An Investigation into Methods to Control Breakup and Droplet Formation in Single and Compound Liquid Jets

by

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Abstract

The disintegration of a thread of fluid into droplets is ubiquitous in modern engineering applications and research in this area stretches back over two centuries. However, understanding the mechanism, and as a consequence being able to control, the breakup and subsequent droplet formation of a liquid jet remains both an inherently interesting and challenging problem in modern science. Whilst the last few decades have seen a growth in novel techniques for droplet generation, in many such cases, these methods are poorly understood. In this thesis we investigate a number of techniques for controlling rupture of jets, including industrial prilling with non-Newtonian fluids, the use of insoluble surfactants, controlled insonification and thermal modulation. In addition we examine the dynamics of rupture in two-fluid or compound threads as well as the breakup of jets on the microscopic scale using the theory of interface formation. We find that in all these cases, a greater degree of control and ability to manipulate droplet sizes and breakup lengths can be achieved.

Chapter 1

Introduction

The disintegration of a thread of fluid into droplets is ubiquitous in modern engineering applications with research in this area stretching back over two centuries. It may therefore come as a surprise that despite such close scrutiny of this classical phenomenon, a good understanding of the mechanisms, along with an ability to control, the breakup and subsequent droplet formation of a liquid jet remains elusive to the scientific community. Whilst the last few decades have seen a growth in novel techniques for the generation of droplets (including variations of existing methods), in many cases, these methods are poorly understood.

The challenge of gaining a greater understanding of the physical mechanisms responsible for the process of rupture in a liquid thread (and consequently droplet formation) is made more difficult for a number of reasons, some of which fundamentally question the foundations of our current understanding of fluid dynamics. Firstly, the process of rupture involves a topological transition of the flow domain, usually from a cylindrical column into a series of spherical droplets. Rupture, by definition, is inherently associated with some length scale, usually the radius of a column of fluid, becoming infinitesimally small whilst at the same time the velocity of a typical fluid element near the point of rupture diverges to infinity. Therefore a singularity is reached in finite time and the familiar Navier-Stokes equations become redundant and no more progress can be made. Secondly, typical length and time scales can change over a number of orders between some initial equilibrium state and rupture. Such obstacles can be classified as 'local' or even 'universal' in the sense that, irrespective of any particular experimental setup, these issues present themselves as the point and time of rupture is approached. The breakup of a liquid thread can also be affected by, what may be termed 'global' mechanisms. These include externally imposed vibrations, the use of special chemicals and so forth, and in general, are much more easily manipulated and can effect both the initial steady equilibrium state and the evolution of disturbances towards pinch-off.

In this thesis we seek to explore a number methods to control droplet formation (and particularly parasitic satellite droplet formation which in many cases is an unavoidable consequence of the generation of droplets) with particular emphasis placed on gaining a deeper mathematical appreciation of rupture in a variety of complex industrial settings. Some of the material within this thesis has already been published (see Uddin *et al.* (2006) and Hawkins *et al.* (2007)) whilst others are under consideration for publication (see Uddin *et al.* (2007*a*) and (2007*b*)) and yet others are under preparation to be submitted (Uddin *et al.* (2007*c*), (2007*d*) and (2007*e*)).

This thesis is arranged in the following manner; the first chapter of this thesis provides a gentle introduction into the world of liquid jets with particular emphasis placed on developments over the last century. An appreciation of linear instability (of disturbances along liquid jets) which forms the backbone of most of the classical literature is followed by more recent non-linear analysis. A review of the most prominent experimental works in this field is then examined highlighting some of the successes of both linear and non-linear theories. We then examine some of the more recent works dealing with similarity solutions to breakup phenomena which become increasingly important as the time and length scales of the flow become very small.

The next chapter introduces the concept of a non-Newtonian fluid. We examine a number of models for the simplest types of non-Newtonian fluids and in particular we consider viscoelastic fluids (which are popular in the literature) as well as some inelastic fluids. We make mention of the Power-Law model which we use as the working fluid for the rest of this thesis.

Chapter Four provides a mathematical introduction to the industrial prilling process. In this process free surface disturbances (which can be artificially imposed to create desired frequencies) are allowed to travel along a rotating liquid jet to stimulate breakup[1]. Droplets formed upon rupture of the jet are later cooled and solidified in order to produce pellets. This technique is amongst the most favoured method for the preparation of fertilizers. Previous attempts to model this process based on Newtonian liquids are reviewed[2]. A mathematical model is developed to incorporate non-Newtonian flow into the spiralling jet equations. A linear temporal analysis is considered and is shown to produce similar results to spatial instability. The dispersion equation for the growth rate and most unstable wavenumber is derived and solved for certain parameter ranges. A brief summary of the main results is also presented.

In Chapter Five we introduce the reader to numerical methods involving finite difference schemes and we discuss some of the inherent restrictions which apply when using these methods. We solve the spiralling jet equations for fluids with power law rheology (derived in the previous Chapter) using a finite difference scheme based on the two-step Lax-Wendroff scheme. We investigate flows with parameter regimes which apply to in-

[1]Collaborators from the School of Mathematics and the School of Chemical Engineering at the University of Birmingham, with support from EPSRC and *Norsk Hydro*, were the first to present a model for this process. In addition further financial support was provided by Nestle and Prismo (who were mainly interested in developing models for liquid jets made of glue). To date only Newtonian fluids have been considered and since many industrial fluids (including those used for the production of fertilizers) are usually non-Newtonian, a better appreciation of rotating non-Newtonian fluids is needed.

[2]There are a number of commercial and environmental reasons why greater insight into the prilling process is necessary, these include the need to produce droplets with a specific chemical composition (due to regulations regarding fertilizer pellet composition) and a desire to eliminate or reduce wastage through satellite production.

dustrial prilling and our simulations allow us to calculate breakup lengths, droplet sizes and time to breakup. We further explore and discuss the relationship between certain fluid parameters and jet breakup characteristics.

In Chapter Six we re-examine the rotating liquid jet problem with the addition of surfactants to the free surface. Since the presence of surfactants reduce the surface tension of the liquid jet we find that depending upon the initial surfactant concentration and the importance of surfactant activity the dynamics of breakup and droplet formation are qualitatively different. In particular the effects on satellite formation is noticeable.

We attempt to generalize and incorporate the ideas of the previous chapters in Chapter Seven by considering a surfactant laden jet emerging from a rotating orifice and falling under gravity. The effects of the acceleration due to gravity on the trajectory and linear instability of the jet are investigated further.

The next chapter looks at how insonification can be used at the nozzle to manipulate droplet sizes and breakup lengths. We consider insonification amplitudes which are an order of magnitude larger than the standard perturbation at the nozzle. We also attempt to numerically simulate breakup of non-Newtonian jets to multiple perturbations at the nozzle (thereby attempting to more closely simulate real life breakup dynamics where an infinite number of wave modes are excited at the nozzle).

In Chapter Nine and Ten we eliminate rotation from our investigations and consider the breakup of straight compound jets. Such two-fluid systems have numerous applications in their own right, especially in connection to encapsulated droplets in the fields of pharmaceutics and biotechnology, however, they can also be used to manipulate the dynamics of the inner liquid thread. We consider both an inviscid-invsiscid and power law-power law system. In particular we examine the formation of compound droplets with multiple cores and pay close attention to what factors lead to their formation.

The dynamics of liquid microjets (jets on the micron scale) are considered in more detail in Chapter Eleven with some references to the latest research and practical applications which exploit such jets. In particular we examine closely the last stages of the topological transition which occurs when a fluid thread ruptures. The creation of *fresh* surface area as the topological transition point is reached allows us to use the theory of interface formation (which allows for a dynamic surface tension and therefore surface tension driven flows) and consider what, if any, differences appear between jets on the macroscopic scale and liquid microjets. We discuss the effects on the most unstable wavenumbers predicted by both theories and make some comparisons to molecular dynamic simulations.

The process of thermally heating a liquid thread at the orifice is examined in detail in Chapter Twelve with special attention paid to the effect on satellite droplet formation. Thermally modulating the free surface of a liquid thread is a technique which is already widely used in ink-jet printing but has particular importance to micro-fabrication in electronics and engineering. We investigate an number of heating 'patterns' at the nozzle including sinusoidal and pulsed heating. The effects of changing the frequency of heating is also considered. We incorporate the changing of viscosity with temperature into our model through an Arrhenius type equation. The effects of changing the thermal properties of the liquid are also examined.

Chapter Thirteen contains a brief summary of all the results we have obtained in the preceding chapters. We attempt to describe the salient features observed for the different techniques considered so far.

Finally, in Chapter Fourteen we look ahead at the future directions of this thesis. We consider a number of different direct extensions of the work presented in previous chapters along with some more novel yet related applications. Particular attention is placed upon techniques for the production of emulsions (or compound drops). We also discuss the technique of creating droplets using a T-shaped junction as well as the creation of mono-disperse droplets through the process of flow focusing.

Chapter 2

A Brief History of Liquid Jets

2.1 Introduction.

Whether it be the formation of stars or the production of nuclear fuel, or even quite simply the dripping of a kitchen tap, the instability of a liquid jet is a universally recognized phenomenon which finds applications in a wide variety of industrial and scientific processes. Whilst there are many scenarios where the applications of the rupture of a liquid thread can be easily identified, like ink-jet printing for example, there are many more which are not so apparent. Take, for example, the targeted delivery of drugs to the site of a disease (Prausnitz (2001)) or the cooling of microchip components (Wang *et al.* (2004)) which on the surface might not appear to have much in common. However, if we delve a little deeper into the underlying physics we find that such processes are among only a few of the more recent applications which take advantage of the properties of a liquid jet. Although it is true that many traditional applications like ink-jet printing (where nearly 10 patents are registered around the world every day) and industrial prilling (where liquid jets are used to make small pellets) continue to remain popular in industry it is the growing success and maturing of microengineering which has captured the attention of scientists and engineers and has resulted in a number of exciting and innovative uses. Applications like

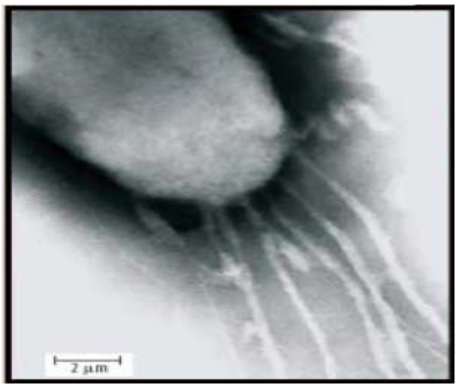

Figure 2.1: Myxobacteria can move at speeds of up to 10 μm a second by squirting out jets of slime from tiny nozzles placed on their bodies. Picture taken from *New Scientist*, April, 2006.

microchip fabrication and modern fuel injection devices are already in common use but more novel and emerging applications in this rapidly growing field include developments in biotechnology, where liquid jets are used to rupture cells, and pharmaceutics where microjets are used to deliver medicine intravenously (see Mortanto (2005) and Fletcher (2002)). Surprisingly liquid jets are even important to the motion of tiny bacteria which can squirt out slime from tiny nozzles located on their bodies to move at speeds up to 10 μm a second (see Fig. 2.1). All in all, such widespread use of liquid jets, especially in the biochemical sciences where tailor-made fluids are common, provides new and subtle challenges not only in appreciating how different types of fluids behave in such applications but also necessitates a greater understanding of liquid jet dynamics.

The dynamics of liquid jets are a special case of *free surface* flows which elicit huge interest not only for their immense practical applications but also for the fundamental issues they raise about the structure and solutions of the Navier-Stokes equation itself. One such complication is that for a liquid jet to disintegrate it must undergo a topological change at rupture, usually changing from a long cylindrical thread into a series of droplets. This process involves infinitesimally small length scales and singularities of the equations of motion. Furthermore, such flows are made more complicated in comparison with other flow situations due to the presence of a unknown *a priori* boundary (i.e the free interface) for which the shape and position must be determined as part of the solution to the flow equations.

Free surfaces (unlike rigid surfaces) can respond to the flow and as such have the tendency to allow for the propagation of free surface waves (or disturbances) and therefore in essentially all such applications an understanding of the stability of a liquid jet to disturbances provides the greatest challenge. This is particularly true in situations where the ultimate aim is the disintegration of the jet and subsequent drop formation. Consequently in a vast majority of such cases the goal is one of two things; the desire for droplets of uniform size and/or the suppression of unwanted satellites. If we take ink-jet printing as an example then in this case droplets produced from tiny nozzles are deposited onto paper to produce the final image. However the process of breakup near the nozzle may create satellite droplets which can reduce quality. Thus in this case the complete suppression of satellites is highly desirable. Alternatively with the advent of increasingly complex printing methods and the need for high quality output the production of uniform or monodisperse droplets can greatly improve image quality.

From everyday observations it is clear that a liquid jet is unable to retain its coherent structure for long periods. Instead the disintegration of a thread of liquid into droplets is a very familiar occurrence with most people observing such a phenomenon every time a kitchen tap is turned on. Nevertheless this apparent universal behaviour of liquid jets was not recognized until 1849[1] by Plateau who also correctly identified the mechanism driving instability; namely surface tension. Surface tension plays an important role in many free surface flows and particularly so in liquid jets. To gain a better understanding of the tendency for a liquid jet to spontaneously decay into droplets, as well as providing a gentle introduction to the linear stability analysis in the next section, we shall begin by considering the simple interplay between surface tension and the radius of a column of

[1]Although well over a century earlier in 1686 the French scientist E. Mariotte wrote a book, wherein he mentioned the breakup of a liquid thread emerging from the bottom of a container.

fluid.

Firstly, surface tension (also known as interfacial tension) is inherently a physical property of an *interface* separating two bulk phases. Here the interface needs further clarification since despite its intuitive nature it is a term which is rather difficult to describe using continuum mechanics. It is normal in continuum mechanics for length scales of the order of intermolecular forces to be ignored in line with the *continuum hypothesis*. However, since the thickness of the interface is determined by the range of intermolecular forces, it must consequently be modelled as a mathematical surface (i.e, a surface of zero thickness). Now the physical properties of the interface must be two dimensional analogues of the physical properties of the bulk fluids and in turn this means that quantities like energy per unit volume become energy per unit area along the interface. Surface tension, which acts along the interface, originates due to asymmetry between intermolecular forces on either side of the *interfacial layer* (see Rowlinson & Widom (1982) and Israelachvili (1995) for discussions of the molecular origins of surface tension). For water, surface tension σ is usually expressed in the units of milli-Newton per meter (mN/m) and takes the value of $\sim$ 70mN/m for a water-air interface. Most oils have a surface tension lower than that of water, typically $\sigma \sim$ 20mN/m, however liquid metals like mercury can have surface tensions up to seven times higher than water.

The relationship between the surface tension of a column of liquid and its associated radius is summarized by the Young-Laplace equation which relates the pressure jump across an interface to its curvature. If we consider a cylindrical co-ordinate system (which is invariably the most popular choice when studying liquid jets), there will necessarily be two principle radii of curvature, one along the cross section of the jet and another along its axis. If we assume an initially homogenous column of fluid then the curvature is dominated by variations across the cross section of the jet.

We now imagine an infinitely long horizontal cylindrical thread of fluid having constant radius in a state of rest. At any given time the surface of this liquid column is prone to tiny fluctuations in its surface properties like density, temperature or pressure. These fluctuations may cause the radius of the column to decrease slightly resulting in a small increase in pressure in the locality of the disturbance. Fluid located in the high pressure region of the liquid column has a tendency to migrate to low pressure regions thus further decreasing the radius of the column and ultimately increasing the pressure differential along the thread. This process culminates in breakup (also known as pinch off) where the radius of the thread goes to zero. Since the coefficient for the curvature term in the Young-Laplace equation is the surface tension, it follows that the magnitude of the surface tension determines the growth of interfacial instability.

Another way of viewing this phenomenon is by considering the linear relationship between surface tension and surface energy. In this case surface tension can be viewed as a measurement of the direct loss of the energy per unit area caused by molecules losing energy as they move from the bulk phase to the surface (see De Gennes *et al.* (2003)). Thus surface tension requires the jet to *favour* any distortion which leads to an overall *reduction* in surface area of the liquid column. It is for this reason that any non-axisymmetric disturbances are damped out since they ultimately lead to an increase in the overall surface area (see Rayleigh (1879)).

2.2 Breakup and Linear Instability.

The local analysis of a liquid column considered above can be translated to the global behaviour of a liquid jet (if we ignore any effects due to the presence of a nozzle or orifice[2]). Thus for a liquid jet the primary source of instability is surface tension and although in

[2] A stress singularity exists at the nozzle-jet interface due to the relaxation of the velocity from poiseuille flow to laminar or plug flow. Depending upon the material parameters the resulting relaxation of the free surface may lead to a contraction or swelling. At any rate the effects of the nozzle are only important a few jet diameters away from the nozzle (see Middleman (1993)). A fuller description of the relaxation process of the free surface, generalised to account for non-Newtonian fluids, is presented in Appendix C.

the next section we shall go on to see that the breakup of a liquid jet is strongly non-linear, it turns out that despite this fact, for many situations involving the breakup of liquid jets, the breakup length (and consequently the time to breakup) along with the size of droplets produced can be estimated to a good degree of accuracy using a linear instability analysis about some equilibrium base state. Such an analysis can produce surprisingly accurate estimates for breakup lengths and drop sizes. Before moving on to take a closer look at linear instability we will, for completeness, begin by introducing the different regimes of breakup in the presence of a surrounding fluid (this will become more important when we consider two-fluid systems in Chapter 9 and 10).

The generation of droplets by dispersing one immiscible fluid (known as the dispersed phase) in another immiscible fluid (the continuous phase) through a nozzle, syringe or orifice occurs in many practical applications. Apart from changing the fluid parameters, the flow rate of the dispersed fluid can also be altered. It is found that, depending upon the values of this flow rate, there exist four distinct regimes of breakup. These are known as the *Rayleigh* regime, the *first wind-induced* regime, the *second wind-induced* regime and the *atomization* regime. The first two of these regimes are characterised by breakup occurring many jet diameters away from the orifice (i.e. breakup is determined by long wavelength disturbances) as well as the size of droplets being of the order of the jet diameter[3] (see Fig. 2.2). The *second wind-induced* regime and the *atomization* regime generally tend to breakup very close to the orifice and produce droplets much smaller than the diameter of the orifice. It is not uncommon in these regimes for the jet surface to become disrupted and for drops to emerge laterally.

In addition to taking account of the effects of a quiescent continuous phase, it can be shown that the dynamics of a liquid jet can also be greatly affected if the flow rate of the continuous phase is allowed to vary (so that a liquid jet emerges into a laminar flow

[3]The Rayleigh regime usually occurs at lower speeds than the first wind induced regime. Moreover, the Rayleigh regime produces droplets slightly larger than the diameter of the jet.

Figure 2.2: A schematic depicting the breakup of a liquid jet under the Rayleigh regime with subsequent main and satellite drop formation. Notice how breakup occurs many jet diameters away from the orifice (far left of picture) and droplet sizes are comparable to the jet radius.

field, say). In this case, droplets are formed at the orifice if the flow rate of the dispersed phase is low (droplets are peeled off as they leave the orifice). However, the breakup length is found to correlate with the viscosity and flow rate ratios between dispersed and continuous phases for higher flow rates (see Cramer *et al.* (2002)) in such cases. Modifying the flow of the continuous phase in conjunction with specially patterned surfaces is an interesting area of research which is still very much in its infancy and will provide fertile ground for further research (we will return to this problem in much more detail in Chapter Fourteen).

For most practical applications (especially those considered in this thesis) the flow rate out of the orifice is never high enough to consider any regime other than the *Rayleigh* regime. However, the fluid parameters of both the dispersed and continuous phase do differ in many applications and usually are of three types; gaseous, low viscosity liquids or high viscosity liquids. A summary of works for different continuous and dispersed liquids is given in Fig. 2.3. The most important case that we will be concerned with is the case of a low viscosity liquid emerging into a gaseous environment so that the effects of the surrounding gas can be ignored. With this in mind, we begin with Rayleigh's classical analysis.

2.2.1 Classical Inviscid Jets.

It was not until the late nineteenth century that Rayleigh[4](1879) considered the mathematical treatment of disturbances along an infinite *inviscid* column of liquid. Rayleigh was able to show that instability was caused by surface tension and that an optimal wavelength ($\lambda_{opt} \approx 4.5$ jet diameters) at which perturbations grew the fastest existed. Under the assumption that such *optimal* (also known as Rayleigh mode) disturbances effectively determined droplet sizes, Rayleigh was able to confirm and show favorable comparisons to the experimental observations of Savart (1833). Although Rayleigh's analysis has traditionally been the foundation of almost all liquid jet studies we will not attempt to reproduce his work here but instead, in this section, we shall mention some relevant results only. The interested reader is encouraged to refer to Rayleigh (1879 and 1945) as well as a number of more modern references such as Yarin (1993), Middleman (1995) and Eggers (1997). For an approach which is different but nevertheless produces the same results the reader is referred to Anno (1977).

In Rayleigh's analysis a cylindrical jet, in a state of equilibrium, having radius $r = a$ (in cylindrical coordinates) is allowed to be perturbed by an initial disturbance which leads to the radius having the form $r = r_s$ where

$$r_s = a + \delta \cos(kz) \cos(n\theta), \tag{2.1}$$

and δ is a small initial disturbance, k is the wavenumber, ρ is the density of the liquid and n is an integer. Since a cylindrical coordinate system is being considered r, z and θ have their conventional interpretation. Using the standard equations of motion and

[4]John William Strutt (Lord Rayleigh) (1842-1919) was a prolific contributor in the field of applied mathematics. He was awarded the Nobel Prize in physics for his discovery of the inert gas Argon after which he was made President of the Royal Society for three years between 1905 and 1908. His book the *Theory of Sound* established the field of acoustics and was written on a houseboat on the River Nile.

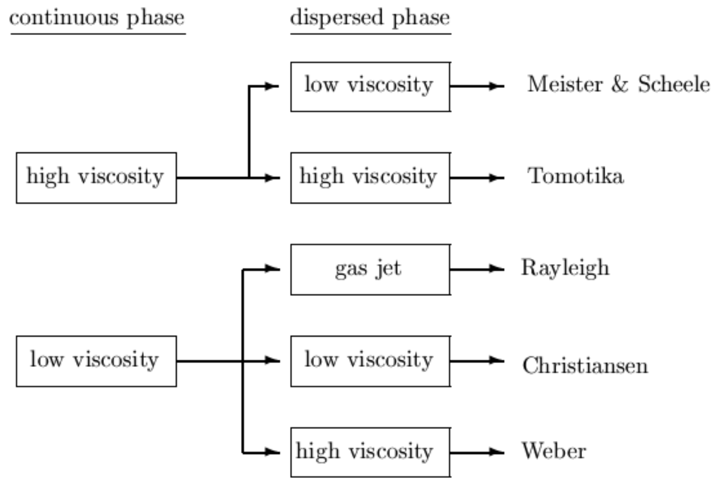

Figure 2.3: Linear stability analysis for liquids with different continuous and dispersed phases. These works constitute the first of their kind and form the foundation of later research.

assuming that disturbances can be written in the form $\exp(\omega t - i(kz - n\theta))$, where t is time, Rayleigh was able to arrive at his famous result. That

$$\omega^2 = \frac{\sigma(ka)}{\rho a^3}(1 - n^2 - (ka)^2)\frac{{I_n}'(ka)}{I_n(ka)}, \tag{2.2}$$

where σ is the surface tension and I_n is the nth modified order Bessel function. If disturbances are axisymmetric then $n = 0$ and we can use the recurrence formulae for Bessel functions,

$$I_{n-1}(x) + I_{n+1}(x) = \frac{2n}{x} I_n(x), \quad I_n^{'}(x) = \frac{1}{2}(I_{n-1}(x) + I_{n+1}(x))$$

to arrive at the dispersion relation for axisymmetric disturbances

$$\omega^2 = \frac{\sigma(ka)}{\rho a^3}(1 - (ka)^2)\frac{I_1(ka)}{I_0(ka)}. \tag{2.3}$$

If ω is plotted against ka for $ka < 1$ we find that the disturbance which grows most rapidly (i.e. has the largest value of ω) occurs for $ka = 0.696$ with a corresponding growth rate $\omega = 0.34(\sigma/\rho a^3)^{\frac{1}{2}}$. For $ka > 1$, ω is imaginary and the disturbances do not grow with time t. In addition if $n \neq 0$, equation (2) gives that ω^2 is always negative and consequently non-axisymmetric disturbances do not grow with time. Thus for a inviscid (or nearly inviscid) liquid jet, issuing from a nozzle, we would expect that from all the tiny disturbances generated along the interface, the one having a non-dimensional wavenumber of $ka = 0.696$ (with a wavelength of about $9.02a$) to dominate and eventually lead to breakup. The characteristic time taken to breakup t_b can be estimated by inverting the growth rate so that $t_b = 2.94(\rho a^3/\sigma)^{\frac{1}{2}}$. Thus, a jet of water (which is generally considered inviscid) emerging from an orifice of radius 5mm will have a characteristic breakup time of about 0.12s.

2.2.2 Classical Viscous Jets.

The above analysis can be repeated with viscosity (Weber (1936)) to arrive at a similar characteristic equation, namely

$$\omega^2 + \frac{3\mu k^2}{\rho}\omega = \frac{\sigma k}{\rho a^2}(1-(ka)^2)\frac{{I_1}'(ka)}{I_0(ka)}. \tag{2.4}$$

In this case the coefficient in front of ω is positive and will thus contribute in stabilizing or dampening disturbances. The disturbance which dominates and eventually leads to breakup is given by[5]

$$ka = 2^{-\frac{1}{2}}\left(1 + \frac{3\mu}{\sqrt{\rho\sigma a}}\right)^{-\frac{1}{2}},$$

which is better represented by introducing the non-dimensional *Ohnesorge* number $Oh = \mu/\sqrt{\rho\sigma a}$ (which is a measure of the relative importance of viscous forces to surface tension forces) so that we have

$$ka = (2(1+3Oh))^{-\frac{1}{2}}.$$

[5]This is really the long wavelength approximation of (2.4). It does however provide a very good approximation.

The observant reader will note that in the limit of $\mu \to 0$ the above equation gives $ka = 1/\sqrt{2} = 0.707$ which slightly overestimates the inviscid limit of $ka = 0.697$ although the growth rate corresponds perfectly. In general, we see that viscosity increases the wavelength of most unstable disturbances (leading to the production of larger droplets) and allows for more viscous jets to have smaller growth rates (and therefore longer breakup times). Both (2.3) and (2.4) provide good qualitative predictions for breakup lengths[6] and droplet sizes.

Tomotika (1935) extended Rayleigh's original analysis to account for the presence of an outside continuous phase and in particular he examined the case of a very viscous fluid encased within another very viscous fluid (i.e., Stokes flow in both fluids). His analysis highlighted the importance of viscosity and density ratios between the two fluids as well as the importance of the ratio of viscous forces to surface tension forces (characterised by the Ohnesorge number) on instability.

So far we have considered what is termed *temporal instability*, that is when considering disturbances, we have assumed they have the form $\exp(\omega t - i(kz - n\theta))$ where the wavenumber k is taken to be real. In this case, the growth rate ω is, in general, complex so that $\omega = \alpha + i\beta$ where α and $-\beta/k$ are known as the *temporal growth rate* and *wave speed* respectively. Normally we find that disturbances grow ($\alpha > 0$) while being convected along the jet ($\beta = kU$, where U is some typical jet velocity).

Keller *et al.* (1973) realised that for liquid jets emerging from a nozzle or orifice disturbances are not just simply created at $t = 0$ and then convected along the jet but instead can occur for later times as well. It therefore follows that disturbances grow spatially as well as temporally and the resulting analysis must now include a complex wavenumber $k = k_r + ik_i$ where k_i represents the spatial growth rate.

[6]An expression for the breakup length of a liquid jet using dimensionless analysis is presented in Appendix B.

2.3 The Non-Linear Dynamics of Breakup.

According to linear stability analysis, a liquid jet should break uniformly along its axis with the size of droplets produced being roughly equal to the wavelength of the initial disturbance. This is plainly not true (see Chaudhary & Redekopp (1980*a*)) and instead breakup occurs non uniformly along the jet and a number of 'satellite' droplets are observed which are much smaller then their adjacent parent drops. An attempt to use higher order perturbation theory (Chaudhary & Maxworthy (1980*b* and 1980*c*)) does not replicate unequal droplet sizes. One reason why linear theory fails to predict satellite droplets is the discarding of the non-linear convective term (see Lin (2003)) in any linear analysis. It is believed that this term can induce higher order harmonics despite the imposition of a pure harmonic disturbance at the orifice. More importantly, as breakup is approached the minimum radius goes to zero and the pressure diverges to infinity (i.e. a singularity is reached in finite time) and as such it is no surprise that linear theory breaks down failing even to give a qualitative description of pinch off.

In order to capture non-linear behaviour near pinch off, a full analysis of the Navier Stokes equation with free boundary conditions is needed. Any analytical approach is virtually impossible and even a full numerical analysis of jet breakup is extremely difficult due to high resolutions needed in neck regions near the singularity. For this reason one dimensional models (which are reduced forms of the momentum equations which depend upon just *one* independent spatial variable, normally the variable along the axis of the jet) are popular although some 2D (Abmraveneswaren *et al.* (2002)) and 3D (Moseler & Landman (2000)) simulations do exist of liquid jet breakup.

In order to arrive at one dimensional models we start by considering the Navier-Stokes equation in cylindrical coordinates with the assumption of axisymmetric flow. This leads to the momentum equation along the radial and axial directions complemented with the

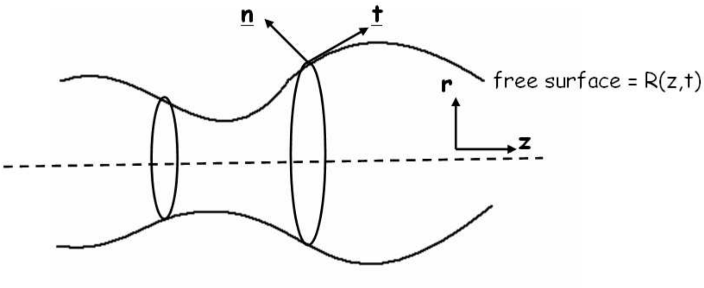

Figure 2.4: A diagram showing the geometry of a free surface in cylindrical co-ordinates. The dashed line represents the axis of symmetry $r = 0$.

continuity equation;

$$\frac{\partial u_r}{\partial t} + u_r \frac{\partial u_r}{\partial r} + u_z \frac{\partial u_r}{\partial z} = -\frac{1}{\rho}\frac{\partial p}{\partial r} + \frac{\mu}{\rho}\left(\frac{\partial^2 u_r}{\partial r^2} + \frac{1}{r}\frac{\partial u_r}{\partial r} - \frac{u_r}{r^2} + \frac{\partial^2 u_r}{\partial z^2}\right), \tag{2.5}$$

$$\frac{\partial u_z}{\partial t} + u_r \frac{\partial u_z}{\partial r} + u_z \frac{\partial u_z}{\partial z} = -\frac{1}{\rho}\frac{\partial p}{\partial z} + \frac{\mu}{\rho}\left(\frac{\partial^2 u_z}{\partial r^2} + \frac{1}{r}\frac{\partial u_z}{\partial r} + \frac{\partial^2 u_z}{\partial z^2}\right), \tag{2.6}$$

$$\frac{\partial u_r}{\partial r} + \frac{\partial u_z}{\partial z} + \frac{u_r}{r} = 0, \tag{2.7}$$

where u_r and u_z are the radial and axial velocities respectively.

The boundary conditions (evaluated at the free surface) express the relationship between the pressure difference (which can be related to the curvature) across the free surface with the normal stress

$$\mathbf{n} \cdot \mathbf{\Pi} \cdot \mathbf{n} = \sigma \kappa, \tag{2.8}$$

where $\mathbf{\Pi}$ is the total stress tensor, κ is the mean curvature of the free surface, σ is the isotropic surface tension and $\mathbf{n}$ is the unit normal vector to the free surface as shown in Fig. 2.4. If the external fluid is a gas then the tangential stresses along the surface of the jet can be equated to zero

$$\mathbf{n} \cdot \mathbf{\Pi} \cdot \mathbf{t} = \mathbf{0}, \tag{2.9}$$

where $\mathbf{t}$ is the tangent vector to the free surface.

Finally, the kinematic condition at the free surface requires that a particle at the free surface to remain there so that

$$\frac{D(r - R(z,t))}{Dt} = 0, \quad \text{i.e.} \ \frac{\partial R}{\partial t} + u_z \frac{\partial R}{\partial z} = u_r \tag{2.10}$$

The above system of equations can be reduced if suitable asymptotic expansions for both the radial and axial velocity components are chosen. If we follow the example of Eggers (1993) we could expand the axial velocity in powers of r so that

$$u_z(z, r, t) = u_{z0}(z, t) + u_{z2}(z, t)r^2 + ...$$

If a similar expansion is made for the pressure and then substituted into the momentum equations and the continuity equation (this will give an expression for u_r in terms of the components of u_z see Eggers (1993)) together with the boundary conditions we arrive at a closed system of equations for u_z (which we write as u from now on) and R, namely

$$\frac{\partial R^2}{\partial t} + \frac{\partial (R^2 u)}{\partial z} = 0 \tag{2.11}$$

$$\rho \left(\frac{\partial u}{\partial t} + u \frac{\partial u}{\partial z} \right) = -\sigma \frac{\partial}{\partial z} \left(\frac{1}{R_1} + \frac{1}{R_2} \right) + 3\mu \frac{\partial^2 u}{\partial z^2} \tag{2.12}$$

where R_1, R_2 are the principal radii of curvature. If we non-dimensionalize our variables so that

$$\bar{t} = \frac{t}{(\rho a^3/\sigma)^{\frac{1}{2}}}, \quad \overline{z} = \frac{z}{a}, \quad \overline{u} = \frac{u}{(\sigma/\rho a)^{\frac{1}{2}}}, \quad \overline{R} = \frac{R}{a}.$$

where a is the initial radius of the jet and ρ is the density of the liquid, then dropping overbars, the system in non-dimensional form is

$$\frac{\partial R^2}{\partial t} + \frac{\partial (R^2 u)}{\partial z} = 0 \tag{2.13}$$

$$\rho \left(\frac{\partial u}{\partial t} + u \frac{\partial u}{\partial z} \right) = -\frac{\partial}{\partial z} \left(\frac{1}{R_1} + \frac{1}{R_2} \right) + 3Oh \frac{\partial^2 u}{\partial z^2} \tag{2.14}$$

where Oh is the Ohnesorge number (which is a ratio of viscous forces to surface tension forces) given by $Oh = \mu/(\sigma a \rho)^{\frac{1}{2}}$.

The first numerical simulation of (2.13) and (2.14) was that of Lee (1974) who investigated the non uniform breakup of an *inviscid* liquid jet. He was able to calculate the profile of the jet at breakup and thus estimate main and satellite drop sizes. After pinch off the fate of a satellite droplet can take one of three paths; it can either merge with the downstream or upstream main droplet (known as forward and rear merging respectively) or alternatively it can move along with the same velocity as the main droplets. A systematic analysis of different merging scenarios for satellite droplets is presented by Pimbley & Lee (1977). The analogous one dimensional viscous case has been investigated by Bousfield and Denn (1987) and Bousfield *et al.* (1990).

Eggers and Dupont (1994) investigated the breakup of a viscous liquid jet and the bifurcation of a drop suspended from an orifice using a one dimensional equation of motion. In particular they were interested in the behaviour of singularities close to the fluid neck in the final stages of pinch off. Simulations of drops of water suspended from an orifice were compared with the experimental photographs of Peregrine *et al.* (1990) with good agreement.

In order to illustrate and discuss some of the important fluid dynamics close to pinch off we will use a method similar to Yarin (1993) to solve (2.13) and (2.14) (details of the numerical method are left till Chapter 5). Figure 2.5 shows the profiles of a Newtonian liquid jet at different times towards the latter stages of pinch-off along with the corresponding pressure. It can be seen that as the surface tension gets to work and causes the jet to contract the curvature increases (since the radius of the jet becomes smaller), this in turn causes the pressure (consistent with the Laplace-Young equation) within the jet to increase. The profiles shown in the figure are taken over half a wavelength and show the radius rapidly decreasing at at point (the pinch-off point) separating a drop on one

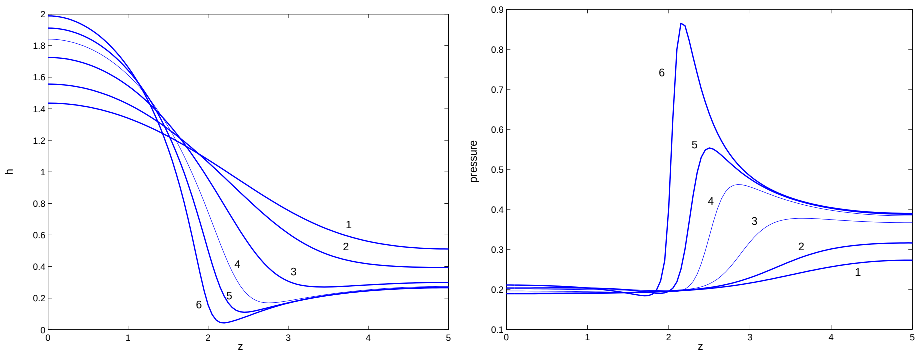

Figure 2.5: The solution of equations (2.13) and (2.14). The profiles of the jet radius close to the pinch off point is shown along with the corresponding pressure. The Ohnesorge number is chosen as 0.62. Profiles are shown for the non-dimensional times corresponding to 1, 2, 3, 4, 5, 6 are 30, 32, 34, 35, 35.5 and 36 respectively.

side with a smaller ligament to the other side. In the locality of the region of pinch-off the pressure can be seen to increase which forces the liquid away from this region into the drop. The rate at which fluid leaves this region is proportional to the magnitude of the pressure and so the drop continues to grow until pinch-off takes place. A closer look at Fig. 2.5 reveals that the ligament connecting the drop stops contracting (while the drop continues to grow), this is in agreement with the experimental results of Goedde and Yuen (1969).

2.4 Experiments.

Experimental studies to investigate the phenomenon of breakup and drop formation in liquid jets generally fall into three categories; those which involve a liquid jet emerging from a nozzle, a dripping faucet and that of a liquid bridge (where a portion of fluid is held between two moving solid plates). All these experiments have much in common and are essentially the same near the locality of the singularity, where the radius goes to zero and rupture occurs, however there exist certain qualitative differences between them.

The earliest known experimental investigation into liquid jets is that of Bidone (1823), who examined the fluid leaving the holes from the bottom of a container. Savart (1833) was the first to observe that the frequency of disturbances along liquid jets could be controlled by varying the frequency of perturbations applied at the orifice. Bidone's experiments and those of other nineteenth century researchers like Savart are remarkable for the accuracy with which observations were made when due consideration is given to the fact that most observations would inevitably have been confined to the human eye or by crude photographic techniques. In contrast, the success of modern photographic equipment to capture images microseconds apart and with resolutions on the micron scale has made experiments on liquid jets comparatively much easier. As such, there have been a number of modern studies.

Following on from Savart's observations regarding the control of disturbances along a liquid jet by modifying the frequency at the orifice, a number of authors have chosen to investigate the decay of a liquid jet using different methods to induce instability. Crane, Birch and McCormack (1964) studied jet instability by using an electrical vibrator to induce disturbances of different wavelengths. Donelly & Glaberson (1966) used a loudspeaker to do the same. They were also the first to measure the growth of surface waves as a function of time. The presence of non-sinusoidal disturbances in their experiments was attributed to higher order harmonics (as opposed to non-linear effects) induced by vibrations at the nozzle.

Goedde & Yuen (1969) introduced perturbations at the orifice using a number of different methods. Apart from the loudspeaker setup, Donelly & Glaberson also utilized an electronically driven vibrator. Experiments were carried out using a short vertical nozzle (allowing the velocity profile to remain uniform on exit) and jet speeds were chosen small enough so that surrounding effects could be ignored and a laminar flow field could be assumed yet high enough so that gravity in experiments could be neglected. Measurements of the diameter of both swell regions (the region of onset of droplets) and neck regions were taken as functions of time. As breakup was approached the swell grew and neck diminished as expected but they found that the exponential growth rate for the swell and neck regions were not constant but the difference between them remained constant. Amongst their observations were that pinch-off did not occur at the center of neck regions but instead away towards the swell region. Later stages of breakup were observed to be dominated by non-linear effects and the neck portion of the jet actually stopped contracting while the swell portion became narrower and bigger.

Rutland & Jameson (1970) investigated the decay of a water jet emerging vertically downwards from a tube of length 30cm and diameter 4mm. The typical speed of the jet when leaving the nozzle varied between 2.5-3.5 m/s. To induce perturbations at the nozzle they used a 10W speaker (a similar method had been used by Donelly & Glaberson previously). The authors' main aim was to use non linear theory (they used the non-linear analysis of Yuen (1968))

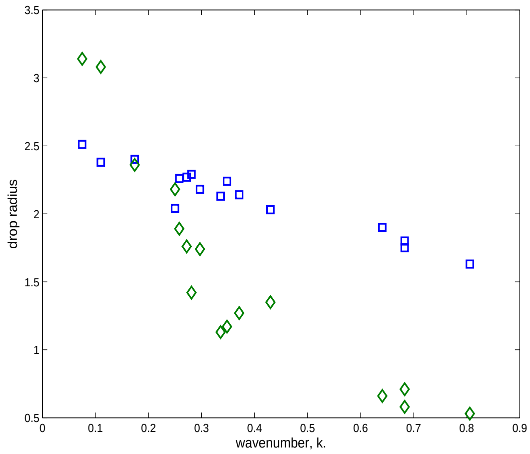

Figure 2.6: Data from Rutland & Jameson (1970) for main (□) and satellite (◊) water droplets. The radius is non-dimensioanlised with respect to the orifice radius

to calculate the profile of waves on the jet at breakup and to use this to predict the volume (and size) of main and satellite droplets. In essence, the theory of Yuen (1968) allows for a non-sinusoidal wave profile at breakup while also ensuring that the volume of the jet remains constant for higher order terms than the initial perturbation amplitude. Using this theory it is possible to calculate wave profiles at the time when the 'trough' meets the centreline of the jet and thus calculate sizes of main and satellite droplets. Rutland & Jameson (1970) found that in general there was good agreement between their experiments and the theory of Yuen (1968) however their experiments revealed satellite droplets through a range of wavenumbers contrary to the non-linear theory which does not predict satellite formation above wavenumbers of 0.7. Figure 2.6 shows some results from Rutland and Jameson (1970) for main and satellite droplets.

Exploiting high speed photographic techniques, Kowalewski (1996), using an experimental setup resembling that of Becker *et al.* (1991), examined the profile of a thin liquid neck joining droplets. Typical jet diameters were up to eighty times smaller (50-900μm) than those considered by Rutland & Jameson (1970) and typical breakup lengths were between 100-200 jet radii. Micro satellite droplets were observed along the necks. The minimum diameter of the jet before breakup was found to be ~ 1 μm even when the viscosity was varied over several orders. The reasons behind this seemingly constant neck radius before breakup are still unknown, although the molecular simulations of Koplik & Banavar (1993) rule out the influence of molecular effects. Indeed, according to Koplik & Banavar (1993) the Navier-Stokes equations remain valid for length scales down to 100 $\mathring{A}$ (10nm) and time scale 10^{-10}s. The theory of interface formation (see Shikhmurzaev (2005)) attempts to provide one explanation for Kowalewski's experiments by introducing the idea of a *relaxation* time for the free surface. Under this theory, pinch-off is the combined result of capillary pressure (with varying surface tension) and flow-induced Marangoni effects. We will return to this issue, and consider this theory in much more detail, in a later chapter.

Experiments involving liquid bridges or hanging pendant droplets (dripping faucet) allow for a much more controlled investigation of the stability of a thread of liquid. Indeed, such studies have created a large body of classical literature in the field of fluid mechanics starting from the early pioneering work of Tate (1864), Rayleigh (1899), Harkins and Brown (1919) and much more recently Shi *et al.* (1994). In particular it has been found that gravity can limit the length of a stable liquid bridge and, as such, there is growing interest in investigating the effects of the growth of single-crystal semiconductors in microgravity. As we have mentioned previously, the breakup of a liquid bridge or a hanging pendant droplet is qualitatively similar to the breakup of a liquid jet near the location of pinch-off; as such, we will not delve too deeply into this topic although the interested reader is referred to Middleman (1995) for a more detailed investigation of this topic. We will however mention here some of the most relevant liquid bridge studies which involve shear thinning liquids namely those of Yildirim & Basaran (2000) and Doshi *et al.* (2003).

2.5 Similarity Solutions.

Experiments like those of Haenlein (1931) and Kowalewski (1996) have shown that near the locality of pinch-off, the geometry of a liquid jet exhibits remarkable similarity. In general, we have a thin ligament connected to a (much larger) drop. This configuration at pinch-off remains similar even if certain initial conditions (e.g. the wavelength of disturbances) are varied. However experiments do show a marked difference when the viscosity of the fluid is altered.

These results suggest that while the flow in a liquid jet may be influenced by initial conditions away from breakup, near pinch-off the flow becomes increasingly dependent on internal fluid parameters (this was first recognised by Peregrine *et al.* (1990)). In general there are three fluid parameters which can affect the flow; the kinematic viscosity ν ($\mathrm{cm^2s^{-1}}$), the surface tension γ ($\mathrm{gm\ s^{-2}}$) and the density ρ ($\mathrm{gm\ cm^{-3}}$). A simple dimensional analysis using these three parameters leads to the so called *natural* length and time scales given by

$$l_\nu = \frac{\nu^2\rho}{\gamma} \quad \text{and} \quad t_\nu = \frac{\nu^3\rho^2}{\gamma^2}.$$

For water at room temperature (20°C) having viscosity 1.004×10^{-2} $\mathrm{cm^2s^{-1}}$ and with density 0.998 $\mathrm{gm\ cm^{-3}}$ and surface tension equal to 72.8 $\mathrm{gm\ s^{-2}}$, we have a *natural* length scale of $1.38\times10^{-6}m$ and time scale $1.91\times10^{-10}s$. The equivalent length scale for glycerol is 10^6 times greater and the time scale can be up to 10^9 times greater.

Thus, irrespective of whether the experiment involves a liquid jet emerging from an orifice or a dripping faucet, the motion near breakup will become independent of initial and boundary conditions. If the fluid is inviscid and irrotational then Keller and Miksis (1983) have shown that the height of an interface decays like 2/3 of the time remaining to breakup. This has been numerically verified by use of numerical simulations based on the boundary element method (BEM) of Day *et al.* (1998). Similarly, if the fluid under consideration is viscous then Eggers (1993) has shown that the minimum radius h_{min} and maximum velocity v_{max} of a liquid thread depends only upon the time Δt remaining from breakup, that is

$$h_{min} = 0.0304\frac{\gamma}{\mu}\Delta t \quad \text{and} \quad v_{max} = 3.07\left(\frac{\mu}{\rho}\right)^{\frac{1}{2}}(\Delta t)^{-\frac{1}{2}}.$$

Under this theory we would expect a thread of glycerol ($\mu = 0.672$kg m^{-1} s^{-1}, $\gamma = 0.0645$ m s^{-2} and $\rho = 1248$ kg m^{-3}) at 0.01s before breakup to have a minimum radius h_{min} of 13.1μm and a maximum velocity v_{max} of 1.06m/s (see Eggers (1993)). The experiments of Kowalewski (1996) show good qualitative agreement with Eggers' findings for pre-breakup thinning of a liquid thread on scales down to the order of microns. However, after rupture, Eggers (1993) and Eggers and Dupont (1994) predict that the pressure and the axial velocity diverge to infinity (so that a singularity is reached in finite time) which does not correspond to the experiments of Kowalewski (1996). These unphysical predictions for the pressure and velocity have been addressed by Shikhmurzaev (2005) with the introduction of the concept of interface formation into the process of thinning. Irrespective of whether the working fluid is viscous or inviscid, Lister and Stone (1998) have shown that the effects due to the dynamics of the surrounding medium (typically air) cannot be neglected once h_{min} reaches some finite minimum value.