Chapter 12

Conclusion and Future Scope
12.1. Conclusion

Microfibrillar composites (MFCs) are a new class of fibre reinforced composites, and the manufacture of in-situ MFCs provides a promising route to enhance properties of general thermoplastics (such as LDPE, HDPE, and PP), using engineering thermoplastics (such as PET, polyamide, polycarbonates, and polybutylene terephthalate). The manufacture of MFCs also provide a technology to make best use of commingled plastic mixtures in which both low melting temperature general plastics and high melting temperature engineering plastics coexist and cannot be easily separated. Having a higher $T_m$ and demonstrating superior mechanical properties, the PET is expected to exist in the microfibril form as the reinforcement for the composite. One can consider the MFCs as a special type of in-situ composite combining the easier processability of the conventional polymer composites with the high aspect ratio of the liquid crystalline polymers (LCPs).

In this work, PP and LDPE were chosen as the general thermoplastics to be converted as matrix for the MFCs. The reinforcement polymer selected was PET. Having a higher $T_m$ and demonstrating superior mechanical properties PET is expected to exist in the microfibrillar form and thus reinforcing the resultant polymer-polymer composite. The effect of draw ratio on the various properties of the composite was analyzed in the case of PP/PET MFCs. In the case of LDPE/PET MFCs, the effect of blend ratio and the effect of a compatibilizer on the properties were studied.

In-situ composites were prepared from the blends of Polypropylene and Poly (ethylene terephthalate) by continuous drawing
followed by injection moulding. The SEM observations show high level of orientation for the drawn blends. The PET fibrils with the lowest mean diameter of 4.1μm were obtained at draw ratio of 8. After injection moulding at a temperature below the $T_m$ of PET, fibrils with high aspect ratio and good orientation were obtained for samples drawn at stretch ratio 5. Beyond stretch ratio 8, the breakage of the fibrils were observed during stretching which produced very short randomly distributed fibrils after injection moulding. In the case of MFBs the sharpness and intensity of the WAXD peaks at 14.2° increases with draw ratio and at high stretch ratios the peaks at 16.8° and 18.4° show a tendency to merge together to create a double peak. WAXD studies on MFCs showed the prevalence of sharp peaks at 14.4° whereas the peaks at other 2θ angles were submissive. The tensile, flexural and impact properties were found increasing with stretch ratio. Neat blends, H2I and H8I exhibited a brittle failure in comparison with H5I and H8I. In the case of H5I and H8I the higher aspect ratio and the abundance of the PET microfibrils reduces the cavitation formation and brittle failure of the MFC. The impact strength of MFCs was found to be higher for H8I on account of the fine (low diameter) microfibrils they possess. The experimental values for tensile strength and tensile modulus were compared with those obtained from the theoretical equations.

The effect of temperature on the dynamic mechanical properties like storage modulus, loss modulus and mechanical loss factor of PP, neat blend and in-situ composites prepared at different draw ratios were studied. The $E'$ values were found decreasing drastically for neat PP beyond $T_g$. The presence of microfibrils showed a positive effect on the
modulus at temperatures above \( T_g \) especially for the samples drawn at stretch ratio 5 and 8. The tan delta peak height at \( \beta \) transition (\( T_g \)) reduced and shifted to higher values with draw ratio. The \( E'' \) curves also showed positive shift in the peak values the highest being for samples drawn at stretch ratio 5. These samples exhibited high loss modulus values beyond 25°C which is indicative of the better energy dissipation. Based on the above studies it can be concluded that microfibrils of PET obtained at draw ratios between 5 and 8 can impart better properties for the MFC. The experimental values storage modulus was compared with those obtained from theoretical equations.

The PET phase in PP/PET blends can act as nucleating agents for the crystallization of PP. Generally, it can make crystallization of bulk PP in the blends at a higher temperature and for shorter time. For all the blends, their nonisothermal crystallization initiates at higher temperature, and thus is dependent on the nuclei density as compared to the neat PP. The presence of PET microfibrils reduces the melting temperature of the PP phase in the blends and composites. The long and thin microfibrils of PET in H5 and H8 can act as heterogeneous nucleating sites for the crystallization of PP. The percentage crystallinity of PP was highest for these samples. The non isothermal crystallization behaviour of the as extruded blend, microfibrillar blend and MFC prepared at a draw ratio 5 was compared. The analysis of the crystallization temperature and time characteristics revealed that the PET fibrils in the microfibrillar s blend had a greater nucleating effect for the crystallization of PP.

From the dynamic rheological studies, the storage modulus and loss modulus values were the highest for MFC prepared at stretch ratio 5
and 8 (H5I and H8I). The dynamic viscosity values were found to be higher for H5I and H8I. The randomly distributed PET microfibrils can form a physical network with the PP matrix which has a significant effect on the elastic behaviour than the viscous behaviour of the composite. The fibrillar morphology of the dispersed phase delays the thermal degradation of the MFBs considerably. The high aspect ratio and the abundance of the PET microfibrils contribute to the delayed degradation in H5 and H8. The activation energy for thermal degradation of the MFBs were found to be higher than PP and neat blends but lower than that of PET.

The morphology of the normal blends and the corresponding MFCs based on LDPE and PET were analyzed at the various stages of their preparation. The dispersed phase exhibited a spherical/nodular morphology in the case of the normal blends. The dispersed phase remained as discrete domains pointing towards the immiscibility of the phases. The average diameter of PET phase increased with an increase in the PET concentration for the normal blends. The morphology of the blends exhibited co-continuity at very high PET concentration of 45 wt%. The fibrillation of the dispersed phase was scantily seen at its low concentration in the case of MFBs. At 15 and 25 wt% of PET loading completely formed microfibrils of PET was observed. At higher PET concentrations the fibrils formed were not uniform in their diameters. During isotropization for the preparation of MFCs the aspect ratio of the PET microfibrils reduced due to the “break up behaviour”. A network of PET microfibrils was seen at 15 and 25 wt% PET concentration. The
diameter of the microfibrils was the lowest for M85 and M75. At 35 and 45 wt% PET concentration the microfibrils are found to be agglomerated.

The static and dynamic mechanical properties of LDPE/PET normal blends and MFCs were compared at varying PET concentrations from 5 to 45 wt%. The tensile strength of normal blends increased up to 35 wt% of PET loading beyond which it dropped. The tensile properties were found to increase with PET concentration up to 25 wt% in the case of MFCs beyond which it declines. The tensile strength obtained from theoretical equations was found to be in agreement with the experimental results only at low PET concentrations. The storage modulus of M75 was found to be the highest amongst the MFCs. The theoretical storage modulus values obtained using certain equations agreed with the experimental results up to 25 wt% PET loading. The tan δ peak values at \( T_g \) of LDPE were found to be lowest for M85 and M75 manifesting good fibre - matrix adhesion. The experimental tan δ\(_{\text{max}}\) values were found to be lower than that obtained by the theoretical equations suggesting the formation of an immobilized layer at the interfacial region of the matrix and fibre.

The melt blending of LDPE and PET in 85/15 and 75/25 w/w% was carried out in the presence and absence of PE-g-MA compatibilizer. In the case of 75/25 w/w % blends, the replacement of LDPE with 4 wt% PE-g-MA facilitated a reduction in the PET microfibril diameter. The addition of compatibilizer in 85/15 w/w% blends reduced the aspect ratio of the microfibrils due to hindrance in the coalescence. The compatibilized normal blends exhibited improved tensile properties than the corresponding non compatibilized ones in both 75/25 and 85/15
w/w% blends. In the case of MFCs, M75CM exhibited enhanced properties than M75M, whereas M85CM displayed inferior properties than M85M. The conversion of blends into microfibrillar composites improved the tensile properties of the resultant product and reduced the impact strength. The PET microfibrils present in the MFCs helps in the stiffening the composite rather than improving its toughness. The dynamic mechanical properties analysis of the blends and composites proved the positive effect of the microfibrils in creating restraints on the relaxation of the LDPE matrix and thereby improving the stiffness of the resultant product. The tensile modulus and storage modulus values of the neat MFCs were found to be higher than the compatibilized blends. The $T_g$ values obtained from the tan $\delta$ peaks shifted to higher temperature in the case of MFCs and compatibilized blends. The shifting of the $T_g$ value was appreciable in the case of MFCs. The activation energy for glass transition was higher for MFCs than the compatibilized blends.

The transport behaviour of xylene through LDPE, neat blends and MFCs were analyzed. As the PET concentration increases the diffusion and permeability coefficients of neat blends and MFCs reduces upto 25 wt% beyond which they increase. The diffusivity and permeability of the MFCs were found to be lower than that of the neat blends owing to the tortuous path they create for the diffusion of the solvent. The increase in solvent temperature enhances the diffusivity and permeability of all the samples. The samples tended to disintegrate very fast at elevated temperature. MFCs especially M75 showed the capability to impede disintegration since the physical network formed by the PET microfibrils binds the LDPE phase together. The diffusion characteristics obtained
experimentally were compared with theoretical equation. The diffusion of the solvent at elevated temperatures was found to be non Fickian. The thermal degradation characteristics of LDPE, neat blends and microfibrillar blends were compared. The activation energy for thermal degradation of the MS 75 was found to be higher than LDPE, neat blends and that of PET.

12.2. Scope of future work

- **Preparation of microfibrillar composites by extrusion-drawing-compression moulding technique.**
  
  o In the present study microfibrillar composites were prepared by the injection moulding process. The composites prepared by compression moulding are expected to maintain orientation of the reinforcing fibrils.

- **Analysis of the transcrystallization behaviour of microfibrillar composites by transmission electron microscopy.**
  
  o The transcrystalline layer of the continuous phase formed surrounding the reinforcing microfibrils could be analyzed using high resolution transmission electron microscopy.

- **Evaluation of the effectiveness of MFC technology for recycled plastics processing.**
  
  o MFCs can be prepared from recycled plastics like waste polyolefin films and waste PET bottles which are posing as a threat to the environment due to its littering. Extruders with
devolatalizing zones are suggested as recycled PET is susceptible to thermolytic and hydrolytic degradation.

- **Permeability studies through MFC membranes.**
  - As MFC films possess a network of the reinforcing microfibrils, apart from the improvement in the mechanical properties the permeability of the gases through them is expected to be restricted due to the “tortuousity”.

- **Studies on the effect of different compatibilizers on the properties of MFCs.**
  - In the present study only one compatibilizer was employed for the preparation of blends. The effect of other compatibilizers may be evaluated.
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Academic Qualifications
B.Tech (Mechanical Engg.)
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Work Experience
1. Industry
7 years (1992-99) in BPL Sanyo Technologies Ltd., Palakkad, Kerala

2. Teaching
11 years (1999 till date) at Amrita Vishwa Vidyapeetham, Coimbatore

3. Research
7 years (2003 till date) in the field of polymer processing and polymer composites

Research Interests
Polymer-polymer composites, Morphology development in polymer blends, Dynamic mechanical analysis of fibre filled polymer systems, Polymer processing optimization, Polymer nanocomposite, Polymer recycling.

Projects Handled
Polymer Nanocomposites for Inflatable Space Structures,
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Publications

In International Journals:


In Conferences/Seminars:


