Chapter 9

Conclusions and Future Outlook

Natural fibre composites, or biocomposites, have recently gained much attention due to their low cost, environmental friendliness, and their potential to compete with glass fibre composites. There has long been an interest in the use of natural fibres for engineering applications. Today, interest in natural fibres has soared with global demand for more efficient and less expensive environmentally friendly products.

The incorporation of vegetable fibres or particles into most polymers presents some difficulties, since dispersion is somehow prevented by the incompatibility between polar fillers and (usually) non-polar matrices. This drawback has been counteracted by chemically modifying the polymer or the filler or by adding compatibiliser agents. The increased compatibility results in improved dispersion of the particles and wetting of the fibres, so that higher concentrations of fibres or particles can be achieved in the composite thus improving the processability and mouldability.

The combination of different reinforcing fibres in one material is a promising trend where reduced mass is needed. The diverse and contradictory requirements for modern composite materials has brought to the forefront the very complex problem of developing new types of composite materials, in which a combination of reinforcing layers from two or more types of fibres is used. Polymer hybrid composite materials have found the widest application in the present time.

Pineapple fibres (PALF) are abundantly available in the tropics and are currently under-utilised. This is a potential candidate to be investigated for its behaviour as reinforcement. In the present work, the effectiveness of PALF as reinforcement in polyester (PER) matrix has been analysed in detail. PALF in the form of short random mats and also as hybrid of PALF and glass fibre (GF) mats has been used as reinforcement in polyester matrix. In our earlier studies, short PALF was found to be effective in improving the mechanical properties of polyester matrix.

Several chemical modifications were done on PALF to improve the interfacial bonding with polyester matrix and properties of the composite. The effectiveness of silane as a coupling agent was directly related to the reactivity of the functional group in co-polymerising with styrene diluted resin. Silane coupling agents with different functional groups were chosen to modify the fibre surface. Acid-base interactions govern fibre/matrix adhesion in natural fibre composites to a great extent. Solvatochromism measurements were done to evaluate the acidity and basicity of the treated PALF. Empirical solvatochromic polarity parameters such as hydrogen-bond donating ability HBD (α) or acidity, hydrogen-bond accepting ability HBA (β) or basicity, dipolarity (π^*), Gutman's acceptor number (AN) and Reichardt's $E_T(30)$ for the PALF and chemically treated PALF were estimated. Solvatochromic studies revealed that chemical modification done on PALF has lowered the HBD or acidity value. The HBD ability was found lowest for alkali treated fibre and highest for methyl triethoxy silane treated fibre. The basicity was found to be higher for methacrylate silane treated fibre. All chemical treatments given to PALF have decreased the polarity of PALF. The FTIR

and SEM studies of PALF revealed the surface morphology after the treatments.

The thermal degradation behaviour of PALF showed that the decomposition was found to take place in two successive stages after the moisture loss. The chemical modification does not show any significant change in the decomposition pattern. However a higher thermal stability was observed for all the treated fibres. The studies revealed that all the treatments improved the thermal stability of PALF indicating higher residues at comparatively lower temperatures upto $300^{o}C$ and shifting of degradation peaks to higher temperatures. The scanning electron micrographs and optical microscopy studies of the fibre showed that the fibre became porous and fibrillated by silane treatments.

A unique combination of PALF and GF was taken to design short hybrid fibre composites in unsaturated polyester matrix. Individual fibre composites namely PALF/PER and GF/PER composites of different loadings were also prepared. All these composites were prepared by compression moulding method. In the present study, the improvement in properties of the hybrid composites on increasing GF loading was evaluated.

Mechanical performance of PALF/PER and GF/PER composites was increased with fibre loading and the optimum loading was found to be 40 wt %. The composite of 50 wt% showed a decrease in properties due to the incomplete wetting of the fibres by the matrix. The impact strength is increased with fibre loading for PALF/PER composites. GF/PER composites showed comparatively higher impact properties than PALF/PER composites. The GF/PER composite of 16 wt% showed maximum value and levelling off is observed on further addition. Mechanical property measurements of the

chemically modified PALF/PER composites were investigated to learn more about the fibre/matrix adhesion. Chemically modified PALF/PER composites showed enhanced tensile and flexural properties compared to untreated fibre composites. While comparing the effect of various treatments done on PALF, vinyl silane treated composite showed higher tensile strength and flexural properties. All the chemically modified PALF/PER composites showed lower impact strength than the untreated composites except methacrylate silane treated composites where an increase is noticed. The tensile and flexural properties were found to be improved in the case of composites treated with vinyl and methacrylate silane. Fibre/matrix adhesion was found to depend on the polarity of PALF. The improved fibre/matrix adhesion of vinyl silane treated composite accounts for the enhancement in properties. The scanning electron micrographs of the tensile fracture surfaces which showed the penetration of the resin into the matrix, less pull out and debonding in these composites. It was further confirmed from the FTIR studies of silane treated fibres.

The effect of hybridising PALF with glass fibres on the tensile, flexural and impact properties of the hybrid composites was studied in detail. The mechanical properties of short PALF/GF hybrid polyester composites were evaluated by varying the relative volume fractions of PALF and GF at a total fibre loading $0.3V_f$. The tensile properties of intimately mixed (IM) composites were considerably enhanced by the incorporation of small amount of GF i.e, $0.1V_f$. Incorporation of 0.9 V_f of GF showed maximum value of tensile strength which is greater than that of 100% GF reinforced polyester composite. The IM hybrid composites based on 0.9 V_f showed the highest flexural strength too. The hybrid composite of higher GF content

showed comparatively higher flexural strength and flexural modulus. The IM hybrid composites have higher impact strength than 100 % PALF or GF composites. The IM composite having 0.3 volume fraction of GF showed maximum impact strength of 1203 J/m which is twice that of GF/PER composite. A synergistic effect of the two fibres leads to the increase in impact strength.

Of the different layering patterns of PALF/GF hybrid polyester composite, GPG of 0.1 volume fraction of GF showed 40 % increase in tensile strength and 0.7 volume fraction of GF showed maximum value of tensile strength which is greater than 100 % GF/PER composite. The highest flexural strength values were observed for composites of 0.9 volume fraction of GF. The tensile and flexural properties of GPG were much higher than the other layering patterns while PGP showed comparatively higher notched impact strength at low volume fractions of GF. Since inclusion of small quantities of GF increased the tensile strength, cost effectiveness could be achieved in this case also. The impact strength of GPG hybrid composite was also increased with increase in GF content. The impact strength of composite of 0.9 volume fraction of GF was 90 % greater than the neat GF/PER composite. Hybrid effect was calculated using the additive rule of hybrid mixtures. A positive hybrid effect was observed for tensile modulus, flexural modulus and impact strength for IM and GPG hybrid composites. A negative hybrid effect for elongation at break is observed for both. Difference of behaviour is shown in the impact strength of IM and GPG composite; addition of small amount of GF highly improves the impact strength of IM composite whereas addition of more amounts shows better property for GPG.

Dynamic mechanical properties of PALF and GF reinforced polyester composites and pineapple leaf/glass hybrid fibre reinforced polyester composites have been analysed. The properties were evaluated as a function of fibre loading, hybrid fibre ratio and hybrid layering pattern. The dynamic modulus and mechanical damping factor of PALF/polyester **c**omposites of varying fibre aspect ratio were also considered as a function of temperature and frequency. Incorporation of PALF increased the storage modulus and the composites of 30 mm fibre length (l/d 600) showed highest modulus values at all temperatures. All composites showed a decrease in modulus values as the temperature is increased. The storage modulus was found to increase with fibre loading in the composites and the composite of 40 wt% and l/d 600 showed higher modulus and least damping at all temperatures. The neat resin showed highest tan δ value indicating good damping characteristics. Dynamic mechanical analysis revealed an additional peak in the case of PALF/PER composites with 40 %. This showed a strong interphase in these composites. GF reinforced composites showed higher storage modulus than that of PALF reinforced composites. The storage modulus and loss modulus of GF/PER composite increased with fibre loading and decreased with temperature; maximum value is observed for 40 wt% as in the case of mechanical properties.

Dynamic mechanical analysis of PALF/GF hybrid fibre polyester composites showed that addition of small amount of GF enhanced the storage modulus values. The IM hybrid composite of 0.2 (V_f) GF showed highest stiffness. The damping values were also decreased for 0.2 volume fractions of GF for IM hybrid composites. An additional relaxation indicating strong interphase, the decrease in the coefficient of reinforcement value and higher value of

activation energy for 0.2 V_f IM hybrid composite are indications of better interfacial interaction. The low impact strength observed for these composite is consistent with the above result. Of the different layering patterns studied, GPG composite showed higher storage modulus and low damping. The GPG composite of 0.2 (V_f) GF showed comparatively higher storage modulus values. However, 0.3V_f GPG composite showed higher stiffness at higher temperatures. The stiffness of PGP composite is increased with increase in GF loading. The E' values of PALF/GF hybrid lie between those of GF reinforced and PALF reinforced composites. The tan delta curve is found to be affected by the layering pattern. Determination of activation energy for the glass transition of the hybrid composites from the Arrhenius plots revealed that activation energy was highest for volume fractions of GF, 0.2, 0.3 and 0.2 for IM, GPG and PGP composites. The effectiveness of reinforcement of IM and GPG composites is higher compared to PGP. The lower storage modulus values of PGP compared to IM and GPG are consistent with the results of tensile strength. The cole-cole plots constructed for the hybrid composites showed relatively good fibre/matrix adhesion. Experimental storage modulus and damping factor were compared with theoretical predictions.

The thermograms of TGA analysis of the PALF/PER composites showed two-stage decomposition similar to PALF. The increased thermal stability of the composites compared to PALF was due to the improved fibre/matrix interaction which is also clear from other properties studied. The chemical treatment of PALF does not show any significant influence in the decomposition pattern of the composite. The treated fibres were found to decompose at higher temperature than the untreated fibre indicating higher

thermal stability. Among the treated composites, NaOH treated composites showed higher thermal stability than the untreated composites. They showed lower water absorption also. The PALF/GF hybrid composites showed thermal behaviour in between that of PALF/PER and GF/PER composites as expected. The IM and GPG hybrid composites of higher GF content are found to be thermally more stable. The GPG hybrid composites of 0.2 volume fraction of GF showed higher thermal stability which is comparable to GF/PER composites. The GPG composite of $0.2V_f$ exhibits good dynamic mechanical thermal properties also. These composites possess higher storage modulus, low damping and higher interfacial adhesion.

The incorporation of PALF in polyester resin showed increase in thermal conductivity and thermal diffusivity. The thermal conductivity, thermal diffusivity and density values were increased with the increase in glass fibre volume fraction whereas specific heat values were decreased. It is found that the glass fibre allows better ability to the heat transport in the composite.

The water absorption studies of chemically modified PALF/PER and PALF/GF hybrid composites revealed that water uptake was dependent on temperature, chemical treatments given and PALF/GF content. The neat polyester shows minimum water absorption. It is observed that the initial rate of water sorption and the equilibrium rate of water uptake of PALF/PER composite increases with increasing temperature. The NaOH and vinyl silane treated composites showed lowest water absorption at 90^{o}C. The diffusion coefficient of PALF/GF hybrid and chemically modified composite increases with temperature upto 60^{o}C and decreased for 90^{o}C. The values are lowest for GPG hybrid composites of higher GF content. At higher temperatures, the hydrogen bond formation tends to disappear and the absorption –

diffusion mechanism in this case will be slow. The moisture uptake for both hybrid systems IM and layered GPG decreased with increase of glass fibre content and the composites of GF content $0.5V_f$ showed minimum water uptake. This is due to the lower hydrophilicity of GF than PALF. These composites showed highest activation energies indicating that entry of water is hindered by the presence of a strong interface. Hybrid composites with greater GF content were found to possess lower water absorption. Among the chemically treated PALF/PER composites, 2% NaOH treated composite showed highest activation energy and PSMA treated composites showed minimum.

The effect of different aggressive environments on the performance of PALF/PER and PALF/GF hybrid polyester composites has been evaluated. Boiling water and cold water ageing result in an increase of the composite weight due to the absorption of water. The thickness swelling of IM and GPG hybrid composites of 0.5 (V_f) GF content was found to be low. A higher retention in tensile strength was observed for all the composites on cold water ageing for 2 hours. Both IM and GPG hybrid composites of 0.5 GF content showed a higher retention in tensile strength and modulus even after hot water ageing. Higher retention in elongation at break is also observed for PALF/PER and the hybrid composites on water ageing. The composite impact strength is decreased on water ageing. The modulus value showed an increase in all the thermally aged composites. Increased temperature leads to increase in crosslinking which increases the modulus of the composites.

The vinyl silane treated composite absorbed only small amount of water on immersion in water for 2 hours. This indicated better moisture resistance of

the treated composite. A higher retention in tensile strength and impact strength was also observed after environmental ageing. The decreased water absorption of these composites indicates strong fibre/matrix adhesion. The silane treated composites also showed significantly lower swelling than the untreated.

It was observed that the mechanical properties of all the composites decreased considerably after three days of thermal ageing. Impact strength was greatly enhanced for the GPG composites after thermal ageing. GPG hybrid composites of higher GF content showed higher retention in tensile strength and elongation at break after outdoor ageing and soil immersion. Overall studies indicate that the short PALF/GF hybrid composite exhibits better resistance to ageing than the neat PALF/PER composite. Thus by hybridising PALF with small amounts of GF the durability of natural fibre composite under environmental ageing could be improved.

The results also reveal that the incorporation of both these fibres improved the interaction at the interfacial region of PALF, GF and PER matrix. As the mechanical properties and the durability of the composites were improved upon GF addition, this may expand the applicability of these composites in automotive and building products industries.

Future Outlook

1. The fibre/matrix interface between PALF and polyester needs to be investigated as the composite performance is determined by the fibre/matrix interface. The interface may be characterised by various techniques like ESCA, microdebond test, single fibre pullout test etc. The relationship between interface and bulk composite properties should be established.

2. Resin transfer moulding (RTM) can be used as an ideal technique to manufacture composites having superior properties, economically.

3. The use of cellulose microfibrils as reinforcement in polyester matrix is a new and emerging field to develop microfibril composites.

4. Fully green composites would be developed by reinforcing PALF nanofibrils in biodegradable polymer resins to produce environmentally friendly materials.

5. Product development – Many useful products like cases and covers for appliances, packaging applications, automotive seats, furniture, table tops etc. can be fabricated using PALF/PER composites. New applications should be found for the PALF/GF hybrid polyester composites.