Mechanical properties of epitaxially crystallized 1,4-polybutadiene on uniaxially oriented isotactic polypropylene

J. Petermann and Y. Xu

Polymer and Composites Group, Technical University Hamburg Harburg, Hamburg, FRG

Abstract: The structure and mechanical properties of polypropylene (PP)-trans-1,4-polybutadiene (PBD) blends were investigated. It was found that PBD can epitaxially crystallize on PP. An increase in mechanical properties (Young's modulus, fracture stress) is observed, despite the fact that pure PBD has much worse mechanical properties compared to PP. The epitaxial morphology is responsible for the synergetic effects in the mechanical properties.

Key words: Polypropylene; trans-1,4-polybutadiene; blend; epitaxy; mechanical property.

1. Introduction

In recent years, the epitaxy between PP and some polymers with zig-zag chains, such as polyethylene, polyoctenamer and nylons, has been a subject of interest [1-4]. In such epitaxies, the zig-zag chains of some polymers incline 50° to the PP-chain direction. This kind of epitaxy is explained by the alignment of the zig-zag chain along rows of methyl groups of PP with 5 Å intermolecular distances for a chain/row matching [2]. Some property improvements as a result of such an epitaxy were demonstrated: Young's modulus and fracture stress showed that a synergism [5] and improved adhesion also resulted [6]. PP-PBD blends attract our interest because PBD also has a zig-zag molecular conformation, but not in a plan, and the intermolecular distance is 4.6 Å apart when the temperature is lower than 76°C [7]. The mechanical properties of PP-PBD have not yet been studied in detail. In the present investigation, we have carried out some TEM, DSC, x-ray and mechanical tests on PP-PBD blends.

2. Experimental details

The materials used for the experiments were polypropylene, type Novolene from BASF AG, Ludwigshafen, FRG, and trans-1,4-polybutadiene (trans content 65%) from Asahi-Kasei Co. Ltd., Japan. The oriented films were prepared according to the method of Petermann and Gohil [8]: a small amount of a 0.5 % solution of the polymers with the appropriate concentration in xylene was poured on a hot glass slide, where the solvent was allowed to evaporate. The resulting thin polymer blend film was then picked up on a motor-driven cylinder at a wind-up speed of 20 cm/s. The resulting thin polymer films were about 80 nm thick and were highly oriented in the drawing direction. Single films were used for TEM investigations. This blending method with PP and the used PBD as components was only possible to a maximum concentration of 50 % PBD, because due to the low viscosity of the PBD melt, the film-drawing method for higher concentrations did not work. For mechanical, thermal, and x-ray measurements about 300 layers were wound up. Thin films of PBD were solution-cast from a 0.1 wt% solution in xylene on the surface of water. For layer films, the PBD film was put on top of PP substrates and the layer was subsequently heated to 90°C for 20 min, and then slowly cooled to room temperature. The materials were investigated using a Philips EM-400 T electron microscope operated at 100 KV. The tensile tests were performed in a Zwick 1445. The strain rate used was 10 mm/min for all measurements, the gauge length of the samples was 25 mm, and the deformation temperature was 20°C.

*) Dedicated to Prof. Dr. W. Pechhold on the occasion of his 60th birthday
The crystallinity and melting temperature of PBD were measured using a Heraeus DSC 500 differential scanning calorimeter and a Philips PW 1729 x-ray diffractometer.

3. Results and discussion

1. Morphology

Figure 1 shows the transmission electron micrograph of a PP substrate film and a PBD film prepared as described above. The PP film has a lamellar morphology and a high crystalline fiber texture, as can be estimated from the electron diffraction pattern (Fig. 1a). In the PBD film some lamellae with random orientation can be seen (Fig. 1b). The crystallinity and melting temperature of PBD measured from DSC and x-ray is about 12% and 80°C, respectively. The conformation of the PBD chain is zig-zag (TS'T'S'). Two crystalline forms are known to exist in PBD [7, 9-11]. One is stable at room temperature and has a monoclinic unit cell with \( a = 8.63 \, \text{Å}, b = 9.11 \, \text{Å}, c = 4.83 \, \text{Å} \) and \( \beta = 114^\circ \). At a temperature of 76°C a reversible crystal transformation occurs to a pseudo-hexagonal unit cell \( (a = 4.95 \, \text{Å}, c = 4.66 \, \text{Å}) \). In the high temperature form a considerable amount of rotational freedom around the molecular axes exists.

The microstructure of the PP-PBD layer, annealed at 90°C and slowly cooled to room temperature is shown in Fig. 2. A cross-hatched lamellar structure arises with the PBD lamellae being inclined about ±40° with respect to the PP c-axis. The above result...