

## FATIGUE LIFE, MORPHOLOGICAL STUDIES, AND THERMAL AGING OF RATTAN POWDER-FILLED NATURAL RUBBER COMPOSITES AS A FUNCTION OF FILLER LOADING AND A SILANE COUPLING AGENT

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Fatigue life, morphological studies, and thermal aging properties of rattan powder-filled natural rubber (NR) composites were investigated as a function of filler loading and a silane coupling agent. NR composites were prepared by the incorporation of rattan powder in the range of 0 to 30 phr into a NR matrix with a laboratory size two roll mill. Thermal aging was carried out for 7 and 14 days at a temperature of 70 °C, and tensile testing was performed in order to determine the aging properties. The results indicated that the fatigue life of rattan powder-filled NR composites decreased with increasing rattan powder loading. Tensile strength and elongation at break decreased whilst tensile modulus, stress at 100% elongation (M100), and stress at 300% elongation (M300) increased after aging. Nevertheless, the addition of the silane coupling agent improved both fatigue life and the aging properties of NR composites due to better adhesion between the rubber matrix and the rattan filler which was confirmed by FTIR studies of composites and SEM studies of fatigue fractured surfaces.

*Keywords: Rattan filler; Natural rubber; Silane coupling agent; Fatigue life; Thermal aging; FTIR; SEM*

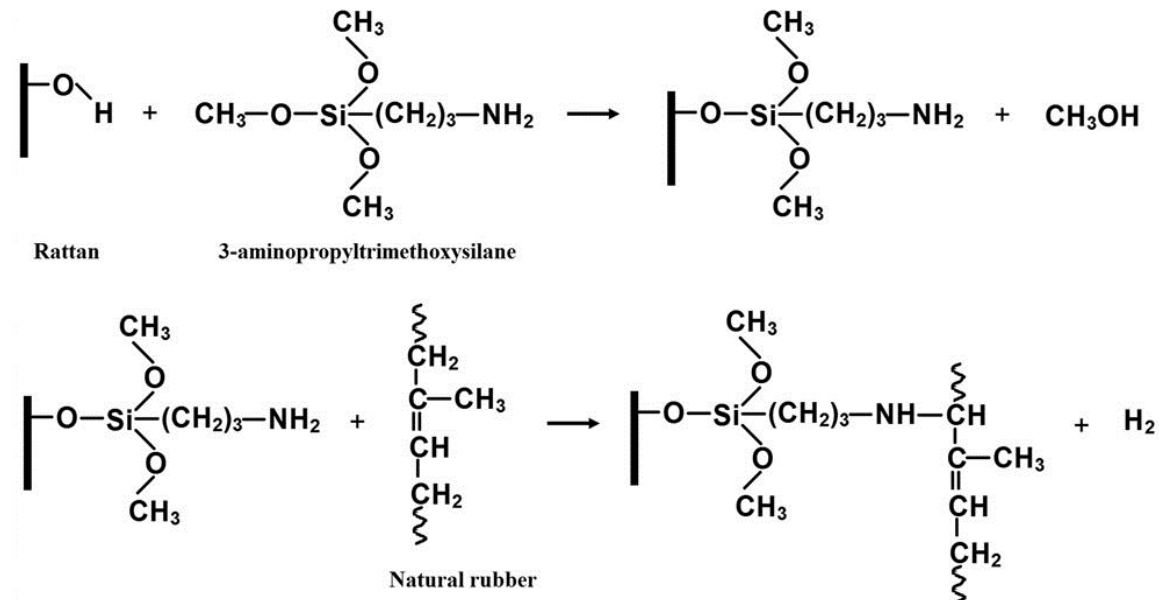
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### INTRODUCTION

Natural fibers are naturally occurring composites consisting of cellulose fibrils embedded in a lignin matrix. Extensive studies have been carried out to investigate the use of natural fibers as filler in polymer composites, as they provide advantages over commercial fillers. Natural fibers are environmentally friendly, biodegradable, readily and abundantly available, cheap, and have lower density than traditional fillers. Despite the advantages, a notable drawback of using natural fibers in composites is the polarity of the fibers, which results in poor compatibility with the hydrophobic matrix (John and Anandjiwala 2008; Li et al. 2007; Saheb and Jog 1999). This naturally causes a drop in the mechanical strength of a composite, and the effect is magnified at a higher filler loading (Da Costa et al. 2001; Ismail et al. 2003). This drawback can be remedied by the use of coupling agents in order to improve the wettability of natural fiber by polymers and promote interfacial adhesion (John and Anandjiwala 2008; Xie et al. 2010). Coupling agents contain bi-functional groups, which can react with both natural fiber and polymer matrices by forming a linkage between them, as illustrated in Fig. 1. This brings an enhancement in adhesion of the natural fiber to the polymer, forming a uniform

composite structure. Silane coupling agent is recognized as an efficient coupling agent used extensively in polymer composites (Barry 1977; Xie et al. 2010).

Interestingly, natural fibers could possibly allow countries to use their own natural resources in their composite processing industries. In this case, for example, rattans are used in Malaysia. Rattans are spiny climbing plants belonging to the palm family, which are abundantly available and considered as the most important non-wood forest product (Ali and Khoo 1995). Rattan canes are extensively used in the furniture industry and account for a significant proportion of the export earnings for Malaysia. The rattan industry, however, produces large amounts of rattan waste materials, especially rattan poles, which often become a burden to the environment through open burning and illegal dumping. Rattan poles even produce waste before the furniture manufacturing process. In some cases, 6 inches of each end are cut off and thrown away. At this stage, the wastage rate could be as high as 10 percent. Financially, 0.20 USD per pole is lost as waste (Ariffin et al. 2001). Thus, the waste rattan cane, being lightweight, strong, and durable, has spurred an interest for use as filler in natural rubber composites.



**Fig.1.** Possible interaction or adhesion of silane coupling agent (3-aminopropyltrimethoxysilane) with rattan filler and natural rubber

Natural rubber maintains its leading position as the most suitable rubber among the general-purpose rubbers available from today's technology. The very uniform, high-cis-polyisoprene molecules, which readily crystallize upon stretching beyond a certain point, provide the rubber with unique and important characteristics. Resistance to fatigue by crack growth and degradation by thermal aging are two important properties that are valued for natural rubber. NR's resistance to fatigue by crack growth mechanism is an important property in dynamic applications of rubber. A major culprit of failure in rubber products under dynamic service conditions is the development of cracks either within the rubber or at its surface. Upon repeated deformation, these cracks grow and lead to complete failure. For many applications rubber must perform consistently over a long period, and this may pose concern (Elliot 1979). Unfortunately, natural rubber contains

double bonds, which render it sensitive to oxidation in the presence of molecular oxygen (Ngolemasango et al. 2006). This oxidative degradation of rubber leads to deterioration in the physical and mechanical properties of vulcanized rubbers.

Nevertheless, a wide variety of fillers such as carbon black, silica, and calcium carbonate are often used in the rubber industry for various purposes, of which the most important are for reinforcement, reduction in material costs, and improvements in processing (Rattanasoma et al. 2006). Thus, for the purpose of this research, waste rattan are collected and used as filler in natural rubber composites. Successful use of waste rattan as filler will help to create a more environmentally friendly product. This paper highlights the use of rattan as a new type of filler as well as the use of 3-aminopropyltrimethoxysilane (AMEO) as silane coupling agent in natural rubber composites. The effect of filler loading and the silane coupling agent on fatigue life, fatigue fracture surface observations using scanning electron microscopy, and thermal aging of rattan powder-filled natural rubber composites were investigated. The results of curing characteristics, tensile properties, studies on fracture surface morphology, and extent of rubber-filler interaction of rattan powder-filled natural rubber composites were reported in a previous article (Ismail et al. 2011).

## EXPERIMENTAL

### Materials and Formulation

Table 1 shows the formulation and materials and their suppliers used in this research study. Rattan wastes were collected, cleaned, ground, and sieved to an average particle size less than 180  $\mu\text{m}$ .

**Table 1.** Formulation of Rattan Powder Filled NR Composites

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

<sup>a</sup>CBS - N-cyclohexyl-2-benzothiazolsulfenamide

<sup>b</sup>BKF - 2,2 methylene-bis-(4-methyl-6-tert-butylphenol)

<sup>c</sup>AMEO – aminopropyltrimethoxysilane

### Sample Preparation

Mixing for both series was carried out in a laboratory model two roll mill according to American Standard of Testing Material (ASTM) designation D 3184-80.

The first series was without the addition of the silane coupling agent, while the second series included the addition of the silane coupling agent. The cure characteristics of the rubber composites were studied using a Monsanto Rheometer, model MDR 2000 at 150°C. Cure time ( $t_{90}$ ), scorch time ( $t_{s2}$ ), and maximum torque ( $M_H$ ) were obtained from the rheographs. The rubber composites were molded into sheets with their respective cure time,  $t_{90}$  at 150°C using a hot press machine.

### Fourier Transform Infrared (FTIR)

The chemical changes in samples were detected using Fourier-transform infrared spectroscopy (FTIR; Perkin Elmer System 2000). The FTIR spectra were recorded in the range of 550-4000  $\text{cm}^{-1}$  wave numbers with an average of 24 scans.

### Thermal Aging

Thermal aging was carried out according to ASTM D 573. The vulcanized rubber sheets were exposed to the deteriorating influence of air at temperature of 70°C in an oven for an interval time of 7 and 14 days. The rubber sheets were then cooled at room temperature, cut into dumbbell-shaped samples, and analyzed for tensile properties in accordance with D 412. The deterioration of physical properties was determined by comparing the tensile properties of aged samples with the properties of unaged samples. The percent retention (%) of tensile properties such as tensile strength, elongation at break, stress at 100% elongation (M100) and stress at 300% elongation (M300) were calculated using the following Equation 1,

$$\text{Tensile properties retention (\%)} = [(P_u - P_a) / P_u] \times 100 \quad (1)$$

where  $P_u$  is the respective tensile property of the unaged sample, and  $P_a$  is the tensile property of the aged sample.

### Fatigue Life

The rubber composites were molded into rectangular sheets ( $22.9 \times 7.6 \times 0.15$  cm) with beaded edges at 150°C for a respective cure time ( $t_{90}$ ). Individual dumbbell samples were cut at right angles to the grain using a BS type E dumbbell cutter. Fatigue tests of the samples were then carried out on a Monsanto Fatigue To Failure Tester (FTFT). The samples were subjected to repeated cyclic strain at 100 rpm. Six samples were used for each test. The numbers of cycles were recorded automatically. The fatigue life in kilocycles (kc) for each sample was computed as the Japanese Industrial Standard (J.I.S.) average, which was obtained from the four highest values, and recorded using the formula in Eq. 2 (Ismail and Haw 2008),

$$\text{J.I.S. average} = 0.5A + 0.3B + 0.1(C + D) \quad (2)$$

where  $A$  is the highest value, followed by  $B$ ,  $C$ , and  $D$ .

### Scanning Electron Microscopy (SEM)

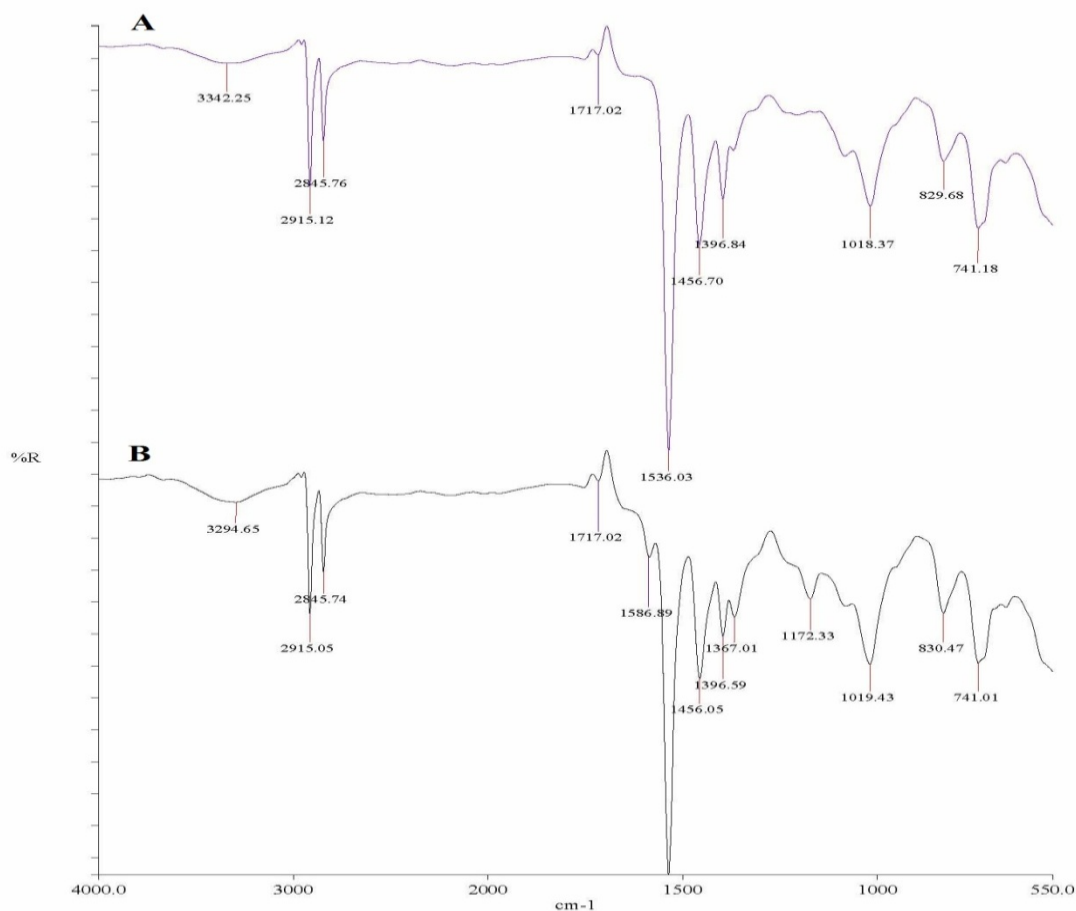
Examination of the fatigue-fractured surfaces was carried out using scanning electron microscopy (SEM), device Zeiss Supra 35vp. The fracture surfaces were sputter-

coated with gold to avoid electrostatic changing and poor image resolution. The fatigue failure, rubber-filler interactions, and filler dispersion were evaluated from the micrograph.

## RESULTS AND DISCUSSION

### Fourier Transform Infrared (FTIR)

Figure 2 shows the results of Fourier Transform Infrared (FTIR) analysis of 30 phr rattan powder-filled natural rubber (NR) composites; A represents the NR composite without the silane coupling agent, and B refers to the NR composite with the silane coupling agent. Formation of two additional peaks was observed in the FTIR spectrum of composite with the silane coupling agent, as compared to composite without the silane coupling agent. The peaks were found in the region of  $1172\text{ cm}^{-1}$ , which can be attributed to Si-O-C bonds, and  $1586\text{ cm}^{-1}$  corresponds to the C-N bond. The existence of Si-O-C and C-N bonds is supported by the possible interaction, which is illustrated in Fig. 1. Thus, the formation of these bonds proves that the silane coupling agent achieved a better interaction between the rattan filler and the rubber matrix.



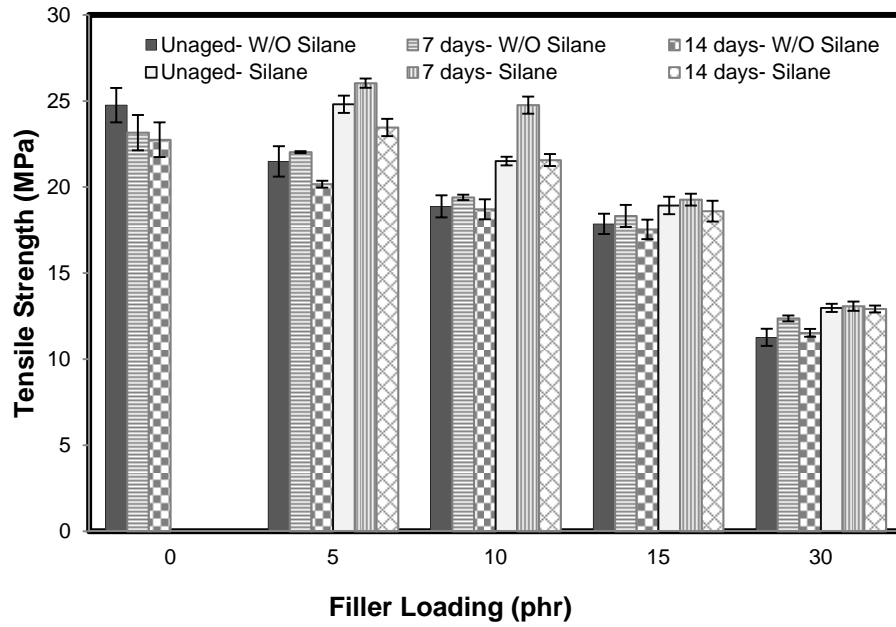
**Fig. 2.** FTIR Spectrum of 30 phr rattan powder-filled NR composite (a) A- without silane coupling agent (b) B- with silane coupling agent

## Thermal Aging

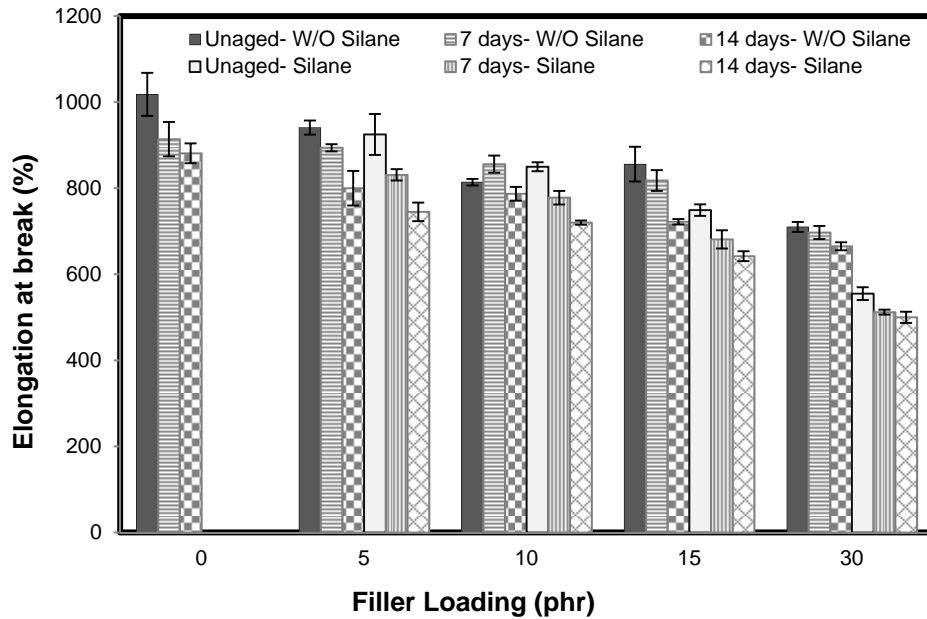
The resistance of rubber compounds to thermal aging is considered to be an essential requirement for better service performance of rubber products. The effect of filler loading and the silane coupling agent on tensile properties such as tensile strength, elongation at break, stress at 100% elongation (M100), and stress at 300% elongation (M300) of the unfilled natural rubber (NR) compound and the rattan powder-filled NR composites after aging are shown in Figs. 3 through 6. The aging properties of rubber compounds were determined by comparing the changes in mechanical properties before and after aging process. Figures 3 through 6 clearly display that for both composites with and without the silane coupling agent, tensile strength and elongation at break decreased, whilst the tensile modulus increased as the aging duration was prolonged to 14 days. Other research (Ismail et al. 1997) reported similar observation using rice husk ash and commercial fillers in epoxidized natural rubber compounds. The legend 'Silane' in Figs. 3 through 6 refers to the NR composites with the silane coupling agent, and 'W/O Silane' indicates the NR composite without the silane coupling agent.

According to Fig. 3, the tensile strength of rattan powder-filled NR composites with and without the silane coupling agent increased when aged for 7 days. The increment in tensile strength is due to the presence of free sulfur in composites, which is not involved in the cross linking reactions and leads to the formation of few new crosslinks during thermal aging. This phenomenon is well known as post-curing during aging (Abdul Kader and Bhowmick 2003). Meanwhile, tensile strength of the unfilled NR compound was reduced after being aged for 7 days, as displayed in Fig. 3. Again, this is due to the effect of post-curing during aging, which resulted in the formation of excessive crosslinks, leading to reduced tensile strength (Rattansom and Prasertsri 2009). The presence of rattan fillers in the composites might have retarded the formation of excessive crosslinks in NR composites as compared to unfilled NR compound. As the aging duration prolonged to 14 days, reduction in tensile strength is seen for all samples due to the dissociation of existing crosslinks and the structural changes in rubber chains. As a consequence, natural rubber loses its elastomeric properties and its ability to act as an effective matrix material to transmit stress (Ismail et al. 1997). Thus, a reduction in tensile strength was clearly observed for all samples.

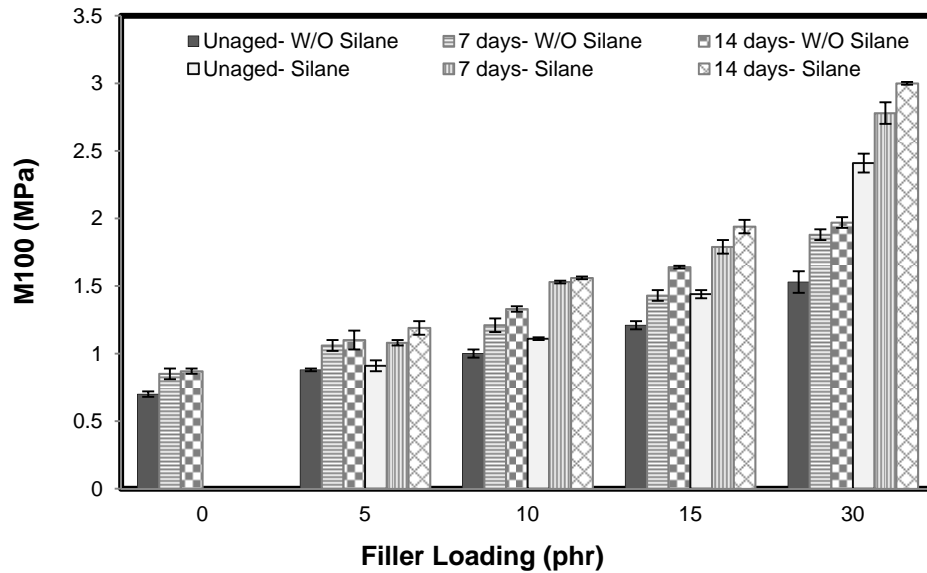
Figures 4 through 6 present the results of elongation at break, stress at 100% elongation, and stress at 300% elongation of unfilled NR compound and rattan powder-filled NR composites with and without the silane coupling agent after aging. Elongation at break decreased with increasing aging, whereas the opposite trend was observed for the tensile modulus. After aging for 7 days, all samples exhibited a higher tensile modulus compared to the unaged samples. This is due to the reduction in mobility of the rubber chains resulting from an increase in crosslink density from post-curing. Meanwhile, a further increase in the modulus was observed when aging was prolonged to 14 days. This is generally attributed to the dissociation and rearrangement of sulfides bonds, which tend to increase the stiffness of the NR composites (Ismail et al. 1997). A reduction in elongation at break was observed for all aged samples, as a consequent of both cross linking and scissioning reactions, which lead to a decrease in elastic nature of the rubber chain (Azura et al. 2008; Khanlari and Kokabi 2011).



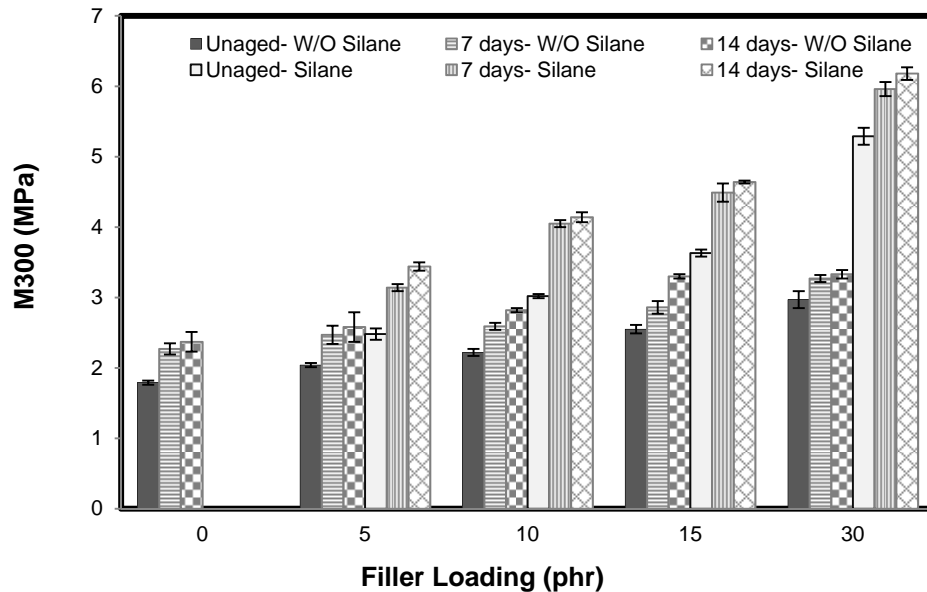
**Fig. 3.** Effect of filler loading on tensile strength of rattan powder-filled natural rubber composites with and without silane coupling agent after aging process



**Fig. 4.** Effect of filler loading on elongation at break of rattan powder-filled natural rubber composites with and without silane coupling agent after aging process



**Fig. 5.** Effect of filler loading on stress at 100% elongation (M100) of rattan powder-filled natural rubber composites with and without silane coupling agent after aging process



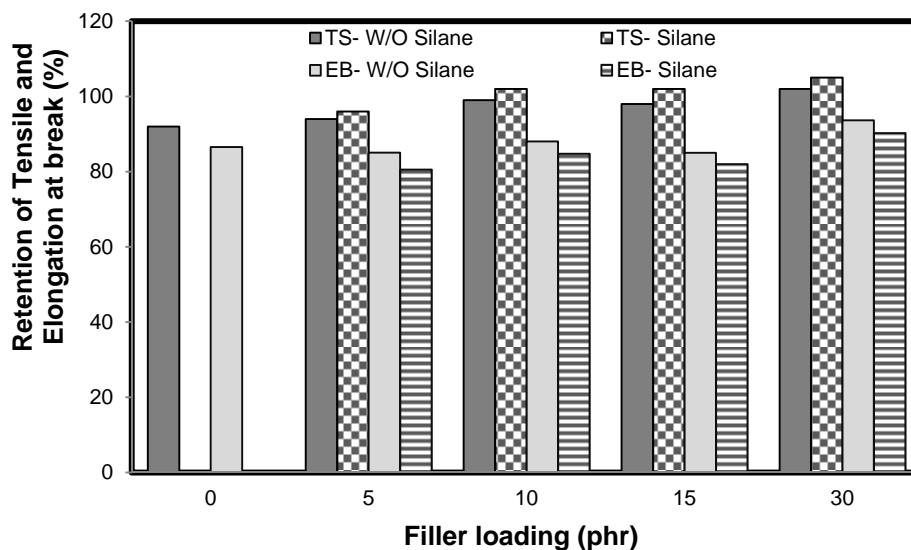
**Fig. 6.** Effect of filler loading on stress at 300% elongation (M300) of rattan powder-filled natural rubber composites with and without silane coupling agent after aging process

In order to quantify the effect of filler loading and the silane coupling agent on thermal aging, the percent retention (%) of tensile properties was investigated. It can be seen from Figs. 7 through 8 that the unfilled natural rubber (NR) compound and all rattan powder-filled NR composites retained an average tensile strength of 99%, an elongation at break of 88%, and about 133% and 125% for M 100 and M 300, respectively for both with and without silane coupling agent. Also, the percentage of retention of tensile properties showed no significant change with an increase in filler loading in the rubber composites. This implies that the changes in tensile properties during aging can be attrib-

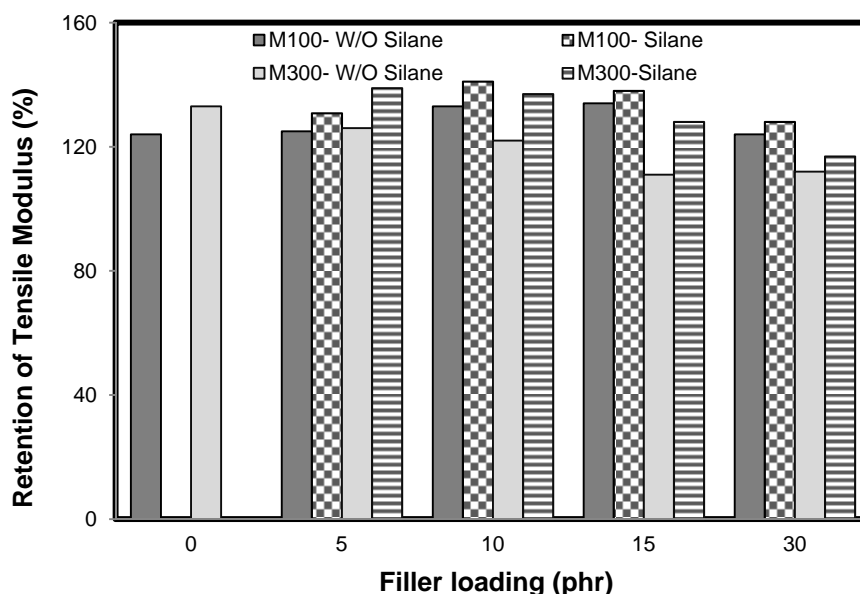


uted solely to the nature of the rubber and are independent of the rattan filler and its loading in composites. This proves that rattan fillers do not undergo degradation upon aging. The deterioration in tensile properties is mainly due to the thermal degradation of natural rubber which has undergone main-chain scission, crosslink formation, and crosslink breakage (Mostafa et al. 2009). Jacob et al. (2007) also reported a similar observation in their study of hybrid biofiber reinforced natural rubber biocomposites. The deterioration of the tensile properties, however, is not severe, as the rubber contains antioxidants to retard aging process.

The percent of retention (%) of composites showed improvement in the case of the silane coupling agent when compared with composites without the silane coupling agent, as shown in Figs. 7 through 8. The observed improvement, however, was very little, about an average increase of only 3% and 6% with respect to tensile strength and tensile modulus, while about 4% of reduction was observed for elongation at break of all composites. Natural rubber and rattan filler are incompatible with each other; thus, the incorporation of the silane coupling agent showed to improve the compatibility between NR matrix and the rattan surface. The silane coupling agent enhanced the wettability of the rattan by the rubber matrix and promoted interfacial adhesion (displayed in Fig.1). As a consequence, the effectiveness of stress being transferred from the matrix to the rattan also was improved. Thus, the tensile strength retention of the samples with silane coupling agent was higher than for samples without silane coupling agent, as presented in Fig. 7. Based on Fig. 8, an increment in retention of tensile modulus is also noted for samples with silane coupling agent as compared to without silane coupling agent. This modulus enhancement can be accredited to better adhesion between the rubber matrix and the rattan filler, resulting in stiffer and more rigid composites. Accordingly, the elasticity of the composites is reduced. Therefore, the retention of elongation at break of the samples with silane coupling agent is lower in comparison to samples without silane coupling agent, as shown in Fig. 7.



**Fig. 7.** Retention of tensile strength and elongation at break of rattan powder-filled natural rubber composites with and without silane coupling agent after aging process

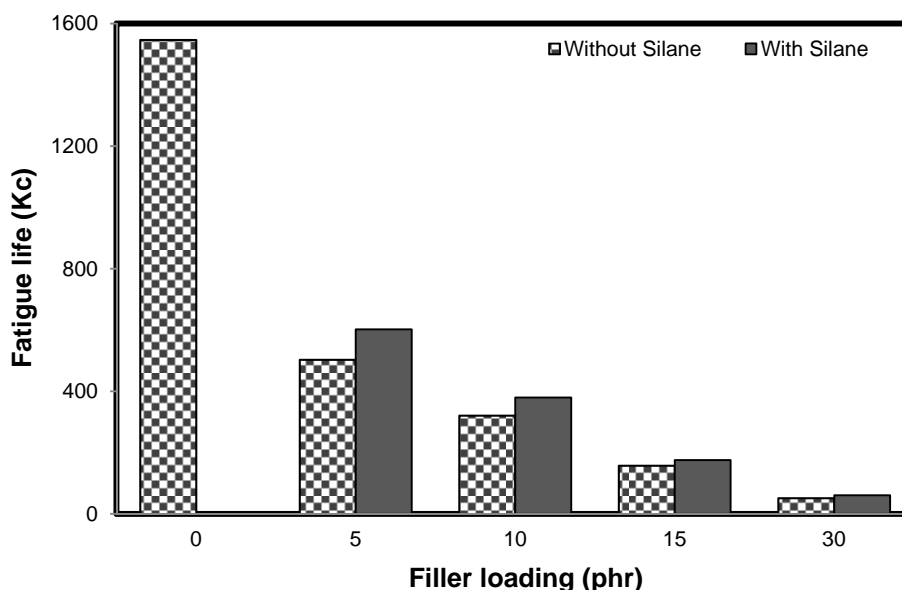


**Fig. 8.** Retention of stress at 100% elongation (M100) and stress at 300% elongation (M300) of rattan powder-filled natural rubber composites with and without silane coupling agent after aging process

### Fatigue Life

Figure 9 represents the dependence of fatigue life of rattan powder-filled natural rubber (NR) composites on filler loading and silane coupling agent. Figure 9 clearly shows that the fatigue life of NR composites decreased noticeably as the filler loading increased. Other research efforts (Ishak et al. 1997; Ismail and Haw 2008; Ismail and Jaffri 1999) also obtained similar results using natural fibers such as palm ash, oil palm wood flour, and rice husk ash as filler in rubber composites. Considering Fig. 9, it can be concluded that the unfilled NR compound exhibits a higher fatigue life value than the rattan powder-filled composites. This is due to the ability of natural rubber to crystallize upon stretching. Strain induced crystallization have a beneficial effect on the fatigue fracture properties of rubber by inducing strong microstructural changes in the crack tip region (Saintier et al. 2011).

A decrease in fatigue life of rattan powder-filled NR composites was also observed as the filler loading increased, as seen in Fig. 9. This reduction of fatigue life proves that rattan filler has a significant influence on fatigue life. The effect of filler on fatigue properties is attributed to pronounced changes induced by fillers on stiffness, non-homogeneity of the rubber-filler composites at crack tip, and agglomeration of filler particles, resulting in increased effective initial flaw sizes (Mars 2004). Adding unstrained and rigid rattan fillers to rubber tends to form aggregates in between flexible chains and restrict the mobility of natural rubber molecules. Thus, the stiffness of composites increases and the ability of rubber to deform elastically under applied cyclic stress is lessened. The fatigue life of natural rubber composites decreases as rubber is unable to relieve cyclic stress efficiently. Hainsworth (2007) mentioned that fatigue life increases as the flexibility of the chain increases, given that more flexible chains have a higher resilience to cyclic deformation.



**Fig. 9.** Fatigue life of rattan powder-filled natural rubber composites with and without silane coupling agent

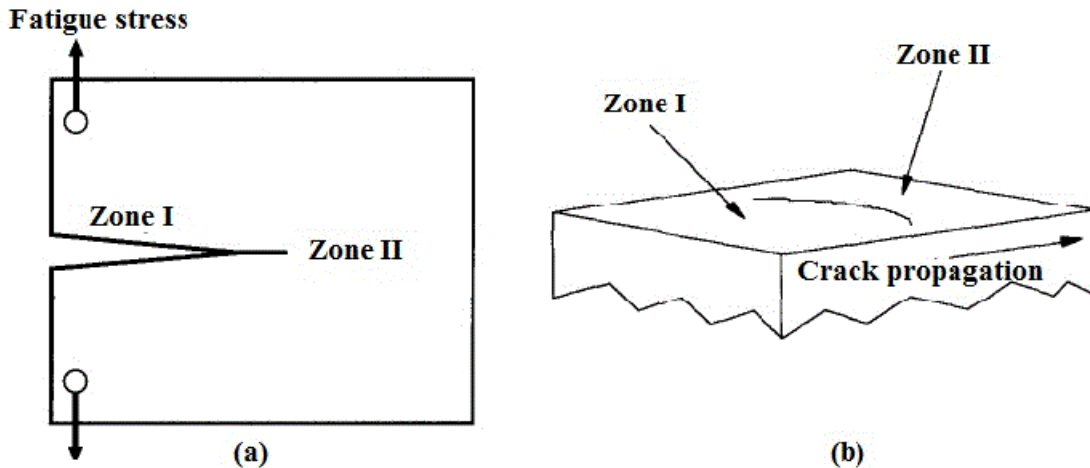
Apart from increase in stiffness of composites, the filler aggregates also act as stress concentrators and become points at which crack growth is initiated when stress applied exceeds a critical value (Mars and Fatemi 2002a). In addition, the decrement of the fatigue life of rattan powder filled natural rubber composites is also due to the heterogeneous nature of the composites, resulting in poor adhesion or incompatibility between polar rattan filler and non polar rubber matrix. As a consequence, a weaker interfacial adhesion is results. Thus, crack growth resulting from filler agglomeration along with weak interfacial adhesion will lead to fatigue failure. The fatigue life of rattan powder-filled NR composites shortens with increase in filler loading.

The fatigue life of the rattan powder-filled NR composites, nonetheless, showed improvement with the addition of the silane coupling agent, as shown by Fig. 9. Similar results have been reported by other researchers as well (Ismail and Haw 2008; Ismail and Jaffri 1999). The addition of the silane coupling agent improved the wettability of the rattan filler in the rubber matrix, promoting interfacial adhesion. Moreover, the silane coupling agent has the ability to reduce filler-filler interaction and facilitate the dispersion of the rattan in the rubber matrix. Furthermore, formations of stress concentration points are reduced due to be the better filler dispersion and improved wettability of the filler and the matrix. No major improvements were imparted by the silane coupling agent in the fatigue life of composites at higher filler loading, especially at 30 phr rattans loading. It has been reported that fiber-reinforced composites have shorter fatigue limit due to poor crack growth resistance under dynamic conditions (Bhattacharya and Bhowmick 1995).

### Surface Morphology Study

Figures 11a, 11b, and 11c present scanning electron microscopy (SEM) micrographs of the fatigue fracture surfaces of the unfilled natural rubber (NR) compound and the rattan powder-filled NR composites at 5 and 30 phr rattan loadings,

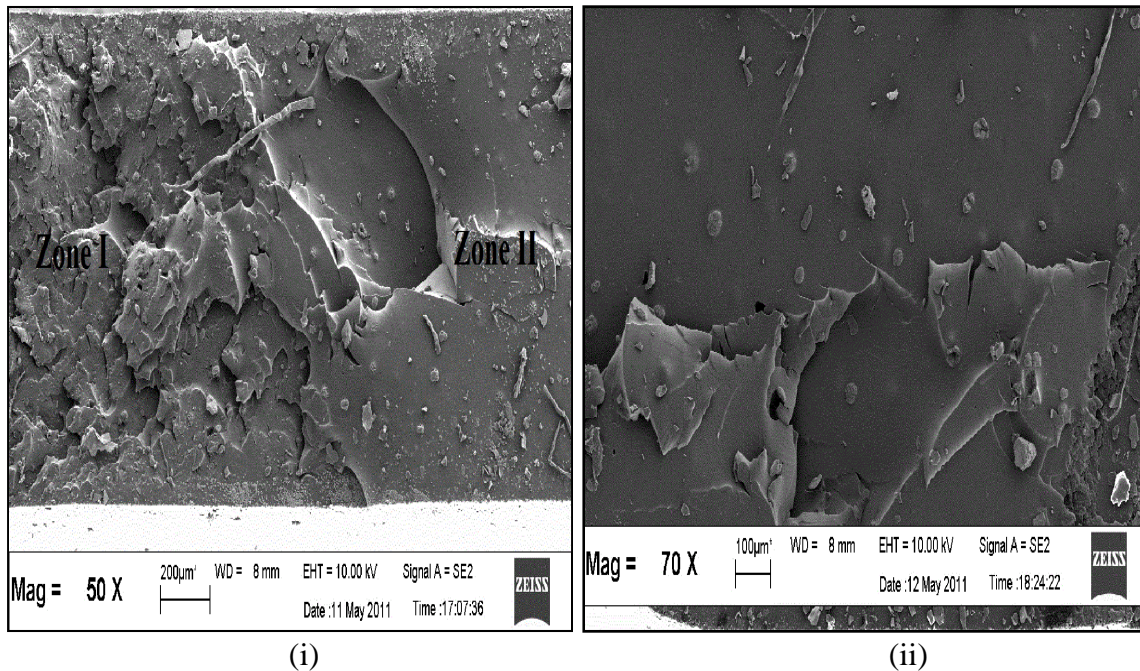
respectively. The fatigue fracture surface morphologies give information about fatigue failure along with the level of matrix and filler adhesion. Generally, studies on the morphology of fatigue fracture surfaces reveal that unfilled NR compound and filled NR composites exhibit two failure zones. Such a description is exemplified by the unfilled NR compound in Fig. 11 (a), whereby, the failure zones are indicated as Zone I and Zone II. Figure 10 represents a simple diagram illustrating the direction of crack propagation and the failure zones in a sample.



**Fig. 14.** Schematic diagram showing the direction of crack propagation and the failure zones (a) side view (b) plan view

Zone I is the first stage and referred to as “fatigue crack growth.” This zone represents the development of cracks under cyclic loading from an initial state in which the crack of interest is known (Harbour et al. 2008). This cyclic deformation results in the propagation of cracks within the samples. The rough nature of the failure surface indicates that the fracture process is relatively slower and some matrix tearing has occurred before fracture, as illustrated in Fig. 11a (i). As the failure process progresses, the stress distribution in the samples are no longer uniform, and the samples is unable to resist the crack propagation. Thus, fast and unstable propagation is believed to occur, leading to catastrophic failure and resulting in the formation of a smoother failure surface, indicated as Zone II in Fig. 11a (i).

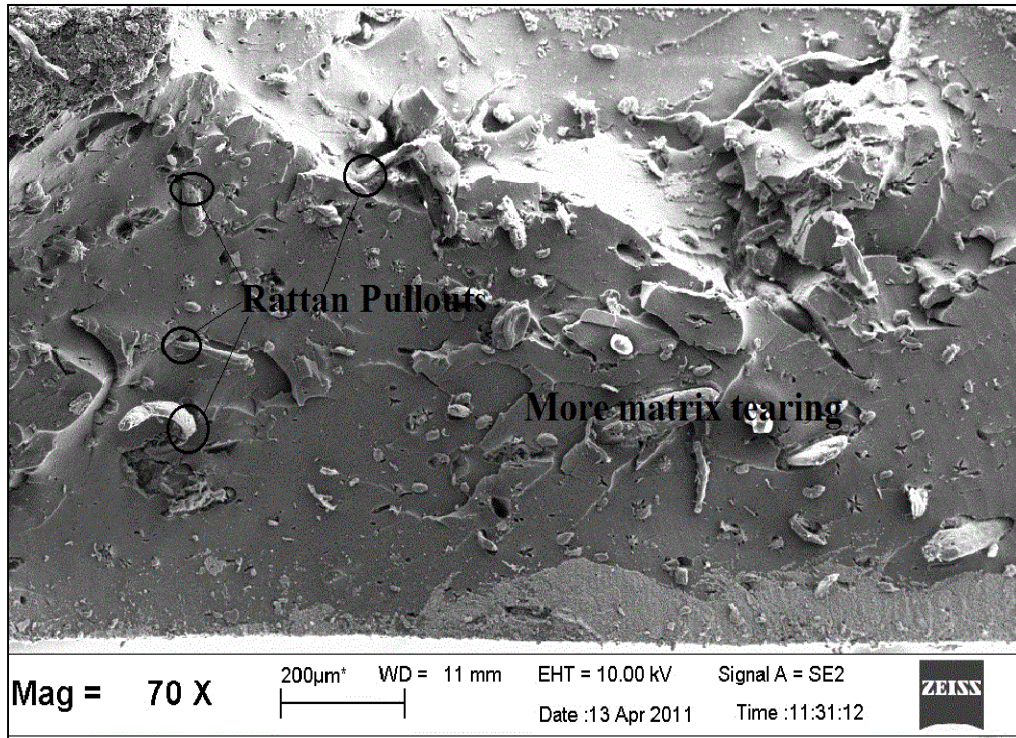
The fatigue fracture surface morphologies for 5 phr and 30 phr rattan powder-filled NR composites with and without the silane coupling agent are illustrated in Figs. 11b and 11c. For this study, the smooth surface fracture at a magnification of 70X will be area of interest, as shown in Figs. 11a(ii), 11b(i), 11b(ii), 11c(i) and 11c(ii). These micrographs reveal that with increasing filler loading, the fracture surfaces become rougher due to the increase in stiffness of the composites. The unfilled NR compound, presented in Fig. 10a(ii), displays matrix tearing with smoother fracture surface. More matrix tearing is observed at lower rattan loading, 5 phr as compared to 30 phr rattan loading.



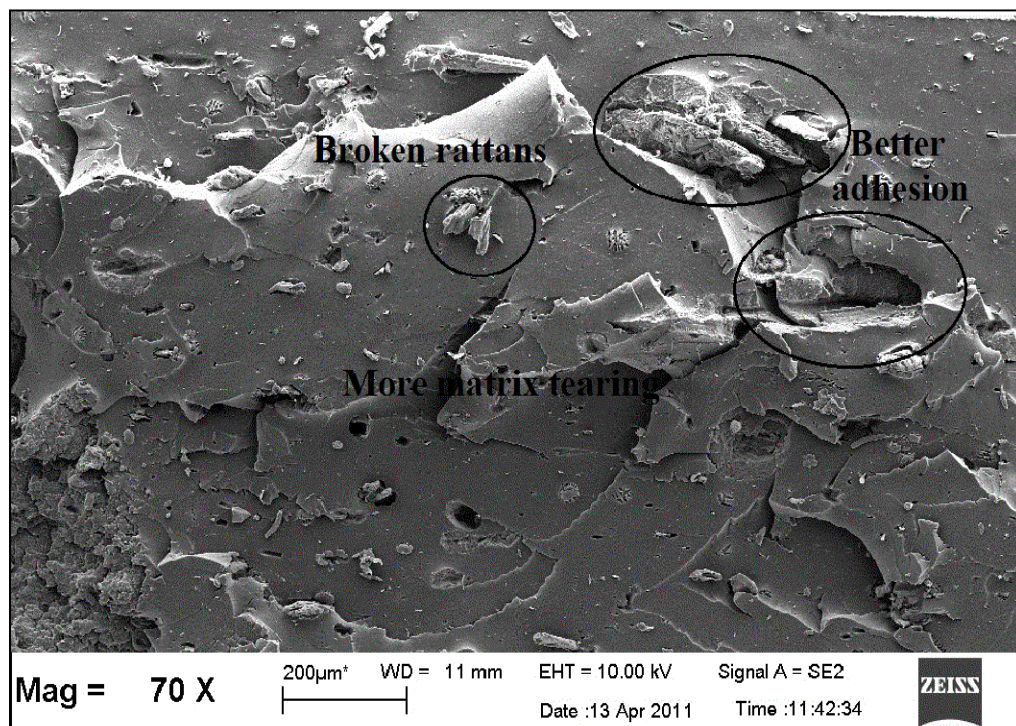
**Fig. 11a.** SEM Micrograph of fatigue fractured surface of unfilled natural rubber compound (i) failure zones at magnification of 50× (ii) smooth failure surface at magnification of 70×

Apart from that, fatigue fracture surfaces also exhibit evidence of more rattan pullouts or detachment of rattan from the rubber matrix and rattan agglomerations with increasing rattan loading. These observations resulted from poor wetting of the filler by the matrix, giving rise to poor interfacial adhesion between the rattan and the rubber matrix and causing the formation of weak interfacial regions and agglomerations. During fatigue testing, cracks travel through the rubber matrix along the weaker interfacial regions. The weaker interfacial regions cannot resist crack propagation as effectively as the rubber matrix, resulting in debonding and frictional pullouts of filler. The extensive pull outs and poor distribution of rattans in the rubber matrix can be seen in Figs. 11c(i) and 11c(ii). NR composite with 30 phr rattan loading (Fig. 11 c) showed more rattan pullouts and rattan agglomeration than the 5 phr rattan powder-filled NR composite (Fig. 10b), providing evidence for the reduction of fatigue life of the composites as more rattan fillers are added to the composites.

The rattan fillers were well dispersed throughout the rubber matrix and fewer agglomerates were observed in the presence of the silane coupling agent, as seen in Figs. 11b(ii) and 11c(ii). The silane coupling agent has the ability to form a uniform composite structure by reducing filler agglomerations. This effect is clearly seen in Fig. 11c(ii), where there is less filler agglomeration and better filler dispersion as compared to NR composite without silane coupling agent, Fig. 11c(i). At 5 phr loading, the rattans show better wetting with the matrix, less detachment of rattan filler from matrix, and some broken rattan at the surface, as shown in Fig. 11b(ii). This is due to the ability of the silane coupling agent to promote higher interfacial adhesion between the filler and the matrix resulting in improved efficiency of crack resistance. Thus, the rattan filler tend to break from rubber matrix rattan, rather than completely pullout from the matrix.

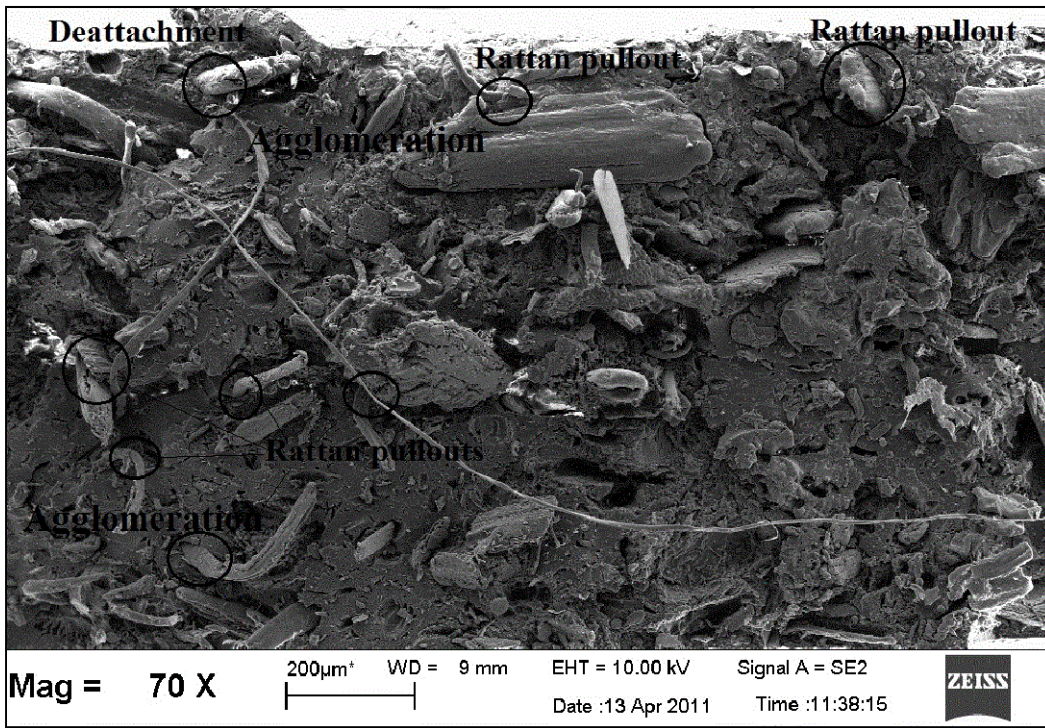


(i)

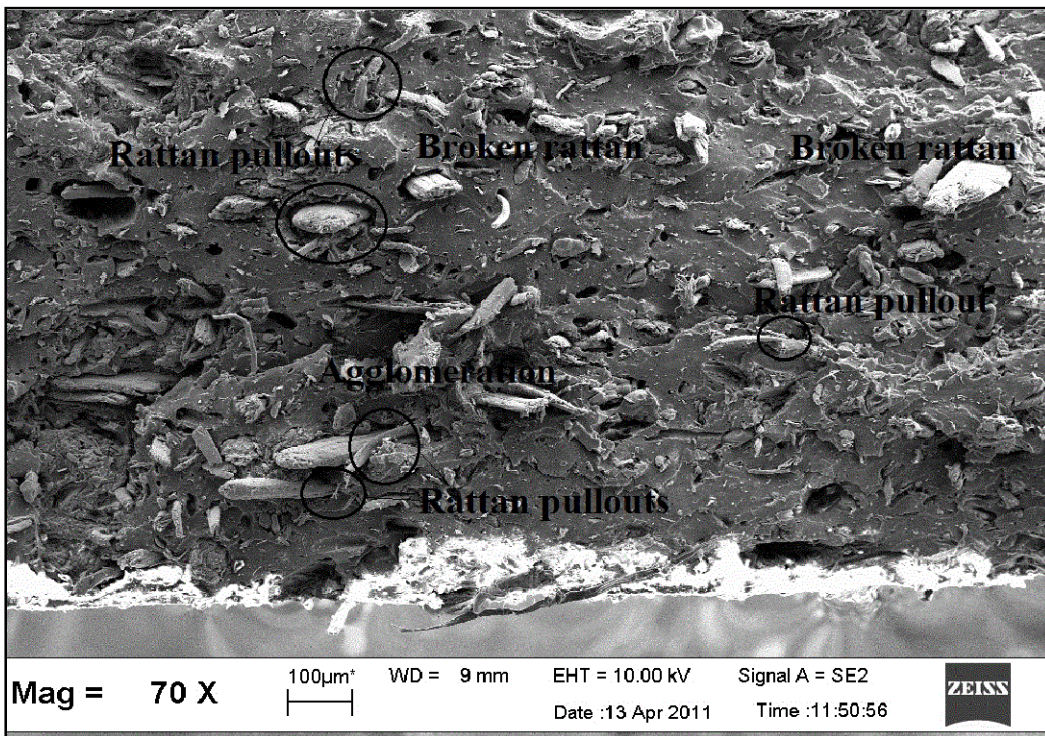


(ii)

**Fig. 11b.** SEM Micrograph of fatigue fractured surface of 5 phr rattan powder-filled natural rubber composites (i) without silane coupling agent (ii) with silane coupling agent at magnification of 70x



(i)



(ii)

**Fig. 11c.** SEM Micrograph of fatigue fractured surface of 30 phr rattan powder-filled natural rubber composites (i) without silane coupling agent (ii) with silane coupling agent at magnification of 70x

This has improved the fatigue life of the composite. As the filler loading increases, however, the efficiency of the silane coupling agent to form uniform structure and promote interfacial adhesion is reduced, which leads to poor improvement in fatigue life. Referring to Fig. 11c(i) and 11c(ii), although there is a reduction in rattan agglomeration and rattan pullouts in 30 phr rattan powder-filled NR composite with silane coupling agent, in comparison to without silane coupling agent, the rattan agglomeration and rattan pullouts still could be observed in the composite, leading to poor fatigue failure.

## CONCLUSIONS

1. The incorporation of rattan powder as a filler in natural rubber shortens the fatigue life of rattan powder-filled NR composites. The formation of agglomerates with poor adhesion between the filler and the rubber matrix, which results in weaker interfacial adhesion, eases the crack growth in the rubber matrix and leads to fatigue failure.
2. Thermal aging of composites increases the tensile modulus but reduces the tensile strength and elongation at break, which can be attributed to the deterioration of the rubber matrix. The deterioration of tensile properties is not influenced by the rattan filler.
3. An improvement in fatigue life and thermal aging properties is observed with the addition of a silane coupling agent, which promotes better adhesion between the rattan filler and the rubber matrix. This observation is supported by SEM studies of fatigue-fractured surfaces and the FTIR studies.

## ACKNOWLEDGMENT

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