

STUDIES ON NATURAL WEATHERING OF RATTAN POWDER-FILLED NATURAL RUBBER COMPOSITES

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This article investigates the effect of natural weathering on mechanical and morphological properties of rattan powder-filled natural rubber (NR) composites as a function of filler loading and silane coupling agent. The rattan powder samples in the range of 0 to 30 phr were compounded with NR using a laboratory size two-roll mill. The natural weathering test was carried out for six months. The degradation of the samples was evaluated by performing a tensile test, a Fourier transform infrared spectroscopy (FTIR), and a scanning electron microscopy (SEM) test. The results indicated that after natural weathering, an increase in stress at 100% elongation (M100) can be seen for samples without the silane coupling agent, whilst M100 was reduced for samples with silane coupling agent. A drastic reduction in tensile strength and elongation at break were observed for all samples due to the photo-oxidation process that occurred during the degradation of the samples. The extent of degradation on the samples' surfaces and the presence of oxygenated products were confirmed by SEM and FTIR studies, respectively.

Keywords: Natural weathering; Natural rubber; Rattan filler; Silane coupling agent; FTIR; SEM

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INTRODUCTION

Natural weathering, also known as environmental degradation, is considered a practical way to obtain information regarding material performances resulting from exposure to natural outdoor conditions. Throughout the service of products, polymeric materials are directly or indirectly subjected to a variety of environmental conditions such as sunlight, heat, oxygen, moisture, and ozone. These factors, along with microorganisms and airborne chemical pollutants such as sulphur oxides and nitrogen oxides, contribute to the degradation of polymeric materials. Generally, the degradation of polymeric materials involves changes in mechanical properties such as strength, stiffness, and elasticity, as well as optical characteristics, which affect the suitability of the materials for specific applications (Awang and Ismail 2009; Noriman *et al.* 2010; Sahin *et al.* 2007).

In the case of natural rubber products, environmental degradation is mainly caused by oxygen and ozone molecules. The degradation proceeds via different chemical mechanisms, resulting in different effects on the final properties of rubber. This is because natural rubber contains an unsaturated backbone that can be easily attacked by ozone and oxygen molecules. Ozone degradation, which is primarily a surface phenomenon, results in discoloration and cracking of rubber. Conversely, oxygen degradation results in hardening or softening of the rubber. For instance, during the

oxidation process, natural rubber (NR) vulcanizates are subjected to cleavage reactions and usually turn softer. Whilst, NR becomes hard and brittle when it is completely oxidized upon progressive aging and the cross-linking mechanism starts to dominate again (Datta and Huntink 2008).

For many applications, filler additions into rubber compounds are essential for various purposes, of which the most important are for reinforcement, reduction in material costs, and improvements in processing (Rattanasom *et al.* 2006). A wide variety of fillers such as carbon black, silica, and calcium carbonate are often used in the rubber industry. Recently, the use of natural fibres as an alternative source of commercial fillers in polymer composites has spurred the interests of many researchers. Extensive work has been carried out to explore the possibilities of natural fibres as filler in polymer composites, as they provide advantages over commercial fillers such as being environmentally friendly, biodegradable, readily and abundantly available, cheap, and having low density (Satyanarayana *et al.* 2009; John *et al.* 2008).

Accordingly, for the purpose of this research, waste rattans were used as filler in preparing natural rubber composites. Rattan canes are extensively used in the furniture industry and account for a significant proportion of the export earnings for Malaysia. Rattan waste from these furniture industries becomes a burden to the environment through open burning and illegal dumping. The wastage rate could be as high as 10%, and financially as much as USD 0.20 per cane is wasted (Ariffin *et al.* 2001). In addition, inherent properties of rattan cane such as its being lightweight, strong, and durable have increased interest for its use as a filler. Successful use of waste rattan as filler will help to create a more environmentally friendly product.

However, a notable drawback of using natural fibre in polymer composites is the polarity of the fibres, which results in poor compatibility with a hydrophobic matrix. This naturally causes a reduction in the mechanical properties of the composites, and the effect is multiplied at higher filler loading. Coupling agents are used as a remedy to overcome this drawback in order to improve the wettability of natural fibre by polymers and promote interfacial adhesion. Coupling agents contain bi-functional groups which can react with both natural fibre and polymer matrix by forming a linkage between them. This brings about an enhancement in adhesion of natural fibre to polymer and forms a uniform composite structure. Silane coupling agent is recognized as an efficient coupling agent used extensively in the polymer industry (Taj *et al.* 2007; Yan *et al.* 2005).

As an extension from a previous paper (Ismail *et al.* 2011), this paper focuses on natural weathering of rattan powder-filled NR composites. The degradation of samples is evaluated by performing tensile test, Fourier transform infrared spectroscopy (FTIR), and Scanning electron microscopy (SEM).

EXPERIMENTAL

Materials and Formulation

Table 1 shows the formulation, materials, and their respective suppliers used in this research study. Rattan wastes were collected, cleaned, ground, and sieved into a

particle size less than 180 μm . The particle size distribution of rattan powder fell in the range of 0.2 μm to 180 μm , as displayed in Fig.1.

Table 1. Formulation of Rattan Powder Filled NR Composites (Ismail *et al.* 2012)

Ingredients	Formulation [phr]		Supplier
	Series 1	Series 2	
Natural Rubber	100	100	Rubber Research Institute Malaysia
Rattan	0, 5, 10, 15, 30	0, 5, 10, 15, 30	Seng Huat Sdn Bhd
Zinc Oxide	1.5	1.5	Bayer (M) Ltd
Stearic Acid	1.5	1.5	Bayer (M) Ltd
CBS ^a	1.9	1.9	Bayer (M) Ltd
BKF ^b	2.0	2.0	Bayer (M) Ltd
Sulphur	1.6	1.6	Bayer (M) Ltd
Silane coupling agent (AMEO) ^c	-	1.0	Bayer (M) Ltd

^aCBS - N-cyclohexyl-2-benzothiazole sulfenamide

^bBKF - 2,2 methylene-bis-(4-methyl-6-tert-butylphenol)

^cAMEO – aminopropyltrimethoxysilane

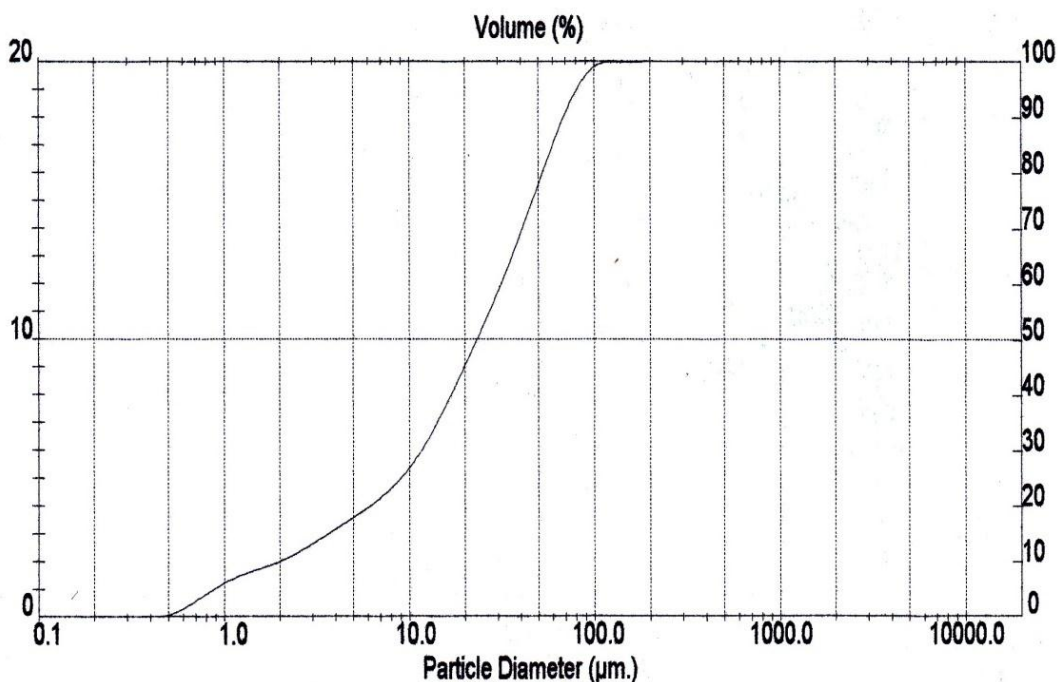


Fig. 1. Graph of particle size distribution of rattan powder

Sample Preparation

Mixing procedures for two series of natural rubber composites were carried out in a laboratory two-roll mill, model XK-160, in accordance to the method described by American Society for Testing and Materials (ASTM), designated D 3184-80. The first series was without the addition of silane coupling agent, while the second series involved the addition of silane coupling agent. The total mixing time was kept to a minimum of about 25 minutes to avoid damage of the rubber compound in the early stage. The water pipe channel was opened to remove excess heat produced during compounding in order to maintain the mill rolls temperature at 70 ± 5 °C and avoid premature vulcanization during mixing. After the compounding process was done, the rubber compound was kept in a freezer for 24 hours before curing assessment and vulcanization. The cure characteristics of rubber compounds were studied using a Monsanto Rheometer, model MDR 2000 at 150°C. Cure time (t_{90}), scorch time (t_{s2}), and maximum torque (MH) were obtained from the rheographs. The rubber compounds were moulded into sheets with respective cure time, t_{90} at 150°C using a hot press machine. The cure characterization results were reported in a previous paper (Ismail *et al.* 2011).

Natural Weathering

The natural weathering test was performed at School of Materials and Mineral Resources Engineering, Universiti Sains Malaysia, Penang, Malaysia (latitude 5°28'N, longitude 100°23'N) for a period of 6 months from December 2010 to May 2011. The natural weathering test was carried out according to ISO 877.2. Dumbbell-shaped samples were cut from vulcanized rubber sheets and arranged on the exposed rack at an inclined angle of 45°C. The rack was located at an open area that was free from being overshadowed by other objects. Thus, the samples were exposed to all environmental effects such as rain, sunlight, wind, *etc.* After six months, the samples were withdrawn, washed with distilled water, and dried at room temperature. The degradation was assessed and evaluated by measuring tensile properties before and after the exposure to environmental conditions. Tensile test were carried out according to ISO 37 at a constant rate of 500 mm/min with an Instron Universal testing machine, model 3366. Five samples were tested, and the average of three results was reported.

Fourier Transform Infrared (FTIR)

The chemical changes in un-weathered and weathered samples were detected using Fourier-transform infrared spectroscopy (FTIR; Perkin Elmer System 2000). A sample with dimension of 5mm × 5mm × 2mm (width × length × thickness) was used for the FTIR analysis. The FTIR spectra were recorded in the range of 550 to 4000 cm^{-1} wave numbers with an average of 24 scans.

Scanning Electron Microscopy (SEM)

Examination of the weathered sample surfaces was carried out using a scanning electron microscopy (SEM), Zeiss Supra 35vp. The surfaces were sputter-coated with gold to avoid electrostatic charging and poor image resolution. The degradation of samples was evaluated from the micrographs.

RESULTS AND DISCUSSION

Tensile Properties

The results for the tensile properties of unfilled natural rubber (NR) compound and rattan powder filled NR composites with and without silane coupling agent, before and after 6 months of natural weathering, are summarized in Figs. 1 to 3. Before natural weathering, the samples without the silane coupling agent showed a decreasing trend for tensile strength and elongation at break, whilst, an increasing trend for M100 was noted as the filler loading was increased. In a previous paper (Ismail *et al.* 2011), the authors had reported the effect of filler loading and silane coupling agent on the mechanical properties of rattan powder filled natural rubber composites.

With reference to Fig. 1, the reduction in tensile strength was attributed to the incompatibility between the rattan filler and the rubber matrix, which resulted in a weaker rubber-filler interaction. As a consequence, the filler was unable to support the uniform transmission of stresses from the rubber matrix, thus lowering the tensile strength. On the other hand, as rattan was added into NR composites, higher restriction to movement was experienced by the rubber chains. Hence, the elasticity of composites was reduced, resulting in more stiff and rigid composites. This led to lower resistance to breakage. Thus, the elongation at break decreased and the modulus increased, which is displayed accordingly in Figs. 2 and 3. However, the addition of 3-aminopropyltrimethoxysilane as the coupling agent promoted a better interaction between the rattan filler and rubber matrix, enabling good stress transfer from the matrix to the rattan filler. This was proven by the improvement in tensile strength as shown in Fig. 1. The better rubber-rattan interaction also increased the stiffness and rigidity of the composites, resulting in further increase in M100 and reduction in the elongation at break, which is exhibited in Figs. 2 and 3, respectively (Ismail *et al.* 2011).

After subjecting the samples to weathering conditions for 6 months, deterioration in tensile properties was observed for both samples with and without the silane coupling agent in comparison with the un-weathered samples, as shown in Figs. 1 to 3. A drastic drop was seen for tensile strength, with an average of 90% for all samples in comparison with un-weathered samples, reduction of about 80% for elongation at break, an average of 22% for M100 of samples with silane coupling agent, and an increment of 4% for M100 of samples without silane coupling agent. This is due to the degradation of natural rubber and the rattan filler upon exposure to outdoor conditions.

During weathering, polymer matrix degradation would be initiated by the breakdown of polymer chains via the photo-oxidation process with the help of ultraviolet radiation (Abu Bakar *et al.* 2005; Beg and Pickering 2008). The photo-oxidation process can easily degrade NR, as it contains an unsaturated backbone that is prone to attack by the oxygen and ozone molecules previously mentioned in the introduction section. Thus, the reduction in tensile strength of all samples was mainly attributable to the combined effects of oxidative and ozone degradation. These degradations resulted in scission of the rubber chains and intermolecular crosslinking under UV exposure. Scission of the larger molecular chains increases the number of shorter chains, which leads to fewer entanglements, while intermolecular cross-linking causes material embrittlement (Abu Bakar *et al.* 2005; Ismail *et al.* 2008; Noriman *et al.* 2010). As a consequence, natural rubber loses

its elastomeric properties and its ability to act as an effective matrix material to transmit stress. Hence, the tensile strength was reduced after weathering for 6 months, as illustrated in Fig.1.

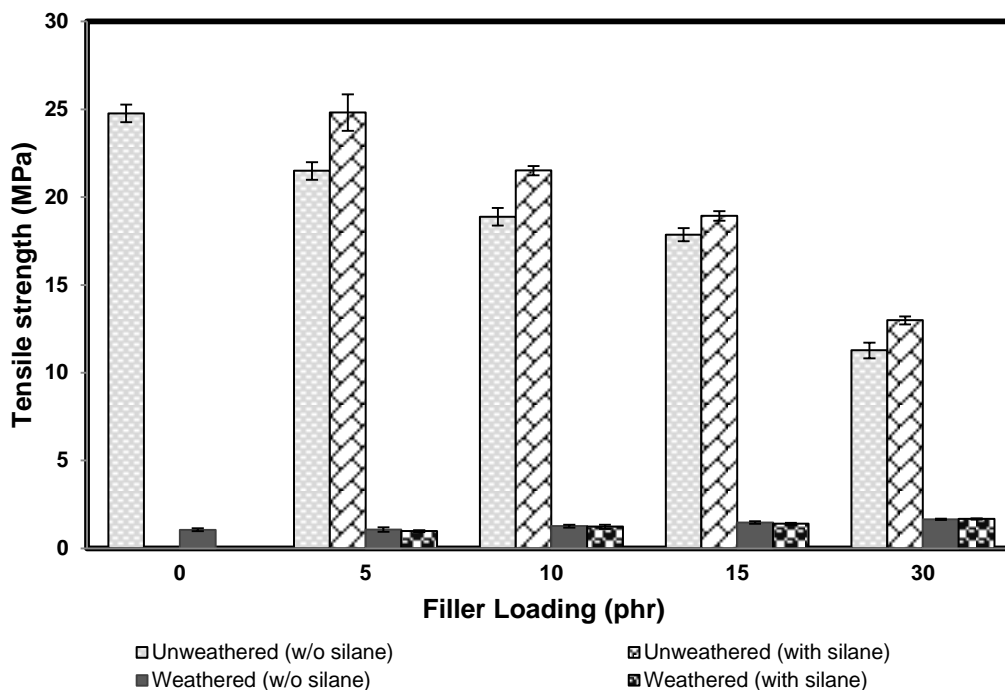


Fig. 1. The effect of filler loading and silane coupling agent on tensile strength of rattan powder-filled natural rubber composites before and after natural weathering

As mentioned for tensile strength, the breakdown of rubber chains and embrittlement reduces the slipping nature and stretch ability of the rubber chains. Accordingly, the elasticity of the samples was reduced and the sample's resistance to break is lowered, as shown in Fig. 2. The result for the M100 of samples was also controlled by the chain cleavage and the crosslinking reactions. The M100 of samples without silane coupling agent were seen to increase after weathering. This increment might be attributed to some crosslinking reactions due to radical recombination becoming dominant during exposure. The M100 of samples with silane coupling agent was noted to decrease after weathering which might be due to the influence of cleavage reactions becoming dominant along with the destruction of interaction provided by silane coupling agent (which will be discussed in FTIR section), making the rubber soft.

Upon longer exposure, the degradation of samples, which started at the outer surface, penetrated gradually into the bulk rubber matrix (Ismail *et al.* 2008). This led to the development of more surface cracks, causing the rattan fillers to be exposed to the outdoor conditions. The exposed rattan fillers were also expected to degrade by natural weathering. One of the components of rattan, lignin was removed by leaching during the weathering period. Leaching of lignin was apparent due to the color fading of the samples.

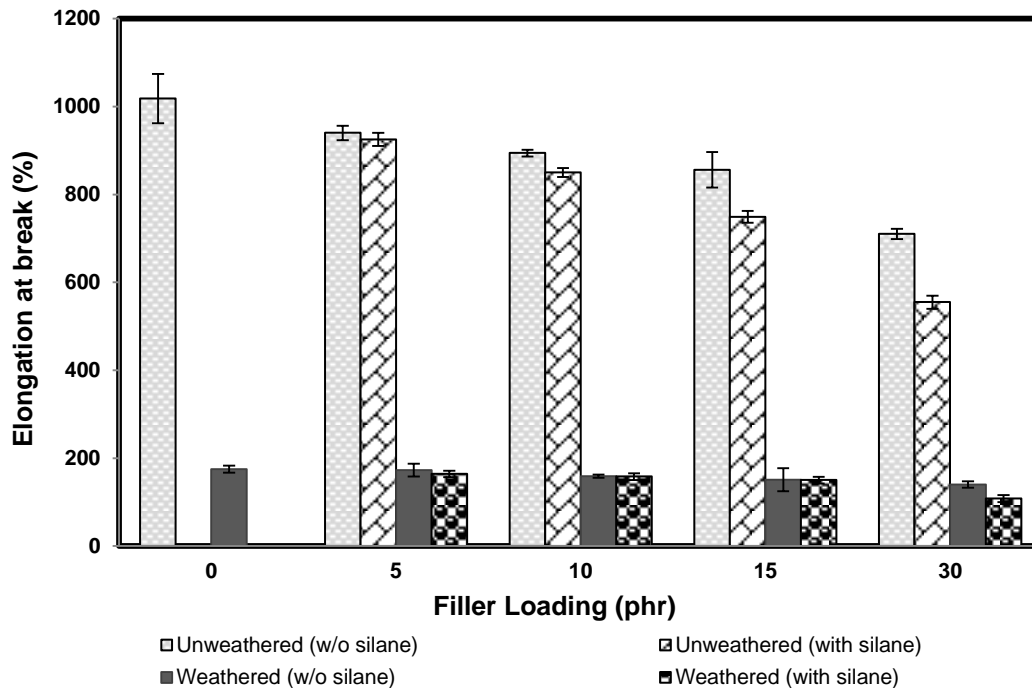


Fig. 2. The effect of filler loading and silane coupling agent on elongation at break of rattan powder filled natural rubber composites before and after natural weathering

Pattamasattayasonthi *et al.* (2011) observed and reported that the photo-degradation of the lignin in wood components was the main reason for the color fading. However, the major contribution of weathering of the rattan fillers toward the properties of composites are the aesthetic effects such as surface checking and growth of sap-staining fungi, which can be seen in SEM micrograph (Fig. 5) and also the change in color. For instance, the results of tensile strength obtained for all samples were in a similar range, proving that degradation of rattan filler did not contribute much to the deterioration of tensile properties. Instead, the major contributor towards the deterioration in tensile properties was the degradation of the rubber matrix.

When considering the effect of filler loading toward weathering, regardless of whether it is with or without silane coupling agent, it can be concluded that the trend of results obtained with increasing filler loading was similar to that of results obtained before weathering, except for the tensile strength. A very slight increase in tensile strength was observed after natural weathering with increasing filler loading. Referring to Crabtree and Kemp (1946), filler has the tendency to lower the rate of oxygen absorption of NR with the exposure to UV light. This is attributed to the ability of filler to make the compound opaque, thus limiting the penetration of UV light into the NR matrix. So, we can conclude that the addition of rattan into rubber matrix diminishes the inner damage of matrix, enabling additional strength to samples as compared to that of without or with less rattan filler.

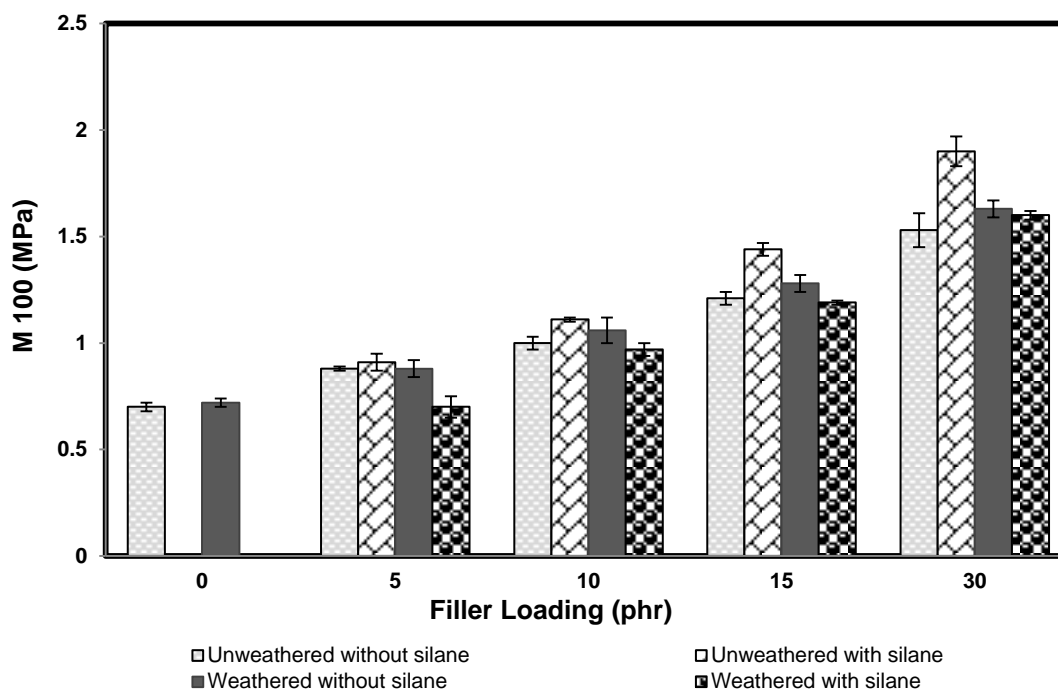


Fig. 3. The effect of filler loading and silane coupling agent on stress at 100% elongation (M100) of rattan powder filled natural rubber composites before and after natural weathering

Referring to Figs. 1 to 3, it is noticeable that addition of silane coupling agent did not improve the tensile properties of the composites after weathering when compared to that of before weathering. This suggests that during weathering, the adhesion formed between the rattan filler and rubber matrix dissolved in rainwater; as a result, silane loses its power as adhesion promoter (George *et al.* 1998). This further reduced the fiber matrix adhesion and resulted in dropping of tensile strength, elongation at break, and M100. The destruction in the rubber-rattan interaction had been demonstrated through FTIR study and will be discussed later in the FTIR section

Fourier Transform Infrared (FTIR)

The results for the FTIR spectrum of rattan powder-filled natural rubber (NR) composites with and without silane coupling agent, before and after exposure to natural weathering are shown in Fig. 4. Before natural weathering, the FTIR spectra of the samples with silane coupling agent (Fig. 4b-i) show the appearance of two additional peaks as compared to composites without the silane coupling agent (Fig. 4a-i). These peaks are found in the region of 1172 cm^{-1} and 1584 cm^{-1} , which is respectively attributed to Si-O-C bonds and deformation modes of NH_2 groups, which bonded to either rattan filler or rubber matrix (Abdelmouleh *et al.* 2004). These bonds prove that the silane coupling agent formed linkages between rattan filler and rubber matrix, thus improving the interfacial adhesion.

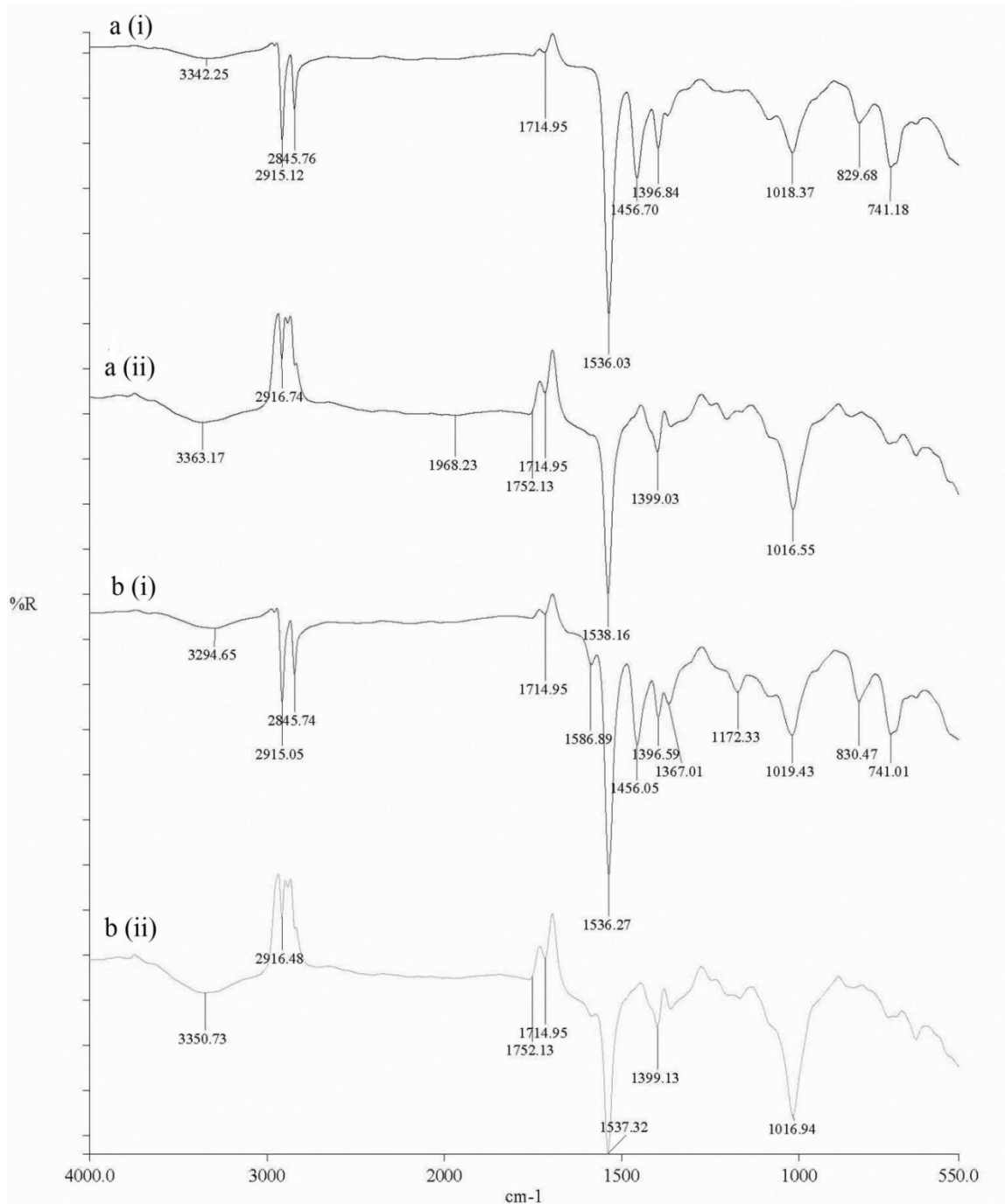


Fig. 4. FTIR spectra of 30 phr rattan powder filled natural rubber composite (a) without and (b) with silane coupling agent (i) before and (ii) after natural weathering

Upon exposure to natural weathering, various chemical reactions took place in the composites, which resulted in the change of FTIR spectrum as compared to un-weathered composites. It can be observed that after weathering there was formation and disappearance of some peaks in the FTIR spectrum. Hydroxyl (OH), carbonyl (C=O), and ether (C-O-C) groups are noticed in the region of 3200-3400 cm^{-1} , 1717 cm^{-1} , and 1020-1050

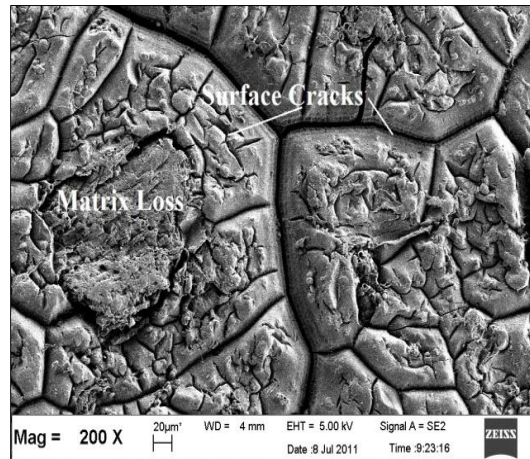
cm^{-1} , respectively, before and after weathering. Referring to Fig. 4a-i and Fig. 4a-ii or Fig. 4b-i and Fig. 4b-ii, after exposure to weathering, a broader peak with a stronger shoulder is observed in the hydroxyl region, whereas the intensity of carbonyl and ether bonds is increased. A new peak at 1762 cm^{-1} is spotted after weathering, which corresponds to the carbonyl (C=O) group from ester bonds. Ozone attack forms unstable ozonides, which undergo cleavage to a variety of oxygenated products, such as acids, esters, ketones, and aldehydes, and also exposes new rubber chains to the effects of ozone (Datta and Huntink 2008). The intensity of peaks at 2845 cm^{-1} and 1456 cm^{-1} , which are attributed to CH_2 and CH_3 groups, disappeared, whilst the intensity of 2916 cm^{-1} , which is assigned to C- CH_3 , is attenuated markedly with exposure, as displayed in Figs. 4a-ii and 4b-ii. These changes indicate that substantial degradation had taken place in the sample. On the other hand, for composites with silane coupling agent, the peaks at 1172 cm^{-1} and 1584 cm^{-1} (Fig. 4a-i and Fig. 4b-i) have disappeared after weathering (Fig. 4a-ii and Fig. 4b-ii). This proves that the linkages formed between rattan filler and rubber matrix were destroyed during weathering.

Surface Morphological Study

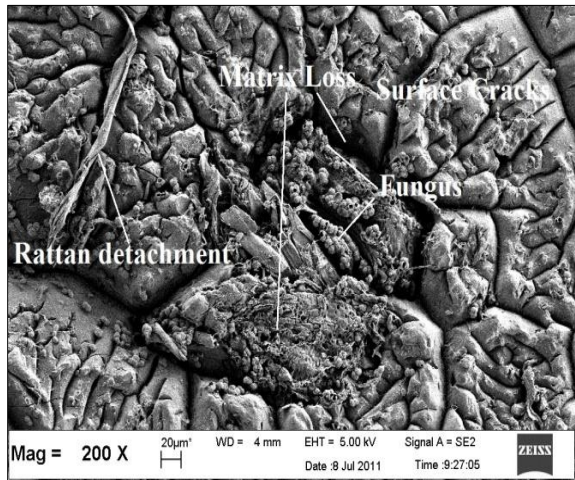
The surface morphology of unfilled natural rubber (NR) compound- and rattan powder-filled NR composites with and without silane coupling agent after 6 months of natural weathering are illustrated through SEM micrographs in Fig. 5.

From these micrographs, it can be observed that all the weathered samples were highly degraded. The surfaces of all samples were rough with formation of extensive surface cracks, which have an appearance similar to a mosaic pattern. As the filler loading was increased, the roughness of surface sample also increased with formation of more micro cracks. The surface roughness was evident due to the increase of M100, and reduction of elongation at break as the filler loading was increased. Apart from oxidative degradation and ozone degradation, dust also was identified to be responsible for the formation of cracks. Dust, once settled on the rubber, can be activated by sunlight to give off oxidizing moieties, which are capable of cracking the rubber (Datta 2008). Furthermore, thermal stress during outdoor exposures due to the day and night variations in temperature can also result in the formation of cracks (Noriman and Ismail 2011).

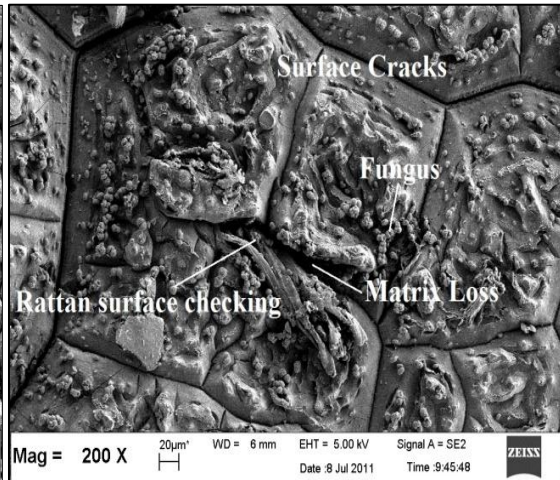
In addition, matrix losses due to degradation were seen in the SEM micrographs. Thus, rattan filler was easily seen, and it became degraded upon weathering. Detachment of rattan filler from the rubber matrix, surface checking, and growth of sap-staining fungi proved that the rattan filler had undergone degradation. The formation of holes on rattan surface and splitting of rattan filler are known as the surface checking of rattan. This will lead to reduced filler-matrix interaction and consequently deteriorate the mechanical properties. More matrix losses and rattan filler degradation were observed at a higher filler loading, 30 phr. From Figs. 5 b-i, 5 b-ii, 5 c-i, and 5 c-ii, it is noticeable that the extent of degradation of samples with the silane coupling agent is similar to that of samples without the silane coupling agent. This indicates that the adhesion provided by silane coupling agent was destroyed by weathering.



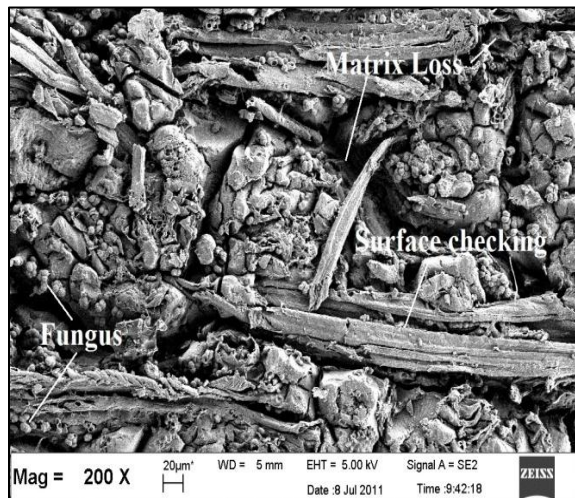
(a)



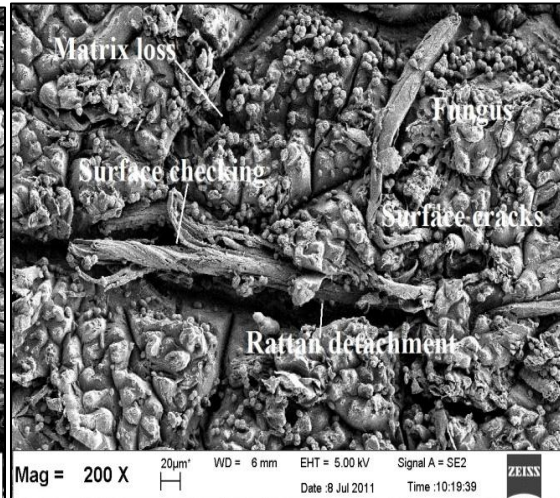
(b-i)



(b-ii)



(c-i)



(c-ii)

Fig. 5. Surface morphology of (a) unfilled natural rubber vulcanizate (b) 5phr; i) with silane coupling agent ii) without silane coupling agent (c) 30phr; i) with silane coupling agent ii) without silane coupling agent of rattan powder filled natural rubber composites after natural weathering at magnification of 200x

CONCLUSIONS

1. Reduction in tensile strength and elongation at break and an increase in M100 were noted as the filler loading was increased for both NR composites with and without silane coupling agent before exposure to natural weathering. The presence of silane coupling agent improved the tensile properties of composites owing to the better wetting and dispersion imparted by silane.
2. After 6 months of natural weathering, an increase in M100 was seen for NR composites without silane coupling agent, whilst a reduction in M100 was seen for NR composites with silane coupling agent as compared to that before weathering. A drastic reduction in tensile strength and elongation at break was observed for all NR composites due to the degradation of the rubber matrix and rattan filler by photo-oxidation. The FTIR results show evidence of degradation.
3. Formation of surface cracks, matrix losses, detachment of rattan filler from rubber matrix, surface checking, and growth of sap staining fungi were observed in the SEM study, which provided supportive evidence to prove the degradation process in the NR composites.

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