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Mechanical behaviour of particulate hybrid composite laminates as potential building materials

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Sisal fibres and red mud were treated with different hydrophobic chelates based on titanium and silicon. The effect of the addition of treated red-mud loading on the mechanical properties of polyester resin has been investigated. Composite laminates have been prepared and their physico-mechanical properties were evaluated as a function of fibre length, fibre content, sisal/glass fibres and sisal/glass/red mud combinations. It was found that particulate hybrid composites had superior physico-mechanical properties than those of sisal/polyester, neat and filled unsaturated polyester resin. The suitability of particulate composites was also assessed in relation to medium-density fibreboard/high-density particleboard.

Keywords: hybrid composite laminates; building materials; fibre reinforcement

The use of natural-fibre-reinforced polymeric building materials has been strongly favoured to promote the utilization of indigenous materials in the developing countries in view of their low cost, easy availability, saving in energy and pollution-free production. Natural fibres are much cheaper than glass fibres and could replace them in applications where cost consideration outweighs strength requirements. However, natural fibres have not been widely used commercially to reinforce more modern resins such as unsaturated polyester, epoxy, etc., despite extensive research. The major problems associated with the applications of natural fibres in the composite industry are poor wettability, high moisture absorption and susceptibility to environmental degradation. In such composites, delamination due to moisture absorption leads to weakening of the interfacial bond, thus causing a reduction in the mechanical properties of the composites. In an effort to overcome some of these disadvantages, several attempts have been made to modify the surface of these fibres either by imparting hydrophobicity[1-5] or by applying a gelcoat/glass fibres as a surfacing layer[6] to the composites. The work carried out by Winfield[7] in India and Bangladesh on jute-reinforced polyester systems for the construction of cheap primary school buildings has been considered to be a major breakthrough in this field. Considerable work[8-14] has also been attempted to develop plastic composite products based on natural fibres for various applications such as roofings, panellings, food grain silos and other low-cost housing units. Work on natural-fibre-reinforced polymer composites was initiated at the Central Building Research Institute (CBRI), Roorkee, with the main objectives of finding cost-effective applications for use in different Indian climatic conditions. This paper describes the systematic work carried out so far on particulate hybrid composites based on glass/sisal fibres, red-mud filler, and unsaturated polyester resin.

Experimental

Materials

Sisal fibres were obtained from a local market (Roorkee), and their physico-chemical, mechanical and surface topographic characteristics are as given in *Table 1* and *Figure 1* respectively. Glass mat (E. glass, weighing 450 g m^{-2}) was supplied by Pilkington Fibre Glass Ltd, India. The sample of red mud (industrial by-product) used in the experiments was collected from the Indian Aluminium Company (Indalco, India). Its chemical analysis is given in *Table 2*. Unsaturated polyester resin (HSR 8131) was received from Bakelite Hylam Ltd, India. Coupling agents such as gamma-Methacryloxypropyltrimethoxy silane, A-174 (Union Carbide, USA), and neopentyl (diallyl)oxy, tri (dioctyl) pyrophosphatotitanate, LICA-38 (Kenrich, USA), were used as supplied.

Fibre/filler coating procedure

Red-mud coating

Red mud was first calcined at 200 °C for 24 hours and washed with distilled water. The sample was dried at

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Table 1 Physico-chemical and mechanical properties of sisal fibre

No.	Properties	Value
A	Physical properties	
	1 Specific gravity	1.10
	2 Fibre diameter (mm)	0.08–0.276
	3 Fibre length (mm)	50–150
	4 Moisture content (%)	12.77
	5 Water absorption (%)	123.73
B	Chemical composition	
	1 Cellulose (%)	66–78
	2 Lignin (%)	8–10
	3 Hemicellulose (%)	10–12
	4 Ash (%)	1.0
C	Mechanical properties	
	1 Ultimate tensile strength (N mm^{-2})	280–370
	2 Elongation (%)	3–5
	3 Modulus of elasticity (GPa)	34–62

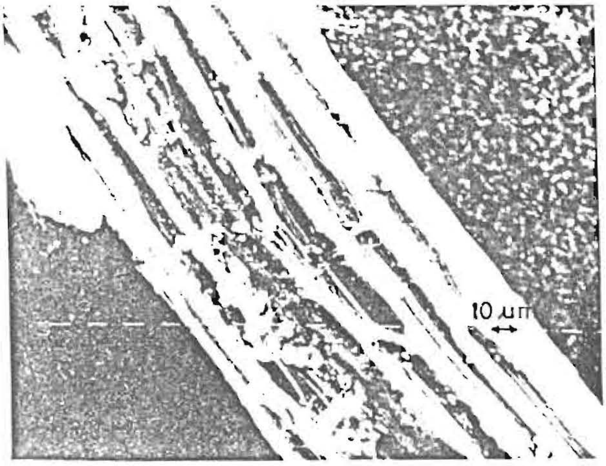

Figure 1 Scanning electron micrograph of sisal fibre showing surface texture

Table 2 Chemical analysis of red mud

No.	Constituent	%
1	Silica (SiO_2)	6–10
2	Alumina (Al_2O_3)	19–26
3	Ferric oxide (Fe_2O_3)	23–31
4	Titanium oxide (TiO_2)	20–27
5	Lime (CaO)	2–4
6	Alkalis (Na_2O, K_2O)	4–7
7	LOI (Loss Of Ignition)	8–11

100 °C for 24 hours and stored in a desiccator before use. Titanate treatment of the red mud was carried out from a 1% (by weight of filler) solution of coupling agent in toluene. The slurry was agitated for 2 hours and filtered on a Buchner filter. The sample was then washed with toluene to remove unreacted coupling agent and dried at 80 °C for 4 hours to complete the condensation process.

Fibre coating

Sisal fibres were washed with distilled water for 24 hours to ensure the complete removal of water solubles from the surface and dried at 60 °C in an oven for 8 hours. The washed fibres were immersed in a 1% (by weight of fibre) coupling agent (A-174) solution in toluene and kept for vigorous stirring for 30 minutes. After treatment with a coupling agent, the fibres were washed with toluene to remove compounds not covalently bonded to the fibres and dried at 80 °C for 4 hours in an oven to constant weight.

Composites preparation

Red mud/polyester system

Treated red mud was added to styrenated polyester and stirred with a mechanical stirrer for 90 minutes; 1.5% methyl ethyl ketone peroxide and 1.5% cobalt naphthenate were used as a catalyst and accelerator respectively. The mixture was slowly rotated to prevent sedimentation until polymerization rendered it viscous. It was then poured into a metal mould of 4 mm thickness, allowed to polymerize for 24 hours and then postcured for 4 hours at 80 °C. PVA1c was used as the mould releasing agent.

Laminates preparation

Treated sisal fibres were cut to the desired length and then accommodated at random to form 30 × 30 cm^2 non-woven mats using polyvinyl acetate emulsion as a binding agent. Normally, the mats have 2–3% moisture content and a weight of 440 g m^{-2}. To make polyester laminates, conventional hand lay-up techniques were employed. These sisal mats were then impregnated with unsaturated polyester resin containing 1.5% methyl ethyl ketone peroxide and 1.5% cobalt naphthenate. Stacks of different impregnated fibre mats were placed between two brass plates. PVA1c was used as the mould releasing agent. The laminates were pressed on a hydraulic press at room temperature for 2 hours at a pressure of 2 MPa. Similarly particulate hybrid laminates were prepared by applying optimized red-mud-polyester mix on the glass/sisal mats (30 × 30 cm^2). Glass mat was used only on the outer surfaces to sandwich the sisal fibre mats. Curing was done at room temperature for 24 hours and then postcuring at 80 °C for 4 hours at a pressure of 1.5–2 MPa.

Test methods

Physical properties such as density and void content were determined in accordance with ASTM D 792-66 and ASTM D 2734-70 respectively. Water absorption was measured as per ASTM D 570-81. Tensile properties of dumb-bell-shaped samples were determined according to ASTM D 638-76 at a cross-head speed of 5 mm min^{-1}. Flexural strength was measured as per ASTM D 790-71. A cross-head speed of 5 mm min^{-1} and span-to-depth ratio of 16:1 was used for the red mud polyester and fibre laminates. A simple accelerated test was performed by immersing the samples into boiling water for 2 hours as per IS 12406-88.

Results and discussion

Effect of red-mud loading

The tensile and flexural strength of particulate composites as a function of red-mud loading are shown in *Figure 2*. On increasing the red-mud concentration, the tensile strength decreases rapidly up to 15% and then slowly with further addition (up to 45%). This behaviour may be attributed to the failure of the matrix at lower stresses and maximum stress concentration around the red mud. In flexure, there was no significant change in strength up to 20% red-mud addition as expected because of an effective interfacial adhesion between the red mud and polyester and higher compressive strength of the red mud, but on further loading of red mud in polyester, the flexural strength decreases from 62 MPa for neat resin to 41 MPa for 45% red-mud-filled composite. The net reduction in tensile strength is about 43% for 45% red-mud-filled composite with respect to the neat resin while the drop in flexural strength was about 34% for the same. Interfacial adhesion is more effective in flexure mode than in tension. Similar results have also been reported for other particulate composites[15-17].

Effect of fibre loading

Figure 3 shows a plot of tensile strength as a function of fibre length of sisal-fibre-reinforced polyester composites. As was expected, the composite containing 3 cm fibre length imparted maximum tensile strength as compared with the other composites made from higher fibre length. There was no evidence of fibre pull-out in the fracture surfaces. This suggested the reason that bonding between the fibres and polyester might be greater than optimum. The void content of laminates made from 3 cm fibre length was roughly twice that of the others, which may affect the mechanical properties and performance in wet conditions.

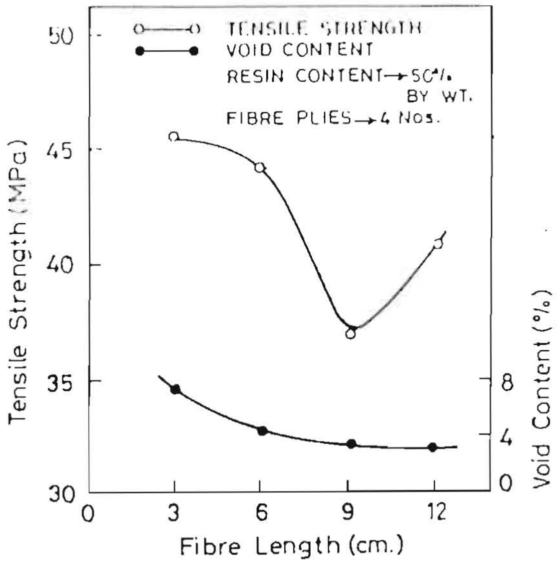

Figure 3 Variation of tensile strength and void content with fibre length of sisal polyester composites (sisal fibre ply weight = 440 g m^{-2})

As seen in *Figure 4* the tensile results indicate a maximum followed by a downward trend. This implies that there is an optimum fibre content (50% by weight) of the laminate above which the tensile properties degrade. An improvement of about 71% in tensile strength was observed for 50% (by weight) fibre content with respect to neat resin. This fact indicates that applied stress is transferred from the matrix to the stronger fibres by shear stress at the fibre–matrix interface. In the absence of bonding, the fibres would act as voids. No significant reinforcement was noticed up to 20–25% fibre content. The void content remains unaltered up to 50% fibre content and then increases rapidly with further loading of fibres. In flexure, the relationship between strength and fibre content appears to be linear (*Figure 4*). Increasing the number of fibre plies brings an increase in tensile and flexural properties of the laminates. This effect is expected because failure of a critically stressed layer in the composite does not necessarily mean that total catastrophic failure of the multilayered composite takes place[5].

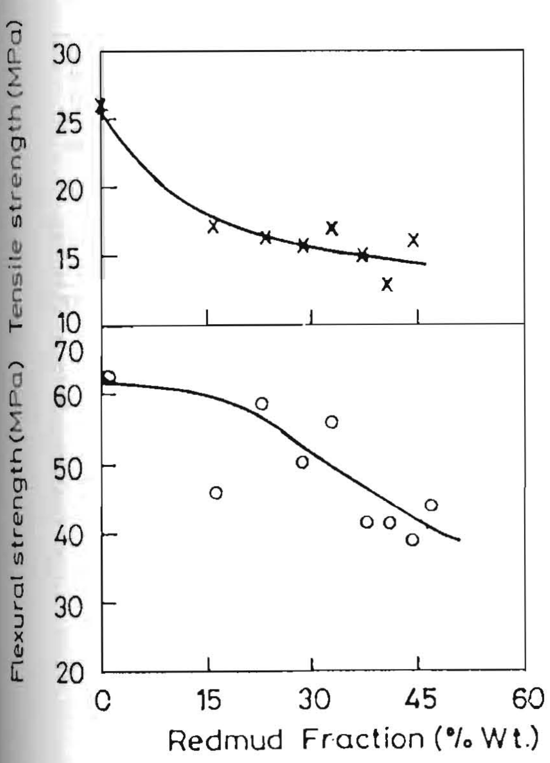

Figure 2 Effect of red-mud loading on flexural and tensile strength of particulate composites

Properties of particulate hybrid composites

Particulate hybrid composites were prepared with the help of 20% red mud, 34% sisal fibres, 8% glass fibres and unsaturated polyester resin, and their physico-mechanical properties were compared with unsaturated

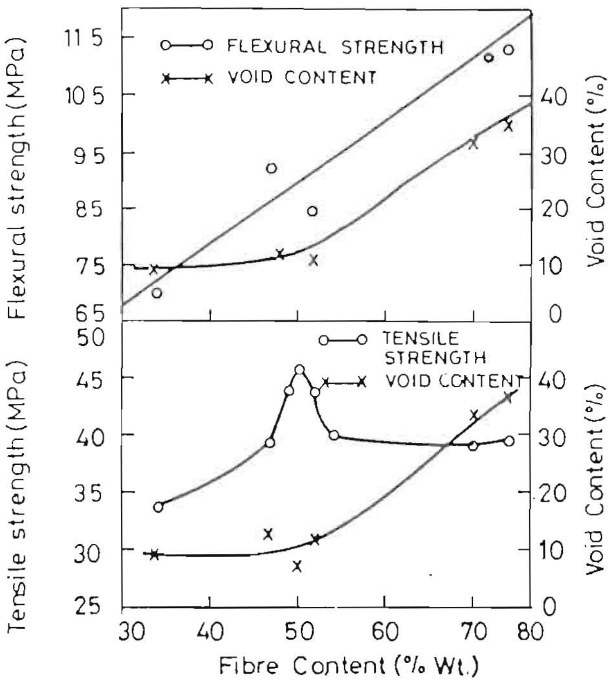

Figure 4 Tensile and flexural strength as a function of fibre content of sisal polyester composites (three plies = 34% FC, four plies = 48% FC, five plies = 70% FC, six plies = 74% FC; sisal fibre ply weight = 440 g m^{-2})

polyester resin and other reinforced composites as shown in *Table 3*. It was found that the tensile and flexural properties of hybrid/particulate hybrid composite increased, probably due to the inherent better properties of glass fibres. The hybrid composite containing a small amount of glass fibres seems to fail by rupture of the sisal fibres followed by that of the glass fibres. An improvement of 15% in flexural strength for particulate hybrid composites was observed over that of sisal/polyester composites. It can be seen from *Table 3* that the properties of particulate hybrid composite are inferior to those of glass-fibre-reinforced plastics (GRP).

The suitability of particulate hybrid composites was also assessed and their physico-mechanical properties were compared (*Table 4*) with medium-density fibreboard and high-density particleboard, satisfying the requirements laid down by the Bureau of Indian Standards (IS 12406-88 and IS 3478-66).

Effect of water on physico-tensile properties

The percentage weight gain as a function of immersion time is shown in *Figure 5*. As is to be expected, laminates containing a substantial number of voids absorb water more readily than those which do not. It is obvious that at high fibre loading, the water uptake will be significant. The dimensions (swelling in thickness) of the specimens also change during immersion in water

Table 3 Comparative physico-mechanical properties of USP and composites

No.	Material	Reinforcement content Red mud (% by wt)	 Sisal fibre (% by wt)	Density (g cm^{-3})	Water absorption 24 h (%)	Swelling in thickness 24 h (%)	Tensile strength (MPa)	Elongation (%)	Mod. of elasticity (GPa)	Flexural strength (MPa)
1	USP	–	–	1.12	0.130	Nil	26.40	2.38	1.91	61.26
2	RM/USP	20	–	1.31	0.203	Nil	17.30	1.49	0.97	58.79
3	SRP	–	50	1.10	8.03	13.19	45.10	2.33	3.11	85.03
4	S/RM/USP	20	38	1.086	4.71	0.58	23.50	7.69	0.64	40.02
5	GRP (wt % 28–32)	–	–	1.6	1.03	–	76.00	–	41.00	178.40
6	S/GRP (G. cont.–15%)	–	46	1.13	4.12	3.03	65.20	11.30	–	89.20
7	S/G/RM/USP (G. cont.–8%)	20	34	1.24	3.65	Negligible	45.20	8.10	5.95	98.10

USP = unsaturated polyester, RM/USP = red mud–polyester, SRP = sisal-reinforced polyester, S/RM/USP = sisal/red mud/polyester, GRP = glass-reinforced polyester, S/GRP = sisal/glass-reinforced polyester, S/G/RM/USP = sisal/glass/red mud/polyester

Table 4 Comparative physical and mechanical properties of particulate hybrid composite, medium-density fibreboard and high-density particleboard

No.	Properties	Required value Medium-density fibreboard (IS 12406-88)^a	 High-density particleboard (IS 3478-66)^b	Particulate hybrid composite
1	Bulk density (kg m^{-3})	500–900	900–1200	1120
2	Moisture content (%)	5–15	5–10	2–3
3	Water absorption 24 h (%)	12–18	10–25	3.65
4	Swelling in thickness 24 h (%)	4–7	–	Negligible
5	Modulus of rupture, min (N mm^{-2})	28	39–44	98.10
6	Tensile strength parallel to surface (N mm^{-2}):			
	(i) dry	–	29–34	45.20
	(ii) 2 h boiling water	–	No sign of delamination	38.20

^{a}Indian Standard Specification for medium-density fibreboards for general purposes
^{b}Indian Standard Specification for high-density wood particleboards

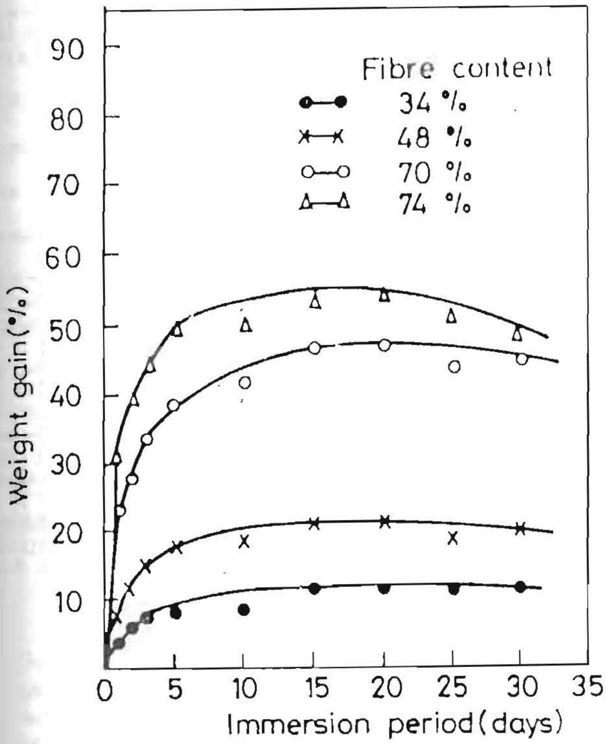

Figure 5 Variation of water uptake versus immersion time in water of sisal/polyester composites

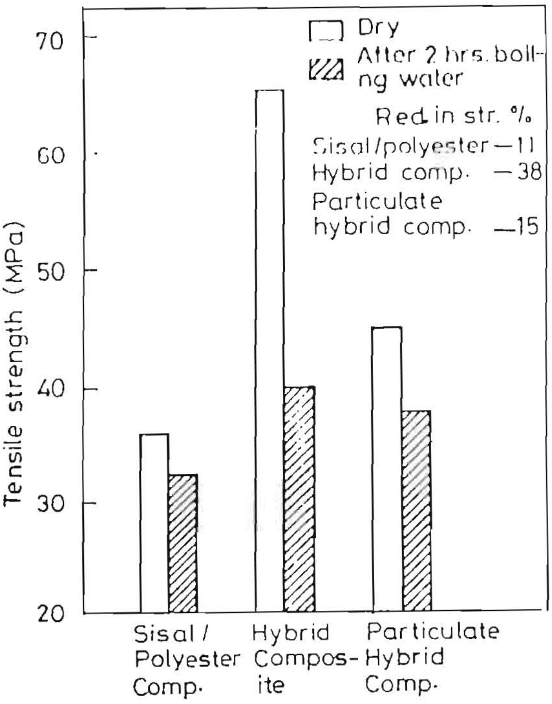

Figure 7 Reduction in tensile strength after 2 h immersion in boiling water of different fibre-reinforced composites

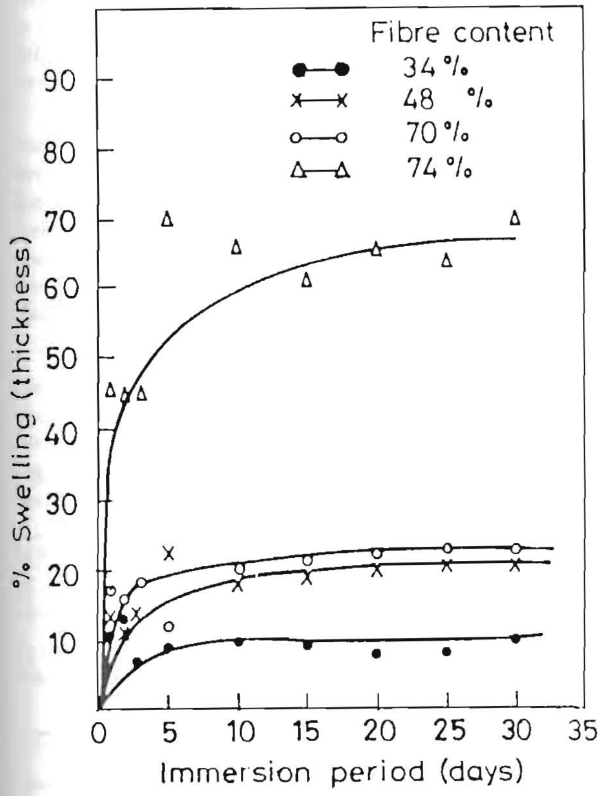

Figure 6 Percentage increase in swelling thickness versus immersion period in water of sisal/polyester composites

(*Figure 6*). The maximum swelling was observed in six-plies laminates containing 74% fibre content.

The tensile strengths of sisal/polyester, hybrid and particulate hybrid composites were evaluated before and after exposure for 2 hours in boiling water (*Figure 7*). It can be seen that a considerable reduction in strength (11–38%) was observed owing to the combined effect of water and temperature and also plasticization of the resin. The partial separation of fibre plies from each other was noticed after exposure owing to the hydrolysis of the polyester resin (hybrid composite reduction in strength 38%). However, the percentage reduction of tensile strength in sisal/polyester was unexpectedly less than that of the other laminates for some unknown reason. The reduction in mechanical properties under wet conditions could be avoided by the development of covalent bonds between the fibre and the matrix.

Conclusions

This work shows that particulate hybrid composite made from sisal fibres and industrial waste is a potential candidate to impart superior physico-mechanical properties compared with sisal-fibre-reinforced composites and other commercially available medium-density fibreboard and high-density particleboard. It can be considered as a potential building material used for partitions, wall panels, claddings, etc. It is therefore suggested that further research is needed to develop an appropriate technology for its large-scale production.

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