A Theory for Wave Propagation and Growth on Particle Laden sheets produced during the Atomization of Suspensions.

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Abstract

The analysis of the physical processes which take place during the early stages of the atomization of suspensions is presented and simplifying assumptions are made in order to create a simplified model which takes account of curvature induced thinning caused the presence of particles in the region where this sheet thickness is less than the size of particles. It is postulated that in the region where sheet thickness is greater than the size of particles (Zone I), the fluid behaves as a single phase and bulk properties can be used to predict wave growth from Squire's theory. Within zone II, where sheet thickness is less than the particles size, the fluid comprises two phases, particles and the suspending medium. The sheet stability and its impact on the propagation of waves is limited by the evolution of the thinnest regions in the inter-particle space. Simple calculations based on the above and Squire's theory demonstrate abnormally high growth rate of waves when large particles are present in sufficiently high concentrations. It is predicted that when the size of suspended particles are small then aerodynamic wave growth can be predicted by the conventional atomization theory and that the troughs of such waves can reduce the local sheet thickness below the size of particles, leading to predictable particle effects such as the onset of perforations. Although particle effects are present, the break-up of sheets contain small particles would be similar but not identical to that of sheets free of particle. Evidence is presented, which affirms the theory.
In many instances, atomization is achieved by using a nozzle to form a thin freely moving sheet of liquid, which subsequently breaks up to form droplets. When sheets are composed of single phase liquids, break-up occurs via action of aerodynamic waves. This mechanism is well understood and there have been numerous reviews on it (Lefevbre 1987, Dombrowski & Fraser 1953).

In contrast much less is known of the mechanism by which sheets composed of two phases break up in spite of the obvious importance of such information for the spray drying industry where slurries and pastes are commonly sprayed to produce particles of desired physical properties.

Several seemingly unexpected observations have been recorded in the literature concerning the atomization of suspensions. Several workers have reported early appearance of waves on sheets of suspensions of solids in liquids (F. Addo-Yobo et. al, Mulhern et. al.). Perforations are known to occur on such sheets, which are produced during the atomization of suspensions (Aidun and Spielbauer, F. Addo-Yobo et. al 2010). Other important observations include the observation that when suspensions are sprayed, some drops are formed, which contain no particles [4]. The explanations of this latter phenomena presented by the two different workers [3,4] who approach the problem from separate perspectives coincide. Since particles cannot be broken during atomization of suspensions, drops produced by the process whose sizes are less than that of the suspended particles, would be free of particles.

To date there remains a gap in the literature regarding the mechanism by which accelerated wave growth can occur on sheets of suspensions. When a coherent mechanism for the above is established, then it becomes possible to anticipate sheets break-up lengths and hence place limits on the size of droplets formed during the atomization of suspensions. This work presents such a mechanism which applies to atomization cases where freely moving sheets are produced. Blast atomizers in which a second fluid, usually air is used to break-up the sheets are excluded. This paper presents a mechanism by which relatively thick sheets can become destabilized due to curvature induced thinning caused by the presence of particles in the region where sheet thickness is less than the size of particles and presents a simplified theory on how this leads to accelerated wave growth on sheet of suspensions compared to the growth rate of waves on sheets of liquids free of particles.
We present a case study of the situation where the volume fraction of particles in the suspensions used to form the sheets are high such that the sheets perforate and use the theory to predict the expected wave growth.

We begin by analyzing the structure of the sheet and introducing assumptions which enable a simple theory to be used to predict wave growth. Case studies consisting of the break-up of sheets on suspensions of large and small particles.

**Structure of the Sheet**

Recent work by Addo-Yobo et al.[4] posits that freely moving sheets of suspensions comprise two regions with different characteristic properties: Zone I, where sheet thickness is greater than particle size, and zone II, where sheet thickness is less than particle size. Within Zone II the above show that when the perforation number of the suspensions, defined in Eqn. (1) below, which depends on the particles size and the liquid properties, exceeds a critical value then the sheet perforate.

\[
A = \frac{1}{\sqrt{d \rho \sigma}}
\]  

(1)

**Zone I**

The location and extent of zone I deserves further attention. When the particles are non-interacting and nearly spherical, such as when particles of glass, or wetted lycopodium seeds are used, then the appropriate dimension to use in determining the location of zone I is the particle diameter. When ultrafine particles such as: Titanium dioxide (ca 0.5micron) or Attapulgite and Bentonite are used then strong hydrogen bonds can lead to the formation of structure which break down incompletely under shear as the as the fluid passes through the nozzle orifice. Under such circumstances, it is the agglomerate size that determines the appropriate location of the zone I. Should any loose association of particles be formed in the nozzle orifice survive into the sheet, then should they cause protuberances at the surface of the sheet, surface tension force would flatten them at the surface. Thus only "hard-agglomerates would survive. In Zone I of the sheet we postulate that, the suspension flows as a single fluid with properties equivalent to those of the bulk suspension.

The sheet thins uniformly according to the hyperbolic law shown in Eqn. (2) below:

\[
h = \frac{K}{R}
\]  

(2)

Where sheets are formed by impacting to jets of liquid, or by impacting a single jet on a plate, then K, the thinning parameter may be approximated by the mass balance law.
Zone II

Within Zone II, where the average sheet thickness is less than the particle or "hard" agglomerate size, curvature variations are imposed on the sheet because the wetted particles move apart at right angles to the direction of the spray. And because the particle are wetted, they would tend to hold liquid around them. Thus the liquid held in the sheet is unevenly distributed in Zone II, leading to curvature variations within the inter-particle space. This leads to curvature driven localized thinning of the sheet, which adds to the thinning due to extensional flow of liquid within the sheet, which is imposed as the liquid departs the nozzle orifice.

Figure 1 depicts the cross section of expanding sheet showing two neighbouring particles P1 and P2, undergoing separation. The liquid between particles may be considered as "cells" of liquid in a bulbous droplet undergoing elongation. The physical description of the evolution of the shapes are similar to those of drop with bulbous ends. The constraints at the boundaries lead to the development of a convex region between A and B, followed by a concavity between B and C. According to the symmetrical boundary conditions, the curvature is zero at centre of the 'cell' (D).

The curvature variations effects between P1 and P2 are intensified by those between the two and the particles which lead and follow the two along the radial streamlines emanating from the nozzle orifice. It is noted that the positioning of the particles is would be the outcome of stochastic processes within the fluid in its passage through the orifice of the nozzle.

The Propagation and Growth of Waves

Disturbances of the two surfaces of the freely moving sheet cause pressure distribution in the ambient air which act on the sheet. The result is that the disturbances grow on the sheets. This paper argues that the conventional theory of wave propagation in freely moving sheets is applicable and that the structure of the sheet need to be taken into account in a proper way!

Waves growth within Zone I

Within zone I, the waves 'see' a medium whose properties are that of the bulk. As the wave growth time is not great particularly when large particles (in excess of 20 microns) are used
(zone I is confined close to the nozzle), wave amplitudes are small and would not lead to sheet thinning below the size of particles.

The motion of the ambient air and the aqueous suspension (low viscosity) are described by an appropriate form of the Navier-Stokes equation in which the viscous term can be dropped.

The boundary conditions are identical to those used in the classical analyses: The surface of the suspension fluid moves with the fluid and the component of normal stress is continuous across the interface, when the surface tension force due to curvature is considered:

The solution of the above equation and the boundary conditions for a parallel sided segment, using perturbation analyses, has been presented by Clark and Dombrowski [ ]. The solution to the case where the viscosity of the medium is significant have been presented by Dombrowski and Johns [ ]. These solution are based on Squire’s theory of atomization, which predicts the development of sinuous and dilatational waves on the two surfaces of the sheet.

**Wave Propagation in Zone II**

From the description of the structure of the sheet in Zone II, stated in the preceding sections, waves which develop in Zone I would encounter particles with liquid held around them and thin segments of liquid containing no particles during their passage into zone II. Because the particles are not deformable, the waves would move then as rigid bodies held by surrounding liquids, i.e. both surfaces of the particles would move together in the same direction. Thus sinuous waves would be transmitted but dilatational waves would be suppressed by the particles. The Sinuous waves would deform the two surfaces of the pure liquid segments and grow and would encounter successive segments of pure liquid which thin rapidly due to particle separation and in all cases, the action of the ambient air on the sheet would not change considerably from that predicted by equivalent sheet free of particles.

In order to determine the stability we consider the growth of waves in zone II and note that the growth rate is dependent on the local sheet thickness. We hypostasize that the stability is determined by the evolution of the thinnest region in the inter particle space. Because the growth rate of waves is inversely proportional to the local thickness of the sheet.

**Evolution of the thinnest region in the inter-particles space.**

The evolution of the sheet thickness in the inter-particle space caused by their separation can only be predicted by the solution of the creeping flow equations for the pure liquid held between several particles- which task is beyond the scope of this work. Instead we note from the empirical observation the perforation occur a distance into Zone II, when the volume fraction of solids exceeds a critical value shown in equation (3) below.

\[
\psi_c = \frac{A_c}{\mu} \sqrt{d \rho \sigma} \quad (3)
\]
We exploit the occurrence of perforation within zone II under appropriate conditions to infer a rate of localized thinning from the perforation distance and the change in sheet thickness from that equal to the size of the particles to the molecular scale i.e. one hundred (100) nanometer thickness. Below this thickness the portion of pure liquid would burst spontaneously due to disturbances on the molecular scale (Madarelli et al.). Thus waves in this region of the sheet encounter a succession of rapidly thinning portions of liquid which is near periodic as liquid within the "cell droplets" thin rapidly from the size of the particles to the nano-scale. This is a further simplification from the reality as it would be expected that deeper into zone II and further away from the nozzle, the perforation times would decrease as the process intensifies.

\[ \text{Re} = \frac{\rho d^3}{\mu} \frac{\pi^2 U}{36 \nu^2 K} \]

\[ \frac{dH}{dt} = \left( \frac{K}{R^2} \right) U \]

**Objectives of Case Study**

In these case studies we test the above theories under the most adverse conditions such that conventional atomization theory would predict an outcome totally at variance with the theory outlined above and also observed in practice. We select sheets prepared from suspensions of neutrally dense Lycopodium particles wetted by water and also glass particles. The sizes of these particles are between 26 and 30 microns. These represent coarse particles. Another set of suspensions prepared using suspensions of fine glass particles of size 4.5 microns, is used to test the theory for fine or small sized particles. In both these cases, the densities and the viscosities of the resulting suspensions are expected to increase marginally therefore conventional atomization theory would predict lower growth rate of waves. Previous work has reported the break-up of such sheets formed from these suspensions by hydraulic pressure nozzle. The results are quoted and compared with calculations presented by this simple theory.

**Method**

We present a model for aerodynamic wave propagation in suspensions sheets derived from the two zone theory and test against experiments.

This work presents a theory of wave propagation and growth in sheets containing particles and models the case where the particle concentration is such that the perforation number exceeds
the critical value. In particular we compare the amplitude ratios at positions on sheets free of particles to those on sheets containing particles but produced under the same conditions by the same nozzle.

Wave propagation within the two zones of the sheets can be accounted for by imposing periodic disturbances represented by a complex Fourier series, on the two surfaces of the sheet and carrying out a perturbation analysis after the method of Squire [8]\textsuperscript{2} and Clark and Dombrowski [7]\textsuperscript{3}.

**The Model**

We make further simplifying assumptions:

- The viscosity of the medium is low therefore the flow is approximately irrotational
- The sheet is ejected into ambient air that is still at typically low velocities between 16m/s to 45m/s therefore flow of liquid and ambient air are laminar.
- The sheet thickness in zone I is given by eqn (1)
- Within Zone II, the particle induced thinning is akin to 2-dimensional necking has been shown to be orders of magnitude larger than thinning due to extension flow [4]\textsuperscript{1}.

**The Results**

The front and side views have been used to demonstrate the appearance of wave growth in the region of 1.5 cm from the nozzle orifice, a position where the equivalent sheet of water produced for comparison showed insignificant wave growth - see Figure [5]. We use the theory above to show that when particle induced thinning is taken into account then the faster wave growth is expected on sheets of suspended particles compare to sheets free of water. The consequence is that shorter sheets break-up lengths and coarser particles are expected due to the presence of particles in the sheets.

Within zone I, wave propagation conforms to the classical analysis. According to Squire’s theory (1955), sinuous and dilational waves grow on freely moving sheets and that sinuous waves grow much faster. In their analysis, the growth rate of disturbances of wavenumber greater than the critical value, $k_c$, can be represented by:
\[ \eta = \eta_0 e^{i(kx - \omega_i t)} e^{\pm \omega t} \]  
\[ \omega_i = \sqrt{-\frac{2\sigma^2}{\rho h} - \frac{4\rho_a k}{\rho^2 h^2} + \frac{2\rho_a U^2}{\rho h} k \left(1 + \frac{2\rho_a}{\rho h k}\right)} \]

Maximum growth rate occurs at the most dangerous wave number, \( k_D \), which is dependent on the sheet thickness but at high velocities is approximated by:

\[ k_D \approx \frac{\rho_a U^2}{2\sigma} \]  
\[ \omega_{im} \approx \frac{\rho_a U^2}{\sqrt{2\rho h \sigma}}. \]

Morris (1981) has shown that when the because the ambient air density is small compared to the density of the medium and the sheet thickness are small, is much less than unity and the wave speed is nearly equal to the sheet velocity. These conditions are met by the experimental conditions used in the work quoted above therefore the growth time of the waves is related to the axial position on the sheet by the relation

\[ t_g = \frac{R}{U}, \text{ where } R \text{ is the radial distance from the orifice of the nozzle.} \]

The sheet thickness varies with radial distance and therefore with growth time, therefore an average value of the growth rate can be found by integration thus:

\[ \bar{\omega} = \frac{1}{t_g} \int_{0}^{t_g} \omega_{im} \, dt \]  
\[ \text{where } t_g \text{ is the growth time.} \]
Sheets Containing Coarse Particles

In zone I the axial waves ‘see’ a sheet whose thickness is determined by bulk expansion therefore the time averaged growth rate, $\overline{\omega_1}$, over a portion of sheet which lies between the radius $R_0$ from the source of the sheet and $R$ at the wave number given in eqn (5) can be calculated analytically from eqn (6) and (7) to give:

$$
\overline{\omega_1} = \frac{\rho_a U^2}{2 \rho \sigma K / R} \frac{2(1 - (R_0 / R)^{1.5})}{3(1 - R_0 / R)}
$$

(8)

In zone II the thickness of the sheet subjected to curvature driven thinning, which is rapid. It is hypostasized that the waves ‘see’ a succession of elongating ‘cells’ which thin at a rate greater than the prediction in eqn (1) due to curvature driven flow and that the growth rate of waves is determined by the thinnest region in the interparticle space. Secondly it is noted that suspended particles cannot deform but will undergo purely translational motion. In determining the average growth rate of waves, $\overline{\omega_2}$, in zone II, the instantaneous rate is evaluated along the temporal evolution curve for the thickness of the thinnest region in the interparticle space according to eqn (7) and averaged numerically over the time required for the local thickness to diminish below 100 nanometres (beyond this point the sheet is likely to either perforate spontaneously or undergo capillary break up locally). Curve B in fig 2 shows an example of the evolution of the thickness of the thinnest region in the interparticle space of a sheet containing particles undergoing curvature driven thinning. The curve was calculated by treating the sheet in zone II as comprising planar droplet with circular cylindrical ends undergoing elongation. The thinning rate due to bulk elongation is shown by curve A in the above figure. It is noted that the wavenumbers calculated in Equation (5) need not necessarily remain the fastest growing wave in zone II, however its use serves the purpose of demonstrating the impact of curvature induced thinning on the wave action. The growth rates and amplitude ratios of waves over a portion of sheet, originating at the nozzle, one centimeter long and 40% of which lies in zone 1 and 60% of which lies in zone II are presented in
Table 1. The ratio of the amplitude of the exponentially growing waves to the amplitude of the initial disturbance was calculated according to:

\[
\frac{\eta}{\eta_0} = e^{\frac{R_1}{U} + \frac{R_2}{U}} \tag{9}
\]

where \(R_1\) is 0.40cm and \(R_2\) is 0.6cm. The growth rates and amplitude ratio on an equivalent sheet free of particles but of the same surface tension and density are also presented in table 1.

Comparison of columns two and three of the above table shows that over the range of ejection velocities encountered, the effect of curvature driven thinning is to double the growth rates. It is apparent from the above table that even at the moderate velocities and small growth times, the predicted amplitude ratios of the waves are increased by a factor varying from 4 to 20 when curvature driven thinning effects are taken into account. This agrees qualitatively with the observed early appearance of low amplitude waves on sheets containing particles compared to sheets free of particles. It is also noted that the calculated amplitude ratios of waves on perforated sheets containing coarse particles are an about two orders of magnitude less than that required to break up a sheet of water free of particles which agrees qualitatively with the observations in Fig. 1 and indicates that the curvature induced thinning significantly reduces sheet stability to the point that very little energy, in the form of low amplitude waves, are required to break up the sheets.

<table>
<thead>
<tr>
<th>Velocity (m/s)</th>
<th>Growth Rate of Waves (s(^{-1})) on Sheet free of particles</th>
<th>Growth Rate of Waves on sheets containing particles, which cause Curvature driven thinning</th>
<th>Amplitude Ratio ((\eta/\alpha)) of waves on sheets free of particles (no Curvature driven flow)</th>
<th>Amplitude Ratio ((\eta/\alpha)) of Waves on sheets containing particles, which cause Curvature driven thinning</th>
</tr>
</thead>
</table>

Author to whom queries should be sent [Type text]
**Table 1.** Comparison of Predicted Growth Rates and Amplitude Ratio of waves on Sheets containing particles and sheets free of particles.

<table>
<thead>
<tr>
<th>Velocity (m/s)</th>
<th>Growth rate of waves (per s)</th>
<th>Amplitude ratio ($\eta/a$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>20</td>
<td>1.12E+04</td>
<td>1.90E07</td>
</tr>
<tr>
<td>25</td>
<td>1.75E+04</td>
<td>1.25E09</td>
</tr>
<tr>
<td>30</td>
<td>2.51E+04</td>
<td>8.25E+10</td>
</tr>
</tbody>
</table>

**Stability of Sheets Containing Fine Particles.**

As stated earlier in the presentation of the theory section, when suspensions comprise fine particles, the growth times would be longer and therefore waves would grow to significant amplitudes before the expansion flow thin the sheets to the size of the particles. In this section we compare the predictions of the simple prediction by Squire theory with the experimental observation recorded in Fig. 1. the sheets containing fine particles perforate at a distance from the nozzle such that if waves were absent then the sheet thickness (calculated according to eqn 3) would be greater than the particle size. This is not surprising as analysis shows that sinuous and dilatant waves of significant amplitude can be expected in zone I of sheets containing fine particles.
Table 2. Variation of Growth rates of Sinuous Waves and Amplitude ratio on sheets of Suspension of fine Glass Particles

<table>
<thead>
<tr>
<th></th>
<th>3.42E+04</th>
<th>5.44E+12</th>
</tr>
</thead>
<tbody>
<tr>
<td>35</td>
<td></td>
<td></td>
</tr>
<tr>
<td>40</td>
<td>4.47E+04</td>
<td>3.59E+14</td>
</tr>
<tr>
<td>45</td>
<td>5.66E+04</td>
<td>2.37E+16</td>
</tr>
</tbody>
</table>

Table 2 above shows significant growth rates and amplitude ratios of waves at the optimum wavelength at a radial distance from nozzle orifice of 5 cm. The Nozzle used is Titan 943. The sheet thickness predicted by eqn 3 is seven microns, which is greater than the particle size of ca 4.5 microns. The prediction have been carried out using conventional atomization theory. Aerodynamic Waves of significant amplitude ratios are predicted. As a results the sheet will be thinned by both sinuous and dilatational waves as shown in photograph in fig 3 thereby reducing the its thickness of sheets in the troughs below the size of particles in this region. Hence the particles effects described above will commence in the region which lies apparently in zone I. Dombrowski and Clark (1972) analysed the action of sinuous and dilatational waves on the two surfaces of a freely moving sheet and showed that the maximum difference in displacement of the two surfaces occur at half wavelength intervals and at a growth time, $t^*$, is given by:

$$\eta_2(x^*,t^*) - \eta_1(x^*,t^*) = k^2 H \eta_0^2 \left[ \cosh(2\beta \eta^*) - 1 \right] / 4$$  \hspace{1cm} (10)

when $kh << 1$, $\rho_a << \rho$ and $\rho_a << \rho \tanh(kh)$.

Therefore replacing $t^*$ by $R^*/U$ and evaluating the average growth rate from eqn (8) and noting that the source of the sheet is only a short distance, $R_0$, from the orifice, it can be deduced that at high phase ratios of fine particles, curvature induced perforation will occur when the sheet thickness equals the particle size at the radial position, $R^*$, such that;
\[ R^{3/2} = \left( \frac{9\rho KU^2}{32(\rho_s U^2 k - \sigma k^2)} \right)^{1/2} \cosh^{-1} \left\{ 1 + \frac{4(1 - R^*/d / K)}{k^2 \eta_0^2} \right\} \]

The above equation was solved by the method of back substitution and showed that the calculated value of \( R^* \) was slightly less that \( K/d \) (i.e. the prediction from eqn 3).

**Discussion and Conclusion**

The physical analyses of the expanding sheets of suspensions leads to adoption of the two zone model to describe the structure of sheets produced during the atomization of suspensions. Zone II can be described as comprising cells of liquids joined end to end undergoing elongation. It is inferred from the analyses presented that sheets containing coarse particles develop waves of significant amplitudes in Zone I, close to the nozzle. These waves are still small in amplitude but noticeable and one would predict that when volume fraction of solids is high, a significant proportion of the sheet is broken by perforations.

When the particle sizes are small, the conventional theory for wave growth can be used to predict the observed wavy nature of sheets and that Zone II lies in the troughs of the waves a greater distance from the nozzle. This suggests that atomization of suspension of fine particles would be similar to that of the suspension medium free of particles. However, they should not be identical because when the perforation number is greater that the critical value perforation would occur in the troughs of the waves leading to thickened rims around the holes and thereby presenting some anomalously large particles.
References


2. Dombrowski, N. and Fraser, R.P., A Photographic Investigation into the Disintegration of Liquid Sheets, Phil. Trans. R. Soc. of London. 1953, 247A,101 state pages

3. Fraser, R.P., Eisenklam, P., Norman Dombrowski, and Hasson, D., Drop Formation from Rapidly Moving Liquid Sheets, A.I.Ch.E.J. 1962; 8: 672-680


Fig 11: Schematic of the thickness profile of liquid within a "cell" before and after elongation - wdt
Figure 2: Example of the Evolution of the thinnest region in the interparticle space
Figure 3: Front and side views of sheets of suspensions and sheet of water free of particles. The Nozzle is Titan 521, Ejection Velocity is 22m/s  Courtesy of AICHEJ
Figure 4: Sheet of suspension of fine glass (4.5 microns) undergoing aerodynamic wave break-up as well as perforation in wavy zone.

Nozzle is Titan 943, velocity is 22m/s. Courtesy of AICHEJ