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Doctoral Dissertation

Doctoral Program in Materials Science and Technology (38th Cycle)

PhD Candidate: Fulvia Cravero

Supervisor: Prof. Rossella Arrigo

Co- Supervisor: Prof. Frache Alberto

Title: Processing-driven structuring of melt compounded polymer-based systems

Summary

Polymer structuring is a valuable cost- and time-saving method for producing high-performance polymer-based materials. The main issue in the diffusion of this approach is the understanding of the relationship between the processing parameters, the microstructure and the resulting properties, which is crucial for the actual definition of the final features.

Therefore, the aim of this PhD dissertation is the study of the processing parameter-microstructure-property relationships in polymer blends-based materials with the purpose of obtaining structured morphologies that can be opportunely tuned in the production step. In particular, after a detailed analysis of the state of the art and considering its importance at an industrial level, it was decided to investigate the solvent-free production of thermoplastic polymer blends by melt blending approach. Specifically, two homopolymer systems were selected. Namely, the first one is based on high-density polyethylene (HDPE) and the other one on polyamide 6 (PA6). With regard to the experimental trial, the role of both the material characteristics and the processing parameters was investigated. Specifically, the polarity of the polymer was

considered along with the mono- or bimodality of the molecular weight (MW) in the blend. Various bimodal distributions were also investigated. From a processing point of view, both the HDPE and PA6 blends were obtained by corotating twin screw extrusion. Therefore, the effect of varying the screw rotation speed and the barrel temperature was evaluated.

Furthermore, the second step of the research focused on the role of the application of a non-isothermal uniaxial elongational flow at the exit of the die. In the case of PA6-based materials, the investigation was firstly performed on the polymeric systems. Then, the evaluation of the morphology of composites containing hydrotalcites (LDHs) characterized by two different surface areas were studied. Thermal, rheological, morphological and mechanical investigations were performed and are reported and discussed in detail.

In addition, the innovative approach adopted in this PhD dissertation refers to the multivariate analysis employed for the interpretation of the results. In fact, this approach has allowed to highlight the impact of the simultaneous variation of two parameters on the resulting morphology, thus considering the role of the combination of two variables in the processing parameter-microstructure-property relationships and enriching the current knowledge available in the literature, which is mainly focused on the univariate analysis. Furthermore, the multivariate approach allows to obtain solutions that are often not obtained by considering the variation of a single parameter alone. This has been appreciated, for instance, in the study of the twin-screw extruded HDPE blends, where the full polymer miscibility was achieved when the highest values of shear stress and processing temperatures were simultaneously exploited. Also, from the analysis of the rheological characterization, the minor role of the screw speed and barrel temperature over the molecular weight was appreciated. In addition, the concurrent increase of the screw rotation speed and MW in PA6-based materials was found to promote the influence of the flow-induced crystallization (FIC) over the memory effect, resulting in the greater increase in final crystallinity and α/γ phase content when the high MW was considered instead of the low molecular weight.

Another novelty of the present PhD research is the extensive use of a simulation software to obtain information about the thermo-mechanical field inside the barrel during compounding. In particular, data on the actual shear rate, temperature and residence time were used to interpret the characterization results according to the processing parameters.