

Surface Modification to Improve the Impact Performance of Natural Fibre Composites

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SUMMARY: Natural fibre-nonwoven -mats made of jute, hemp and flax were used for the study. Composite plates of jute/epoxy, hemp/epoxy and flax/epoxy at different volume fractions were prepared by autoclave moulding technique. In jute/epoxy composites, it was found that the impact strength decreases with increasing fibre volume fraction. There was an increase of about 60% in strength on increasing the fibre volume fraction from 0.15 to 0.22 in hemp/epoxy composites. But in the case of flax composites the increase was marginal. Flax fibres were subjected to different chemical treatments such as alkali, isocyanate and silane. In order to improve the technical fibre strength, the fibre is impregnated with the matrix resin. To reduce the surface cracking and to improve the impact strength, latex modification on the fibres was tried. Fibre surfaces were characterised by SEM and FTIR. Impact fracture surfaces were examined using scanning electron microscopy. Morphological studies indicated that a coating is developed on the fibre after treatment and as a result surface roughness is increased.

KEYWORDS: impact properties, coupling agent, characterisation, interface modification, natural fibre, flax, jute, hemp .

INTRODUCTION

In recent years, composites made of natural fibres have received increasing attention in light of the growing environmental awareness. Especially with respect to the specific strength and specific stiffness, natural fibres can compete with glass fibres. The advantages of these natural fibres are their biodegradability, combustibility, light weight, non-toxicity, decreased environmental pollution, low cost and recyclability. Moreover, natural fibre composites have a low density about half of that glass fibre composites and are less abrasive. Their utilisation in composite materials gives good opportunities for the enhancement of agricultural by-products. N.F.C are 100% combustible without the production of toxic gases nor solid residues. But their high level of moisture absorption, poor wetting and insufficient adhesion between untreated fibres and the polymer matrix leads to debonding and significant degradation after aging. A natural fibre is in fact a composite itself. Formation of mechanical and

chemical bonding at the interface is mainly dependent on the surface morphology of the fibres, chemical composition of fibres and polarity or presence of reactive functional groups in the matrix resin. Therefore microscopic analysis of the fibre surface morphology is important in fibrous composites.

Natural fibres are amenable to modifications as they consist of several hydroxyl groups in the cellulose and lignin. Chemical modifications may activate these groups or introduce new functional groups which can effectively interact with the matrix. Chemical bleaching of fibres may lead to major changes in the fibre surface roughness. The irregularities of the fibre surface play an important role in the mechanical interlocking at the interface. Usually the effect of this fibre surface modification is characterised by its influence on the mechanical and thermal properties of the composites. Jindal [1] studied the impact properties of bamboo fibre reinforced epoxy composites and found that impact strength of these composites is poor. The effect of silane treatment on the mechanical and physical properties of sisal/epoxy composites was reported by Bisanda and Ansell [2]. Use of sisal fibre as a reinforcing agent in different polymeric systems such as polyester, epoxy and phenol formaldehyde were reported by Thomas and coworkers [3]. Gassan and Bledzki [4] studied the properties of jute/epoxy composites. The present article deals with the impact performance of N.F.composites. The influence of different fibre treatments was studied. In order to improve the technical fibre properties, fibres were impregnated with the matrix resin. Latex modifications were also done on the fibre to improve the impact performance.

MATERIALS AND EXPERIMENTAL TECHNIQUES

In this study random non woven mats of jute, hemp and flax were used. Epoxy resin used was in the form of adhesive film (HM 533) obtained from Hexcel. To obtain a higher fibre-matrix interaction, fibres were subjected to different chemical modifications.

Alkali Treatment (NaOH)

Nonwoven mats were immersed in 1% sodium hydroxide solution for one hour. Then the mats were washed thoroughly with water and finally with water containing a few drops of HCl to remove the final traces of alkali. Then the fibres were dried in a hot air oven.

Silane Treatment (3-amino propyl tri ethoxy silane)

Flax nonwoven mats were allowed to react with silane by immersing in silane dissolved in a water-acetone mixture for 2 h. The pH of the solution was 9.0. After that the solution is decanted and the fiber is dried.

Isocyanate Treatment (Phenyl Isocyanate)

Nonwoven mat was allowed to swell in dimethyl formamide (DMF) for 30 min and was treated with isocyanate using dibutyl tin dilaurate as catalyst at a temperature of 125C for 1h. Dimethyl formamide was used as solvent for the chemical modification processes due to its high boiling point and its good solubility for isocyanates. After the

treatment, the mats were immersed in hot DMF and washed thoroughly before being dried at 105°C in an air oven.

Resin Impregnation

Prepared a solution of silane in acetone. A dilute solution of epoxy in acetone is also prepared. Both solutions were mixed together and the nonwoven mat was dipped in the solution for 2h. Then the mat was dried.

Latex modification

Fibres were given a latex coating by dipping the mats into a rubber latex solution having 10 % dry rubber content. Both natural rubber latex and epoxidised natural rubber latex were used for the treatment.

Preparation of epoxidised natural rubber (ENR)

100 ml of NR latex was taken in an RB flask equipped with a stirrer, thermometer and condenser. The latex was stabilised by adding polyvinyl alcohol. The ammonia in the latex was neutralised by adding a quantitative amount of formalin. The latex was diluted in water to get the required solid rubber content and the required amount of formic acid followed by hydrogen peroxide were added. The set up was kept in a constant temperature bath, at 50°C. The experiment was allowed to continue for 2 h. The epoxidation was confirmed by IR studies.

Composite preparation and Characterisation

Composite plates of jute/epoxy, hemp/epoxy and flax/ epoxy at different volume fractions were prepared by autoclave moulding technique. Moulding conditions are optimised to a temperature of 125°C, pressure of 3 bars and vacuum. Impact properties were measured using an Instron -Wolpert charpy impact tester, with specimen dimensions of 80*10*4 mm³. At least 6 specimens of every composition were tested. Fibre surfaces and fracture surfaces of impact specimens were sputter coated and examined using scanning electron microscopy. A scanning electron microscope Philips 515 was used at an acceleration voltage of 20kV. Infra red spectra were recorded using Philips infra red spectrophotometer. KBr disc method is used in taking the spectrum.

RESULTS AND DISCUSSION

The impact strength of jute/epoxy, hemp/epoxy and flax/epoxy composites are depicted in Fig. 1. It is clear that the impact properties of hemp and flax composites vary with fibre volume fraction. There was an increase of about 60% in strength on increasing the fibre volume fraction, V_f from 0.15 to 0.22 in hemp/epoxy composite. But on further increase of V_f the improvement is marginal (70% at 0.3 volume fraction). In the case of flax composites the increase of fiber volume fraction does not have much influence on the impact properties. But in the jute/epoxy composite, a different trend is observed. It is clear that the impact strength decreases with increasing fibre volume fraction. This may be associated with the poor wetting and

fibre to fibre interaction at higher loading. In general, the obtained fracture energies are low, demonstrating the necessity to alter the fibre-matrix interaction. To improve the wettability and interfacial strength fibres were subjected to different treatments.

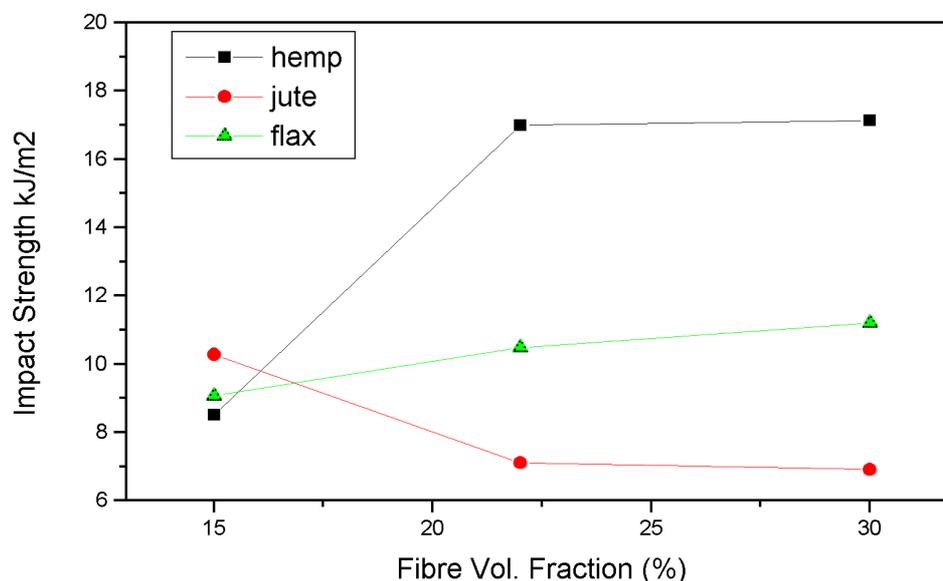
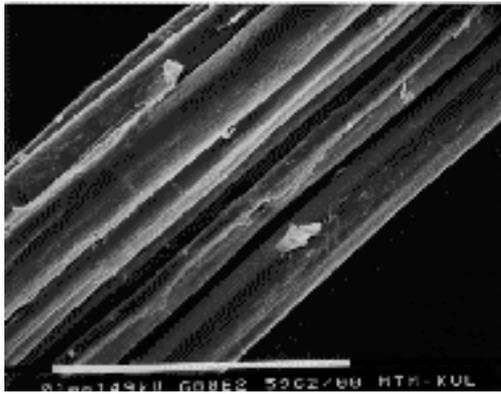


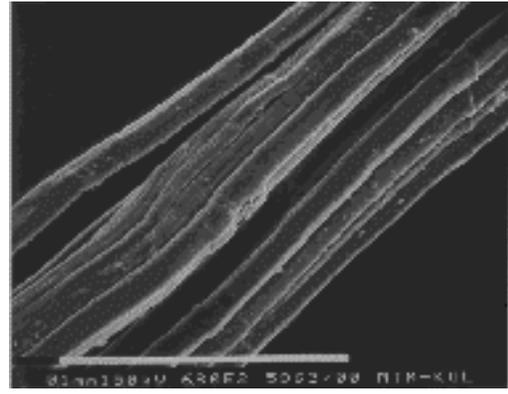
Fig. 1. Effects of fibre volume fraction on impact strength of jute/epoxy, hemp/epoxy and flax/epoxy composites.

In Fig. 2 the SEM photographs show the fibre surface morphology before and after treatment. The micrograph of untreated fibres shows their multicellular nature (Fig. 2a). But in Fig. 2b, where the fibres are alkali treated, the fibrillar structure of the individual fibre is even more revealed due to the better dissolution of waxy materials and lignin. SEM photographs given in Fig. (c) & (d) give a clear picture of the morphological changes of the fibre surface after silane and isocyanate treatment. It is clear that these fibres have a rough surface topography. As a result the effective surface area available for contact with the matrix increases.

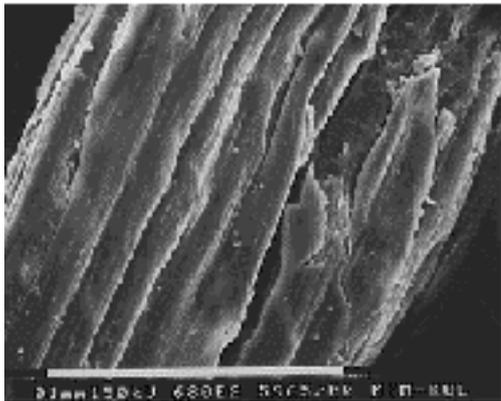
FTIR spectra of untreated and NaOH treated fibres are given in Fig. 3. Untreated fibre shows a sharp peak at 3450 cm^{-1} corresponding to -OH group stretching in cellulose. After treatment with alkali, the amount of amorphous cellulose at the expense of crystalline cellulose increases. The important modification observed here is the removal of H-bonding in the network structure. This is evident from the increased intensity of peak at 3450 cm^{-1} (b). The peak at 1730 cm^{-1} is the characteristic band for carbonyl stretching. With the alkali treatment this band disappears. It appears that in alkali treatment, a substantial portion of uranic acid, a constituent of hemicellulose is removed. The generation of band at 2260 cm^{-1} indicates the reaction of isocyanate with the fibre.



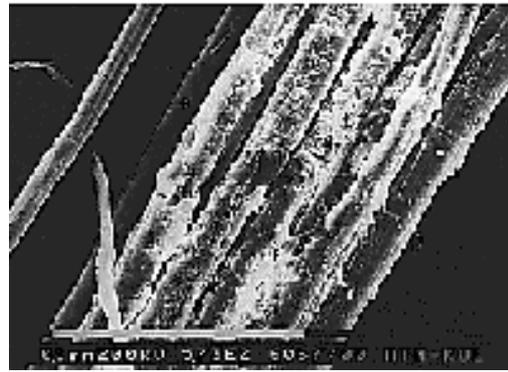
(a)



(b)



(c)



(d)

Fig. 2. SEM photographs of (a) untreated (b) NaOH treated (c) silane and (d) isocyanate treated flax fibres.

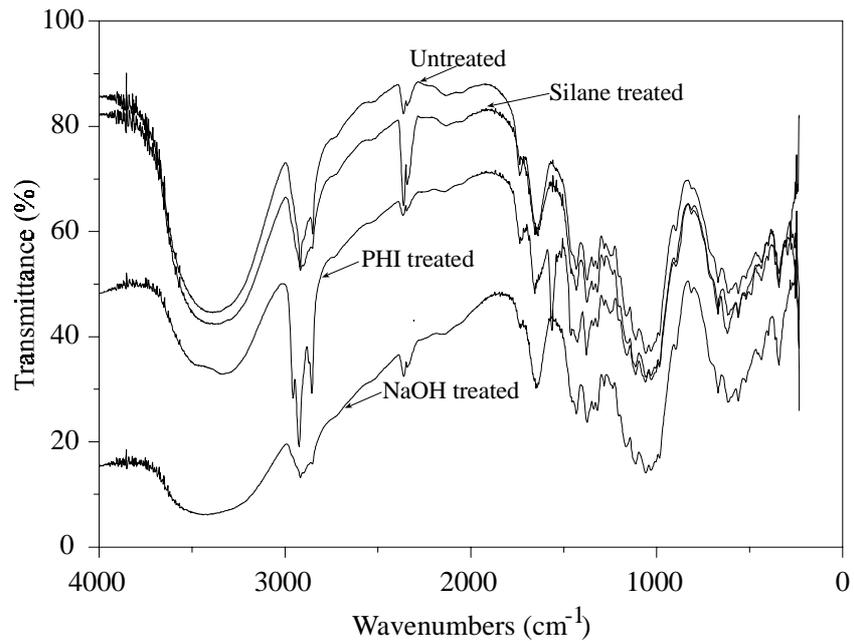


Fig. 3. FTIR spectra of (a) untreated, (b) NaOH treated, (c) PHI treated and (d) silane treated flax fibre.

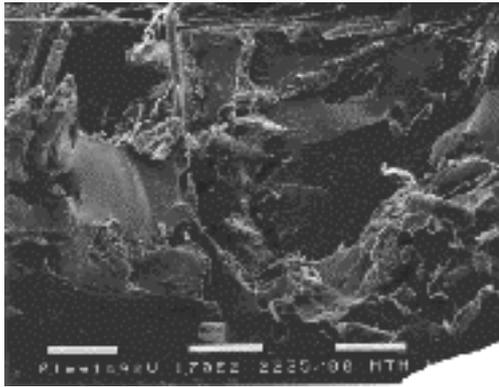
The effect of different chemical treatments on the impact performance of flax/epoxy composites are given in Table 1. It is found that the sodium hydroxide treatment increases the impact strength with about 25%. But treatment with coupling agents such as silane and isocyanate do not have much influence on the impact performance. In both cases the impact strength is reduced about 24% and 21%, respectively. But interestingly, in the case of resin impregnated fibre there is an increase in strength of about 30%. The most significant improvement is observed in the case of natural rubber latex coated fibre. In this case the increase is 76%. However, the impact strength of composite is reduced again when the fibres are coated with epoxidised natural rubber latex. Due to the treatment with coupling agents highly reactive functional group of silane (silanol) and isocyanate (-N-C=O) lead to strong hydrogen and chemical bonds with the hydroxyl group of cellulose. The modified surface can further bonds with epoxy resin and establishes a good bonding. As a result the stiffness of the composite will increase at the expense of impact strength. But after latex modification interface become more elastic and will be able to absorb more energy and hence show a high impact strength. After epoxidation, the ring opening reaction will lead to the production of hydroxy formates and the epoxy groups. These in turn can crosslink with epoxy resin matrix and a strong interface is established.

Table1. Impact strength of flax/epoxy composites. Fibre vol. fraction 22%.

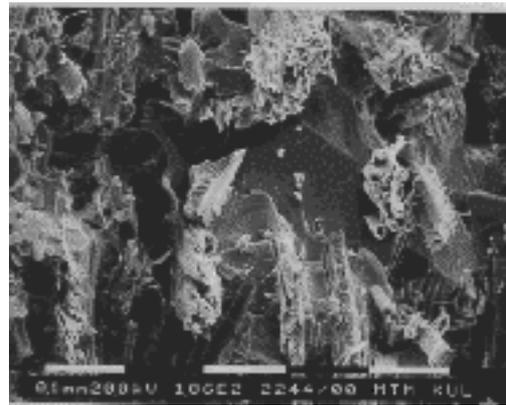
Fibre	Impact Strength (kJ/m ²)	Standard Deviation
Untreated	10.47	± 1.11
NaOH treated	13.09	± 1.59
Silane treated	8.41	± 0.20
Isocyanate treated	8.65	± 0.61
Resin impregnated	13.69	± 0.60
NR modified	18.43	± 0.40
ENR modified	9.69	± 0.50

Fracture Surface Morphology

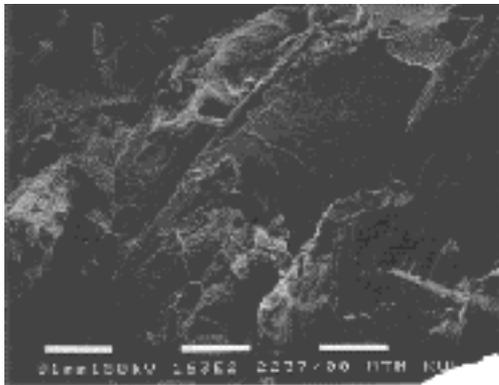
Figure 4a shows a typical fracture surface of composite. It can be seen that there is a lot of fibre pullouts in untreated fibre composite. This suggests a poor adhesion between fibre and matrix. A brittle fracture is observed in Fig. 4b& 4c in the case of silane treated and ENR treated composite due to the strong interface. A high impact strength of NR treated composite is attributed to the formation of an elastic coating and pullout of the fibre.



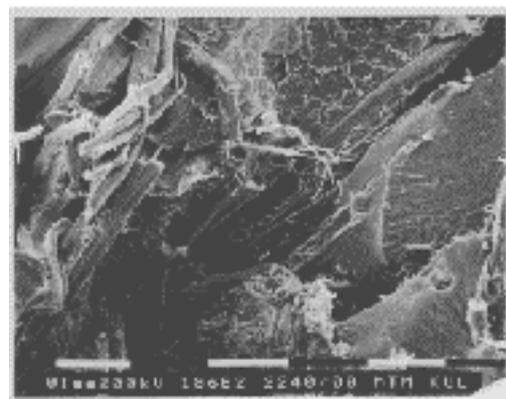
(a)



(b)



(c)



(d)

Fig. 4. SEM photographs showing impact fracture surfaces of flax fibre (a) untreated (b) silane treated (c) ENR treated and (d) NR treated fibre.

Conclusion

Composites of jute/epoxy, hemp/epoxy and flax/epoxy composites were prepared by autoclave moulding technique. It was found that impact strength of hemp and flax composites increases with increase of fibre volume fraction. Different fibre modifications were carried out to improve the fibre-matrix interaction. Modified fibre surfaces were characterised by scanning electron microscopy. It was found that impact strength of natural rubber latex modified fibre composites was improved significantly. Impact fracture surfaces were examined by scanning electron microscopy.

References

1. U. C. Jindal, *J. Comp. Mater.*, 20 (1986) 19.
2. E. T. N. Bisanda and M. P. Ansell, *Comp. Sci. Technol.*, 41 (1991) 165.
3. K. Joseph, C. Pavithran and S. Thomas, *Eur. Polym. J.*, 32 (1996) 1243.
4. J. Gassan and A. K. Bledzki, *Polym. Comp.*, 18 (1997) 179.

