NUMERICAL AND EXPERIMENTAL STUDY OF AGGLOMERATE DISPERSION IN POLYMER EXTRUSION

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Abstract: A model for agglomerate dispersion in screw extruders was developed and superimposed on the flow patterns as simulated using the FIDAP software. A particle tracking algorithm with an adaptive time step was used to follow the agglomerates trajectory. Along this flow path, the breakup probability was estimated using a Monte Carlo method and in conjunction with the local fragmentation number. Particle size distributions and Shannon entropy were computed along the screw channel. The results show good qualitative agreement between model predictions and experimental data.

Introduction

Extruders are required to process efficiently increasingly more complex polymeric systems, such as polymer blends and highly filled polymers containing various additives. Thus, among other process performance criteria, good mixing is required, both in terms of the distribution and the dispersion of the minor component in the polymeric matrix [1, 2].

This work presents a model for agglomerate dispersion that combines a Monte Carlo simulation with a mechanistic rupture/erosion model, superimposed on a particle tracking algorithm based on the fourth order Runge-Kutta method [3] with an adaptive time step and used in conjunction with the flow field profile. In turn, the flow filed profile was computed using the FIDAP software. Shannon entropy was used to quantify distributive mixing, while dispersive mixing was characterized using a mechanistic rupture and erosion model [4]. Given the encouraging results obtained, the same model was then applied to a plasticating extrusion software and the predictions were compared with experimental data.

Agglomerate Dispersion Model

Commercial solid additives/fillers are usually clustered into agglomerates, which are broken when the hydrodynamic forces are higher than their cohesive strength. Break-up can occur via erosion (detachment of indivisible small particles from the agglomerate surface) [5] or rupture (splitting into large fragments comprising several particles). The model of agglomerate dispersion involves a Monte Carlo method combined with a mechanistic rupture/erosion model. A breakup probability is computed in each interval Δt [4]:

$$P_{break} = \lambda . \Delta t \qquad (1)$$

The probability per unit time λ is proportional to the agglomerate surface. Assuming that each agglomerate comprises N particles, the surface of the agglomerate scales with $N^{2/3}$ [4]:

$$\lambda \propto N^{2/3} \qquad (2)$$

The time interval is always chosen so that $\lambda \Delta t << 1$. The Monte Carlo method generates a random number R_{nd} in the interval [0, 1]. If $R_{nd} > P_{break}$ the agglomerate remains intact. If $R_{nd} \leq P_{break}$ the agglomerate can either erode or rupture. The probability of breakage increases as the agglomerate residence time increases. Rupture and erosion depend on the local fragmentation number and on the agglomerate trajectory. The former is defined as:

$$Fa = \frac{\sigma_h}{\sigma_c} \qquad (3)$$

where σ_h and σ_c are the hydrodynamic stress induced by the flow and the agglomerate cohesive strength, respectively. Erosion takes place when Fa stays in the interval [2, 5[, while rupture occurs for $Fa \geq 5$ [6].

To quantify the agglomerates/particles distribution, an entropic measure based on the Shannon entropy was used [7, 8]. The latter is defined as:

$$S = -\sum_{j=1}^{M} p_j \, ln \, p_j \qquad (4)$$

where p_j is the probability of finding a particle in region (bin) indexed by j, and M is the total number of regions (bins) in which the system was divided. A normalized Shannon entropy is used by dividing S by $\ln(M)$.

Case Study

Numerical Simulations

Numerical simulations were performed using FIDAP software [9, 10], assuming a 3D Newtonian isothermal flow (at 190ºC the HDPE used has a viscosity of 3800 Pa.s [4]) and open discharge. For each simulation, 1000 silica agglomerates were positioned near the barrel at the entrance of the channel. Each agglomerate contains 100 indivisible particles clustered by a cohesive force of 1000 Pa [6]. The unwounded metering section of an extruder was considered as equivalent to a rectangular channel [11] with a cross-section of 24x5 mm and a length of 1200 mm, on top of which a surface slides at an angle (16.7º) and constant speed (equivalent to 20 and 60 rpm) in the down-channel direction.

A particle tracking algorithm with an adaptive time step was applied on the velocity profiles to follow the agglomerates trajectory. The agglomerates were taken as having no mass and being non-interacting. The Monte Carlo method was used at every 0.01 seconds on each agglomerate. The agglomerate break-up model was applied to those agglomerates that could potentially break [4].

The same method was applied on a 2D software that computes temperature, velocity and pressure profiles along a plasticating extruder [12], which takes into consideration solids conveying, melting and melt conveying stages. Calculations were performed using the geometry of an available prototype modular single screw extruder. The axial lengths of the feed, compression and metering sections (L_1, L_2 and L_3) are the same. The screw feed and metering sections had channel depths of 5 and 2 mm, respectively, and a helix angle of 16.7º. The entire screw channel has a length of 2610 mm. A total of 10.000 agglomerates (corresponding to a concentration of circa 1.2%) were uniformly distributed in the matrix as melting progresses.

Experimental Procedure

The single screw extruder described above was used in the experimental studies. The polymer system consisted of a high density polyethylene (grade T-100 from Repsol YPF, with MFI of 0.4g/10 min) filled with 2 w/w % of a HDPE-silica masterbatch (containing 40 w/w % of silica). The two components were pre-mixed in a tumble mixer and then fed to the hopper of the extruder.

The material was processed using a uniform barrel temperature of 190ºC and with the screws rotating at 20 or 60 rpm. Upon reaching the operating steady state, the screws were stopped, the die was disassembled and the screw extracted with the assistance of a hydraulic device. Figure 1 shows a typical material helix separated from the screw channel. Numbers indicate the helix number, from hopper to die. Thin films (10 μm thick) were cut from cross-sections of the helices at regular intervals and observed by optical microscopy (bright field technique) and image analysis. Figure 2 presents one example.

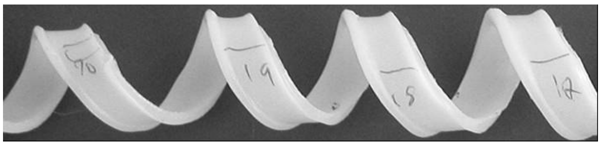

Figure 1 – Portion of the helix of material removed from the screw channel. The screw turns (#) are identified from hopper (#1) to screw tip (#30).

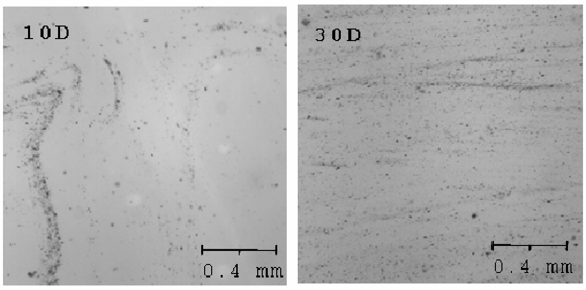

Figure 2 – Images of part of the cross-section of the screw channel at the beginning of the compression zone (left) and at the end of the metering zone (right), for a screw speed of 60 rpm.

Results and Discussion

When using the FIDAP software and considering the screw rotating at 20 and 60 rpm, 1000 agglomerates, each comprising 100 particles, were “injected” into the fluid. The various particulate species predicted to exist at the channel outlet are identified in Figure 3. The percentage of agglomerates and fragments is predominant by comparison with the primary/indivisible particles. Also, the model is sensitive to variations in screw speed, predicting, as anticipated, that a higher screw speed will promote break-up.

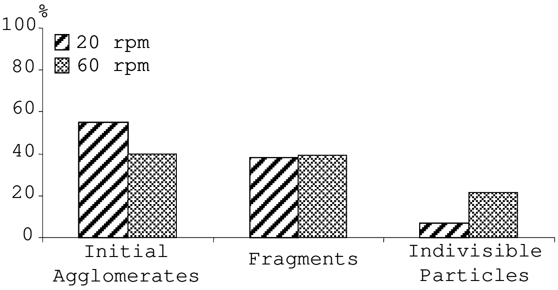

Figure 3 – Particulate species present at the outlet of the rectangular channel (flow computed with FIDAP software).

We have also applied the model to a plasticating extruder. In this case, circa 10000 agglomerates, each comprising of 100 particles were inserted in the matrix.

As shown in Figure 4, the percentage of fragments is predicted to be higher than that of indivisible particles.

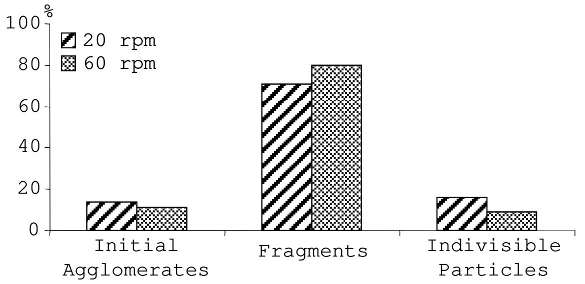

Figure 4 – Particulate species predicted to be present at the outlet of the plasticating extruder.

The corresponding experimental results (Figure 5) are quite encouraging in terms of the global relative percentage of particulate species. The effect of screw speed is also predicted correctly, except for the percentage of indivisible particles.

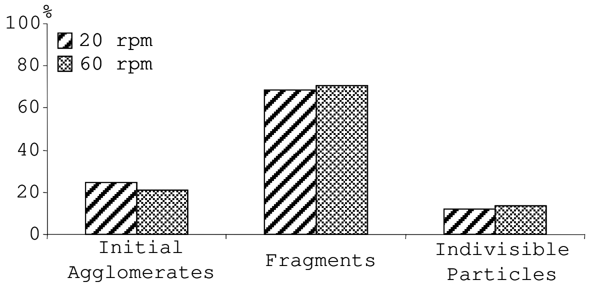

Figure 5 – Particulate species observed experimentally at the outlet of the plasticating extruder.

To quantify the agglomerate distribution in each channel cross-section, from the melting onset to the screw tip, the Shannon entropy was applied directly to the micrographs obtained with optical microscopy (Figure 6). Each image was divided into 50000 small regions (bins). As expected, Shannon entropy increases along the screw, and reaches higher values for 60 rpm.

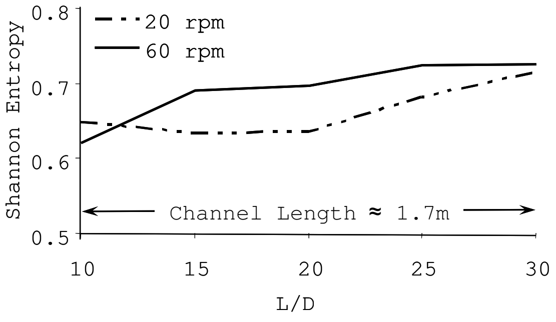

Figure 6 – Shannon entropy along the screw channel.

The dispersive mixing was quantified using the equation:

$$mix_{disp} = \frac{\sum_{j}^{N}\left[\left(\frac{d}{d_i}\right)_j \times \left(1-\frac{d}{d_i}\right)_j\right]}{\sum_{j}^{N}\left(\frac{d}{d_i}\right)_j} \qquad (1)$$

where d and d_i are the current and the original size of the agglomerates. This index reflects the reduction in agglomerates/particles size and ranges from 0 to 1. As expected, dispersion progresses along the screw, particularly in the compression zone (from 10 to 20 L/D), due to the increasing shear rate.

The effect of a higher screw speed is visible towards the end of the compression zone, again because the average shear rate becomes higher.

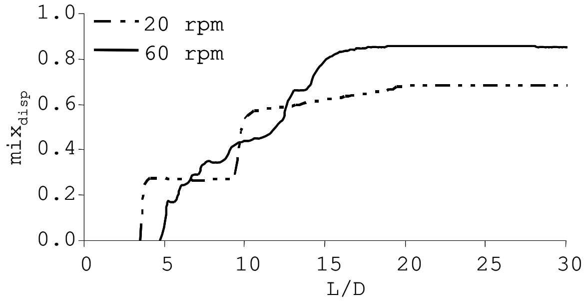

Figure 7 – Influence of screw speed on the intensity of dispersive mixing.

Conclusions

A model to predict agglomerate dispersion in a plasticating extruder was developed. It combines a Monte Carlo algorithm with a model of agglomerate rupture and erosion superimposed on simulated flow profiles. The model was implemented on the unwinded metering section of a single screw extruder as well as in conjunction with the simulated flow profiles in a plasticating extruder. The predicted results were compared with experimental data showing good qualitative agreement.

Acknowledgements

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