Industrial production of nanocomposites could revolutionize the polymer industry. The NASA Goddard project Oriented Nanocomposite Extrusion (O.N.E.) is developing a process to produce desirable nanocomposites that can easily be scaled up and transferred to industry. This development uses a research-scale twin-screw extruder (TSE) based at the University of Maryland. A major need for this process is a method for online characterization of the polymer composition.

This project focuses on using pressure measurements, in combination with correlations for inferential, online characterization of the composition. Pressure measured near the entrance of the die is dependent on the temperature, flow velocity, and composition of the nanocomposite being produced. Since temperature and flow velocity can be directly measured, the correlation allows for online calculation of the composition of the polymer composite.

A correlation was developed that related pressure to the flow velocity and screw speed. The temperature probe was found to be faulty, so the correlation used screw speed measurements as a substitute for the temperature of the polymer melt. Temperature of the melt is affected by viscous heating due to the rotating screws, enabling screw speed to be used as a possible substitute. During the research, major findings include the faulty temperature probe and a periodic variation in pressure. The periodic variation in pressure was caused by variations in the polymer flow rate that results in thickness variation of the extrudate on the order of 20% (±10%). Recommendations are made for investigations of the temperature probes, the feeder flow rate, and the installation of the pressure probe.

I. INTRODUCTION

The main objective of this project was to develop a future control system for polymer composition based on an online characterization technique. Characterization of composition is difficult to achieve because composition is not a directly measurable property. However, composition affects several properties of a material, including viscosity. Viscosity, in turn, affects the pressure drop in a flow, which is directly measurable. The characterization technique developed as part of this project relates composition to the measured pressure drop through the die. Other factors that affect the pressure drop in the die were accounted for by the development of a correlation.

This objective aimed to enhance the production of high quality materials with properties specific to composition. The following steps were fulfilled:

- Installation of the pressure probe and corresponding equipment
- Experimentation with the pressure probe
- Development of a correlation to isolate compositional effects on pressure

Fulfillment of this objective provided consistency and accuracy to the polymer process, which is a major goal of NASA’s Oriented Nanocomposite Extrusion (O.N.E.) project. Implementation of the pressure probe also served to enhance the combinatorial approach that is being investigated for the O.N.E. project. Finally, the research uncovered several issues that have a critical role in the O.N.E. project.

Online characterization was done using a pressure probe located at the beginning of the die, as shown in Figure 1 in the Appendix. The pressure outside the TSE will be atmospheric, and the pressure probe will be measuring the gauge pressure. As a result, the pressure change will be equal to the gauge pressure that is measured. Since the flow velocity, or \( u \), is known, equation 1 can be used to solve for viscosity (\( \mu \)).

\[
\frac{\Delta P}{u} = k \cdot \mu \tag{1}
\]

The viscosity is a function of temperature and composition, as shown in equation 2a; if the temperature is known, the composition can be determined from the viscosity. Rearranging, as shown in equation 2b, the pressure change is a function of flow velocity (\( u \)), temperature, and composition. Rheology data has already been obtained for the nanocomposites, so the temperature and compositional terms have been accounted for. This rheology data can be seen in Figure 4 of the Appendix.

\[
\mu = f(T, \text{composition}) \tag{2a}
\]
\[
\Delta P = u \cdot g(T) \cdot h(\text{composition}) \tag{2b}
\]

1 The apparent viscosity is shear rate dependent. Since the screw design does not change, the shear rates do not change between runs, and the average viscosity can be determined.
Since temperature and flow velocity are known quantities, any pressure fluctuations based on changes in temperature and flow velocity can be compensated for by using the correlation. Any other pressure changes will be a result of a change in composition, related in the h(composition) term. Reiterating, these correlations allow online characterization of the composition via measurements of pressure by accounting for fluctuations in temperature and flow velocity.

II. EXPERIMENTATION

A. Methods

Experiments were run with pure polystyrene (PS) flowing through the TSE with constant temperature settings. The temperature zones of the TSE were set at the following temperatures: the five barrel zones were set at 425°F and the die zone at 325°F. The screw speeds and flow rates varied according to Table 1.

<table>
<thead>
<tr>
<th>Experimental Runs</th>
<th>Screw Speeds</th>
<th>Flow Rate</th>
</tr>
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<td>1.0 lb/hr</td>
</tr>
<tr>
<td>2</td>
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</tr>
<tr>
<td>9</td>
<td>60 rpm</td>
<td>1.0 lb/hr</td>
</tr>
</tbody>
</table>

Table 1: Experimental matrix for pressure experiments

B. Experimental Results

A noticeable decrease in the pressure drop occurred when the screw speed was increased, as shown in Figures 2a and 2b in the Appendix. Figure 2a shows that for a flow rate of 1.0 lb/hr, the pressure drop is almost cut in half (about 110 psi to 60 psi) when increasing the screw speed from 30 rpm to 60 rpm. At 2.0 lb/hr, Figure 2b shows an even larger decrease in the pressure drop (from 220 psi to 80 psi) when increasing the screw speed from 30 rpm to 60 rpm. For the higher flow rate, the magnitude of the pressure drop is higher for all screw speeds, but most noticeably for the lower screw speeds. Therefore, the higher screw speeds have a larger impact on the pressure drop. Also, the lower flow rate of 1.0 lb/hr has a narrower range of pressure values (between 50 psi and 150 psi). The higher flow rate of 2.0 lb/hr has a much higher pressure range (between 60 psi and 270 psi).

When the flow rate is increased, the pressure drop also increases, though the effects are less noticeable for higher screw speeds. In the Appendix, Figures 3a and 3b show that increasing the flow rate will increase the pressure drop with screw speeds of 30rpm and 45rpm. Also, Figure 3c also shows this trend for a screw speed of 60 rpm. Repeated runs were observed in Figure 3a for 1.0 lb/hr and in Figure 3c for 1.0 lb/hr. The observed overlap shows the reproducibility of the measurements.

Figure 3a shows more than a 100% increase in the pressure drop (from 100 psi to 225 psi) as the flow rate increases from 1.0 lb/hr to 2.0 lb/hr. Figure 3b shows an 80% increase in the pressure drop (from 75 psi to 140 psi). Figure 3c shows an even smaller increase of less than 35% in the pressure drop (from 60 psi to 80 psi). As mentioned earlier, the higher screw speeds tend to outweigh the effect of the flow rate changes.

III. DISCUSSION

A. Screw Speeds Effects on Pressure

Surprisingly, the screw speed strongly affected the pressure drop. As described in Equation 2b, the pressure drop should change only if the flow velocity, the temperature or the composition changes. The screw speed does not affect the flow velocity because of a simple material balance: the amount of material coming into the extrudate equals the amount of material coming out. Since the composition does not change either, it was concluded that the screw speed is significantly affecting the temperature.

Increasing screw speeds results in an increase in the viscous heating of the material. High screw speeds cause the material to be pushed against the screws with a higher force. This force causes an increase in heat from friction, and therefore, a higher temperature of the melt. An increase in temperature leads to a decrease in viscosity, causing a lower pressure. Table 1 in the Appendix shows the temperatures at the die taken for each run.

The temperature is slightly higher at the higher screw speeds, except for run #8, which had a high temperature because the run immediately before it had a high temperature. However, according to the rheometric investigation, a temperature change of two or three degrees Kelvins would not account for such a large change in the pressure drop (which was as much as a factor of 2 at a given flow rate). In the Appendix, Figure 4 shows the rheometry results determined earlier, as well as the highest and lowest viscosities found in the TSE experiments. The viscosity of the TSE melt was found using Equation 3.

$$\eta = \frac{\sigma_w}{\dot{\gamma}_w} = \left( \frac{-\Delta P}{L} \right) \frac{h^3 w}{12Q}$$

(3)

The higher frequencies of 100 rad/sec and 10 rad/sec correspond closely to the relevant viscosities. Figure 4 indicates that a change of one or two degrees correspond to a viscosity change of about 100 Pa·sec at 100 rad/sec and a viscosity change of 250 Pa·sec at 10 rad/sec.

Changing the screw speed from 30 rpm to 60 rpm caused a change in the pressure drop of a factor of 2, which would correspond to a viscosity change in the thousands of Pa·sec range. The calculated viscosities also show a viscosity...
change of about 2000 Pa-sec. However, the measured die temperature change corresponded to a viscosity change of only a few hundred Pa-sec. Therefore, the measured die temperature alone does not account for the change in the pressure drop. A change of a thousand Pa-sec would correspond to a change of at least 10 degrees Celsius. As a result, my professors and I believe that the temperature probe located at the entrance of the capillary die is not giving an accurate measurement of the melt temperature of the polymer. The polymer temperature must be changing by at least ten degrees to give such a large change in the pressure drop.

The temperature probes are touching other parts of the TSE and are unable to directly touch the melt without interfering with the extrusion process. If the temperature probes are touching the metal in the TSE, which is likely, the temperature will not accurately reflect the melt temperature.

In addition, the probe in the last barrel zone (the zone immediately before the die) was reading a temperature of over 400°F (477 K), and only six inches later, the probe in the die zone was only 325°F (436 K). A 75°F temperature change of the polymer in a span of six inches is not physically possible at the flow rate of 1.0 lb/hr. Therefore, the temperature measured by the probes is incorrect. In conclusion, the temperature probes are not accurately calculating the temperature of the melt and should be examined as soon as possible.

B. Correlation Development

According to equation 1 and 2, the pressure is directly dependent on flow velocity and indirectly dependent on temperature and composition. However, as noted in section IIIA, the temperature was not accurately given by the temperature probe in the TSE. Instead, the screw speed measurement gives a more accurate correlation of the melt temperature than the temperature probe readings. As a result, the screw speed will be used as a substitute for the temperature measurement.\(^4\) The average pressure measurements for each run are shown in Table 2 in the Appendix.

The correlation followed the form of equation 4a, and the exact correlation values are shown in equation 4b.\(^5\) The correlation fit best when screw speed had an exponential power of one; the screw speed was shown to be proportional to temperature. In the equation, the composition term for pure PS is unity.

\[
\Delta P = k_1 \cdot \text{FR} \cdot \exp\left(\frac{k_2}{\text{SS}}\right) \cdot h(\text{composition}) \tag{4a}
\]

\[
\Delta P = 50.8 \cdot \text{FR} \cdot \exp\left(\frac{21.4}{\text{SS}}\right) \cdot h(\text{composition}) \tag{4b}
\]

\(^4\) This substitution of screw speed into temperature should be thoroughly tested before implementation of the correlation.

\(^5\) FR is flow rate, SS is screw speed, and \(k_1\) & \(k_2\) are constants found for the correlation.

The correlation fit the data well as Figure 5 (from the Appendix) shows. Though the data fits well, the correlation should be thoroughly investigated. Investigation is required to ensure that screw speed is an accurate substitute for temperature. Also, reproducibility of data that would fit this correlation is necessary. Most importantly, the correlation should only be used when the temperatures are set at 425°F for the five barrel zones and 325°F for the die zone. Finally, the correlation may change when composition is taken into account.

IV. FINDINGS

A. Oscillations in Pressure

Short-term oscillations occurred in the pressure measurements. These oscillations occur with a frequency of about forty seconds, which can best be seen in the runs with a flow rate of 2.0 lb/hr and screw speeds of 45 rpm and 60 rpm (Figure 1b).\(^6\) These oscillations have an amplitude of about 20% of the average pressure. The oscillatory trends were investigated and were found to be associated with the flow rate variations. Early in the experimentation, I noticed that the flow rate out of the feeder oscillated. The flow rate was measured in a few of the experiments, with a sampling of one measurement every ten seconds. In the Appendix, Figure 6 compares the oscillations in pressure with the oscillations in flow rate.

Figure 6 shows that the oscillations in the pressure only slightly correlate to the flow rate changes. The pressure oscillations have a magnitude of about 20%, from 59 to 69 psi, while the flow rate varies by about 60%, from 0.7 lb/hr to 1.3 lb/hr. However, the period of the flow rate oscillations is similar to the forty second period of the pressure oscillations. During the flow rate investigation, I noticed that the flow rate would change several times between my measurements. The changes were significant, which affected the validity of these results.

The thickness of the extrudate composition investigation was also measured to associate the changing flow rates to the oscillation in the noise of the pressure measurements.\(^7\) If the flow rate is changing, the extrudate coming out of the TSE will change in total volume. Because the extrudate is coming out of a 1-inch wide slit, any change in the extrudate volume can be found in a changing extrudate thickness. The thickness measurements had static, measurable readings, while the flow rate readings changed too quickly to have accurate readings. The thickness of the polymer extrudate was measured for three meters and compared to the pressure measurements made during that time. The measurements were made every 1.8 inches, which corresponds to about 10 seconds at a 1.0 lb/hr flow rate. These measurements are plotted with the pressure changes in Figure 7 in the Appendix.

\(^6\) All the runs have these oscillations. The runs mentioned simply show the oscillations the clearest through the noise.

\(^7\) The maximum change in the thickness was found to be less than 20%; therefore, the flow rate out of the feeder could not change more than 20% over a 10 second period.
Interestingly, the oscillations in the pressure and thickness had about the same period. The pressure had an oscillatory period of about 0.35 m, while the thickness oscillated every 0.41 m. More importantly, the thickness varied from 1.1 to 1.3 mm, or about 20%. Likewise, the pressure varied from 210 to 250 psi, which is also about 20%. The correlation between the pressure and thickness in Figure 7 shows the pressure oscillations are most likely due to flow rate changes. The periods line up closely, and both vary about 20%. Further investigation should be done to confirm that flow rate changes account for the pressure oscillations, and that the origin of the flow rate changes lie in the polymer feeder system.

Also, in a polymer extrusion process, the polymer extrudate should have an even thickness. The fact that the thickness changes at all is cause for concern; a 20% variation in the polymer thickness is a major quality issue in the extrusion process. This issue should be investigated further. An extrudate that has a 20% change in thickness may not meet product specifications.

B. Noise

All the runs contain significant noise. The noise seems to be oscillatory, which is most noticeable for the 2.0 lb/hr runs. The oscillations were determined to be attributed to an oscillatory polymer flow rate. An analysis was done to minimize this noise. Noise occurs from random fluctuations in the pressure measurement and should balance out over a certain period of time. Intuitively, longer time scales result in a closer average to the overall average (which is calculated using the full time scale). However, shorter time scales produce a more efficient process. In the Appendix, Figure 8 shows how the average pressure measurement gets closer as the time scale of the measurements is increased.

For this particular example in Figure 8, if you took measurements for 3 seconds, the average of those measurements would have been 15% different than the total average of the measurement. If you took measurements for 20 seconds, the measurement average would only be 5% different. However, Figure 8 shows only the comparison for one specific run and one specific starting time.8 Table 3 from the Appendix averages the noise difference for all nine runs from the second data set and uses three different starting points.

As expected, measuring a longer time period will result in a more accurate average. Only taking measurements for 15 seconds will result in an average that is less than 5% different than the overall average.

V. RECOMMENDATIONS AND CONCLUSIONS

Most importantly, the correlation provided should act as a stepping stone for the O.N.E. project. More measurements should be made to ensure that the correlation accurately predicts the measured pressure before solving for the composition function of the correlation. Furthermore, a thorough investigation should be conducted to confirm that screw speed can serve as a substitute for temperature. Promisingly, the correlation could serve as a useful tool to account for the melt temperature without using the faulty temperature probe.

Another important finding is the discovery that the temperature probe located near the die entrance does not provide an accurate measure of the polymer temperature. An investigation should be made to confirm this discovery. One explanation is that the temperature probe is touching the metal surrounding the screws, which would read a different temperature than the temperature of the polymer melt. Other possible explanations are that the temperature probe is too close to the heater or that the temperature probe is too far from the melt to get accurate readings. If this discovery is confirmed, the result may be to (1) move the temperature probe, (2) find a different method to obtain temperature readings, or (3) change the combinatorial approach to rely less on melt temperature.

In addition, an important discovery was that the thickness of the extrudate varied by 20%. To fix this issue, an investigation of the polymer feeder should be conducted. This investigation may also reduce the large scale oscillations in pressure. Experiments should be made to confirm that the oscillations in pressure are related to oscillations in the flow rate out of the polymer feeder. As discussed earlier, measuring the flow rate from the polymer feeder display does not give an accurate flow rate; instead, the thickness of the polymer extrudate should be further investigated. If the investigation concludes that the thickness varies, the polymer feeder should be either calibrated or replaced. Having a constant thickness of the extrudate is vital to the O.N.E. goal of scaling the process up to industry.

Finally, this report recommends an experimental investigation to relate the pressure discrepancies to the installation of the pressure probe. Experiments should be made to test the hypothesis that a tighter installation of the pressure probe will cause a higher pressure reading. The experiment can be carried out by measuring the pressure readings while changing the amount of rotations to install the pressure probe. In addition, this experiment will allow for an analysis of noise in the pressure measurements. Tighter installation of the pressure probe will hopefully decrease the noise in the pressure measurements. If the installation procedure does not significantly reduce the noise, an average pressure can be taken over a fifteen second time frame to negate the noise.

8 The starting time refers to the data points where the each second interval would begin at. In this example, the 3 second time interval would average the 120th, 121st, and 122nd second data points.
Figure 1: Diagram of the TSE with the pressure probe

Figure 2a: Pure PS runs with changing screw speeds at 1.0 lb/hr

Figure 2b: Pure PS runs with changing screw speeds at 2.0 lb/hr

Figure 3a: Pure PS runs with changing flow rates at 30 rpm

Figure 3b: Pure PS runs with changing flow rates at 45 rpm
Figure 3c: Pure PS runs with changing flow rates at 60 rpm

Table 1: Average measured die temperatures

<table>
<thead>
<tr>
<th>Run #</th>
<th>Die Temp (K)</th>
<th>Screw Speed (rpm)</th>
<th>Flow Rate (lb/hr)</th>
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<td>9</td>
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Table 2: Average pressure measurements from each run

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<tr>
<th>Run #</th>
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<th>Average Pressure (psig)</th>
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Figure 4: Rheometry results at high frequencies and the calculated viscosities from the TSE runs
Figure 5: Experimental averages vs Predicted Values calculated from equation 4b

Figure 6: Comparison of flow rate changes to pressure oscillations

Figure 7: Thickness measurement oscillations compared to pressure noise

Figure 8: Noise Comparison for Run #5 with parameters of 2.0 lb/hr and 45 rpm starting at 120 seconds after the beginning of the run

Table 3: Deviations from the total pressure average
ACKNOWLEDGMENTS

I would like to first acknowledge Dan Powell of NASA GSFC and Arun Kota of UMCP who first brought the O.N.E. project into fruition. I would like to thank my technical advisors, Donald J. Kirwan and Mark T. Aronson, and my Science, Technology, and Society advisor, Ingrid H. Townsend. I would like to thank all who has assisted me on this project and my parents who, in a way, sponsored the project.

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10] Private communication with Arun Kota. Graduate student at the University of Maryland mechanical engineering department.
12] Private communication with Donald J. Kirwan and Mark T. Aronson. Professor / Associate Professor at the University of Virginia chemical engineering department.