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Hydrothermal ageing of jute–glass fibre hybrid composites – an acousto-ultrasonic study

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Studies of damage under hydrothermal ageing incurred by random fibre glass-reinforced laminates and jute–glass fibre hybrid composites have been carried out using an acousto-ultrasonic technique. It is shown that the "stress wave factor" is a sensitive indicator of flexural strength reduction due to hydrothermal effects in both these composites. The incorporation of jute into glass fibre composites brings about a reduction in the rate of degradation of these composites. The treatment of jute with a silane coupling agent marginally improves the strength properties of the hybrids.

1. Introduction

The possibility of using jute as a cheap reinforcing fibre for fabrication of composites has been investigated by several authors [1–5]. Although the composites have low specific strength (340 MPa compared to 1.36 GPa of glass), the low specific gravity of jute (1.3) compared to that of glass (2.5) makes it an attractive candidate for jute–glass hybridization [5]. While detailed studies of tensile, flexural, compressive and impact behaviour of jute-reinforced plastic (JRP) and jute–glass hybrids have been reported in the literature [1–7], very little work has been done on the hydrothermal degradation of these composites. Shah and Lakkad [4] have reported reduction in tensile strength and Young's modulus of JRP, jute–glass hydrid and glass-reinforced composites (GRP) after 2 h boiling in water. Their results indicate that jute–glass hybrids are less susceptible to hydrothermal degradation compared to either JRP and GRP laminates. In an earlier study [8] it was shown that the treatment of jute with silane coupling agents improves the hydrothermal degradation characteristic of JRP. Different treatments of natural fibres have also been reported in the literature. Belmares [9] investigated the effect of polyvinyl alcohol, polyvinyl acetate and 2-hydroxyethyl methacrylate (HEMA) as an additive to polyester resin on palm fibre–polyester composites. Coutts and Campbell [10] have reported the effect of alkoxides of titanium and silane coupling agent treatment on wood fibre in wood–fibre reinforced cement composites. A coupling mechanism has also been proposed through the formation of metal hydroxide species on fibre surface and covalent chemical linkages with the matrix.

The purpose of this paper is to report the hydrothermal degradation of both untreated and silane-treated jute–glass hybrid composites along with GRP, studied by an acousto-ultrasonic technique.

2. Experimental methods

The material used in the preparation of hybrid laminates for the present study were "E"-glass chopped strand mat (CSM) of density 450 g m^{-2} (FGP Ltd, India) isopthalic polyester resin HSR8131 (Bakelite Hylam Ltd, India) and commercially available woven jute cloth of density 400 g m^{-2}. Large panels were fabricated using three layers of CSM and two layers of jute cloth by standard hand lay-up technique with the jute cloth sandwiched between two layers of CSM. CSM formed the outermost layers. The basis for selection of this lay-up was that jute having the lower strength, was kept in the vicinity of the neutral axis so that it was subjected to relatively low stresses during testing in flexure. The central CSM layer was used for good bonding between two layers of jute cloth. Methyl ethyl ketone peroxide and cobalt napthanate were used as catalyst and accelerator, respectively. The laminates were cured in laboratory atmosphere for 18 h and then post-cured at a temperature of 100° C for 4 h. The nominal thickness of the finally cured laminate was 3.9 ± 0.1 mm having a glass content of 22% by weight and a jute content of 9% by weight as determined from the weight of individual constituents. For a second panel, the jute cloth was treated with a hydrolysed solution of γ-methacryloxy propyl trimethoxy silane (Union Carbide – A174) at pH 5 and dried at 80° C before being incorporated into the laminate. Similar GRP panel was also fabricated using three layers of CSM only.

Flexure test specimens of length 80 mm and width 10 mm were cut out of these panels and the edges were smoothened by sanding. Each specimen was subjected to measurements of the acousto-ultrasonic stress wave factor using acousto-ultrasonic equipment Model 206AU (AET Corporation, USA) by placing the receiving transducer at the centre of the specimen and the transmitting transducer near the end of the specimen. The equipment has been described elsewhere [11]. The instrument was set up as follows: gain 50 dB, threshold voltage 0.24 V (auto mode operation), pulse rate 2000 sec^{-1}, pulse energy 50 V, gate width 187.2 sec, time window 1 sec to obtain readings in a convenient range for the material studied.

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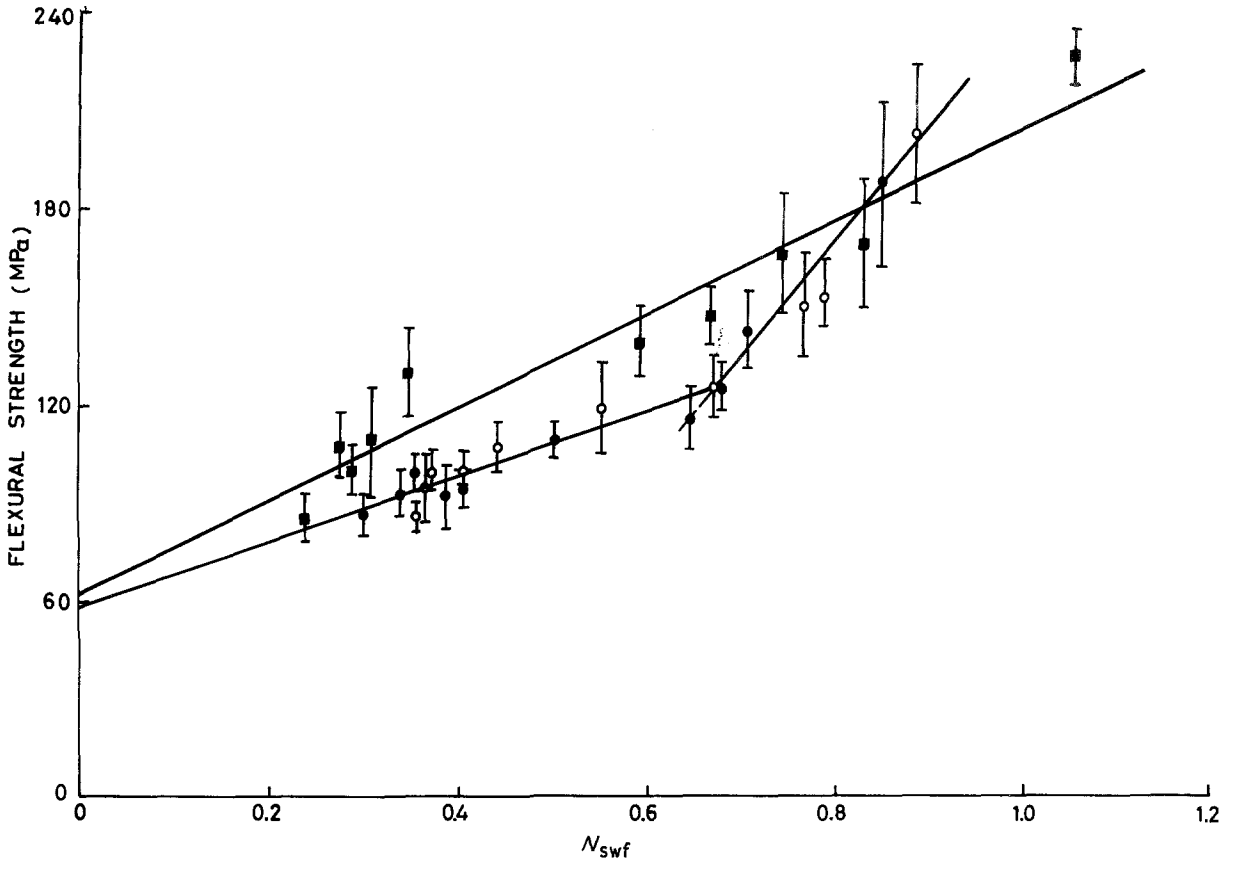

Figure 1 Variation of flexural strength with N_{swf}. (■) GRP, (○) jute/GRP (silane-treated), (●) jute/GRP (untreated), $\bar{\Phi}$ average with 95% confidence limit.

The stress wave factor is a measure of stress wave energy propagation in the material and gives the integrated effects of microflaws which controls the strength of the system. In stress wave factor measurement the pulses are generated at a fixed repetition rate, g, with each successive pulse identical to its predecessors. After amplification, the signals received are sent to an electronic counter, which determines the number of oscillations, n, received in each time window, which exceeds the fixed threshold value. The time window of the counter is reset after each interval, r. The number that is displayed, $E = grn$, is defined as the stress wave factor. This value is normalized relative to the maximum E value formed for all specimens, i.e. $N_{swf} = E/E_{max}$. The numbers E and N_{swf} are both relative and depend on factors such as probe pressure, signal gain, reset time, threshold voltage, repetition rate. All these factors have been kept constant in this study.

Ten specimens were tested dry in three-point flexure using an Instron 1195 machine at a displacement rate of $2\,mm\,min^{-1}$. 90 specimens were kept in boiling distilled water. Ten samples were drawn at random from these after a specified number of hours of boiling. They were subjected to stress wave factor measurement and flexure test after cooling to laboratory temperature. The flexure strength was calculated by using standard beam formula and reported as average of ten values.

The procedure was repeated for all types of laminates described earlier.

3. Results

The values of average flexural strength, σ, obtained for GRP, untreated jute–glass hybrid (UGJG) and silane treated jute–glass hybrid (TGJG) laminates after different hours of ageing (t) in boiling water are plotted against the normalized stress wave factor, N_{swf}, in Fig. 1. The stress wave factor was normalized with respect to the highest value obtained for TGJG laminates. The plot indicates that σ for GRP laminate increases monotonically with N_{swf}. The least square analysis of the data yielded the relation

$$\sigma = 144.8\,N_{swf} + 61.9 \quad (1)$$

with coefficient of determination of 0.97.

There is also monotonic increase of σ with N_{swf} for both UGJG and TGJG laminates; however, the data fits two different regression lines giving the relations

$$\sigma = 347.7\,N_{swf} - 108.4 \quad (2)$$

for $0 \leqslant t \leqslant 70\,h$, and

$$\sigma = 99.4\,N_{swf} + 58.9 \quad (3)$$

for $t > 70\,h$, with coefficient of determination 0.92 and 0.98, respectively. Analysis of the values of normalized stress wave factors at different hours of ageing showed a linear relationship between $\ln N_{swf}$ and t with the coefficients of determination of 0.95, 0.99 and 0.98 for GRP, UGJG and TGJG laminates, respectively. Thus a relation of type $N_{swf} = A \exp(-bt)$ where A and b are constants fitted to the data following the method given in [12], yielding the relations

$$N_{swf} = 1.012 \exp(-7.433 \times 10^{-3}\,t) \quad (4)$$

$$N_{swf} = 0.88 \exp(-3.788 \times 10^{-3}\,t) \quad (5)$$

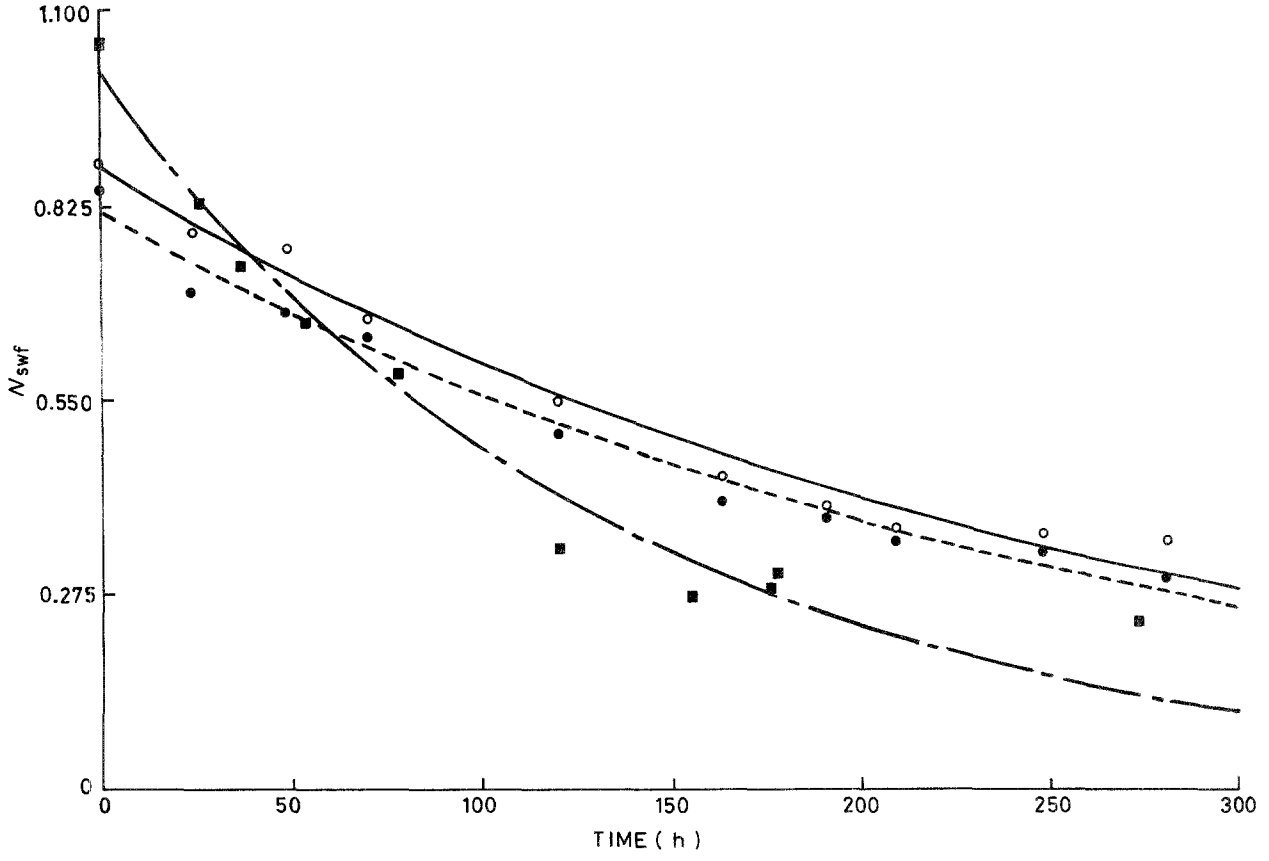

Figure 2 Variation of N_{swf} with time of boiling in water. (■, –·–) GRP; (○, ——) jute/GRP (silane-treated); (●, – – –) jute/GRP (untreated).

and

$$N_{swf} = 0.817 \exp(-3.862 \times 10^{-3} t) \quad (6)$$

for GRP, TGJG and UGJG laminates, respectively. Equations 4 to 6 have been plotted in Fig. 2.

Combining Equations 4, 5 and 6 with Equations 1, 2 and 3, respectively, the strength degradation with hours of ageing in boiling water are obtained as:

(a) GRP laminates

$$\sigma = 146.48 \exp(-7.433 \times 10^{-3} t) + 61.9 \quad (7)$$

(b) TGJG laminates

$$\sigma = 305.84 \exp(-3.788 \times 10^{-3} t) - 108.42 \quad (8)$$

for $t < 70$ h and

$$\sigma = 87.49 \exp(-3.788 \times 10^{-3} t) + 58.9 \quad (9)$$

for $t > 70$ h

(c) UGJG laminate

$$\sigma = 283.9 \exp(-3.862 \times 10^{-3} t) - 108.42 \quad (10)$$

for $t < 70$ h and

$$\sigma = 81.22 \exp(-3.862 \times 10^{-3} t) + 58.9 \quad (11)$$

for $t > 70$ h.

Equations 7 to 11 are plotted in Fig. 3 together with the experimentally measured values. For TGJG and UGJG laminates two separate curves are obtained and they have been joined by a smooth curve. The strength and modulus values obtained for these two types of laminates are also given in Table I.

4. Discussion

In stress wave factor measurement, the sending transducer injects a repeating series of pulses, each of which produces simulated stress waves that resemble

TABLE I Flexural strength and modulus of GJG hybrid composites after water immersion

Time in boiling water (h)	TGJG (silane-treated)				UGJG (untreated)			
	Flexural strength (MPa)	Standard deviation	Flexural modulus (GPa)	Standard deviation	Flexural strength (MPa)	Standard deviation	Flexural modulus (GPa)	Standard deviation
0	205.4	9.883	7.6	0.704	189.5	12.032	7.5	1.696
24	154.4	8.729	7.3	0.861	144.42	9.877	6.6	0.534
48	151.6	22.758	7.7	0.793	126.26	11.147	6.1	0.585
70	126.4	9.32	5.9	0.405	116.1	8.124	6.2	0.360
120	120.4	18.830	6.3	0.751	109.8	7.645	5.6	0.386
164	108.2	9.998	5.5	0.533	94.3	8.732	5.3	0.499
191	101.1	8.588	5.8	0.782	92.3	18.677	5.3	1.475
209	100.4	9.094	5.6	0.523	99.5	7.162	5.3	0.613
248	95.4	15.072	5.8	1.057	93.26	10.377	5.3	0.563
281	86.6	5.943	5.0	0.395	86.6	9.281	5.1	0.655

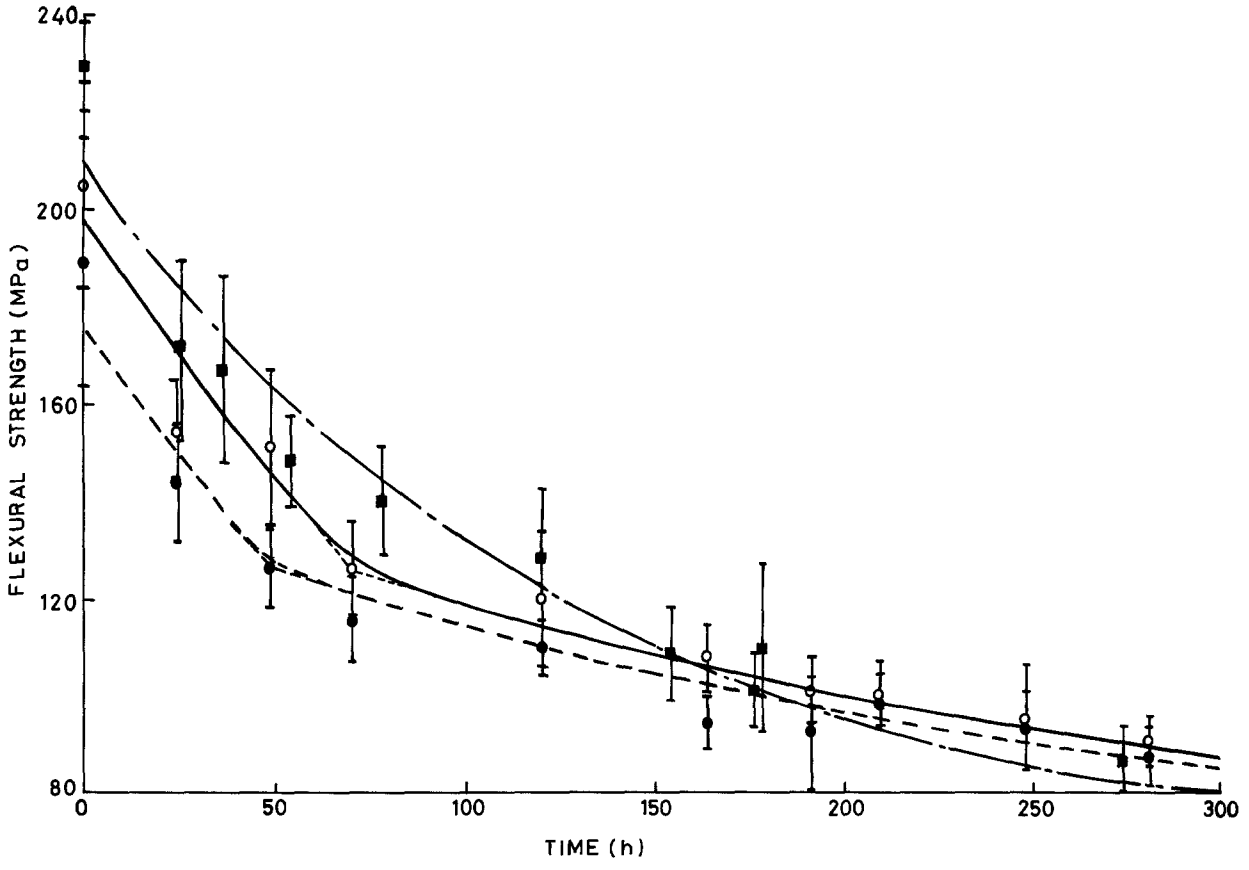

Figure 3 Variation in flexural strength with time of boiling in water. (■, ---) GRP; (○, ——) jute/GRP (silane-treated); (●, ——) jute/GRP (untreated); (Φ) average with 95% confidence limit.

acoustic emission in the material [13, 14]. Any defect or discontinuity present in the material under investigation will lead to the attenuation of energy with consequent reduction in energy received by the receiving transducer. Thus stress wave factor may be described as a measure of the efficiency of stress wave energy transmission. In a random fibre GRP composite, energy transmission essentially takes place from matrix to the fibre and again fibre to the matrix. The fibre, having higher modulus, is more efficient in transfer of energy. Any void or weak interface between fibre resin will act as a source of attenuation of energy, giving a low value of stress wave factor. Because the strength of a random fibre composite is essentially controlled by the fibre strength and load transfer efficiency at the fibre–resin interface, the weak interface also corresponds to lower strength of the composite. Thus attenuated stress wave energy flow corresponds to decreased fracture resistance [15]. On subjecting the GRP composite to ageing in water, the polyester component of GRP is subjected to water attack. Ashbee *et al.* [17] have shown that when the resin is free from inorganic impurities the water merely acts as a swelling agent and plasticizes the resin; however, in the presence of traces of salts, the resin can act as a semi-permeable membrane and the resulting osmotic pressure within the resin leads to cracking. There is also some evidence in the literature [18, 19] that the strength of E-glass reinforcement is time-dependent in the presence of small amounts of moisture. Additionally, the dissolution of water-soluble constituents of E-glass (i.e. K_2O and Na_2O) embedded in the resin, are known to generate osmotic pressures in hot water sufficient to grow resin cracks [17]. Moreover, the alkalinity of such a solution, besides chemically aiding fracture at the interface, is likely to enhance the dissolution of B_2O_3 from the fibre to form boric acid, hence boosting osmotic pressure [17]. Ashbee *et al.* [17] and Ishai [20] have also shown evidence of chemical attack on E-glass fibre when the composite is exposed to hot water. All these contribute to the reduction of the strength of the composite. As mentioned earlier, a debonded or weak interface attenuates the energy more, thereby transmitting less energy to the fibre. Also the ageing of the fibre makes it less efficient in transfer of stress wave energy. Both these factors reduce the simulated stress wave energy at the receiving transducer giving low values of stress wave factor. The correlation given by Equation 1 supports this view.

In the case of jute–glass hybrids (both UGJG and TGJG), the presence of continuous strands in woven fabric should provide a path for efficient transfer of energy. However, jute will be less efficient than glass fibre in this respect because of its low modulus compared to glass. Jute is also more hydrophilic than glass and the polymer has to compete with water for complete wetting of the surface. The incomplete displacement of air at the fibre surface possibly leaves microvoids at the interface with dimensions comparable to a fibre diameter. These voids not only act as a stress-riser to reduce the strength of the composite but also as a source of energy attenuation, reducing the stress wave factor. On ageing in boiling water, the jute fibre will absorb more water compared to a glass fibre, leading to the fibre swelling. This will induce cracking

as well as debonding at the interface, causing the composite strength to deteriorate further with consequent reduction in the stress wave factor. An earlier study [8] has shown that in the case of JRP laminates, the strength reduces at a drastic rate to 54% of its original value even after 8 h boiling and then continues to reduce slowly. Thus for the initial period, the degradation of jute plays the predominant role, and accounts for the correlation given by Equation 2. After the initial rapid degradation, further deterioration of jute takes place at a much slower rate and comparatively faster deterioration of glass fibre in the composite plays the predominant role. On prolonged exposure, the swelling rupture of the resin and glass–resin interface exposes the glass fibres to a high humid atmosphere which, when coupled with the swelling-induced tension in the fibres, can lead to stress corrosion of the glass fibre as discussed earlier. Equations 1 and 3 give almost identical values of σ for long-term ageing, thus indicating that the jute fibre makes little contribution towards the strength of the composite in such cases and strength is mainly provided by glass fibre. From Fig. 3 it can be seen that for ageing time below 70 h the rate of decrease of strength of both TGJG and UGJG laminates with time is higher than that of GRP. This, as explained before, is accounted for in terms of fast deterioration of the jute fibre and jute–resin interface in the composite on exposure to humid conditions. On the other hand, over an ageing time of 70 h, the TGJG and UGJG have a lower rate of decrease in strength with time compared to GRP laminates, possibly due to: (a) the swollen jute fibre layer acting as a cushion to absorb some of the swelling strain in the resin; (b) jute fibre layers providing additional protection to the sandwiched glass-fibre layer.

However, for long-term ageing, all three composites approach the same strength. From Table I, it can be seen that during the initial hours of ageing, TGJG laminates have a consistently higher strength compared to UGJG laminates. A statistical analysis of the strength data up to 70 h shows this difference to be significant at the 5% level. Thus silane treatment brings about a slight improvement in both dry and wet strengths of the hybrids during the initial period of boiling. The improvement brought about by silane treatment may be due to a similar mechanism as is operative in glass–polymer composite. The hydrated silanol group of A-174 may form a bond with the lignin hemicellulose and cellulose of jute fibre through hydrogen bonding, whereas the organic end tightens up the resin at the interface. A reduction in moisture absorption is also possibly brought about by the formation of a hydrophobic polysiloxane coating on the jute fibre surface, shifting the degradation process along the time axis.

5. Conclusions

The hydrothermal degradation of glass fibre and jute–glass fibre hybrid composites was studied through an acousto-ultrasonic technique. The method involved characterization of stress wave propagation in the material. Measurement yielded the stress wave factor which was used to determine the rate of the degradation process.

Specific conclusions drawn from this study are:

1. The stress wave factor is a sensitive indicator of glass-fibre and jute–glass fibre hybrid composite strength reduction due to hydrothermal effects.

2. In the case of jute–glass fibre hybrid composites the initial strength reduction is controlled by the rapid degradation of the jute fibre. Subsequent reduction is controlled by the glass fibre.

3. Incorporation of jute brings about a reduction in the rate of degradation of these hybrids over that of glass-fibre composites.

4. Treatment of jute with a silane coupling agent brings about a marginal improvement in strength properties of jute–glass hybrids.

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