

Morphology Development of Immiscible Polymer Blends during Melting in Single-Screw Extruders: Effect of Composition and Compatibilization

S. M. Cunha^{a,*}, A. Gaspar-Cunha^a, J. A. Covas^a

^a I3N/IPC, (Institute for Polymers and Composites, Department of Polymer Engineering, University of Minho),
Campus de Azurém, Guimarães, Portugal

*Corresponding author: denebe@dep.uminho.pt

Abstract. Melting in single screw extruders began to be studied in the fifties, based on the pioneering work of Maddock. Most theoretical and experimental studies used homopolymers as model systems. However, in practice, there has been a considerable evolution in terms of the complexity of the materials being extruded. In the case of polymer blends, the morphology developed during melting should determine the final blend properties. Therefore, this work aims at investigating the morphology evolution during the melting stage of immiscible physical and chemically compatibilized PA6/PP blends. In general, the sequence of steps of morphology evolution reported for twin screw extruders and batch mixers was observed, though adapted to the flow kinematics along a helical single screw channel. The global morphological development is not affected by blend composition, but distinct domains seem to be formed when *in situ* reactive compatibilization takes place.

Introduction

The current understanding of the melting stage in single screw extruders results from pioneering research efforts that were initiated in the fifties and continued for more than thirty years on relatively simple polymer systems, such as homopolymers [1] or copolymers [2]. Nowadays, these machines can process complex systems, such as polymer blends, that were either previously prepared in a compounding stage, or that are processed directly into final products, such as monofilaments, tapes and raffia. Many authors have shown that the melting stage is crucial to morphology development and, consequentially, to the final properties of polymer blends. A morphology evolution mechanism for the initial polymer blending stages was proposed by Scott and Macosko for batch mixers [3] and confirmed by Sundararaj and Macosko for twin-screw extruders [4]. However, and despite some efforts (for example, by Gosh and Tyagi (2002) [5]), the morphology evolution mechanism and the importance of melting stage on final blend

morphology in the case of single-screw extruders, is still not well understood.

This work reports a study of the morphology development in a prototype modular single screw extruder, during the melting stage of immiscible and compatibilized PA6/PP blends with varying compositions. The progression of melting is analyzed at various length scales, from the existence of solid or melted material, down to the actual morphological characteristics at micrometer level, which is the topic of the present work.

Experimental

Materials

Polyamide 6 / Polypropylene (immiscible physical blends, PA6/PP and *in situ* compatibilized with PP/PP-g-MA) were chosen as model systems for this study. The polymer components are identified in Table 1. The viscosity ratio between PA6 and PP is near one in the shear rate range of interest.

Table 1. Raw Materials

Material	Trade name	Supplier	MFI (2.16 kg, 230°C) g/10min	T _m (°C)
PP	ISPLEN 030 G1E	Repsol	5,48	166
PP-g-MA	OREVAC CA 100	Arkema	292,30	167
PA6	Akulon F 130	DSM	8.46	220

Equipment

The blends were prepared in a prototype modular single screw extruder, with diameter $D = 30$ mm, $L/D = 30$, equipped with material sampling devices and pressure transducers along the barrel. The screw can be removed quickly by means of an hydraulic engine, in order to perform Maddock-type experiments. The screw profile used in the experiments below had a compression ratio of 2.5.

Procedure

The PA6 was dried overnight at 70°C. The polymers were pre-mixed in a drum mixer for 10 minute before processing. The experiments involved the following steps:

- reach operating steady state;
- record pressure and melt temperatures along the extruder;
- stop screw rotation, switch-off the heating system and remove quickly the die;
- extract the screw from the barrel;
- cool the polymer with compressed air;
- identify screw turns in the polymer helix;
- remove the polymer from the screw and freeze it in liquid nitrogen;
- obtain cross-sections at regular down-channel intervals.

The morphology evolution was observed by SEM, following the procedure presented in Figure 1.

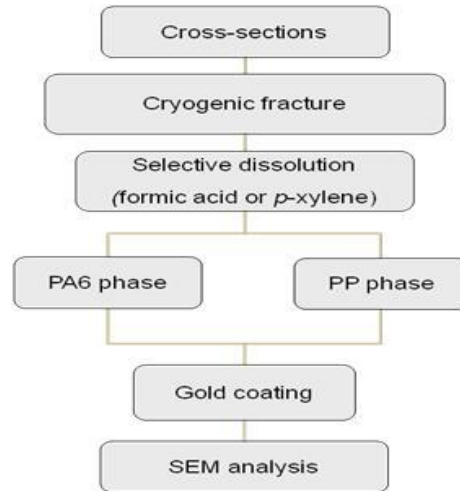


Fig. 1. Procedure for sample characterization.

Results and Discussion

The evolution of polymer blends morphology during melting is complex, as the components have different melting rates and exhibit distinct rheological responses to the same thermomechanical environment. As an example, Figure 2 shows the morphology of a cross-section of PA6/PP (80/20 wt.%) at screw turn 7, after extraction of PA6. The various morphological structures that are usually reported in literature for batch mixers and twin-screw extruders [3,4] are perceived here.

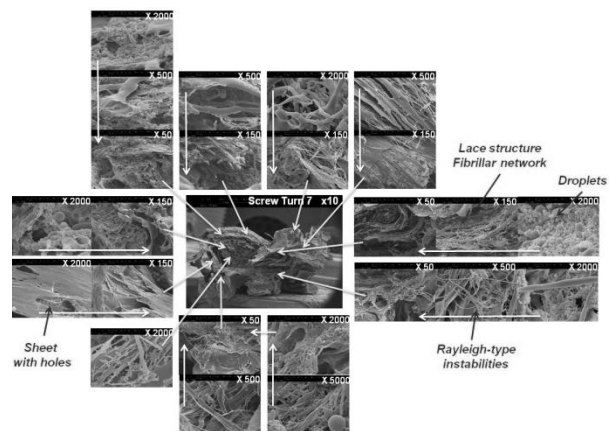


Fig. 2. Morphology of PA6/PP 80/20 wt.% at screw turn 7, after PA6 extraction.

Therefore, this means that in single screw extrusion these features co-exist, while for other processing techniques they usually develop sequentially.

In fact, at any cross-section, the morphological evolution is location dependent, as the flow kinematics and the heat transfer dictate differences in local residence times, temperatures and type and intensity of the thermomechanical stresses. As illustrated in Figure 3, the entire morphological sequence from sheets and ribbons to droplets of the dispersed phase reported by Scott and Macosko (1991) can be observed in one single micrograph.

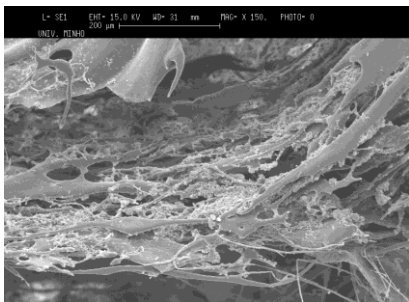


Fig. 3. Micrograph of a location in screw turn 7 showing the the morphology evolution from sheets to droplets (PA6/PP 80/20 wt.%).

The general morphology evolution mechanism is independent of blend composition. As seen in Figure 4 for the PA6/PP 50/50 and 80/20 w/w blends, the same final morphological entities are present, even if with different coarseness.

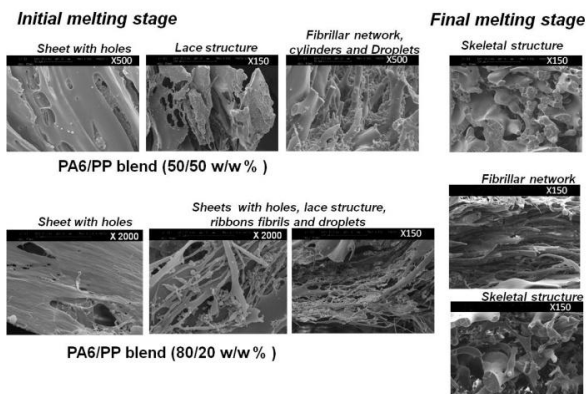


Fig. 4. Morphology of the PP phase during melting of the PA6/PP 50/50 and 80/20 wt.%.

During *in-situ* compatibilization, the morphology development during the early melting stages involves the same type of structures observed for the equivalent physical blends, even though new structure types seem to be detected. These new domains (Figure 5) are possibly related with the formation of the copolymer at the interface. In fact, the copolymer phase is resistant to both solvents used for selective dissolution.

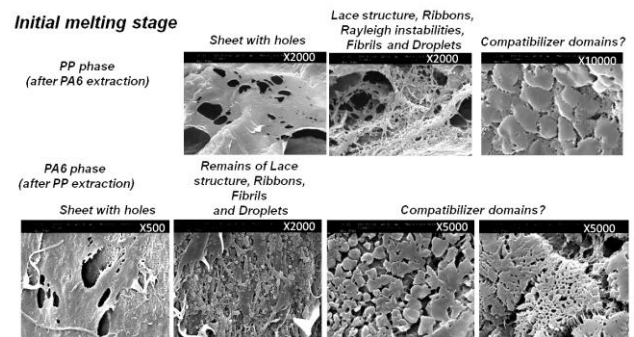


Fig. 5. Morphology of the PP and PA6 phases during melting of the PA6/PP/PP-g-MA 80/5/15 wt.% blend.

Conclusions

Physical and chemically compatibilized polymer blends prepared by single-screw extrusion exhibit the same morphology development mechanism reported for twin screw extruders and batch mixers during the melting stage. Contrarily to these machines though, the morphology development rate in single screw extruders depends on the location on the channel cross-section, due to the particularities of the flow kinematics. The blend composition does not affect the above mechanism, but determines the coarseness of the morphology entities. The same applies to compatibilization, where new structures - possibly related to the formation of a copolymer - were detected.

Acknowledgments

The authors are grateful to Portuguese Fundação para a Ciência e Tecnologia for supporting this work under grant SFRH/BD/19997/2004, to DSM, the Netherlands, for supplying PA6 and ARKEMA, Spain, for supplying PP-g-MA.

References

1. Z. Tadmor and C. Gogos, *Principle of Polymer Processing*, p. 178, Wiley Interscience, New York, (1979).
2. W. -Lu, S. H. Bumm, J. L. White, *Inter. Polymer Processing*, XXIII, 72 (2008).
3. C.E. Scott, C.W. Macosko, *Polymer Bulletin*, **26**, 341 (1991).
4. U. Sundararaj, C.W. Macosko, *Polym. Eng. Sci.*, **32**,1804 (1992).
5. A. K. Gosh, S. Tyagi, *Polym. Eng. Sci.*, **42**, 1309 (2002).