Aerospace propulsion often involves the spray and combustion of liquids. When a liquid is sprayed, large drops form first, in a process known as primary atomization. Then, each drop breaks up into smaller droplets, in a process known as secondary atomization. This determines final drop sizes, which affect the liquid’s evaporation and mixing rates and ultimately influence combustor efficiency. Little has been published concerning the secondary atomization of visco-elastic non-Newtonian liquids, such as gels. These substances have special potential as aerospace propellants, because they are safer to handle than their Newtonian liquid counterparts, such as water. Additionally, they can be injected at varying rates, allowing for more control than solid propellants. To learn more about the atomization process of these liquids, a liquid drop generator and a high-speed camera were used to create and measure the conditions at which different breakup modes occurred, as well as the time required for the process. These results were compared to experimental and theoretical results for Newtonian liquids. Based on the data, one can conclude that solutions that are more elastic require higher shear forces to break up. In addition, while Newtonian liquids form droplets as they atomize, visco-elastic non-Newtonian solutions form ligaments. As a result, a combustion system utilizing these types of propellants must be capable of generating these forces. It may also be necessary to find a way to transform the ligaments into more spherically-shaped droplets to increase combustion efficiency.


Keywords
Aerospace
Spray
Propulsion
Liquid
non-Newtonian
Viscous
Elastic
Visco-Elastic
Secondary
Atomization
Motivation

Many aerospace propulsion systems today operate by combusting liquid propellants. Before they can be combusted, these liquids must first be sprayed in a two-part process known as atomization. The first part, primary atomization, occurs when a liquid jet breaks up into large drops. The second part, known as secondary atomization, occurs after a drop is already formed but undergoes further breakup due to instability. This process determines final drop sizes, which affect the liquid’s evaporation and mixing rates, ultimately impacting the efficiency of the combustor in which the liquid is used (Guildenbecher, Lopez-Rivera, & Sojka, 2009). As the cost of using an aerospace vehicle is closely tied to its efficiency, the secondary atomization of propellants is of paramount importance.

Research problem

While much is known about the atomization of Newtonian liquids, such as water, very little is understood about the secondary atomization of non-Newtonian liquids, such as gels. Non-Newtonian liquids are increasingly being used as aerospace propellants for two reasons. First, they are more easily contained than Newtonian liquids, making them safer for humans to handle (Snyder, Arockiam, & Sojka, 2010). Second, they can be injected at varying rates, allowing for more control than solid propellants.

The aim of this study was to determine the breakup modes of visco-elastic non-Newtonian liquids, the conditions at which they occur, and the characteristic times of the atomization process. This is part of the overall goal to control the final droplet sizes, eventually enabling non-Newtonian liquids to be effectively used as propellants.

Theory

There are many non-dimensional parameters that can be used to describe secondary atomization. These include Weber number, Ohnesorge number, Deborah number, and Weissenberg number. Dimensionless initiation time is also a common parameter.

Weber number (We) measures the shear forces acting on a drop relative to its surface tension.

\[ We = \frac{\rho_d v_{rel}^2 d_0}{\sigma} \]

- \( \rho_d \) = density of liquid drop
- \( v_{rel} \) = velocity of surrounding fluid relative to drop
- \( d_0 \) = drop diameter
- \( \sigma \) = liquid surface tension

Depending on the Weber number, a drop will exhibit different breakup modes, which result in different final droplet sizes. Newtonian liquids have five distinct breakup modes (Pilch & Erdman, 1987), as illustrated in Figure 1.
The conditions at which these modes occur have been well-researched, and there is an overall consensus among researchers regarding the approximate conditions at which each mode occurs. Table 1 shows the transition Weber (We) numbers for Newtonian drops with Ohnesorge (Oh) numbers less than 0.1 (Guildenbecher et al., 2009).

The Ohnesorge number (Oh) is another important non-dimensional parameter. It is a measure of the viscosity of a liquid relative to its surface tension. In the case of visco-elastic liquids, a non-constant viscosity is used, since the viscosity of a visco-elastic liquid depends on the shear rate, which is a function of both the drop’s diameter and the relative velocity of the surrounding fluid.

\[
Oh = \frac{\mu}{\sqrt{\rho d_0\sigma}}
\]

\(\mu\) = viscosity of the liquid  
\(\rho_d\) = density of liquid drop  
\(d_0\) = drop diameter  
\(\sigma\) = liquid surface tension

Another parameter is the initiation time (T\(_{\text{ini}}\)), which is the time required for the drop to deform from a sphere to an oblate spheroid (similar to the shape of a red blood cell) once it is subjected to shear forces, as shown in Figure 2 (Lopez-Rivera & Sojka, 2009).

Although there are many formulas that can be used to calculate non-dimensional initiation time, this study used the following equation by Ranger and Nicholls (1969).

\[
T_{\text{ini}} = t * \frac{v_{\text{rel}}}{\epsilon^{0.5} d_0}
\]

\(t\) = dimensional initiation time  
\(v_{\text{rel}}\) = velocity of surrounding fluid relative to drop  
\(d_0\) = drop diameter  
\(\epsilon\) = a non-dimensional constant representing acceleration due to drag

The previously mentioned non-dimensional parameters are used for all non-Newtonian liquids, regardless of their elasticity. The following non-dimensional parameters are used for elastic non-Newtonian liquids.

Deborah number (De) is a non-dimensional measure of the relaxation time of a liquid. The higher the Deborah number, the more elastic a liquid is, so the longer it takes to return to its original state when acted on by shear forces.

\[
De = \frac{t_c}{t_p}
\]

\(t_c\) = relaxation time  
\(t_p\) = Rayleigh time scale

The Weissenberg number (Wi) is a non-dimensional parameter that is the product of the deformation rate and the relaxation time of the liquid drop.

\[
Wi = \dot{\gamma} * \lambda
\]

\(\dot{\gamma}\) = deformation rate  
\(\lambda\) = relaxation time

Overall experimental approach

This study of visco-elastic non-Newtonian liquids was primarily experimental and involved the use of a drop generator, an air jet, a high-speed camera, and computer software to record and analyze the breakup process of various solutions designed to simulate gelled aerospace propellants. Specifically, for each solution, the conditions at which different breakup modes occurred and the time required for drops of the solutions to deform and break were recorded. These results were then compared to those of water, a Newtonian liquid.

Solution creation and characterization took place in the Rheology Laboratory in the Food Science Building at Purdue University. Solution atomization took place in the Combustion Laboratory, part of the Maurice J. Zucrow Laboratories at Purdue University.

Solution creation

The non-Newtonian liquids used in the experiments were mixtures of xanthan gum and distilled water at concentrations of 0, 0.1, 0.3, and 0.5% xanthan gum by mass. For each run, 750 mL of solution were created. The mass of the xanthan gum powder was measured using an

<table>
<thead>
<tr>
<th>Mode</th>
<th>We Range</th>
</tr>
</thead>
<tbody>
<tr>
<td>Vibrational</td>
<td>0 &lt; We &lt; ~11</td>
</tr>
<tr>
<td>Bag</td>
<td>~11 &lt; We &lt; ~35</td>
</tr>
<tr>
<td>Multimode</td>
<td>~35 &lt; We &lt; ~80</td>
</tr>
<tr>
<td>Sheet-thinning</td>
<td>~80 &lt; We &lt; ~350</td>
</tr>
<tr>
<td>Catastrophic</td>
<td>We &gt; ~350</td>
</tr>
</tbody>
</table>

Table 1. Transition We for Newtonian drops with Oh<0.1.
analytical balance with a resolution of 0.1 mg. The mass of the distilled water was measured using a standard balance with a resolution of 0.1 g.

To create the solutions, the water and xanthan gum were combined in a large beaker (at least 1L) using a magnetic stirring rod and plate that ran for at least 24 hours before characterization. Since xanthan gum solutions are known to deteriorate over time, special effort was taken to ensure that solutions were created and characterized as close as possible to the experiment date (see Table 2).

**Solution characterization**

After the solution mixed for at least 24 hours and could be considered homogeneous, the density of the solution was measured. A graduated cylinder (with a resolution of 1 mL) was placed on the standard balance (which has a resolution of 0.1 g). The mass was set to 0, and then 25 mL of solution was added to the cylinder. The mass of the added solution was divided by 25 mL to get the density in g/mL (or g/cm$^3$).

Next, it was characterized using the AR-G2 Rheometer, manufactured by TA Instruments. For the characterization, standard rotational mapping was used, and the temperature was set to 20°C. First, a flow procedure was run, with shear rate ranging from 1,000 to 0.1. Measurements were taken such that there were 5 data points per decade, and the percentage tolerance was set to 5%. Second, an oscillation procedure was run, with strain ranging from 1 to 0.01. As before, measurements were taken such that there were 5 points/decade, with a 5% tolerance. Third, another oscillation procedure was run, with angular frequency ranging from 50 to 0.1. As before, measurements were taken such that there were 5 points/decade, with a 5% tolerance. The data file from each procedure was analyzed using the Carreau model, which gives zero-rate viscosity, infinite-rate viscosity, consistency index, and rate index. Relaxation time was found by studying the second oscillation test and determining at what angular rate the delta value was 45 degrees. The reciprocal of this angular rate was the relaxation time. All the characterization data files were saved in case another model is chosen in the future.

The solution’s surface tension was measured using the CSC Precision Tensiometer, which utilizes a DuNouy ring. For the surface tension measurement, a glass dish and the DuNouy ring were sterilized using a solvent (ethanol), distilled water, and a propane torch. The solution was poured into the dish, and the dish was placed on the stand. The ring was placed on the tensiometer arm, and the tension in the wire was set to 0 dynes/cm. The height of the stand was adjusted such that the ring was submerged in the solution. The dial below the stand and the tension in the wire were adjusted slowly and simultaneously until the ring broke free of the solution. This surface tension was recorded (the device measures to the nearest 0.1 dyne/cm). This process was repeated three times per solution, and the average of the three trials was taken. A summary of the characterization results appears in Table 3.

<table>
<thead>
<tr>
<th>Solution (wt %)</th>
<th>Created</th>
<th>Characterized (&amp; Surface Tension Measured)</th>
<th>Experiment Ran</th>
</tr>
</thead>
<tbody>
<tr>
<td>Distilled water</td>
<td>9/6/10</td>
<td>9/6/10</td>
<td>9/10/10</td>
</tr>
<tr>
<td>0.1% xanthan gum</td>
<td>9/22/10</td>
<td>9/23/10</td>
<td>9/24/10</td>
</tr>
<tr>
<td>0.3% xanthan gum</td>
<td>10/20/10</td>
<td>10/21/10</td>
<td>10/22/10</td>
</tr>
<tr>
<td>0.5% xanthan gum</td>
<td>10/19/10</td>
<td>10/21/10</td>
<td>10/22/10</td>
</tr>
</tbody>
</table>

Table 2. Solution creation.

<table>
<thead>
<tr>
<th>Solution (wt %)</th>
<th>$\eta_0$ (Pa-s)</th>
<th>$\eta_\infty$ (Pa-s)</th>
<th>$\lambda$ (s)</th>
<th>$n$</th>
<th>$\sigma$ (dynes/cm)</th>
<th>$\rho$ (g/mL)</th>
<th>$\tau$ (s)</th>
</tr>
</thead>
<tbody>
<tr>
<td>water</td>
<td>0.001532</td>
<td>0.001089</td>
<td>1.00E+35</td>
<td>0.1544</td>
<td>74.1</td>
<td>0.988</td>
<td>N/A</td>
</tr>
<tr>
<td>0.1% XG</td>
<td>0.4553</td>
<td>0.001769</td>
<td>1.53</td>
<td>0.686</td>
<td>71.96666667</td>
<td>0.96</td>
<td>0.97632</td>
</tr>
<tr>
<td>0.3% XG</td>
<td>2.711</td>
<td>0.002934</td>
<td>2.206</td>
<td>0.7588</td>
<td>77</td>
<td>0.972</td>
<td>3.822468</td>
</tr>
<tr>
<td>0.5% XG</td>
<td>2.921</td>
<td>0.00000966</td>
<td>1.714</td>
<td>0.7282</td>
<td>79.23333333</td>
<td>0.96</td>
<td>9.00836</td>
</tr>
</tbody>
</table>

$\eta_0$ = Zero-Rate Viscosity  \quad \sigma = $Surface Tension

$\eta_\infty$ = Infinite-Rate Viscosity  \quad \rho = $Density

$\lambda$ = Consistency Index  \quad \tau = $Relaxation Time

$n$ = Rate Index

Table 3. Characterization results.
Solution atomization

After each solution was characterized, it was brought to the Combustion Lab for atomization. The lab is outfitted with an air supply system that is connected to an air nozzle, as shown in Figure 3 (Lopez-Rivera, 2010). In order to set up the experiment, the solution was first poured into a stainless steel pressure vessel, which was connected via a tubing system to a drop generator. The pressure vessel was then closed and pressurized until the internal pressure reached approximately 60 psi.

In order to see the drops, it was necessary for them to fall between a high-speed camera and a lighting system consisting of a lamp, a mirror, and a light-diffusing plate, as shown in Figure 4 (Lopez-Rivera, 2010). The camera was a Vision Research Phantom 7.1 high speed digital camera outfitted with a 105 mm Nikon lens, which recorded 800 x 600 pixel images at 4,700 frames per second. In order to calculate the drop sizes, a calibration image of a 5 mm square grid was used to provide a relationship between camera pixels and length (Snyder et al., 2010).

For each air-jet flow rate ranging from 15.2 to 58.0 m/s, videos of three drops falling and breaking up were recorded. The height of the dropper tip was increased as the air velocity increased so that the drop broke up entirely within the air jet. Each video was screened immediately after recording so that the air-jet velocity at which each breakup mode initiated could be recorded. The videos were then saved for more intensive data analysis.

Data analysis

Each video was viewed closely, and several data items were recorded. First, the initial spherical diameter of the drop was noted. Then, the initiation time (the time it takes for the drop to deform from a sphere into an oblate spheroid) was measured. For liquids that exhibited bag breakup, additional parameters related to bag formation and breakup were also recorded. Each video had a certain air flow rate, and the corresponding air velocity and density were calculated in a Microsoft Excel spreadsheet. This information, together with the recorded data, was used to calculate all the non-dimensional parameters mentioned in the theory section, allowing for direct comparison of the various liquids.
**Results**

Figures 5, 6, and 7 show some of the different types of breakup that were observed. Although the actual breakup modes for visco-elastic non-Newtonian liquids were similar to those of Newtonian liquids, one key difference is that the visco-elastic liquids formed ligaments instead of smaller droplets.

Several interesting trends were also noted in the various non-dimensional parameters, as seen in Figures 8, 9, and 10.

The transitions into bag and multimode breakup are exactly what one would expect (Figure 8). As the concentration of xanthan gum increases, the solution increases in elasticity, so a higher Weber number is required to reach a certain breakup mode. The transition into sheet breakup generally follows the same trend, except for the 0.1% solution, which requires the highest Weber number. The transition into catastrophic breakup also generally follows the same trend, except the 0.1% solution requires a lower Weber number than the distilled water.

The dimensionless initiation times for all the solutions are roughly the same, but the Ohnesorge numbers do vary considerably (Figure 9). The Ohnesorge numbers for the 0.3% and 0.5% solutions are approximately one order of magnitude higher than those for the water and 0.1% solution. In addition, the 0.3% and 0.5% solutions cover a much wider range of Ohnesorge numbers than the 0.1% solution and water.

Much higher Weber numbers were reached in this research compared to previous studies, and these additional results indicate that initiation time does in fact decrease slightly as the Weber number increases (Figure 10).

As expected, as the concentration of xanthan gum increases, the elasticity, and therefore the Deborah number, increases. However, it also appears as the concentration increases, the Deborah number range becomes wider and the initiation time range becomes narrower (Figure 11).

As expected, as the concentration of xanthan gum increases, the elasticity—and therefore relaxation time and Weissenberg number—increases. As was the case with the Deborah number, as the concentration increases, the Weissenberg number range becomes wider and the initiation time range becomes narrower (Figure 12).
Figure 9. Dimensionless initiation versus Ohnesorge number.

Figure 10. Dimensionless initiation versus Weber number.

Figure 11. Dimensionless initiation versus Deborah number.
Conclusion

From the recorded videos, it is clear that basic breakup modes are similar in Newtonian and non-Newtonian liquids. However, while Newtonian liquids form droplets as they atomize, visco-elastic non-Newtonian solutions form ligaments, which cannot be combusted as efficiently. Based on the data, one can conclude that solutions that are more elastic require higher shear forces to break up, and as shear forces increase, initiation time does decrease slightly.

In sum, a combustion system utilizing visco-elastic non-Newtonian liquids as propellants must be capable of generating very high shear forces to create the smallest fragments possible for the most efficient combustion. It may also be necessary to find a way to break the ligaments into more spherically-shaped droplets to further increase combustion efficiency, ultimately resulting in better performance and lower cost for aerospace vehicles in the future.

References


