# POLYMER BLENDS WITH MICROFIBRILLAR-PHASE MORPHOLOGY

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## ABSTRACT

The work focuses on the possibilities of microfibrillar-phase formation in polymer blends during extrusion through a die with semihyperbolic-converging channel. For these purposes, three polyethylene/polypropylene blends with the mixing ratios (wt./wt.) 80/20, 70/30 and 60/40 were prepared. Final structure and properties of extruded blends were examined using electron microscopy, wide-angle X-ray scattering and differential scanning calorimetry. It is demonstrated that the formation of microfibrillar-phase morphology is achieved in all the blends, independent of their mixing ratios. However, the structure and properties of these microfibrillar-phase composites are strongly influenced by mixing parameters.

Keywords: in-situ composite, polyethylene, polypropylene

### **1. INTRODUCTION**

Technical and financial incentives make polymeric materials an attractive alternative in an everincreasing number of applications. The development of new resins, which, for several decades, attracted most of research efforts, now tends to be supplemented by new approaches. The strength of these new tendencies is clearly shown by the rapid emergence of polymer blends and composites as engineering materials.

Noteworthy, one of the greatest research interests in blending field was dedicated to studies of properties and morphology of polyolefin blends. This was not only due to wide applicability of these materials in industry but rather due to their rich and fascinated morphology depending on molecular structure, thermal history, and external stress field. Nowadays when plastic waste recycling is one of the main issues of environmental concerns, studies of polypropylene and polyethylene blends play important role, due to their consumed amount and difficult separation of these materials apart.

Polymers compounding with special morphology governed by processing conditions, commonly known as in-situ composites preparation, offers extra possibilities of improvement of blend properties. Relatively successful in-situ composites preparation was achieved via blending of common thermoplastic resins with liquid crystalline polymers. Liquid crystalline polymers represent special macromolecular material containing rigid mesogenic units that in molten state keep defined conformation. During processing these rigid domains can be oriented along the flow which results in a fine fibril structure reinforcing the thermoplastic matrix. Besides this approach, in-situ composites were effectively prepared also from common polymeric materials several times cheaper than liquid crystalline polymers. One of such example is the combination of polypropylene with polyamide 6. Nevertheless, only a few works dealt with improvement of mechanical properties of polyethylene by the polypropylene microfibrillar-phase reinforcement.

Present work focuses on in-situ composites prepared by blending of linear high-density polyethylene with isotactic polypropylene using extrusion die with semihyperbolic-converging channel. Beside proper preparation of in-situ composites, interrelations between material composition, processing conditions and resulting structure and properties of polyethylene-polypropylene blends are studied.

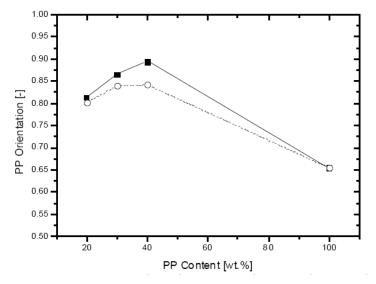
#### **2. EXPERIMENTAL**

High-density polyethylene LITEN TB 38 and isotactic polypropylene MOSTEN GB 003, both produced by Chemopetrol Litvínov, Czech Republic, were chosen as starting materials in this study. PP/PE blends were prepared using counter-rotating twin-screw extruder at screw speed 35 rpm. The temperature of extruder ranged from 190 °C to 210 °C. The extruded blends with 20, 30 and 40 wt.% of PP were quenched and granulated.

One of the ways to prepare blend of incompatible polymer pair in which the dispersed phase forms in-situ reinforced fibres is utilisation of extrusion line equipped with converging die. The whole extrusion line used in this study consisted of <sup>(i)</sup> a Brabender measuring single-screw extruder 30/25 D, <sup>(ii)</sup> a Zenith PEP II gear pump 1.2 cm /rev and <sup>(iii)</sup> a flat extrusion die with 2x20 mm outlet cross-section. The polymer melt passing through die was firstly accelerated in semi-hyperbolic convergent section and subsequently cooled in calibration section. Final extrusion rate was governed by gear pump (20 rpm). Temperatures of extruder (150, 170, 190, 210 °C), gear pump (200 °C), and die sections (185 °C in cylinder, 170 °C in converging and calibration sections) were constant throughout the experiment. For determination of the effects of compounding on resulting morphology and properties, extrudates were prepared from both compounded blends and hand-mixed polymers.

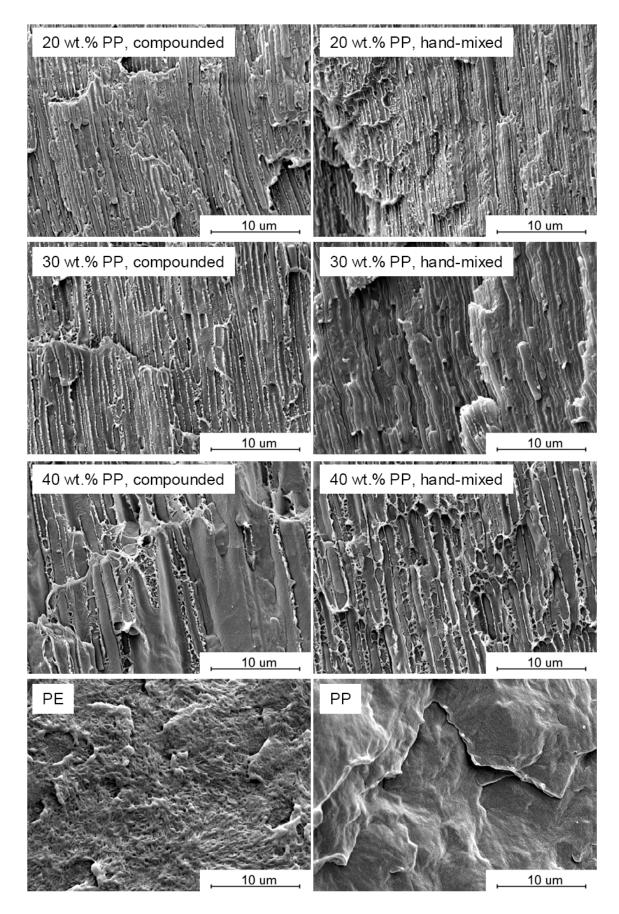
#### **3. RESULTS AND DISCUSSION**

Wide-angle X-ray scattering was applied to determine degree of orientation of the specimens. Figure 1 display orientation of PP in extruded tapes as a function of PP content. Generally, the orientation in blends is significantly higher compared to pure PP extrudates, which indicates the formation of oriented PP fibrils in PE matrix upon extrusion. The PP orientation increases with rising amount of PP in blends. However, this tendency is more pronounced in compounded blends reflecting a higher lever of mutual phase interaction, compared to that of hand-mixed blends.



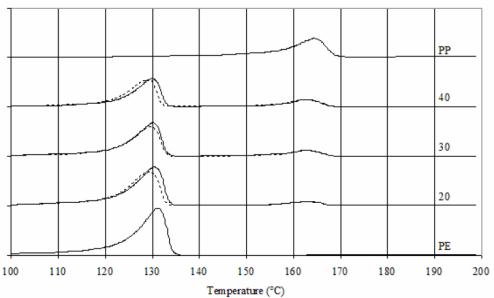
*Figure 1. Orientation of PP phase in extrudates as a function of PP content;* — **–** *compounded blends, ---o--- hand-mixed blends.* 

Structure of crack surfaces of extruded tapes was examined by scanning electron microscopy. The scanned micrographs are shown in Figure 2. While the micrographs of pure extrudates show isotropic structure, the morphology of the tapes extruded from blends is oriented along the flow direction. This is important result and the first prerequisite to rate these blends as microfibrillar-phase composites. As can be seen, the structure of compounded-blend extrudates gradually changes from fine-fibril morphology to coarse co-continuous phase structure, in respect of the PP content increase. On the other hand, this structure gradation is not clearly shown in the extrudates prepared from hand-mixed materials. Although the transition between fibrillar and co-continuous morphology can be recognized, the diameter of PP domains is not directly controlled by the PP content.



*Figure 2. Structure of extrudates prepared from pure materials and compounded and hand-mixed blends.* 

Differential scanning calorimetry was employed for the measurements of thermal properties of extrudates. As can be seen in Figure 3, shapes of the melting peaks vary with the composition of the blends. In comparison with the thermal scans of hand-mixed blend extrudates, the melting peaks of individual phases in compounded blends are slightly shifted towards themselves. This fact indicates a limited miscibility at interfaces and subsequent co-crystallization occurring in these blends during extrusion. Consequently, this co-crystallization of both components can increase dramatically interface cohesion which can results in improvement of mechanical properties. The heats of fusion (i.e. the areas under melting peaks) of PP and PE components in compounded blends show slightly higher values than those of hand mixed blends, which reflects generally higher level of macromolecular order resulting from enhanced phase mixing upon twin-screw extruder blending.



*Figure 3. Thermal scans of tapes extruded from pure and blended polymers. Solid line represents samples prepared from compounded blends and dashed line is used for hand-mixed blends.* 

## 4. CONCLUSIONS

The aim of the present work was preparation of microfibrillar-phase morphology in the polypropylene/polyethylene blends with polypropylene as a minority component. For these purposes an extrusion line based on conventional single-screw extruder and semihyperbolic-converging die was employed. It was proved that the formation of fibrillar structure in one processing step can be achieved using <sup>(i)</sup> blends with various content of polypropylene (the blends with 20, 30 and 40 wt.% of PP were studied) and <sup>(ii)</sup> compounded and hand-mixed materials (both twin-screw extruder compounded and non-compounded materials were processed).

The extruded tapes with microfibrillar morphology were examined by several methods of structure analysis and measurements of properties. The results can be summarized as follows: <sup>(i)</sup> Wide-angle X-ray scattering showed a high orientation of PP phase in blends. The degree of orientation increases with rising amount of PP. <sup>(ii)</sup> The structure of extruded blends observed using electron microscopy gradually changes from fibril morphology to co-continuous phase structure, in respect of the PP content increase. In the compounded-blend extrudates, the aspect ratio of PP domains decreases with the rising PP amount. <sup>(iii)</sup> Differential scanning calorimetry revealed a slight shift of the melting peaks of individual phases towards themselves in compounded blends, which indicates a limited miscibility at interfaces and subsequent co-crystallization during extrusion.

#### **5. ACKNOWLEDGEMENT**

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