Non-Newtonian Impinging Jet Spray Formation at Low Generalized Bird-Carreau Jet Reynolds Numbers

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Abstract

Non-Newtonian impinging jet spray formation was experimentally investigated using two different grades of Carboxymethylcellulose (CMC) mixed with deionized (DI) water (0.5 wt.-% CMC-7HF, 0.8 wt.-% CMC-7MF, and 1.4 wt.-% CMC-7MF). DI water was also tested as a reference liquid. Experimental rheological data obtained using rotational and capillary rheometers was characterized using the Bird-Carreau rheological model and a generalized Bird-Carreau jet Reynolds number $Re_{j, gen}$ was used to correlate atomization behavior. The resulting sprays were qualitatively and quantitatively studied using shadowgraphy. The general behavior exhibited by Newtonian impinging jet atomization was not observed when using non-Newtonian liquids; differences are ascribed to the shear-thinning nature of the non-Newtonian liquids employed. Depending on $Re_{j, gen}$ the observed spray patterns include: perforated sheet, ruffled sheet, tangled web, open rim, and ligament web. The experimentally measured maximum instability wavelength and sheet breakup length were observed to decrease with increasing $Re_{j, gen}$.

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Introduction

Adding a non-Newtonian agent to a liquid with typical Newtonian properties transforms the liquid into one exhibiting non-Newtonian behavior. Newtonian liquids have a constant value for viscosity independent of strain rate, whereas the viscosity of non-Newtonian liquids varies with strain rate. The ability of shear-thinning, non-Newtonian liquids to not flow easily unless under pressure is desirable for applications such as liquid rocket propulsion, since containment and cleanup would be more manageable in the event of an accident. Non-traditional liquids for use with aerospace propulsion systems have been investigated for decades [1]. One main disadvantage of non-Newtonian liquids, however, is the difficulty in atomization due to the increased effective viscosity of the liquid [2]. Non-Newtonian agents can generally be divided into two groups: organic and inorganic [3].

The Bird-Carreau model is a suitable choice to account for shear-thinning behavior of liquids that do not have a yield stress, because it can adequately capture the details of the experimentally measured viscosity. The expression for the Bird-Carreau model is:

\[
\eta(\dot{\gamma})_{BC} = \left[1 + (\lambda \dot{\gamma})^n\right]^{\frac{1}{n}} \left(\eta_0 - \eta_\infty\right) + \eta_\infty. \tag{1}
\]

In the above equation \(\eta(\dot{\gamma})_{BC}\) is the effective viscosity as a function of the strain rate \(\dot{\gamma}\). Zero- and infinite-strain rate viscosity limits are symbolized by \(\eta_0\) and \(\eta_\infty\) respectively. The parameter \(n\) is called the flow behavior index and describes the rapidly decreasing part of the viscosity curve. The parameter \(\lambda\) describes the fluid time response to a change in strain rate [4].

The general primary atomization behavior for impinging jet atomization involves two jets colliding to form a liquid sheet. Instabilities on the sheet cause it to fragment into ligaments, which then breakup into drops. The dimensionless parameters that are used for impinging jet atomization studies are the jet Reynolds number (inertial force to viscous force) and the jet Weber number (inertial force to surface tension force). Due to the strain-rate dependence of viscosity, the Reynolds number must be modified for non-Newtonian liquids. The generalized Reynolds number based on the Bird-Carreau model \(Re_{\gamma,\text{gen}-BC}\) [5] is used in this work as the primary dimensionless parameter for atomization. The expression for \(Re_{\gamma,\text{gen}-BC}\) is:

\[
Re_{\gamma,\text{gen}-BC} = \frac{\rho L U_j d_j}{\left[1 + \left(\frac{3n+1}{4n} \frac{\dot{\gamma}}{\lambda}\right)^\gamma \left(\eta_0 - \eta_\infty\right) + \eta_\infty\right]^{\frac{2n+1}{4n}}}. \tag{2}
\]

The terms in the above expression are the Bird-Carreau parameters along with the liquid density \(\rho_L\), jet velocity \(U_j\), and jet diameter \(d_j\). The expression for the jet Weber number \(We_j\) is:

\[
We_j = \frac{\rho L U_j d_j}{\gamma}. \tag{3}
\]

In the above equation the liquid/air surface tension is symbolized by \(\gamma\). The Weber number is unaffected from the Newtonian expression because the surface tension does not vary with strain rate.

Spray characterization of jet impingement with Newtonian liquids has been studied extensively in existing literature [6-9]. Four distinct regimes have been identified by Heidmann et al. (1957) [6]: Closed Rim with Drop Formation, Periodic Drop, Open Rim, and Fully Developed [6]. The jet Reynolds number has been observed to have a great effect on the flavor of the breakup process. For instance, in the “Closed Rim” regime, a distinct sheet was formed and drops were shed from the rim of the sheet. In contrast, for the “Fully Developed” regime the sheet was no longer even visible; ligaments and drops were observed to directly emit from the impingement point.

Although the atomization behavior of non-Newtonian liquids has been studied in recent years [10-16], there is still an incomplete understanding regarding the variety of effects that non-Newtonian behavior has on atomization. Furthermore, atomization regimes for non-Newtonian liquids have not been observed to be nearly as clear-cut [15]. The available literature indicates an inverse relationship between the jet Reynolds number and atomization characteristics such as maximum instability wavelength and sheet breakup length [10-13].

Chojnacki and Feikema (1995) [16] and Mallory and Sojka (2014) [12, 13] previously conducted investigations on water-based solutions of carboxymethylcellulose (CMC). This work seeks to expand understanding of the atomization behavior of CMC solutions by experimentally investigating the spray patterns and spray characteristics at low generalized Bird-Carreau jet Reynolds numbers.

Experimental Apparatus

A low-shear mixer was used in this work to formulate the non-Newtonian liquids. Special care was taken during the mixing process to ensure homogeneous mixing. The solutions were left to stir until they were determined to be homogenous by visual inspection. Two grades of carboxymethylcellulose, CMC-7HF (700 kDa) and CMC-7MF (250 kDa), were used as the non-Newtonian agents. The water based solutions used in this work were: 0.5 wt.-% CMC-7HF, 0.8 wt.-% CMC, and 1.4 wt.-% CMC-7MF. Deionized (DI) water was also tested as a reference liquid. The three CMC solutions can be described as homogenous, soluble, and highly viscous liquids.
A rotational rheometer was used to determine the bulk rheological properties at low strain rates. The cone-and-plate configuration (60 mm, 2.025° angle) was used in controlled-rate mode. A 5% tolerance was set for all measurements. The criterion for a data point to be considered valid was three consecutive measurements within the tolerance. A Sweep Up test was conducted with increasing strain rates. Sweeping the strain rates from lowest to highest values preserved the solution structure during testing. In order to determine if any thixotropic behavior was present, a Sweep Down test was also conducted.

A capillary rheometer was used to determine the viscosity of the investigated solutions at high strain rates. A 0.66 mm capillary die was used for all measurements. The pressure drop was recorded once fully developed flow was achieved in order to determine the liquid viscosity at a particular strain rate. The Weissenberg-Rabinowitsh correction factor as outlined by Morrison (2001) [4] was used for all measurements.

A du Noüy ring tensiometer was used to experimentally determine the surface tension of all three CMC solutions. The uncertainty in the measurement was calculated based on one standard deviation from a sample size of ten.

The unique experimental facility used to create the impinging jet atomization for this study was identical to the facility used by Rodrigues et al. (2015) [17]. Figure 1 provides a schematic of the facility. Rotational stages were used to set the impingement angle $2\theta = 100^\circ$. Translation stages were used to set the free jet length-to-orifice diameter ratio $x/d_0 = 60$. Designated tip elements were used to set the internal length-to-orifice diameter ratio $L/d_0 = 20$ and orifice diameter $d_0 = 0.686$ mm. The operating pressure was controlled in order to vary the mean jet velocity $U_j$. The flow rate from the orifices were measured for test durations of 30 seconds using a stopwatch. Measurements were repeated three times to ensure statistical significance and the mean jet velocity was calculated based on the measured flow rate.

![Figure 1. Schematic of experimental apparatus.](image)

<table>
<thead>
<tr>
<th>Quantity</th>
<th>Uncertainty (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$Re_{jgen-BC}$</td>
<td>6.4</td>
</tr>
<tr>
<td>$We_j$</td>
<td>4.5</td>
</tr>
</tbody>
</table>

**Table 1.** Experimental uncertainty for present work.

Still photography using the shadowgraphy technique with image analysis has been extensively used for atomization studies [7-13]. In this work the imaged were captured in the plane of the sheet formed by the two jets using a CCD camera. A double-pulsed Nd:YAG laser beam provided the back illumination. In order to reduce the coherence of the laser beam, the laser beam was first expanded and then projected to a diffuser. Figure 2 provides a schematic for the shadowgraphy set-up.

The percent uncertainty for the operating conditions was calculated using the Kline and McClintock method [18]. Table 1 presents the calculated uncertainty for the generalized Bird-Carreau jet Reynolds number and the jet Weber number. Further details on calculating the uncertainty is provided in Rodrigues [19]. One standard deviation was used as the experimental uncertainty for maximum instability wavelength and sheet breakup length measurements. A sample size of ten images was used at each test condition.

**Results and Discussion.**

The Bird-Carreau rheological model was used to characterize the strain-rate dependency of viscosity for the three non-Newtonian viscous liquids. The effective viscosity versus strain rate for 0.5 wt.-% CMC-7HF, 0.8 wt.-% CMC-7MF, and 1.4 wt.-% CMC-7MF are presented in Figures 3 - 5. The Bird-Carreau model satisfactorily characterized the non-Newtonian behavior of the three liquids. For all three non-Newtonian liquids, a Newtonian plateau was observed at very low strain rates. As the strain rate increased, shear-thinning behavior was observed. Since a Newtonian plateau at high strain rates was not observed, it was assumed that the infinite strain rate viscosity was that of the base fluid (water). This assumption is commonly used in literature in work such as Madlener and Ciezki (2012) [20]. The viscosity of water was taken to be the literature value of 0.001 Pa-s. Table 2 provides the Bird-Carreau rheological parameters for all three CMC solutions. Note that...
Table 2. Bird-Carreau rheological parameters for investigated non-Newtonian solutions.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>0.5 wt.-% CMC-7HF</th>
<th>0.8 wt.-% CMC-7MF</th>
<th>1.4 wt.-% CMC-7MF</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\eta_0$ [Pa-s]</td>
<td>0.576 ± 0.029</td>
<td>0.0596 ± 0.0030</td>
<td>0.309 ± 0.015</td>
</tr>
<tr>
<td>$\eta_\infty$ [Pa-s]</td>
<td>0.001</td>
<td>0.001</td>
<td>0.001</td>
</tr>
<tr>
<td>$n$ [-]</td>
<td>0.169 ± 0.008</td>
<td>0.427 ± 0.021</td>
<td>0.397 ± 0.020</td>
</tr>
<tr>
<td>$\lambda$ [s]</td>
<td>0.334 ± 0.017</td>
<td>0.173 ± 0.009</td>
<td>0.324 ± 0.016</td>
</tr>
</tbody>
</table>

for the Newtonian DI water: $\eta_0 = \eta_\infty = 0.001$ Pa-s and $n = 0$. Both sweep up and sweep down tests were conducted using the rotational rheometer in order to determine the thixotropic nature of the non-Newtonian liquids. All three CMC solutions were not observed to show any significant thixotropic behavior.

It was experimentally observed that all three CMC solutions have surface tension values very close to the literature surface tension value of water – 0.0728 N/m. Therefore, for jet Weber number calculations, the surface tension of all three liquids was taken to be the surface tension of water. The generalized Bird-Carreau jet Reynolds number and jet Weber numbers for the test conditions in this work are presented in Table 3.

Two impinging jets of 0.5 wt.-% CMC-7HF at $Re_{\text{gen-BC}} = 3,580$ and $We = 1,380$ were observed to form a liquid sheet bounded by a thick rim, as shown in Figure 6. Instabilities were observed on the liquid sheet. However, these instabilities led to sheet perforations rather than sheet breakup due to the dominating viscous and surface tension forces. Three-dimensional structures that can be described as wavy rims were observed at the regions of perforation. The rim surrounding the sheet was observed to detach into string-like ligaments,
which were then observed to breakup into a few small drops. This atomization behavior was called the *perforated sheet* pattern.

The spray formation from the collision of two jets of 1.4 wt.% CMC-7MF at \( Re_{j,gen-BC} = 4,320 \) and \( We_j = 1,380 \) was observed to be that of a distinct ruffled circular sheet – and therefore called the *ruffled sheet* pattern. Instabilities were once again observed on the sheet. However, instead of perforations, long ligaments that maintained connectivity with parts of the liquid sheet were observed. The ligaments then experienced breakup into several drops. This spray pattern is shown in Figure 7.

Moderately increasing the inertial force for the 0.5 wt.% CMC-7HF impinging jets resulted in spray patterns that contained tangled ligaments. Figure 8 shows the *tangled web* patterns at \( Re_{j,gen-BC} = 5,110 \) and \( We_j = 2,760 \). Instabilities on the liquid sheet led to the formation of large three-dimensional structures that resembled a tangled web. Downstream from the impingement point, long string-like ligaments were observed to detach from the web. A few large drops were observed to form from the ligaments. Further increasing the jet Reynolds and Weber numbers to \( Re_{j,gen-BC} = 6,280 \) and \( We_j = 4,120 \) as shown in Figure 9 and \( Re_{j,gen-BC} = 7,040 \) and \( We_j = 5,160 \) as shown in Figure 10, resulted in changes to the morphology of the web. At these generalized Bird-Carreau jet Reynolds number and jet Weber number it was somewhat difficult to determine where the liquid sheet experienced breakup and where the ligaments began to form. The structures inside the tangled web were observed to become increasingly dense with an increase in \( Re_{j,gen-BC} \) and \( We_j \). In addition, by increasing the inertial force a greater number of ligaments and drops were observed downstream of the web.

The spray formation of two impinging jets of 0.8 wt.% CMC-7MF at \( Re_{j,gen-BC} = 4,770 \) and \( We_j = 1,100 \) is presented in Figure 11. This atomization behavior was called the *open rim* pattern. A circular sheet with instability waves was observed near the impingement point. Downstream from the impingement point the sheet was observed to show bow-shaped ligaments that were connected at the sheet centerline. The ligaments were then observed to detach from the sheet and breakup into several drops. Striking similarities can be observed between this spray pattern and the spray

\[\text{Table 3. Bird-Carreau rheological parameters for investigated non-Newtonian solutions.}\]

<table>
<thead>
<tr>
<th>Liquid</th>
<th>( Re_{j,gen-BC} )</th>
<th>( We_j )</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.5 wt.% CMC-7HF</td>
<td>3.58E+03 - 7.04E+03</td>
<td>1.38E+03 - 5.16E+03</td>
</tr>
<tr>
<td>0.8 wt.% CMC-7MF</td>
<td>4.77E+03 - 7.81E+03</td>
<td>1.10E+03 - 2.76E+03</td>
</tr>
<tr>
<td>1.4 wt.% CMC-7MF</td>
<td>4.32E+03 - 8.10E+03</td>
<td>1.38E+03 - 4.12E+03</td>
</tr>
<tr>
<td>DI Water</td>
<td>9.13E+03</td>
<td>1.69E+03</td>
</tr>
</tbody>
</table>
Figure 8. Tangled web pattern of 0.5 wt.-% CMC-7HF at $Re_{j,\text{gen-BC}} = 5,110$ and $We_j = 2,760$: (a) spray formation, (b) drop formation.

Figure 9. Tangled web pattern of 0.5 wt.-% CMC-7HF at $Re_{j,\text{gen-BC}} = 6,280$ and $We_j = 4,120$.

Figure 10. Tangled web pattern of 0.5 wt.-% CMC-7HF at $Re_{j,\text{gen-BC}} = 7,040$ and $We_j = 5,160$.

Figure 11. Open rim pattern of 0.8 wt.-% CMC-7MF at $Re_{j,\text{gen-BC}} = 4,770$ and $We_j = 1,100$: (a) spray formation, (b) drop formation.
formation of 1.4 wt.-% CMC-7MF at $Re_{j,\text{gen}} = 4,320$ and $We_j = 1,380$ (Figure 2). Slightly increasing the jet Reynolds and jet Weber numbers for the 0.8 wt.-% CMC-7MF impinging jets to $Re_{j,\text{gen}} = 5,400$ and $We_j = 1,380$ led to a stark difference in the spray pattern. As shown in Figure 12, the liquid structure that was observed at the lower $Re_{j,\text{gen}}$ and $We_j$ was now observed to be very unstable. The structure was observed to breakup into bow shaped ligaments, which then experienced breakup into many small drops. Interestingly, this pattern was similar to the spray formation pattern of DI water at $Re_{j,\text{gen}} = 9,190$ and $We_j = 1,690$, which is presented in Figure 13. This DI water pattern is commonly referred to as the Open Rim pattern in literature.

Increasing the jet Reynolds and jet Weber numbers for the 1.4 wt.-% CMC-7MF impinging jets from $Re_{j,\text{gen}} = 4,320$ and $We_j = 1,380$ (Figure 7) to $Re_{j,\text{gen}} = 6,450$ and $We_j = 2,760$ drastically changed the spray formation pattern. As presented in Figure 14, a ligament web with various three-dimensional morphologies was observed. This atomization behavior was called the perforated sheet pattern. It was again difficult to determine where the liquid sheet experienced breakup and where the ligament web began. The ligaments were observed to separate from the web further downstream and eventually drops were formed from the ligaments. Interestingly, a similar spray pattern was observed for the 0.8 wt.-% CMC-7MF jets at $Re_{j,\text{gen}} = 7,810$ and $We_j = 2,760$, as shown in Figure 15. This is noteworthy because even though two different concentrations of polymers were used for the liquid formulation, the generalized Bird-Carreau jet Reynolds number satisfactorily accounted for atomization behavior and polymer

Figure 12. Open Rim pattern of 0.8 wt.-% CMC-7MF at $Re_{j,\text{gen}} = 5,400$ and $We_j = 1,380$.

Figure 13. Open Rim pattern of DI Water at $Re_{j,\text{gen}} = 9,130$ and $We_j = 1,690$.

Figure 14. Ligament Web pattern of 1.4 wt.-% CMC-7MF at $Re_{j,\text{gen}} = 6,450$ and $We_j = 2,760$.

Figure 15. Ligament Web pattern of 0.8 wt.-% CMC-7MF at $Re_{j,\text{gen}} = 7,810$ and $We_j = 2,760$. 
concentration itself does not appear to be a discriminating factor. Figure 16 shows the spray pattern of 1.4 wt.-% CMC-7MF at $Re_{j,gen-BC} = 8,100$ and $We_j = 4,120$. Further increasing the inertial force resulted in an increase in: the denseness of the ligament web, the number of ligaments separating from the web, and the number of drops formed. Note that these are all common characteristics to the spray patterns of the 0.5 wt.-% CMC-7HF impinging jets, but those spray formation were observed to be different. This was believed to be due to CMC-7HF possessing a higher molecular weight compared to the molecular weight of CMC-7MF.

Figure 17 presents the dimensionless maximum instability wavelength versus the generalized Bird-Carreau jet Reynolds number. The dimensionless maximum instability wavelength was generally observed to decrease with increasing $Re_{j,gen-BC}$. The dimensionless sheet breakup length was also generally observed to decrease with increasing generalized Bird-Carreau jet Reynolds number, as presented in Figure 18. Note that the maximum instability wavelength and the sheet breakup length were made dimensionless by the orifice diameter. Considerable variation was observed in the experimental measurements due to the unsteady nature of the impinging jet spray and one standard deviation was used for the vertical bars in Figures 17 and 18. The trends for both the maximum instability wavelength and the sheet breakup length agreed with previous Newtonian and non-Newtonian literature [6-13].

**Summary and Conclusions**

The impinging jet spray formation of three non-Newtonian liquids was investigated in this work. The rheology of the 0.5 wt.-% CMC-7HF, 0.8 wt.-% CMC-
7MF, and 1.4 wt.-% CMC-7MF solutions were characterized using the Bird-Carreau rheological model. Spray patterns based on the generalized Bird-Carreau jet Reynolds numbers were presented and qualitatively discussed. Depending on $Re_{j,gen-BC}$ the observed spray patterns include: perforated sheet, ruffled sheet, tangled web, open rim, and ligament web. Experimental measurements were presented for maximum instability wavelength and sheet breakup length. Both spray characteristics were observed to decrease with increasing generalized Bird-Carreau jet Reynolds number.

Nomenclature

- **BC**: Bird-Carreau rheological model
- $d_0$ : orifice diameter [mm]
- **DI**: Deionized
- **n**: BC flow behavior index [-]
- $Re_{j,gen-BC}$ : generalized BC jet Reynolds number [-]
- $Ub$ : mean jet velocity [m/s]
- $We_j$ : jet Weber number [-]
- $X_b$ : sheet breakup length [mm]
- **g**: liquid/air surface tension [N·m⁻¹]
- $\eta_0$ : BC zero-rate viscosity [Pa·s]
- $\eta_\infty$ : BC infinite-rate density [Pa·s]
- $\lambda$: BC time constant [s]
- $\lambda_m$: maximum instability wavelength [mm]
- $\rho_l$: liquid density [kg·m⁻³]

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References