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Stress Relaxation Behavior of Banana Fiber-reinforced Polyester Composites

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ABSTRACT: Stress relaxation behavior of banana fiber-reinforced polyester composites were investigated with special reference to the effect of fiber loading, fiber treatment, hybridization with glass fiber and also as woven fabric composites. It was observed that incorporation of fiber in the polyester matrix reduces the rate of relaxation. The nature of the relaxation curve was found to depend on the quantity of fiber as well as the surface characteristics of fiber used as reinforcement. The decrease in stress relaxation modulus was greatest for the neat polyester sample whereas it was at a minimum for composites with 40% fiber loading. Chemical modification of the fiber surface was found to affect the degree of adhesion and thereby the nature of relaxation at the initial stages. Of the various chemical modifications chosen in the present study, the decrease in stress was found to be the lowest for composites made out of banana fiber treated with NaOH. Hybridisation of banana fiber with glass was found to decrease the rate of relaxation further. However, stress relaxation values of woven glass and banana gave even lower values.

KEY WORDS: stress relaxation, banana fiber, polyester, hybrid composite, woven fabric.

INTRODUCTION

POLYMERIC MATERIALS ARE replacing conventional engineering materials and information about the response of the material over a long period of time is important. Meaningful data about the behavior of the materials can be obtained by accelerated testing methods. Creep and stress relaxation are the widely employed testing methods for this. Since stress relaxation represents the basic time-dependent response of

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the material, the measurement of stress relaxation is considered very important [1]. Moreover, creep and stress relaxation are the most fundamental experiments used for characterizing the viscoelastic properties of materials. The decrease in stress with time when a solid is subjected to constant strain can be measured using these experiments. The stress relaxation modulus of polymers is increased by rigid fillers and decreased by elastomeric ones. The rate of stress relaxation for rigid and elastomeric fillers increases after the onset of dewetting [2,3]. Flink and Stenberg used stress relaxation experiments to measure the adhesive strength of cellulose fiber-reinforced natural rubber by analyzing the relaxation mechanism [4]. Bhagawan et al. studied the stress relaxation behavior of short jute fiber-reinforced rubber composites [5]. The stress relaxation behavior of polyacetal–polyurethane blends have been studied in detail by Kumar et al. In their experiments, the rate of loss of the relaxation modulus was found to be a non-linear function of time [6].

Natural fibers have already established a track record as simple filler material in automobile parts. Although in general, the use of natural fiber-reinforced plastics is increasing, certain aspects of their behavior are poorly understood. The time dependent behavior like the creep and fatigue behavior for natural fiber-reinforced plastics has not been studied in detail except for a few reports [7]. Natural fibers like sisal, jute, pineapple leaf fiber, oil palm fiber have all been proved to be good reinforcements in thermoset and thermoplastic matrices [8–11]. Our earlier studies have proved banana fibers to be an effective reinforcement in polyester matrix [12]. Studies were carried out on the dynamic mechanical properties of banana fiber-reinforced polyester composites to have an insight into the adhesive behavior as well as the viscoelastic nature of these materials [13].

Extensive research is being carried out by Thomas and co-workers on natural fiber composites [10–14]. The stress relaxation studies of sisal fiber-filled natural rubber, pineapple leaf fiber-reinforced polyethylene composites, coir fiber-filled natural rubber, oil palm fiber-reinforced phenol formaldehyde etc. have all been studied and results reported [14–17].

In the present article we report on the stress relaxation behavior of banana fiber-reinforced polyester composites with special reference to the effect of fiber content, fiber treatment, hybridization with glass fiber, woven fabrics of banana and glass and also the effect of strain level.

EXPERIMENTAL

Materials

Banana fiber was obtained from Sheeba Fibers and Handicrafts, Poovancode, Tamil Nadu. Unsaturated polyester HSR 8131 based on isophthalic acid was obtained from Bakelite Hylam, Hyderabad. Methyl ethyl ketone peroxide and cobalt naphthenate were of commercial grade and obtained from Sharon Engineering Enterprises, Cochin. The details of the material are given in Table 1.

Hybrid Composites

Hybrid composites were prepared by using different volume fractions of glass and banana. Alternate mats of glass fiber as well as banana fiber were made for the preparation of hybrid composites. In all these samples, glass was used as the core material

Table 1. The mechanical properties of banana fiber.

Sample Number	Diameter of Fiber (μm)	Initial Young's Modulus (GPa)	SD* Initial Young's Modulus (GPa)	Breaking Strength (MPa)	% Strain
1	50	32	8.190	779	2.7
2	100	30	4	711	2.4
3	150	29	8	773	3.5
4	200	27	7	789	3.3
5	250	29	4	766	3.2

Table 2. Description of composite samples with different glass volume fraction.

Sample Marking	Volume Fraction of Glass
A	0.03
B	0.15
C	0.17

and banana as the skin. Samples with different layering patterns were also made in combinations A, B, and C as given in Table 2.

Preparation of Composites

Banana fibers were neatly separated by hand; adhering pith, if any, were removed and the fibers were cut to a uniform size of 30 mm length. The fibers were then evenly arranged in a mould measuring $150 \times 150 \times 3 \text{ mm}^3$ and pressed into a mat. Composite sheets were prepared by impregnating the fiber with the polyester resin to which 0.9 volume percent cobalt naphthanate and 1% methyl ethyl ketone peroxide were added and the air bubbles were removed carefully with a roller. The closed mould was kept under 1 bar pressure for 12 h, samples were post-cured and test specimens of the required size were cut.

Chemical Modification

Chemical modification done on the fibers and details of the hybrid samples have already been reported [18,19]. Silane treatment was done by preparing 0.6% solutions of the various silanes in alcohol–water mixture and adjusting the pH of the solution to the required value for silanol formation depending on the silane. The various silanes, A174 (γ -Methacryloxypropyltrimethoxysilane), A151 (Vinyl triethoxysilane), A1100 (γ -Aminopropyltriethoxysilane), were obtained from Sigma-Aldrich, India., Si 69 bis(triethoxysilyl propyl) tetra sulphide was obtained from Bayer(I) Ltd, Dynasylan F8261 (1*H*, 1*H*, 2*H*, 2*H*-Perfluorooctyl triethoxy silane) was obtained from ABCR GmbH and Co. NaOH and all other chemicals were of commercial grade. The silanes were allowed to hydrolyse using an ethanol–water mixture. The fibers were dipped in the respective silanols for one and a half hours, taken out, dried in air followed by drying in a vacuum oven at 70°C. The treated fibers were used for the preparation of composites.

Treatment with NaOH

Cleaned and well-separated cellulose fibers were dipped in 0.5 and 1% solution of NaOH for half an hour and then washed in very diluted acid to remove any excess alkali. Washing was continued till the fibers were alkali free. The fibers were finally washed in distilled water. The washed fibers were then dried in the oven at 70°C for 3 h.

Polarity or acid base characteristics are found to have a profound influence on the fiber–matrix interactions in composites. ζ -potential measurement and solvatochromic techniques were used to investigate the hydrogen bond donating ability, hydrogen bond accepting ability and the overall polarity of the modified fiber surface [18,24]. Treatment of the banana fiber surface with various chemical agents and the polarity parameters obtained are reported in our earlier publication. The electrokinetic measurements of the banana fibers were also done and the results were found to be mutually agreeing with those of the polarity parameter measurements.

STRESS RELAXATION

Zwick Universal Testing machine (model 1465) was employed for stress relaxation measurements in uniaxial tension at 28°C. Specimens measuring $120 \times 120 \times 2.5 \text{ mm}^3$ were used. The test specimens were pulled to a strain level of 2% using crosshead speed of 5 mm/min. Once the appropriate strain was reached, the strain was held constant, and the decay in stress was recorded as a function of time for a time span covering about 10,000 s. The stress decay was calculated as σ_t/σ_o , where σ_o is the initial stress at zero time ($t=0$) and σ_t is the stress at subsequent times. Also the stress value was converted to corresponding modulus $Er(t)$ by dividing stress σ_t by the appropriate strain (ϵ_o). The stress decay was recorded starting from 1 s after the predetermined strain was attained.

Effect of Fibre Loading

Figure 1 shows the effect of fiber loading on the stress relaxation behavior of the neat polyester sample and the composites with different banana fiber loading. The relaxation curve of the neat polyester sample shows a regular decrease in stress with time. There are reports in the literature that unfilled materials usually only have one relaxation mechanism [20]. With the incorporation of fiber, the nature of the relaxation curves changes. The rate of stress relaxation is found to be at a maximum for composites with 20% loading in the present study. However, the nature of the relaxation curve changes during the final stages. At relatively low fiber loading, the fibers instead of acting as reinforcements, act as flaws. The increased rate of stress relaxation in the case of composites with low fiber loading can be attributed to the lower contribution of the fibers in stress transfer. In addition, the characteristics of the fiber, namely fiber rigidity and flexibility, are not reflected in strength and modulus values at low fiber content. Compared to the neat polyester sample, there is hindrance to the flow of polymer molecules and the hindrance becomes noticeable only after a higher time span. When higher fiber loading is incorporated, the nature of the relaxation curve is found to change with the lowest rate of relaxation at 40% loading. Addition of higher fiber content helps in effective stress transfer between the fiber and the matrix and thereby in increasing the strength values. It has been reported by other authors that natural fiber-reinforced plastics with higher fiber fraction possess higher critical loads [7]. The tensile strength values obtained for the composites, reported in our earlier

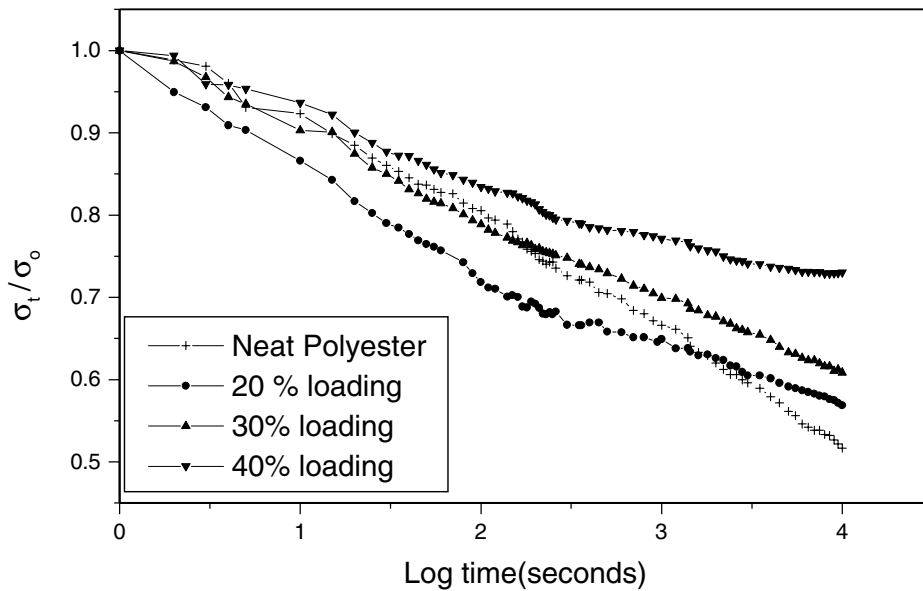


Figure 1. Stress relaxation curves of composites with different fiber loading (strain level 2%).

publication agree with the stress relaxation data obtained [12]. Cellulosic fibers are viscoelastic with the elastic nature predominating. Incorporation of this cellulose fiber induces more of elastic nature into the material. The change in free volume accompanying a uniaxial tensile deformation also induces stress induced decrease of the viscoelastic relaxation [7]. The inter winding and coiling of the polymer particles in compression lead to lesser free volume. On application of stress on the other hand, the molecules unwind and more free volume is produced. The relaxation curve shows an abrupt change in the pattern in all the samples with fiber incorporated. The initial relaxation has been reported to be globally faster for higher tensile rate [22]. The change occurs after a time span of about 200 s in all the cases. In the case of banana fibers, the elementary fibers overlap like other cellulose fibers and are glued together by other low molecular weight polysaccharides. On application of stress, physical transformations can occur in these fibers. The physical transformations include the change in the nature of the elementary fiber bundles. These elementary fiber bundles in turn are made up of micro fibrils. The micro fibrils will be pulled out from the cementing inter-phase and this leads to change in the relaxation behavior. Other authors have reported the effect of stress and strain on the structure of the fibers [7]. The reinforcing cellulose micro fibrils diminish the role of the cementing matrix as a load-bearing component by continually transferring the stress to adjacent layers, and hence acting to reduce the energy loss therein. It has further been reported by other authors that the damage phenomena characteristic of the fibers are a consequence of the micromechanical degradation and subsequent structural breakdown of the micro fibrils. All these transformations lead to changes in the nature of the relaxation curves. The polymer chains tend to get attached to the filler particles as well. In other words, the change in the slope can be attributed due to two reasons. One, the elastic nature of the fiber which induces more stress relaxation effect and the other, the improved stress transfer between the fiber and matrix. At higher fiber loading, the slope of the curve is decreased considerably at the final stages of relaxation due to better stress transfer at the

critical fiber loading [12]. At lower fiber loading, fibers instead of acting as reinforcements act as flaws which facilitates faster relaxation. This is evident from the slope changes of the curves in Figure 1 and the rate of relaxation of the respective composites at a strain rate of 2%, which is shown in Table 3. The rate of relaxation of the neat polyester sample and the composites with different fiber loading are compared in Table 3. In all the cases the initial decrease in stress is found to be faster than the decrease during the final stages. Table 4 gives the cross over times of the different mechanisms involved in the composites. The relaxation mechanism can also be attributed to the intrinsic stress relaxation behavior of the fiber. Under the stressed condition, the individual fiber can undergo various molecular and cellular rearrangements within the three dimensional multicellular network. A similar trend in the rate of relaxation is reported in the case of pineapple–polyethylene composites [15]. The modulus relaxation also shows the same trend and is shown graphically in Figure 2. While the neat polyester sample shows a 48% reduction in the stress relaxation modulus, composites with 40% fiber loading shows a 31% reduction.

Effect of Fibre Treatment

Figure 3 shows the effect of fiber surface modification on the stress relaxation curves. The fibers have been treated with various silanes and also with alkali. Silane treatment of glass fibers have been found to reduce the relaxation rates especially after longer periods, due to improved adhesion [23]. The addition of coupling agents and the treatment with alkali reduces the relaxation rate compared to the neat polyester sample. The rate of relaxation is reduced appreciably during the initial stages by treatment with silane A174 (γ -Methacryloxypropyltrimethoxysilane) and NaOH. The reduction in the slope during the initial stages can be attributed to the improved adhesion between fiber and matrix.

Table 3. Rate of stress relaxation in banana–polyester composites at a strain level of 2% at different fiber loading.

Range (s)	Fiber Loading			
	Neat Polyester	20	30	40
Slope $\times 10^{-2}$				
0 – 10 ¹	8.9	12.9	10	6.93
10 ¹ – 10 ²	12.43	18.6	13.3	11.17
10 ² – 10 ³	16.20	8.48	11.53	7.78
10 ³ – 10 ⁴	21.28	11.9	12.75	4.93

Table 4. Cross over time of the relaxation mechanism in composites with different fiber loading (strain level 2%).

Sample	Cross Over Time (s)
Neat Polyester	Nil
20%	2.6
30%	2.4
40%	2.3

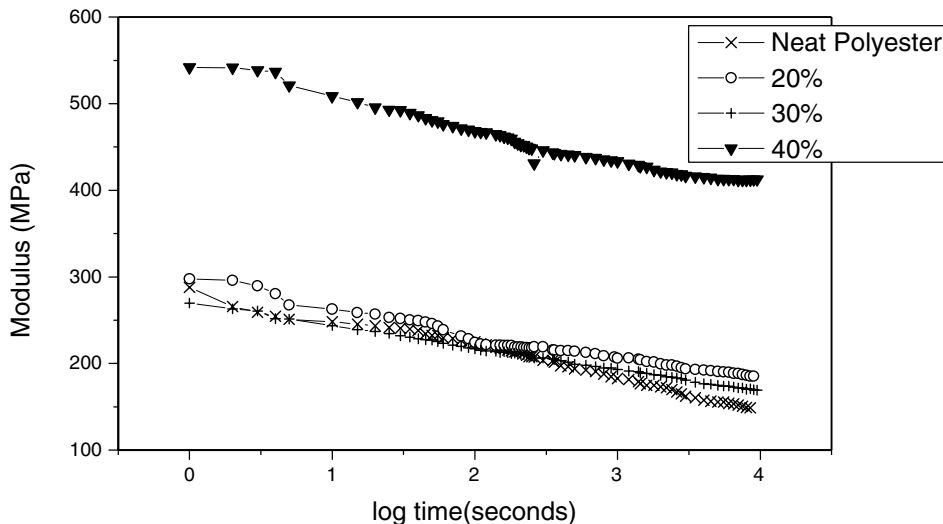


Figure 2. Stress relaxation modulus of composites with different fiber loading (strain level 2%).

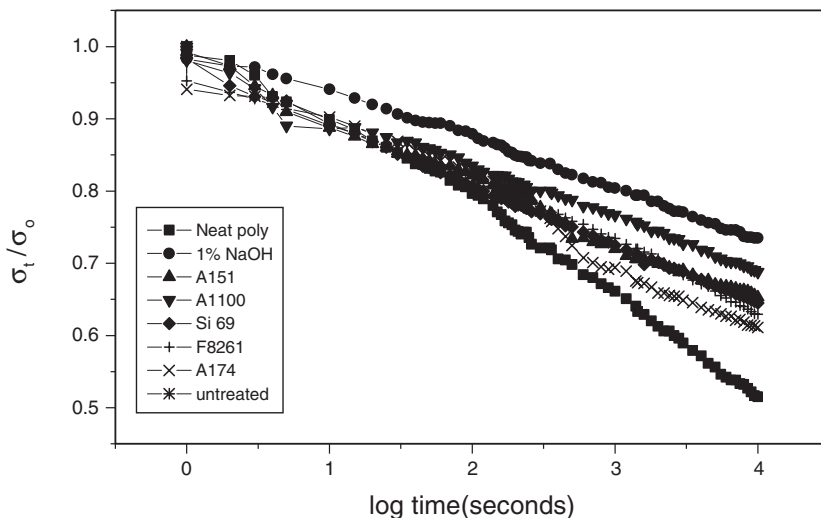


Figure 3. Stress relaxation curves of composites with different fiber treatment (strain level 2%).

Obviously, due to alkali treatment, mechanical interlocking is the predominant mode of attachment and on application of stress the mechanical bonding gets disturbed after the initial application of stress. Figure 4 gives the SEM of the alkali treated fibers. The surface of the alkali treated fibers shows a rough surface topography. In addition, the improved adhesion is evident from the polyester particles attached on the banana fibers. We have reported in an earlier paper regarding the chemical modification and the surface characterisation of banana fibers [18,24]. The reduction in the decay in stress during the initial stages of relaxation can be attributed to the improved fiber–matrix adhesion in the case of the alkali treated fibers. Unlike treatment with other coupling agents, treatment with alkali brings about the dissolution of the lignin and the hemicellulose and thereby the

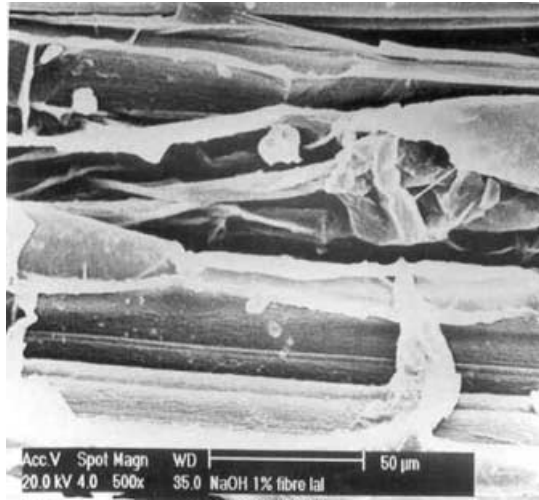


Figure 4. SEM of the alkali treated fiber showing evidence of enhanced adhesion after alkali treatment.

availability of other replaceable hydrogen atoms within the cellulose. This brings about better adhesion of the fiber and the matrix leading to lower rate of relaxation at all stages. However, in the case of composites made out of the silane treated fibers, the bonding is less effective or application of stress leads to the scission of bonds, which is established, between the fiber and the matrix. The type of bonding in silane treated composites can be explained by the reaction between silanol from the silane and the hydroxyl groups of the cellulose. Figure 5 shows the schematic representation of the bonding between the fiber and the matrix. The stress decay at the initial portion is found to be more or less the same irrespective of the chemical treatment involved. The stress relaxation is affected only after longer periods of time. After longer periods of time, on the application of the stress, bond scission occurs. There are theories in the literature regarding the formation of a flexible deformable phase between the fiber and the matrix on silane treatment [25]. The application of stress stretches this deformable layer initially. After longer periods of time, this deformable layer retracts which is felt as the increased decay in stress. The response of all the silane treated composites is more or less the same. The rate of relaxation of the different chemically modified fiber composites is compared in Table 5.

Effect of Hybridisation

Figure 6 shows the effect of hybridization on the stress relaxation of the composites. Composites with glass volume fraction 0.03, 0.15, and 0.17 were used in the preparation of composites with glass mat as the core material. The nature of the stress relaxation curve seems to be more or less the same for composites with glass volume fraction 0.15 and 0.17. Composites with a glass fiber volume fraction of 0.03 have given a stress relaxation curve with a change in the relaxation mechanism at around a time span of 250 s. The difference in the relaxation curve could be explained by the difference in the behavior of the glass fibers, which form the core. When uniaxial tensile stress is applied, because the breaking strain of glass and banana fibers is different, both the fibers behave differently. The

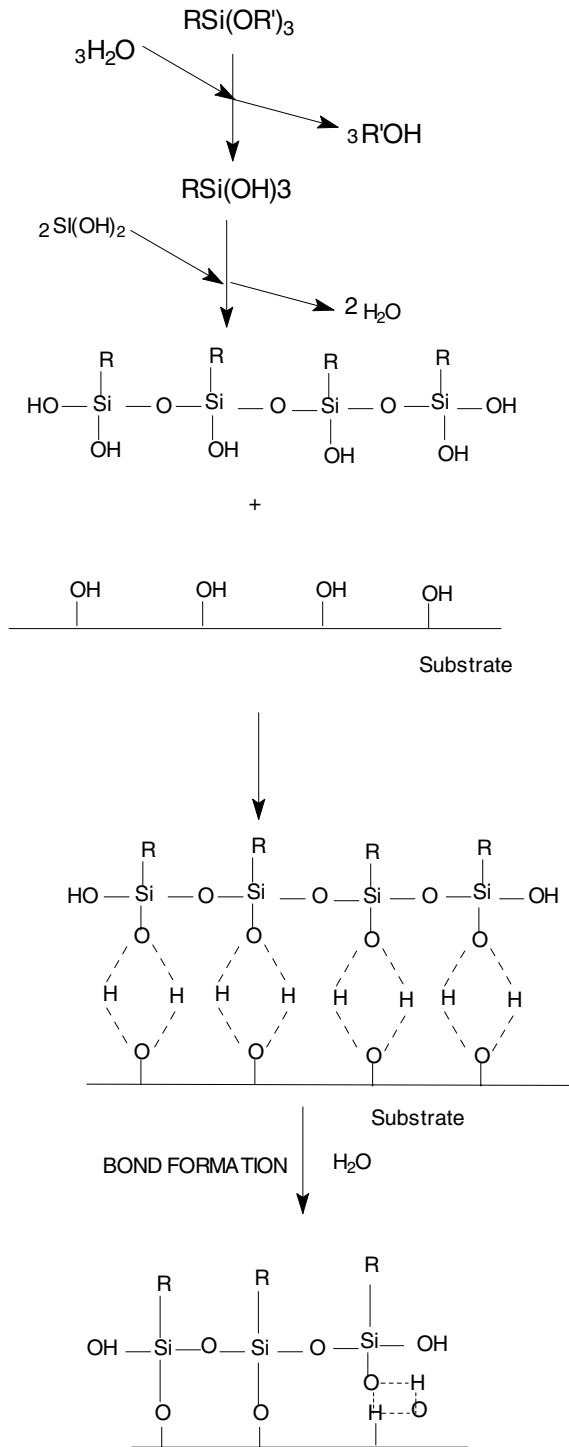


Figure 5. Schematic representation of the bonding of silane coupling agent with the fiber on treatment with silanes.

Table 5. Rate of stress relaxation in banana–polyester composites at a strain level of 2% for different chemical treatments. (Fiber loading 40%).

Range (s)	Fiber Treatment							
	A151	A174	Si69	A1100	F8261	NaOH	Neat Polyester	Untreated
$0 - 10^1$	11.65	3.82	9.41	11.8	6.87	4.15	8.45	6.93
$10^1 - 10^2$	6.5	8.07	9.9	4.08	8.10	6.34	12.39	11.17
$10^2 - 10^3$	13.59	16	10.0	9.10	9.92	8.23	17.6	7.78
$10^3 - 10^4$	6.79	11.6	10.0	10.013	14.28	8.97	21.6	4.93

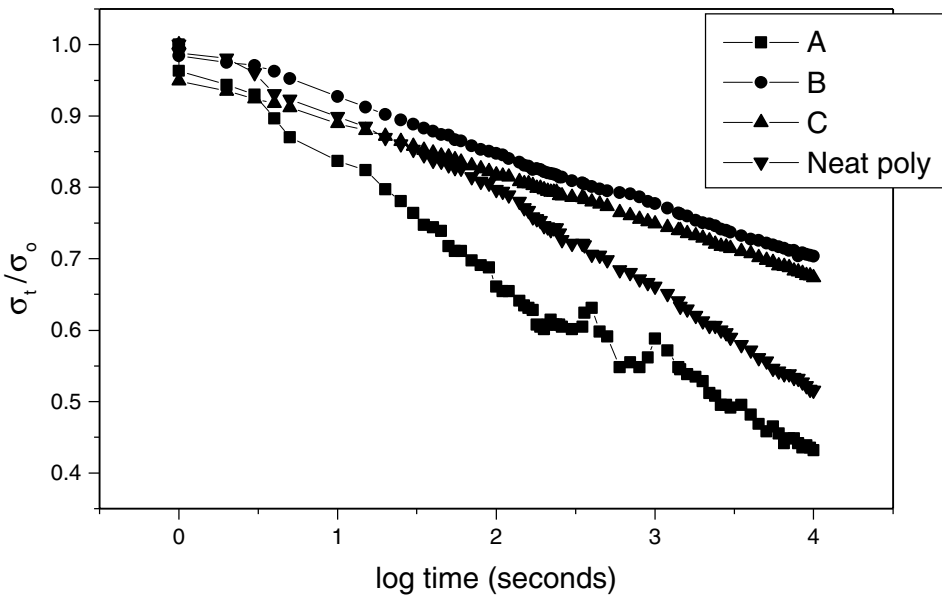
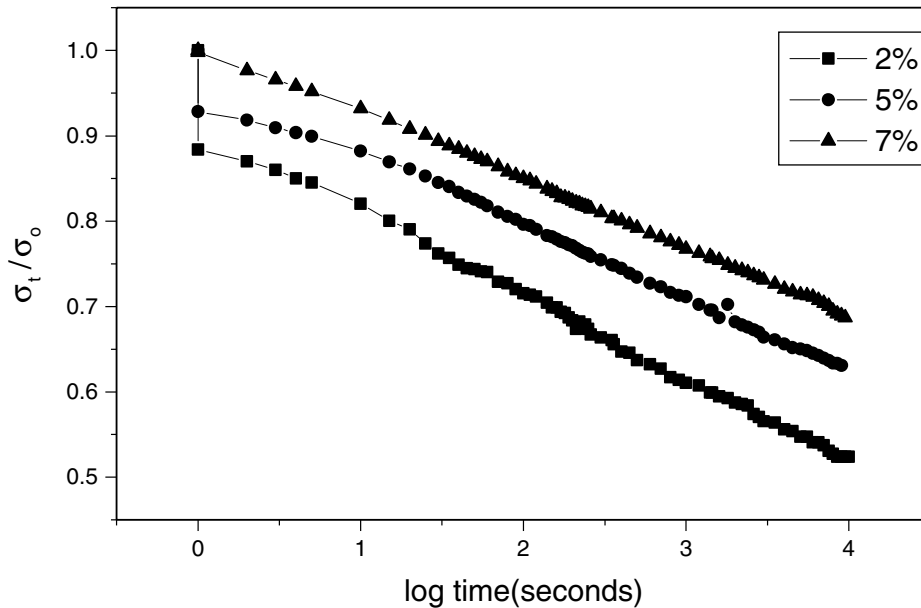


Figure 6. Stress relaxation curves of composites with different ratios of glass fiber (strain level 2%).

difference in response of the two fibers is felt prominently in the stress relaxation curve of composites with fiber volume fraction 0.03 i.e., sample marked A. Apart from the intrinsic properties of the inter-face or inter-phase, the stress distribution along the embedded fiber also plays an important role in the overall performance of the composite [25]. Glass fibers being brittle and of low elongation, behave differently from the banana fiber when subjected to tensile stress and the change in the slope of the relaxation curve can be explained by this. Other than the mechanical response of the two fibers when subjected to stress, the difference in the behavior of the interphase also affects the stress decay. Changes in the shape of the fiber tip produce maximum stress. The point of maximum stress concentration in a square ended fiber system is located a short distance from the fiber end, whilst that of a round-tip fiber system is at the fiber tip [26]. The two fibers being different in shape, another reason for the difference in response is the difference in stress concentration in the two. However, at higher glass volume fractions, the effect of the fibers oppose each other and the relaxation curve gives a single slope. Table 6 gives the rate of stress relaxation of the different hybrid composite samples.

Table 6. Rate of stress relaxation in banana–polyester hybrid composites at a strain level of 2% with different glass volume fraction.

Range (s) Slope $\times 10^{-2}$	Sample			
	Neat Polyester	A	B	C
$0 - 10^1$	8.9	16.3	7.3	11.1
$10^1 - 10^2$	12.43	21.0	8.5	8.09
$10^2 - 10^3$	16.20	11.0	8.3	8.44
$10^3 - 10^4$	21.28	26.7	9.2	10.02

**Figure 7.** Effect of strain level on the stress relaxation curves of composites.

Effect of Strain Level

Figure 7 gives the effect of strain level on the stress relaxation curve of the hybrid composite with glass volume fraction. The rate of relaxation is found to be marginally faster at 2% strain level. The slopes of the different samples vary little with strain. The stress relaxation is found to decrease with time and the relaxation is found to be highest at lower strain level. At lower strain level, the molecules have more time for rearrangement, the main relaxation in the polymer being that of physical rearrangement.

Woven Fabric Composite

The stress relaxation curves of woven fabric composites of banana and glass fabric have been investigated, as shown in Figure 8. Figure 9 shows the sketch of the weaving pattern followed by banana in the warp and bundles of glass and banana in the weft direction. The relaxation behavior has been found to be more or less the same for both samples. The rate of relaxation of the samples is found to increase towards the end. Table 7 shows the relaxation rate of the woven fabric composites at various time spans.

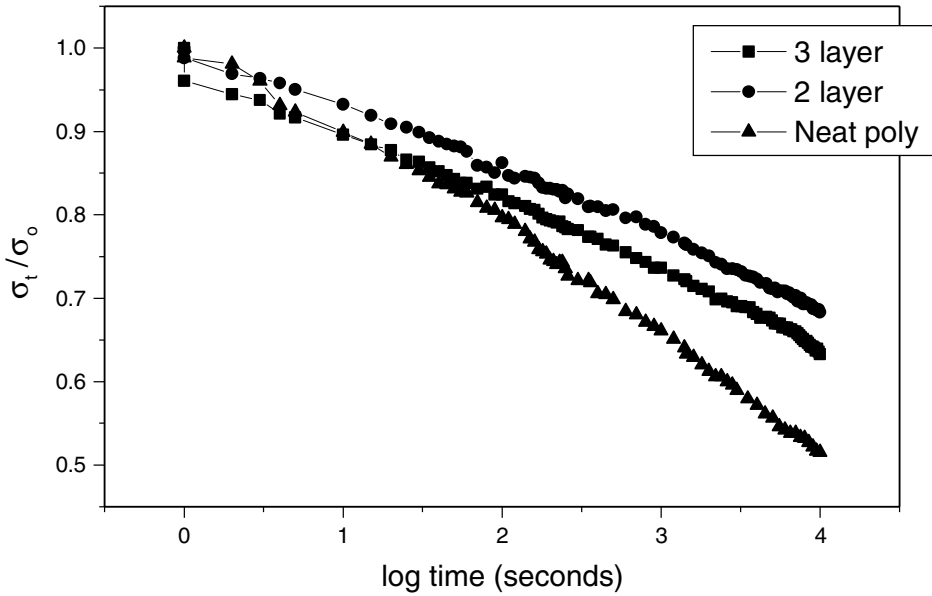


Figure 8. Stress relaxation curves of woven fabric composites. (Strain level 2%).

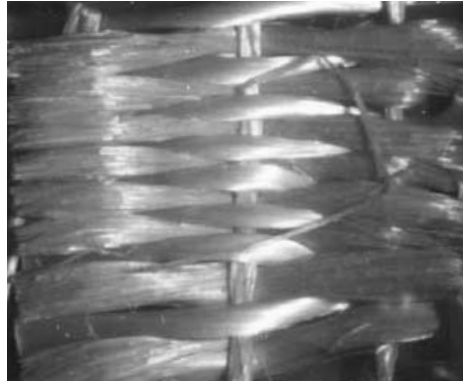


Figure 9. Sketch of the weaving pattern in the weft direction.

Table 7. Rate of stress relaxation in banana–polyester woven fabric composites at a strain level of 2% with different layering pattern.

Range (s) Slope $\times 10^{-2}$	Sample		
	2 Layer	3 Layer	Neat Polyester
0 – 10 ¹	6.76	10.41	8.45
10 ¹ – 10 ²	7.5	8	12.39
10 ² – 10 ³	9.7	10.6	17.6
10 ³ – 10 ⁴	12.24	14.05	21.6

CONCLUSION

The stress relaxation behaviors of banana fiber-reinforced polyester composites have been found to be dependent on the amount of banana fiber. The rate of stress relaxation was found to be at a maximum during the initial stages. Incorporation of fiber reduced the rate of stress relaxation and the highest reduction was observed in the case of composites with the highest fiber loading. Chemical treatment on the fibers reduced the rate of stress relaxation and the lowest rate was observed in the case of NaOH treated fiber composites. This was associated with the improved fiber–matrix adhesion due to better interlocking between the fiber and the matrix. An increase in strain level was found to reduce the rate of relaxation. In the case of woven fabric composites of glass and banana, the nature of the relaxation curve was found to be more or less the same, the highest rate being for the three layer composites. Hybridization with glass fiber reduced the relaxation rate.

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