Chapter

3

INFLUENCE OF RELATIVE VOLUME FRACTION, INTIMATELY MIXED ORIENTATION AND HYBRID EFFECT ON THE MECHANICAL PROPERTIES OF SHORT SISAL/GLASS HYBRID FIBRE REINFORCED LOW DENSITY POLYETHYLENE COMPOSITES

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3.1. Introduction

The reinforcement of two or more fibres into a single matrix leads to the development of hybrid composites with a great diversity of material properties. The potential advantages of combining two kinds of fibre in a single polymer matrix have been described by many researchers. Most of them reported that the behaviour of hybrid composites appeared to be simply a weighted sum of the individual components. There is a more favourable balance between the advantages and disadvantages inherent in any composite material when it is a hybrid fibre system. It is generally accepted that the properties of hybrid composites are controlled by many factors such as the nature of matrix, nature, length and relative composition of the reinforcements, fibre-matrix interface, hybrid design etc. Zweben reported that during the failure of the hybrid composites, high elongation fibres in the composites enhance the strain level required for the propagation of cracks through the composites. So high elongation fibre functioned as crack arrestors on a micromechanical level. Marom et al. reported that aramid/carbon/aramid (ACA) type sandwich hybrid composite with aramid fibre reinforced skin and carbon fibre reinforced core exhibited a positive hybrid effect in their flexural strength. Peijs and De Kok found that intimately mixed hybrid composites based on epoxy resin/carbon fibre/chromic acid treated polyethylene fibres, exhibited better tensile and fatigue properties, than those containing untreated polyethylene fibres.

It is important to mention that only limited studies were carried out in the field of discontinuous fibre reinforced hybrid composites. Ritcher investigated on the properties of hybrid composites of epoxy resin containing short oriented carbon and glass fibres. He obtained tensile strength values up to 70% and modulus values up to 90% of those obtained with continuous hybrid fibre composites.
Investigation on natural fibre incorporated hybrid composites revealed that their properties can be effectively utilised in hybrid composites. Clark and Ansell\(^8\) reported the improvement of various mechanical properties of jute/glass hybrid laminate with different arrangements of plies in the laminate. According to them, glass core laminates offered exceptionally low cost in terms of impact energy. Pavithran and coworkers\(^9\) carried out a detailed investigation of sisal/glass hybrid fibre and coir/glass hybrid fibre reinforced polyester resin and showed that the addition of small amounts of glass fibres to the natural fibres not only enhanced the mechanical properties but also improved the weathering characteristics of the composites. It is important to mention that the performance of hybrid fibre reinforced composites can be improved to a great extent by proper chemical modifications. Varma et al.\(^{10}\) investigated the effect of three coupling agents viz., silane, titanate, and toluene diisocynate on the mechanical properties of jute/glass hybrid fibre reinforced polyester composites. They concluded that composites containing titanate treated jute fabric exhibited better mechanical properties than those reinforced with silane or TDI modified jute fibres.

This chapter discusses the changes in mechanical properties and water absorbing capacity of short sisal/glass hybrid fibre reinforced LDPE composites. Emphasis is given to the relative volume fraction of the two fibres, fibre orientation, hybrid effect and chemical modification of sisal fibre.

3.2. Results and discussion

Mechanical properties of two sets of hybrid composites were studied in this chapter. In the first and second sets, tensile testing was carried out at a crosshead speed of 5 mm/min and 50 mm/min, respectively.
(a) **First set**

In this set, the properties of the hybrid composites were studied as a function of relative change in the volume fraction of SRP and GRP.

(b) **Second set**

In second set, the properties have been studied as a function of the increasing volume fraction of one type of fibre by keeping the other as constant.

In the first set, the total volume fraction of sisal and glass fibres is almost same in all compositions of SRP and GRP (90/10, 80/20, 70/30, 60/40, 40/60 and 30/70). Figure 3.1 shows the stress-strain behaviour of pure LDPE, pure sisal, pure glass, SRP, GRP and their first set of hybrid composites in the longitudinal orientation. It can be seen from the figure that pure LDPE exhibits quite a large necking behaviour. Necking of LDPE cannot be seen in the Figure 3.1 due to the change in the strain percentage scale. In the case of pure sisal and glass fibre, the stress increases proportional to strain and a sharp break is observed. No necking behaviour is observed in the case of sisal and glass fibre. When sisal fibre is incorporated in the LDPE, the necking decreases to a large extent as the volume fraction of glass fibre (GRP) in the hybrid composites increases the necking again decreases for composite containing glass alone (GRP). This can be explained by the fact that the glass fibre shows more elasticity than sisal fibre which in turn is more elastic than LDPE. It can also be seen in the figure that the yield stress of the hybrid composite increases as the volume fraction of GRP increases. Also there is a reduction in the percentage strain of the composites with increase in the volume fraction of glass. This is due to the fact that glass is highly brittle. As the volume fraction of glass increases, the yield stress of the composite system increases, but at the same time the system becomes more brittle. The increase in the value of yield
stress with volume fraction of GRP is attributed to the high modulus of glass fibres oriented perpendicular to the direction of crack propagation. Figure 3.1 clearly shows that as the volume fraction of GRP increases, the area under the curve decreases. This indicates a reduction in the overall ductility of the samples. The shapes of the stress-strain curves in Figure 3.1 also indicate that, as the volume fraction of GRP increases, the hybrid composites (GSRP) become more strong and hard compared to SRP.

Figure 3.1. Stress-strain curves of LDPE, sisal fibre, longitudinally oriented SRP GRP and GSRP (80/20, 70/30 and 30/70 compositions of SRP/GRP) composites.

Stress-strain curves of second set of hybrid composites under tension are shown in Figures 3.2-3.4. Figure 3.2 shows the stress-strain behaviour of longitudinally oriented fibre composites. Stress strain curves in this figure also exhibit a similar tendency as in the previous case (Figure 3.1).
Figure 3.2. Stress-strain curves of SRP, GRP and GSRP with longitudinal fibre orientation.

Figure 3.3. Stress-strain curves of SRP, GRP and GSRP with random fibre orientation.
Figure 3.4: Stress-strain curves of SRP, GRP and GSRP with transverse fibre orientation.

The stress-strain behaviour of randomly oriented composites is depicted in Figure 3.3. All the samples show necking behaviour. It is observed that these composites exhibit lower value of yield stress than longitudinally oriented ones, for all compositions. The randomly oriented fibres act as barriers and prevent the stress transfer between fibre and matrix, and thus contribute to higher concentration of localised stresses. This explains the reduction in yield stress value and necking behaviour of random type hybrid composites. Randomly oriented composites exhibit high value of percentage strain compared to longitudinal and transverse type composites (Figures 3.2-3.4). This can be due to the decreased effect of randomly oriented fibres on matrix strain than transversely or longitudinally oriented fibres. It may be noted that the fibres have considerably lower percentage strain compared to the matrix, and their alignment in the direction of the applied stress effectively reduces the percentage strain of the matrix.

Figure 3.4 shows the stress-strain behaviour of transverse type hybrid composites. In this case, the strength of the composites is very much lower than that of the longitudinal and random oriented types. In transverse orientation, the stress application and fibre orientations are opposed to each other. Therefore, the
stress transfer from matrix to fibre is not effective due to the poor interfacial bonding between transversely oriented fibres and matrix. This eventually causes an instant failure of hybrid composites during tension. This clearly accounts for the low value of yield stress and absence of necking behaviour in transversely oriented hybrid composites. Percentage strain of transverse type is lower than that of longitudinal or random type. The perpendicular orientation of fibres to the applied stress direction and the weak interfacial bond strength between fibre and matrix effectively reduces the overall straining of the matrix. In order to analyse the influence of fibre orientation on the stress-strain behaviour, the stress-strain properties of GSRP at three orientations are compared. Figure 3.5 shows the stress-strain curves of longitudinal, random and transverse GSRP containing equal percentage of glass fibre (glass volume fraction = 0.015). The figure clearly indicates that the randomly oriented composites exhibit maximum percentage strain while transversely oriented composites, the minimum. The modulus and ultimate strength properties are maximum in the case of longitudinal and the minimum in the case of transversely oriented composites.

![Stress-strain curves of GSRP](image)

Figure 3.5. Stress-strain curves of GSRP with longitudinal random and transverse fibre orientation containing 0.015 volume fraction of glass fibres.
Figure 3.6 shows the variation of tensile strength of second set of composites upon the addition of glass fibres to SRP and GRP containing equivalent volume fraction \(V_f = 0.14\) of sisal and glass fibres. The values on the y-axis of the graphs at zero level of x axis indicate the properties of SRP and GRP containing 0.14 volume fraction of sisal and glass respectively. SRP becomes hybrid composite of sisal and glass (GSRP) on addition of glass fibres. The tensile properties of GSRP also shown in the figure.

![Diagram](image.png)

**Figure 3.6.** Variation of tensile strength with volume fraction of glass for GRP and GSRP with 0.14 initial volume fraction of glass in GRP and 0.14 initial volume fraction of sisal in GSRP at three orientation of the fibres.
A typical strength composition curve of intimately mixed aligned short fibre hybrids is shown in Figure 1.3.\textsuperscript{10}

Figure 1.2. Different hybrid configurations

Figure 1.3. General form of the strength of intimately mixed two-fibre hybrids.
Incorporation of glass fibre by about 0.03 volume fraction of SRP containing about 0.14 volume fraction of sisal fibres increases the ultimate tensile strength by 80% (18.7 to 33.97 MPa) for longitudinally oriented composites, by 37% (13.22 to 18.12 MPa) for randomly oriented composites and by less than 1% (1.92 to 2.53 MPa) for transversely oriented composites (Figure 3.6). In Figure 3.6 it can also be seen that at low glass content (volume fraction of glass around 0.03) the tensile strength of longitudinally oriented hybrid composite (GSRP) is close to that of GRP containing equivalent volume fraction of glass fibres. In the case of randomly oriented one, the values are lower as compared to the latter one. But the tensile strength values of transversely oriented GSRP and GRP are nearly same at all volume fractions of glass.

Figures 3.7a-c represent the photomicrographs of tensile fracture surfaces of the SRP, GRP and GSRP in the second set of composites respectively. From these photographs it is seen that as a result of poor interfacial bonding between fibre and matrix shear failure is occurred in SRP, GRP and GSRP composites. The micrograph given in Figure 3.7c shows failure surface of GSRP containing 0.05 $V_r$ of glass fibres. It is seen that irregularity of fracture surface is more marked in the case of hybrid composites. This is due to the difference in crack propagation mechanism in GSRP compared to SRP and GRP. In GSRP, the interface between glass and polyethylene is weak compared to sisal-polyethylene due to the smooth surface of the glass fibre compared to the surface of the sisal fibre. Therefore failure initiates and randomly propagates through the weak glass-polyethylene interface. Scanning electron micrographs of the similar composites in the second set are shown in Figures 3.8a-c. High degree of fibre pull out seen in these photographs gives a clear evidence for the shear failure in all these composites.
Figure 3.7. Photomicrographs of tensile failure surfaces of (a) SRP ($V_f$ sisal = 0.14), (b) GRP ($V_f$ of glass = 0.14) and (c) GSRP ($V_f$ sisal : $V_f$ of glass = 0.14 : 0.05).

Figure 3.8. Scanning electron micrographs of failure surfaces of (a) SRP ($V_f$ sisal = 0.14), (b) GRP ($V_f$ of glass = 0.14) and (c) GSRP ($V_f$ sisal : $V_f$ of glass = 0.14 : 0.05).

Figure 3.9 shows the variation in tensile strength of first set up hybrid composites as a function of relative change in the volume fraction of SRP and GRP. There also, the tensile strength increases with increase in volume fraction of glass fibres. Addition of glass in the sisal- LDPE system helps to attain a uniform
dispersion of the sisal fibre and prevent fibre to fibre contact in the matrix. This can be understood from the optical photographs shown in Figure 3.10. Figures 3.10a and 3.10b represent GRP and SRP respectively. Figure 3.10c indicates the decreased fibre to fibre contact on addition of glass fibres. As the volume fraction of glass increases, agglomeration of glass fibre takes place (Figure 3.10d). This in fact accounts for the decrease in positive hybrid effect at high glass fibre loading which will be discussed in the coming section. The increase in tensile strength of hybrid composites (Figure 3.6 and 3.9) is due to the higher tensile strength of glass fibre than sisal fibre and also to the high degree of dispersion of the sisal by the additional incorporation of glass fibres. Figure 3.9 also shows that in the case of tensile strength of randomly oriented composite, addition of glass fibre results in no remarkable improvement in tensile strength. This clearly highlights the fact that if glass fibres are not oriented, hybridisation makes no improvement in the tensile strength of the composites. Scanning electron micrographs of the tensile fracture surfaces of first set of hybrid composites are shown in Figure 3.11. The Figure also confirms the fact that tensile failure of untreated composites is mainly due to fibre pull out.

![Figure 3.9. Variation of tensile strength (experimental and theoretical) of GSRP with relative volume fraction of GRP.](image-url)
Figure 3.10. Optical micrographs of the cross-section of (a) GRP, (b) SRP, (c) GSRP containing 0.3 volume fraction of GRP and (d) GSRP containing 0.6 volume fraction of GRP (Magnification × 60).

Figure 3.11. Scanning electron micrograph of fracture surface of GSRP (50/50 composition of SRP/GRP) composites containing untreated fibres.
Figure 3.12 shows the variation of Young's modulus of second set of hybrid composites as a function of volume fraction of glass fibres. Tensile moduli of all composites increase with increase in volume fraction of glass fibre. Generally, as the volume fraction of non-bonded fibres increases, the tensile moduli decrease because non bonded fibres act as failure initiators. In the present case there is no chemical interaction between the fibre and the matrix. However, there exist only physical bonding due to the mechanical interlocking of fibre and matrix. Addition of glass fibre increases the effective mechanical interlocking between sisal and polyethylene. This inturn increases the frictional force between the fibre and matrix. Therefore, one of the reasons for enhancement in tensile moduli is attributed to the frictional force existing between fibre and matrix. Tensile modulus of glass fibre is higher than that of sisal fibre. This also may be another reason for the enhancement in modulus upon glass fibre addition. Variation in Young’s modulus values of longitudinal and randomly oriented composites (first set) with volume fraction of GRP is seen in the Figure 3.13. It is observed that increase in Young’s modulus of both the directions (longitudinal and random) is due to higher Young’s modulus of glass fibre compared to sisal fibre.

![Figure 3.12](image)

**Figure 3.12.** Variation of tensile modulus with volume fraction of glass for GRP and GSRP with 0.14 initial volume fraction of glass in GRP and 0.14 initial volume fraction of sisal in GSRP at three orientation of the fibres.
Figure 3.13. Variation in Young's modulus with relative volume fraction of GRP in GSRP composites.

Elongation at break values of the second and first sets of hybrid composites is shown in Figures 3.14 and 3.15. Figure 3.14 shows the elongation at break values of composite as a function of increasing amount of GRP. In all cases, addition of glass fibre reduces the elongation at break value because of the lower elongation at break value of glass fibre compared to those of sisal fibre and polyethylene. Figure 3.15 reveals the reduction in elongation at break values of hybrid composites by the addition of glass fibres. It is interesting to see that in both cases (longitudinal and random), elongation at break values were decreased by the addition of GRP. Figure 3.15 also shows that elongation at break values of longitudinally oriented composite are lower than that of randomly oriented composites.
Figure 3.14. Variation of elongation at break with volume fraction of glass for GRP and GSRP with 0.14 initial volume fraction of glass in GRP and 0.14 initial volume fraction of sisal in GSRP at three orientation of the fibres.

Figure 3.15. Variation in elongation at break with relative volume fraction of GRP in GSRP composites.
Figure 3.16 shows the tear strength of longitudinally aligned hybrid composites (first set). The property increases with relative increase in volume fraction of GRP. The tear strength of LDPE is very low compared to hybrid composites. At the time of tearing, fibres in the composite prevent the growth of crack front because the fibres are aligned perpendicular to the direction of crack growth. This in fact increases the tear strength of the composite. Tear strength of glass fibre composites is higher than that of sisal filled composites. Load-displacement curves (tear curves) of the samples are given in Figure 3.17. Polyethylene tears at the minimum force with maximum displacement while GRP, tears at the highest force with minimum displacement. This indicates that glass reinforced composites have high resistance to tearing. It is seen that the tearing force decreases and displacement increases with the decrease in the amount of glass fibres.

![Figure 3.16. Variation in tear strength with relative volume fraction of GRP in GSRP composites.](image)
Figure 3.17. Load displacement curves of LDPE, SRP, GRP and GSRP (different relative composition of GRP) composites.

Figure 3.18 shows the hardness values of first set of hybrid composites with relative increase in volume fraction of GRP. Increase in hardness may be due to the higher stiffness of glass fibre compared to sisal fibre. Diameter of the glass fibre is very much lower than that of sisal fibre. Hence glass fibres were more closely packed than sisal fibre in the composite. This also may be another reason for improving the hardness of hybrid composites.

Figure 3.18. Variation in hardness with relative volume fraction of GRP in GSRP composites.
Figures 3.19 and 3.20 depict the flexural properties (second set) of GSRP and GRP (longitudinal oriented) as a function of volume fraction of glass fibres. Figure 3.19 shows that the flexural strength of GRP is almost unaffected by the incorporation of glass fibres. However, in the case of GSRP it goes on increasing with addition of glass fibres. The incorporation of glass fibre by about 0.03 volume fraction in SRP containing about 0.14 volume fraction of sisal increases the flexural strength by 65% (14.12 to 23.23 MPa). However, beyond 0.04 volume fraction of glass, the value increases only slightly and then levels off. This levelling of tendency indicates that the optimum volume fraction of glass fibre for maximum flexural strength is 0.04. It is interesting to note that when the volume fraction of glass reaches 0.09, the flexural strength of GSRP is very much close to that of GRP. Figure 3.20 indicates that in the case of GRP, the flexural modulus does not change considerably up to 0.04 volume fraction of glass. Beyond 0.04 volume fraction of glass, the value increases and finally levels off. For hybrid composites (GSRP), the flexural modulus regularly increases with increase in volume fraction of glass and then levels off. This regular increase is due to the hybrid effect caused by the incorporation of glass fibres into SRP.

![Figure 3.19](image)

**Figure 3.19.** Variation of flexural strength with volume fraction of glass for GRP and GSRP with 0.14 initial volume fraction of glass in GRP and 0.14 initial volume fraction of sisal in GSRP at longitudinal orientation.
Figure 3.20. Variation of flexural modulus with volume fraction of glass for GRP and GSRP with 0.14 initial volume fraction of glass in GRP and 0.14 initial volume fraction of sisal in GSRP at longitudinal orientation.

Photographs of the flexural tested samples of SRP, GSRP and GRP (second set) are shown in Figure 3.21a-c. Figure 3.21a indicates the compressive side of a flexural tested SRP. A small whitening region of the surface of the sample denotes the formation of a craze at the failure of the sample. Figure 3.21b indicates the compressive side of GSRP. In this case crazing increases a little more which can be observed from a clear white marking on the surface of the sample. Figure 3.21c indicates the failure surface of the compressive side of GRP. A sharp white marking on the material indicates that the crazing is still intensive in GRP. In other words, as glass content increases, crazing of the composite also increases. This may be due to the more brittle nature of glass fibre than sisal fibre.
3.2.1 Comparison with theoretical predictions

Tensile modulus of the second set of hybrid composite was calculated using the modified Halpin-Tsai equation. According to the Halpin-Tsai equation

\[
\frac{M_H}{M_m} = \frac{1 + A \eta V_f}{1 - \eta \psi V_f}
\]

(3.1)

where \(M_H\) and \(M_m\) are the modulus of the hybrid composite and matrix, \(V_f\) is the volume fraction of the fibres. In this case \(V_f\) is the total volume fraction of the two fibres. \(A\) is a constant called Einstein’s coefficient for composites which is
governed by the geometry of the reinforcement. In this equation the value of $A$ is found to be 2.5. It was reported that the $A$ value for aggregates of spheres can have the same value as that of short fibres or rods.\textsuperscript{11} The values of $\eta$ and $\psi$ are given by the equations,

$$\eta = \frac{M_f / M_m - 1}{M_f / M_m + A}$$

(3.2)

$M_f$ and $M_m$ are the modulus values of the fibre and matrix respectively.

$$\psi = 1 + \left[ \frac{1 - \phi_m}{\phi_m^2} \right] V_f$$

(3.3)

where $\phi_m$ is the packing fraction of fibres.

The value of packing fraction for different fibre arrangement in the matrix has already reported in the literature.\textsuperscript{12} The different arrangements of fibres in the matrix are shown in Figure 3.22. These include square packing, hexagonal packing and random packing. In the present case, it is assumed that the fibres are randomly close packed in the matrix. Therefore the value of $\phi_m = 0.82$ is substituted in the equation (3.3). Figure 3.23 shows the theoretical and experimental variation of tensile modulus with volume fraction of glass fibres. The figure shows that the experimental values of tensile modulus is higher than the theoretical predictions. The reason may be attributed to the different diameters of the sisal and glass fibre which affect the packing fraction. The mixtures of fibres with different diameter can pack more densely than one type of fibres having larger diameter. Diameter of the sisal fibre is very much larger than that of glass fibre and hence glass fibre can fill the interstitial space between the closely packed sisal fibres to form an agglomerate. These agglomerated fibres may be able to carry a larger proportion of the load there by increasing the modulus of the system.
Figure 3.22. Different packing arrangements of fibres in the composite.

Square packing
\( \phi_m = 0.785 \)

Hexagonal packing
\( \phi_m = 0.907 \)

Random packing
\( \phi_m = 0.820 \)

Figure 3.23. Variation of experimental and theoretical (Halpin-Tsai) tensile modulus with volume fraction of glass for GSRP containing an initial volume fraction of 0.14 sisal in longitudinal orientation.
3.2.2 Hybrid effect calculation

The hybrid effect was theoretically calculated using the law of additive rule of hybrid mixtures. According to this law hybrid properties are calculated using the equation.

\[ X_H = X_1 V_1 + X_2 V_2 \]  

(3.4)

where, \( X_H \) is the characteristic property of hybrid composite, \( X_1 \) and \( X_2 \) are the characteristic properties of individual composites. \( V_1 \) and \( V_2 \) are hybrid volume fractions of reinforcements of the individual composites. The above equation is justified only if the total volume fraction of the reinforcement is almost equal for all the hybrid composites. In the first set of hybrid composites, this is true for all compositions. When the values of theoretically calculated properties of hybrid composites using equation (3.4) are higher than those of the experimental values, a positive hybrid effect is observed. If the properties are lower than the theoretical, a negative hybrid effect is observed. The theoretical curves drawn according to the rule of additivity (equation 3.4) are also shown in Figures 3.9, 3.13, 3.15, 3.16 and 3.18. It is evident from these figures that the hybrid composites exhibit a positive hybrid effect for all the mechanical properties except for elongation at break. This positive deviation is mainly attributed to the higher degree of dispersion of sisal fibre in SRP with additional incorporation of glass fibre (GRP) as discussed before. But it is seen that as the volume fraction of GRP increases, there is a reduction in positive hybrid effect. Agglomeration of glass fibre (Figure 3.10d) at high volume fraction of GRP prevents the uniform stress distribution throughout the matrix and this in turn decreases the positive hybrid effect. The negative hybrid effect observed in the case of elongation at break is due to the improved reinforcing effect of sisal fibres in the presence of glass fibres.
3.3. References