Fracture Behavior of Vinylester Resin Matrix Composites Reinforced with Alkali-Treated Jute Fibers

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ABSTRACT: Jute fibers were treated with 5% NaOH solution for 4 and 8 h, respectively, to study the mechanical and impact fatigue properties of jute-reinforced vinylester resin matrix composites. Mechanical properties were enhanced in case of fiber composites treated for 4 h, where improved interfacial bonding (as evident from scanning electron microscopy [SEM]) and increased fiber strength properties contributed effectively in load transfer from the matrix to the fiber; but their superior mechanical property was not retained with fatigue, as they showed poor impact fatigue behavior. The fracture surfaces produced under a three-point bend test and repeated impact loading were examined under SEM to study the nature of failure in the composites. In case of untreated fiber composites, interfacial debonding and extensive fiber pullout were observed, which lowered the mechanical property of the composites but improved their impact fatigue behavior. In composites treated for 4 h under repeated impact loading, interfacial debonding occurred, followed by fiber breakage, producing a sawlike structure at the fracture surface, which lowered the fatigue resistance property of the composites. The composites with fibers treated with alkali for 8 h showed maximum impact fatigue resistance. Here, interfacial debonding was at a minimum, and the fibers, being much stronger and stiffer owing to their increased crystallinity, suffered catastrophic fracture along with some microfibrillar pullout (as evident from the SEM micrographs), absorbing a lot of energy in the process, which increased the fatigue resistance property of the composites. © 2002 Wiley Periodicals, Inc. J Appl Polym Sci 85: 2588–2593, 2002

Key words: jute fiber; fracture behavior; flexural test; impact fatigue; fiber pullout

INTRODUCTION

There is a growing interest in natural fiber–based composites, such as jute, sisal, ramie, flax, PALF, coir, and so forth, because of their low cost, light weight, high specific modulus, renewability, and biodegradability; but their high level of moisture absorption, poor wettability, and an inadequate level of adhesion with the nonpolar resin matrix lead to poor bonding at the interface. Various surface treatments of natural fibers have, therefore, been attempted, such as alkali treatment;[1–4] silane treatment;[5] and the use of different coupling agents such as polyester amide polyol,[6] trichloro-s-triazine,[7] and so forth, to improve the bonding at the interface. Among these treatments, alkali treatment is the most economically viable. Some authors have employed this technique for jute,[1,2] coir,[3] and coconut fiber,[4] and the changes occurring in the composite properties

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have been reported. Gassan and Bledzki[1,2] treated isometric jute yarns with 25% NaOH solution for 20 minutes and reported 60% improvement in the treated jute/epoxy composite properties. Prasad et al.[3] treated coir fibers with 5% NaOH solution for 72–96 h and reported 40% improvement in the flexural strength and 5%–10% improvement in the flexural modulus of the alkali-treated coir/polyester composites. The improvements have been attributed to the greater reactivity of the treated fibers, with the resin promoting superior bonding with alkali-treated fibers.

Whilst the investigators mainly reported the mechanical properties of the composites containing both untreated and treated fibers, they did not, however, examine in detail the nature of failure in the composites. The fracture behavior of natural fiber is much more complex than that of the planar failure obtained for synthetic fibers such as glass, carbon, and so on. But very few studies have been reported on the fracture behavior of natural fibers and natural fiber–reinforced composites. A. K. Mukhopadhyay et al.[8] studied the fracture of jute fibers at various test lengths and various rates of extension. They observed that at a shorter test length (2 cm), the diameter variation along the fiber axis being less, the stress distribution was better, the probability of developing cracks at any point was less, and hence, the incidence of slippage was predominant. At a higher test length (4 cm) and higher rate of extension (5 mm/min), catastrophic fracture was observed. At a lower rate of extension (2 mm/min), slippage-type failures mixed with occasional sharp breaks at weak points were evident. Mukherjee et al.[9] classified the fracture behavior of various cellulose fibers into two groups. One is the intercellular fracture, where the crack propagates between the cells, and the other is intracellular fracture, where the crack propagates through the cells causing microfibrillar pullout. The fracture behavior of the natural fibers largely affects the fracture behavior of natural fiber–reinforced composites. Sanadi et al.[10] examined the impact-fractured surface of sunhemp/polyester composites and observed fibrillar pullout with the plastic deformation of the matrix. Pavithran et al.[11] investigated the impact-fractured surface of sisal-, PALF-, banana-, and coir-reinforced polyester composites. They explained the variation in the impact properties of various natural fiber composites in terms of the microfibrillar angle of the fiber. The fracture surfaces of straw/polyester composites were studied by White and Ansell.[12] They observed that the tensile failures were relatively flat and transverse, whereas the bend failure surfaces were considerably greater in area and indicated that extensive shear delamination had occurred at the fiber/resin interface. Roe and Ansell[13] showed that the tensile fracture of 15% jute/polyester composites were mostly brittle with very little fiber pullout. They observed that with the increase in the jute content, the pullout lengths were increased and the crack paths were macroscopically longer and more complex, involving delamination between the strands of jute sliver.

In this article, an attempt has been made to study the fracture behavior of raw and alkali-treated jute fiber composites under a three-point bend and impact fatigue test. The nature of failure at the fiber/matrix interface, fiber fracture behavior, and matrix cracking were investigated under SEM.

EXPERIMENTAL

Materials

Jute fibers (white jute, *Corchorus capsularis*) were wrapped in black paper, kept in sealed polythene bags, and stored at 65% RH and 25°C temperature. Vinylester resin used was of grade HPR 8711, a Bakelite Hylam product. Methyl ethyl ketone peroxide (MEKP), Co napthenate, and N,N-dimethylaniline were used as catalyst, accelerator, and promoter, respectively.

Alkali Treatment

Jute fibers were cut to 50 cm in length and soaked in a 5% NaOH solution at 30°C, maintaining a liquor ratio of 15 : 1. The fibers were kept immersed in the alkali solution for 4 and 8 h. The fibers were then washed several times with fresh water to remove any NaOH sticking to the fiber surface, neutralized with dilute acetic acid, and finally washed again with distilled water. A final pH of 7 was maintained. The fibers were then dried at room temperature for 48 h, followed by oven drying at 100°C for 6 h.

Sample Preparation

Jute/vinylester composites containing raw and alkali-treated jute fibers were fabricated in the form of cylindrical rods 6 mm in diameter. Hollow cylindrical glass tubes with an internal diameter of 6 mm were used as moulds. The resin was

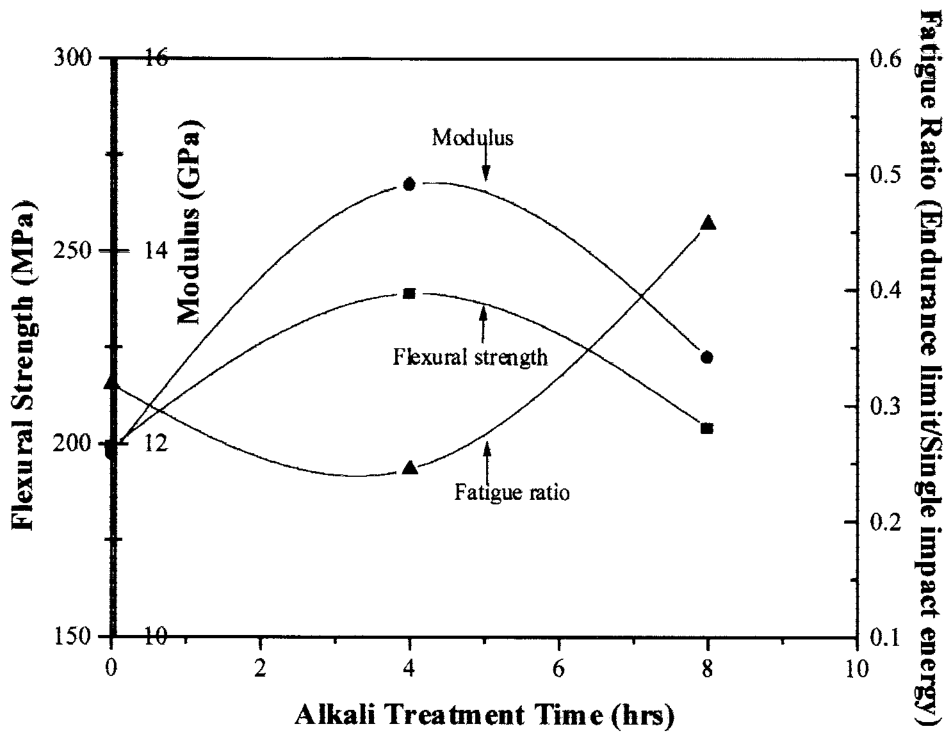

Figure 1 Variation of flexural strength, modulus, and fatigue ratio of 35 vol% composites with the alkali-treatment time.

mixed with accelerator, promoter, and catalyst (1% each). The jute fibers were dried in an oven at 100°C for 4 h before use and soaked in the mixed resin, and the wetted fibers were pultruded through the glass tube by hand.

The pultruded samples within the glass tube were cured at room temperature for 24 h, followed by postcuring in an oven at 80°C for 4 h. The glass tubes were broken clean to release the composite rods. Composites with 35 vol% of untreated and treated jute fibers were prepared for investigation. In this case, vol% is equal to wt%, as the density of the jute fiber and the vinylester resin both are equal to 1.3 gm/cm^3.

Test Methods

Flexural Test of Composites

Composites with treated and untreated fibers were tested for their flexural strength under three-point bend tests in an Instron 4303 machine in accordance with ASTM D790M-81. Test specimens were 120-mm-long cylindrical rods having a diameter of 6 mm. A span of 100 mm was employed, maintaining a crosshead speed of 2 mm/min.

Impact Fatigue Test

For impact fatigue tests, a swinging pendulum-type repeated impact tester described earlier[14,15] was designed and fabricated essentially on the basis of the principles of the Charpy Impact Tester.

SEM

The fracture surfaces of the composites were examined by SEM in a LEO S-440 (UK), using a voltage of 15 kV.

RESULTS AND DISCUSSION

There were significant changes in the chemical composition, crystallinity, and strength properties of the jute fibers in the alkali treatment. Loss in weight was observed after the alkali treatment of the fibers, with a drop in the hemicellulose content from 22.0% to 12.9% by 41%. The fibers were somewhat leached and were finer, resulting a drop in linear density from 33.0 to 14.5 denier after 6 h of treatment. By 8 h of treatment, the I_{002} peak height increased by 23.4% compared with that of the untreated fibers. The modulus and the tenacity of the fibers increased by 79% and 46%, respectively, after 8 h of treatment. The fibers treated for 8 h were thus found to be more stiff and brittle owing to their increased crystallinity. The changes occurring in the fibers have

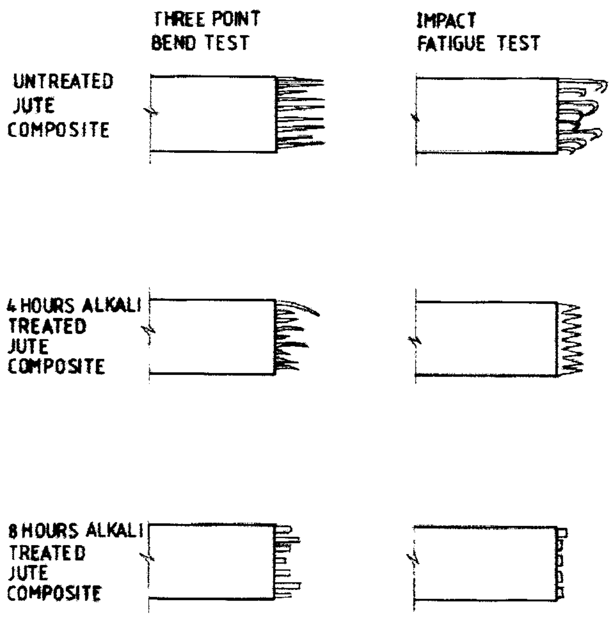

Figure 2 A schematic diagram showing the fracture surfaces of vinylester resin matrix composites reinforced with untreated and alkali-treated jute fibers under a three-point bend test and impact fatigue test.

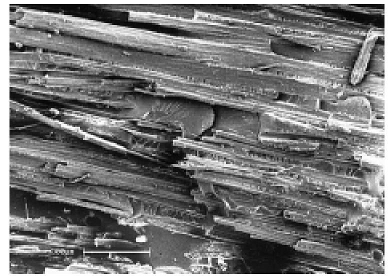

Figure 4 The fracture surface of 4-h-alkali-treated jute/vinylester composite showing improved bonding at the fiber/matrix interface (three point bend test).

been discussed in detail in our previous work.[16] Maximum improvement was observed for composites with fibers treated with alkali for 4 h, where both increased fiber strength property and good interfacial bonding contributed effectively to increase the mechanical strength of the composites.[17] However, composites reinforced with jute fibers treated with alkali for 8 h were found to have maximum fatigue resistance under repeated impact loading. The percentage defects present in the composites were found to be 30%, 18.5%, and 28.5% for the untreated, 4-h-treated, and 8-h-treated fiber composites, respectively.[17] The defects lowered the flexural strength properties of the composites under slow loading (three-point bend test) but also acted as a damper to the shock to improve the impact fatigue property of the composites, as shown in Figure 1.

The composites reinforced with untreated, 4- and 8-h-alkali-treated jute fibers fractured under three-point bending and repeated impact loading were investigated with SEM. The fracture behavior of the composites containing untreated and alkali-treated jute fibers under a three-point bend and impact fatigue test have been shown schematically in Figure 2.

Fracture under Three-Point Bending Test

In case of composites reinforced with untreated jute fibers, interfacial debonding occurred with extensive fiber pullout.[17] Poor wetting of the fibers by the resin was responsible for such weak bonds at the interface (Fig. 3). In case of fiber composites treated for 4 h, improved interfacial bonding was observed (Fig. 4) with much less fiber pullout having shorter pullout lengths. For composites with fibers treated for 8 h, the interfacial bonding was highly improved (Fig. 5), resulting in a shear fracture of the composite.[17]

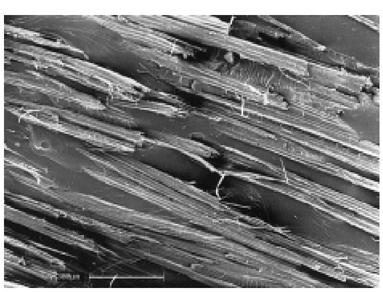

Figure 3 The fracture surface of untreated jute/vinylester composite showing weak bonding at the fiber/matrix interface (three-point bend test).

Figure 5 The fracture surface of 8-h-alkali-treated jute/vinylester composite showing strong bonding at the fiber/matrix interface (three point bend test).

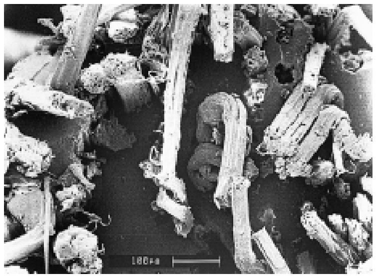

Figure 6 The repeated impact–fractured specimen of untreated jute fiber composites showing long fiber pullout with the fibers bent in the direction of the shear stress acting on the specimen (impact energy → 350.36 *N*mm, endurance → 93 cycles).

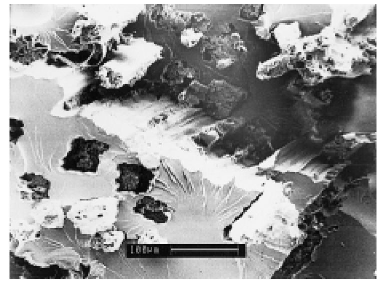

Figure 8 The fiber slippage and shear fracture of 4-h-treated fiber composites under repeated impact loading (impact energy → 204.73 *N*mm, endurance → 126 cycles).

Fracture under Impact Fatigue

The repeated impact fractured specimen of untreated fiber composites showed extensive fiber pullout with long pullout lengths, and the fibers were bent in the direction of the shear stress acting on the specimen, as shown in Figure 6. Here the interfacial bonding was weak, and the crack propagated along the fiber/matrix interface, causing debonding, thus providing a composite with low strength properties. Such extensive debonding lead to a significant increase in the energy-absorbing capacity of the composites as a result of the large new surfaces produced and frictional work resulting from differential displacement between the matrix and the fiber, increasing the impact fatigue resistance of the composites. This conforms with the observations of Mukherjee et al.[18] and Marston et al.,[19] who reported that the composites with weak interfacial bonding show poor strength properties but high impact strength.

In case of fiber composites treated for 4 h, both the fiber strength characteristics and the interfacial bonding were in the intermediary stage between the untreated and 8-h-treated fiber composites, as we have discussed in our earlier work.[17] Here, the propagation of the crack occurred both along the interface as well as normal to the fibers, lowering the fatigue resistance property of the composites. In case of both single and repeated impact fractured specimens, interfacial debonding occurred, followed by fiber breakage, producing a sawlike fracture surface (Fig. 7). Fiber slippage from the matrix and shear fracture of the fibers were observed, as shown in Figure 8. Figure 9 shows the "corrugated" fracture surface of the fiber and the gap created between the fiber and the resin matrix during repeated impact fracture. As a result of this clearance, no frictional work was possible between the fiber and the matrix affecting the impact strength of the composites. Thus, these fiber composites treated for 4 h showed poor impact fatigue resistance. This is in agreement with Sanadi et al.,[10] who showed a similar corrugated fiber fracture in the case of impact fractured sunhemp fiber–reinforced polyester composites and concluded that the clearance between the fiber and the matrix, caused by some permanent lateral contraction of the fiber on ap-

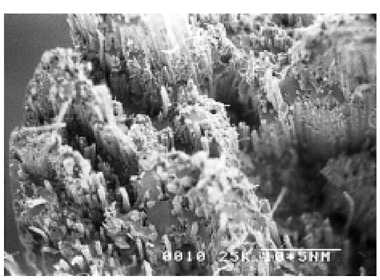

Figure 7 The sawlike fracture surface of a 4-h-alkali-treated jute/vinylester composite under repeated impact test (impact energy → 137.24 *N*mm, endurance → 2088 cycles).

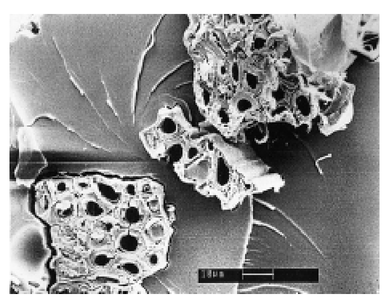

Figure 9 The corrugated fracture surface of the fiber in 4-h-treated fiber composites under repeated impact loading (impact energy → 204.73 *N*mm, endurance → 126 cycles).

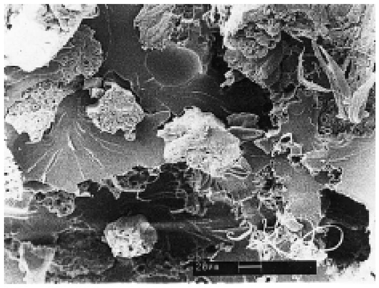

Figure 10 The catastrophic fracture of 8-h-treated fiber composites with some microfibrillar pullout under repeated impact loading. (impact energy → 256.09 *N*mm, endurance → 128 cycles).

plication of stress, reduced the frictional work between the fiber and the matrix, lowering the impact strength of the composites.

The composites with fibers treated for 8 h had the strongest interface with minimum debonding characteristics. The crack, therefore, propagated perpendicular to the fibers, and the fibers suffered catastrophic fracture with some microfibrillar pullout, as shown in Figure 10, absorbing a significant amount of energy in the process and, thus, highly improving the fatigue resistance property of the composites.

CONCLUSION

The fracture behavior of 35 vol% composites reinforced with untreated, 4-h-, and 8-h-alkali-treated jute fibers under a three-point bend test and a repeated impact test were examined under SEM. Flexurally fractured composites with untreated jute fibers showed poor interfacial bonding with extensive fiber pullout, whereas 8-h-treated fiber composites with strong interfacial bonding exhibited a transverse shear fracture. Composites with fibers treated for 4 h showed a fracture behavior intermediate between the untreated and 8-h-treated fiber composites, with much less fiber pullout having shorter pullout lengths and improved interfacial bonding compared with untreated fiber composites. Under repeated impact loading, interfacial debonding was predominant in the case of untreated fiber composites showing long fiber pullout. For fiber composites treated for 4 h, both interfacial debonding and fiber breakage occurred in parallel, producing a sawlike fracture surface. A catastrophic failure was observed for 8-h-treated fiber composites with some microfibrillar pullout, as evident from SEM photographs.

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