Flow Behaviour of Gas-Containing LDPE/i-PP Melts

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Summary: An experimental study was conducted to investigate the rheological behaviour and extrudate swell of polyolefin blends based on two grades of low-density polyethylene (LDPE) and an isotactic polypropylene (i-PP). Blending was carried out on a twin-screw extruder "Brabender" at different composition ratios in the temperature range from 140 to 190 °C. The LDPE/i-PP blends mixed with 0.5 wt. % blowing agent were extruded by means of "Brabender" extrusiograph at melt temperature of 200 °C and different extrusion rates.
The influence of composition content on the viscosity and extrudate swell was considered. The foam structure and morphology are discussed in terms of shear rate, molecular characteristics and composition content. The presence of layered structure was observed: an outer smectic layer and an inner partially crystalline layer. The thickness of smectic layer and size of spherulites were determined.

Introduction

Whereas extensive studies have been reported in the literature dealing with the rheological properties of polymer blends,[1-3] little has been reported about the flow behaviour of gas-containing polymer blends. Polymer blends have been recently used for producing microcellular foams. The studies have shown that the morphology and crystallinity of semicrystalline polymers have a great influence on the solubility and diffusitivity of the blowing agent and on the cellular structure. [4]

The aim of this study is to investigate the rheological properties of gas-containing LDPE/PP blends and their influence on the macrostructure and morphology.

Experimental

The polymers used were two grades of LDPE: "Ropoten" FB-7-104 [MFI (190/2.16) = 5.8 g/10 min, M_W/M_n = 20] and "Ropoten" OV-3-110 [MFI (190/2.16) = 0.2 g/10 min, M_W/M_n = 29], and an i-PP: "Buplen" 6631 [MFI (230/2.16) = 1.1 g/10 min,

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M_w/M_n = 6.8], Bulgarian products. Blowing agent used was azodicarbonamide "Genitron" AC-4 (Fisons Ltd.). Blends from LDPE and i-PP were prepared using a twin screw extruder "Brabender" DSE 35/17D at different composition ratios of 0/100; 25/75; 50/50; 75/25 and 100/0 in the temperature range from 140 to 190 °C.
The LDPE/i-PP compositions mixed with 0,5 wt. % blowing agent were extruded by means of "Brabender" extrusiograph 30/25D trough cylindrical die with diameter of 1.5 mm and length-to-diameter ratio (L/D) 16. The experiments were carried out at melt temperature T_m of 200 °C and at extrusion rate in the range from 0.1047 to 10.47 rad/s. The mass throughput rate and die pressure drop were measured for each extrusion rate. From the viscosity plot for gas-containing blends[5] we obtained the concentration dependence of viscosity at constant shear rate.
Extrudate swell B is defined as the ratio between the extrudate diameter D_e and die diameter D. Extrudate diameters were measured on frozen samples by micrometer with 5.10^{-3} cm resolution.
Microtomed cross-sections (40 μk) taken from the extrudate were investigated by means of optical microscopy with small magnification (x20). The bubble morphology (size, shape, and size distribution) was determined. Morphology was investigated by optical microscopy with polarized light at 200-fold magnification (x200).

Results

Concentration dependence of viscosity

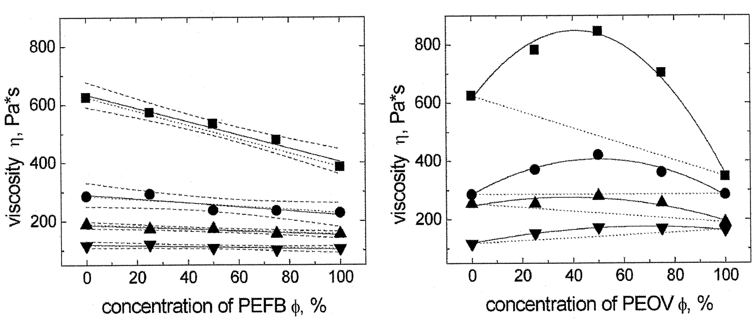

Figure 1. Viscosity variation with PE content in the blends: *left* - PEFB/PP and *right* - PEOV/PP at different shear rates: ■ – 100 s^{-1}, ● – 300 s^{-1}, ▲ – 500 s^{-1}, ▼ – 1000 s^{-1}.

Figure 1 shows the concentration dependence of viscosity of gas-containing blends at different shear rates. LDPE/i-PP blends behave either as mono-phase or bi-phase system in dependence on the flowability of the type of LDPE. The viscosity vs. concentration dependence was found to be additive for the blends with LDPE FB-7-104. The relationship shows positive quadratic deviation from linearity for the blends with LDPE OV-3-110.

Extrudate swell

The extrudate swell of foamed blends is of great importance in the shape and quality control of extruded products. The relationship between processing conditions, material variables, and extrudate swell is complex because the extrudate swell is associated with the elastic property of the polymer melt and with the expansion of gas-containing melt. It was found that the extrudate swell decreases with increasing PE content in the blend, this effect more strongly expressed for PEOV as it is less elastic than PEFB. The extrudate swell correlates inversely with the foam density in a certain range of shear rate (Figure 2). The deviations observed are due to the expansion in the flow channel at low shear rates or to the coalescence of gas bubbles before solidification of extrudate at high shear rates. In both cases the results are an increase of density and poor foam structure.

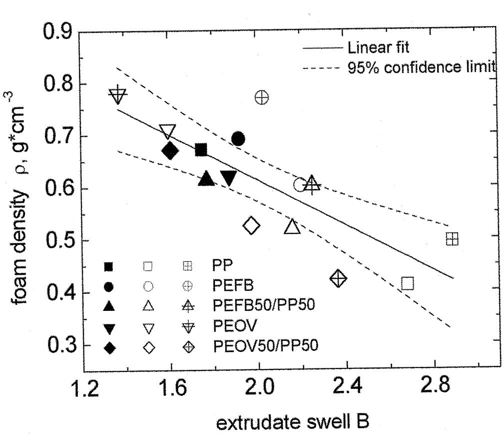

Figure 2. Foam density vs. extrudate swell for gas-containing polyolefin blends at different shear rates: a/ closed symbols – 100 s^{-1}; b/ open symbols – 500 s^{-1} ; c/ crossed symbols – 800 s^{-1}.

Figure 3 shows a representative cross-section for the PEOV50/PP50 blend extruded at different shear rates. It can be seen that bubble size decreases with increasing of shear rate.

Morphology

The presence of a two-layered structure was observed: an outer smectic layer and an inner partially crystalline layer. The thickness of the smectic layer decreases with increasing PE contents in the blends and decreasing the shear rate. PP forms spherulitic crystals of different modifications in the blends. A hexagonal β-PP-phase was observed along with its predominant monoclinic α-phase. β-PP spherulites are brighter in cross-polarized light and bigger in size then α-PP crystals. The contents of β-PP phase decreases with increasing PE in blends and with decreasing of the shear rate The size of the α-PP spherulites also depends on these two factors – blend composition and shear rate. Increasing the PE content and shear rate causes a reduction in α-PP spherulite size, as shown on Figure 4.

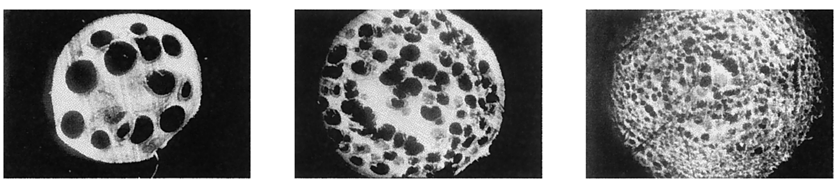

Figure 3. Photomicrographs of PEOV50/PP50 extrudate cross-section at different shear rates: *left* – 100 s^{-1}; *centre* – 500 s^{-1}; *right* – 800 s^{-1}.

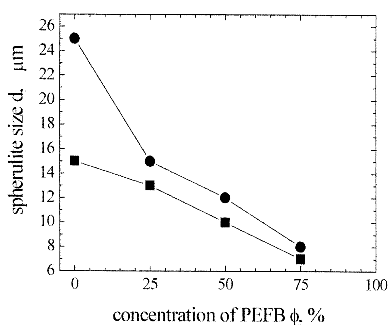

Figure 4. Spherulite size of PP vs. contents of PEFB at different shear rates:
● – 100 s^{-1}; ■ – 500 s^{-1}.

Conclusion

The investigations confirm that the rheological properties of blends greatly affect the foam structure. Melt viscosity is the key for controlling the bubble growth inside the die and melt elasticity is crucial for controlling the extrudate swell. The foam structure and morphology are discussed in terms of extrusion conditions and composition contents. The observations show a strong connection between extrusion rate, composition content, and some macro- and microstructural parameters (density, bubble size, thickness of smectic layer, spherulite size). A correlation was established between foam density and extrudate swell at a certain range of shear rate.

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