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Environmental effects in oil palm fiber reinforced phenol formaldehyde composites: Studies on thermal, biological, moisture and high energy radiation effects

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Abstract—Accelerated weathering studies of untreated and treated oil palm fiber reinforced phenol formaldehyde composites and oil palm/glass hybrid fiber reinforced phenol formaldehyde composites were conducted. Thermal, water, biological and γ radiation effects on the composite properties were analysed. Mechanical properties such as tensile, flexural and impact properties of thermal and water aged samples were investigated. The extent of biodegradation and γ irradiation effects was estimated from variations in tensile and impact properties. The tensile and impact fracture mechanism and changes in fiber-matrix adhesion were studied using scanning electron microscopic analysis. The changes in the tensile and flexural stress-strain characteristics as well as deformation behaviour of aged composites are well explained by respective stress-strain curves. Mechanical performance of the composites decreased upon thermal ageing. Marked decrease in these properties of the composites is observed upon radiation ageing. However water immersion leads to an increase in strength properties in some treated composites such as on acetylation, silane, acrylonitrile grafting, isocyanate, permanganate treatment, alkali treatment etc. It is found that oil palm fiber increases the biodegradability of the composites.

Keywords: Oil palm fiber; weathering studies; mechanical properties; fiber treatment; biodegradability.

1. INTRODUCTION

Natural fibre reinforced polymer composites are superior to synthetic fibre reinforced composites in properties, such as enhanced biodegradability, combustibility, light weight, non-toxicity, decreased environmental pollution, low cost, ease of recyclability, etc. These advantages place the natural fibre composites among the high

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*To whom correspondence should be addressed. E-mail: sabut@sancharnet.in and sreekalams@yahoo.co.in performance composites having economic and environmental advantages. The versatile high performance applications of natural fibre composites, which can replace glass and carbon fibres, were listed in an article by Hill [1]. Vegetable fibre has a density of about one-half that of glass fibre. During the processing of natural fibre composites there will be no abrasion of the processing machines. These fibres can withstand processing temperatures up to 250°C. Reinforcement of polymers with vegetable fibres gives good opportunities for the effective utilisation of agricultural by-products. They are cent percent combustible without the production of either noxious gases or solid residues.

The growing interest in the field leads to the development of a new class of composites which are fully biodegradable, eco-efficient 'Green composites' [2]. To alleviate problems resulting from the incorporation of synthetic fibres, such as high abrasiveness, health hazards, machine wear, disposal problems, etc., incorporation of natural fibres in polymers is proposed. They are abundant, renewable, biodegradable, cheap and have low density. Oil palm fibres are highly cellulosic and can reinforce polymeric matrices. As they are organic waste materials in the palm oil industry, and would cause great environmental problems if left unutilised, their utilisation will eliminate the waste disposal problem. Owing to the versatile applications of natural fibre reinforced polymer composites, studies on the environment impact on the composites becomes significant.

The response of polymer composites to environmental exposure must be known to utilise their full potential as a structural material. Polymers often undergo degradation rapidly in outdoor applications. The environmental degradation of polymeric materials includes thermal, mechanical, photochemical, radiation, biological, and chemical degradation. Polymer degradation is mainly caused by chemical bond scission reactions in macromolecules [3]. Several studies have been reported on the thermal, photo, biological and outdoor weathering of polymer composites [4–8]. It was found that long-term exposure of the composites to elevated conditions affect the mechanical properties. Recently Hancox reviewed the effects of temperature and environment on the performance of polymer matrix composites [9].

Natural fibres are susceptible to moisture, temperature, high energy radiation and attack from bioorganisms. Most of the natural fibres are stable up to 300°C. Hence incorporation of natural fibres into plastics changes their environmental stability. Phenolic resins are highly heat resistant and they find applications in aerospace as ablative heat shields for protection of space vehicles.

The present work describes various ageing effects on the mechanical properties of oil palm fibre reinforced phenol formaldehyde composites. Thermal, water, boiling water, γ-irradiation and biodegradation effects are investigated.

2. MATERIALS AND EXPERIMENTAL

The oil palm empty fruit bunches were obtained from Oil Palm India Limited, Kottayam, Kerala, India. The oil palm empty fruit bunch (OPEFB) fibres were processed by retting method. They were cleaned off from pithy materials and dried. The phenol formaldehyde resole type resin was used for composite preparation, which was supplied by West Coast Polymers Pvt. Ltd., Kannur, Kerala, India. The resin has been prepared under alkaline conditions with a molar excess of formaldehyde to phenol. Chopped E-glass strand mat was used for hybrid composite preparation and was procured from Ceat Ltd., Hyderabad, India. The glass fibre mat used was chopped strand mat made up of glass fibre bundles of 50 mm length. The physical, chemical and mechanical properties of oil palm fibre, glass and neat phenol formaldehyde resin are given in Tables 1a and 1b. Chemicals used for the fibre surface modifications (sodium hydroxide, acetic acid and acetic anhydride) were of reagent grade. NR latex having 10% DRC content was used for latex coating on the fibers. The coupling agents used were toluene 2,4-diisocyanate and triethoxyvinyl silane. They were supplied by PolySciences, USA and Union Carbide Co., Montreal, Canada, respectively.

2.1. Fiber treatment

2.1.1. Mercerisation. Fibres were immersed in 5% solution of sodium hydroxide for 48 h at room temp. Fibres were then washed many times in distilled water and finally washed with water containing little acid and dried.

2.1.2. Acetylation. Fibres were pre-treated with 2% sodium hydroxide for about half an hour in order to activate the —OH groups on the cellulose and lignin. Fibres were subjected to acetylation with acetic anhydride in acetic acid medium. Conc. H_2SO_4 acts as a catalyst in this reaction. Fibres were washed in distilled water and then dried.

Table 1a.
Physical and chemical characteristics of glass and oil palm fibers

	Lignin content (%)	Cellulose content (%)	Density (g/cc)	Average diameter (μm)	Ash content (%)
Oil palm fiber	19	65	0.7–1.55	150–500	2
Glass fiber bundle in chopped strand mat	—	—	2.54	400	—

Table 1b.
Mechanical properties of raw materials used for composite preparation

	Tensile strength (MPa)	Young's modulus (MPa)	Elongation at break (%)	Flexural strength (MPa)	Flexural modulus (MPa)	Izod impact strength (kJ/m^2)
PF Resin (Resole)	10	375	2	10	1875	20
Oil palm fiber	100–400	1000–9000	8–18	—	—	—

2.1.3. Peroxide treatment. Fibres were coated with benzoyl peroxide from acetone solution after alkali pre-treatment. Saturated solution of the peroxide in acetone was used. Finally, the fibres were dried.

2.1.4. Permanganate treatment. Fibres were pre-treated with alkali and then dipped in permanganate solution in acetone for about 2–3 min. Permanganate solutions of concentrations 0.01, 0.05 and 0.1% were used. Fibres were washed in distilled water and finally dried.

2.1.5. Radiation treatment. Fibres were exposed to ^{60}Co γ radiation at a dose rate of 0.1 Mrad per hour for about 30 h.

2.1.6. Isocyanate treatment. The alkali treated fibres were taken in a round-bottomed flask and soaked in chloroform containing dibutyl tin dilaurate catalyst. Toluene diisocyanate was added dropwise into the flask using a pressure equalising funnel. The reaction was allowed to take place for two hours with continuous stirring. Fibres were purified by refluxing with acetone and then washed with distilled water and dried in oven.

2.1.7. Silane treatment. The pre-treated fibres were dipped in alcohol water mixture (60 : 40) containing triethoxyvinyl silane coupling agent. The pH of the solution is maintained in between 3.5 and 4. Fibres were washed in distilled water and dried.

2.1.8. Acrylation. Fibres were mixed with 10% NaOH for about 30 min at room temperature. The solution was decanted and the wet product was treated with solution containing different concentrations of acrylic acid. The reaction was allowed to proceed for about one hour at 50°C. Fibres were washed with water/alcohol mixture and dried.

2.1.9. Acrylonitrile grafting. Fibres are bleached with 2% alkali for 30 min and then oxidised with 0.02 ml^{-1} $KMnO_4$ (liquor ratio, 1 : 150) for 10 min. The fibres were further washed with water and then put into 1% H_2SO_4 containing acrylonitrile in the ratio 30 : 1. The sample is then placed in a thermostatic water bath, the temperature of which was kept at 50°C for 2 h without any disturbance. The sample was then washed with water thoroughly and then dried. The grafted fibre was isolated from any homopolymer formed (PAN) by soxhlet extraction using dimethyl formamide.

2.1.10. Latex modification. Fibres were given a latex coating by dipping it into natural rubber latex having 10% dry rubber content after pre-treatment with NaOH.

2.2. Preparation of composites

OPEFB fibre was used for the composite preparation. Hand lay-up method followed by compression moulding at 100°C for about 30 min was adopted for composite fabrication. First, fibres were chopped to desired lengths and randomly oriented fibre mat was prepared. The prepreg route was followed for the composite preparation. Oil palm fibre/PF composites with 40 mm fibre length and fibre loading, 10, 20, 30, 40 and 50 wt% were prepared. Composites were prepared with treated fibres having 40 wt% fibre loading and 40 mm fibre length. Hybrid composite was prepared by arranging randomly oriented short glass and oil palm fibre mats as alternate layers. The volume fraction ratio of glass/oil palm fibre in hybrid composite was 0.3 : 0.7.

2.3. Ageing studies

2.3.1. Thermal ageing. Composite samples were cut into specified dimension according to ASTM standards for mechanical testing. It was then placed in an air oven for about three days at 100°C. Samples were then allowed to cool at room temperature.

2.3.2. Water ageing. Composite samples were kept in distilled water at room temperature for two weeks to attain a saturation level. Samples were taken out and air dried.

2.3.3. Boiling water ageing. Samples were kept in boiling water for 2 h, then taken out and air dried.

2.3.4. Radiation ageing. Composite samples were irradiated from a ^{60}Co γ-ray source. The samples were given 0.1, 1, 10, 50, and 100 Mrad doses.

2.3.5. Biodegradation. The composite samples were inoculated with two fungi *Tricoderma* and *Aspergillus*. Inoculation was done at the cut portions of the composites and the samples were kept in a moist environment. Samples were kept for several months under the same condition.

2.4. Mechanical tests

Tensile and flexural properties were tested on a FIE electronic tensile testing machine TNE-500 at a cross-head speed of 50 mm/min. Tensile, flexural and izod impact test specimens having dimension 120 × 12 × 2.5 mm size were cut from the composite sheets. Tensile testing was carried out according to ASTM D 638-76. The tensile testing of the composites was done at a gauge length of 40 mm. The tensile load–elongation curve was plotted. The tensile stress at any point is determined as

$$TensileStress = \frac{F\,(\mathrm{N})}{A\ (\mathrm{mm}^2)},$$

where F is the applied force in N, A is the area of cross-section in mm^2. The tensile strain at any point of stress is calculated as

$$TensileStrain\ (\%) = \frac{IncreaseInElongation}{GaugeLength} \times 100.$$

Three-point flexure properties were tested according to ASTM D 790. A load–deflection curve was plotted. The flexural stress at a particular load was calculated as follows.

$$FlexuralStress = \frac{3PL}{2bd^2},$$

where P is the applied load, L is the gauge length, b is the breadth of the specimen and d is the thickness of the specimen. The flexural strain at any point of stress is calculated as follows.

$$FlexuralStrain = \frac{6Dd}{L^2},$$

where D is the deflection, d is the thickness of the specimen and L is the gauge length.

Unnotched izod impact strength of the composites was measured on an impact tester from Ceast Torino, Italy according to ASTM D 256.

2.5. Scanning electron microscope studies

Scanning electron micrographs of the untreated and treated fibre surfaces were taken to study the fibre surface morphology. The tensile and impact fracture mechanisms and interface adhesion of the composites were analysed by scanning electron microscopy.

3. RESULTS AND DISCUSSION

Characterisation of the treated fibre surface were done by Scanning Electron Microscopy and Infrared Spectroscopy and were reported elsewhere [10].

3.1. Ageing effects on tensile properties

The tensile stress–strain behaviour of unaged and aged composites are given in Figs 1–6. The unaged composite shows necking followed by catastrophic failure (Fig. 1). Alkali and permanganate treatments improve the fibre–matrix interaction leading to the brittle fracture of the composites for which the stress–strain curves are evidence. Presence of oil palm fibre in hybrid composites makes the composite

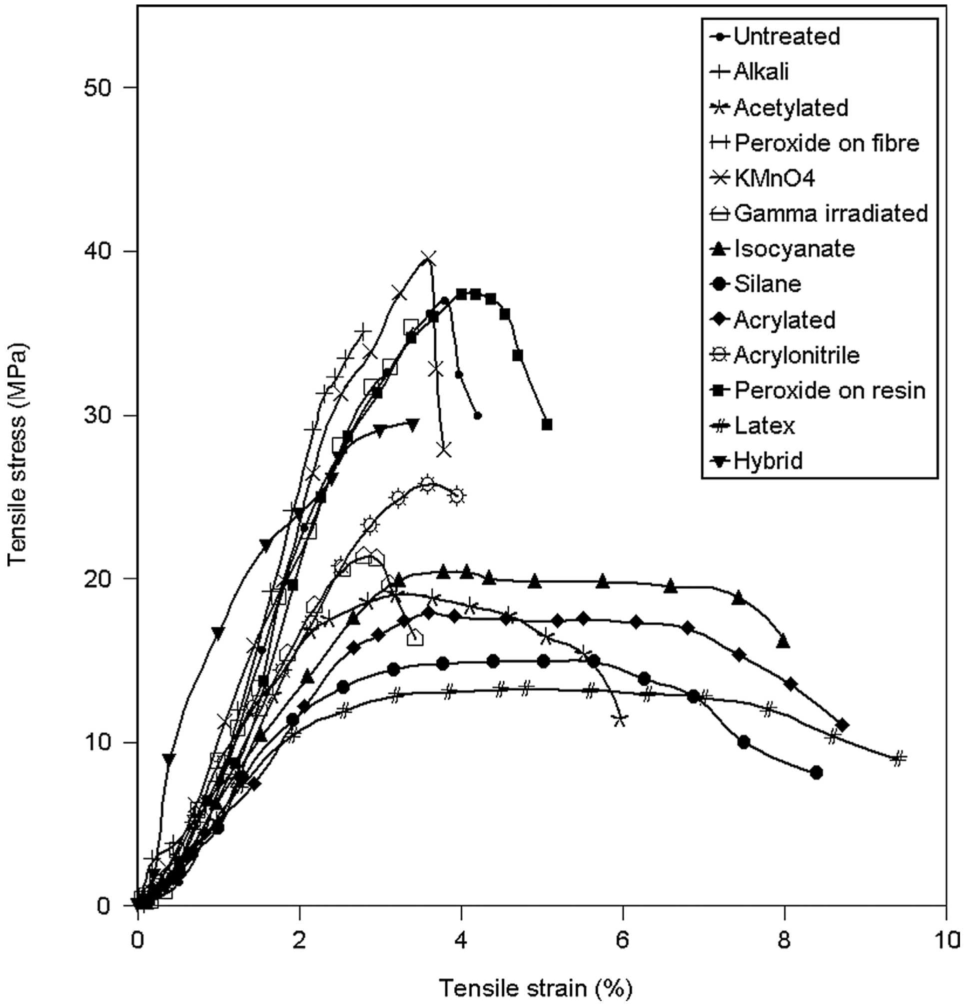

Figure 1. Tensile stress–strain characteristics of unaged composites.

less brittle. Treatments such as peroxide on resin, acrylonitrile grafting and gamma irradiation cause a necking effect. The stress–strain curves of unaged, isocyanate, acrylated, acetylated, silane and latex treated composites show major slope change at an early stage. These composites exhibit yielding and higher extensibility. A dramatic change in the stress–strain behaviour of the composites was observed upon thermal ageing (Fig. 2). Except latex treated composites all composites show clear brittle failure. Crack propagation in a thermal aged composite is faster, which leads to catastrophic failure of the system. Fibre breakage will also more easily in a thermal aged composite. In latex treated composite, latex can form a protective coating on the fibre surface. Crack propagation can be hindered by the presence of elastic phase and leads to a higher elongation. On water immersion the necking effect becomes more prominent (Fig. 3). In this case the decreased brittle nature

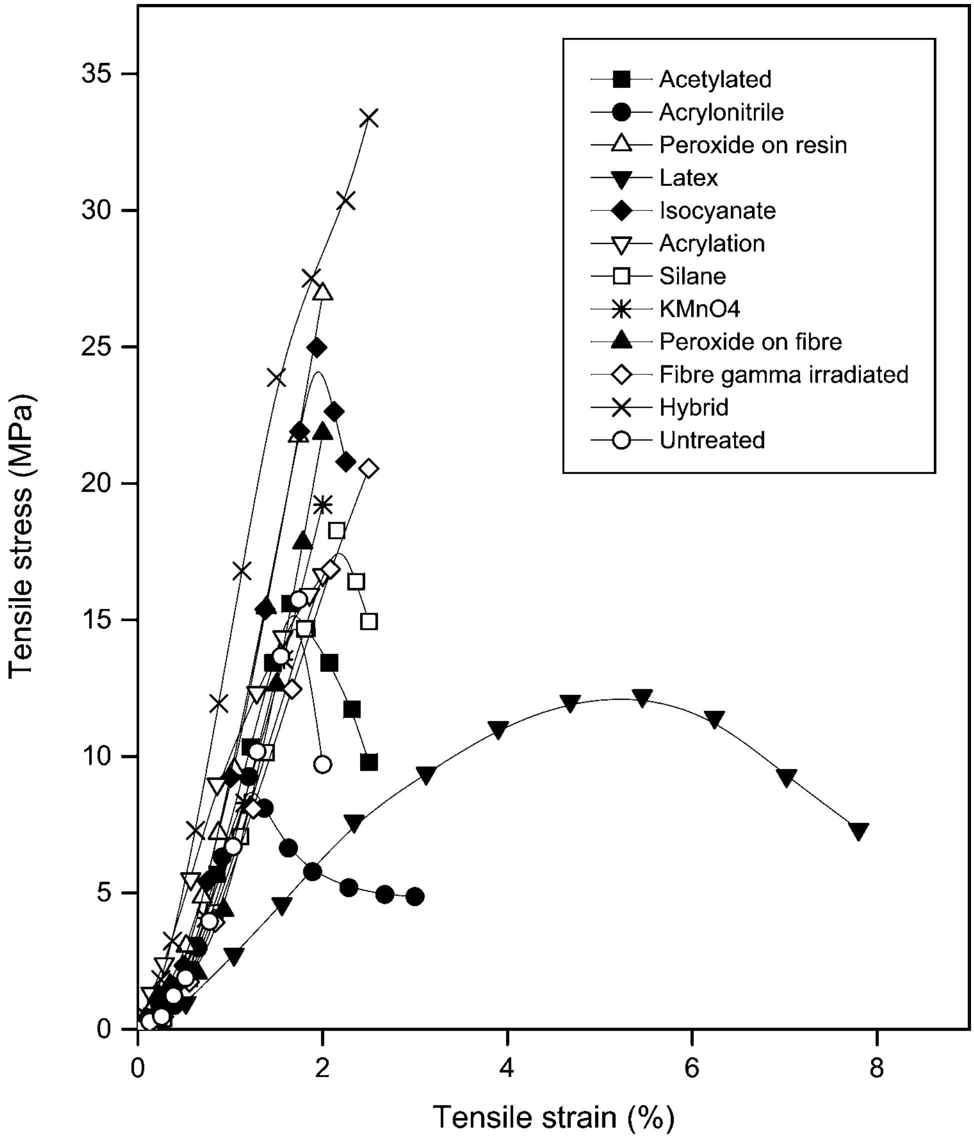

Figure 2. Tensile stress–strain characteristics of thermal aged composites.

is attributed to the plasticising effect of water in composites. Effect of boiling water ageing on the tensile stress–strain behaviour of the selected composites is given in Fig. 4. Catastrophic failure occurred in untreated and in peroxide treated fibre composites. It is found that biological effects of the fungi affect the stress–strain behaviour of the composites. A prominent necking effect is observed in silane treated composite upon biodegradation (Fig. 5). Necking is a local reduction in cross-sectional area and corresponds to a drop in load observed at the yield point. An unstable necking is observed in the present case where the original

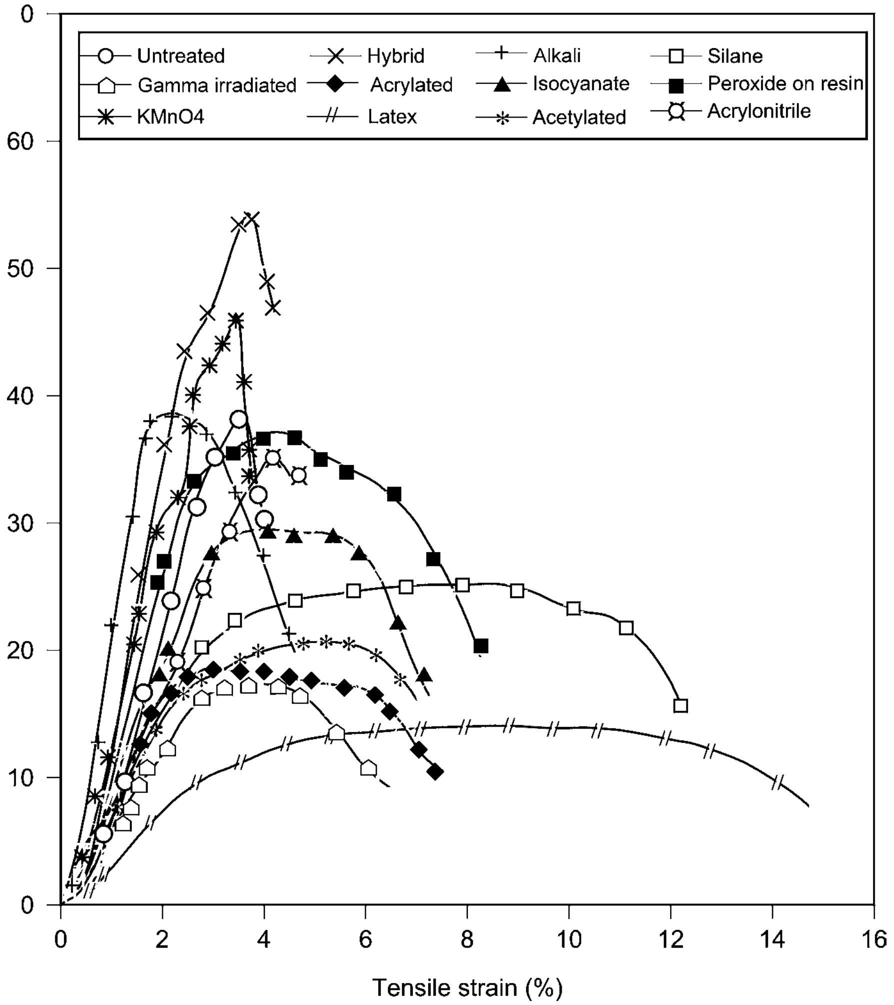

Figure 3. Tensile stress–strain characteristics of cold water aged composites.

neck continues to get thinner, the load decreases and eventually failure of the fibre occurs. The radiation effects on the stress–strain behaviour are evident from Fig. 6. Gamma irradiated acrylated composite shows exceptionally high elongation. Hybrid composite shows higher brittle behaviour.

The effect of ageing on the tensile strength values of the composites is given in Table 2. Untreated and most of the other treated composites show changes in properties upon thermal ageing, biodegradation and radiation ageing. However, on water immersion the tensile strength is retained and in most cases enhancement in strength value is observed. This is due to the swelling of the fibre on water immersion thereby decreasing the void size at the fibre–matrix interphase. This can exert a radial pressure leading to higher tensile strength values. Among water aged composites, boiling water aged composites show comparatively lower values.

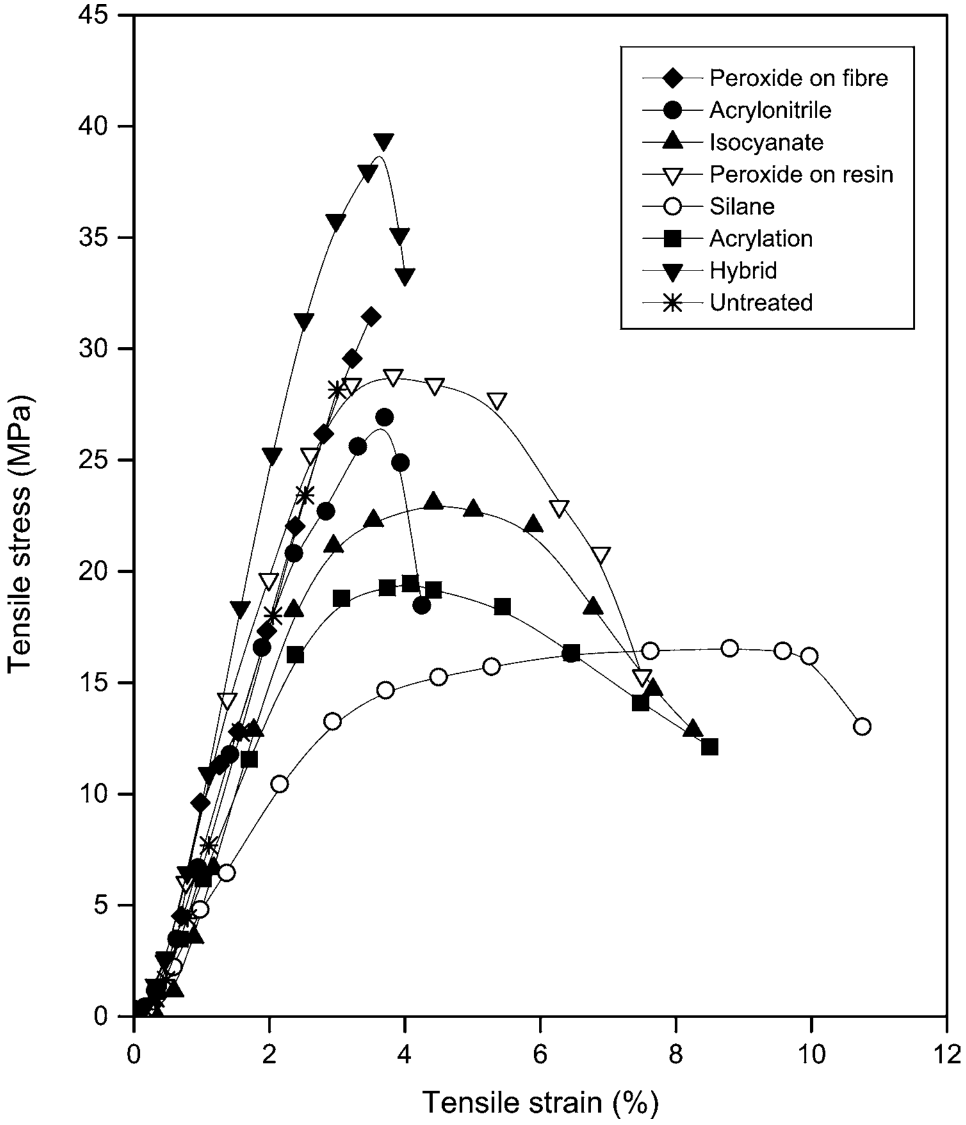

Figure 4. Tensile stress–strain characteristics of boiling water aged composites.

Enhancement in strength properties of various natural fibre reinforced polymer composites upon water immersion was reported [11, 12]. The retention in tensile properties can be attributed to the plasticising effect of water and better fibre–matrix adhesion at lower water absorption stage. Thermal degradation occurred in composites except hybrid, isocyanate and silane treated composites. Glass fibre is stable above 900°C without any weight reduction. Major degradation in phenolics occurs at 500°C. Oil palm fibres were found to be stable up to 325°C. Upon thermal ageing, degradation in interface properties may occur from the

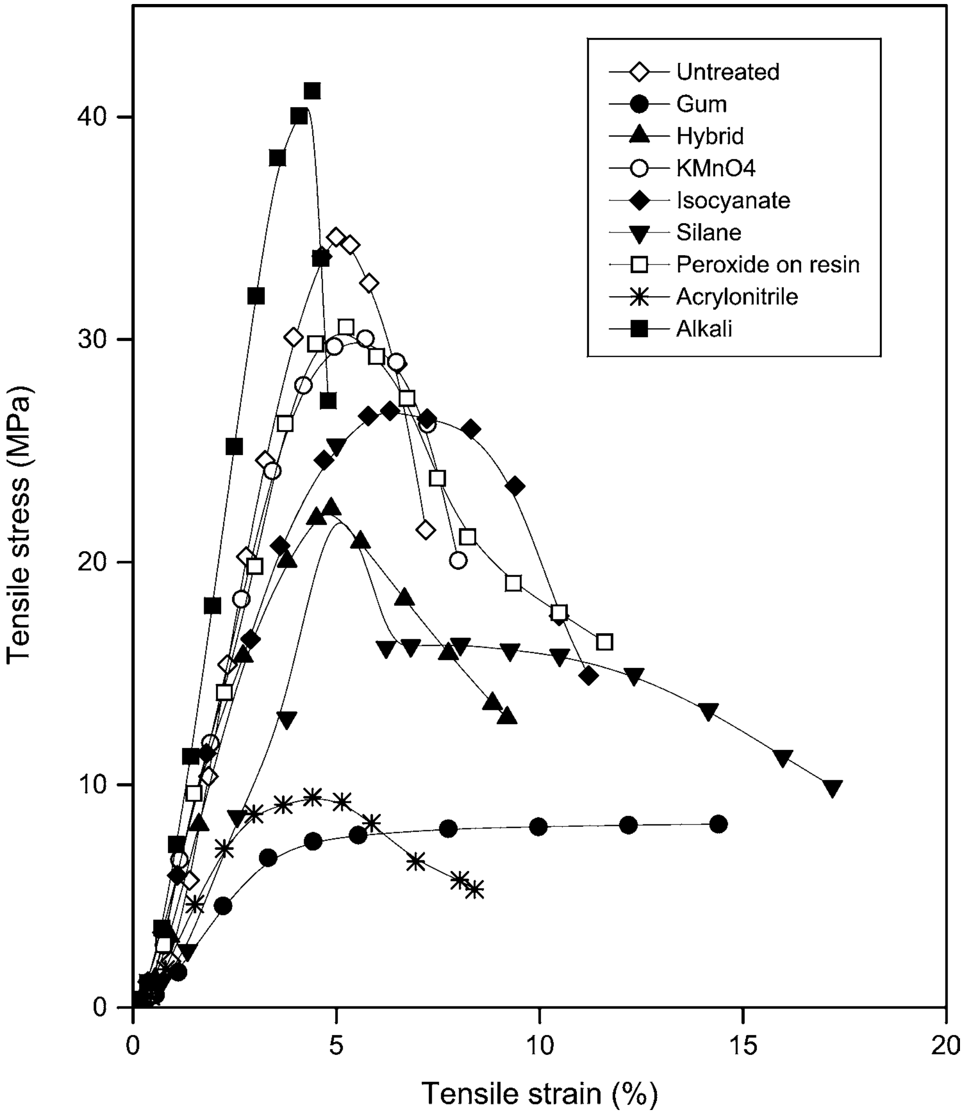

Figure 5. Tensile stress–strain characteristics of biodegraded composites.

decrease in fibre–matrix adhesion. Physical interaction between the fibre and matrix decreases owing to the shrinkage of the oil palm fibre in thermal environment. But fibre coatings like silane and protective layers by glass etc. can eliminate this type of thermal deterioration in composites. Among the biodegraded composites, isocyanate, silane and alkali treated composites show enhanced tensile strength values. Presence of moisture can lead to a more stable system by plasticising effect. Studies have been reported on the degradation behaviour in various thermosetting composites under thermal and chemical environments [13, 14]. It was found that thermal degradation is a complex phenomenon that changes thermal and mechanical

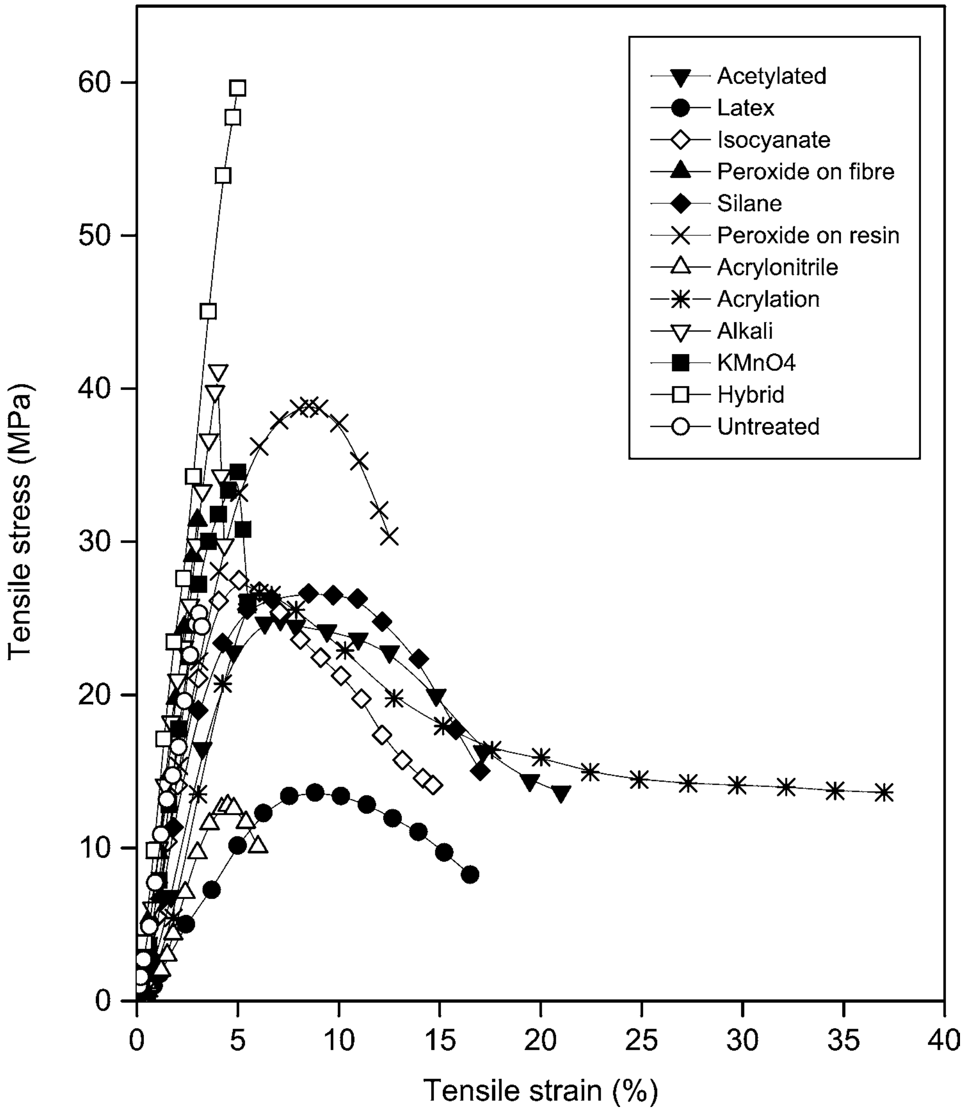

Figure 6. Tensile stress–strain characteristics of gamma irradiated composites.

properties of the material in an irreversible form. A small dose of gamma irradiation on composite increases the tensile strength, except in untreated and resin peroxide treated composites. However, at higher doses of radiation, the strength decreased considerably. It is reported that gamma irradiation of polymers can result in chain scission, molecular weight changes, molecular rearrangements, free radical initiation, etc. [15]. The free radicals formed can enhance the crosslinking between the fibre and matrix thereby enhancing the strength. But at higher radiation doses, disintegration of fibre and matrix causes degradation in properties. Similar results have been reported in carbon fibre reinforced epoxy composites in which case the

Table 2.
Tensile strength values (MPa) of unaged and aged composites

Sample	Unaged	Thermal aged (100 °C; 72 h)	Cold water aged (2 weeks)	Boiling water aged (2 h)	Biodegradation (8 months)	Gamma irradiation				
						Dose (Mrad)				
						0.1	1	10	50	100
Untreated	37	16	38	28	35	28	25	20	5	6
Hybrid	30	33	54	39	22	39	60	45	40	33
Acetylated	19	16	21	—	—	22	19	24	14	11
Acrylonitrile	26	9	35	27	9	29	13	29	10	11
Peroxide on resin	37	27	37	29	31	27	39	32	26	18
Latex	13	12	14	—	—	16	14	18	13	11
Isocyanate	20	25	29	23	27	22	23	28	21	10
Acrylation	18	17	19	19	—	20	27	22	12	10
Silane	15	18	25	17	16	25	27	20	14	11
$KMnO_4$	40	19	46	—	30	46	35	32	19	16
Mercerisation	35	—	39	—	41	40	41	34	31	30
Peroxide on fiber	35	22	—	31	—	33	31	33	30	12
Fiber γ irradiated	21	21	17	—	—	—	—	—	—	—

ultimate tensile strength keeps its value until the first influence and then falls to 60–70% at higher dose levels [16]. Also, in graphite reinforced epoxy composite, the graphite fibre strength is not affected by radiation, but the tensile properties of the epoxies were adversely affected by the radiation [17]. However, in this system, it is reported that the interfacial shear strength value increases with the radiation dose.

Variation in elongation at break of the composites upon various ageing is given in Table 3. The elongation of the composites decreased upon thermal ageing. In most cases the values show enhancement upon water ageing and biodegradation. The decreased elongation upon thermal ageing is due to the decreased elongation of the fibre on thermal degradation. It has been found already that the elongation at break of oil palm fibre considerably enhanced upon water immersion [18]. The intrinsic elongation behaviour of the fibre on water ageing leads to composites with higher elongation. Gamma irradiation increases the elongation of the composites. Changes in the fibre–matrix interactions are responsible for the enhancement in elongation of the composites. However, in the case of thermoplastics such as polyethylene, a decrease in elongation at break is reported with absorbed dose of high energy ion irradiation [19].

The tensile moduli of the composites at 1% strain of the unaged and aged composites are compared in Table 4. Thermal ageing leads to enhanced modulus in acetylated, peroxide treated resin, isocyanate, acrylated and silane treated composites. Increased temperature leads to increase in cross-linking, which increases the modulus of the composites. The stiffness of the peroxide, isocyanate,

Table 3.
Elongation at break values (%) of unaged and aged composites

Sample	Unaged	Thermal aged (100 °C; 72 h)	Cold water aged (2 weeks)	Boiling water aged (2 h)	Biodegradation (8 months)	Gamma irradiation				
						Dose (Mrad)				
						0.1	1	10	50	100
Untreated	4	2	4	3	7	8	5	5	26	21
Hybrid	4	3	4	4	9	12	5	6	6	7
Acetylated	6	3	7	—	—	11	15	11	5	25
Acrylonitrile	4	3	5	4	8	9	6	23	4	26
Peroxide on resin	5	2	8	8	12	11	13	5	5	10
Latex	9	8	15	—	—	18	17	11	4	9
Isocyanate	8	2	7	8	11	9	11	8	6	3
Acrylation	9	2	8	9	—	12	37	13	15	12
Silane	8	3	12	11	17	20	17	15	5	3
$KMnO_4$	4	2	4	—	8	8	6	6	5	4
Mercerisation	3	—	5	—	5	4	4	4	4	3
Peroxide on resin	3	2	—	4	—	6	3	6	5	18
Fiber γ irradiated	3	3	7	—	—	—	—	—	—	—

Table 4.
Tensile modulus values (MPa) of unaged and aged composites

Sample	Unaged	Thermal aged (100 °C; 72 h)	Cold water aged (2 weeks)	Boiling water aged (2 h)	Biodegradation (8 months)	Gamma irradiation (1 Mrad)
Untreated	746	600	688	702	286	873
Hybrid	1665	1438	1217	1010	412	1223
Acetylated	682	740	633	—	—	304
Acrylonitrile	810	670	605	758	259	164
Peroxide on resin	685	938	1022	996	552	528
Latex	575	262	299	—	—	136
Isocyanate	660	938	828	535	552	528
Acrylation	468	623	661	632	—	234
Silane	532	542	633	521	189	500
$KMnO_4$	1045	646	1189	—	552	667
Mercerisation	853	—	2107	—	678	947
Fiber γ irradiated	746	530	578	—	—	—
Peroxide on fiber	858	565	—	954	—	947

acrylated, silane, permangate and alkali treated composite increased upon water ageing. Upon biodegradation, decrease in modulus is observed in all systems. The modulus of the untreated composite is enhanced on gamma irradiation. Peroxide on fibre and alkali treated composite also showed enhanced modulus values upon gamma irradiation.

3.2. Tensile fracture mechanism

Tensile fracture mechanism in aged condition is studied by scanning electron microscopy. Tensile fractograph of water aged alkali treated composites is given in Fig. 7. Better fibre–matrix interaction is evidenced from the fracture surface. Major failure occurred by fibre breakage. Effect of gamma radiation on the matrix failure is clear from the scanning electron micrograph of tensile fracture of PF gum sample (Fig. 8). Multiple fracture arising from the disintegration of the sample by radiation is evident from this figure. Tensile fracture of gamma irradiated alkali treated composite shows matrix cracking (Fig. 9). Brittle fibre failure is observed here. In gamma ray irradiated permanganate treated composite, fibrillation is observed (Fig. 10). Fibre–matrix debonding and ductile failure of the fibre is seen. In hybrid composites, radiation causes disintegration. This is clear from the micrograph (Fig. 11). Glass fibre–matrix debonding becomes easier upon radiation ageing, which is evident from the debonded paths found in the micrograph.

3.3. Ageing effects on the flexural properties

The flexural stress–strain behaviour of unaged, thermal aged and water aged composites is shown in Figs 12–15. Due to the presence of glass fibre, the hybrid composite undergoes catastrophic failure upon application of flexural force, which is a combination of tension and compression. Fibre exposed to gamma

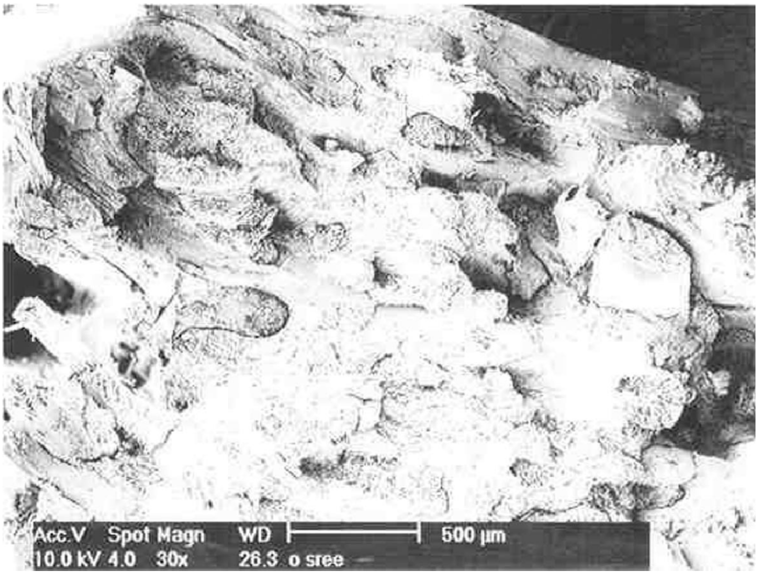

Figure 7. SEM of tensile fracture of cold water aged alkali treated oil palm fiber/PF composite (×30).

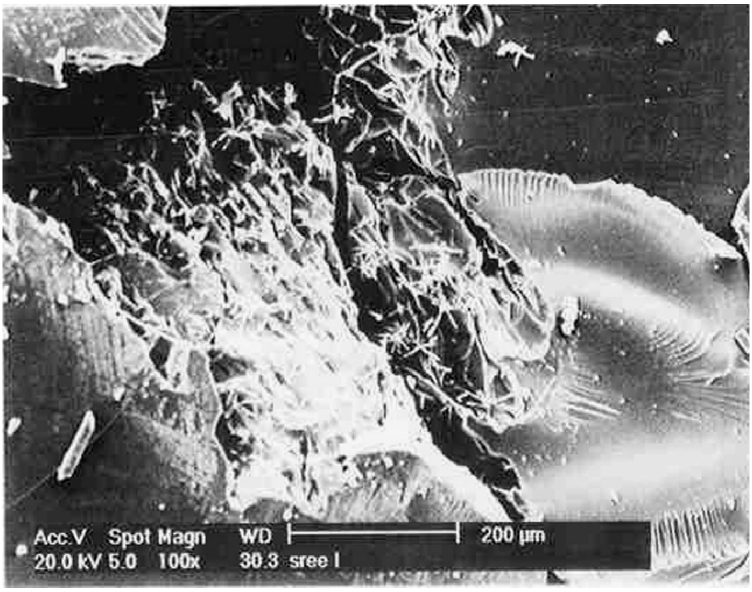

Figure 8. SEM of tensile fracture of gamma irradiated neat PF sample (×100).

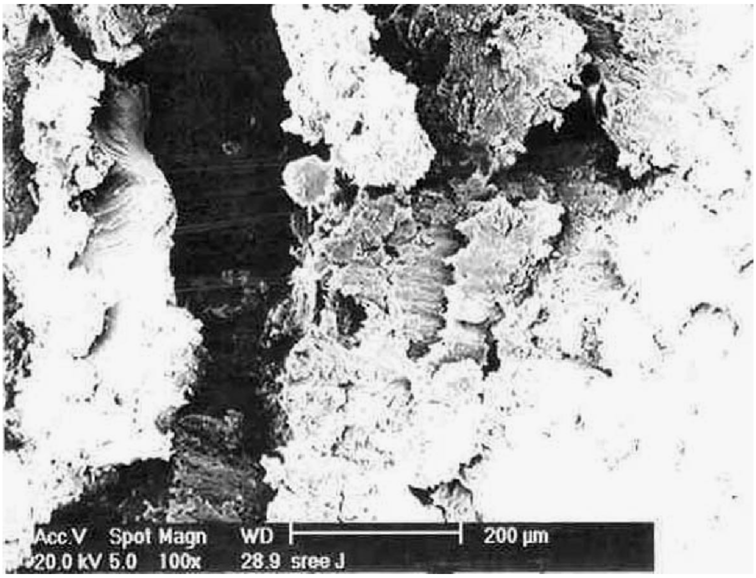

Figure 9. SEM of tensile fracture of gamma irradiated alkali treated oil palm fiber/PF composite (×100).

irradiation and permanganate treated composites failed at lower flexural strain in unaged condition. On thermal ageing, the failure strain decreased to a very low value. However, in thermal aged composites, acetylated and gamma irradiated fibre reinforced composites withstand higher strain (Fig. 13). On water ageing,

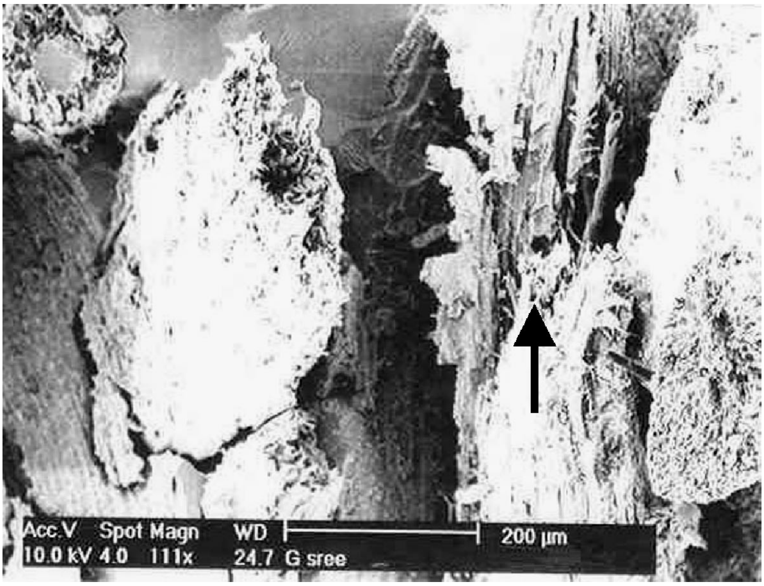

Figure 10. SEM of tensile fracture of gamma irradiated permanganate treated oil palm fiber/PF composite (×111).

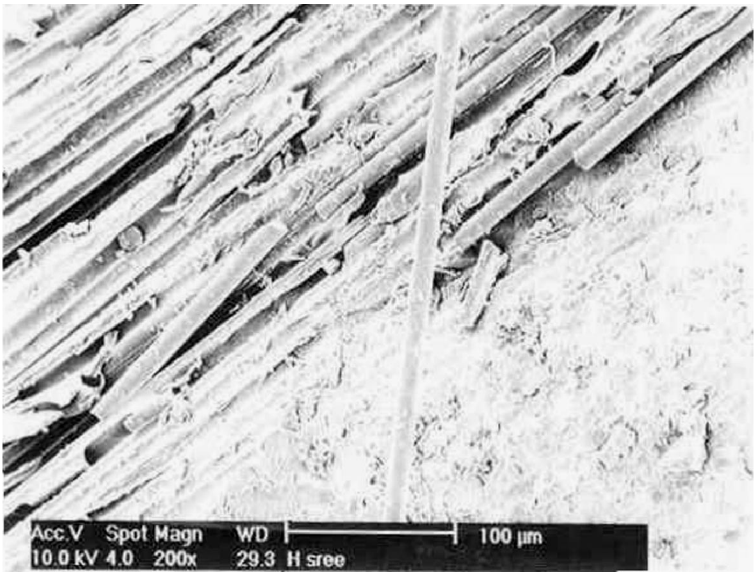

Figure 11. SEM of tensile fracture of gamma irradiated oil palm/glass hybrid fiber PF composite (×200).

the composite became more flexible due to the presence of water and therefore less brittle behaviour is observed (Figs 14 and 15).

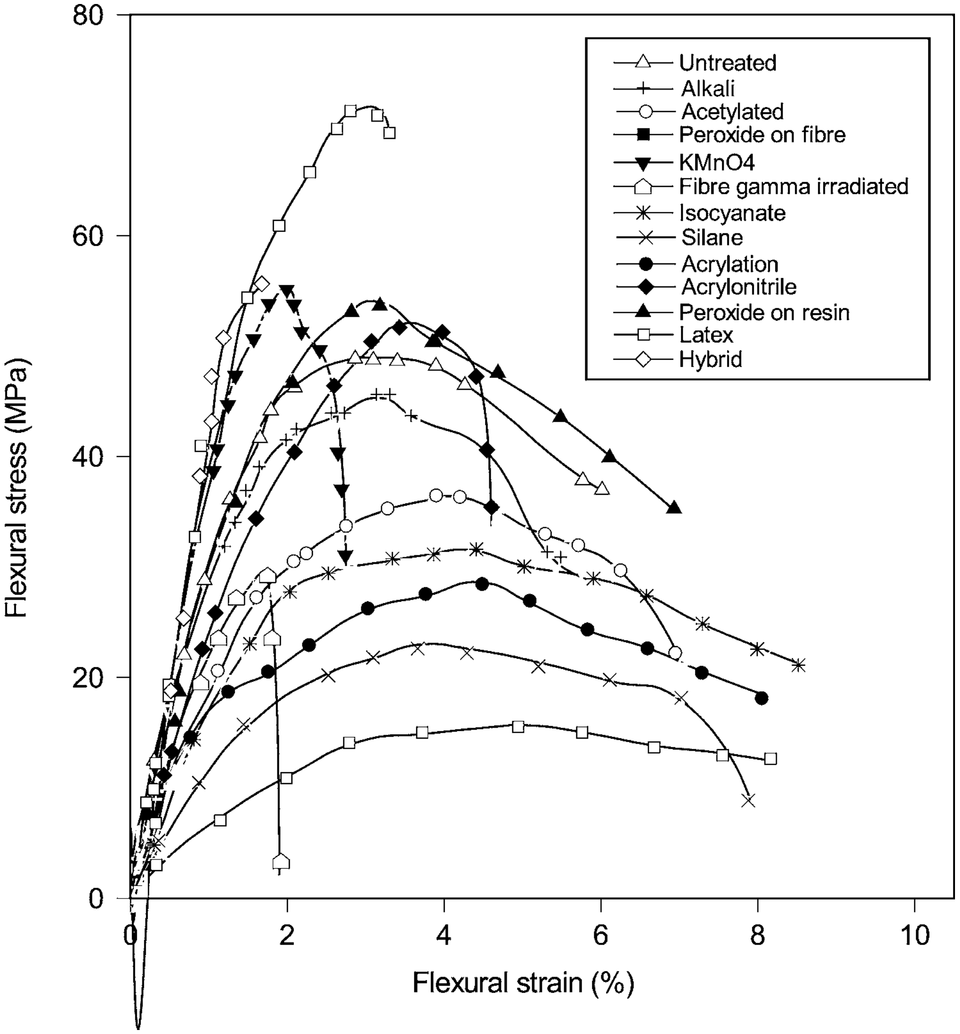

Figure 12. Flexural stress–strain characteristics of unaged composites.

Variation of flexural strength on thermal and water ageing is given in Table 5. Flexural strength of untreated composites decreased upon ageing. The flexural properties of peroxide, latex modified and acrylated fibre composites showed increase upon thermal ageing. However, untreated and most of the treated composites show decreased flexural performance on water ageing. Flexural modulus shows a similar trend (Table 6). In the case of hybrid composites, cold water ageing increased the flexural properties. Water can act as a plasticizer at the fibre–matrix interface. The absorbed water affects the interlaminar properties of the composites and hence the compressive failure of the composites.

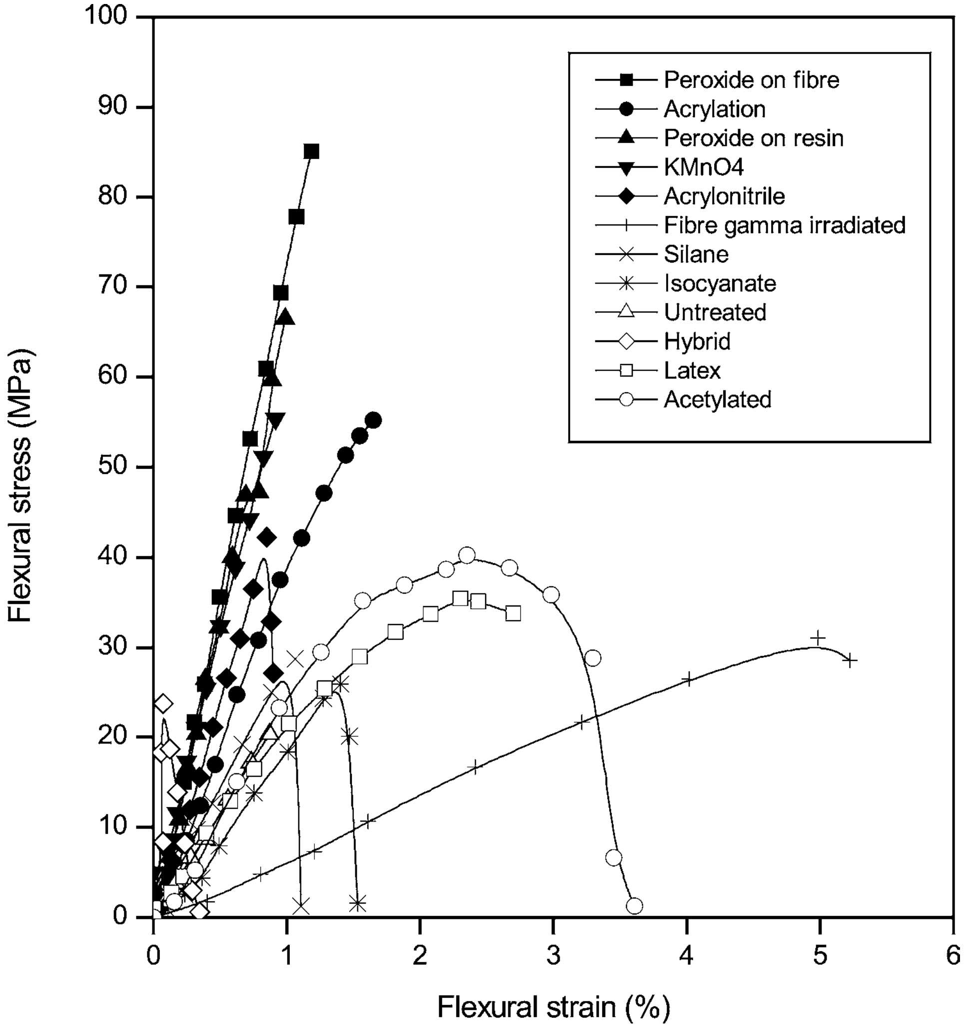

Figure 13. Flexural stress–strain characteristics of thermal aged composites.

3.4. Ageing effects on the impact properties

Izod impact strength values of composites on various ageing are given in Table 7. Impact strength was enhanced upon water ageing in untreated, acrylonitrile, peroxide and isocyanate treated composites. Composite impact strength degraded upon thermal, biodegradation and on gamma irradiation. The decrease is prominent at higher doses of radiation and on thermal ageing. Izod impact strength is a measure of the energy required to initiate and propagate a crack. Phenolics exhibit superior heat resistance compared to polyesters and epoxies. This is directly related to the structure and thermal degradation mechanism of phenolics. Several research studies have been reported in improving the low impact strength of the phenolics [20, 21]. It is found that impact strength is strongly influenced by the fibre–matrix interface and

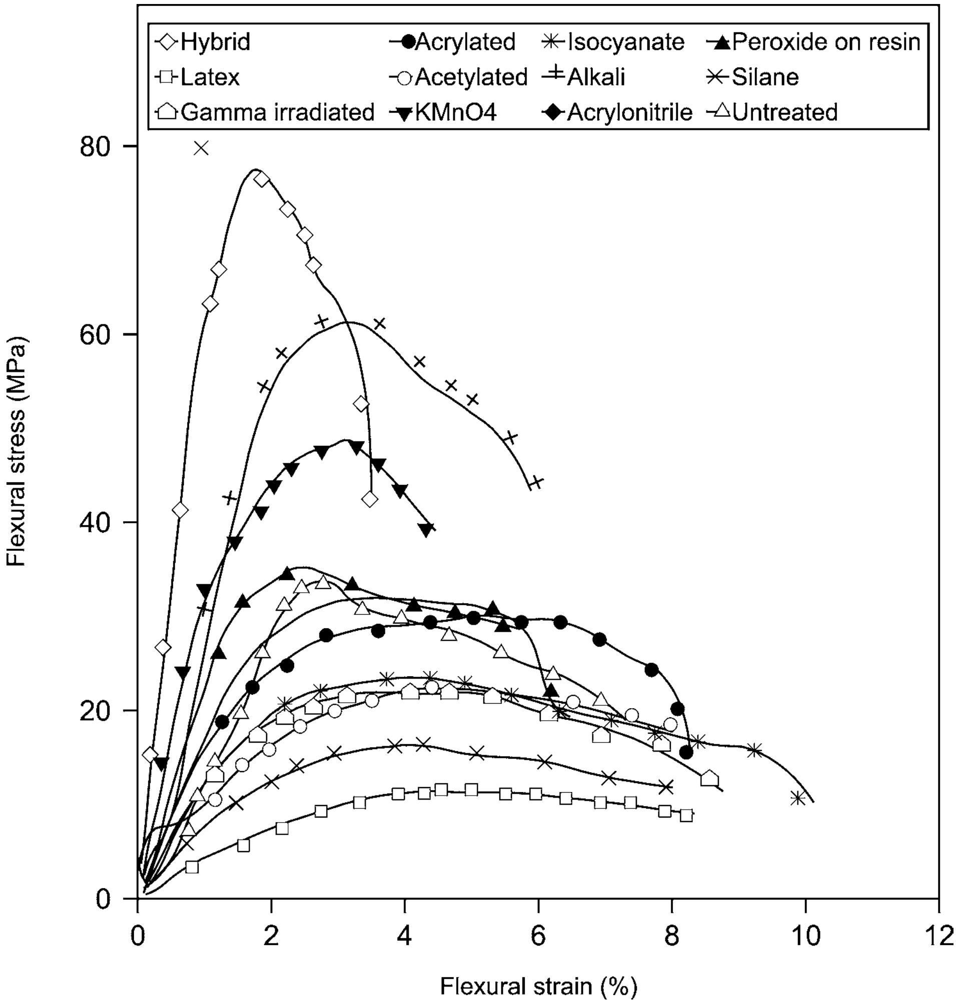

Figure 14. Flexural stress–strain characteristics of cold water aged composites.

fibre length. Maximum energy can be dissipated by mechanical friction during the pull out process and by debonding of the fibres [21]. The decline in the properties of the phenolic composite upon thermal ageing is due to the breakdown of the bonds at the fibre–matrix interface. The unnotched and notched izod impact performance of phenolic composites following thermal exposure was studied in detail by Kuzak *et al.* [22]. They observed that the impact performance of the S-glass (Strength-glass) reinforced phenolic composite declined when exposed to 300°C temperature. They prepared a phenolic blend from Resole and Novolac and found to have best retention in impact properties on thermal exposure. The izod impact strength of all the composites declined to a very low value upon gamma irradiation. This is due to the bond scission and disintegration at the fibre–matrix interface.

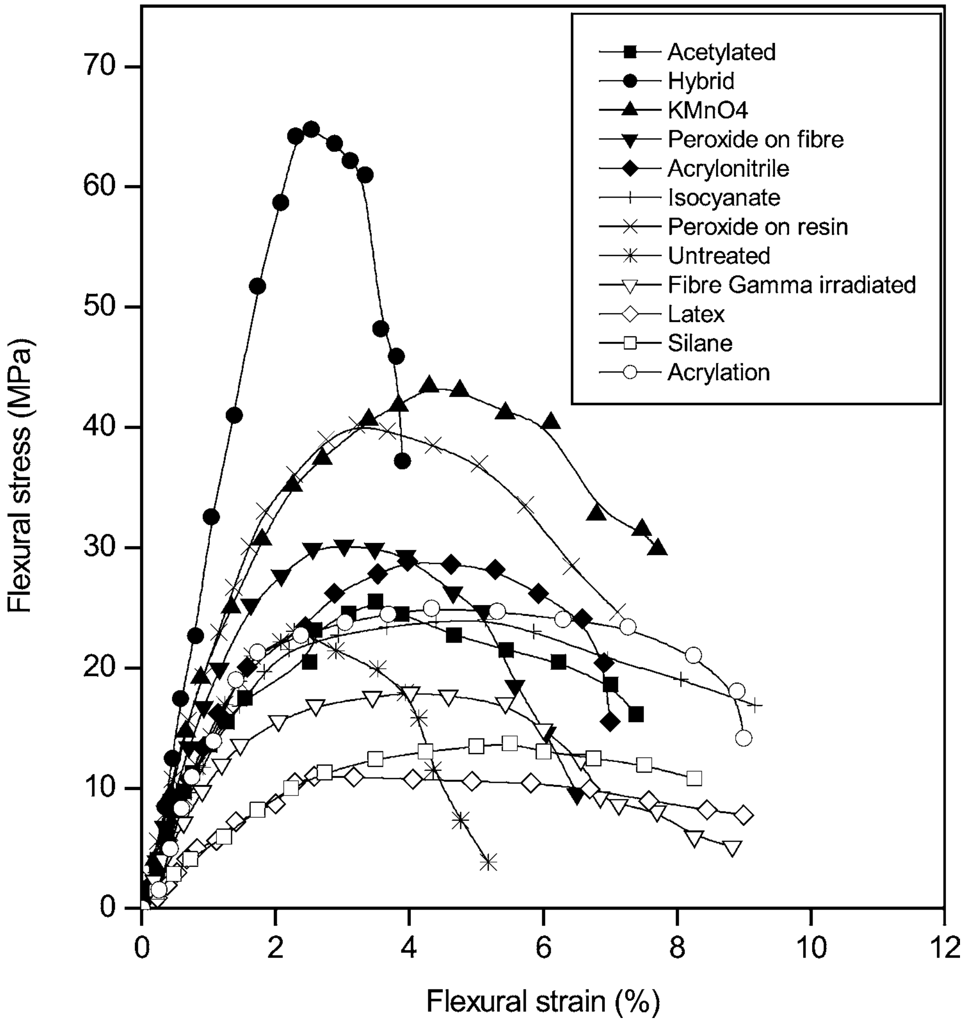

Figure 15. Flexural stress–strain characteristics of boiling water aged composites.

3.5. Impact fracture mechanism

Scanning electron micrographs of the impact fracture surfaces were taken to study the fracture mechanism (Figs 16–21). Pulled out fibres are seen in water aged, acetylated and latex treated composites (Figs 16 and 17). Moderate retention in impact strength in these composites is observed. The exposed fibre indicates weak adhesion at the interface. The impact fracture lines are clearly visible on failure surface of gamma irradiated neat PF sample (Fig. 18). The interfacial disintegration on gamma irradiation is evident from the fractograph of acetylated composite (Fig. 19). Fibrillation is also observed. Fibre failure due to the cracking and splitting of the fibres is visible in the failed surface of the gamma irradiated, latex coated composite (Fig. 20). The fibre strength is decreased considerably in silane treated

Table 5.
Flexural strength values (MPa) of unaged and aged composites

Sample	Unaged	Thermal aged (100 °C; 72 h)	Cold water aged (2 weeks)	Boiling water aged (2 h)
Untreated	49	20	33	23
Hybrid	—	23	78	65
Acetylated	36	40	22	26
Acrylonitrile	52	42	32	29
Peroxide on resin	54	67	35	40
Latex	16	35	11	11
Isocyanate	32	26	24	24
Acrylation	29	55	30	25
Silane	23	29	16	14
$KMnO_4$	55	55	49	43
Mercerisation	75	—	61	—
Fiber γ irradiated	30	31	22	18
Peroxide on fiber	71	85	—	30

Table 6.
Flexural modulus values (MPa) of unaged and aged composites

Sample	Unaged	Thermal aged (100 °C; 72 h)	Cold water aged (2 weeks)	Boiling water aged (2 h)
Untreated	3596	2292	1184	1402
Hybrid	4194	—	7652	2740
Acetylated	2280	2180	1568	1522
Acrylonitrile	2578	—	2032	1562
Peroxide on resin	3656	6596	2648	2218
Latex	782	2292	490	620
Isocyanate	1860	1622	1568	1316
Acrylation	2100	3800	2032	1358
Silane	1260	2906	952	490
$KMnO_4$	4136	6484	4034	2218
Mercerisation	3478	—	2724	—
Fiber γ irradiated	2578	504	1568	1098
Peroxide on fiber	3814	7156	—	1912

composite upon gamma irradiation as is evident from the breaking pattern of the fibre (Fig. 21).

4. CONCLUSIONS

Environmental effects on the mechanical properties of oil palm fibre reinforced PF composites have been investigated. Ageing effects were compared with those of glass/PF and oil palm/glass hybrid PF composites. Various fibre modifications

Table 7.
Impact strength values (kJ/m^2) of unaged and aged composites

Sample	Unaged	Thermal aged (100 °C; 72 h)	Cold water aged (2 weeks)	Boiling water aged (2 h)	Biodegradation (8 months)	Gamma irradiation Dose (Mrad)				
						0.1	1	10	50	100
Untreated	41	16	48	59	17	7	18	6	3	2
Hybrid	280	106	202	115	52	53	47	34	14	25
Acetylated	181	46	119	135	53	46	53	40	7	3
Acrylonitrile	59	12	63	54	11	20	23	15	4	5
Peroxide on resin	56	21	199	152	28	38	27	18	4	2
Latex	190	65	102	105	35	31	47	23	12	2
Isocyanate	155	26	162	149	62	47	39	27	7	4
Acrylation	129	14	119	78	19	28	30	11	3	3
Silane	165	25	160	131	60	99	70	46	4	24
$KMnO_4$	53	18	45	52	27	27	20	28	3	43
Fiber γ irradiated	105	21	69	64	28	—	—	—	—	—
Peroxide on fiber	44	22	—	53	17	17	16	7	4	4

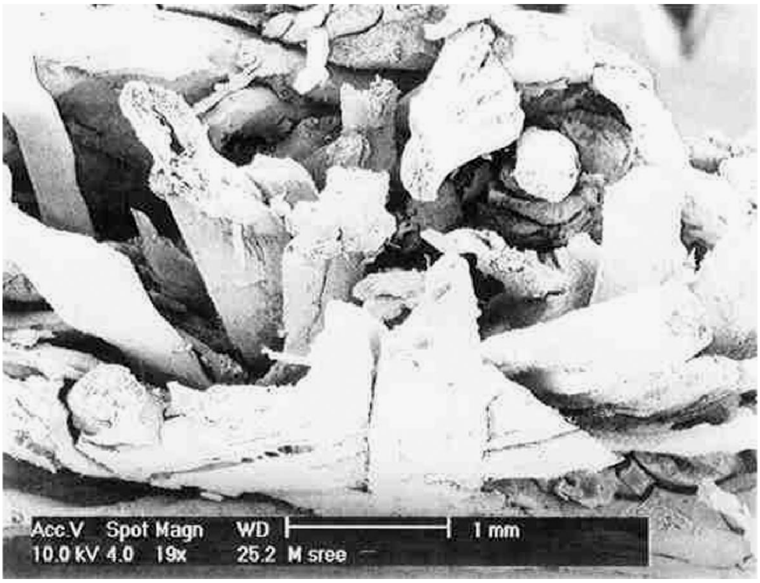

Figure 16. SEM of impact fracture of water aged, acetylated oil palm fiber/PF composite (×19).

were carried out and their effect on the ageing process were studied. Thermal ageing, cold water and boiling water ageing, biodegradation effects and gamma irradiation effects on the composite properties were analysed. Influence of ageing on the tensile, flexural and impact properties of the composites was investigated.

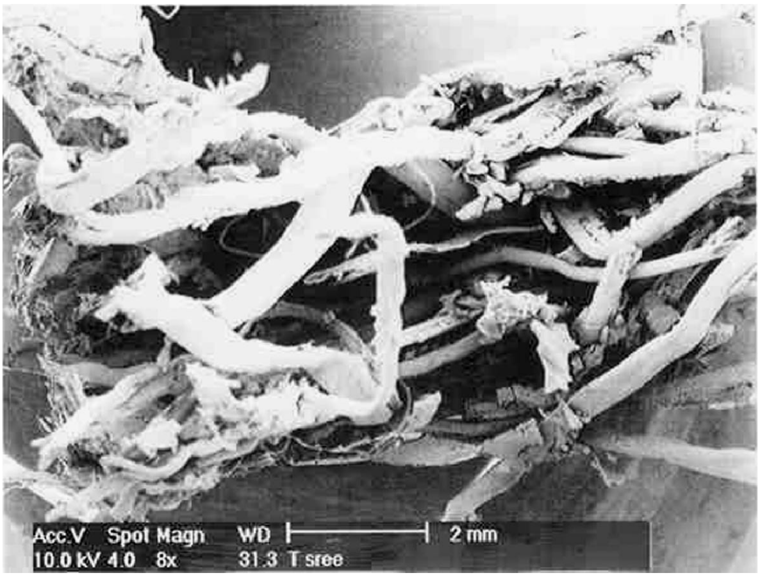

Figure 17. SEM of impact fracture of water aged, latex coated oil palm fiber/PF composite (×8).

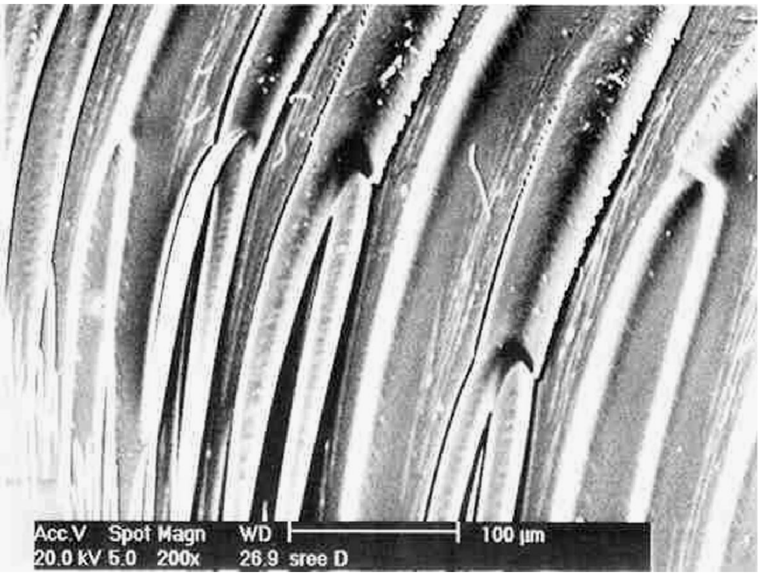

Figure 18. SEM of impact fracture of gamma irradiated, neat PF sample (×200).

The tensile and flexural stress–strain behaviour of the composites was affected by ageing. The tensile strength of the composites decreased upon thermal, biodegradation and gamma irradiation. However, water ageing did not decrease the properties of the composites and in some cases enhancement is observed. The extensibility of the composites decreased upon thermal ageing. However, the value

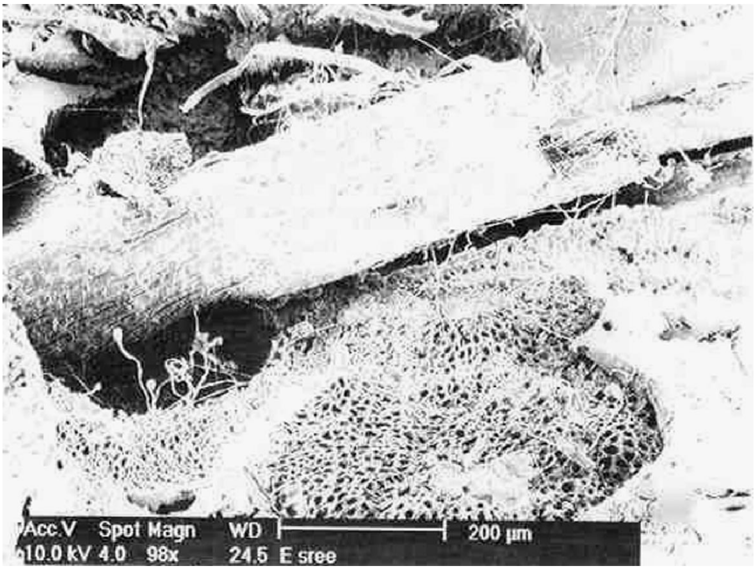

Figure 19. SEM of impact fracture of gamma irradiated, acetylated oil palm fiber/PF composite (×98).

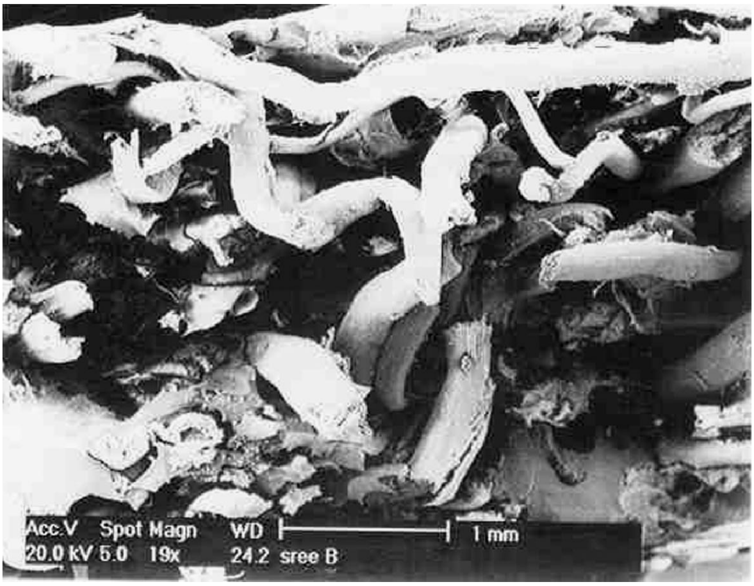

Figure 20. SEM of impact fracture of gamma irradiated, latex coated oil palm fiber/PF composite (×19).

increased upon biodegradation, water ageing and gamma irradiation. Thermal and water ageing increase the tensile modulus of some treated composite such as silane, peroxide, isocyanate and acrylated samples. Biodegradation decreases the modulus

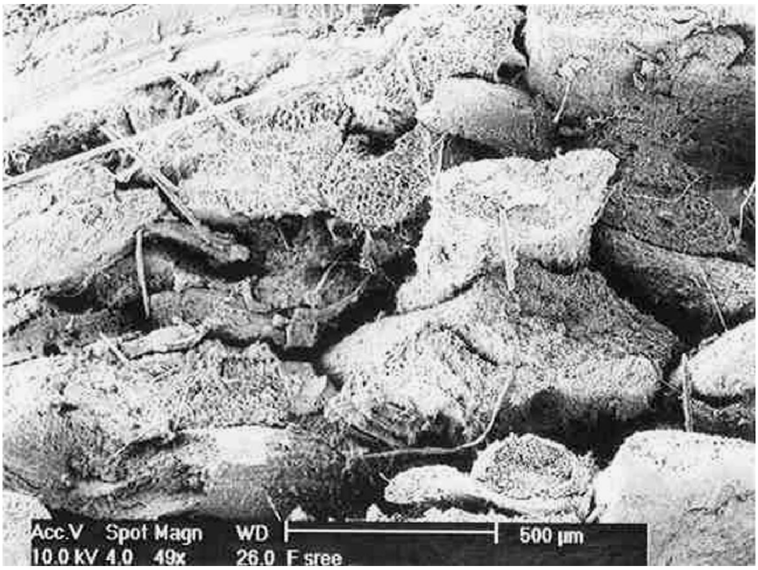

Figure 21. SEM of impact fracture of gamma irradiated, silane treated oil palm fiber/PF composite (×49).

values of all systems. Untreated, alkali and peroxide treated fibre composite show enhancement in tensile modulus values upon gamma irradiation. Various failure processes were studied by scanning electron microscopy. The flexural properties of the composites decrease upon water ageing. Thermal ageing leads to higher flexural properties in peroxide, latex and acrylated composite. Izod impact strength of the untreated, acrylonitrile, peroxide and isocyanate treated composites increased upon water ageing. Biodegradation, thermal ageing and gamma irradiation decreased the impact resistance of the composite. Scanning electron micrograph of the impact fracture surface revealed the failure mechanism. The present study reveals that environmental conditions have a significant effect on the composite properties. Biodegradable studies showed that growth of the fungi on the composites is favoured under moist conditions. By incorporation of oil palm fibre, the composite became more biodegradable, ecofriendly and cost efficient. Hence research based on composites made from natural materials will receive more attention as they are ecofriendly.

REFERENCES

1. S. Hill, *New Scientist* (1 Feb.), 36 (1997).
2. A. N. Netravali and S. Chabba, *Materials Today* (April), 22 (2003).
3. W. Schnabel (Ed.), *Polymer Degradation, Principles and Practical Applications*. Macmillan, New York (1981).
4. L. Barral, J. Cano, J. Lopez, P. Nogueira and C. Ramirez, *J. Appl. Polym. Sci.* **63**, 1841 (1997).
5. T. C. Uzomah and G. C. Unuoha, *J. Appl. Polym. Sci.* **69**, 2533 (1998).

6. H. Lin, D. E. Day and J. O. Stoffer, *Polym. Comp.* **14**, 402 (1993).
7. T. Nakashima and M. Matsuo, *J. Macromol. Sci., Phys.* **B 35**(3&4), 659 (1996).
8. H. Parvatareddy, J. Z. Wang, D. A. Dillard, T. C. Ward and M. E. Rogalski, *Compos. Sci. Technol.* **53**, 399 (1995).
9. N. L. Hancox, *Plast. Rubber Comp. Process. Appln.* **27**, 97 (1998).
10. M. S. Sreekala, M. G. Kumaran and S. Thomas, *Appl. Comp. Mater.* (in press).
11. J. George, S. S. Bhagawan and S. Thomas, *Comp. Sci. Technol.* **58**, 1471 (1998).
12. K. Joseph and S. Thomas, *Compos. Sci. Technol.* **53**, 99 (1995).
13. S. P. Sonawala and R. J. Spontak, *J. Mater. Sci.* **31**, 4745 (1996).
14. S. P. Sonawala and R. J. Spontak, *J. Mater. Sci.* **31**, 4757 (1996).
15. L. Dong, D. J. T. Hill, J. H. O'Donnell, P. J. Pomery and K. Hatada, *J. Appl. Polym. Sci.* **59**, 589 (1996).
16. S. M. Spieberger, K. Humer, E. K. Tschegg, H. W. Weber, K. Noma and Y. Iwasaki, *Cryogenics*, **37**, 135 (1997).
17. A. N. Netravali and A. Manji, *Polym. Compos.* **12**, 153 (1991).
18. M. S. Sreekala, M. G. Kumaran and S. Thomas, Water sorption kinetics in oil palm fibers, *Eur. Polym. J.* (submitted).
19. H. Kudoh, T. Sasuga, T. Seguchi and Y. Katsumura, *Polymer* **37**, 3737 (1996).
20. J. T. Carter, *Plast. Rubber Comp. Proc. Appln.* **16**, 157 (1991).
21. A. Matsumoto, K. Hasegawa, A. Fukuda and K. Otsuki, *J. Appl. Polym. Sci.* **44**, 1547 (1992).
22. S. G. Kuzak, J. A. Hiltz and P. A. Waitkus, *J. Appl. Polym. Sci.* **67**, 349 (1998).