## Microfibrillar reinforced polymer–polymer composites with amorphous matrix: morphological studies

K. Friedrich<sup>1</sup>, M. Evstatiev<sup>1</sup>, O. Evstatiev<sup>2</sup>, S. Fakirov<sup>2</sup>, N. Stribeck<sup>3</sup>, S. S. Funari<sup>4</sup>

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

A new type of polymer–polymer composites, the microfibrillar reinforced composites (MFC), was recently developed from polymer blends[1]. Unlike the classical macro-composites (e.g. fibre-reinforced ones) and the "molecular" or in situ composites (with "single" rigid rod-like macro-molecules as reinforcing elements), the MFC are reinforced by microfibrils of flexible chains. The latter are created during MFC manufacturing by drawing (fibrilization step) followed by melting of the lower-melting component (isotropization step) with preservation of the oriented microfibrillar structure of the higher-melting component. In the case of polycondensates, in addition to the isotropization during the thermal treatment, additional condensation and transreactions in the melt and solid state take place at the interface, resulting a copolymeric interphase playing the role of a self-compatibilizer.

Only very recently [2] one attempt was undertaken to prepare MFC with completely amorphous matrix. Suitable partners for this purpose are noncrystallizable polymers with a relatively high glass transition temperature  $T_g$  (around or above  $200^{\rm o}$ C) because of their attractiveness as engineering plastics. Another challenge was to reinforce such a matrix with liquid crystalline polymers (LCP). The goal of this study is to characterize the morphology of the MFC based on amorphous polymer (XYZ)<sup>1</sup> ( $T_g$  around  $220^{\rm o}$ C) (matrix) reinforced by LCP<sup>1</sup> by means of scanning electron microscopy (SEM), wide–angle X–ray scattering (WAXS) and small–angle X–ray scattering (SAXS).

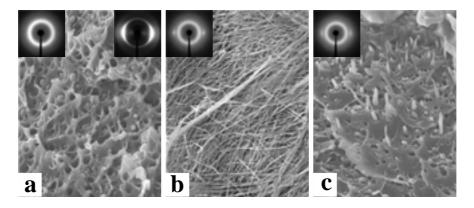


Figure 1: SEM micrographs of LCP/XYZ blends taken from cryogenic fracture surface and WAXS patterns from various stages of MFC preparation: a) after mixing and extrusion, X-ray patterns taken from the neat isotropic components XYZ (left) and LCP (right), b) after drawing (the matrix is removed by means of a selective solvent), c) after compression molding of the drawn bristles.

Figure 1 shows the typical morphologies for the steps of MFC preparation[1, 3]: rather perfect spherical particles homogeneously dispersed in the matrix after mixing and extrusion (Figure 1a),

<sup>&</sup>lt;sup>1</sup>The supplier of the materials does not disclose the chemical composition during various stages of the preparation of these polymers

well-defined microfibrils from LCP after drawing (Figure 1b), which basically preserve their shape after compression molding (Figure 1c).

The most striking conclusion derived from WAXS (Figure 1) is that the matrix preserves its isotropic amorphous state regardless of the processing steps in contrast to the LCP. Figure 2 shows some of the SAXS patterns of the neat components XYZ and LCP, their drawn blend and the injection molded sample with MFC structure taken at HASYLAB, beamline A2.

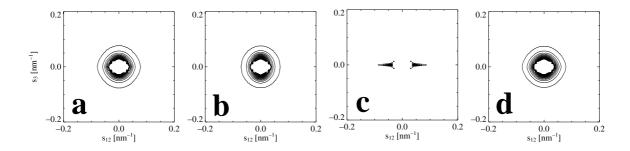


Figure 2: SAXS patterns taken at HASYLAB beamline A2 of: (a) neat matrix XYZ, (b) neat reinforcement LCP, (c) extruded and cold drawn blend LCP/XYZ (20/80 by wt.), (d) injection molded sample from the drawn blend with MFC structure

The SAXS patterns show that the neat components and the injection molded samples (Figure 2a,b and d) are isotropic in contrast to the drawn material (Figure 2c). In the last case the scattering is caused from an ensemble of uncorrelated, perfectly oriented microfibrils, similar to those observed by the SEM method (Figure 1b and 1c). The apparent contradiction regarding the orientation state derived from the SAXS (Figure 2d) and the SEM and WAXS (Figure 1c) is not a real one, because in the second case the processing of MFC is performed by compression molding where the microfibrils preserve their original uniaxial orientation, whereas in the first case (Figure 2d) the sample is prepared by means of injection molding resulting in random displacement of the microfibrils as already observed on other MFC systems [4].

**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, M. Evstatiev, K. Friedrich, "From Polymer Blends to Microfibrillar Reinforced Composites" in: D. R. Paul, C. B. Bucknall (Eds.) "Polymer Blends", vol. 2, chap. 33, Wiley, New York, 2000
- [2] K. Friedrich, M. Evstatiev, O. Evstatiev, S. Fakirov, unpublished data
- [3] M. Evstatiev, N. Nikolov, S. Fakirov Polymer, 37, 4455 (1996)
- [4] S. Fakirov, A. Apostolov, Z. Denchev, D. Sapoundjieva, M. Evstatiev, N. Stribeck, A. Meyer, J. A. Martins, Hasylab Annual Report 1, 723 (1999)