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In-line measurement of residence time distribution in twin-screw extruder using non-destructive ultrasound

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Abstract

In this study, we performed RTD measurement at the die exit of co-rotating twin-screw extruder using a non-destructive ultrasonic device. The ultrasonic device was attached at slit die and was composed of a steel buffer rod and 10 MHz longitudinal piezoelectric ultrasonic transducer. This in-line ultrasonic monitoring method is based on the ultrasonic response of $CaCO_3$ filled in polymer. The RTD is evaluated by variation of ultrasonic attenuation with time caused by change of the tracer concentration during extrusion. The ultrasonic tracer, pellet type of compounded $CaCO_3$ in polymer was used in this study. The effects of tracer concentration on RTD and flow patterns were studied. Evaluation for the residence functions at different screw speeds, feeding rates and screw configurations were also carried out.

Keywards : RTD, ultrasound, extruder, tracer, calcium carbonate

1. Introduction

The twin-screw extruder is one of the most important equipments for polymer processing, which is used mainly for blending and compounding operations and most recently for reactive processing. It is possible to assemble different screw profiles, including conveying and kneading elements as occasional demands. The standard setup for the extrusion process consists of an extruder, a die or shaping devices, and auxiliary equipments which include sizing, taking-up, and cooling units. During extrusion process, a material undergoes very complex thermo-mechanical changes which affect the qualities of final product. The problems in the final product are related to processing variables such as screw profile, screw speed and feed rate. Also, the levels of dispersive and distributive mixings are quite different depending on their variables. In the course of achieving optimum performance condition, the residence time distribution (RTD) curves give informations on the level of the axial mixing, initial, average and final residence times, presence of stagnation zones, etc., and they are efficiently used (Cassagnau *et al.*, 1991). RTD data also can be used for scale-up and improving equipment design (Todd, 1975). Many in and off-line evaluation methods have been proposed for RTD measurement (Todd, 1975; Gendron *et al.*, 1996; Hu *et al.*, 1999; Mélo *et al.*, 2002; Sun *et al.*, 2003). Some off-line techniques such as colorimetric and microscopic methods etc. are time consuming and yield few data points, not enough to allow for a detailed analysis at the tail region of the RTD curves. On the other hand, in-line methods are faster, but the technique that is used should be reliable. The oldest in-line technique is probably the γ-ray detection of irradiated samples (Thompson *et al.*, 1995) or the change in magnetic susceptibility using iron powder as tracer (Puaux *et al.*, 2000). Optical methods (light transmittance (Chen *et al.*, 1995) or light reflectance (Gao *et al.*, 1999; Gassner *et al.*, 1999)), changes in electrical properties (capacity (Curry *et al.*, 1991), conductivity (Unlu and Feller, 2001), dielectric constant (Choudury and Gautam, 1998)) and spectrometric methods (infrared attenuation (Nietsch *et al.*, 1997) and fluorescence (Bur and Galland, 1991; Cassagnau *et al.*, 1999; Hu *et al.*, 1999)) have also been used. Recently, ultrasonic technique, sensitive to the filler concentration of polymer suspensions, was proposed (Gendron *et al.*, 1996; Sun *et al.*, 2003). Besides, ultrasonic device have been effectively used as a powerful tool for the monitoring of the composition of polymer blends, the extrusion flow instabilities and the injection molding process, etc. (Franca *et al.*, 2000; Chen *et al.*, 1999; Youssef, 2001).

The objective of this study was to analyze the performance of a twin-screw extruder in terms of mixing and conveying according to different screw configurations, and to evaluate the flow patterns as a function of various processing conditions such as feed rate and screw speed, using

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ultrasonic monitoring system.

2. Ultrasonic monitoring system

Ultrasonic non-destructive test introduces high frequency sound wave into a test object to observe without altering or damaging it any way. Ultrasonic monitoring system for RTD measurement was attached on slit die in twin screw extruder as shown in Fig. 1. Ultrasonic system was composed of ultrasonic transducer, buffer rod, pulser/receiver, and data acquisition PC. Fig. 2 shows schematic diagram of slit die with ultrasonic transducer and buffer rod. Slit die also contains three pressure transducers and melt temperature transducer. In order to avoid deterioration of the piezoelectric ultrasonic transducer by high temperature during extrusion, a steel buffer rod was inserted between the transducer and the polymer melt. As shown in Fig. 3, the air cooling control system containing jet-cooler is attached to buffer rod, which play an important role in protecting the conduction of heat from slit die to ultrasonic

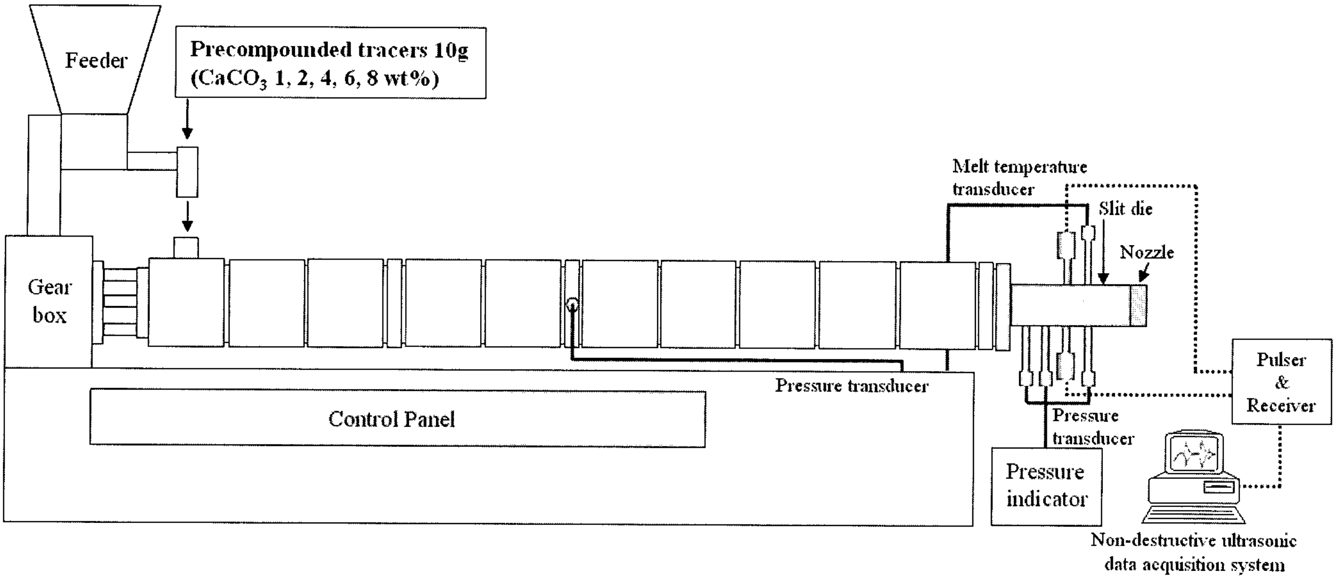


Fig. 1. Schematic diagram of co-rotating intermeshing twin-screw extruder with in-line ultrasonic monitoring system.

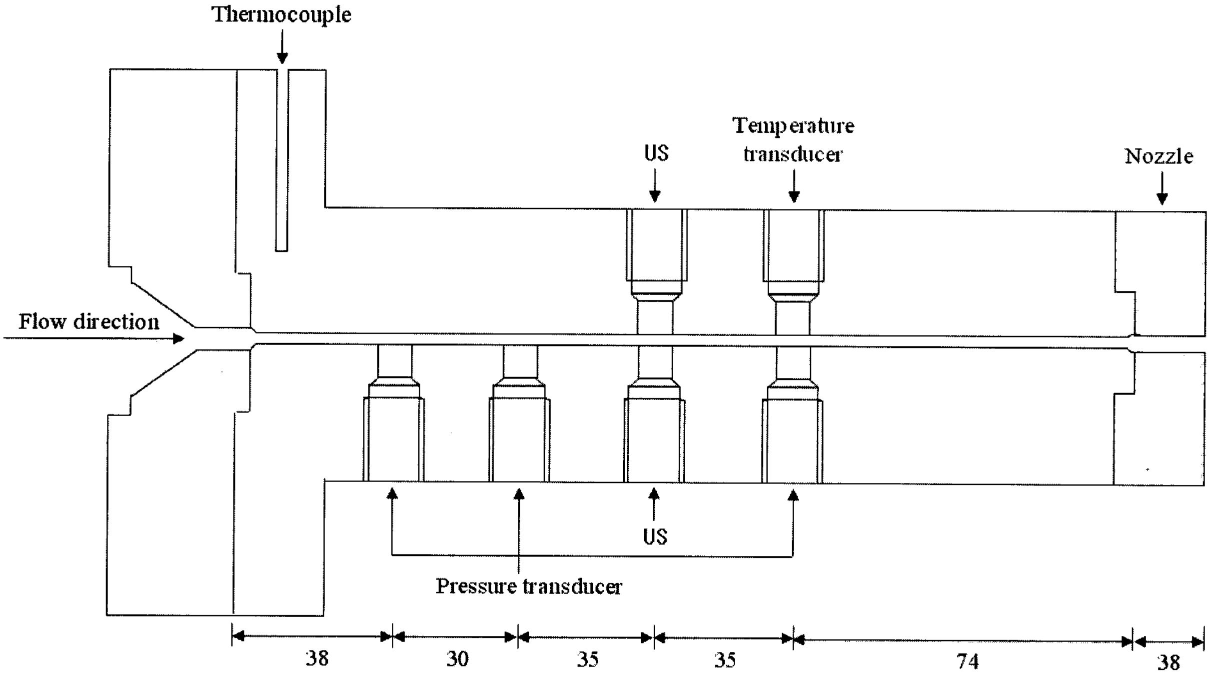


Fig. 2. Schematics of slit die designed for a twin-screw extruder.

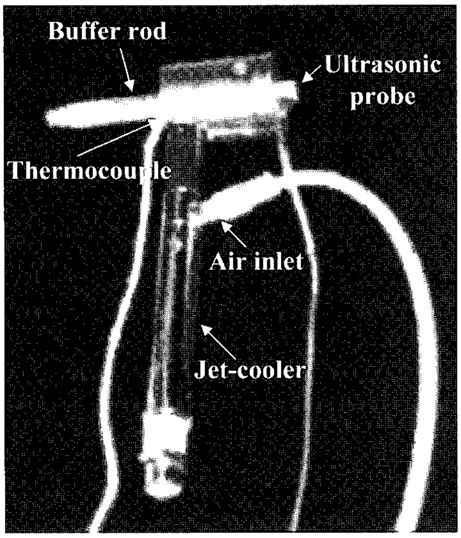


Fig. 3. Ultrasonic device with buffer rod and cooling system.

transducer. Two ultrasonic transducers were attached to buffer rods in the upper and lower plates of slit die. As illustrated in Fig. 4(a), the polymer sample is held in confinement between two axially aligned buffer rods, on which the ultrasonic transducer is mounted. Piezoelectric ultrasonic transducer has a center frequency of 10 MHz. RTD experiment was performed at transmission mode for longitudinal wave. The emitting transducer was repetitiously energized approximately every 0.01 s to produce a short burst of ultrasound, lasting typically 2-6 periods, as shown in Fig. 4(b). The acoustic pulses with amplitude A_0 travel down to the steel/polymer interface where part of the energy is transmitted in the polymer sample of thickness *e*. The residual acoustic pulses are then reflected back and forth between the two interfaces, its energy being diminished with reflection (Gendron *et al.*, 1996). This produces a series of echo signals, A_1, A_2, ..., exiting from the second interface and directed toward the receiving transducer. The echo signals were acquired with a PC-based data acquisition system. Fig. 5 shows a real signal which was measured during extrusion by ultrasonic monitoring system. Trigger indicates A_0 and successive echo signals show A_1 and A_2.

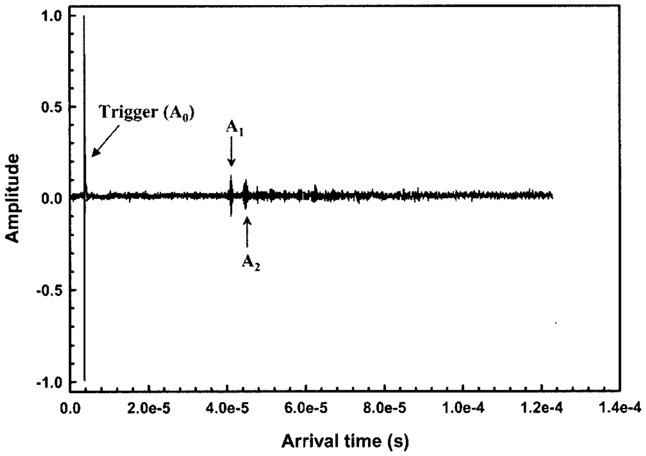
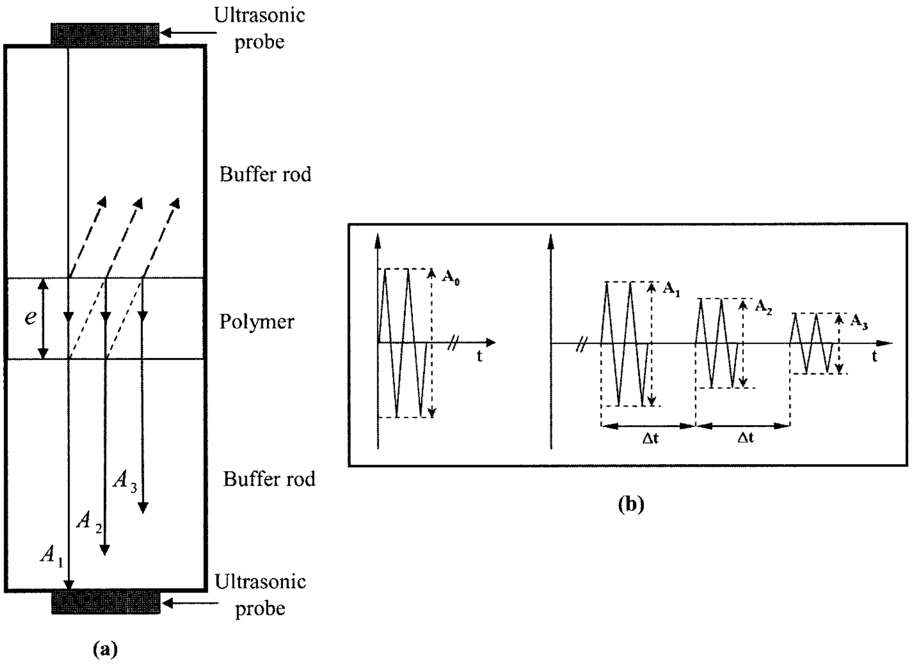


Fig. 5. Actual ultrasonic signal, which is measured by our ultrasonic monitoring system.



(a)

(b)

Fig. 4. Schematics of ultrasonic measurement.
(a) Technique, (b) Detection method

3. RTD determination

The attenuation (α) can be obtained by measuring the amplitude ratio of successive echoes as shown in Fig. 5.

$$\alpha = 20[\log_{10}(A_1/A_2)]/2e \tag{1}$$

In this study, we are only interested in detecting the presence of a tracer, relative to the attenuation level of tracer free during steady state extrusion conditions. A measure of the relative variation of the attenuation level can be obtained also by tracking only the amplitude of the first echo A_1, and it is possible to increase the sensitivity of the measurement. The relative variation of the attenuation level, α_{rel}, can be described as follows

$$\alpha_{rel} = 20[\log_{10}(A_1(0)/A_1(t))]/e \tag{2}$$

where $A_1(0)$ is the first echo A_1 which is measured in the absence of the tracer during steady-state extrusion and $A_1(t)$ is the first echo A_1 as a function of time (t) after injection of tracer into the extruder.

RTD includes the residence time distribution, $E(t)$, and the cumulative residence time distribution function, $F(t)$. The RTD curves are indicative of the flow behavior of the product passing through the extruder and provide a foundation for evaluating the extrusion process. $E(t)$ was mathematically defined by Dankwerts (1953). $E(t)$ curves represent the relative tracer concentration versus time at the die exit, similar to the relative attenuation value (α_{rel}). $E(t)$ can be defined in connection with the ultrasonic signal strength as follows

$$E(t) = \frac{C(t)}{\int_0^{\infty} C(t)dt} \cong \log_{10}[A_1(t)/A_1(0)]/\int_0^{\infty} \log_{10}[A_1(t)/A_1(0)]dt \tag{3}$$

By integrating $E(t)$, the cumulative residence time distribution function $F(t)$ can be obtained

$$F(t) = \int_0^t E(t)dt \cong \sum_0^t E(t)\Delta t \tag{4}$$

The mean residence time ($\bar{t}_m$) which is used to indicate the location of the distribution can be obtained from the following mathematical expression

$$\bar{t}_m = \int_0^{\infty} tE(t)dt \cong \sum_0^{\infty} tE(t)\Delta t \tag{5}$$

The variance of the residence time (σ^2) which is used to measure the spread of the distribution can be calculated by

$$\sigma^2 = \sum_0^{\infty}(t-\bar{t}_m)^2 E(t)\Delta t \tag{6}$$

The F function can also be described by dimensionless time variable (θ), which is the ratio of the measuring time (t) to the mean residence time ($\bar{t}_m$)

$$\theta = \frac{t}{\bar{t}_m} \tag{7}$$

$$F(\theta) = F(t) \tag{8}$$

It is worthwhile mentioning that the theoretical analysis of RTD are usually based on the ideal assumption of either plug flow or perfect mixing conditions expressed by, for plug flow

$$F(t) = 0; t < \bar{t}_m \tag{9}$$

$$F(t) = 1; t \geq \bar{t}_m \tag{10}$$

and for perfect mixing

$$F(t) = 1 - e^{-t/\bar{t}_m} \tag{11}$$

Plug flow usually takes place in tubular reactor and a fair approximation to perfect mixing can be easily achieved in a continuous stirred tank reactor (CSTR). Generally, F function in extruder shows axial mixing behavior between plug flow and perfect mixing flow.

4. Experimental

4.1. Materials

The polymer used for the RTD experiments was an extrusion grade low density polyethylene (PL1845 from Dow Plastics) with 0.91 g/cm^3 density and MFI=3.5 g/10 min. It had number and weight average molecular weights of 48,000 and 105,700, respectively, as determined by gel permeation chromatography. The filler used as a tracer was calcium carbonate (OM-1T from Omya Korea) and its particle size is from 1.0 to 1.8 micron. It was stearate coated in order to impart better particle dispersion in polymer matrix.

4.2. Procedure

The twin-screw extruder used in this study was a co-rotating, intermeshing twin screw extruder, SM Platek TEK25 with diameter of 25 mm, total screw length of 1025 mm and a ratio of screw length to diameter (L/D) of 41. The screws were assembled in a typical combination of conveying, shearing, and mixing elements. Twin-screw extruder instrumented with slit die, where ultrasonic sensor attached buffer rods, melt temperature and pressure transducers were flush-mounted (See Fig. 2), was used for the measurements filled polymer melt flow. The dimension of slit die channel was 2.5 mm height and 15.0 mm width. The temperature profile for the extruder varied from 90°C at the feed section to 185°C at the die. RTD experiments were performed for various feed rates, screw speeds, and screw configurations. Fig. 6 shows two screw configurations evaluated in this study. Their screw configurations were composed for the purpose of performing gas or liquid assisted polymer blend and alloy. The 1st screw configuration (See Fig. 6(a)) was assembled in combination of

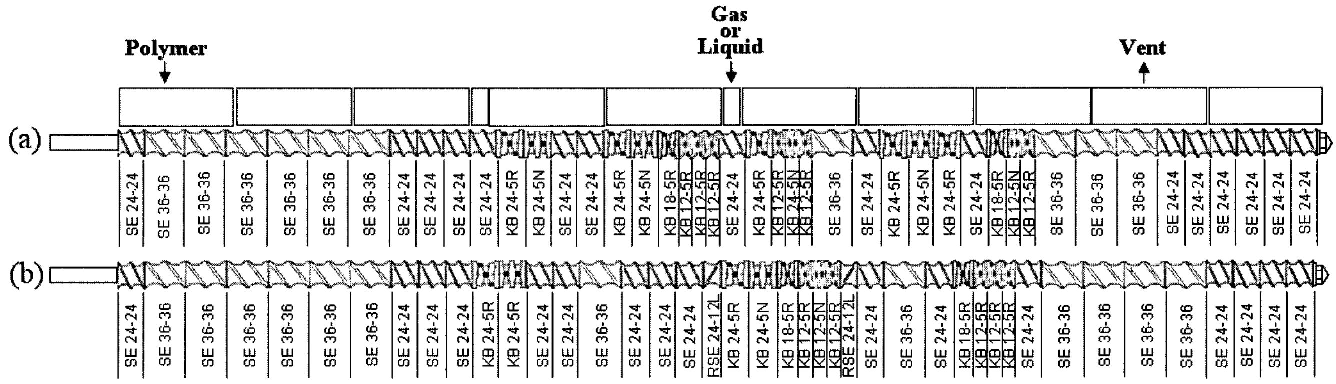
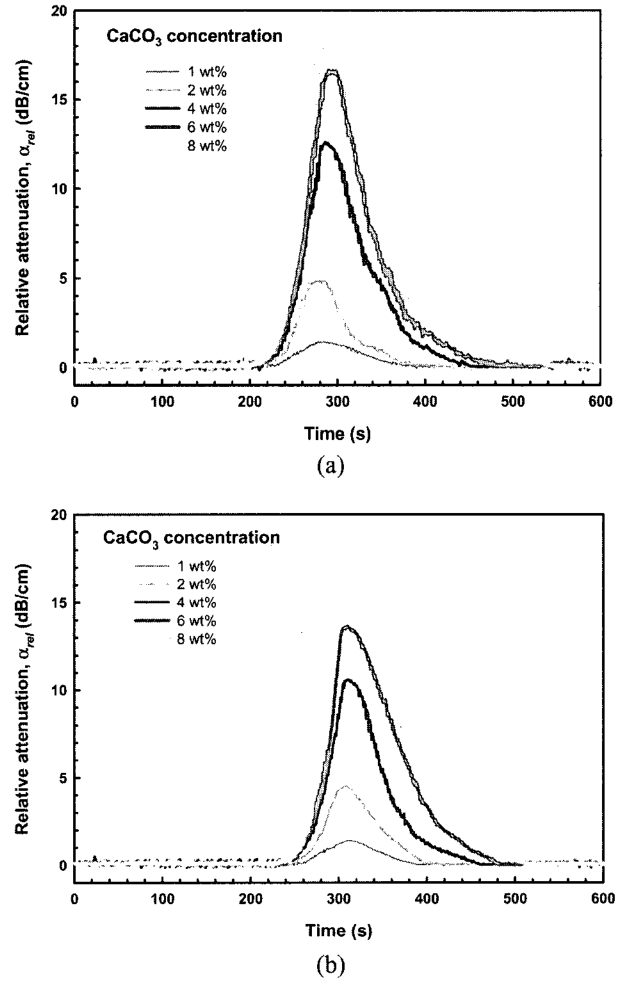


Fig. 6. Schematic representations of two screw configurations of the twin-screw extruder used.
(a) 1st screw configuration, (b) 2nd screw configuration

conveying, shearing, and kneading elements of some different types. The 2nd screw configuration (See Fig. 6(b)) has a different arrangement compared with the 1st screw configuration and contains additional two reverse elements. Ultrasonic tracer used in this study was pellet type, which is prepared by compounding appropriate amounts of $CaCO_3$ and polyethylene using extruder. The precompounded tracer of pellet type were used because $CaCO_3$ tracer of powder form and its behavior may different from that of precompouded pellets, flow to the clearance between the screw flight and the barrel wall. It may also stick to the walls of the feed throat. For all experimental cases, the compounded tracers (10 g) of pellet type containing various $CaCO_3$ concentrations (1, 2, 4, 6, 8 wt%) were injected directly in the feeding zone during extrusion (See Fig. 1).



(a)

(b)

Fig. 7. Effect of $CaCO_3$ concentration on RTD curves (screw speed of 40 rpm, feed rate of 2.1 kg/h).
(a) 1st screw configuration, (b) 2nd screw configuration

5. Results and discussion

5.1. Effect of tracer concentration

First of all, we carried out study on the relationship between the relative attenuation and the stearate coated $CaCO_3$ concentration. Figs. 7(a) and 7(b) display the resulting curves for two screw configurations at processing conditions of feed rate of 2.1 kg/h and screw speed of 40 rpm. For both screw configurations, the relative attenuation level was increased with increasing $CaCO_3$ concentration. Contrary to the result of off-line RTD test, we could observe tail region in the in-line RTD measurement using ultrasonic monitoring system. It was also shown that the tail region was enhanced as the $CaCO_3$ concentration increased. The mean residence time and the variance were slightly increased with increasing $CaCO_3$ concentration for both screw configurations. Their results were represented in Figs. 8 and 9. The mean residence time and the variance of 2nd screw configuration showed generally higher values compare to those of the 1st screw configuration.

5.2. Effect of screw speed and feed rate

The effect of screw speed and feed rate on the RTD curves was verified in two screw configurations. The effect

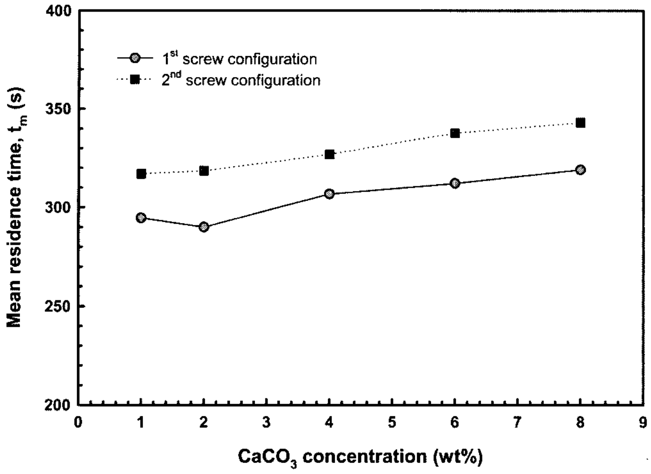


Fig. 8. The mean residence time ($\bar{t}_m$) as a function of $CaCO_3$ concentration (screw speed of 40 rpm, feed rate of 2.1 kg/h).

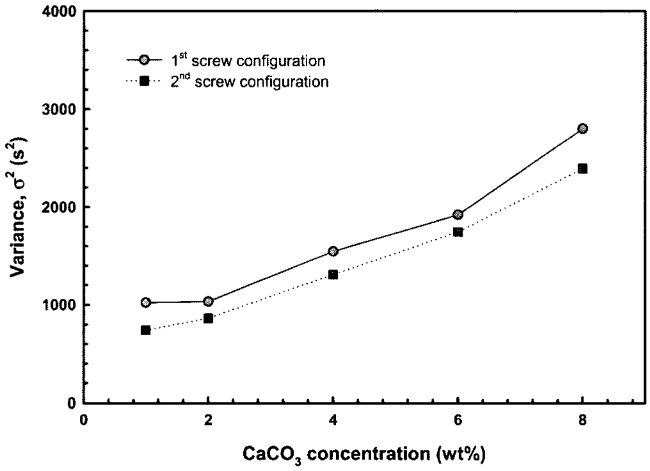
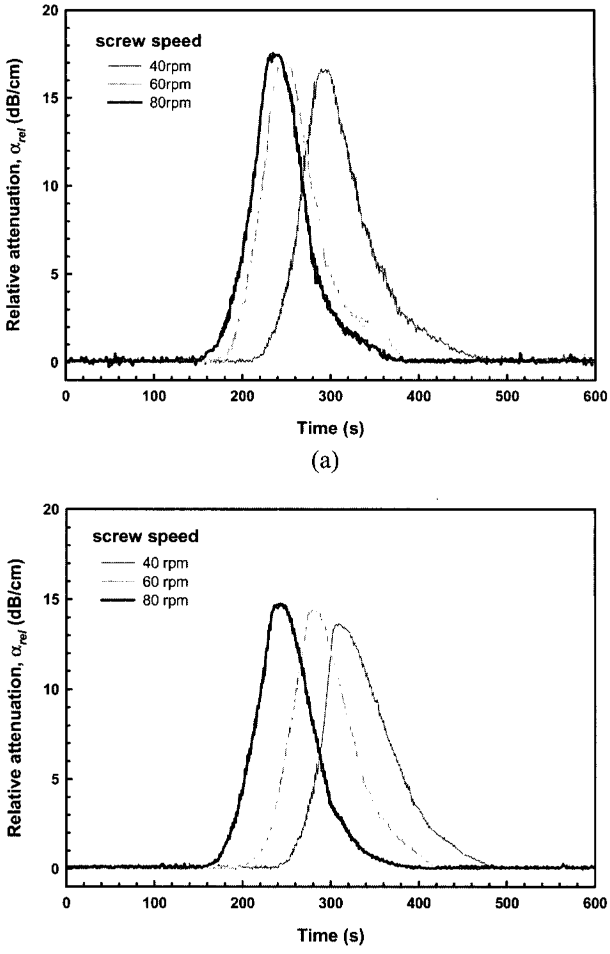


Fig. 9. The variance of residence time (σ^2) as a function of $CaCO_3$ concentration (screw speed of 40 rpm, feed rate of 2.1 kg/h).

of screw speed on the relative attenuation curve at the feed rate of 2.1 kg/h is given in Figs. 10(a) and 10(b). As expected, all the RTD curves shift to lower time values when screw speed is increased. Their mean residence time and variances were slightly decreased as the screw speed was increased (See Table 1). The mean residence time for 2nd screw configuration exhibited generally higher values than that of 1st screw configuration. On the other hand, the variance, which exhibits the degree of spread, did not show specific difference between two screw configurations. The cumulative residence time distribution function $F(t)$ curves for 1st screw configuration is presented in Fig. 11(a). To analyze flow behavior for the result of Fig. 11(a), $F(t)$ was described as a function of a dimensionless time as shown in Fig. 11(b). $F(\theta)$ for 2nd screw configuration was also described in Fig. 12. The flow in extruder shows behavior between mixed flow and plug flow. As shown in results, the changes in screw speed affected the flow pattern in the extruder. The flow slightly approached the mixed flow as increasing the screw speed. This may be due to the



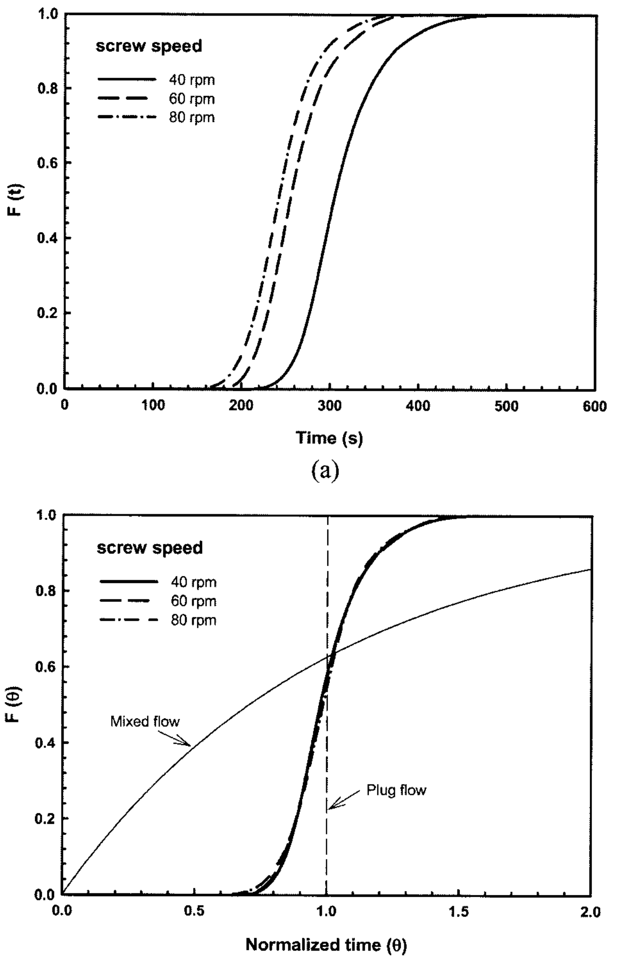
(a)

(b)

Fig. 10. Effect of the screw speed on RTD curves (feed rate of 2.1 kg/h, the precompounded tracer containing $CaCO_3$ 6 wt%).
(a) 1st screw configuration, (b) 2nd screw configuration

Table 1. The mean residence time ($\bar{t}_m$) and the variance of the residence time (σ^2) as a function of screw speed for two screw configurations. (feed rate of 2.1 kg/h, the precompounded tracer containing $CaCO_3$ 6 wt%)

Screw	Screw speed (rpm)	$\bar{t}_m(s)$	$\sigma^2(s^2)$
1st configuration	40	312.1	1921.9
	60	260.1	1350.8
	80	245.2	1232.0
2nd configuration	40	337.1	1745.6
	60	295.6	1378.3
	80	251.6	1240.8



(a)

(b)

Fig. 11. RTD curves as a function of screw speed (1st screw configuration, feed rate of 2.1 kg/h, the precompounded tracer containing $CaCO_3$ 6 wt%).
(a) $F(t)$ curves, (b) $F(\theta)$ curves

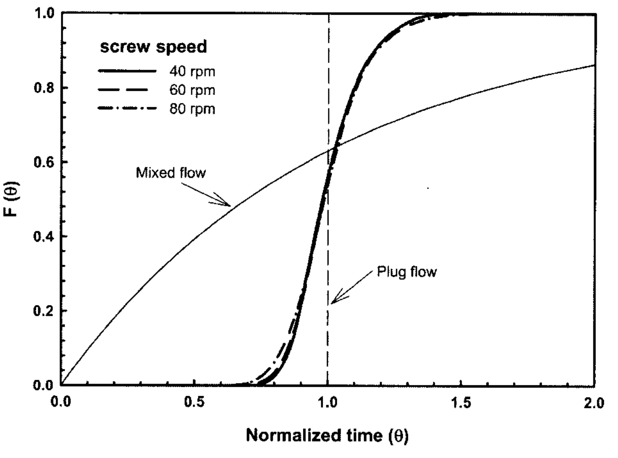
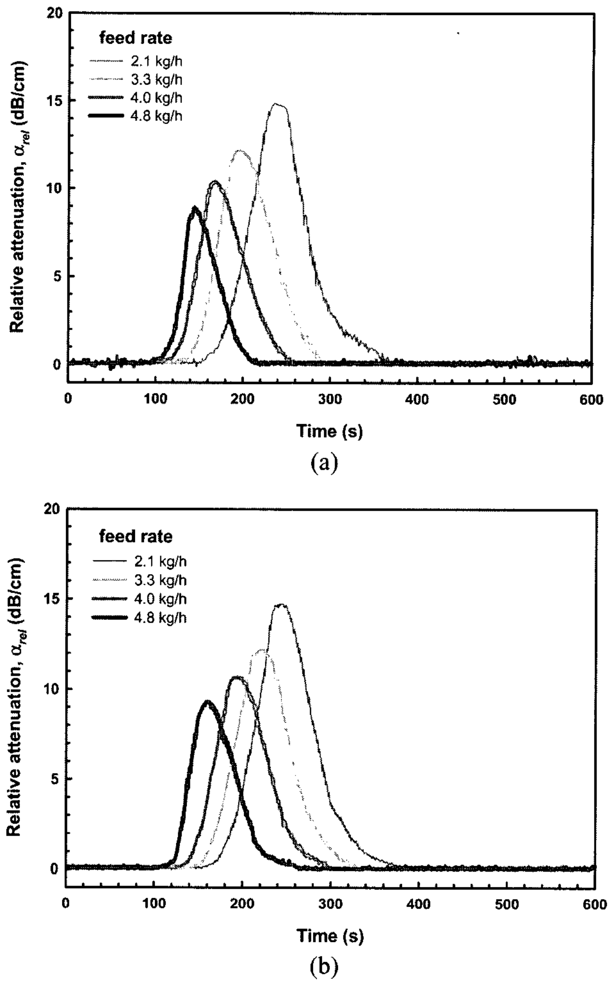


Fig. 12. $F(\theta)$ curves as a function of screw speed (2nd screw configuration, feed rate of 2.1 kg/h, the precompounded tracer containing $CaCO_3$ 6 wt%).



(a)

(b)

Fig. 13. RTD curves as a function of feed rate (screw speed of 80 rpm. the precompounded tracer containing $CaCO_3$ 6 wt%).
(a) 1st screw configuration, (b) 2nd screw configuration

increase in degree of filling in the barrel at low screw speed. Also, high screw speed may result in more slip, thus more backward leakage, resulting in more axial mixing. The effects of feed rates on the RTD curves for two screw configurations are given in Figs. 13(a) and 13(b). The increase of feeding rate shifted the relative attenuation curves to left, similar to the result for the screw speed effect. The level of relative attenuation was decreased with increasing feed rate. Figs. 14(a) and 14(b) show the $F(\theta)$ values of the RTD curves given in Figs. 13(a) and 13(b). As the feed rate increased, the curve slightly approached that of plug flow. This may be due to an increase in degree of fill of the barrel at high feed rate as discussed above. Increasing screw speed and decreasing feed rate cause a decrease in the fraction filled. The materials in the extruder with a low degree of fill could be less compacted than those in the extruder with a high degree of fill (Ainsworth *et al.*, 1997). This could cause more axial mixing in the extruder. As shown in Table 2, the mean residence time

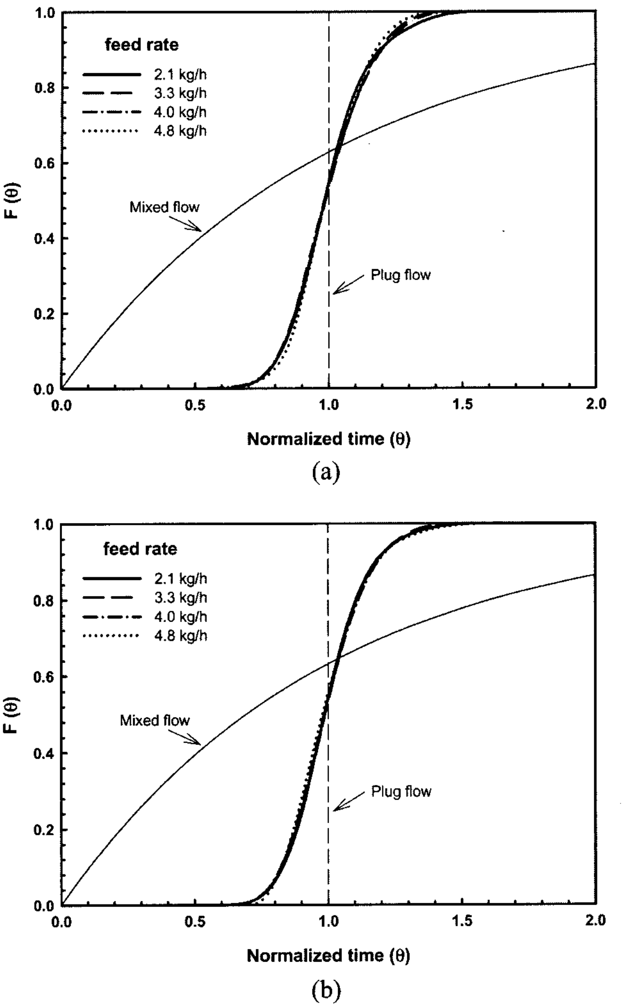


Fig. 14. $F(\theta)$ curves as a function of feed rate (screw speed of 80 rpm. the precompounded tracer containing $CaCO_3$ 6 wt%).
(a) 1st screw configuration, (b) 2nd screw configuration

Table 2. The mean residence time ($\bar{t}_m$) and the variance of the residence time (σ^2) as a function of feed rate for two screw configurations. (screw speed of 80 rpm, the precompounded tracer containing $CaCO_3$ 6 wt%)

Screw	Feed rate (kg/h)	$\bar{t}_m(s)$	$\sigma^2(s^2)$
1st configuration	2.1	245.2	1232.0
	3.3	205.6	858.6
	4.0	178.1	688.0
	4.8	153.2	414.8
2nd configuration	2.1	251.6	1240.8
	3.3	226.3	978.8
	4.0	202.1	832.7
	4.8	171.1	616.5

and the variance for two screw configurations were decreased as feed rate was increased. The mean residence time and the variance in 2nd screw configuration exhibited generally higher values compared to those in 1st screw configuration. This may be due to the existence of reverse screw elements in 2nd screw configuration.

6. Conclusions

In this study, we carried out RTD measurement in a co-rotating twin-screw extruder using a non-destructive ultrasonic monitoring system and used $CaCO_3$ as ultrasonic tracer. In-line determination of RTD by ultrasound during extrusion was successfully realized. RTD measurements were taken with various tracer concentrations, screw speeds, feed rates and screw configurations. Contrary to off-line RTD measurement, we could observe tail region of RTD curve, when RTD using ultrasonic system measured. The flow in the extruder approached plug flow as the feed rate increased, whereas an increase in the screw speed resulted in the flow approaching mixed flow. Increasing the screw speed or feed rate reduced the mean residence time ($\bar{t}_m$) and the variance of the residence time (σ^2), whereas increasing $CaCO_3$ concentration increased $\bar{t}_m$ and σ^2. The presence of reserve screw elements in the screw configuration also increased $\bar{t}_m$ and σ^2.

Acknowledgements

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Nomenclatures

A_1	: Amplitude of first echo
A_2	: Amplitude of second echo
$A_1(0)$	: First echo A_1 measured in the absence of tracer
$A_1(t)$	: First echo A_1 measured at time t after the injection of a pulse of tracer
C	: Tracer concentration
e	: Thickness of slit die [cm]
$E(t)$	: Residence time distribution
$F(t)$	: Cumulative residence time distribution function
$F(\theta)$	: Normalized cumulative residence time distribution
t	: Time [s]
$\bar{t}_m$	: Mean residence time [s]

Greek Letters

α	: Attenuation [dB/cm]
α_{rel}	: Relative attenuation [dB/cm]
σ^2	: Variance of residence time distribution [s^2]

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