

Mixing conditions of polymer and ceramic powder determined by ultrasound

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Abstract: Mixing process in the internal mixer has been monitored by ultrasonic technology real-time, non-intrusively and non-destructively. Visual observation, mechanical torque measurement, and ultrasonic signatures, such as amplitude of transmission and reflection echoes, were used for the diagnosis of the melting process of low density polyethylene (LDPE) and the mixing process of the melted LDPE with a calcium carbonate (CaCO_3) powder. Phenomena during the melting and mixing processes, including phase change from solid to melt, and distributing progress of CaCO_3 powder were successfully monitored by ultrasound. The ultrasonic signature was able to determine when the polymer had melted thoroughly and mixed with CaCO_3 powder completely. The presented ultrasonic technique can be utilized to optimize the melting and mixing processes.

Keywords: Mixing condition; internal mixer; ultrasound.

1. Introduction

Mixing, including both solids and liquids, is a fundamental and important process for many fields: combustion, the chemical, food and pharmaceutical industries, biotechnology, polymer processing, environmental engineering, and so on. (1-2). The internal mixer is one of the essential static mixing instruments widely used in most laboratories working on polymer or powder material process. It is usually used as a preliminary testing step for verifying the melting and mixing behaviors of polymers, chemical reactions in polymers, cross-linking and degradation of polymer before the actual processing in extruder. Ultrasonic technique is one of the candidates for monitoring the different melting and mixing stages during such process, because its basic signatures are associated with properties of materials, process dynamics, and product qualities (3-5). The objective of this research

is to investigate ultrasonic signatures during melting and mixing processes in the internal mixer for optimization.

2. Experiments

An ultrasonic delay line probe, as shown in Figure 1 (a), is installed into the mixing chamber of internal mixer. The central frequency of the employed ultrasonic transducer (UT) is 10MHz. The ultrasonic delay line probe can operate up to 600°C, and the UT end of the probe can be cooled down to a temperature lower than 40°C. A rotating blade with roller shape, as shown in Figure 1 (b), was installed into the internal mixer for grinding and mixing material. The dimensions of the head of the rotating blade are 36.7mm in diameter and 47mm in length.

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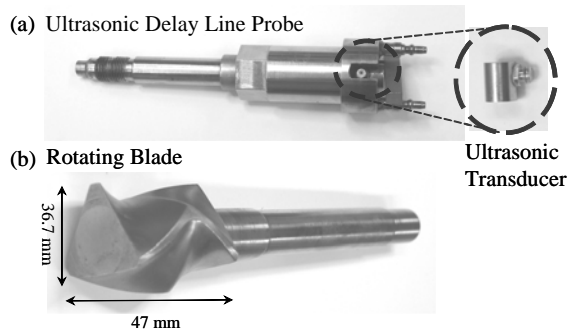


Figure 1. Photographs of ultrasonic delay line probe with ultrasonic transducer and rotating blade.

The internal mixer, as shown in Figure 2, is equipped with a mixing chamber, an ultrasonic delay line probe, a thermocouple and a torque meter connecting to the driving motor. The mixing chamber has two cylindrical housings for melting and mixing processes. These two housings have the capacity of 60ml. The internal mixer can operate up to 300°C and rotate up to 108 rotation per minute (RPM).

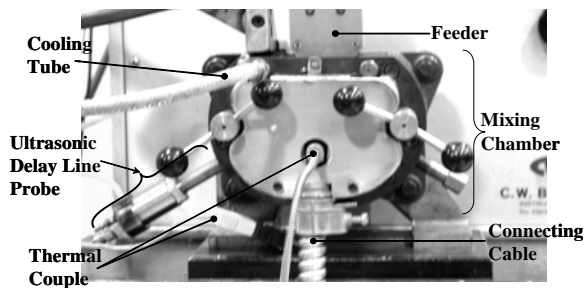


Figure 2. Photograph of mixing chamber with thermocouple and ultrasonic delay line probe.

A schematic drawing of a cross-section of the mixing chamber is given in Figure 3. It illustrates the paths of ultrasound propagating in the probe and the polymer melt. The first round trip ultrasonic longitudinal-wave echo, indicated by L^1 , is reflected at the probe end/polymer interface. The 1st round trip echo from the flight of the rotating blade half

through the polymer melt and reach back to UT is L_{2f} . The torque meter was attached to the rotator of the driving motor, and a K-type thermal couple was located at 0.2mm-depth from the inside surface of mixing chamber bottom, as shown in Figure 3. The torque and temperature profiles are used for comparison with ultrasonic data.

The material employed is LDPE and CaCO_3 . The procedures of melting and mixing processes were as follows: First, the empty mixing chamber was heated up to the setting temperature of 200°C and the blades were rotating at the desired speed of 14 RPM. Then the LDPE polymer pellets, 45g, were fed into the housings for melt. After the LDPE pellets being melted completely, the additional 4g CaCO_3 powder were fed into the mixing chamber for mixing with the melted LDPE.

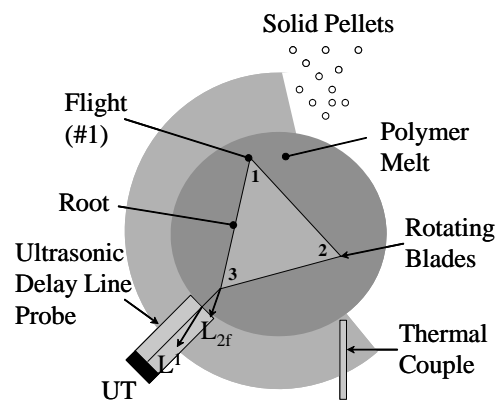


Figure 3. Schematic drawing of cross-section of mixing chamber, including ultrasonic delay line probe, rotating blade and thermocouple, presents ultrasonic paths in probe and polymer.

3. Results and discussions

Figure 4 shows the ultrasonic signal acquired by the ultrasonic transducer at the process time from 83.90 to 84.05s when LDPE had melted. The ultrasonic signal L^1 was observed in the entire process. When the flight of rotating blade arrived below the probe location, the L_{2f} echo appeared at time

delay of $50.4\mu\text{s}$ from process time 83.94 to 84.03s. Echoes reflected from the other two flights had the similar waveforms.

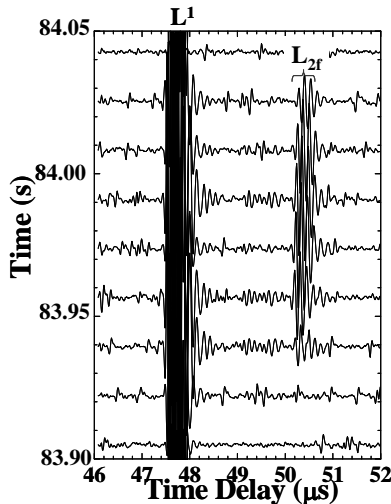


Figure 4. Sample traces of ultrasonic waveforms showing L_1 and L_{2f} echoes.

In order to investigate the correlation between ultrasonic signals observed and the processing cycle, the amplitude values of the L_{2f} echoes with respect to the process time were obtained. Figure 5 presents the amplitude of the L_{2f} echo from one of the three flights with respect to the process time, when polymer was melted. At process time around 77.71s, peak amplitude of the L_{2f} echo was observed. The duration of pulse was around 0.1s.

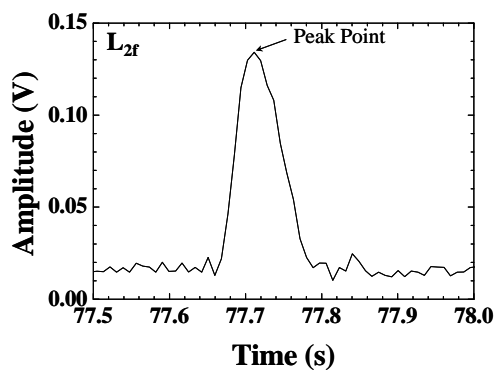


Figure 5. Amplitude of L_{2f} echo from rotating blade (see Figure 3) in process time after polymer is melted.

Figure 6 (a) ~ (d) present the amplitudes variation of torque, temperature, ultrasonic L_1 echo, and peak value of ultrasonic L_{2f} echo, respectively. They were recorded during the melting and mixing processes under the temperature setting of 200°C and rotation speed of 14 RPM. It is noted that the presented peak values of L_{2f} echo were from the same flight of the blade. Figure 7 (a) ~ (f) are photographs of the melting and mixing processes in Figure 6 at the process times of 37, 64, 85, 154, 172, and 180s, respectively. In the following, the torque and temperature profiles will be compared with the visible observation to explain the melting and mixing processes. Then the ultrasonic signals will be added for further investigation.

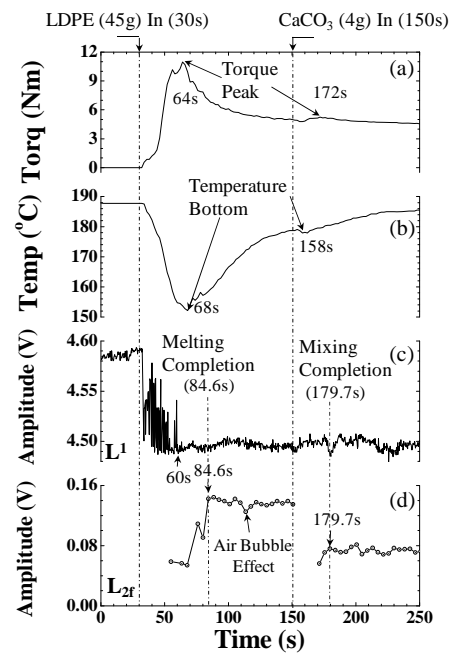


Figure 6. Variation of (a) torque, (b) temperature, (c) amplitude of L_1 and (d) peak value of L_{2f} echo during melting and mixing processes. 45g LDPE introduced at 30s; 4g CaCO_3 at 150s.

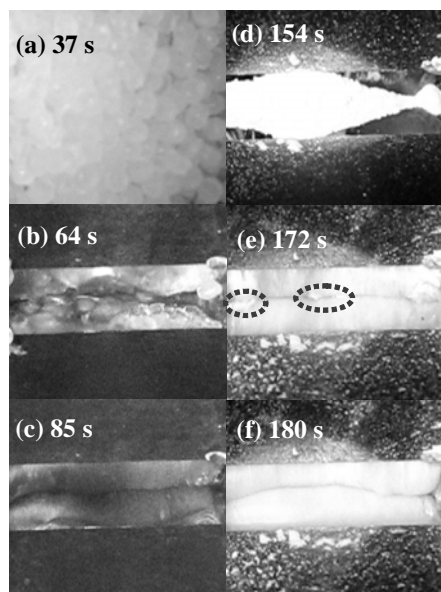


Figure 7. Photographs from feed window of melting and mixing processes in Figure 6 at process time of 37, 64, 85, 154, 172, and 180s.

In Figure 6 (a) and (b), before the process time of 30s, the mixing chamber was heated up to 188°C and the blade rotated at the speed of 14RPM. One can notice that even under the temperature setting of 200°C, the actual chamber temperature was 188°C, due to the heating loss from the feed window. At the process time of 30s, 45g of LDPE were fed into the mixing chamber. From process time of 30 to 64s, the torque value increased due to the friction, and temperature decreased due to the heat transfer between polymer pellets, rotating blades and mixing chamber. At process time of 64 and 68s, the torque and temperature reached their peak and bottom values of 10.9Nm and 152.1°C, respectively. After that, the torque decreased due to the reduction of viscosity of the polymer melt, and temperature increased due to the heat coming from the heating coils, friction and shear forces exceeding that absorbed by the polymer. At process time of 150s, additional 4g CaCO₃ powder were fed into the mixing chamber for the mixing process. At the process time of 158s, the torque and temperature values de-

creased to the bottom value of 4.8Nm and 177.9°C, respectively. The introduced CaCO₃ powder caused that viscosity reduced and heat transferred. After this point, the temperature increased due to the reason mentioned above. At process time of 172s, torque reached to peak value of 5.3Nm. After that, the torque decreased due to the reduction of viscosity. At process time of 250s, the mixing process stopped since the torque reached a steady state and the temperature almost recovered its original value.

The above results will be compared with photographs in Figure 7. In Figure 7 (a), at the process time of 37s, some of the pellets were still in the feed window and did not enter into the mixing chamber yet. In Figure 7 (b), at process time of 64s, which was the time that torque reached its peak value, most of the pellets were in the elastic solid form and bound with rotating blade. In Figure 7 (c), at process time of 85s, most of the solid pellets had been melted and dispersed in the mixing chamber. After the process time of 85s, even though the torque and temperature still varied, there were no solid or partially melted pellets left and the polymer melt didn't appear to change significantly. This may indicate that, after the process time of 85s, most pellets were already melted.

At process time of 150s, additional 4g CaCO₃ powder were fed into the mixing chamber. In Figure 7 (d), at the process time of 154s, some of the CaCO₃ powder were still in the feed window and not entered into the mixing chamber yet. In Figure 7 (e), at process time of 172s, most of the CaCO₃ powder was during the distributing process with the melted LDPE. But some of CaCO₃ powder still didn't mix with the melted LDPE, as indicated by the dashed circles. In Figure 7 (f), at process time of 180s, most of the CaCO₃ powder had been distributed into the melted LDPE. After the process time of 180s, even though the torque and temperature still varied, there were no undistributed CaCO₃ powder left and the mixed polymer melt didn't appear

to change significantly. This may indicate that, after the process time of 180s, most of the CaCO_3 powder was already mixed with melted LDPE.

The ultrasonic signals will be compared with the above results. In Figure 6 (a), at the process time of 32s, the torque value rose up quickly. In Figure 6 (b) and (c), at the process times of 32.4 and 34s, the amplitude of L^1 echo and temperature started to decrease, respectively. The drop in the value of L^1 echo was due to the fact that part of the ultrasonic energy was transmitted into the polymer melt through the probe end/polymer melt interface. The fluctuation of L^1 echo indicated the random contact of soft pellets and molten polymer to the probing end surface.

When the torque and temperature value reached to their peak and bottom values, the amplitude of L^1 echo also reached its lower level, but kept fluctuating. According to Figure 7 (b), most of the pellets were still solid with partially melting during this period. The fluctuations of the amplitude of L^1 stopped at the process time of 60s. At the process time of 55s, the ultrasonic L_{2f} echo, reflecting from the polymer melt/flight of the blade interface, started to appear. In the process time from 55 to 84.6s, the peak value of L_{2f} echo fluctuated, indicating the non-uniformity of the melting condition. After the process time of 84.6s, the peak value of L_{2f} echo reached its upper level, indicating that there were no solid or partially melted pellets, and all the LDPE pellets were melted. Figure 7 (c) also supported those diagnosed by ultrasonic technique. Therefore, when the peak value of L_{2f} echo reached its upper stable level, all polymer pellets had completely melted and further heating or rotating may not be necessary. The information of melting completion may be useful to optimize the process.

After the feeding of additional 4g CaCO_3 powder, the amplitude of L^1 echo kept at its lower level, but the peak value of L_{2f} echo disappeared due to the undistributed CaCO_3 powder. According to Figure 7 (d), around

this period, most of the CaCO_3 powder was still undistributed. At the process time of 171.4s, the ultrasonic L_{2f} echo started to appear again. In the process time from 171.4 to 179.7s, the peak of L_{2f} echo increased monotonically, indicating the progress of mixing condition. After the process time of 179.7s, the peak value of L_{2f} echo reached its upper level again, indicating that there was no undistributed powder and all the CaCO_3 powder was mixed with melted LDPE. Figure 7 (f) also supported those diagnosed by ultrasonic technique. Therefore, when the peak value of L_{2f} echo reached its upper stable level, all CaCO_3 powder had well mixed with the melted LDPE and further heating or rotating may not be necessary. The information of mixing completion may be useful to optimize the process.

4. Conclusions

Diagnosis of melting and mixing process in the internal mixer has been performed on-line, non-intrusively and non-destructively. The ultrasonic delay line probe with commercial UT, thermocouple, and torque meter were installed into the mixing chamber of internal mixer. The melting process of 45g LDPE from solid pellets to polymer melt, and the mixing process of the melted LDPE with additional 4g CaCO_3 powder were diagnosed by visual observation, torque, temperature, and ultrasonic signatures.

The phase change from solid to melt during melting process, and the distribution of CaCO_3 powder during mixing process can be detected by ultrasonic signatures. The timings of melting and mixing completion can be clearly determined by peak value of ultrasonic L_{2f} echo. Therefore, the presented ultrasonic technique might be an excellent approach to diagnose melting and mixing processes of polymer with powder in the internal mixer nondestructively and noninvasive.

5. References

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