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# Sisal Fiber/Natural Rubber Composites: Effect of Fiber Content and Interfacial Modification

Wittawat Wongsorat[1,2,a], Nitinat Suppakarn[1,2,b] and Kasama Jarukumjorn[1,2,c*]

[1]School of Polymer Engineering, Institute of Engineering, Suranaree University of Technology, Nakhon Ratchasima 30000, Thailand

[2]Center for Petroleum, Petrochemicals, and Advanced Materials, Chulalongkorn University, Bangkok 10330, Thailand

[a]w.wongsorat@gmail.com, [b]nitinat@sut.ac.th, [c*]kasama@sut.ac.th

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**Abstract.** Sisal fiber/natural rubber (NR) composites were prepared by the incorporation of sisal fiber into NR at contents of 10-30 phr. Fiber treatment (alkalization) and adding maleic anhydride grafted natural rubber (NR-g-MA) were used to improve interfacial adhesion between sisal fiber and NR matrix. Mechanical properties, morphologies, and cure characteristics of the composites were studied. With increasing fiber content, modulus at 100% strain (M100), modulus at 300% strain (M300), and hardness of the composites increased whereas tensile strength and elongation at break decreased. Cure time of the composites decreased with increasing fiber content but scorch time was not much affected by fiber content. Alkali treated sisal fiber/NR composite exhibited higher tensile properties and hardness than untreated sisal fiber/NR composite at all fiber content due to the improved adhesion between fiber and NR matrix through the mechanical interlocking mechanism. Alkalization showed no effect on scorch time and cure time of the composites. The addition of NR-g-MA into the composites increased M100, M300, tensile strength, and hardness but prolonged scorch time and cure time. NR-g-MA provided more effective improvement of the mechanical properties of the composites when compared to fiber alkalization.

## Introduction

Nowadays, natural fibers are received much attention as alternative fillers for plastic and rubber products. Sisal fiber is one of the most interesting candidates for reinforcing natural rubber due to its high specific strength and modulus, low density, renewability, and no health risk. In addition, sisal fiber is a tropical plant which can cultivate in a wide range of area in Thailand. Short natural fiber reinforced natural rubber composites have gained more interest due to low specific weight, renewable resource, ease of processing, and economic advantages [1]. The main problem of using natural fiber to reinforce NR is the incompatibility between a hydrophilic natural fiber and a hydrophobic NR, leading to poor mechanical properties of the composites. The interfacial adhesion between the natural fiber and NR can be improved by various methods such as fiber surface treatment [1-3], matrix modification [4,5], and addition of compatibilizer [6,7]. In this work, the effect of fiber content and interfacial modification (alkalization and addition of NR-g-MA) on mechanical properties, morphologies, and cure characteristics of sisal fiber/NR composites were investigated.

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## Materials and Method

**Materials.** Natural Rubber (NR, STR 5L) was supplied from Thai Hua Co., Ltd. Sisal fiber (SF) was purchased from Sisal-Handicraft OTOP group, Nakhon Ratchasima, Thailand. NR-g-MA prepared in house was used as a compatibilizer at content of 5 phr. The other ingredients used to vulcanize natural rubber were stearic acid, zinc oxide, N-cyclohexyl-2-benzothiazole-2-sulphenamide (CBS), and sulfur.

**Fiber Preparation.** Sisal fiber was cut into an approximate length of 2 mm then dried in an oven at 60$^{o}$C overnight. This fiber was called untreated fiber (UT). In order to eliminate hemicellulose, waxes, and low molecular weight species, the UT fiber was treated with 2 wt% sodium hydroxide (NaOH) solution for 2 hrs. Then, the UT fiber was washed with water and dried at 60$^{o}$C overnight. This fiber was called alkali treated fiber (AT).

**Sample Preparation.** Formulations for compounding are shown in Table 1. The compounds were mixed using a two-roll mill (CHAICHAREON) at room temperature. The fiber contents used in this study were 10, 20, and 30 phr. The test specimens were prepared using a compression molding machine (GOTECH: GT-7014-A30) under a pressure of 130 MPa and a temperature of 150$^{o}$C.

**Properties Mesurement.** Cure characteristics were measured on a moving die rheometer (MDR, GOTECH: M2000F) at temperature of 150$^{o}$C. Tensile properties were tested according to ASTM D412 on a universal testing machine (INSTRON: 5565). Hardness was determined according to ASTM D2240 on an international rubber hardness degrees tester (IRHD, BAI EISS: DIGI TEST). Morphologies of the composites were examined using a scanning electron microscope (SEM, JEOL: JSM-6400). The samples were coated with gold before analysis.

Table 1. NR formulations.

| Materials | Contents [phr] |
|---|---|
| Natural rubber | 100 |
| Zinc oxide | 5.0 |
| Stearic acid | 1.5 |
| CBS | 0.5 |
| Sulfur | 2.5 |
| Sisal fiber (UT or AT) | 0, 10, 20, 30 |
| NR-g-MA | 0, 5 |

## Results and Discussion

**Cure Characteristics.** Maximum torque (MH), minimum torque (ML), scorch time ($t_{s1}$), and cure time ($t_{90}$) of NR and NR composites are listed in Table 2. With increasing fiber content, MH and ML values increased due to an increase in stiffness of the composites and a reduction of the deformation of NR molecules. As fiber content increased the scorch time of the composites was not much affected but the cure time decreased. At the same fiber content, AT fiber filled NR composites exhibited higher MH and ML values than the NR composites filled with UT fiber. When fibers were treated with alkali solution, hemicellulose, wax, and other impurities covering the external surface of fibers were removed resulting in increased surface roughness of the fibers. So, the adhesion between fiber and NR matrix was improved through the mechanical interlocking mechanism. The alkalization had no significant effect on the scorch time and cure time of the composites. NR-g-MA compatibilized NR composites showed higher MH and ML values than uncompatibilized NR composites (NR composites filled with UT fiber). This might be due to the formation of a good interaction between fiber and NR matrix with the presence of compatibilizer. Scorch time and cure time of the NR-g-MA compatibilized NR composites were longer than those of the NR, uncompatibilized NR composites, and NR composites filled with AT fiber. This may be due to the acidity of maleic acid and the interaction between maleic anhydride and the accelerator [8].

**Mechanical Properties.** Modulus at 100% strain (M100), modulus at 300% strain (M300), tensile strength, and elongation at break of NR and NR composites are listed in Table 3. When fiber content was increased, M100 and M300 of the composites also increased. Tensile strength of NR was higher than those of all NR composites. NR inherently has high tensile strength owing to strain-induced crystallization. With incorporating the fibers into NR, the regular arrangement of rubber molecules was disrupted; thus, the efficiency of strain-induced crystallization was lost. So, the fiber reinforced NR composites had lower tensile strength than NR. In addition, tensile strength of UT fiber/NR composites decreased with increasing fiber content. High population of fiber led to fiber agglomeration resulting in difficult stress transmission from matrix to fiber and disturbance of the continuity of the matrix phase [3]. This led to a reduction of the tensile strength of the composites. Moreover, when fiber content was increased the composites became stiffer and harder leading to a reduction of resistance to break of the composites. With increasing AT fiber content, M100, M300, tensile strength, and elongation at break of AT fiber/NR composites also showed the same trend as the UT fiber/NR composites. However, all of the composites filled with AT fiber exhibited higher M100, M300, tensile strength, and elongation at break than the composites filled with UT fiber due to the enhancement in the interfacial adhesion between the fiber and NR matrix through mechanical interlocking. M100, M300, tensile strength, and elongation at break of NR-g-MA compatibilized NR composites drop off after fiber content of 10 phr. This was probably because the compatibilizer content was insufficient to improve the interfacial adhesion between the fiber and NR matrix. The addition of NR-g-MA into UT fiber/NR composites gave a positive impact on M100, M300, tensile strength, and elongation at break of the composites. This was attributed to the improvement in interfacial adhesion between the fiber and NR matrix by the presence of compatibilizers. AT fiber filled NR composite lower M100, M300, and tensile strength than NR-g-MA compatibilized NR composites. This suggested that adding NR-g-MA into NR composite may have more efficiency to improve interfacial adhesion between fiber and NR matrix than the fiber alkalization. Hardness of the composites increased with increasing UT fiber content. Moreover, alkalization and adding NR-g-MA resulted in an increase in hardness of the NR composites. This was due to the improvement of adhesion between the fiber and rubber.

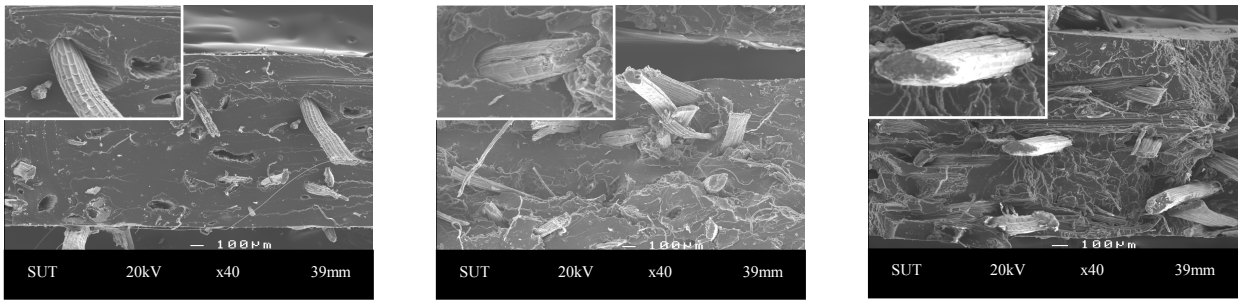
Table 2. Cure characteristics of NR, NR composites filled with UT and AT fiber, and NR-g-MA compatibilized NR composites.

| Formulation | MH [dN-m] | ML [dN-m] | Time [min] | |
|---|---|---|---|---|
| | | | $t_{s1}$ | $t_{90}$ |
| NR | 16.662 | 2.310 | 4.27 | 7.54 |
| NR/10 UT | 19.741 | 2.620 | 3.35 | 7.16 |
| NR/20 UT | 21.170 | 3.373 | 3.29 | 6.33 |
| NR/30 UT | 22.790 | 3.722 | 3.20 | 6.02 |
| NR/10 AT | 23.650 | 3.757 | 3.33 | 7.08 |
| NR/20 AT | 26.641 | 3.919 | 3.23 | 6.21 |
| NR/30 AT | 28.410 | 4.022 | 3.18 | 5.93 |
| NR/5 NR-g-MA/10 UT | 26.348 | 4.376 | 4.22 | 8.43 |
| NR/5 NR-g-MA/20 UT | 28.612 | 4.847 | 4.06 | 8.05 |
| NR/5 NR-g-MA/30 UT | 29.734 | 5.446 | 4.03 | 7.72 |

**Morphological Properties.** Tensile fracture surfaces of NR composites filled with UT and AT fiber, and NR-g-MA compatibilized NR composites at 10 phr of sisal fiber are shown in Fig. 1. From Fig. 1(a), there were many holes from fiber pull out after the tension was applied. This indicated the weak interfacial adhesion between the sisal fiber and NR matrix. For NR composite with the addition of NR-g-MA and AT fiber/NR composite, the better adhesion between the sisal fiber and NR matrix was observed as shown in Fig. 1(b) and 1(c).

Table 3. Mechanical properties of NR and NR composites.

| Formulation | M100 [MPa] | M300 [MPa] | Tensile strength [MPa] | Elongation at break [%] | Hardness [IRHD] |
|---|---|---|---|---|---|
| NR | 0.82±0.068 | 1.65±0.138 | 20.13±0.113 | 1350±17.67 | 39.34±0.195 |
| NR/10 UT | 1.09±0.081 | 1.75±0.132 | 11.05±0.121 | 1090±21.22 | 45.96±0.955 |
| NR/20 UT | 1.13±0.032 | 1.78±0.112 | 9.67±0.092 | 1050±19.03 | 49.14±0.776 |
| NR/30 UT | 1.22±0.046 | 1.92±0.171 | 8.70±0.146 | 920±23.49 | 55.18±0.756 |
| NR/10 AT | 1.36±0.022 | 1.83±0.075 | 13.09±0.082 | 1111±19.21 | 50.74±1.775 |
| NR/20 AT | 1.48±0.051 | 2.02±0.061 | 11.04±0.104 | 1070±24.60 | 52.82±0.726 |
| NR/30 AT | 1.59±0.044 | 2.26±0.052 | 9.42±0.093 | 950±18.44 | 59.16±0.896 |
| NR/5 NR-g-MA/10 UT | 1.67±0.033 | 2.52±0.035 | 15.75±0.087 | 1139±18.34 | 52.38±0.925 |
| NR/5 NR-g-MA/20 UT | 1.35±0.082 | 2.03±0.100 | 11.01±0.184 | 1107±17.67 | 56.89±0.939 |
| NR/5 NR-g-MA/30 UT | 1.43±0.007 | 2.04±0.056 | 9.44±0.219 | 961±19.29 | 61.12±0.676 |




(a) (b) (c)

Fig. 1 SEM micrographs of (a) NR/10UT, (b) NR/10AT, and (c) NR/5NR-g-MA/10UT composites.

## Summary

Maximum torque, minimum torque, modulus at 100% strain, modulus at 300% modulus, and hardness of the sisal fiber/NR composites increased with increasing fiber content while scorch time, cure time, tensile strength, and elongation at break decreased. NR-g-MA improved tensile properties, hardness, and prolonged scorch time and cure time of the composites. When compared to fiber alkalization, NR-g-MA resulted in better enhancement in the mechanical properties of the NR composites.

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