PET/LDPE microfibrillar reinforced polymer–polymer composites

M. Evstatiev[1], O. Evstatiev[1], S. Fakirov[1], K. Friedrich[2], N. Stribeck[3], S. S. Funari[4]

[1] *Laboratory on Polymers, University of Sofia, 1126 Sofia, Bulgaria*
[2] *Inst. of Compos. Materials Ltd., University of Kaiserslautern, 67663 Kaiserslautern, Germany*
[3] *Inst. of Techn. and Macromol. Chemistry, University of Hamburg, 20146 Hamburg, Germany*
[4] *Max–Planck Institute of Colloids and Interfaces, c/o HASYLAB, 22603 Hamburg, Germany*

Microfibrillar reinforced composites (MFC) comprising an isotropic matrix from a lower melting polymer reinforced by microfibrils of a higher melting polymer were manufactured under industrially relevant conditions and processed via injection molding. Low density polyethylene (LDPE) (matrix) and recycled poly(ethylene terephthalate) (PET) from bottles were melt blended (in 30/70 and 50/50 PET/LDPE wt. ratio) and extruded, followed by continuous drawing, pelletizing and injection molding of dogbone samples. Samples of each stage of MFC manufacturing and processing were characterized by means of scanning electron microscopy (SEM), wide-angle X-ray scattering (WAXS), dynamic mechanical thermal analysis (DMTA), and mechanical testing. SEM and WAXS (cf. Figure 1) show that the extruded blend is isotropic but becomes highly oriented after drawing, being converted into a polymer-polymer composite upon injection molding at temperatures below the melting temperature of the PET. This MFC is characterized by an isotropic LDPE matrix reinforced by randomly distributed PET microfibrils, as concluded from the WAXS patterns and the SEM observations.

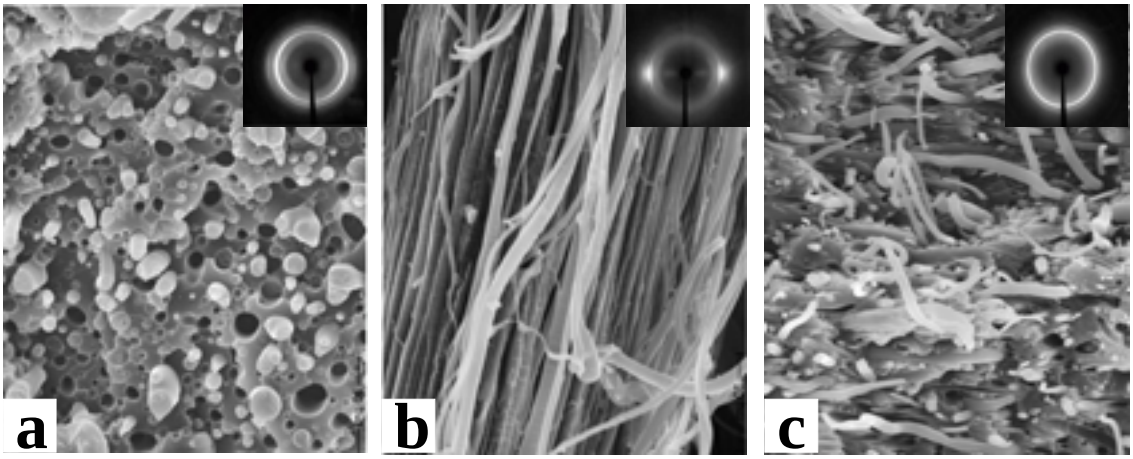

Figure 1: Morphological peculiarities according to SEM and WAXS (inserted patterns) during manufacturing (mixing with extrusion (a), and drawing (b)) as well as their processing by injection molding (c) for the blend PET(recycled)/LDPE

Materials were studied at HASYLAB, beamline A2 in several stages of the manufacturing process and as a function of strain during cold drawing. Figure 2 shows some of the SAXS scattering patterns.

The results differ considerably from those of a similar study where LDPE was replaced by HDPE [1, 2]. Here no nanostructure of PET is found in the SAXS diagrams. Because of the fact that SEM images show microfibrils from PET in every stage after extrusion, but crystalline reflections are missing in WAXS [3] we conclude that here PET is in the amorphous state.

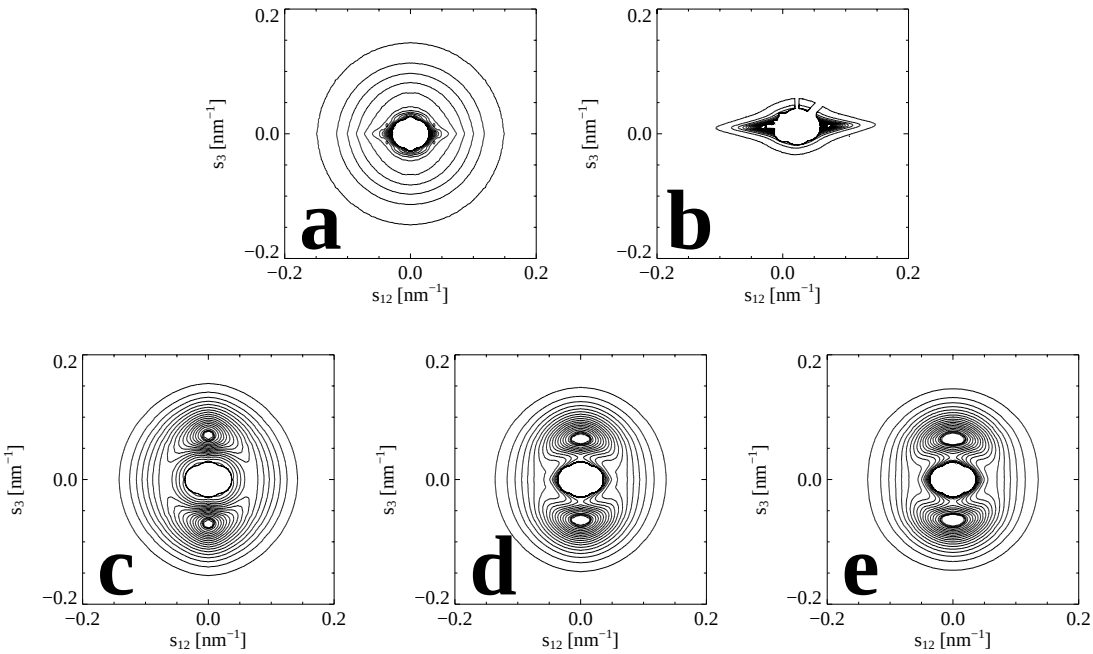

Figure 2: SAXS scattering patterns of PET/LDPE blends in various stages of the formation of a microfibrillar reinforced composite (MFC). (a) after blending and extrusion (b) after straining at 85°C to $\varepsilon \approx 350\%$ highly oriented microfibrils are formed (c) Pure LDPE, injection molded showing highly oriented stacks from semicrystalline LDPE lamellae (d) MFC PET/LDPE 30/70 (e) MFC PET/LDPE 50/50. A PET nanostructure is not seen in the MFCs

Acknowledgements. This investigation was supported by the Bilateral Cooperation Program between the University of Hamburg, Germany, and the University of Sofia, Bulgaria, which is funded by the DAAD. SAXS investigations were supported by HASYLAB, Hamburg, Project II–01–041. One of us (S.F.) expresses his warmest thanks to the Alexander von Humboldt Foundations for the "Humboldt Research Award", enabling the finalization of this study.

References

[1] S. Fakirov, O. Samokovlijsky, N. Stribeck, A. A. Apostolov, Z. Denchev, D. Sapoundjieva, M. Evstatiev, A. Meyer, M. Stamm, Macromolecules, 34, 3314 (2001)

[2] S. Fakirov, M. Evstatiev, K. Friedrich, "Nanostructured Polymer Composites from Polymer Blends: Morphology and Mechanical Properties" in: S. Fakirov (Ed.) "*Handbook of Thermoplastic Elastomers*", vol. 2, chap. 23, Wiley-VCH, Weinheim, 2002

[3] M. Evstatiev, S. Fakirov, B. Krasteva, K. Friedrich, J. A. Covas, A. M. Cunha, Polym. Eng. Sci., 42, 826 (2002)