CHAPTER 12:

HYBRID COMPOSITES

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1. INTRODUCTION

The incorporation of several different types of fibres into a single matrix has led to the development of hybrid biocomposites. The behavior of hybrid composites is a weighed sum of the individual components in which there is a more favorable balance between the inherent advantages and disadvantages. Also, using a hybrid composite that contains two or more types of fibre, the advantages of one type of fibre could complement with what are lacking in the other. As a consequence, a balance in cost and performance can be achieved through proper material design [1]. The properties of a hybrid composite mainly depend upon the fibre content, length of individual fibres, orientation, extent of intermingling of fibres, fibre to matrix bonding and arrangement of both the fibres. The strength of the hybrid composite is also dependent on the failure strain of individual fibres. Maximum hybrid results are obtained when the fibres are highly strain compatible [2].

The properties of the hybrid system consisting of two components can be predicted by the rule of mixtures.

$$P_H = P_1 V_1 + P_2 V_2 \qquad [1]$$

where P_H is the property to be investigated, P_1 the corresponding property of the first system and P_2 the corresponding property of the second system. V_1 and V_2 are the relative hybrid volume fractions of the first and second system and

$$V_1 + V_2 = 1 \qquad [2]$$

A positive or negative hybrid effect is defined as a positive or negative deviation of a certain mechanical property from the rule of hybrid mixture.

The term hybrid effect has been used to describe the phenomenon of an apparent synergistic improvement in the properties of a composite containing two or more types of fibre [3]. The selection of the components that make up the hybrid composite is determined by the purpose of hybridization, requirements imposed on the material or the construction being designed. The problem of selecting the type of compatible fibres and the level of their properties is of prime importance when designing and producing hybrid composites. The successful use of hybrid composites is determined by the chemical, mechanical and physical stability of the fibre / matrix system.

There are several types of hybrid composites characterized as: (1) interply or tow-by-tow, in which tows of the two or more constituent types of fiber are mixed in a regular or random manner; (2) sandwich hybrids, also known as core-shell, in which one material is sandwiched between two layers of another; (3) interply or laminated, where alternate layers of the two (or more) materials are stacked in a regular manner; (4) intimately mixed hybrids, where the constituent fibers are made to mix as randomly as possible so that no over-concentration of any one type is present in the material; (5) other kinds, such as those reinforced with ribs, pultruded wires, thin veils of fiber or combinations of the above.

The concept of hybrid systems for improved material or structural performance is well-known in engineering design. However, it is the inspiration from natures' own materials that is recently motivating the path towards innovative material and structural designs. Studies on natural materials show how high structural performance can be achieved with non-exotic materials through hybrid combinations assembled in optimized hybrid hierarchical configurations [4].

2. DESIGNING HYBRID COMPOSITES

Although hybrid fiber reinforced polymer composites are gaining interest, the challenge is to replace conventional glass reinforced plastics with biocomposites that exhibit structural and functional stability during storage and use and yet are susceptible to environmental degradation upon disposal. An interesting approach in fabricating biocomposites of superior and desired properties include efficient and cost effective chemical modification of fibre, judicious selection if fibers, matrix modification by functionalizing and blending and efficient processing techniques. (Figure 1)

Another interesting concept is that of “engineered natural fibres” to obtain superior strength biocomposites [5].This concept explores the suitable blending of bast (stem) and leaf fibres. The judicious selection of blends of biofibres is based on the fact that the correct blend achieves optimum balance in mechanical properties for e.g., the combination of bast and leaf fibre is expected to provide a stiffness-toughness balance in the resulting biocomposites.

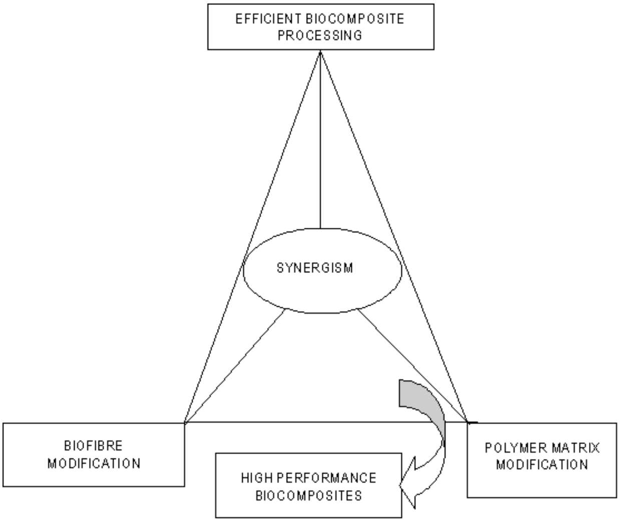


Figure 1. Tricorner approach in designing of high performance biocomposites.

3. HYBRID BIOCOMPOSITES

3.1. Biofiber-Synthetic Fiber Composite

Hybrid biocomposites can be designed by the combination of a synthetic fibre and natural fibre (biofibre) in a matrix and a combination of two natural fibre / biofibre in a matrix. Hybridization with glass fibre provides a method to improve the mechanical properties of natural fibre composites and its effect in different modes of stress depends on the design and construction of the composites [6]. The effect of hybridization of glass fibre in thermoset biocomposites has been discussed in detail [7].

The tensile and impact behavior of oil palm fibre-glass fibre-reinforced epoxy resin was investigated by Bakar et al [8].. The hybridization of oil palm fibres with glass fibres increased the tensile strength, Young's modulus, and elongation at break of the hybrid composites. A negative hybrid effect was observed for the tensile strength and Young's modulus while a positive hybrid effect was observed for the elongation at break of the hybrid composites. The impact strength of the hybrid composites increased with the addition of glass fibres.

The tensile and impact properties of thermoplastic natural rubber reinforced short glass fiber and empty fruit bunch hybrid composites was reported by Anuar et al [9]. The study also focused on the effect of fiber (glass and EFB) treatment using silane and maleic anhydride grafted polypropylene (MAgPP) as a coupling agent. In general, composite containing 10% EFB/10% glass fiber gave an optimum tensile and impact strength for treated and untreated hybrid composites. Tensile properties were found to increase with addition of coupling agent.

Cellular biocomposite cores fabricated from industrial hemp or flax fibres with unsaturated polyester were hybridized with woven jute, chopped glass, and unidirectional carbon fabrics [10]. Material characterization showed improved stiffness, strength, and moisture-absorption stability, while flexural tests on laboratory-scale plates demonstrated enhanced structural behavior. These hybrid cellular biofibre-based composites were found to provide an economic and environmentally friendlier alternative to entry-level synthetic composites.

Mehta et al [11]. have looked into the properties of hybrid hemp/glass fiber reinforced polyester composites. They observed a balance of properties for the hybrid systems when compared to single systems. The interface modification and mechanical properties of newsprint, kraft pulp and hemp fibre reinforced polyolefin composite products was investigated by Sain et al [12]. The effect of a low-molecular weight, maleated type coupling agent, on the mechanical properties of these natural fibre-filled PP composites was also investigated and it was found that the optimum level of the coupling agent was around 3-4 percentage by weight of the composite. Hybrid composites were also produced using 10wt% of glass fiber and 30wt% of hemp fiber and surprisingly showed only a marginal improvement in the mechanical properties.

The hybrid effect of glass fibre and oil palm empty fruit bunch (OPEFB) fibre on the tensile, flexural and impact response of the phenol formaldehyde (PF) composites was investigated by Sreekala et al [13]. Figure 2 shows the variation of izod impact with volume fraction of fibers. With the addition of a small amount of glass fibre, the impact performance of the OPEFB/PF composite was increased by more than 100%. The maximum impact strength is observed for hybrid composites having a 0.74 volume fraction of the OPEFB fibre. The value is higher than glass/PF composites. The fibres play an important role in the impact resistance of the composites as they interact with the crack formation in the matrix and act as stress transferring medium.

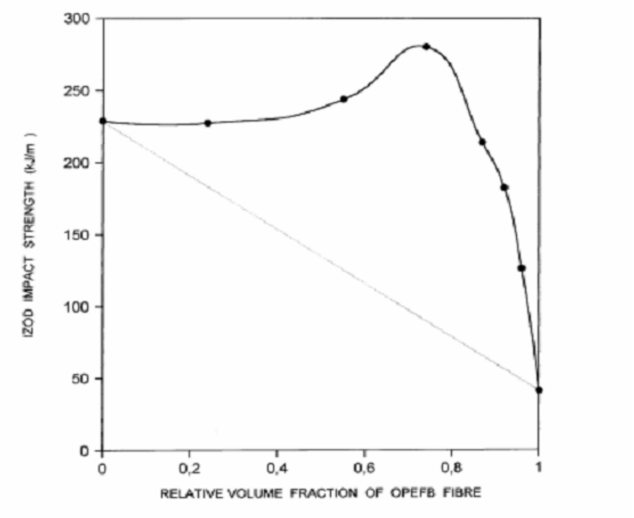


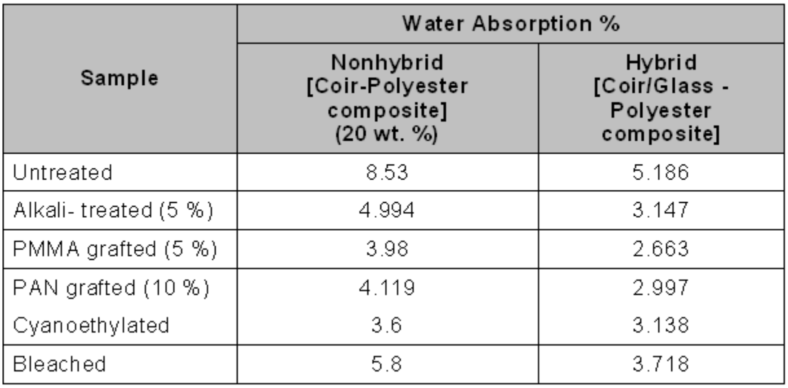
Figure 2. Variations in impact properties of glass IOPEFB hybrid PF composites with relative volume fractions of glass and OPEFB fibre.
[Reference: Sreekala et al. Composites Science and Technology 62 339— 353 2002]

Hybridization with a synthetic fiber also has a profound effect on the water absorption property of composites. Mishra et al [14] studied the moisture uptake characteristics of hybrid composites. The systems chosen were sisal / glass and pineapple /glass fibre reinforced polyester composites. Composites were prepared by varying the concentration of glass fibre and by subjecting the bio-fibres to different chemical treatments. The authors observed that water uptake of hybrid composites were less than that of unhybridized composites.

A comparative study of the water absorption of the glass fibre (7 wt. %) / natural biofibre (13 wt. %) with that of non-hybrid composites is given in Table 1. A lowering in water absorption is evident [15].

Table 1 Comparative study of the water absorption of the glass fibre / natural biofibre with that of non-hybrid composites

[Reference: Rout J. et al., Composites Science & Technology 61, 1303, 2001]



Sample	Water Absorption %	
	Nonhybrid [Coir-Polyester composite] (20 wt. %)	Hybrid [Coir/Glass - Polyester composite]
Untreated	8.53	5.186
Alkali- treated (5 %)	4.994	3.147
PMMA grafted (5 %)	3.98	2.663
PAN grafted (10 %)	4.119	2.997
Cyanoethylated	3.6	3.138
Bleached	5.8	3.718

In an interesting study, the mechanical reinforcement obtained by the introduction of glass fibers in biofiber (silk fabric)-reinforced epoxy composites was investigated by Padma Priya and Rai [16]. The addition of a relatively small amount of glass fabric to the silk fabric reinforced epoxy matrix enhanced the mechanical properties of the resulting hybrid composites. It was also observed that the properties increase with the increase in the weight fraction of reinforcement. The water uptake of hybrid composites was found to be less than that of unhybridized composites.

The hybridization of palmyra fiber waste with glass fiber in polyester matrix was reported by Velmurugan and Manikandan [17]. Samples were prepared by sandwiching the fiber waste between chopped strand glass fiber mats by varying both glass fibers and waste material content, keeping total fiber content as 60% by weight. Mechanical properties of the composites were found to increase with increase in the amount of glass fiber in the hybrid. Hybrid composites containing more of waste fiber showed good reinforcement effect than the composites reinforced with more of glass fiber.

The viscoelastic properties of oil palm fibre/glass hybrid phenol formaldehyde composites as a function of fibre content and hybrid fibre ratio was investigated by Sreekala et al [18]. The incorporation of oil palm fibre shifted the glass transition towards lower temperature value. The glass transition temperature of the hybrid composites was found to be lower than that of the unhybridized composites.

Storage modulus of the hybrid composites was also found to be lower than unhybridized oil palm fibre/PF composite.

In an interesting report, the mechanical and thermo-mechanical properties of the hybrid composites from poly (lactic acid) PLA/Talc/Recycled newspaper fiber were investigated and compared with that of composites from either of the reinforcement [19]. The tensile, flexural and impact strength of these hybrid composites were found to be significantly higher than that made from either PLA/recycled newspaper fiber or PLA/talc. The hybrid composites exhibited encouraging properties with a flexural strength value of 94 MPa and flexural modulus of 10.8 GPa while unhybridized PLA/recycled newspaper fiber based composites exhibited flexural strength and modulus values as 77 MPa and 6.7 GPa respectively.

In an interesting report [20], hybrid blends of natural and synthetic fibers were fabricated and found to significantly improve the characteristics of biocomposites with minimal cost and environmental impact. It was observed that hierarchical cellular designs can maximize material efficiency in structural components. Periodic and hierarchical cellular plate designs made from natural fibers and unsaturated polyester resin was evaluated experimentally and analytically. Stiffness, strength, and dimensional stability of all-biocomposite and hybrid natural–synthetic material systems were evaluated through material tests while structural performance of cellular plate designs was assessed through flexural tests on laboratory-scale samples. The experimental results were correlated with analytical models for short-fiber composites and cellular structures. The results showed that biocomposites have adequate short-term performance and that they can efficiently compete with housing panels made from conventional structural materials.

Figure 3 illustrates the performance of cellular biocomposite panels against conventional systems used for building and residential construction, namely a pre-cast pre-stressed hollow core concrete slab (PC/PS HC slab), a precast prestressed solid concrete slab (PC/PS solid slab) and an oriented wood strand board insulated structural panel (ISP). All panels were assumed to have the same overall dimensions: The comparison shows that while the bio-panel is out-performed by the PC/PS HC slab, it effectively competes with the solid PC/PS slab and it considerably outperforms the ISP board.

The effect of replacing glass fibers with flax fibers in composites and its influence on impact performance was investigated by Santulli et al [21]. Figure 4 shows the effect of impact resistance on flax/epoxy, glass/epoxy and hybrid system. Previous results in literature suggested that untreated plant fibers are more prone to debonding in correspondence with defects, and this reduces their resistance to repeated impacts. In Figure 5 the vertical crack in the matrix appears to lead to efficient stress redistribution in the flax core, so that the energy dissipated is sufficient in this case (impact at 50 J) to hinder damage propagation in the whole of the sandwich. Conclusive observations point to the fact that the flax fiber reinforced core in the hybrid laminates showed an appreciable action of impact damage dissipation. Flax-epoxy laminates and hybrid E-glass/epoxy-flax/epoxy laminates provided a sufficient impact performance with a considerable weight reduction with respect to fiberglass.

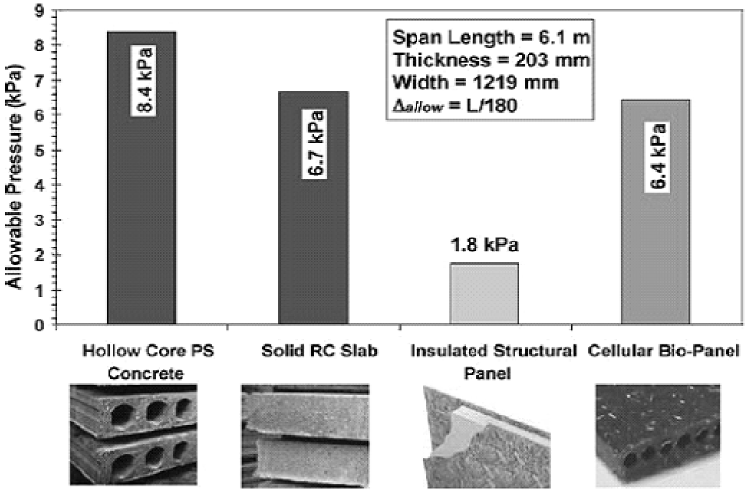


Figure 3. Performance of cellular biocomposite panels against conventional slab and panel systems for commercial and residential construction
[Reference: Burgueno et al. Journal of Polymers and the Environment 13 2,2005]

The novel development of a photofabrication process of biofibre composites, based on oil palm empty fruit bunch fibres was recently reported [22]. The process consisted of the following steps: (1) the preparation of nonwoven mat of biofibre, either alone or in combination with glass and nylon; (2) drying the mat; (3) preparation of photocurable resin matrix, consisting of vinyl ester and photoinitiator; (4) impregnation of the mat by photocurable resin; and (5) irradiation of the impregnated mat by UV radiation to effect the cure of the composite. Oil palm fibre, glass, and nylon fibres were mixed in different proportions. A ˝mixture experimental design" was used to generate experimental compositions of the reinforcing fibres and to model dependency of the response variables on the components through mathematical relationships.

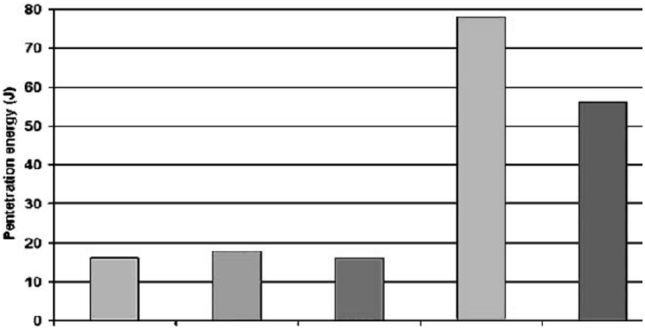


Figure 4. Comparison of impact resistance in different laminates
[Reference: Santulli et al. Journal of Materials Science 40 3581 —3585 2005]

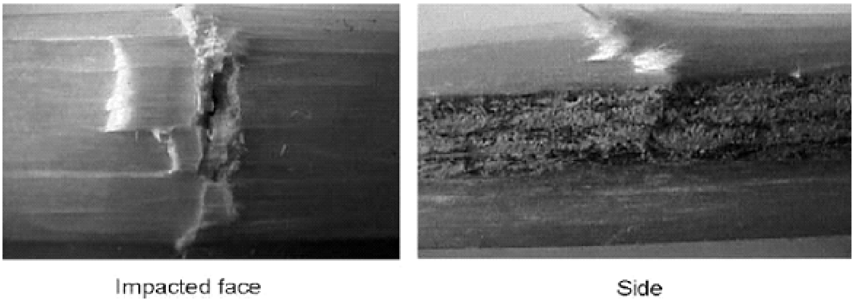


Figure 5. Impact damage in E-glasslflax fiber reinforced hybrid laminate
[Reference: Santulli et al. Journal of Materials Science 40 3581 — 3585 2005]

Scientists [23] at the Affordable Composites from Renewable Resources (ACRES) program at the University of Delaware investigated the mechanical properties of glass/flax hybrid composites based on a novel modified soybean oil matrix material. Composites with different glass/flax ratios and different fibre arrangements were made using a modified soybean oil matrix material. The fibre arrangement was varied to make symmetric and unsymmetric composites. The latter were tested in different modes in flexural tests and drop weight impact tests. The mechanical properties of the composites were found to depend upon the glass/flax ratio and the arrangement of fibres in the composite. On proper selection of the arrangement of fibres in the composite, the glass fibres and flax fibres were found to act synergistically resulting in an improved flexural and impact performance.

Li et al [24]. developed and characterized a super-hybrid (natural composite/fibre-reinforced composite/metal hybridization) eco-material, reformed bamboo/glass fabric/aluminum (RB/GF/Al). The addition of a sparse glass fabric/epoxy resin layer between reformed bamboo and aluminum proved to be effective in increasing the compressive tensile strength of the composite material. In particular, the interfacial shear strength between the reformed bamboo and aluminum was improved.

The influence of chemical modifications on the tensile properties of intimately mixed short sisal/glass hybrid fibre reinforced low density polyethylene composites was investigated by Kalaprasad et al [25].Chemical surface modifications such as alkali, acetic anhydride, stearic acid, permanganate, maleic anhydride, silane and peroxides were given to the fibres and matrix and were found to be successful in improving the interfacial adhesion and compatibility between the fibre and matrix. It was found that the extent of improvement in tensile properties of SGRP varied with respect to the nature of chemical modifications between fibre and matrix. Improved mechanical anchoring and physical and chemical bonding between fibre and polyethylene matrix are the reasons for superior tensile strength and Young's modulus in treated composites. Among the various chemical modifications, the best tensile strength and modulus was exhibited by composite containing benzoyl peroxide treated fibres. This is attributed to the peroxide-initiated grafting of polyethylene on to the fibres.

Idicula et al [26]. have investigated the thermophysical properties of pineapple (PALF)/glass fiber composites as a function of fiber loading and chemical treatments. The results show that chemical treatment of the fibres reduces the composite thermal contact resistance. Hybridisation of natural fibre with glass fibre was found to increase heat transport ability of the composite. This is depicted in Table 2 which shows the thermophysical and density measurements of PALF/glass fibre hybrid composites. There is an increasing of thermal diffusivity and density values with the glass fibre volume fraction. In fact, the glass fibre has a high value of thermal conductivity, diffusivity and density compared to the PALF fibre.

Table 2: Thermophysical and density measurements of PALF/glass fibre hybrid composites

[Idicula et al. Composites Science and Technology 66 2719–2725 2006]

	Thermal Conductivity k (W m^{-1} K^{-1})	Thermal Diffusivity a (m^{2}s^{-1}) 10^{-7}	Specific heat Cp (J kg^{-1} K^{-1})	□ (kg m^{-3})
0.40 V_f PALF	0.184 ± 0.003	1.60 ± 0.25	979 ± 155	1175 ± 155
0.36 V_f PALF + 0.04 V_f glass fibre	0.198 ± 0.002	1.64 ± 0.20	972 ± 121	1243 ± 121
0.20 V_f PALF + 0.20 V_f glass fibre	0.216 ± 0.003	1.68 ± 0.20	925 ± 113	1390 ± 113
0.40 V_f glass fibre	0.277 ± 0.003	2.14 ± 0.24	798 ± 92	1622 ± 92

3.2. Biofiber- Biofiber Composite

Another innovative approach to hybrid composites is the incorporation of two natural fibres in a matrix system. The mechanical performance of short randomly oriented banana and sisal hybrid fibre reinforced polyester composites [27] was investigated with reference to the relative volume fraction of the two fibres at a constant total fibre loading of 0.40 volume fraction (V_f), keeping banana as the skin material and sisal as the core material. A positive hybrid effect was observed in the flexural strength (Figure 6) and flexural modulus of the hybrid composites. The tensile strength of the composites showed a positive hybrid effect when the relative volume fraction of the two fibres was varied, and maximum tensile strength was found to be in the hybrid composite having a ratio of banana and sisal 4 : 1.

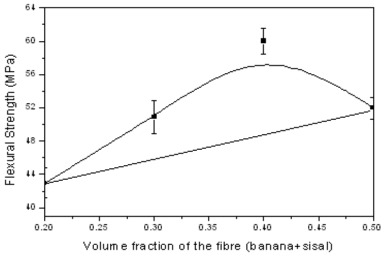
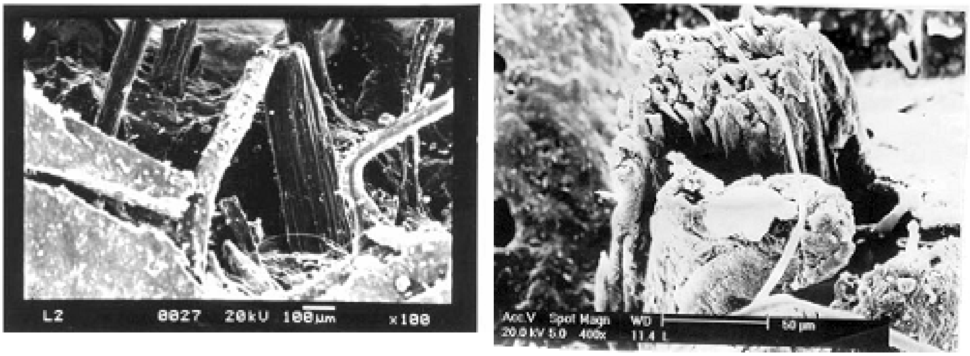


Figure 6: Variation of flexural strength with volume fraction of fibre
[Reference: Idicula M., et al. J. Appl. Polym. Sci. 96, 5, *1699,* 2005]

As a continuation to the above study the dynamic and static mechanical properties of randomly oriented intimately mixed banana and sisal hybrid fibre reinforced polyester composites were reported [28]. Maximum stress transfer between the fibre and matrix was obtained in composites having volume ratio of banana and sisal as 3:1. The storage modulus was found to increase with fibre volume fraction above the glass transition temperature of the composites.

In an interesting study [29], hybrid biocomposites from natural fibers were fabricated by a novel high volume processing technique named ‘biocomposite sheet molding compound panel’ (BCSMCP) manufacturing process. The natural fibers used were jute and hemp and were found to have promising impact and thermal properties.

In an innovative study, a unique combination of sisal and oil palm fibres in natural rubber has been utilized to design hybrid biocomposites. It was seen that the incorporation of fibres resulted in increased modulus [30]. Chemical modification of both sisal and oil palm fibres was imperative for increased interfacial adhesion and resulted in enhanced properties [31]. Figures 7 a and b represents the untreated and alkali treated (4%) sisal-oil palm hybrid fiber reinforced natural rubber composites.



a b

Figures 7: SEM micrographs of fracture surfaces

The viscoelastic [32,33] , biodegradation [34], water sorption [35], dielectric [36] and stress relaxation [37] characteristics were also studied.

Researchers have also designed novel rubber biocomposites by using a combination of leaf and fruit fibre in natural rubber [38,39]. The incorporation of sisal and coir fibre in NR was seen to increase the dielectric constant of the composites. These hybrid biocomposites were found to have enormous applications as antistatic agents. The researchers also used the swelling technique to estimate interfacial adhesion was reported in the case of sisal / coir fibre reinforced natural rubber composites [40]. The bonding agent added mixes showed enhanced restriction to swelling and it was seen that the ratio of change in volume fraction of rubber before and after swelling to the volume fraction of rubber before swelling (V_o - V_r/V_0) was lower for bonding agent added composites, when compared to an unbonded one.

In another interesting study, the static and dynamic mechanical properties of a kenaf fiber-wood flour/polypropylene hybrid composite was investigated by Mehdi [41]. The hybrid composite exhibited tensile and flexural moduli and strength values closer to those of the kenaf fiber [KF] reinforced composite, which indicated a higher reinforcing efficiency of kenaf fiber compared with wood flour [WF]. DMA studies revealed that although the glass-transition temperature remained unchanged by the replacement of half of the WF by KFs, the α-transition temperature of the hybrid composite was identical to that of WF composite.

3.3. Hybrid Textile Biocomposites

The development of textile technologies such as weaving, knitting and braiding has resulted in the formation of composites that have superior mechanical properties, as continuous orientation of fibres is not restricted at any point. In applications where more than one fibre orientation is required, a fabric combining 0° and 90° fibre orientations is useful. Woven fabrics are produced by the interlacing of warp (0°) fibres and weft (90°) fibres in a regular pattern or weave style. The fabric's integrity is maintained by the mechanical interlocking of the fibres. Drape (the ability of a fabric to conform to a complex surface), surface smoothness and stability of a fabric are controlled primarily by the weave style.

Researchers have looked into tensile strength of ramie-cotton hybrid fibre reinforced polyester composites [42] .They observed that tensile behaviour was dominated by volume fraction of ramie fibres aligned in the test direction. The fabric and diameter of the thread did not play any role in tensile characteristics. Cotton fabric was found to have minor reinforcement effect due to weak cotton/polyester interface. Similar studies were performed by Mwaikambo and Bisanda [43] on kapok- cotton fibre reinforced polyester composites.

Novolac type phenolic composites reinforced with jute/cotton hybrid woven fabrics were fabricated and its properties were investigated as a function of fibre orientation and roving/fabric characteristics [44]. Results showed that the composite properties were strongly influenced by test direction and rovings/fabric characteristics. The anisotropy degree was shown to increase with test angle and to strongly depend on the type of architecture of fabric used, i.e., jute rovings diameter, relative fibre content, etc. The best overall mechanical properties were obtained for the composites tested along the jute rovings direction. Composites tested at 45° and 90° with respect to the jute roving direction exhibited a controlled brittle failure combined with a successive fibre pullout, while those tested in the longitudinal direction (0°) exhibited a catastrophic failure mode. The researchers are of the opinion that jute fibre promotes a higher reinforcing effect and cotton fibre avoids catastrophic failure. Therefore, this combination of natural fibres is suitable to produce composites for lightweight structural applications.

The thermal diffusivity, thermal conductivity and specific heat of jute/cotton, sisal/cotton and ramie/cotton hybrid fabric-reinforced unsaturated polyester composites were investigated by Alsina et al [45]. These properties were measured both parallel and perpendicular to the plane of the fabrics. The results obtained show that higher values were obtained parallel to the plane of the fibres. Sisal/cotton composites showed a particular behavior, with thermal properties very close to those of the resin matrix. The thermal properties of the fabrics, i.e. without any resin, were also evaluated and were used to predict the properties of the composites from the theoretical series and parallel model equations. The effect of fabric pre-drying on the thermal properties of the composites was also evaluated. The results showed that the drying procedure used did not bring any relevant change in the properties evaluated.

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