

# **MODIFICATION OF JUTE FIBRES FOR APPLICATIONS IN FIBRE-REINFORCED COMPOSITES**

by

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## CERTIFICATE

This is to certify that the thesis entitled "MODIFICATION OF JUTE FIBRES FOR APPLICATIONS IN FIBRE-REINFORCED COMPOSITES" submitted by S.R. Anantha Krishnan to the Indian Institute of Technology, Delhi for the award of degree of Doctor of Philosophy is a record of bonafide research work carried out by him. S.R. Anantha Krishnan has worked under our guidance and supervision and has fulfilled the requirements for the submission of the thesis, which to our knowledge, has reached the requisite standard.

The results contained in this thesis have not been submitted, in part or full, to any other University or Institute for the award of any degree or diploma.

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## ABSTRACT

Jute fibres constitute an ubiquitous and renewable natural resource. The fibres are obtained from the stem of the jute plant. The traditional market areas of jute are being slowly captured by synthetic fibres, particularly polypropylene. In order to save the fibre, alternative avenues have to be devised in non-traditional areas of application. One of the ways of doing this is to use jute in combination with glass as reinforcement in composite materials.

However, there is a major drawback associated with the application of jute fibres for reinforcement purposes. Due to the presence of hydroxy and other polar functionalities in various constituents of the fibre, the moisture regain is high, which leads to (i) poor wettability with organic matrix resins and (ii) weak interfacial bonding. Environmental performance of such composites is also poor due to delamination under humid conditions. Therefore, in order to develop composites with better mechanical properties and environmental performance, it is necessary to impart hydrophobicity to jute fibres by reaction with suitable chemical reagents. The present thesis embodies the results of chemical modifications of jute fibres. Surface modification of jute fibre is expected not only to decrease moisture adsorption, but also concomitantly increase

interfacial bond strength, which is a critical factor for obtaining better mechanical properties of composites.

Two varieties of jute fibre, i.e., *Corchorus Capsularis* (Capsularis jute) and *Corchorus Olitorius* (Olitorius jute) were treated with isopropyl triisostearoyl titanate (titanate),  $\gamma$ -aminopropyl trimethoxy silane (silane), sebacoyl chloride (SC) and tolylene diisocyanate (TDI). Coating of fibres with unsaturated polyester (USP) and vinyl ester (VE) resin from dilute methyl ethyl ketone solution was also investigated. These reactions were carried out in organic solvents under various reaction conditions of time, temperature and concentration of reagent. Titanate treatment of jute fibres was done using benzene as solvent at 78°C for 3 or 6h. Silane reaction was also done in benzene at 70°C for 3 or 5h. Dioxan was used as a solvent for sebacoyl chloride, and reaction was carried out at 30°C for 3h, or 80°C for 2h. TDI treatment was carried out in DMSO at 35°C for 3h, or 100°C for 2h. Coating with USP resin (conc. 1, 4, 8, 12 & 16% w/w) and with VE resin (conc. 1, 2.5, 5, 9, & 16% w/w) was performed in methyl ethyl ketone. The modified fibres were characterised by weight gain experiments, elemental analysis and ir spectroscopy.

The moisture regain (MR)-relative vapour pressure (RVP) hysteresis of treated fibres was studied vis-a-vis

untreated fibre. All modified fibres showed hydrophobicity to a greater or lesser extent. Reduction in moisture regain of fibres was more when modification was done at elevated temperature [for e.g. TDI (100°C)], or for greater reaction time [silane (5h), titanate (6h)] or with higher concentration of reagents [USP (16%), VE (16%)]. SC (80°C) treated fibres also showed greater hydrophobicity than SC (30°C) treated fibres upto about 60% RVP. In all cases, the % AR (% adsorption regain) and % DR (% desorption regain) reduction values of treated with respect to untreated fibres were higher at lower ranges of RVP, but progressively decreased over the higher ranges of RVP.

The mechanical properties of modified jute fibres remained, more or less, unaffected by silane, titanate and TDI treatments. However, a deterioration was observed upon SC, USP (16%) and VE treatments.

Density and X-ray crystallinity studies were also conducted to supplement the moisture regain and mechanical properties' data. The density of all treated fibres showed a small increase over those of untreated fibre. The degree of crystallinity (D.C.) of jute fibres remained unchanged upon titanate (6h), silane (5h), TDI (100°C) and SC (30 & 80°C) treatments, but dropped significantly upon USP (16%) and VE (16%) treatments.

Thermal behaviour of modified jute fibres vis-a-vis untreated fibres was investigated by dynamic thermogravimetry (TG) and differential scanning calorimetry (DSC). In TG studies, Capsularis and Olitorius jute fibres showed initial weight loss (2.5 - 3.5%) in 50-150°C temperature region in both nitrogen and static air atmospheres. Major weight loss in these fibres, however, occurred above 200°C.

Thermal degradation of Capsularis and Olitorius jute above 200°C proceeded in two steps in nitrogen atmosphere. The temperature of maximum rate of weight loss corresponding to the two steps ( $T_{\max-1}$  and  $T_{\max-2}$ ) were ~300 and 375°C. The final decomposition temperature of the two steps ( $T_{f1}$  &  $T_{f2}$ ) were ~325 and 400°C. The residual weight at  $T_{\max-1}$  was ~90%. Residual weight at  $T_{\max-2}$  was 46% for Capsularis jute and 50% for Olitorius jute. Char yield in nitrogen atmosphere at 550°C was 19% for Capsularis jute and 23% for Olitorius jute.

In static air atmosphere, degradation proceeded in three steps.  $T_{\max-2}$  decreased by approximately 25°C, and residual weight at  $T_{\max-2}$  was ~55%, i.e. higher in static air compared to nitrogen atmosphere. Temperature of maximum rate of weight loss for the third degradation step ( $T_{\max-3}$ ) was ~475°C; residual weight at  $T_{\max-3}$  was 12.5% for Capsularis jute and 16% for Olitorius jute;  $T_{f-3}$  was 510°C for Capsularis jute and 540°C for Olitorius jute. Char yield

at 550°C was 3.0% & 1.5% for Capsularis and Olitorius jute, respectively.

Modified fibres of Capsularis and Olitorius jute showed lower weight loss in 50-150°C region. Further, SC treatment of Olitorius jute brought about a decrease in  $T_{\max-1}$ , residual weight at  $T_{\max-3}$ , and in the char yield. TDI treatment showed an additional decomposition step with  $T_{\max}$  at 185°C. An increase in residual weight at  $T_{\max-2}$ , decrease in weight loss upto  $T_{f-2}$  and increase in the char yield was observed in silane treated jute.  $T_{\max}$  observed around 300°C in Capsularis jute was absent in USP treated fibres. A decrease in residual weight at  $T_{\max-2}$ , increase in weight loss upto  $T_{f-2}$  and lower char yield was observed on USP treatment of jute fibres. The TG behaviour of VE treated Olitorius jute incorporated the thermal degradation features of Olitorius jute and VE resin. Increase in residual weight at  $T_{\max-1}$  &  $T_{\max-2}$ , and decrease in weight loss upto  $T_{f-1}$  &  $T_{f-2}$  of Olitorius jute was observed upon treatment with VE resin of different concentrations.

DSC studies of modified fibres showed broad endothermic transition in the temperature range of 60-150°C, which is due to desorption of water. The area under this endotherm was used to evaluate enthalpy of desorption ( $-\Delta H$ ). Modification of jute fibres by various treatments described earlier resulted in a decrease in  $-\Delta H$  values.

Hybrid composites were fabricated using glass and untreated/TDI(100°C)/titanate(6h)/silane(5h) treated jute fabric having similar jute (~25-27%), glass (~6-8%) and resin contents (~66-69%) by hand lay-up technique. A Carver laboratory press was used for fabrication of laminates. A stacking sequence of glass on the outer layers and jute in the inner layers was maintained. Commercially available styrenated unsaturated polyester resin (USP) was used as the matrix. Mechanical properties (tensile, flexural and inter-laminar shear strength) of the composites were evaluated using standard ASTM procedures.

Composites made from glass/untreated jute fabric (jute content 27.2%) showed improvement of 91, 214 & 78%, respectively, in flexural strength, flexural modulus and tensile strength over those of USP resin. Further, composites made from glass/titanate treated jute fabric showed a marginal improvement (5,16,15 & 4% increase in values of flexural strength, flexural modulus, tensile strength and ILSS, respectively) over those of glass/untreated jute fabric control. On the other hand, composites made from glass/silane,TDI treated jute fabric showed a decrease in mechanical properties.

The durability of the hybrid composites was studied by exposing them to an environment of relative humidity ~95% for a period of 9 weeks by placing them in a

closed vessel containing water in a tray. Changes in weight and thickness were monitored at intervals of 1 week. The mechanical properties were evaluated at the end of 9 weeks of exposure.

Exposure to water vapour resulted in an increase in weight and thickness of all composites. The weight & thickness gain were, however, lower for composites from glass/titanate treated jute fabric, whereas they were higher for glass/silane/TDI treated jute fabric composites compared to glass/untreated jute fabric control.

Exposure to water vapour also resulted in a drop in mechanical properties of all composites. But glass/titanate treated jute fabric composite still showed better properties (16% and 14% higher values in flexural modulus and tensile strength) over the glass/untreated jute fabric composite. However, the silane and TDI treated counterparts showed inferior mechanical properties.

Amongst the various chemical treatments of jute fibre studied in the present work, the most promising treatment was based on isopropyl triisostearoyl titanate. Such modification of jute reduced moisture regain, and hybrid composites with better retention of mechanical properties in wet condition could be fabricated from such fibres.

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