then also the
main droplets start to leave the straight line. The jet starts to show a whipping behavior. This is the fifth jet break-up regime. Figure 6.12 c shows an example of this spiraling, whipping motion of the jet. This figure clearly shows that the break-up in this regime is influenced by both varicose and kink instabilities. In this regime the width of the droplet size distribution started to increase. These transitions between the various jet break-up regimes will be discussed later.

6.4.1 The Influence of the Applied Potential Difference

Gomez (1991) reported that the whipping of the jet increased with increasing applied potential difference. In the setup presented in figure 6.9, this effect was not observed. The jet break-up mechanism was independent of the potential difference. Figure 6.11 showed that the current through the liquid cone determines which jet break-up regime will occur. Gomez used a nozzle with an outer diameter of 0.45 mm. The nozzle diameter in the presented experiments was 8 mm. In Chapter 3 it was shown that the nozzle diameter has an influence on the current through the liquid cone. It was shown that for an 8-mm nozzle, the current was almost independent of the applied potential difference. The current for a nozzle with a diameter of 1 mm was about 15 percent higher than for an 8-mm nozzle, and depended more on the applied potential difference. So, in case of a 0.45 mm nozzle, the current through the liquid cone strongly depends on the potential difference. With increasing potential difference, the current increases. With increasing current, the whipping of the jet increases. In case of an 8-mm nozzle, the current is almost independent of the potential difference. So, the jet break-up regime is not changed.

In order to reduce the number of secondary droplets and satellites, the current through the liquid cone should be as low as possible. This can be obtained by using larger nozzles, at the lowest applied potential difference possible, and at liquid flow rates which are near to the minimum flow rate of the liquid.

6.5 COMPARISON BETWEEN SIMULATIONS AND EXPERIMENTS

The Cone-Shape model presented in Chapter 3 was used to calculate the shape of the liquid cone and jet, the current through the liquid cone, and the surface charge on the liquid surface. Figure 6.13 shows the comparison between the currents calculated and measured. In most cases, simulations and measurements agree very well. The 20 percent overestimation of the current for iso-butanol K=13 \( \mu \text{S m}^{-1} \) can be caused either by a mistake in the conductivity measurement, or by effects inside the liquid that create deviations from the assumptions used in the model. The current measured for iso-butanol is relatively low compared to n-butanol K=15 \( \mu \text{S m}^{-1} \). The droplet size was relatively large. This indicates that the current was really low for iso-butanol K=13 \( \mu \text{S m}^{-1} \). When LiCl was added to the iso-butanol, then the model predicted the current well again. The model predicts the current the best for jets that break-up due to varicose instabilities. The simulation results start to deviate from the measurements at higher flow rates, where the jet starts to make a whipping motion. For 2-butane the lowest flow rates are already near to the flow rate were the whipping starts. So the deviations are relatively larger.

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Figure 6.13 Comparison of the current measured and simulated.

The Mathematical Jet Break-Up model described in the theory section was used to simulate the jet break-up. The cone shape, and the surface charge distribution on the jet were used as an input. Droplet size, droplet velocity at jet break-up, and the wave number at jet break-up were calculated. The droplet size represents a droplet with the volume of a main droplet and a secondary droplet together. The mathematical model cannot distinguish between main droplets and secondary droplets. Figure 6.14 shows the simulated droplet sizes, and compares them to the droplet sizes measured for the main and secondary droplets. The measured sizes were an average
of only a small number of droplets. Since, the size distributions of main droplets and secondary droplets are relatively narrow, this low number will not cause a large statistical mistake. However, some scattering in the measured sizes cannot be avoided. The Mathematical Jet Break-Up Model requires a start position, and a start amplitude of the perturbation. The start position was taken equal to $dr_{jet}/dz=0.02$. The amplitude of the perturbation was taken equal to $\alpha_r/r_{jet}=0.001$. The influence of these assumptions will be discussed later.

![Ethylene Glycol](Image)

![iso-Butanol](Image)

![n-Butanol](Image)

![2-Butanone](Image)

*Figure 6.14 Comparison of the droplet size measured and simulated.*
The droplet velocity was measured just below the jet break-up region, because the measurement at break-up proved to be difficult. It was not easy to relate the droplets on consecutive pictures. The fast change in droplet shape, and the formation of secondary droplets at that position, make it difficult to recognize them. For n-butanol K=29 and iso-butanol K=30 \( \mu \text{S m}^{-1} \) an attempt was made. The velocity proved to be on average 10 percent lower than at the measurement position. This difference can be explained from the acceleration of the droplets in the electric field. Figure 6.15 shows the comparison between simulations and measurements. The
model underestimates in most cases the droplet velocity. However, the model calculates the velocity at jet break-up, where the measurements are done just below this area. With a correction for this effect equal to 10 percent, model and measurements agree quite well with each other. The results seem to indicate that the velocity depends less on the flow rate for the low conducting liquids than for the higher conducting liquids. The results also show that the droplet velocity increases with increasing conductivity, and the droplet velocity decreases with increasing liquid flow rate.

The dimensionless wave number at break-up was calculated with the Mathematical Jet Break-Up Model. This wave number was compared to the measurements in the following way. The measured droplet velocity was multiplied by 0.9 in order to compensate for the measurement position. Then, the liquid flow rate was used to calculate the radius of the unperturbed jet at break-up. At the jet break-up position, the wavelength was measured, and the dimensionless wave number calculated. Figure 6.16 shows the dimensionless wave numbers measured for all liquids as function of the liquid flow rate. The scattering in the data comes from the fact that there was quite some scattering in the wavelength, and the presented results are only the average of a few measurements.

\textbf{Figure 6.16 The dimensionless wave number calculated from the measurements.}
In spite of the scattering, some trends can be clearly seen. Rayleigh's theory predicts a wave number of 0.7 for low viscous liquids. 2-Butanone with its relatively high conductivity and low viscosity comes close to this value, but is lower. Weber's theory, equation (6.3), predicts for a jet diameter of 15 μm, for n-butanol a wave number of 0.62, and for ethylene glycol a wave number of 0.44. Figure 6.16 clearly shows that the wave numbers are below the values predicted by Weber and Rayleigh. Figure 6.17 shows the wave numbers calculated with the Mathematical Jet Break-Up Model. This model also predicts values of the dimensionless wave number, which are smaller than predicted by Rayleigh and Weber. The same trends can be seen. The low viscous liquid 2-butanone breaks up with the largest wave number. Ethylene glycol with the highest viscosity breaks up with the lowest wave number. The model predicts also that the conductivity has some influence on the dimensionless wave number. This effect is not contradicted by the measurements, but the scattering in the data is too big to confirm this effect. Also the model predicts an increasing wave number with increasing flow rate, which seems to be confirmed by the measurements.

The low wave numbers can be explained from the following effect. Rayleigh and Weber considered uncharged jets with a constant velocity. They did not consider a jet with a surface
charge, which is accelerated by the electric field. At the starting point on the jet, where the perturbation starts to grow, there is a certain perturbation with a certain wavelength. With increasing velocity, the wavelength increases, and the jet radius decreases. As a result, the wave number decreases along the jet. At jet break up, the perturbation, that causes the jet break-up, has a dimensionless wave number which is much smaller than at the position where the perturbation started to grow. If we assume an accelerating jet with a low viscosity and a low surface charge, then the fastest growing perturbation is the one with a wave number of 0.7. However, the wave number of the perturbations is not constant. For the accelerating jet, the fastest growing perturbation has a wave number at the starting point which is larger than 0.7. At the jet break-up position, the wave number is lower than 0.7. Together, with the effect of viscosity, this explains the low wave numbers measured and calculated.

When the wave number is known, then the ratio of the droplet diameter over the jet diameter at jet break-up can be calculated. For a wave number of 0.7 this yields a ratio of 1.89. For 2-butanoine with a wave number of 0.6 this ratio is 1.99. So, it is not surprising that Cloupeau and Prouhet Foch (1989) and Gomez and Tang (1991) both found agreement between their measurements for low viscous liquids and 1.89 predicted by Rayleigh’s theory. However, the results for n-butanol already show quite some influence of the viscosity, even more than can be expected from Weber’s theory. So, the acceleration of the jet has to be taken into account, when calculating the wave numbers at break-up.

6.5.1 Stresses in the Jet Surface

Cloupeau and Gomez both claimed that there was no influence of the charge on the jet break-up. Melcher’s theory clearly shows that there is an influence of charge. The influence of charge on the jet break-up cannot be deduced from the measurements. According to Melcher’s theory, the dominant wave number will increase with increasing charge. On the other hand, the acceleration of the jet makes the wave number lower at jet break-up. These two effects work against each other. The model requires a starting position, and a starting amplitude of the perturbation. The choice of the amplitude, and of the position has a strong influence on the wave number at break-up. This influence will be discussed in one of the next paragraphs. So, it is impossible to know, whether the differences and agreements in wave number between simulations and measurements come from the assumptions in the model, from the choice of the starting conditions, or from a real effect of the charge on the jet. However, the occurrence of the whipping motion of the jet, predicted by Melcher’s theory, proves that the charge has an influence on the jet break-up.

Melcher’s theory predicted that the whipping motion of the jet was possible if the ratio of the normal electric stress over the surface tension stress, $R_e$, in equation (6.32), was larger than 0.3. This ratio was checked with the experimental data. The jet diameter was again calculated in the same way as for the wave numbers. The Cone-Shape model showed that 95-99 percent of the current in the jet is transported by the surface charge. So, the measured current, the jet diameter at break-up, and the velocity at break-up can be used to calculate the surface charge at jet break-up. Two transitions in the jet break-up regime were investigated. The first transition was the
occurrence of the whipping filament, see Figure 6.12.b, and the second transition was the occurrence of the whipping jet, Figure 6.12.c. Figure 6.18 shows the stress ratios $R_{E_{\text{d}}}$ equation (6.32) at which these transitions occurred. This figure shows that the whipping starts around a stress ratio of 0.3. Although for ethylene glycol with a conductivity of 21 $\mu$S m$^{-1}$ the value was a bit lower. Melcher’s theory predicts that the growth rate for the varicose $m=0$ perturbations at a stress ratio of 0.3 is considerably larger than the growth rate for the kink $m=1$ perturbations. However, the measurements show that the amplitude of the whipping is already of the same order of magnitude. A possible explanation is that the amplitude of the initial perturbation for kink instabilities is larger than for varicose instabilities. Another explanation might be the fact that Melcher’s theory was derived for perturbations with a small amplitude compared to the jet radius. Figure 6.18 shows that the whipping of the filament between the main droplets starts at even lower stress ratios. This figure also seems to indicate that there is some influence of the wave number at jet break-up.

### 6.5.2 Stresses in the Droplet Surface

The whipping motion of the jet is of importance for the droplet size distribution. It has been observed that the whipping motion of the jet increases the width of the main droplet size distribution. In the whipping filament regime, not only the size distribution of the satellites and secondary droplets is changed, but also the size distribution of the main droplets is influenced. So, it is interesting to know when this whipping will start without having to take pictures of the jet break-up. The stress ratio on the surface of the jet can be used to predict this whipping motion. However, this requires the velocity at break-up. So, still pictures have to be taken. Droplets cannot have a charge larger than the Rayleigh Charge Limit. This limit can be calculated by taking the ratio of the electric normal stress over the surface tension stress equal to 1. This ratio is for a droplet in the Cone-Jet mode equal to

$$R_{E_{\text{d}}\text{, droplet}} = \frac{\sigma^2d_{\text{d}}}{8\epsilon_0\gamma} = \frac{I^2d_{\text{d}}^3}{288\epsilon_0\gamma Q^2}$$  \hspace{1cm} (6.47)
In order to calculate this ratio, the current, and droplet diameter, have to be known. These values could also be measured with other measurement tools. Figure 6.19 shows droplet stress ratios at which the transitions between the jet break-up regimes occur. These ratios were calculated from the current and droplet size calculated by the Cone-Shape model and the Mathematical Jet Break-Up model. So, it was assumed that no satellites are formed. Figure 6.19 shows that the whipping of the filament starts at a droplet stress ratio equal to 1. The higher stress ratio for iso-butanol K=13 μS m⁻¹ is caused by the overestimation of the current by the model. As a result, the calculated stress ratio is also higher than the real stress ratio.

The theory of the Rayleigh Charge Limit yields that no droplets can exist, which have a stress ratio larger than 1. In the Mathematical Jet Break-Up Model the formation of secondary droplets and satellites cannot be simulated. In reality, secondary droplets and satellites are formed. These droplets also carry a part of the charge. As a result, the stress ratio calculated for a droplet in the simulations is higher than the stress ratio for the main droplets in the experiments. This might explain why the whipping starts at a droplet stress ratio larger than 1. Figure 6.20 shows a comparison between the droplet stress ratios calculated by the model and calculated from the measurements. In order to calculate the stresses from the measurements, the droplet diameter was taken equal to a droplet with the volume of a main droplet and a secondary droplet together. The model only takes the varicose jet break-up into account. As a result, the simulated droplet can have a stress ratio larger than 1. For the measurements, the stress ratio is almost always below 1. It is thought that these stress ratios above 1 are the result of measurement inaccuracy, or statistical effects. This figure clearly shows that the model is not correct, when a stress ratio is calculated, which is larger than 1. This can be explained from the fact that the model does not take the whipping motion of the jet into account.

6.5.3 Scaling of the Droplet Size

Section 2.3.1 and equation (3.1) showed the scaling laws for droplet size and current. For the current scaling laws, agreement between researchers exists about the fact that the current depends
Figure 6.20 Comparison of the droplet stress ratio measured and simulated as function of the flow rate.

on the liquid flow rate to the power 0.5. This was also confirmed by the results of the Cone-Shape model. The present droplet size scaling law predicts a dependence of the main droplet diameter as function of the flow rate, which scales like $d_0 \sim Q^{1/3}$. The Rayleigh Charge Limit can also be used to calculate the droplet size. When equation (6.47) is considered, and when it is assumed that $I_Q^{1/2}$, then the Rayleigh Charge Limit, $R_{\text{Rayleigh}}$, will yield $d_0 \sim Q^{1/3}$. This means that the droplet size calculated with the scaling law as function of the flow rate should be parallel to the droplet size calculated with the Rayleigh Charge Limit. Gomez and Tang (1994), Gañán-Calvo et al. (1997), and Chen et al. (1995) all noticed that this was only the case at high flow
rates. At high flow rates, and when the presence of secondary droplets is neglected, then the droplet charge was equal to 0.7 to 0.8 times the Rayleigh Charge Limit. The stress ratio on such droplets is then equal to 0.49 to 0.64. At lower flow rates, the droplet diameter decreases faster as a function of decreasing flow rate than predicted by the scaling law. Gañán-Calvo tried to solve this discrepancy by introducing an extra fitting parameter \( c_i \) into equation (3.1).

A fit was made of the main droplet size measured as function of the liquid flow rate for the three liquids with the lowest conductivity. Only those results were considered that yield a jet, which breaks up in the varicose jet break-up regime. For the four highest conducting liquids, only a few measurement points were available. Consequently, measurement mistakes would have a large influence on the obtained fitting results. The trends for these four liquids were exactly the same as for the three liquids with a lower conductivity. Table 6.2 shows the fitting results for both measurements and simulations, where \( d_d \sim Q^a \).

<table>
<thead>
<tr>
<th></th>
<th>Measured</th>
<th>Model</th>
</tr>
</thead>
<tbody>
<tr>
<td>n-Butanol (nB15)</td>
<td>0.50</td>
<td>0.48</td>
</tr>
<tr>
<td>iso-Butanol (iB13)</td>
<td>0.47</td>
<td>0.48</td>
</tr>
<tr>
<td>Ethylene Glycol (Eg21)</td>
<td>0.46</td>
<td>0.48</td>
</tr>
</tbody>
</table>

Table 6.2 The comparison between measurements and simulations for the power \( a \) for jets that break-up due to varicose instabilities.

These results show that the main droplet diameter for jets that break-up due to varicose instabilities scales with the flow rate as \( d_d \sim Q^{0.48} \). The results of figures 6.19 and 6.20 show that, when the whipping motion becomes of importance, then the droplet stress ratio approaches a fixed value. If it is assumed that no secondary droplets are present, then the size of the droplets can be calculated with the assumption that each droplet has a fixed stress ratio. For the results presented in this chapter, a droplet stress ratio of 0.8 seems to be the best. However, the scattering in the data is too big to establish accurately this value. Also, when the pictures were not perfectly focussed, then the measurement system was liable to overestimate the droplet size. This could result in a 4 \( \mu \)m overestimation of the droplet size. If this was really the case, then the stress ratio would have been somewhere between the values of 0.49 to 0.64 calculated from the measurements of Gañán-Calvo et al. (1997) and Gomez and Tang (1994). More precise measurements are needed to correctly establish this stress ratio in the whipping jet break-up regime. For the whipping jet break-up regime, and using the stress ratio of 0.8, the droplet size can be calculated from the flow rate and the current using

\[
d_d = (0.8 \frac{288 \varepsilon_0 \gamma Q^2}{I^2})^{1/3}
\]

Figure 6.21 shows the measured main droplet size, the droplet size calculated with the Mathematical Jet Break-Up Model for the axisymmetric varicose jet break-up regime, and the
The whipping filament regime started at a droplet stress ratio, calculated from the simulations, equal to 1. Figures 6.20 and 6.21 show that the stresses on the droplets, calculated from the measurements, never reached this stress ratio. So, the jet break-up in the varicose jet break-up regime seems to be already influenced by the surface charge before the whipping becomes really visible. In general, it can be said that the droplet size produced in the Cone-Jet mode can be calculated with a varicose jet break-up scaling law that should fit to \( d_\text{d} \sim Q^{0.48} \). When, the droplet size calculated with such a law becomes larger than the droplet size calculated with equation (6.48), then the jet break-up enters the transition zone from varicose to whipping jet break-up. In that case equation (6.48) should be used to calculate the droplet size.

### 6.5.4 The Dimensionless Wave Number at Jet Break-Up

Rutland and Jameson (1970), and Parkin (1973), have investigated the jet break-up as function of the wave number. Their liquid jets had a relatively large diameter (~1 mm), there was no acceleration of the jet, and there was no surface charge. In that case, the number and volume of the secondary droplets and satellites depends on the dimensionless wave number. In the presented experiments, the jet diameter was in the order of tens of microns. This small jet diameter might have some influence on the jet break-up. Especially, the effect that the liquid filament between two main droplets flows back into one of the main droplets, might be the result of the small jet diameter. For larger uncharged jets, the wave numbers of Figure 6.16 would lead to the formation of secondary droplets. However, in the break-up of thin jets, the liquid filament can flow back into the main droplet. If this happens, no secondary droplets are formed.

Figures 6.16 and 6.17 showed that the wave number increased with increasing flow rates. For an uncharged jet, this would mean less secondary droplets and satellites. However, in the Cone-Jet mode an increase in secondary droplets and satellites was observed. The reason for this increase, is the increase in the stress ratio of the electric normal stress over the surface tension stress on the jet. Figure 6.11 shows that the wavelengths, for iso-butanol \( K=13 \) and 30 \( \mu \)S m\(^{-1}\) at the same flow rate, are in the same order of magnitude. Figure 6.16 shows that the dimensionless wave number was lower for \( K=13 \) than for \( K=30 \) \( \mu \)S m\(^{-1}\). Without surface charge, the liquid with \( K=13 \) \( \mu \)S m\(^{-1}\) should produce more secondary droplets and satellites. However, this is not the
case. The liquid with $K=30 \, \mu S \, m^{-1}$ produces much more secondary droplets. This can be seen in Figure 6.11. Figure 6.20 shows that the stress ratio on the droplets is much higher for iso-butanol $K=30$ compared to iso-butanol $K=13 \, \mu S \, m^{-1}$. This means that the stress ratio on the jet is also much higher. Melcher's theory predicts that the charge favors the shorter wave lengths. As a result, a high stress ratio enhances the break-up of the filament. Also, the repelling force of the charge might reduce the velocity of the liquid flowing back into the main droplet. These two effects could explain the observation that more secondary droplets are formed with an increasing stress ratio.

The transition between a flow rate that yields a small number of secondary droplets, and one that yields one secondary droplet for every main droplet, was not a sharp transition. The number of secondary droplets increases gradually with increasing flow rate. This can be explained from the fact that the perturbation wavelength is not perfectly constant. It depends on the disturbance that creates the perturbation on the jet. This perturbation creation process is very irregular. As a result, the relatively long wavelengths will create secondary droplets, where the small wavelengths will not produce them. With increasing current through the liquid cone, the ratio of the electric normal stress over the surface tension stress increases. As a result, the dimensionless wave number that just creates secondary droplets will increase. Therefore, more waves will create secondary droplets and satellites. So, no criterion can be given, whether a stress ratio will give rise to secondary droplets and satellites or not. However, the lower the flow rate for a certain liquid, the lower the current through the liquid cone, the lower the stress ratio on the jet, the lower the number of secondary droplets and satellites. If an oscillating electric field, with the right frequency, is superimposed on the present electric field, then it is possible to reduce the fluctuations in the wavelength, Cloupeau (1994). In that case, it should be possible to create a sharp transition between a jet break-up regime without secondary droplets, and a regime with secondary droplets.

Table 6.3 The influence of the initial conditions in the Mathematical Jet Break-Up Model on the obtained droplet size, the dimensionless wave number at jet break-up, and the liquid velocity.

<table>
<thead>
<tr>
<th>Start Position $\frac{dr_{pl}}{dz}$</th>
<th>Start Amplitude $\alpha/r_{jet}$</th>
<th>Velocity at Break-Up $m , s^{-1}$</th>
<th>$d_d$ $\mu m$</th>
<th>$k_{r_{jet}}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.02</td>
<td>0.001</td>
<td>12.2</td>
<td>27.5</td>
<td>0.279</td>
</tr>
<tr>
<td>0.05</td>
<td>0.01</td>
<td>10.7</td>
<td>32.6</td>
<td>0.211</td>
</tr>
<tr>
<td>0.02</td>
<td>0.01</td>
<td>10.4</td>
<td>28.8</td>
<td>0.307</td>
</tr>
<tr>
<td>0.01</td>
<td>0.01</td>
<td>10.7</td>
<td>26.7</td>
<td>0.365</td>
</tr>
<tr>
<td>0.008</td>
<td>0.01</td>
<td>10.9</td>
<td>26.1</td>
<td>0.392</td>
</tr>
<tr>
<td>0.005</td>
<td>0.01</td>
<td>11.4</td>
<td>25.0</td>
<td>0.424</td>
</tr>
<tr>
<td>0.005</td>
<td>0.006</td>
<td>11.7</td>
<td>25.0</td>
<td>0.406</td>
</tr>
<tr>
<td>0.005</td>
<td>0.004</td>
<td>12.0</td>
<td>25.0</td>
<td>0.388</td>
</tr>
<tr>
<td>0.005</td>
<td>0.002</td>
<td>12.4</td>
<td>24.4</td>
<td>0.399</td>
</tr>
</tbody>
</table>

The starting position on the jet and the initial amplitude of the perturbation have an influence on the wavelength, and on the liquid velocity at jet break-up. Table 6.3 shows this
influence on the velocity, on the dimensionless wave number, and on the droplet diameter. The results were calculated for iso-butanol $K=13 \\mu S \ m^{-1}$ at a liquid flow rate of $1.11 \times 10^9 \ m^3 \ s^{-1}$. These results show that the initial amplitude mainly influences the velocity at jet break-up, where the starting position has an influence on the velocity, the wave number at break-up, and the droplet size.

The initial conditions in the Mathematical Jet Break-Up Model were $\frac{dr_{j}}{dz}=0.02$ and $\frac{\alpha}{r_{j}}=0.001$. Careful comparing of the simulation results with the measurement results revealed that the measured droplet size and velocity agreed well with the simulation results. However, the wavelength at break-up appeared to be 26 percent lower for the measurements than for the simulations. If the calculated velocity is right, but the calculated wavelength is too long, then the calculated droplet size should be 9 percent too big. However, on average they were exactly the same. The experiments with the polymer particles showed, that when the camera system is not perfectly focussed, then the system overestimates the droplet size by 4 $\mu m$. This could explain why the droplet sizes calculated and measured are in agreement, where the wavelength is overestimated by the simulations. More precise droplet size measurements are needed to solve this discrepancy between droplet size and wavelength. When the simulated velocity and wave number have to be matched with the measured velocity and wave number, then the best initial conditions in the Mathematical Jet Break-Up Model are expected to be $\frac{dr_{j}}{dz}=0.005$ and $\frac{\alpha}{r_{j}}=0.002$.

6.5.6 The Droplet Size and Charge of Secondary Droplets

The Numerical Jet-Break-Up model was used to calculate the diameter and charge for both main droplets and secondary droplets. The model did not predict that the liquid of the filament would flow back into one of the main droplets. A few reasons could be found. Firstly, it is difficult to model the shock wave that occurs in the filament at the moment of break-up. The model shows that the liquid starts to flow back, but the effect in the model is not very strong. As a result, the filament breaks a second time before the liquid can flow back into the main droplet. Secondly, the effect of the surrounding air is not taken into account. Thirdly, the acceleration of the liquid was neglected. Some simulations were done with an accelerating jet. In that case, the number of grid points was insufficient to reach an acceptable accuracy. In spite of this low number of grid points, the acceleration seems to have an effect on the shape of the filament. However, when in the simulations with and without acceleration the same wave number is used at the moment of break-up, then the acceleration of the jet seemed to have little influence on the size and charge simulated for the secondary droplets.

The numerical model was used to estimate the sizes and charges of the main and secondary droplets as functions of the wave number at jet break-up. The results were calculated for iso-butanol $K=13 \ \mu S \ m^{-1}$ at a liquid flow rate of $1.11 \times 10^9 \ m^3 \ s^{-1}$. Figure 6.22 shows the sizes
and charges simulated as functions of the wave number for both main and secondary droplets. The results show that a change in wave number creates a small change in the main droplet size, but a larger change in the size of the secondary droplet. Figure 6.14 shows that ratio of the secondary droplet size over the main droplet size decreases with increasing flow rate. This confirms that the dimensionless wave number at break-up increases with increasing flow rate. For iso-butanol K=13 μS m⁻¹, and for flow rates close to Q= 1.11 10⁸ m³ s⁻¹, the size ratio of the secondary droplet size over the main droplet size is about 0.65. When using initial conditions dr/dz=0.02 and α/τµ=0.001, the Mathematical Jet Break-Up Model calculated a wave number of 0.28. From Figure 6.22 can be deduced that a wave number of 0.33 yields the best size ratio. This result indicates that the Mathematical Jet Break-Up Model underestimates the wave number by 18 percent. This is in reasonable agreement with the 26 percent underestimation that came out of the comparison with the measurements.

Figure 6.22 The influence of the dimensionless wave number at jet break-up on the size and charge of the main and secondary droplets.

Figure 6.23 The influence of viscosity on the size and charge of the main and secondary droplets.

Figure 6.23 shows the effect of viscosity on the charge, and on the size of the secondary droplets. This figure shows that, with increasing viscosity, the amount of liquid flowing out of the filament increases. This can be explained from the fact that the growth rate of perturbations with a high wave number is more reduced than the growth rate of perturbations with a low wave number. When a perturbation is considered, then the filament is formed from a perturbation with a relatively low wave number. The break-up of the filament near the main droplet is caused by a secondary perturbation with a much higher wave number. As a result, the breaking of the filament is more reduced by the viscosity than the formation of the filament. Therefore, more liquid can flow out of the filament before the filament breaks. The charge is much less affected by viscosity. The charge depends on the liquid flow, because of charge convection. On the other hand, it depends also on the conduction due to the electric field. In a rough approximation, it
could be said that for a conductor, the electric field between main droplet and filament is equal to zero. This electric field consists of two fields caused by the charge on the main droplet and by the charge on the filament. For a case with equal wave numbers, the length of the filament is almost independent of the viscosity. So the charge on both main droplet and filament should not change too much. This means that the charge on the secondary droplet should be almost independent of the viscosity.

A droplet charge to diameter relation equal to \( q \sim d_d \) \(^{1,5} \) for one spray of ethylene glycol was discussed in Chapter 5. If the current and the diameters of the main and secondary droplets are known, then this relation can be used to calculate the charge on the main and secondary droplets in this spray. This relation was calculated from the movement of the droplets in the electric field. The main droplet size was 18 \( \mu \text{m} \), and the secondary droplet size was 8 \( \mu \text{m} \). The numerical model was used to simulate the break-up of an ethylene glycol jet with the same conductivity. A wave number and jet radius were chosen, that yield the same droplet sizes as the measured sizes. This resulted into a charge to diameter relation equal to \( q \sim d_d \) \(^{1,2} \). The measurements were done in another experimental setup. In this case, it was not known, whether always one secondary droplet was produced per main droplet, and whether the jet was still in the varicose jet break-up regime or in the early stages of the whipping jet regime. Apart, from the measurement accuracy, and apart from the simplifications in the model, also satellites, and some influence of whipping jet break-up could explain the difference between 1.5 and 1.2. In spite of this, the results of figures 6.22 and 6.23 clearly show that simple charge to diameter relations as discussed above do not exist for all sprays. The droplet size ratio of the main droplets over the secondary droplets, and the viscosity have a strong influence on this relation. If the jet breaks-up in the varicose jet break-up regime, and if the droplet sizes are known, then the results of figures 6.22 and 6.23 can be used to make some estimations of the charge on the secondary droplets. However, more research is necessary to establish a relation between liquid properties, liquid flow rate, and the size and charge of main and secondary droplets.

### 6.6 Conclusions

The jet break-up mechanism in the Cone-Jet mode was investigated, as function of the liquid flow rate. For all seven liquids, the same trend was observed. The jet break-up mechanism went through five different regimes with increasing flow rate. The first regime is the regime without secondary droplets and satellites produced. The small filament between two main droplets breaks at the first main droplet produced. The surface tension in the tip of the broken filament creates an area of high pressure. Due to this high pressure, possibly due to the influence of the surrounding air at high jet velocities, and possibly due to the acceleration caused by the electric field, the liquid in the filament starts to flow back into the other main droplet. The second regime occurs when the filament also breaks near the second main droplet before the liquid can flow back into this droplet. Between every pair of main droplets one smaller secondary droplet is formed. The third regime occurs when the filament between two main droplets forms a secondary droplet. The liquid filament between the secondary droplet and one of the main droplets forms
Jet Break-Up

an even smaller satellite droplet. The fourth regime occurs when lateral or kink instabilities started to become important. The liquid filament and the secondary droplets start to leave the straight line at jet break-up. This jet break-up regime will be referred to as the whipping filament regime. The fifth observed regime occurs when the whole jet starts to show a whipping motion. The jet break-up in this regime is influenced by both varicose and kink instabilities. In the last two regimes the width of the size distribution of the main droplets starts to increase.

The wave number at break-up increases slightly with increasing flow rate. In literature for uncharged jets, fewer secondary droplets and satellites are produced with increasing wave number. In the Cone-Jet mode, the opposite occurs. This is explained from the surface charge on the jet. With increasing flow rate, the current through the liquid cone increases. This increases the stress ratio of the electric normal stress over the surface tension stress in the liquid jet. As a result, short waves can break-up faster and the filament breaks a second time before the liquid in the filament can flow back into the main droplet. Satellites and secondary droplets are formed. At even higher flow rates, surface charge induces the whipping motion of the jet.

Whether the jet break-up regime depends on the applied potential difference, depends on the nozzle diameter. For an 8 mm nozzle the current is almost independent of the applied potential difference. Since, the current determines the surface charge, the jet break-up regime was not changed. For a 0.45 mm nozzle, the current depends on the applied potential difference. So, the jet break-up regime also depends on the this potential difference.

In order to reduce the number of secondary droplets and satellites, the current through the liquid cone should be as low as possible. This can be obtained by using larger nozzles, at the lowest potential difference possible, and at liquid flow rates which are near to the minimum flow rate of the liquid.

The current calculated with the Cone-Shape model agrees very well with the measurements.

The Mathematical Jet Break-Up model, with initial conditions equal to $dr_{jet}/dz = 0.02$, and $\alpha_{i}/r_{jet} = 0.001$, predicted well the droplet velocity and the droplet size. It also predicted, the right trends for the perturbation wavelength at jet break-up. However, the wavelengths were on average 26 percent to long. This could be solved by choosing initial conditions equal to $dr_{i}/dz = 0.005$, and $\alpha_{i}/r_{jet} = 0.002$. More precise droplet size measurements are needed to find the exact starting position and starting amplitude that will yield the best results.

It was observed that the velocity at jet break-up increases with increasing conductivity, and the velocity decreases with increasing liquid flow rate.

The wave number at jet break-up cannot be calculated with the simple assumptions of Rayleigh's theory. Viscosity, surface charge and the acceleration of the jet have to be taken into account. Increasing viscosity yields decreasing wave numbers at break-up. The acceleration of the jet yields a lower wave number, where increasing surface charge yields a larger wave number.

Melcher's theory predicted a whipping motion of the jet, if the ratio of the normal electric stress over the surface tension stress on the jet was larger than 0.3. This ratio was confirmed by the experiments. However, the amplitude of the whipping motion was larger than expected from this theory. The whipping filament regime occurred at a stress ratio of about 0.23.

If a model is used that only considers varicose jet break-up, then the first signs of
whipping behavior can be detected in a real spray, when the simulated stress ratio on the droplets is close to 1. However, the real droplet stress ratio never reaches this value. Even when the whipping is not detected, then the jet break-up is already influenced.

It was found that the droplet size scales with the flow rate in the following way

\[ d_d \sim Q^{0.48} \quad \text{varicose break-up regime} \]

\[ d_d \sim Q^{0.33} \quad \text{whipping break-up regime} \]

The droplet size in the whipping regime can be calculated from the current and the liquid flow rate with the assumption that the droplet charge is equal to a fixed percentage of the Rayleigh Charge Limit. This percentage ranges from 70 to 90 percent, depending on the different authors. Very precise size measurements are needed to establish this percentage.

It was found that no simple charge to diameter relation exists for the charge of secondary droplets produced in the varicose jet break-up regime. The influence of the wave number and the viscosity at break-up have to be taken into account.
CHAPTER 7

SCALING OF DROPLET SIZE AND CURRENT

This Chapter presents a new derivation of the current scaling, \( I/I_o = (Q/Q_o)^{\gamma} \) with \( I_o = (e_o \gamma^2 / \rho)^{\gamma} \), and \( Q_o = e_o \gamma / K \rho \). The difference with the present scaling laws is that the liquid permittivity has disappeared from the scaling. Simulations and derivations proved that the influence of the liquid permittivity is small. This current scaling is extended to liquids with low conductivities and low viscosities.

A relation is derived, which enables the calculation of the droplet size from the current and the liquid flow rate, for the varicose jet break-up regime, \( d_{d, \text{varicose}} = c_d \rho e_o Q^\gamma / \gamma \). The droplet size and current scaling are fitted, and compared to measurements for nozzles with diameters of 1 mm, Gañán-Calvo (1997), \( c_d = 1.76 \), and of 8 mm, Chapter 6, \( c_d = 2.05 \).

For the additive HCl in ethylene glycol and n-butanol, the current was at low flow rates up to 50 percent lower than for the same liquid with the same conductivity, but with LiCl as additive. It is shown that the polarity of the applied potential does not have an influence on the current.

7.1 INTRODUCTION

The droplet size produced in the Cone-Jet mode depends on the jet break-up mechanism. In order to judge, which kind of jet break-up occurs, the current through the liquid cone has to be known. Chapter 3 introduced the physical numerical Cone-Shape model, which is able to calculate the shape of the liquid cone and jet, and the current through this cone. Chapter 6 presented a physical mathematical model, which is able to calculate the droplet size and charge. The input to this model is the output of the Cone-Shape model. Disadvantages of these models are the calculation time that is required, and the time needed for defining the input into the models. Scaling laws offer the possibility to come to less accurate, but much easier obtainable results.

In the past, various equations have been derived, to estimate the produced main droplet size, and the electric current required for a liquid sprayed in the Cone-Jet mode. These relations were discussed in section 2.3.1. Combination of equations (2.3), (2.4), and (2.7), yields
\[ \frac{d_{\text{jet}}}{d_o} = 0.4 \epsilon_r^{\frac{1}{3}} \left( \frac{Q}{Q_o} \right)^{\frac{1}{3}} \]
\[ \frac{I}{I_o} = f \left( \frac{\epsilon_r}{\epsilon_r^{1/2}} \right) \left( \frac{Q}{Q_o} \right)^{\frac{1}{2}} \]

where, \( I \) is current [A], \( d_{\text{jet}} \) is jet diameter [m], \( Q \) is liquid flow rate \([m^3 \text{ s}^{-1}]\), \( \epsilon_r \) is relative permittivity of the liquid, \( Q_o, I_o, \) and \( d_o \) refer to the characteristic flow rate, current and droplet diameter, which were defined in equation (2.7). Comparison of equations (7.1) and (2.6) shows that these scaling laws for highly conductive or highly viscous liquids are equivalent. The differences between these relations are the influence of the relative permittivity of the liquid, and extra constants in equation (2.7). Chen and Pui (1997) noticed this difference with respect of the liquid permittivity, and tried to measure the influence of permittivity. This resulted in again another relation with respect of the permittivity.

Chapter 6 showed that the droplet size in the varicose jet break-up regime scales with \( d_j \sim Q^{0.48} \). In the whipping jet break-up regime the droplet size scaled with \( d_j \sim Q^{0.33} \). In Chapter 6 a droplet size scaling was presented for the whipping jet break-up regime. This size scaling equals

\[ d_{\text{d,whipping}} = (0.8 \frac{288 \epsilon_r \gamma Q^2}{I^2})^{\frac{1}{3}} \]

where, \( d_{\text{d,whipping}} \) is the droplets size predicted for a jet that breaks up in the whipping jet break-up regime [m]. The constant 0.8 was calculated from the measured droplet sizes. However, other measurements of Gomez and Tang (1994), and Gañán-Calvo et al. (1997), indicate that this value can also be 0.49 or 0.64.

In this Chapter new scaling laws will be presented. A current scaling law is derived, and the discrepancy with respect to the relative permittivity will be clarified. The current law is extended with a scaling of the current increase due to the liquid velocity profile in the jet. This extension makes it possible, to use this scaling law for all Newtonian liquids. A droplet size scaling law for the varicose jet break-up regime will be derived. The current and the droplet size laws are fitted and compared to experimental results. These laws are also fitted to experimental results presented by Gañán-Calvo et al. (1997), to which sometimes is referred to as GC (1997). Also, the influence of the ion species in the liquid, and the influence of the nozzle diameter, on the current will be discussed.

### 7.2 THEORETICAL SCALING OF THE CURRENT

Chen and Pui (1997) compared the scaling laws of equations (2.6) and (7.1). They observed the difference with respect of the influence of the electric permittivity of the liquid between these two scaling laws. They measured indirectly this influence, and found again a different relation
with respect of the liquid permittivity. In order to clarify the effect of electrical permittivity, the scaling law is derived in another way than before. The shape of the liquid cone is mainly determined by the tangential electric stress, the surface tension, the normal electric stress, and the velocity pressure in the cone. As pointed out by Gañán-Calvo et al. (1997), this shape is determined by the one dimensional axial momentum equation of the jet which may be written as

\[
\frac{2\tau}{r_s} = \frac{\partial}{\partial z} \left( \frac{\gamma}{r_s} - \frac{1}{2} \frac{\sigma^2}{\varepsilon_o} + \frac{1}{2} \frac{\rho u_z^2}{\varepsilon_o} \right)
\]

(7.3)

Where, \( r_s \) is the radius of the surface of the liquid cone and jet in the cylindrical coordinate system [m], \( z \) is the axial coordinate along the jet [m], \( u_z \) is liquid velocity in the axial direction [m s\(^{-1}\)], \( \tau \) is tangential stress [N m\(^{-2}\)], and \( \sigma \) is surface charge [C m\(^{-2}\)].

The electric field of a ring of charges scales with

\[
E_z = \frac{q}{\varepsilon_o \Delta z^2}
\]

(7.4)

where, \( q \) is the charge on the ring [C], \( E_z \) is electric field strength in the axial direction [V m\(^{-1}\)], and \( \Delta z \) is the distance in the axial direction from this charged ring [m]. When a position is considered in the middle between two charged rings with different radii and different charge, then the electric field scales with

\[
E_z = \frac{2\pi \Delta (r_\sigma) \Delta z}{\varepsilon_o \Delta z^2} \sim \frac{\sigma}{\varepsilon_o} \frac{dr_s}{dz} + \frac{r_s \; d\sigma}{\varepsilon_o \; dz}
\]

(7.5)

In the derivation of equation (7.5), the relative permittivity of the liquid can be neglected, because at the beginning of the jet, \( dr/dz \) is small. So, the electric field strength in the axial direction depends mainly on the tangential electric field. Since, the permittivity of a liquid only influences the normal electric field strength inside the liquid, and not the tangential electric field strength at that position, the electric field strength in the axial direction is almost independent of the permittivity of the liquid.

In the jet, the value of \( dr/dz \) is small. So, the axial electric field strength is almost equal to the tangential electric field strength. Simulations with the Cone-Shape model, Chapter 3, showed that in the jet, the tangential viscous stress is much smaller than the tangential electric stress. So, the tangential stress in equation (7.3) comes mainly from the axial electric field strength, and the surface charge. When it is assumed, that the axial velocity at the liquid surface
is equal to the axial liquid velocity in the jet center, then the liquid velocity can be calculated with

$$u_z = \frac{Q}{\pi r_s^2} \quad (7.6)$$

If this is the case, then equations (7.3), (7.5), and (7.6) can be used to derive the following differential equation

$$\left( -\frac{\gamma \varepsilon_o}{r_s \sigma^2} - \frac{2\rho \varepsilon_o Q^2}{\sigma^2 \pi^2 r_s^4} \right) \frac{dr_s}{dz} - \frac{r_s \, d\sigma}{\sigma \, dz} - \frac{dr_s}{dz} + \frac{r_s \, d\sigma}{\sigma \, dz} \quad (7.7)$$

In this equation, two dimensionless numbers can be distinguished. The first number describes the relation between the surface tension and the normal electric stress. The second dimensionless number describes the relation between the velocity pressure, and the normal electric stress. This equation can be made dimensionless with the following set of equations

$$\sigma = \left( \frac{\varepsilon_o \gamma}{r_s^2} \right)^{\frac{1}{2}} \sigma' \quad r_s = r_s^*, \quad r_s' = r_s^*, \quad z = z^* \quad (7.8)$$

where, $\sigma'$ is the dimensionless surface charge, $r_s'$ is the dimensionless radius of the liquid surface, $z'$ is the dimensionless axial coordinate of the jet, and $r_s^*$ is a characteristic radius [m]. In the cone, the shape is determined by the surface tension stress. On the other hand, in the jet, the shape is determined by the velocity pressure. At the position on the jet, where the value of the first dimensionless number of equation (7.7) is equal to the value of the second dimensionless number, the velocity pressure becomes dominant over the surface tension stress. This radius $r_{sv}$ [m] is equal to

$$r_{sv} \sim \left( \frac{\rho Q^2}{\gamma} \right)^{\frac{1}{3}} \quad (7.9)$$

This radius $r_{sv}$ symbolizes the cone radius, where the jet emerges from the liquid cone.

The current through the liquid cone is not determined by this characteristic radius. The current depends on the charge transport in the liquid cone. The charge is transported in two ways. In the base of the cone, the charge is mainly transported through conduction. In the jet, the charge is mainly transported by charge convection. The current through the cone equals the sum of these two contributions. So, the current equals
Scaling of Droplet Size and Current

\[ I = \pi r_s^2 E_z K + 2\pi r_s u_z \sigma \quad (7.10) \]

Along the cone and jet, the sum of the changes in the charge convection and the conduction current should be equal to zero. Together with equations (7.5), and (7.6), equation (7.10) yields

\[ \frac{d(r_s^2 d(r_s) \sigma)}{dz} - \frac{Q \varepsilon_o}{K} \frac{d(\frac{\sigma}{r_s})}{dz} \quad (7.11) \]

When this equation is made dimensionless with the relations of equation (7.8), then it can be shown that the characteristic radius \( r_s \) scales with

\[ r_s^* = \left( \frac{Q \varepsilon_o}{K} \right)^\frac{1}{3} \quad (7.12) \]

This radius \( r_s^* \) represents the radius, where the conduction becomes too low to supply more current into the jet. Equations (7.5), (7.6), (7.8), (7.10), and (7.12), can be used to derive the current scaling law for liquid cones with flat radial profiles of the axial velocity. This equation equals

\[ I = \frac{Q}{r_s^*} \frac{\gamma \varepsilon_o}{r_s^*} + \frac{2\sigma'}{r_s^*} + K \frac{2}{\varepsilon_o r_s} \frac{\gamma}{r_s^*} \frac{1}{3} \pi r_s^2 E_z' \quad (7.13) \]

\[ = (\gamma KQ)^\frac{1}{2} \left( \frac{2\sigma'}{r_s^*} + \pi r_s^2 E_z' \right) - (\gamma KQ)^\frac{1}{2} \]

where, \( E_z' \) is the dimensionless electric field strength in the axial direction. The difference with the scaling laws of equations (7.1) and (2.6) is that the liquid permittivity has completely disappeared from the scaling law.

Without the kinetic energy of the jet, the cone cannot exist, because the surface tension would relax the cone into a round shape again. The relations of equation (7.8) can be substituted into the second dimensionless number of equation (7.7). In that case, the second dimensionless number represents the velocity pressure over the surface tension stress. At the position on the
cone, where the cone radius starts to limit the conduction, equation (7.12), this ratio becomes

\[
\frac{\rho Q^2}{\gamma r_j^3} - \frac{\rho Q K}{\gamma \varepsilon_o} = \frac{Q}{Q_o}
\]

(7.14)

So, when this ratio is equal to 1, this relation would yield almost the same relation for the minimum flow rate as equations (2.8) and (2.9). The difference is that the liquid permittivity term has disappeared. The minimum flow rate was not investigated experimentally. So, a final conclusion of which of these relations is valid cannot be given. However, the presented theory yields that the minimum flow rate is most likely a result of the relative reduction in the velocity pressure in the jet compared to the surface tension stress with decreasing liquid flow rate.

Relation (7.13) can also be written in the form of equation (7.1), and is then equal to

\[
\frac{I}{I_o} = (\frac{Q}{Q_o})^{\frac{1}{2}}
\]

(7.15)

7.3 EXTENSION OF THE CURRENT SCALING FROM NUMERICAL SIMULATIONS

The current scaling derived in the previous section is valid for liquids with a flat radial profile of the axial velocity in the jet. This current scaling can be fitted to measurements in the following way

\[
\frac{I}{I_o} = a_L (\frac{Q}{Q_o})^{b_L}
\]

(7.16)

where, L is the current for a liquid cone with a flat radial profile of the axial velocity [A], and \(a_L\) and \(b_L\) are constants, which will be discussed in section 7.5. As pointed out by Gañán-Calvo et al. (1997), this radial profile is not always flat. In case of a low viscous liquid, the shear stresses in the jet are too small to obtain a flat profile. Also, when the conductivity is low, and the flow rate is high, then the jet has a relatively large diameter. In that case, the shear stresses are also too small to maintain a flat profile. If the axial velocity at the liquid surface is higher than the velocity in the jet center, then the charge convection current is relatively larger. As a result, the total current is larger than expected from equation (7.16). In order to estimate the current for liquids with a steeper velocity profile the following procedure can be used.

The Cone-Shape model, which was presented in Chapter 3, was used to investigate the influence of the experimental conditions on the electric field strength in the axial direction. The results of the Cone-Shape model showed that the electric field reaches a maximum somewhere
along the jet. In this area of the jet, the charge convection increases very fast, and the conduction current decreases very fast. In this area the total value of the current is determined. So, the ratio of the axial liquid velocity at the liquid surface over the average velocity in that area of the jet determines the increase in the current compared to a liquid with a flat liquid velocity profile. This velocity profile depends on the tangential electric stress and the viscous shear stress. The tangential electric stress was evaluated in the following way. The influence of various parameters on the maximum electric field strength was investigated with the Cone-Shape model. This investigation resulted in the following relation

\[ E_z = E_{ref} \left( \frac{Q}{Q_{ref}} \right)^{-0.44} \left( \frac{K}{K_{ref}} \right)^{-0.15} \left( \frac{\gamma}{\gamma_{ref}} \right) \]  

(7.17)

where, \( E_{ref} = 4 \times 10^6 \) V m\(^{-1} \), \( Q_{ref} = 1.4 \times 10^9 \) m\(^3\) s\(^{-1} \), \( K_{ref} = 69 \) µS m\(^{-1} \), and \( \gamma_{ref} = 0.048 \) N m\(^{-1} \). These constants were chosen, because in the simulations, a case was used with these experimental conditions. The dependency on the parameters was investigated by changing the parameters one by one. For liquids with a relatively flat radial profile of the axial velocity, the accuracy of this relation was about 20 percent. Deviations were mainly caused by the effect of the nozzle diameter on the electric field, and the applied potential difference. However, these effects cannot be easily caught into an equation, so they were neglected. For liquids with an almost flat radial profile of the axial velocity, the maximum axial electric field strength always appeared close to the position where the charge convection current was equal to 59 percent of the total current. This result yields

\[ r_j = \left( \frac{0.41}{\pi E_z K} \right)^{1/2} \]  

(7.18)

where, \( r_j \) is the radius of the jet at the position where the conduction current is equal to 41 percent of the total current [m]. The surface charge at that position then equals

\[ \sigma = \frac{0.59 I_s r_j}{2Q} \]  

(7.19)

For liquids with a steeper axial velocity profile, the scaling of the last three equations was less accurate. However, in the investigated cases, and when looking at the influence of this scaling on the velocity profile in the jet, then the influence of the over and underestimations of the components in these equations were averaged out. So, this scaling seems to yield a suitable basis
to continue the calculation of the axial velocity at the jet surface.

The tangential stress, due to the axial electric field strength, and the surface charge, is passed into the liquid by means of the shear stresses in the liquid. These shear stresses consists of two contributions. The first contribution is due to the viscosity. For viscosity, the tangential stress equals

$$\tau = \mu \frac{\partial u_r}{\partial r} \quad (7.20)$$

The second contribution comes from the velocity pressure. Equation (7.3) shows that the faster the liquid, the slower the increase in the velocity. So, for low viscous liquids with a much higher axial velocity at the surface than in the center of the jet, this effect will reduce the steepness of the velocity profile. In order to scale this effect, the radial liquid velocity has to be known. At the liquid surface, this velocity can be calculated from the axial velocity in the following way

$$u_r = \frac{dr}{dz} u_r \quad (7.21)$$

Also, the continuity equation for an axisymmetric jet at the liquid surface is needed. This relation yields

$$2 \frac{\partial u_r}{\partial r} + \frac{\partial u_z}{\partial z} = 0 \quad (7.22)$$

In the jet, the velocity pressure is dominant over the surface tension stress and the normal electric stress. When these two stresses are neglected, in equation (7.3), and when the last two equations are used, then a scaling can be made for the influence of the velocity pressure on the axial velocity profile

$$\mu \frac{\partial u_r}{\partial r} - \frac{1}{2} \rho u_r r \frac{\partial u_z}{\partial z} = -\rho Q \frac{dr}{nr_j} \frac{\partial u_r}{\partial r} \quad (7.23)$$

This scaling yields

$$\mu' = \mu + c \rho \frac{Q}{nr_j} \frac{dr}{dz} \quad (7.24)$$

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where, \( c_p \) is a constant and \( \mu' \) is the effective viscosity. If the normal electric stress in the axial momentum equation (7.3) is neglected, then this equation can be used for the calculation of the derivative of the liquid surface of the cone and jet.

\[
\frac{dr_j}{dz} = \frac{\sigma E_z}{\gamma + \frac{\rho Q^2}{2r_j \pi^2 r_j^4}} \tag{7.25}
\]

If it is assumed that the velocity profile in the jet can be described by a parabolic profile, then the liquid velocity at the liquid surface can be calculated with

\[
u_{z, surface} = Ar_j^2 + B \quad A = \frac{E_z \sigma}{2\mu' r_j} \quad B = \frac{Q - 0.5\pi r_j^4 A}{\pi r_j^2} \tag{7.26}\]

If the velocity at the liquid surface is known, then the following equation can be used to calculate the current though the liquid cone.

\[
I = \pi r_j^2 E_z K + 2\pi r_j \rho u_{z, surface} \tag{7.27}\]

The current calculated with equation (7.27) is valid for Newtonian liquids independent of the velocity profile in the jet. This velocity profile can either be flat or steep. In order to obtain this current \( r_j \) is calculated with equations (7.16), (7.17), and (7.18), \( \sigma \) with equations (7.16), (7.18), and (7.19), \( E_z \) with equation (7.17), and \( u_z \) with equations (7.24), (7.25), and (7.26).

### 7.4 THEORETICAL SCALING OF THE DROPLET SIZE IN THE VARICOSE JET BREAK-UP REGIME

The droplet size depends on the jet radius and on the jet break-up mechanism. Rayleigh’s jet break-up theory (1878) can be used for low viscous jets with a constant jet velocity. In this theory, a small harmonic perturbation is applied on the surface of the jet. In case of a jet that breaks up due to axisymmetric varicose instabilities, the one dimensional axial momentum equation, and a relation for the surface tension, can be used to derive the following equation

\[
\frac{r_{jet}^3}{\gamma} \omega^2 = \frac{1}{2} k^2 r_{jet}^2 (1 - k^2 r_{jet}^2) \tag{7.28}\]
where, \( r_p \) is jet radius \([m]\), \( \omega \) is the growth rate of the disturbance \([s^{-1}]\), and \( k \) is the wave number of the perturbation \([m^{-1}]\). This equation yields the time constant for droplet formation to be

\[
\tau_d = \left( \frac{r_{jet}^3 \rho}{\gamma} \right)^{\frac{1}{2}}
\]

(7.29)

In an accelerating jet, the perturbation wavelength grows with increasing velocity. As long as the acceleration time constant is shorter than the droplet formation time constant, the perturbation wavelength increases too fast. There is not enough time for the jet break-up to occur. However, with increasing velocity, the acceleration time constant increases, and the droplet formation time constant decreases. As a result, somewhere along the jet, the droplet formation time will be in the same order of magnitude as the acceleration time constant. In that case, there is enough time for the jet break-up. Also, down the jet, the velocity pressure is much larger than the surface tension stress, and the normal electric stress. So, these two stresses can be neglected. In the jet, the charge convection current is almost equal to the total current. These assumptions, and equations (7.3), (7.5), (7.6), and (7.8), yield

\[
\left( \frac{\gamma}{\varepsilon_0 r_{jet}} \right)^{\frac{1}{2}} \frac{r_{jet} I}{Q} \frac{2E'_s \sigma'}{r_{jet}' s} = \frac{\rho Q}{r_{jet}^2 \tau_d} u'_z \frac{\partial u'_z}{\partial z'}
\]

(7.30)

Equations (7.29) and (7.30) yield that the characteristic jet radius at the position where the jet breaks up into droplets \( r_{jet} \) \([m]\) equals

\[
r_{jet}^3 \sim \left( \frac{\varepsilon_0 \rho Q^4}{I^2} \right)^{\frac{1}{2}}
\]

(7.31)

When it is assumed that droplet diameter relates directly to the jet radius at the jet break-up position, then the droplet diameter scales with

\[
d_d \sim r_{jet} \sim \left( \frac{\varepsilon_0 \rho Q^4}{I^2} \right)^{\frac{1}{6}}
\]

(7.32)

Assuming that the current scales according to equation (7.13), this relation yields
Scaling of Droplet Size and Current

\[ d_d - \left( \frac{\rho e_o Q^3}{\gamma K} \right)^{\frac{1}{3}} \quad \frac{d_d}{d_o} - \left( \frac{Q}{Q_o} \right)^{\frac{1}{2}} \]  

(7.33)

This relation confirms the relation \( d_e \sim Q^{0.48} \) presented in Chapter 6.

### 7.5 THE CURRENT THROUGH THE LIQUID CONE

#### 7.5.1 The Effect of the Ion Species in the Liquid

Chen and Pui (1997) reported that the ion species that are used to enhance the liquid conductivity, influences the current through the liquid cone. Based on order of magnitude comparison, it was claimed that \( H^+ \) ions in pure water yield a deviation of 25 percent of the current for liquids where no ion effect occurs. \( Li^+ \) would yield a difference of 15 percent. This difference was explained from the fact that the ion mobility of \( H^+ \) in pure water is 8 times higher than for \( Li^+ \).

This effect was investigated experimentally. A nozzle to plate electrode configuration was used. The nozzle diameter was 8 mm, and the distance between the nozzle and the plate was 3.4 centimeters. The potential difference was 20 kV for ethylene glycol, and 15 kV for n-butanol.

Table 7.1 lists the liquid properties taken from literature.

<table>
<thead>
<tr>
<th></th>
<th>( \rho ) (kg m(^{-3}))</th>
<th>( \gamma ) (N m(^{-1}))</th>
<th>( \varepsilon_r )</th>
<th>( \mu ) (mPa s)</th>
</tr>
</thead>
<tbody>
<tr>
<td>n-Butanol</td>
<td>810</td>
<td>0.0252</td>
<td>17.8</td>
<td>2.76</td>
</tr>
<tr>
<td>Ethylene Glycol</td>
<td>1109</td>
<td>0.048</td>
<td>37</td>
<td>20</td>
</tr>
</tbody>
</table>

Chemically pure, and technically pure ethylene glycol, have been used. Only, the conductivity of the chemically pure ethylene glycol has been enhanced with additives. The current reaching the counter electrode was measured by means of an oscilloscope. Figure 7.1 shows the current as function of the liquid flow rate for ethylene glycol with different additives. The legend shows the additive and the conductivity of the liquid. In order to correct for the differences in conductivity the scaling of equation (7.13) has been used. Figure 7.2 shows the result of this correction. The currents in this figure are normalized with the fit to the measurements of the technical pure ethylene glycol. This figure clearly shows that the type of additive influences the current considerably. Especially, in cases where \( H^+ \) and \( OH^- \) were used to enhance the conductivity, the current was lower than expected.

In Electrohydrodynamic Atomization in the Cone-Jet mode, the current is calculated from the surface tension, the conductivity and the liquid flow rate. In order to reach a certain liquid
conductivity, a liquid that contains ions with a high ion mobility, will have a relatively low ion concentration. This means that the number of ions in the liquid flow entering the cone is also relatively low. When a positive potential is applied to the nozzle, then negative ions react at the nozzle surface, and positive ions form the free charge at the liquid surface. As a result, the ion concentration in the liquid of the cone is reduced. This reduction in ion concentration reduces the conductivity of the liquid in the cone. That is why, the current is lower than expected. The number of ions entering the liquid cone scales with the flow rate to the power 1. However, the current scales with the power 0.5. So, the effect of the reduction of the conductivity in the cone is stronger at low flow rates. Figure 7.2 confirms that this effect occurs.

![Figure 7.1 The influence of additives on the current for an ethylene glycol cone.](image1)

![Figure 7.2 The influence of additives on the current which is normalized with the current of technically pure ethylene glycol.](image2)

In pure water the ion mobilities are relatively high. Table 7.2 shows the ion mobility of ions in pure water. This table shows that H⁺ and OH⁻ have relatively high mobilities. Figure 7.2 shows that the currents for H⁺ and OH⁻ are the lowest. This is in agreement with the above presented hypothesis. If these ion mobilities are used, then the effects of figure 7.2 can be explained. However, these measurements were not done in water, but in ethylene glycol. Other measurements showed exactly the same effect in n-butanol. In these liquids, the ion mobilities are a factor 10 lower than for water. This was confirmed by the amount of salt that had to be added, in order to reach a certain conductivity. Another important fact is that higher ion mobilities of H⁺ and OH⁻ in water are a result of the special nature of water. In ethylene glycol and in n-butanol, the ion mobilities of H⁺ and OH⁻ are not much larger than for the other ions. These two facts contradict the above presented hypothesis.
Table 7.2 The ion mobility of ions in pure water at room temperature.

<table>
<thead>
<tr>
<th>Ion Mobility ( m^2 \text{ S mole}^{-1} )</th>
<th>Ion Mobility ( m^2 \text{ S mole}^{-1} )</th>
</tr>
</thead>
<tbody>
<tr>
<td>H(^+)</td>
<td>349.7</td>
</tr>
<tr>
<td>Na(^+)</td>
<td>50.8</td>
</tr>
<tr>
<td>Li(^+)</td>
<td>38.7</td>
</tr>
</tbody>
</table>

In a few experiments, the influence of the polarity of the applied potential difference on the current was investigated. For ethylene glycol, no stable cone could be obtained when a negative potential was applied to the nozzle. Electrical discharges in the surrounding air disturbed the formation of a liquid cone. Ethylene glycol has a relatively high surface tension. So, the electric fields around the cone are higher than for liquids with a low surface tension. Since, negative discharges occur at lower electric field strengths than positive discharges, it is possible to spray ethylene glycol with a positive potential applied to the nozzle, where it cannot be sprayed with a negative potential applied. n-Butanol has a much lower surface tension. So, it can be sprayed with a positive as well as with a negative potential difference. Figure 7.3 shows that the polarity of the applied potential difference has no influence on the current. It also has no influence on the effect of H\(^+\) on the current. This is in agreement with the above presented hypothesis, because the liquid in the cone can be considered to be neutral. So, the effect of the reduction in conductivity in the cone only depends on the number of ions and not on the polarity of the applied potential difference.

Concluding, the ion mobilities in pure water can be used to estimate the effect of the ion species on the current for n-butanol and ethylene glycol. However, the real ion mobilities in these liquids are completely different. So, a solid physical explanation of this effect was not found.

7.5.2 Current Scaling for a 1 mm Nozzle

Gañán-Calvo et al. (1997) presented current measurement results for liquids with a low conductivity. A nozzle to plate electrode configuration was used. The nozzle diameter was 1 mm, and the distance between nozzle and plate was a few centimeters. Table 7.3 lists the liquid properties used in these experiments.
The table shows that the viscosity was varied over a wide range. This makes these results very suitable to fit the current scaling of section 7.3 to these measurements. Figure 7.4 shows the currents normalized and measured as function of the normalized flow rate. This figure clearly shows that the current for low viscous heptane is above the current for high viscous sunflower oil and dodecanol. In order to fit the scaling to the measurements, the constants \( a_i \) and \( b_i \) of equation (7.16), and constant \( c_\mu \) of equation (7.24), have to be determined. Table 7.4 shows the result of this fitting.

![Graph showing current as function of normalized flow rate](image)

- ▽ Heptane 0.7 [\( \mu \)S/m]
- ▼ Sunflower Oil
- △ Dodecanol
- □ Heptane 4.5 [\( \mu \)S/m]
- ● Heptane 1.9 [\( \mu \)S/m]
- ■ Dioxane + 4% Formamide
- ▲ Dioxane + 2% Formamide

*Figure 7.4 The current for an 1 mm nozzle for liquids with \( K<10 \) \( \mu \)S m\(^{-1}\), Gañán-Calvo et al.(1997).*

*Table 7.3 Liquid properties taken from Gañán-Calvo et al.(1997).*

<table>
<thead>
<tr>
<th></th>
<th>( \rho ) kg m(^{-3})</th>
<th>( \gamma ) N m(^{-1})</th>
<th>( \epsilon_r )</th>
<th>( K ) ( \mu )S m(^{-1})</th>
<th>( \mu ) mPa s</th>
</tr>
</thead>
<tbody>
<tr>
<td>Heptane + 0.1 % Stadis</td>
<td>684</td>
<td>0.021</td>
<td>1.9</td>
<td>0.77</td>
<td>0.39</td>
</tr>
<tr>
<td>Heptane +0.4 % Stadis</td>
<td>684</td>
<td>0.021</td>
<td>1.9</td>
<td>1.9</td>
<td>0.39</td>
</tr>
<tr>
<td>Heptane +1.0 % Stadis</td>
<td>685</td>
<td>0.021</td>
<td>1.9</td>
<td>4.5</td>
<td>0.39</td>
</tr>
<tr>
<td>Dioxane + 2 % Formamide</td>
<td>1030</td>
<td>0.030</td>
<td>2.3</td>
<td>0.24</td>
<td>1.39</td>
</tr>
<tr>
<td>Dioxane + 4 % Formamide</td>
<td>1030</td>
<td>0.031</td>
<td>2.5</td>
<td>1.1</td>
<td>1.39</td>
</tr>
<tr>
<td>Sunflower Oil + Stadis</td>
<td>920</td>
<td>0.029</td>
<td>3.4</td>
<td>2.4</td>
<td>21</td>
</tr>
<tr>
<td>Dodecanol(doped)</td>
<td>830</td>
<td>0.028</td>
<td>6.5</td>
<td>2.85</td>
<td>12</td>
</tr>
</tbody>
</table>

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Table 7.4 The value of the constants $a_r, b_r,$ and $c_r$ of the scaling for a nozzle with a diameter of 1 mm, and for liquids with $K < 10 \, \mu S \, m^{-1}$.

<table>
<thead>
<tr>
<th>Range of Application</th>
<th>$a_r$</th>
<th>$b_r$</th>
<th>$c_r$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$Q/Q_0 &lt; 70$</td>
<td>0.454</td>
<td>3.49</td>
<td>0.635</td>
</tr>
</tbody>
</table>

Figure 7.5 shows the comparison of this scaling with the measurements. In this figure, the current obtained in the experiments is normalized with the current calculated with the scaling of section 7.3. This figure shows that the accuracy for these liquids and this electrode configuration is about 10 percent. The value of $c_r$ is kept equal to 0.635 in all other calculations.

Figure 7.5 The current measured and divided by the current calculated from the scaling for liquids with $K < 10 \, \mu S \, m^{-1}$.

Gañán-Calvo et al. (1997) also presented current measurement results for liquids with a higher conductivity. Table 7.5 lists the liquid properties used in these experiments. Figure 7.6 shows the currents normalized and measured as function of the normalized flow rate. This figure shows that the influence of the viscosity is much smaller than for the liquids with a lower conductivity. Figure 7.7 shows the comparison of the measurements with the current scaling. In
this case only the constants \( a_i \) and \( b_i \) were determined by fitting the scaling of section 7.3 to the measurements. The constant \( c_p \) was kept equal to 0.635, because the influence of this constant on the current for these liquids is very small. So, a small error in the current measurement would lead to a large mistake in \( c_p \). Table 7.6 shows the result of this fitting.

![Graph](image1)

**Figure 7.6** The current for the 1 mm nozzle for liquids with \( K \geq 10 \mu S \text{ m}^{-1} \) Gañán-Calvo et al. (1997).

![Graph](image2)

**Figure 7.7** The current measured and divided by the current calculated from the scaling for liquids with \( K \geq 10 \mu S \text{ m}^{-1} \).

**Table 7.5** Liquid properties, Gañán-Calvo et al. (1997).

<table>
<thead>
<tr>
<th></th>
<th>( \rho ) \text{ kg m}^{-3}</th>
<th>( \gamma ) \text{ N m}^{-1}</th>
<th>( \varepsilon_r )</th>
<th>( K ) \text{ ( \mu S \text{ m}^{-1} )}</th>
<th>( \mu ) \text{ mPa s}</th>
</tr>
</thead>
<tbody>
<tr>
<td>Dioxane + 25 % Water</td>
<td>1022</td>
<td>0.037</td>
<td>14.2</td>
<td>170</td>
<td>2.05</td>
</tr>
<tr>
<td>Propylene Glycol</td>
<td>1036</td>
<td>0.036</td>
<td>31.2</td>
<td>13.3</td>
<td>30</td>
</tr>
<tr>
<td>Methanol</td>
<td>795</td>
<td>0.021</td>
<td>33.6</td>
<td>85</td>
<td>0.59</td>
</tr>
<tr>
<td>Dioxane + 50 % Water</td>
<td>1015</td>
<td>0.043</td>
<td>34.5</td>
<td>570</td>
<td>2.35</td>
</tr>
<tr>
<td>Dioxane + 66.7 % Water</td>
<td>1010</td>
<td>0.044</td>
<td>49.1</td>
<td>1300</td>
<td>1.95</td>
</tr>
</tbody>
</table>

**Table 7.6** The value of the constants \( a_i \) and \( b_i \) of the scaling law for a nozzle with a diameter of 1 mm, and for liquids with \( K \geq 10 \mu S \text{ m}^{-1} \).

<table>
<thead>
<tr>
<th>Range of Application</th>
<th>( a_i )</th>
<th>( b_i )</th>
</tr>
</thead>
<tbody>
<tr>
<td>( Q/Q_0 \leq 50 )</td>
<td>0.508</td>
<td>2.17</td>
</tr>
<tr>
<td>( 50 &lt; Q/Q_0 &lt; 250 )</td>
<td>0.547</td>
<td>1.87</td>
</tr>
<tr>
<td>( 250 &lt; Q/Q_0 )</td>
<td>0.468</td>
<td>2.53</td>
</tr>
</tbody>
</table>
The difference in the constants between table 7.4 and 7.6 can most likely be explained from the fact that the flow rate for liquids with a low conductivity is higher than the flow rate for liquids with a high conductivity. Also, if the flow rate is the same, then the current through the liquid cone is much lower for the low conducting liquids. As a result, the electrical power for the formation of the cone is lower. That is why, the potential difference has to be increased. Gañán-Calvo et al. (1997) showed that for a 1 mm nozzle, the current increases with increasing potential difference. At the same normalized flow rate, this effect results in a higher normalized current for liquids with a low conductivity.

![Graph showing current increase vs normalized flow rate](image)

- Heptane $K=0.77$, $\mu=0.39$
- Methanol $K=85$, $\mu=0.59$
- Dodecanol $K=2.8$, $\mu=12$
- Prop.Gly. $K=13.3$, $\mu=30$
- Dioxane $K=1300$, $\mu=1.95$
- Dioxane $K=2.4$, $\mu=1.39$
- Heptane $K=1.9$, $\mu=0.39$
- Heptane $K=4.5$, $\mu=0.39$

*Figure 7.8 The current increase due to the increase in the axial liquid velocity at the jet surface.*

Figure 7.8 shows the current increase compared to the current calculated with equation (7.16). This represents the current increase, due to a larger liquid velocity at the jet surface. The figure shows that the lower the viscosity, or the higher the normalized liquid flow rate, the lower the current increase. Surprisingly, the relative current increase for heptane with different conductivities, appears to depend on the value of the normalized flow rate alone. Figure 7.9 shows the normalized value of $\mu^*$ of equation (7.24), using $c_\mu=0.635$. 

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Figure 7.9 The effective viscosity over the absolute viscosity, equation (7.24).

7.5.3 Current Scaling for an 8 mm Nozzle

Chapter 3 presented current measurement results for an electrode configuration, which is the same as the one described in the section about the influence of the ion species. The nozzle diameter was 8 mm. The liquid properties were already listed in Table 7.1. The conductivity ranged from 50 to 200 $\mu$S m$^{-1}$. The constants $a_i$ and $b_i$ of the scaling of section 7.3 were determined by fitting the scaling of section 7.3 to the current measurements. $c_p$ was kept constant at 0.635. Table 7.7 shows the result of this fitting. Figure 7.10 shows the measured current and the current scaling as function of the normalized flow rate.

Chapter 6 presented current and droplet size measurements. In these experiments, a nozzle to plate electrode configuration was used. The nozzle diameter was 8 mm. The distance between nozzle and plate was 3.4 centimeters for the ethylene glycol measurements. In this case, the applied potential difference was 17 kV. For the other measurements, the nozzle-plate distance...
was 2.4 cm, and the potential difference was 10.8 kV. Table 7.8 lists the liquid properties taken from literature.

**Table 7.7 The value of the constants \( a_i \) and \( b_i \) of the scaling law for a nozzle with a diameter of 8 mm.**

<table>
<thead>
<tr>
<th>Range of Application</th>
<th>( a_i )</th>
<th>( b_i )</th>
</tr>
</thead>
<tbody>
<tr>
<td>( Q/Q_o &lt; 50 )</td>
<td>0.493</td>
<td>2.22</td>
</tr>
<tr>
<td>50 &lt; ( Q/Q_o &lt; 250 )</td>
<td>0.518</td>
<td>1.93</td>
</tr>
<tr>
<td>250 &lt; ( Q/Q_o )</td>
<td>0.427</td>
<td>3.20</td>
</tr>
</tbody>
</table>

**Table 7.8 Liquid properties.**

<table>
<thead>
<tr>
<th>Liquid</th>
<th>( \rho ) (kg m(^{-3}))</th>
<th>( \varepsilon_r )</th>
<th>( \gamma ) (N m(^{-1}))</th>
<th>( \mu ) (mPa s)</th>
<th>( K ) (μS m(^{-1}))</th>
</tr>
</thead>
<tbody>
<tr>
<td>n-Butanol</td>
<td>809.8</td>
<td>17.8</td>
<td>0.0246</td>
<td>2.948</td>
<td>15</td>
</tr>
<tr>
<td>n-Butanol + LiCl</td>
<td>809.8</td>
<td>17.8</td>
<td>0.0246</td>
<td>2.948</td>
<td>29</td>
</tr>
<tr>
<td>iso-Butanol</td>
<td>801.8</td>
<td>18.5</td>
<td>0.0230</td>
<td>4.703</td>
<td>13</td>
</tr>
<tr>
<td>iso-Butanol + LiCl</td>
<td>801.8</td>
<td>18.5</td>
<td>0.0230</td>
<td>4.703</td>
<td>30</td>
</tr>
<tr>
<td>2-Butanone</td>
<td>805.4</td>
<td>18.5</td>
<td>0.0246</td>
<td>0.417</td>
<td>45</td>
</tr>
<tr>
<td>Ethylene Glycol+LiCl</td>
<td>1108.8</td>
<td>38.0</td>
<td>0.0477</td>
<td>19.9</td>
<td>21</td>
</tr>
<tr>
<td>Ethylene Glycol + LiCl</td>
<td>1108.8</td>
<td>38.0</td>
<td>0.0477</td>
<td>19.9</td>
<td>52</td>
</tr>
</tbody>
</table>

Figure 7.11 shows the current normalized and measured as function of the normalized flow rate. The relatively high current for 2-butaneone is a result of the low liquid viscosity. In this case, the axial velocity profile is steeper than for the other liquids of table 7.8. Figure 7.12 shows the measured current divided by the current calculated with the scaling of section 7.3, the values of table 7.7, and \( c_i = 0.635 \). This figure shows that the calculated current is again in most cases within 10 percent of the measured currents. The only exception is iso-butanol with a conductivity of 13 μS m\(^{-1}\). The increasing tendency of the ratio of the measured current over current scaling with increasing flow rate, indicates that the ion
Figure 7.11 The current measured for the liquids of table 7.8 sprayed with an 8 mm nozzle.

Figure 7.12 The current measured and divided by the current calculated from the scaling.
effect is playing a role. However, which ion species is responsible is not known. The isobutanol was chemically pure. But this does not mean that no other substances are present in the liquid. A very small pollution of the liquid can easily be responsible for this effect.

7.5.4 The Influence of the Nozzle Diameter

In the previous tables, values were given for $a_i$ and $b_i$ of equation (7.16). Chapter 3 showed that the nozzle diameter and the potential difference have an influence on the current through the liquid cone. Smaller nozzle diameters will yield larger currents. Figure 7.13 shows that this was also the case, when the current scaling for the 1 mm nozzle is compared to the current scaling for the 8 mm nozzle.

7.5.5 The Influence of Liquid Permittivity

When it is assumed that the current scales with $I = Q^{0.5}$ and when $Q/Q_o = 100$ then the value of $b_i$ obtained in the previous sections can be recalculated. $Q/Q_o$ was taken equal to 100 because this a very common ratio for Electrohydrodynamic Atomization in the Cone-Jet mode. This current factor $b_i$ can be compared to the measurements presented by Fernández de la Mora and Loscertales (1994) and Chen and Pui (1997). Figure 7.14 shows this current factor as function of the relative permittivity of the liquid. The figure shows that, due to the scattering in the data, no influence of the permittivity can be detected.

The Cone-Shape model, Chapter 3, has been used to investigate the effect of the liquid permittivity. The electrode configuration is already presented in the section about the influence of the ion species. The liquid properties were equal to the properties of ethylene glycol in table 7.1. The liquid conductivity was 69 $\mu$S m$^{-1}$, the flow rate was $1.4 \times 10^{-3}$ m$^3$ s$^{-1}$, and the applied potential difference was 20 kV. Figure 7.15 shows the effect of the liquid permittivity on the current. This figure shows that the influence is small. Concluding, the effects of the nozzle diameter, the ion species, and the applied potential difference on the current are larger than the effect of the relative permittivity of the liquid. (7.16) to the various measurement results.

![Figure 7.13 Comparison of the fit of equation (7.16) to the various measurement results.](image)
7.6 MAIN DROPLET SIZE

In section 7.4, a scaling of the main droplet size as function of the flow rate, and the current has been presented. This scaling is valid for jets that break-up due to varicose instabilities. The following relation can be used to fit this scaling to experimental results,

$$d_{d, varicose} = c_d \left( \frac{\rho \varepsilon_n Q^4}{I^2} \right)^{\frac{1}{6}}$$  \hspace{1cm} (7.34)

where, $d_{d, varicose}$ is the droplet size for the varicose jet break-up regime, and $c_d$ is a constant. The current scaling presented in section 7.3 can be used to calculate the current. However, in the previous section, it has been shown that the nozzle diameter, the applied potential difference, and the ion species in the liquid, cause deviations from the current scaling. So, it is better to validate relation (7.34) from the measured currents. Gañán-Calvo (1997) not only measured the current for the liquids of table 7.3 but also measured the droplet size by means of a PDA. Figure 7.16 shows the droplet diameter as function of the liquid flow rate. The constant was determined to be

$$c_d = 1.76$$
The figure shows that the droplet size measured for heptane fits very well to the scaling of equation (7.34). For dioxane with 2% formamide, the result was less accurate, but also the measured droplet size was not as stable as for heptane.

Chapter 6 presented droplet size measurements for the liquids of table 7.8. The droplet sizes were measured by means of a high resolution digital camera with a long distance microscopic lens. At low liquid flow rates, the jet was observed to break-up in the varicose jet break-up regime. With increasing flow rate the whipping motion of the jet occurred. So, in order to scale the

Figure 7.16 The measured droplet size, Gañán-Calvo et al. (1997), compared to the varicose droplet size scaling.

Figure 7.17 The droplet size normalized and measured as function of the normalized flow rate.
droplets size, equation (7.2) as well as equation (7.34) has to be used. It was observed that the equation yielding the smallest droplet size is the one that has to be used. If equation (7.34) yields the smallest droplet size then the jet is in the varicose jet break-up regime. If equation (7.2) yields the smallest droplet size, then the jet is in the whipping jet break-up regime. Figure 7.17 shows the droplet size normalized and measured as function of the normalized flow rate.

Figure 7.18 shows the normalized droplet size calculated with equation (7.2) and (7.34). The constant $c_d$ was obtained from these measurements and was found to be equal to

$$c_d = 2.05$$

The main difference between the droplet sizes calculated with equations (7.2) and (7.34) from the measured current and the measured droplet sizes, is the lower scattering in the data. The scattering in the measurement results is a result from two effects. Experiments with polymer particles on a glass plate showed that a badly focussed picture would yield an overestimation of the droplet size by 4 $\mu$m. Since, it is hard to focus on fast moving droplets, this effect cannot be avoided. Secondly, the sizes of only a few droplets were measured. So, statistical mistakes lead to a scattering in the results. The value of $c_d$ was found to be 15 percent higher than for the measurement results of Gañán-Calvo et al. (1997). The difference can be explained from the measurement accuracy. So, very precise droplet size measurements are needed to establish the

![Graph showing normalized droplet size vs. Q/Qo]

**Figure 7.18** The droplet size normalized and calculated from equations (7.2) and (7.34).
correct value of the constant $c_\phi$. Figure 7.19 shows the droplets size measured and divided by the droplet size calculated with equations (7.2) and (7.34), when using the measured current. This figure shows that the droplet size can be calculated well with equations (7.2) and (7.34). This figure also shows that the influence of viscosity is small. The difference between highly viscous ethylene glycol and 2-butaneone with a low viscosity is smaller than the measurement accuracy. This effect can be explained from two effects that seem to keep each other in balance. On the one hand, the jet of a highly viscous liquid is longer. This results in a smaller jet radius at jet break-up, and a smaller droplet size. On the other hand, Chapter 6 showed that the normalized wavelength at jet break-up increases with viscosity. A longer wavelength results in a larger droplet size. Since, the sum of these two effects is small, very precise size measurements are necessary to be able to detect the influence of these two effects on the droplet size that is produced.

### 7.7 CONCLUSIONS

Scaling of the droplet size and current yields relations, which can be easily used to calculate the current and the main droplet size produced in Electrohydrodynamic Atomization in the Cone-Jet mode. A new derivation of the current scaling was made for liquids with a flat radial profile of the axial liquid velocity in the jet. The difference with the present scaling laws, is that the liquid permittivity has completely disappeared from the scaling law. This scaling equals
\[
\frac{I}{I_o} \sim \left(\frac{Q}{Q_o}\right)^{\frac{1}{2}}
\]

The current scaling was extended to liquids with low conductivities and low viscosities. For low viscous and low conducting liquids, the radial profile of the axial liquid velocity is steeper. So, the velocity at the jet surface is larger than this velocity in the jet center. As a result, the charge convection current is larger than in a case with a flat radial profile of the axial liquid velocity. The extension of the scaling law was obtained by scaling this liquid velocity profile in the jet. This was done by using simulation results of the Cone-Shape model and by fitting to experimental results. This extension makes it possible to use this scaling for all Newtonian liquids.

The scaling of the current, was compared to measurements for an 1 mm nozzle, Gañán-Calvo et al. (1997), and an 8 mm nozzle. As predicted in Chapter 3, the fit of scaling relations for the 1 mm nozzle was slightly above the fit of the scaling for the 8 mm nozzle. This is caused by the effect of the nozzle on the electric field around the cone apex. Also, for liquids with a low conductivity K< 10 \( \mu \)S m\(^{-1}\) and a nozzle diameter of 1 mm, the current was relatively larger than expected. The most likely explanation comes from the fact that the lower the conductivity, the lower the electric power, which accelerates the liquid. So, the potential difference has to be increased. For a 1 mm nozzle, the increase in potential will yield an increase in current. For the 8 mm nozzle, no experiments or simulations were conducted at low conductivities. So, whether this effect occurs for the 8 mm nozzle cannot be said.

Inside the liquid, ions are responsible for the charge transport. Measurements proved, that the ion species influences the current for ethylene glycol and n-butanol. For the additive HCl, the current was at low flow rates up to 50 percent lower than for the same liquid with the same conductivity, but with LiCl as additive. Also, NaOH was found to yield a lower current. This effect might be explained from the ion mobility. However, a solid physical explanation was not found.

The polarity of the applied potential did not have an influence on the current. Also, the effect of HCl on the current was not changed by the polarity of the potential difference.

Since, a negative electrical discharge in the air occurs at lower electric field strengths than a positive discharge, some liquids with a surface tension close to 0.048 can be sprayed in air with a positive potential applied to the nozzle, but not with a negative potential applied.

If the effect of the ion species does not occur, and when one electrode configuration is used, then the accuracy of the presented current scaling was about 10 percent for all liquids tested.

The influence of the liquid permittivity on the current is small. The liquid permittivity was varied in a range from 1.25 to 50. The deviation of the current from the average was less than 5 percent.

Also, a relation is derived, which enables to calculate the droplet size from the current.
and the liquid flow rate for the varicose jet break-up regime. This relation equals

\[ d_{d,\text{varicose}} = c_d \frac{\rho e_o Q^4}{l^2} \]

The constant \(c_d\) was found to be equal to 1.76 for the experiments of Gañán-Calvo et al. (1997) and equal to 2.05 for the experiments presented in Chapter 6. Combined with the already presented scaling for the whipping jet break-up regime, this yields a complete main droplet size scaling.

In Chapter 6 it was observed that when the droplet size relation for the whipping jet break-up regime yields the smallest droplet size then the jet breaks up in the whipping jet break-up regime. On the other hand, when the droplet size relation for the varicose jet break-up regime yields the smallest droplets size, then the jet breaks up in the varicose jet break-up regime.

When the measured current is used as an input, then the accuracy of the droplet size relations is about 15 percent. More accurate droplet size measurements are required to find out if this accuracy is even lower than 15 percent.

The liquid viscosity can have a large influence on the current through the liquid cone. The above presented droplet size relation for the varicose jet break-up regime, shows that this influence on the current also changes the droplet size. However, when the droplet size is calculated directly from the current, then the viscosity has little influence on the droplet size. When comparing highly viscous ethylene glycol with 2-butanol with a low viscosity, the difference in the particle size measured and normalized with the particle size calculated from the measured current, is smaller than the measurement accuracy. This effect can be explained from two effects that seem to keep each other in balance. On the one hand, the jet of a highly viscous liquid is longer. This results in a smaller jet radius at jet break-up, and a smaller droplet size. On the other hand, the normalized wavelength at jet break-up increases with viscosity. A longer wavelength results in a larger droplet size.
CHAPTER 8

PRODUCTION OF MONODISPERSE POLYMER POWDER

This chapter describes the production of monodisperse polymer powder by means of electrical hydrodynamic atomization in the Cone-Jet mode. It will be shown that discharging of the particles has no influence on the produced particle size, as long as only a few ions produced for discharging reach the liquid cone. The production of spherical polymer particles is discussed. The influence of the liquid flow rate, of premature solvent evaporation from the liquid cone, and of the initial polymer concentration, on the produced particles will be shown. A low polymer concentration will lead to the formation of elongated particles, or bursted balloons. A high polymer concentration will lead to the formation of long strings, where the produced particles are still connected to each other with thin polymer filaments. The production rates of Electrohydrodynamic Atomization in the Cone-Jet mode are much too low to meet industrial demand. Since, the liquid flow rate is a parameter for the droplet size, the flow rate cannot be used for enhancing the production rate. So, scaling up to industrial production rates can only be reached by increasing the number of spraying points. The influence of neighboring nozzles is investigated, and a first design of a multi nozzle reactor will be presented.

8.1 INTRODUCTION

The results presented in this chapter are just intended as an example of the possibility to produce monodisperse powders by means of Electrohydrodynamic Atomization in the Cone-Jet mode. That is why, some of the explanations of certain observations are of a speculative nature. More research is needed to be able to prove all the assumptions and explanations in this chapter.

The big disadvantage of all production processes for monodisperse powders, based on spraying techniques, is the low production rate per spraying point. Electrohydrodynamic Atomization in the Cone-Jet mode is no exception to this rule. It is impossible to enhance the production rate for one spraying point. Increasing the flow rate, will lead initially to changes in the droplet size, and later to much wider size distributions. Changing the liquid properties only leads to a different range of flow rates, at which the liquid can be sprayed, and to changes in the produced droplet size.
When Electrohydrodynamic Atomization in the Cone-Jet mode is used to produce a monodisperse powder with a certain particle size, then the conductivity of the solution should be in a certain range. The conductivity of the solution can only be controlled if the solute does not have a large effect on the conductivity of the solution. If the effect of the solute on the conductivity is small, then a spray can be produced with a monodisperse size distribution. Evaporation of the solvent from the droplets can lead to a monodisperse particle size. However, the droplets are highly charged. When the solvent evaporates, the droplet diameter is reduced but the amount of charge remains unchanged. The droplet comes closer to the Rayleigh Charge Limit, equation (2.1). This can lead to droplet fission. In that case, the produced particle size distribution will be much wider than the initial droplet size distribution. This problem can be solved in two ways. The first way is by discharging the droplets before most of the evaporation has taken place, Chen et al.(1995). The second way is by using solutions with sufficiently high solute concentrations. In the last case, the droplet becomes solid before droplet fission can take place.

In chapter 6, it was shown that in order to produce a monodisperse spray, it is best to spray a liquid just above the minimum flow rate. The best way to produce a monodisperse spray is to find a liquid, which yields the required droplet size at the minimum flow rate. The minimum flow rate can be roughly estimated from the liquid flow rate $Q_o$, introduced by Gañán-Calvo et al. (1997), equation (2.7). The scaling of chapter 7, and $Q_o$, can be used to calculate the required droplet size. However, in the previous chapters it has been shown that the current through the liquid cone depends on the electrode configuration, the applied potential difference, and on the ion species used to enhance the conductivity of a liquid. When the right solution and the right liquid flow rate are found, then some measurements will be needed to investigate, the lowest possible flow rate, the current, and the produced particles.

The production rates in the Cone-Jet mode are much too low to meet industrial demands. The only way to reach these production rates is by creating thousands of spraying points. This can be obtained in two ways. The first way is by increasing the number of nozzles. The second way is by enhancing the number of spraying points per nozzle, like in the Multiple-Jet mode and the Rim-Emission mode.

In this chapter, the results of a study into the production of polymer powder will be presented. A solution of a solvent and a polymer was sprayed in the Cone-Jet mode. The solvent in the droplets evaporates, and solid particles are formed. The produced droplets are highly charged. The effects of discharging these droplets will be discussed. The influence of premature solvent evaporation from the liquid cone, the influence of the liquid flow rate, and the influence of the polymer concentration will be shown. A first design for a multi nozzle system will be presented.

8.2 MEASUREMENT METHOD

Polymer solutions were sprayed. The applied potentials were just above the minimum onset potential for a stable Cone-Jet mode. The nozzles were cylindrically shaped. At the bottom side,
the nozzle was closed with a flat surface. In the middle of this surface there was a small hole of a few hundred micrometers. The liquid was supplied by means of a syringe pump. The minimum flow rate of this pump was 0.1 ml/h. However, only above 1 ml/h, the liquid flow was sufficiently steady in time. The produced polymer particles were collected on a thin glass plate. The width of the glass plate was 2 cm and the length was 5 cm. The back, and the edges of this glass plate were covered with aluminum foil. The aluminum foil was electrically grounded. In this way, the charge build up on the glass was avoided, and the electric field, due to the image charges on the aluminum foil, and due to the electric charges on the nozzle and the liquid cone, was enhanced. This electric field helped to guide the charged particles to the glass plate. In case of discharged particles, the collection time had to be much longer, because the particles were not guided by the electric field toward the glass slide.

The above described particle collection method has a few disadvantages. Firstly, it is possible, that the collection method favors larger particles compared to smaller particles. If this is the case, then the collection method will lead to the measurement of the right particle sizes, but it will not lead to the right number distribution. Secondly, the charged particles reach the glass plate in a much shorter time than the uncharged particles. So, it is possible that the charged particles still contained solvent at the moment of collection. In that case, the droplet shape could change at impact. This could lead to a larger average size. The collected particles were examined under a microscope, and the size was measured with an image analyzing system. The minimum measurable size was 1 μm. When the distance between the collection position and the nozzle was longer than 10 cm, then all collected polymer particles smaller than 10 μm appeared to be spherical. Thirdly, in the spray, a size segregation effect occurs, see Chapter 5. That is why, the collected size distribution also depends on the measurement position in the spray. Concluding, the collection method, the percentage of solvent in the particles at the moment of collection, the lower limit of the measurement system, and the measurement position in the spray, can all yield deviations between the measured particle size distribution, and the produced size distribution. However, for the comparison between two sprays, this method proved to yield good results.

8.3 DISCHARGING OF THE AEROSOL

Droplets produced with Electrohydrodynamic Atomization in the Cone-Jet mode are highly charged. The solvent evaporates, and particles are formed. For powder production, the highly charged particles can be collected on a grounded plate. Disadvantage of this system is the short residence time of the particles in the air. So, the evaporation of the solvent must be very fast in order to produce dry particles, which do not stick to the plate surface. The particle residence time in the air can be increased by increasing the distance to the grounded plate. However, when the residence time has to be increased by a factor 2, then the potential difference, which is necessary to guide the droplets to the grounded plate, increases with a factor somewhere between 4 and 8. When the distance between the electrodes is only a few centimeters, then the required potential difference is already in the order of kilo volts. So, increasing the residence time in the air will soon lead to unacceptable high potential differences. Only when really small particles have to be
produced, then the evaporation time might be shorter than the residence time in the air. Concluding, for many applications discharging of the aerosol is necessary.

Chen et al. (1995) used a radioactive neutralizer for discharging the droplets. However, this system seems to be only suitable for use in a laboratory. Borra et al. (1996a, 1998a) introduced bipolar coagulation. In this method, both positively charged as well as negatively charged sprays are produced. The coalescence of the charged droplets of opposite polarity yields a discharged particle, and offers the possibility to induce chemical reactions inside the droplets at the moment of coalescence. At this moment, bipolar coagulation is still being developed. So, it is too early to judge the efficiency of discharging and collection.

Noakes et al. (1989), Meesters et al. (1992), and Tang and Gomez (1994) used electrical discharges to produce ions for droplet discharging. Corona discharges were induced in the air at a sharp metal needle. The produced ions have an opposite polarity compared to the droplets. As discussed in chapter 4, these electrical discharges occur in a strong divergent electric field. So, the droplet production as well as the ion production, require specific electric fields, Therefore, at least one extra shielding electrode has to be introduced into the electrode configuration. Figure 8.1 shows the electrode configuration used by Meesters et al. (1992). Tang and Gomez (1994) used a plate with a hole instead of a ring. Noakes et al. (1989) used a grounded plate with a hole. They applied a negative potential to the needle.

![Diagram of electrode configuration](image)

**Figure 8.1** Electrode configuration used by Meesters et al. (1992).

**Figure 8.2** The influence of the level of discharging (from without to severe discharging), on the produced droplet size distribution, measured with the PAS system.

Ethanol was sprayed in an electrode configuration similar to Figure 8.1. However, the needle was not grounded. The level of discharging was controlled by applying a negative potential to the needle. The nozzle diameter was 5 mm, the ring diameter was 5 cm, and the distance between the ring, and the nozzle was 3 cm. The distance between ring, and needle was 9 cm. The liquid flow rate was 5 ml/h. The Particle Analysis System described in Chapter 5 was used to measure the droplet size. The measurement position was 1.2 cm below the nozzle, in the
spray center. Figure 8.2 shows the influence of the level of discharging of the aerosol on the produced droplet size distribution. Without discharging, the droplets move in a straight line in the direction of the needle. The nozzle potential was 8 kV. The ring and the needle potentials were 0 kV. In the other experiments the nozzle potential was 11.7 kV, and the ring potential was 5.1 kV. The little discharging level means that some droplets lose their velocity, but most of them still move in a straight line. The needle potential was 0 kV. With the normal discharging level is meant, that a cloud of droplets is formed. The droplets of this cloud move in all directions. The needle potential was -3 kV. The severe discharging level means that droplets are already discharged close to the liquid cone. The needle potential was -5 kV. Figure 8.2 shows that the average droplet size is only slightly changed for the various levels of discharging. This change is not necessarily caused by the ions produced for discharging the droplets. In Chapter 5, it was shown that the measured droplet size depends on the position in the spray. When the droplets are discharged, the spray becomes more irregular, and the size segregation in the spray disappears. Secondly, discharged droplets lose their velocity, this could result in more evaporation, and smaller droplets, because it takes longer before droplets reach the measurement position. Thirdly, it was observed that there were always some small fluctuations in the droplet size distribution.

It was also attempted to discharge the droplets with a flow of ions, which was perpendicular to the flow of droplets. This was done by putting the needle horizontally at the edge of the spray, not very far from the nozzle and the ring. This resulted in a jet with length of several centimeters, which was visibly thicker. In this case, not only the droplets were discharged, but also the jet at the liquid cone.

When a solution of a polymer in acetone is sprayed in the electrode configuration of figure 8.1, then some particles will be collected on the needle. As a result, the production of ions is reduced. This problem could be solved by introducing an extra metal grid close to the needle. Figure 8.3 shows this electrode configuration. The whole setup was built in a glass reactor. In that way, charge can be collected on the walls. This seemed to stabilize the discharging. If the charge on the walls is positive, then the electric field near the grounded grid increases. As a result, negative ions produced near the needle can pass the grid more easily, and the aerosol is discharged. If too many ions pass the grid, then the wall gets discharged, and the electric field near the grid is reduced. As a result, fewer ions can pass the grid. The liquid flow rate was 5 ml/h. The polymer concentration was 20 weight percent. The acetone evaporated, and the particles were collected on a glass slide. Figures 8.4 and 8.5 show the size distributions collected with and without discharging.

Concluding, aerosol discharging has no influence on the produced particle size, as long as only a few ions produced for the discharging of the droplets can reach the liquid cone.
8.4 THE PRODUCTION OF POLYMER PARTICLES

8.4.1 The Influence of the Liquid Flow Rate

The influence of the liquid flow rate on the produced particle size was investigated in the electrode configuration of figure 8.1. The nozzle diameter was 8 mm. A 20-weight percent polymer in acetone solution was sprayed. The particles were collected on a glass plate.
Figures 8.8 Liquid flow rate 5 ml/h.  
Figures 8.9 Liquid flow rate 2.5 ml/h.

Figures 8.6 to 8.9 show the collected particle size distributions. These figures show two things. Firstly, the size distribution becomes wider with increasing flow rate. Secondly, the modal particle size is smaller at a flow rate of 5 ml/h than at a flow rate of 10 ml/h. However, for a flow rate of 2.5 ml/h, the modal particle size is larger than for 5 ml/h. This is against the normal theory of Electrohydrodynamic Atomization in the Cone-Jet mode, discussed in Chapters 6 and 7. This can possibly be explained from premature solvent evaporation from the liquid cone. At high liquid flow rates, this evaporation can be neglected, but at a flow rate of 2.5 ml/h this is not the case. Due to this evaporation, the polymer concentration in the liquid cone increases. As a result, the flow properties of the liquid change. In another experiment, the polymer concentration has been increased in the initial solution. In that case, the modal particle size also increased with the polymer concentration.

When, the polymer concentration is too high at the moment of production, then thin polymer filaments are formed between the droplets. Figure 8.10 shows an example of these filaments. The figure shows main droplets that are connected to secondary droplets and satellites.

Figure 8.10 Particles collected when the polymer concentration was too high.
In open air the lowest possible flow rate was 2.5 ml/h. At lower flow rates, the concentration of the polymer in the cone became too high, and the solution could not be sprayed. In order to reach lower flow rates, the evaporation from the liquid cone has to be diminished. This can be accomplished by taking a nozzle with a much smaller diameter, and by enhancing the solvent vapor concentration around the liquid cone. The influence of the evaporation was investigated in the electrode configuration, which is shown in Figure 8.11. The nozzle diameter was 5 mm. The evaporation was controlled by an air flow, which was saturated with acetone vapor. Two versions were used, which were almost identical. The only difference was an extra rubber ring. This ring was introduced in version 2 in order to guide the saturated air flow along the cone. This flow was obtained by guiding an air flow through 3 bottles of acetone. The first bottle was heated to 50 °C, while the other two were kept at room temperature. In this way, an almost complete saturation could be achieved. This can be concluded from the condensation of acetone vapor in the tubes toward the cone, from a small amount of condensation above the rubber ring, and from flow rate experiments. In these flow rate experiments no electric field was applied. The droplet frequency was measured as function of the liquid flow rate. In an open setup, the relative decrease in frequency was faster than the relative decrease in flow rate, because a larger

**Figure 8.11** Electrode configuration used for suppressing of premature solvent evaporation.

**Figure 8.12** Flow rate 1 ml/h, 20 % polymer in acetone, version 1.

**Figure 8.13** Flow rate 1 ml/h 20 % polymer in acetone, version 2.
percentage of the solvent evaporated before a droplet fell down from the nozzle. For version 2 of figure 8.11, the relation between the droplet frequency and the liquid flow rate was linear. Figures 8.12 and 8.13 show the particle size distributions produced in the Cone-Jet mode for a liquid flow rate of 1 ml/h, for version 1 without a rubber ring and for version 2 with a rubber ring.

![Microscope picture](image)

*Figure 8.14 Microscope picture of the particles produced in version 1 at a flow rate of 1 ml/h.*

Less premature solvent evaporation from the liquid cone occurred in version 2 than in version 1. So, the polymer concentration in the liquid cone should have been higher in version 1 than in version 2. This is in agreement with the larger modal particle size for version 1 compared to version 2.

The polymer concentration influences the liquid flow properties. So, changes in the liquid flow properties might yield changes in the produced particle size. However, it is also possible, that this change in flow properties reduces the mobility of the ions in the solution. This would lead to a lower conductivity of the liquid, and a lower current through the cone. A lower current results in a larger droplet size. Figure 8.14 shows a microscope picture of the particles produced in version 1 of the electrode configuration of Figure 8.11. The size distribution of Figure 8.14 is given in Figure 8.12.

**8.4.2 The Influence of the Polymer Concentration on the Particle Shape**

In the previous section it was shown that the polymer concentration in the solution influences the produced particle size distribution. However, the polymer concentration was also found to have an influence on the shape of the produced particles. The electrode configuration of figure 8.11 was used to investigate this effect. The nozzle diameter was 5 mm. The distance between the ring and the grounded plate was 21 cm. This distance allows the droplets to be almost dry at the moment of collection. So, they will stick to the surface on which they are collected, but their shape is not changed at the moment of impact. A solution with 13.4 wt % polymer, 53.4 wt % dichloromethane, and 33.2 wt % acetone was sprayed. No air or solvent vapor flow was supplied to the system. Figure 8.15 shows the obtained particles for a liquid flow rate of 3 ml/h. The microscope picture does not show spherical particles, but some kind of bursted balloons.
Figure 8.15 Particles produced with the polymer, dichloromethane, acetone solution at a liquid flow rate of 3 ml/h.

Figure 8.16 Particles produced with the polymer, dichloromethane, acetone solution at a flow rate of 1 ml/h.

Figure 8.17 Particles produced with the polymer, dichloromethane, acetone solution at a flow rate of 0.7 ml/h.

Figure 8.18 Particles produced with the polymer, dichloromethane, acetone solution at a flow rate of 3 ml/h, with supply of an air flow.
The shielding around the nozzle reduces the evaporation compared to a completely open system. However, without the supply of solvent vapor to the nozzle, there will be some premature evaporation from the liquid cone. So, when the liquid flow rate is reduced, then the concentration of polymer in the cone will rise due to premature solvent evaporation. Figure 8.16 shows the microscope picture of the produced particles at a flow rate of 1 ml/h. The figure shows much less bursted balloons. However, some particles are not spherical, but they are elongated. Figure 8.17 shows a microscope picture of particles produced at a flow rate of 0.7 ml/h. The flow rate is even lower, so it can be expected that the polymer concentration in the liquid cone is even higher. In this case, most of the produced particles are spherical. Bursted balloons are not present anymore, and only a few elongated particles are present. The influence of the polymer concentration was also investigated by supplying a strong air flow to the cone. As a result, the evaporation increases. In that case, the polymer concentration in the liquid cone will go up. Figure 8.18 shows the particles produced at a liquid flow rate of 3 ml/h with supply of an air flow.

The elongated particles of figure 8.18, were also observed for a solution of 10 wt % polymer, 54 wt % acetone, and 36 wt % ethyl acetate. The solution was sprayed at a liquid flow rate of 2 ml/h in a completely open system, without shielding. A solution of 18 wt % polymer, 50 wt % acetone, and 32 wt % ethyl acetate sprayed at the same flow rate resulted in spherical particles.

The effects presented in the previous paragraphs can possibly be explained in the following way. If the electric stresses in the droplet surface become larger than the surface tension stress, then fission will take place. So, solvent evaporation brings the droplet closer to the Rayleigh Charge Limit. In the solidifying particle, the forces that keep the particle together increase very fast. If the polymer concentration in the liquid cone is around 20 weight percent, then the polymer particle has become solid before the electric stresses can deform the particle. At a polymer concentration somewhere between 13 and 18 weight percent, the droplet starts, due to the repulsive forces of its charge, to elongate before the solidifying process has been completed. During this elongation process, more solvent evaporates. As a result, the polymer solidifies, and it becomes more difficult to increase the deformation of the droplet. At the same time, the distance between the two ends of the droplet increases. This means a larger distance between the charge on both ends of the droplet. An increasing distance results in a decreasing electric force. As a result, the droplet could become solid during the fission process. This could explain the elongated particles. The bursted balloons of figure 8.15 could possibly be explained from the following effect. During evaporation of the solvent, a polymer layer is formed at the surface of the droplet, while in the center the polymer is still fluid. When the droplet reaches the Rayleigh Charge Limit, then droplet fission takes place. After droplet fission, the solidified polymer layer at the surface of the droplet prevents the droplet from attaining a spherical shape again.

8.5 SCALING-UP TO INDUSTRIAL PRODUCTION RATES

The production rates for Electrohydrodynamic Atomization in the Cone-Jet mode are very low.
In order to produce 100 tons of polymer particles per year at a liquid flow rate of 2 ml/h requires 40000 spraying points. There are two ways to solve this problem. The first way is to increase the number of spraying points per nozzle. The second way is by increasing the number of nozzles.

8.5.1 Multiple-Jet and the Rim-Emission Mode

In Chapter 2, the Multiple-Jet and the Rim-Emission mode were discussed. In some preliminary experiments, a 2 mm nozzle has been used to induce a Multiple-Jet mode with 4 jets. The liquid flow rate was 8 ml/h. It was possible to produce a bimodal aerosol with main droplets in the center of the individual sprays. Secondary droplets were found to be trapped between these sprays of main droplets. However, the system was not very stable. A few times, the number of jets changed, and not every jet received the same amount of liquid. Based on this experiment, and based on the fundamental research of the Cone-Jet mode presented in the previous chapters, the following conclusions can be drawn.

The Rim-Emission, and the Multiple-Jet mode, are similar to the Cone-Jet mode. With respect of the current through the liquid cone, there is one difference. In these two modes, the applied electric fields are stronger than for the Cone-Jet mode. A larger electric field strength will lead to a higher current per jet. So, the current scaling, presented in Chapter 7, has to be extended for the Rim-Emission and the Multiple-Jet mode. A higher current per jet also influences the produced droplet size distribution. However, the same droplet size to current relations are valid.

When the electric field strength, and the liquid flow rate, are equal for each jet, then the Rim-Emission, and the Multiple-Jet mode, can be used to produce an aerosol with a narrow size distribution. However, it is not easy to reach a situation with equal electric fields and equal flow rates for all spraying points. Cox and Kidd (1975), and Zafran et al. (1975) presented results concerning colloid thrusters in which they used these spraying modes in order to obtain larger droplet production rates.

8.5.2 Multiple Nozzle Reactor

The design of a multi nozzle reactor requires two things. Rulison and Flagan (1993) presented a linear array of 8 capillaries, with a grounded counter electrode in which the particles were collected. In this setup, the electric fields were not equal for each individual cone. The cones at the edge of the array showed a different shape than the cones in the center of the array. This could lead to the disturbance of the droplet production process. Secondly, the more jets, the higher the space charge in the gap between the cone and the counter electrodes. The higher the space charge, the higher the required potential differences necessary for the formation of the cone. The space charge in the reactor could also lead to differences in the electric fields at the nozzles. This would lead to the same problem as observed by Rulison and Flagan (1993). The problem of the electric field can be solved by introducing a ring electrode close to the nozzle. In that case, the electric field is determined by the field between the nozzle and the ring. Neighboring nozzles have no longer an influence on the field at the nozzle. The problem of space charge can be solved
in two ways. Firstly, the particles could be collected on an electrode shortly after production. As discussed in section 8.3, this is often not possible, because the solvent in the droplets does not have sufficient time to evaporate. The second possibility is to discharge the particles, and to transport them with a gas flow to the required collection position. Figure 8.19 shows a schematic representation of a multiple nozzle reactor.

![Diagram of multi nozzle reactor](image)

**Figure 8.19** A schematic representation of the multi nozzle reactor.

The principles of the reactor presented in Figure 8.19 have been used in an experimental reactor. This experimental reactor consists of a linear array of nozzles at different distances from each other. The maximum distance was 10 cm and the minimum distance was 5 cm. This allowed to investigate the influence of the distance between the nozzles on the electric fields at these nozzles. The electric field for the droplet production was created between the nozzle and a plate with holes, plate II. The nozzle diameter was 5 mm. The size of the holes was 3.7 cm, and the distance between the tip of the nozzle and plate II was variable between 1 and 1.5 cm. Around the nozzle, and in between plate I, and plate II, there was a cylinder of an electrically insulating material. This cylinder was used to guide gas flow II around the cone from the top of the nozzle. This gas flow was used to avoid discharged droplets from entering the space between plate I and plate II. Gas flow II was introduced because in preliminary experiments in another set-up there were some leakages. As a result, there was a gas flow from the area between plate II and III toward the nozzle, and some of the discharged particles were collected on the nozzle. When gas flow II was applied this collection on the nozzle was not observed.

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The droplets could be discharged with ions produced by corona discharges. These discharges were induced in the electric field between a needle, and a grid. This grid was put on another plate with holes, plate III. The grid was grounded through a large resistance. The resistance was in the order of 200 MΩ. The current through this resistance was about 6 μA. 44 needles were used in an array of 50 cm by 10 cm. An array of needles is required in order to reach a more homogeneous ion distribution in the discharging zone. The needles were not placed directly below one of the nozzles. In this way, it was prevented that droplets of polymer fall down on the grid above a needle when the spraying process was started. The function of the resistance is to stabilize the electric field around the needle. When many negative ions are collected on the grid, then the potential of the grid is reduced. Consequently, the electric field between the needle and the grid decreases, and fewer ions are produced. The distance between each needle, and the grid could be changed individually by changing the vertical position of the needle. In this way, a similar current can be obtained for each needle. In preliminary experiments it had been observed that particles were collected on the bottom side of plate II. In the discharging experiments, presented in section 8.3, it had been observed that the electrically insulating glass reactor seemed to favor the stability of the particle discharging process. That is why, the bottom side of plate II, and the top side of plate III were covered with a layer of electrically insulating material. Many of the produced ions were collected on the grid. However, some ions passed the grid. These ions discharged the droplets. The discharged droplets were transported out of the reactor with gas flow I. Gas flow III was used to avoid that discharged particles pass the grid. When particles pass the grid, they can be collected on the needle. In that case, the electrical discharge around the needle will be disturbed.

In the first experiment a solution of 18 wt % polymer 50 wt % acetone, and 32 % ethyl acetate was sprayed at a flow rate of 2 ml/h, using only one nozzle. Figure 8.20 shows the size

![Figure 8.20](image1.png)  
**Figure 8.20** Particle size distribution, collected without discharging.  

![Figure 8.21](image2.png)  
**Figure 8.21** Particles collected on a filter after discharging.
distribution of the produced particles without discharging. The particles were collected on a glass plate and measured under a microscope. Figure 8.21 shows the collected particles after discharging and after collection on a filter. The blur particles on the picture are of the same shape and size as the other particles. They are just out of focus of the microscope. The particle size distribution was similar to the size distribution of figure 8.20.

In other experiments, the particles were again collected on a glass plate without discharging. These particles were again measured with an optical microscope. The liquid flow rate was 3.5 ml/h. Figure 8.22 shows the size distribution collected in the spray center. Then a second spray was started at a neighboring nozzle at a distance of 5 cm. The liquid cones at both nozzles looked exactly the same. So, there is no visually detectable interference of the electric field of the neighboring nozzle. Figure 8.23 shows the size distribution produced with two sprays. The size distribution is obtained at a few places in the spray. In the spray center, the size distribution was exactly equal to figure 8.22. At the spray edge smaller particles were collected. The next step was to start the
discharging of the particles. Figure 8.24 shows the size distribution collected on a glass slide. The size distribution for the main droplets is very similar to Figures 8.22 and 8.23. Also, the size distribution of the secondary droplets is similar to Figure 8.23. However, the number of secondary droplets is much larger. This could be explained from the collection method. Figure 8.23 shows the particle size distribution, which was collected at only a few different positions in the spray. A few sampling points in a spray with a size segregation can easily lead to a wrong ratio of main droplets and secondary droplets. In case of a discharged spray, the size segregation effect is lost. The spray is a mixture of main droplets and secondary droplets. This could explain the difference between Figure 8.23 and 8.24.

The reactor could be operated for hours without change of the produced particle size distribution. The corona current, produced near the needles, increased during this period with about 10 percent. This current was reduced to its original value by reducing the potential difference between the needle and the grid. A few discharged particles could pass the grid. However, they were collected mainly on the base of the needle, and not on the tip. There was no collection of particles in the area between plate I and plate II. The electrically insulating layer on the bottom side of plate II worked well, because almost no particles were collected on this side of the plate.

When the polymer solution is sprayed without discharging, then a grounded metal plate was put on top of plate III. After a short period of time, a layer of polymer particles was formed on this grounded plate. When the particles were discharged, no layer of polymer particles was directly collected on plate III or on its grid. This indicates, that the particles were really discharged to a large extent, and that this way of discharging can yield a high discharging efficiency. After some time, particles were deposited everywhere in the reactor. The particles, which were collected without discharging, were sticky, and could not be removed from the grounded plate. The discharged particles could be removed easily from the surface on which they were deposited. So, these particles must have been dry at the moment of impact. This indicates that they were discharged, and deposited due to other forces than the electric field. The most likely explanations for this deposition are gravitation, and turbulent gas flows inside the reactor.

**Agglomeration**

In the multi nozzle reactor an aerosol cloud is formed. The particles in this cloud, are positively or negatively charged. Mutual electric attraction could lead to agglomeration. When particles are collected on a glass plate, then the first particles will certainly hit the glass. Particles arriving later, can also hit already collected particles. Under the microscope, such particles look like agglomerates. In one experiment, it was attempted to collect discharged particles during a very short period of time. In this way, the number of particles on the glass plate was very low. Due to this low number, the chance of agglomeration on the glass plate itself was small. The measurement was done in an aerosol cloud produced by two sprays. The aerosol was discharged. The distance between these sprays was 5 cm. In this case, almost no agglomerates were collected. The experiment indicates, that agglomeration in the discharged aerosol cloud is low. However,
qualitative data cannot be deduced from such an experiment.

When more sprays are put next to each other, or when the distance between the nozzles is reduced, then the concentration of particles in the aerosol cloud will go up. A higher concentration means smaller distances between the particles. This will result in more agglomeration. Another important parameter for agglomeration is the particle charge. When positively charged droplets are produced, then the positive charge just depends on the original amount of charge minus the charge of the collected discharge ions. The discharging rate mainly depends on the ion concentration in the air. Some of the particles will attain a negative charge. The amount of negative charge depends on two charging mechanisms. The first mechanism is called Diffusion Charging. The thermal motion of the ions makes it possible for ions to reach a particle with the same charge polarity. The particle charge mainly depends on the particle size, and the ion concentration. The second mechanism is called Field Charging. The electric field changes the charge distribution on the particle. Due to polarization of the particle, one side of the particle will be negatively charged, and the other side will be positively charged. With increasing negative particle charge, the area of positive charge on the particle will decrease. With increasing field strength the area of positive charge will grow. So, the particle charge mainly depends on the electric field, and the particle size. For particles larger than approximately 1 μm, Field Charging is dominant. For smaller particles Diffusion Charging is dominant. Concluding, agglomeration of particles larger than 1 μm can be reduced, by reducing the electric field in the space between plate II and plate III of Figure 8.19. When this is not sufficient to avoid agglomeration, then wider spacing between the nozzles, a higher gas flow to transport the particles, and bipolar discharging, are possible solutions. The first two solutions reduce the particle concentration in the cloud. The third solution brings positively as well as negatively charged ions into the particle cloud. An ion cloud of both positively charged ions as well as negatively charged ions is very effective in reducing the particle charge to values close to zero.

The Concept of a Multi Nozzle Reactor

The above presented experiments show that the multi nozzle reactor of Figure 8.19 can in principle be used for the production of polymer powders. However, in the experiments, the liquid flow rate was supplied by syringe pumps. On an industrial scale this is impossible. There are two ways to solve this problem. The first solution is to reduce the inner diameter of the nozzle. The nozzles can be connected to a common reservoir with a certain liquid pressure. When, the gravitational pressure is not strong enough to induce a liquid flow, an equal and stable flow rate to each nozzle can be obtained. A vital condition is that each nozzle has the same inner diameter. Disadvantage of such a system is the small size of the holes. This can easily lead to clogging, and disturbance of the liquid flow rate. If the viscosity of the liquid is very high, then a larger inner diameter of the nozzle is possible.

The other possible solution is turning the reactor of Figure 8.19 upside down. The nozzles can be connected to a reservoir with a certain pressure. In that case, the inner diameter of the nozzles can be larger. All nozzle tips should be on the same level, because a difference in height
Production of Monodisperse Polymer Powder

will result in a difference in flow rate. The nozzle diameter has to be decreased to at least 1 mm. The electric stresses in the cone are approximately proportional to the inverse of the radius of the cone. When the base of the cone has a diameter of 5 mm, then at the base of the cone the gravitational force is dominant over the electrical forces. As a result the cone will never be stable in time. For 1 mm nozzle the electric forces are dominant in the whole cone, and the cone can be created against the gravitational force. So, a 1 mm nozzle can be used to spray upside down.

For the reactor of Figure 8.19, the smallest possible distance between two nozzles was 5 cm. Smaller than 5 cm was not possible, because the size of the hole in plate II was the limiting factor. For a 5 mm nozzle, this size cannot be reduced a lot. However, when the nozzle size is reduced, then the size of the liquid cone is reduced. In that case, the nozzle can be brought much closer to plate II, and the size of the hole can be reduced considerably.

The neighboring nozzles have an influence on the electric field around the liquid cone. The extend of this influence depends mainly on the ratio of two distances. The first distance is the distance between the nozzle and plate II. The second distance is the distance between two neighboring nozzles. So, when the distance between the nozzle and plate II is reduced with a factor 5, then the distance between the nozzles can also be reduced with a factor 5. In that case, the distance between the nozzles can become equal to 1 cm. However, a limiting factor could still be the particle concentration in the aerosol cloud. With decreasing distance between the nozzles, the particle concentration goes up, which enhances the particle agglomeration in this cloud.

8.6 CONCLUSIONS

Electrohydrodynamic atomization in the Cone-Jet mode can be used to produce a powder consisting of polymer particles with a narrow size distribution.

For large nozzle diameters, 5 to 8 mm, premature solvent evaporation from the liquid cone can cause problems. The polymer concentration in the liquid cone increases. A high polymer concentration will lead to the formation of long strings of polymer. It has been observed, that the produced particles are still connected to each other with thin filaments of polymer. Smaller nozzle diameters, and the supply of a gas flow saturated with solvent vapor to the liquid cone, can be used to reduce this evaporation.

The produced polymer particle size depends on the concentration of the polymer in the solution. A higher concentration yields a larger particle size. The polymer concentration changes the flow properties of the liquid. This could influence the mobility of the ions in the solution. A change in the ion mobility changes the conductivity of the liquid. This effect and the change in the flow properties itself, could explain the larger modal particle size than expected.

Due to solvent evaporation, a charged droplet comes closer to the Rayleigh Charge Limit. When a normal droplet exceeds the Rayleigh Charge Limit, then droplet fission takes place. For a droplet consisting of polymer in a solvent, three different situations were observed. When the polymer solution is sufficiently high, then spherical particles are formed. The particles become solid before droplet fission can take place. When the polymer concentration is lower, then
elongated particles are formed. A possible explanation is that the droplet shape is transformed by the electric stresses. The droplet becomes elongated. During this process, more solvent evaporates, and it becomes more difficult to change the droplet shape. At the same time, the distance between the two ends of the elongated particle increases. An increasing distance results in a decreasing electric force. As a result, the deformation rate is reduced, and the droplet solidifies as an elongated particle. At even lower concentrations, particles are produced, which look like bursted balloons. This could possibly be explained in the following way. When the solvent evaporates, then a layer of polymer is formed at the droplet surface. Due to the droplet charge droplet fission will take place. After droplet fission, this polymer layer at the surface prevents the droplet from returning to a spherical shape. As a result, the produced particles look like bursted balloons.

The production rates of Electrohydrodynamic Atomization in the Cone-Jet mode are much too low to meet industrial demands. Since, the liquid flow rate is a parameter for the droplet size, and since each liquid has a maximum flow rate, below which a narrow size distribution can be obtained, the flow rate cannot be used for enhancing the production rate. So, the scale-up to industrial production rates can only be reached by increasing the number of spraying points. A multi nozzle reactor was presented. The principles of this reactor can be used for the scale-up to industrial production rates.

Discharging of the produced aerosol has no influence on the produced particle size, as long as only a few ions produced for the discharging of the droplets can reach the liquid cone.

The droplet discharging method in the multi nozzle reactor works well. So, a high discharge efficiency is possible.

If the distance between the nozzle and plate II in Figure 8.19 is much smaller than the distance between two neighboring nozzles, then the neighboring nozzles have little influence on the electric field at the nozzle. For a ratio of the distance between plate II and the nozzle over the distance between two neighboring nozzles equal to 0.2, the influence of the neighboring nozzle on the electric field could not be detected.

The Rim-Emission, and the Multiple-Jet mode, are similar to the Cone-Jet mode. Due to stronger electric fields the current per jet will be larger. So, the current scaling, presented in Chapter 7, has to be extended for the Rim-Emission and the Multiple-Jet mode. On the other hand, the same droplet size to current relations are valid as for the Cone-Jet Mode.

When the electric field strength, and the liquid flow rate, are equal for each jet, then the Rim-Emission, and the Multiple-Jet mode, then it is theoretically possible to produce an aerosol with a narrow size distribution.
CHAPTER 9

CONCLUSIONS

9.1 CONCLUSIONS

Electrohydrodynamic Atomization in the Cone-Jet mode can be modeled by means of several physical models. Four physical models were developed, and found to be in most cases in agreement with experimental results. The Cone-Shape model, described in Chapter 3, simulates the processes inside the liquid cone and jet. For liquid cones with a flat radial profile of the axial liquid velocity in the jet, the model predicts accurately the cone shape, the surface charge distribution, the electric fields in and outside the liquid cone, and the current through the liquid cone. Two jet break-up models were presented in Chapter 6. These models are able to predict most of the phenomena that occur in the varicose jet break-up regime. The Mathematical Jet Break-Up Model is able to predict the main droplet size, the jet velocity at break-up, and the total charge of one main droplet and the accompanying secondary droplets and satellites. When for each main droplet one secondary droplet is produced, then the Numerical Jet Break-Up Model predicts the sizes and charges of main and secondary droplets. The Spray Development Model, presented in Chapter 5, simulates the evolution of the spray produced in the Cone-Jet mode. The model calculates the trajectories of the charged droplets. It is able to show the size segregation effect in the spray, and the dispersion of the droplets in the radial direction.

Due to the droplet charge, a space charge exists in the gap between the liquid cone and the counter electrode. The influence of this space charge on the electric field at the cone surface, can be simulated by neglecting this space charge, and reducing the potential difference with approximately 5 to 7 percent. So, the influence of the space charge is relatively small.

In Chapter 4, it was concluded that when the total current produced by electrical discharges is of the same order of magnitude or larger than the current through the liquid cone without discharges present, then the influence of electrical discharges on the droplet production process cannot be neglected anymore. When the discharge current consists of pulses of charge, then the droplet production is completely disturbed. When the discharge is in the glow mode, then it is sometimes possible to produce a spray with a narrow size distribution. However, the produced droplet size and charge will be different from the Cone-Jet mode without discharges present. The highest discharge probability can be found near the cone apex, where the jet emerges.
from the cone.

The Cone-Jet mode produces droplets with a high droplet charge. Due to the electric forces between these droplets, and due to differences in inertia, a size segregation effect occurs in the spray just after production. In the first few centimeters of this spray, air velocities of several meters per second can occur. When in the spray an area of low droplet concentration occurs, then the aerosol has a bimodal size distribution. The jet breaks up in the varicose jet break-up regime or in the early stages of the whipping jet break-up regime, because the area with a low droplet concentration only occurs when the amplitude of the whipping motion of the jet is small.

In Chapter 8 a first design of a reactor for the production of monodisperse polymer powder is presented. The influence of neighboring nozzles can be reduced by bringing a plate with holes close to the nozzles. It is shown, that it is possible, to produce a polymer powder with a narrow size distribution. Discharging of the droplets does not have an influence on the droplet production, as long as only a few ions used for discharging the droplets can reach the liquid cone. High discharging efficiencies seem to be possible. The initial polymer concentration in the solvent has a strong influence on the shape of the produced particles. At high concentrations, strings of polymer are produced instead of droplets. At low polymer concentrations, the particles are elongated or look like bursted balloons. Spherical particles are produced at a polymer concentration of about 20 percent.

The Rim-Emission and the Multiple-Jet mode are the most promising spraying modes for the production of an almost monodisperse aerosol at higher production rates. However, they are more difficult to control than the Cone-Jet mode. For applications that require sprays with truly monodisperse size distributions, the Cone-Jet mode seems to the most promising spraying mode.

9.1.1. Parameters Influencing the Cone-Jet Mode

The objective of this thesis was to investigate the parameters that influence the droplet size, the width of the droplet size distribution, and the current through the liquid cone. The results of computer simulations and experiments helped to improve the insight into the phenomenon of Electrohydrodynamic Atomization in the Cone-Jet mode. The influence of the various parameters on the droplet size, the width of the droplet size distribution, and the current through the liquid cone could be established.

New droplet size and current scaling laws were presented in Chapter 7. The scaling is extended to low viscosity and low conductivity liquids. These liquids have a higher axial velocity at the jet surface than in the jet center. When accounted for the electrode configuration, and the ion species used to enhance the conductivity of a liquid, then this scaling deviates less than 10 percent from the measured current for Newtonian liquids. A relation is presented which enables to calculate the main droplet size in the varicose jet break-up regime from the current and the liquid flow rate. The accuracy of this relation is better than 15 percent. In Chapter 6, a relation is presented which enables to calculate the main droplet size in the whipping jet break-up regime from the current and the liquid flow rate. The transition between the varicose and the whipping
Conclusions

jet break-up regime can be found at the flow rate where the droplet size relations for the whipping jet and the varicose jet break-up regime both predict the same droplet size. At lower flow rates, the relation for the varicose jet break-up regime predicts the smallest droplet size. At higher flow rates, the relation for the whipping jet break-up regime predicts the smallest droplet size. So, when the droplet size is calculated for both regimes then the smallest droplet size is the right droplet size.

The scaling laws quantify the influence of the liquid conductivity, the surface tension, the viscosity, the liquid flow rate, the liquid density, and the nozzle diameter, on the droplet size, and on the current through the liquid cone. The influence of these parameters will be discussed more qualitatively in the following sections.

Liquid Flow Rate

When the liquid properties are kept constant but the liquid flow rate increases, then the current through the liquid cone increases. This effect occurs because a higher flow rate requires more electric power to accelerate the liquid in the jet. As a result, a new equilibrium is found with a larger jet diameter, and a larger current through the liquid cone. The pressure inside the jet mainly depends on the jet diameter, and on the surface charge density on the jet. The surface charge comes from the current through the liquid cone. The liquid surface tension and the jet diameter determine the surface tension stress in the jet surface. The surface tension stress increases the pressure in the jet. The surface charge induces an electric stress in the jet surface. This electric stress reduces the pressure in the jet. Below the minimum flow rate for a stable Cone-Jet mode, the jet diameter is small, the surface charge, and the kinetic energy of the jet are low. As a result, the pressure in the jet is relatively large compared to the velocity pressure of the jet. In that case, the cone is unstable, because small disturbances of the process can cause the conical shape to be relaxed to a normal droplet shape. The atomization process is not in the Cone-Jet mode. With increasing flow rate, the jet diameter, and the kinetic energy of the jet increase. On the other hand, the surface charge density changes only slightly. The atomization process becomes stable in time, because the velocity pressure of the jet becomes dominant over the surface tension stress. Five different jet break-up regimes have been observed. Which jet break-up regime occurs, depends on the ratio of the electric stress over the surface tension stress. At low flow rates this ratio is relatively low. The jet breaks up due to axisymmetric or varicose instabilities. In that case, the first jet break-up regime has been observed. At jet break-up, the liquid filament between two main droplets flows into the last produced main droplet. If each perturbation on the jet has the same wavelength, then a truly monodisperse spray can most probably be produced. However, natural perturbations on the jet are not of equal length. As a result, every now and then a secondary droplet is produced. This effect of secondary droplets flowing into one of the main droplets might be the result of the small jet diameter. Uncharged jets with a diameter of about 1 mm, that break-up into droplets due to perturbations with the same dimensionless wave number as observed in the Cone-Jet mode, are reported to produce relatively large secondary droplets. With increasing flow rate, the jet diameter increases, the surface charge density remains
approximately constant, and the surface tension stress decreases. So, the ratio of the electric stress over the surface tension stress increases. With increasing stress ratios the other jet break-up regimes occur. The second jet break-up regime occurs when the filament between two main droplets breaks a second time, before it can flow into the main droplet. Secondary droplets are produced. The third regime occurs when the filament between a main droplet and a secondary droplet breaks two times. In this case, main droplets, secondary droplets, and satellites are produced. The transitions between these first three regimes are not very sharp, because the wavelength of the consecutive perturbations fluctuate around an average. When all experimental parameters are kept constant, then the ratio of the electric stress over the surface tension stress is constant. In that case, a longer wavelength is likely to give rise to more secondary droplets and satellites than a shorter wavelength. In the second and third regime, the electrical forces become more important. So, probably due to the repulsive electric force, the secondary droplets cannot flow back into the main droplets. The fourth regime occurs when asymmetric kink instabilities become more important. The secondary droplets, and the filaments between the main droplets, start to leave the jet axis. This regime was called the whipping filament regime. The fifth regime occurs when the whole jet started to make a swirling whipping motion. In the last two regimes, the width of the main droplet size distribution increases with increasing liquid flow rate.

In the previous section it was shown that when a monodisperse spray has to be produced, then it is best to choose a flow rate just above the minimum flow rate. In Chapter 4 the influence of electrical discharges in the air was discussed. These discharges can completely destabilize the droplet production process. One of the conclusions in this chapter was that the discharge probability increases with increasing flow rate. So, it is again best to choose a flow rate just above the minimum flow rate. The third possible reason to choose a flow rate just above the minimum flow rate is the acceleration of the air inside the spray just after production. The air velocity in the spray depends on the current through the cone. The air velocity in the spray increases with increasing current.

The phenomena discussed in this paragraph, which occur as function of flow rate, occur for all liquids. The liquid properties, the nozzle diameter and the potential difference, influence the flow rate at which a certain phenomenon will occur. These parameters also influence the current, and droplet size produced at this flow rate.

**Conductivity**

The liquid conductivity is the main parameter for the droplet size and the flow rate at which the above described phenomena occur. When the conductivity increases, then the current through the jet increases. As a result the electric energy of the jet increases, the surface charge density increases, and consequently the kinetic energy of the jet increases. As a result the minimum flow rate decreases, and the ratio of the electric stress over the surface tension stress increases. Since it is best to spray just above the minimum flow rate, the flow rate has to be reduced. So, the jet diameter decreases and smaller droplets are produced. The liquid conductivity is the main parameter to control the droplet size that is produced.
Conclusions

Surface Tension

With increasing surface tension, the current through the liquid cone will increase, because the electric stresses in the liquid cone surface have to become larger in order to obtain a conical shape. Since the surface tension determines the pressure inside the jet, the minimum flow rate will increase with increasing surface tension. However, the variation in the surface tension between several liquids is small compared to the possible variation in the liquid conductivity. So, the influence of the surface tension on the current, and the droplet size, has been incorporated into the scaling laws. However, this parameter cannot be used to control the produced droplet size.

On the other hand, the liquid surface tension has a large influence on the probability for electrical discharges in the air. Liquids with a larger surface tension than approximately 0.05 N m\(^{-1}\) cannot be sprayed in air without the occurrence of discharges. This threshold value for the surface tension is not a fixed value, because it also depends on the liquid flow rate, and on other parameters that influence the current through the liquid cone.

Density

The equations for the minimum flow rate in Chapters 2, and 7, indicate that with increasing liquid density, the minimum flow rate decreases. However, for the liquids used in this thesis, the differences in density are small. The old scaling laws indicated that the density could be neglected as a parameter. The new scaling laws presented in Chapter 7 show that the liquid density has a small influence on the droplet size in the varicose jet break-up regime. This could be expected, because the density is a parameter in the acceleration of the jet, and the jet diameter at jet break-up determines the produced droplet size.

Electrical Permittivity

The old scaling laws indicated that the electrical permittivity of the liquid was an important parameter. Computer simulations and a new derivation of the scaling law revealed that the influence of the liquid permittivity on the current is small. The influence of the permittivity can be neglected at this moment, because the influence of ion species, and of the electrode configuration, creates more uncertainty about the current than the electrical permittivity.

Viscosity

Viscosity was found to have little influence on the current, and the droplet size, as long as the radial profile of axial liquid velocity in the jet was almost flat. With increasing viscosity, the wavelength at jet break-up increases as can be expected from jet break-up theory. However, with increasing viscosity, the jet break-up also becomes slower. As a result the jet becomes longer, and has more time to be accelerated by the electric field. So, at jet break-up the jet diameter has decreased. As a result the increase in droplet size due to a longer wavelength is largely
compensated by a decrease in the jet diameter. So, the droplet size is almost not affected by viscosity.

When the liquid properties are kept constant, then there is a threshold value for the influence of the viscosity. When the viscosity is below this threshold value, then the radial profile of the axial liquid velocity is not flat anymore. The velocity at the jet surface is larger than in the jet center. As a result, the charge convection current is larger, and the total current is larger. A larger current means also a smaller droplet size. The threshold value for the influence of the viscosity depends on the jet diameter. This jet diameter depends mainly on the liquid flow rate and the liquid conductivity.

Ion Species

Chapter 7 shows the influence of the ion species on the current. HCl has been added to n-butanol and ethylene glycol. HCl as additive yields up to 50 percent lower currents, compared to the same liquids, with the same conductivity, but with LiCl as additive. A solid physical explanation of this phenomenon was not found.

Applied Potential Difference and Electrode Configuration

The influence of the potential difference on the current, and the droplet size, depends on the nozzle diameter. The normal electric field at the cone apex depends on the surface charge density, and on the shape of the jet at the cone apex. The tangential electric field is determined by two contributions. The first contribution is again the surface charge and the shape of the jet at the cone apex. The second contribution is the electric field due to the electrode configuration, and the nozzle in particular. The tangential electric field determines the current through the liquid cone. For a large nozzle the tangential electric field at the cone apex is mainly determined by the shape and the surface charge of the liquid cone. When the potential difference is increased, then at first the tangential electric field increases. However, this new electric field enhances the transport of liquid toward the jet. As a result, the cone becomes smaller. A smaller cone means a lower tangential electric field. The electric field decreases until a new equilibrium is found. For a nozzle with a large diameter, and compared to the initial situation, the electric field at the cone apex has only slightly increased. In case of a nozzle with a smaller diameter, the tangential electric field at the cone apex is also determined by the nozzle. When the potential difference is increased, then the contribution of the nozzle to the tangential electric field at the cone apex also increases. However, when the cone becomes smaller, then the contribution of the nozzle to the electric field at the cone apex remains approximately the same or maybe increases a bit, while the contribution of the cone shape and charge density decreases. So, for a large nozzle, the electric field at the cone apex does not increase much with an increasing potential difference. Consequently the current through the liquid cone does not increase much. On the other hand for a small nozzle, the electric field at the cone apex increases with increasing potential difference. Consequently, the current increases with increasing potential difference. With increasing current,
the jet break-up can come into another jet break-up regime. This can influence the droplet size
distribution that is produced. Chapter 3 showed that a 1 mm nozzle can induce a 15% higher
current than an 8-mm nozzle. This difference can also be explained with the contribution of the
nozzle to the electric field at the cone apex.

When a large nozzle and a relatively large potential difference are used, then the liquid
cone is relatively small. In that case, it has been observed that the potential difference starts to
have an influence on the current. This means that in the Multiple-Jet or the Rim-Emission mode,
the current per spraying point is relatively large compared to the situation with the same nozzle
with only one large cone attached. A larger current for the same flow rate means a higher stress
ratio of the electric stress over the surface tension stress. This means that the size distribution of
a spray produced in the Rim-Emission or the Multiple-Jet mode is most likely wider than for a
spray produced in the Cone-Jet mode using the same nozzle.

In order to reduce the current it is best to use a nozzle with a large diameter, and with a
relatively low potential difference applied. This will yield the most narrow size distribution.
However, nozzles with large diameters also have two disadvantages. Firstly, there is more
premature evaporation from the liquid cone. This yields especially a problem at low flow rates.
Secondly, these nozzles require a larger distance between two neighboring nozzles. Another
effect of a large nozzle diameter is a smaller divergence of the electric field, while the electric
field at the cone apex is approximately independent of the nozzle diameter. As a result, the air
velocity inside the spray just after production, and the probability of electrical discharges in the
air, are larger. When these disadvantages cause too many problems, then it is best to use a nozzle
with a smaller diameter, and to reduce the potential difference as much as possible.

9.2 POSSIBLE FUTURE RESEARCH

9.2.1 Improvements of the Physical Models

The Cone-Shape model, described in Chapter 3, simulates the processes inside the liquid cone
and Jet. The model predicts accurately the cone shape, and the current through the liquid cone
for liquid cones with a flat radial profile of the axial liquid velocity in jet. For liquids with a
steeper radial profile of the axial velocity the model predicts the right trends but needs still some
improvement. This improvement has most likely to be found in the procedure which estimates
the liquid velocity profile inside the liquid cone.

The two jet break-up models presented in Chapter 6 are able to predict most of the
phenomena that occur during jet break-up in the varicose jet break-up regime. For the
Mathematical Jet Break-Up Model, the initial conditions still need more investigation. These
initial conditions determine the position on the jet where the perturbations start to grow. More
pictures have to be taken from the jet break-up, but this time in combination with accurate droplet
size measurements. The Numerical Jet Break-Up Model still needs some improvement with
respect of the formation of secondary droplets and satellites. The model is able to predict the size
of the secondary droplets. However it is not able to predict whether the filament between two
main droplets flows into one of the main droplets or not, and the model is not able to predict the formation of satellite droplets.

The Spray Development Model presented in Chapter 5 is able to show the size segregation effect in the spray, and the model is able to show the dispersion of the droplets in the radial direction. The model can be improved by including a simulation of the air flow in and around the spray. Comparison between the measurements and the simulations showed that in the spray center at 1 cm from the cone apex, air velocities occurred up to 8 m s⁻¹. This air flow disturbs the evolution of the spray.

9.2.2 Improvements of the Scaling Laws
Presented in this Thesis

The scaling laws presented in Chapter 7 are an improvement compared to the previous scaling laws. However, also the new droplet size and current laws need further improvement. The droplet size laws for the varicose and the whipping jet break-up regime still have to be verified with more precise droplet size measurements. The current scaling laws still needs further evaluation with respect of the prediction of the increase in the charge convection current due to a higher axial liquid velocity at the jet surface than at the jet center. In the new scaling law, this increase was partly obtained by fitting to simulation results. This fitting has to be evaluated in more detail. The influence of the ion species on the current has been observed in Chapter 7. However, a solid explanation was not found. When a solid explanation is found, this phenomenon could possibly be implemented in the current scaling law. The scaling laws have been derived for two nozzle diameters. Further improvement of the scaling laws can be obtained by really incorporating the nozzle diameter into the current law.

In one of the previous paragraphs, it was concluded that the most narrow size distribution can be obtained just above the minimum flow rate for the Cone-Jet Mode. In Chapter 7 a relation was derived to estimate this minimum flow rate. More research aimed at this subject has to be done to verify this relation and to improve it.

9.2.3 Possible Future Research

Electrohydrodynamic atomization in the Cone-Jet mode can be scaled up for the industrial production of a monodisperse powders or can be scaled up for other applications. The main task is to find an application which has commercial potential, because quite some engineering has to be done.

The investigation of the wavelength at jet break-up in chapter 6 revealed that quite some variation occurred in the wavelength at jet break-up. When an application has to be developed, which requires a very narrow size distribution, or when the jet break-up regimes have to be studied in more detail, then field modulation with an AC field superimposed on the DC field will be of interest.


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