Residence-time Measurements and Detection of Melt Heterogeneities

Quality Assurance. A multi-channel ultrasonic measuring system was used to study the homogenisation of polymer blends. It was demonstrated that this method is suitable for analysing the condition of the melt along the extruder screw.

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Consistent melt quality is vital for optimum extrusion. This calls not only for appropriate design features, but also for the capability to detect and process suitable controlled variables. Direct monitoring of the chemical and physical processes along the extruder screw during compounding is difficult, however. The classic approach has been to take samples at various positions along the screw and to analyse them off-line by methods, such as optical and electron microscopy or mechanical measurement.

In-line studies dealing with the detection of spatial and temporal melt inhomogeneities in the extruder are few [1]. While in-line transillumination methods [2] are described in the literature, these are restricted to transparent materials.

Ultrasonic techniques have been used for some time to perform in-line monitoring of extrusion [3,4,5]. It has been shown that ultrasonic velocity and attenuation can be linked to the viscoelastic properties, the melt viscosity, the filler content, the blend composition and the copolymer content. It seems obvious, then, that ultrasonic measurements might also be suitable for revealing melt inhomogeneities in the extruder screw. Added to which, ultrasound offers the possibility of continuously measuring residence time in the extruder [6]. Until now, this entailed adding some sort of marker (e.g. coloured plastics, mineral additives, metal shavings or radio tracers).

In-line measuring system

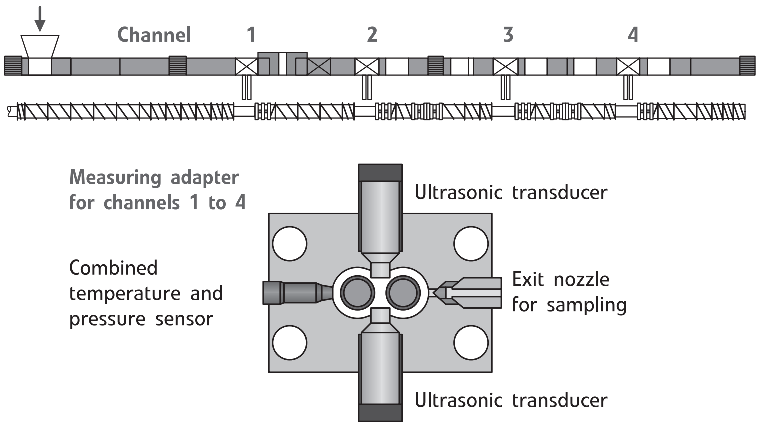

Fig 1. Location of measuring and sampling panels

Experimental Set-Up and Electronics

The in-line measuring system for detecting melt heterogeneities in the extruder screw consists of four barrel sections (Fig. 1), which are fitted with ultrasonic transducers and temperature/pressure sensors. The barrel sections are located in the plasticating and homogenizing zones of a twin-screw extruder (grade: ZSK 30, manufacturer: Werner und Pfleiderer). Pulse through-transmission requires that the ultrasonic signal pass between the two screws. The conveying and mixing elements in the vicinity of the backing plate were therefore replaced by a spacer sleeve. The transmitter transducer is activated by a spike pulse produced by means of a PC plug-in card (Matec SR9000) [5]. The ultrasonic waves penetrate the melt and are damped and delayed (Fig. 2). The second ultrasonic transducer receives the signal, which is amplified and then digitised by a fast AD transducer (Matec SR-9010). To be able to detect the signals from the four channels along the extruder screw, measuring-point selectors are switched between the sensors (ultrasound, pressure and temperature) and the measuring electronics (switching interval: 15 s).

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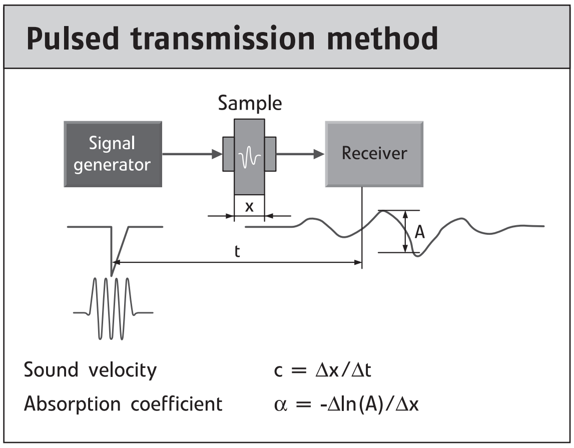

Fig. 2. The ultrasonic waves penetrate the melt and are damped and delayed

Residence–time Measurements

The residence time of the melt was determined by adding a small quantity of glass beads (Ø 150 mm) to polyethylene pellets (PE) at the feed end over the time interval of a few seconds ($t_0 = 0$ min). As the PE/glass bead mixture passes through signal channels 1 to 4, it brings about changes in sound velocity and attenuation due to sound scattering [5]. Figure 3 shows the curves for both parameters as a function of time for channel 3. Sound velocity and attenuation at time t_0 are the levels for pure polyethylene at 220 °C and 9 bar. After about 3 minutes, ultrasonic attenuation in channel 3 increases markedly, passes through a maximum at about 5 minutes and then slowly decays to the original value again. The minimum for the sound velocity at 5 minutes is typical of sound scattering [5].

Figure 4 shows how sound attenuation for the 4 signal channels along the extruder screw changes with time. The time shift in the maxima can be used to determine the mean melt speed. Dispersion of the glass beads in the polymer matrix differs in each of the signal channels. This is why the attenuation maxima have different heights and the time distribution functions for ultrasonic attenuation have different widths in the channels.

Figure 5 plots the times for the attenuation maxima t_{max} (above) and the full width at half maximum (below) for the 4 channels against the screw path length. The t_{max} values can be fitted into a straight line that returns a melt rate of 16.9 cm/min. The full width at half maximum for attenuation can be used to infer the degree of homogenisation or residence-time distribution. The lower section of Figure 5 shows a linear increase full width at half maximum with increasing screw path length. This is equivalent to a broadening of the glass-bead distribution along the screw.

The measured time-dependencies of ultrasonic attenuation $\alpha_{i\,(t)}$ in channels i = 1, 2, 3, 4 (Fig. 4) were used to calculate the auto-correlation (i = j) and the cross-correlation functions (i ≠ j):

$$A_{ij}(\tau) = \langle \alpha_i(t)\alpha_j(t+\tau)\rangle = \int_0^\infty \alpha_i(t)\alpha_j(t+\tau)dt$$

with the integral replaced by the sum for the discrete readings.
Figure 6 shows how channel 1 cross-correlates with the following channels $A_{1j}(\tau)$ (j = 2, 3, 4). The auto-correlation function $A_{11}(\tau)$ of channel 1 is shown additionally. The maxima of the cross-correlation functions $A_{1j}(\tau)$ reflect the time delay for an interruption (in this case: addition of glass beads) in channel 1. The shape and width of the cross-correlation functions are related to the residence-time distribution.

Heterogeneities in Blends

Homogenisation along the screw was determined by studying model blends of PE and polystyrene (PS) containing a mass fraction of 75 % PS. The pellets were mixed directly at the extruder feed section with the aid of separate metering devices for PE and PS.

The sound velocity/time curves relating to the PE/PS blends are shown in Fig. 7 for all channels. Preliminary tests have shown that the sound velocity increases with increasing PS fraction. To facilitate comparison, the coefficients for the sound velocity/pressure and sound velocity/temperature were used to convert the sound velocity c_i (t,T,p) to a reference state (T_0 = 220 °C, p_0 = 0 bar). The corrected sound velocity is $c_{0,i}$ (t), where i denotes the channel (1 to 4). The time curves for $c_{0,i}$ (t) for the various channels show a decrease in fluctuation about the mean of the time interval under inspection $\langle c_{0,i}\rangle$ with increasing path length. Since these fluctuations are not attributable to slow pressure and temperature fluctuations in the extruder, the only explanation is structural heterogeneities – similar to sound attenuation – or "propagating temperature fluctuations" (cold or warm zones) in the melt. The sound velocity responds very sensitively to changes in melt temperature on account of the very large coefficient of sound velocity/temperature.

Stationary fluctuations and propagating fluctuations in material and/or process parameters through the extruder can be distinguished by calculating correlation functions from the readings of the different channels. The correlation functions were calculated with the aid of the deviation in sound velocity $\Delta c_{0,i}(t) = c_{0,i}(t) - \langle c_{0,i}\rangle$ from its mean value in each channel (i = 1, 2, 3, 4). The resultant auto-correlation and cross-correlation functions

$$\Delta c_{ij}(\tau) = \langle \Delta c_{0,i}(t)\Delta c_j(t+\tau)\rangle = \int_0^\infty \Delta c_i(t)\Delta c_j(t+\tau)dt$$

for channels i=1 to 4 are shown in Fig. 8. Although only small fluctuations in sound velocity are recognisable for channels 2 to 4, the cross-correlation functions reveal a "correlation maximum" (arrows in diagram) that is shifted towards longer times with increasing path length ("channel number"). This correlation maximum corresponds to material fluctuations propagating through the extruder. The time shift of the "correlation maximum" with melt path length can be used to determine the residemce time, without the need for adding "markers".

Summary

Residence times for homogenisation of polymer blends (e. g. melting of granules and homogenising of granule blends) were studied with the aid of a multi-channel ultrasonic measuring system based on the pulse through-transmission method, which can register the sound velocity and ultrasonic attenuation at various locations along the screw of a twin-screw extruder during extrusion. At the same time, ultrasonic parameters were used to additionally measure the mean pressure and temperature.

The usefulness of the ultrasonic through-transmission method for analysing the state of the melt along the extruder screw was demonstrated. Application of cross-correlation functions enabled local heterogeneities to be distinguished from heterogeneities propagating through the extruder. The method can be used to determine the residence times from small material or parameter fluctuations, even without the addition of “markers”. ■

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Fig. 3. Curve for channel 3: Sound velocity and attenuation in a polyethylene melt (channel 3) after fast addition of glass beads (t_0 = 0 min)
Schallgeschwindigkeit = Sound velocity; Zeit = Time; Dämpfung = Attenuation; Kanal = Channel

Fig. 4. Curve for 4 signal channels: Temporal change in ultrasonic attenuation for different screw path lengths after fast addition of glass beads at time t_0 = 0 min to a polyethylene melt
Dämpfung = Attenuation; Zeit = Time; Kanal = Channel

Fig. 5. Evaluation: Time of attenuation maximum and full width at half maximum (from Fig. 4) as a function of screw path length
Halbwertsbreite = Full width at half maximum; Schneckenweg = Screw path length; Schmelze = Melt

Fig. 6. Auto-correlation and cross-correlation function of ultrasonic attenuation after fast addition of glass beads
Korrelationsfunktion = Correlation function; Kanal = Channel

Fig. 7. Comparison: Temporal fluctuation in sound velocity in a PE/PS blend (25 % / 75 %) at various positions along the extruder (channels 1 to 4)
Schallgeschwindigkeit = Sound velocity; Zeit = Time; Kanal = Channel

Fig. 8. Auto-correlation and cross-correlation of the sound velocity deviation for a PE/PS blend
Kanal = Channel