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Influence of Filler Types on Mechanical Properties and Cure Characteristics of Natural Rubber Composites

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Abstract. Natural rubber (NR) was reinforced with three types of filler: carbon black, calcium carbonate, and sisal fiber. NR composites were prepared on a two-roll mill. Filler content was 20 phr. Mechanical properties and cure characteristics of NR composites were studied. All NR composites had higher maximum torque than NR. NR filled with carbon black showed the highest maximum torque. However, scorch time and cure time of the NR composites were not much affected by filler types. In addition, influence of fiber treatment (alkalization) on mechanical properties and cure characteristics of sisal fiber-NR composites was investigated. Alkali treated sisal fiber-NR composite exhibited higher tensile properties and hardness than untreated sisal fiber-NR composite due to improved adhesion between the fiber and NR matrix. Moreover, alkali treated sisal fiber-NR composite had superior specific modulus and strength than NR composites filled with carbon black and calcium carbonate.

Introduction

Natural rubber (NR) is one of the versatile materials widely used in many applications. However, raw NR has poor mechanical properties. To improve its mechanical properties, ingredients such as accelerators, activators, crosslinking agents, and fillers are commonly mixed into rubber to obtain suitable properties [1]. In particular, the incorporation of fillers in NR matrix leads to significant improvement in mechanical properties of the NR composites [2]. Conventional fillers used in NR industry are carbon black, silica, and calcium carbonate ($CaCO_3$). Reinforcement of NR with natural fiber has gained more interest due to many advantages of the natural fiber such as low density, biodegradability, renewable nature, reduced tool wear. However, the main drawback of using natural fiber to reinforce NR is the incompatibility of hydrophilic natural fiber and hydrophobic NR matrix leading to poor mechanical properties of the composites [3,4,5]. In addition, the natural fiber has high sensibility to water absorption [6,7]. To overcome these problems, it is necessary to modify the fiber surface through the employment of physical and chemical methods to reduce the hydrophilic nature of natural fiber and improve the fiber-matrix bonding. Jacob et al. [8] reported that the treatment of sisal fibers with alkali solution such as sodium hydroxide (NaOH) significantly increased the mechanical properties of the NR composites. Lopattananon et al. [4] found that alkali treated pineapple fiber-NR composites had higher elongation at break than untreated pineapple fiber-NR composite. The improvement of tensile strength of the alkali treated pineapple fiber-NR composites was not found when the concentration of NaOH was less than 5% (w/v). This improvement was due to an increase in interfacial bond strength through mechanical interlocking between the fiber and the NR. De et al. [9] reported that alkali treated grass fiber-NR composites showed higher maximum torque and mechanical properties than the composites with untreated fiber

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due to the better adhesion between the fibers and the NR matrix. Cure time of the alkali treated grass fiber composites was lower than that of NR compounds. This can be attributed to the influence of pH on the grass fibers, since it is known that a vulcanization reaction becomes faster in an alkaline pH. The objectives of this study were to investigate the effect of filler types: carbon black, calcium carbonate, and sisal fiber and the influence of alkalization of sisal fiber on mechanical properties and cure characteristics of NR composites.

Materials and Method

Materials. Natural rubber (NR, STR 5L) was supplied from Thai Hua Co., Ltd. Carbon black N330 (Thai Tokai Carbon Product Co., Ltd.), calcium carbonate (Asia Pacific Specialty Chemical Limited), and sisal fiber (Sisal-Handicraft OTOP group, Nakhon Ratchasima, Thailand) were used as fillers. The average particle sizes of carbon black and calcium carbonate were 28 nm and 5.35 μm, respectively. 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 with an approximate length of 370 μm was dried in an oven at 60°C overnight. This fiber was called untreated fiber (UT). In order to prepare alkali treated fiber (AT), the UT fiber was treated with 2 wt% sodium hydroxide solution (NaOH) for 2 hrs. Then, the fiber was washed with water and dried at 60°C overnight.

Sample Preparation. All compounds were mixed under the same conditions on a two-roll mill (CHAICHAROEN) at room temperature. Formulations of rubber compounds used are shown in Table 1. Filler content used in this study was 20 phr. Test specimens were prepared by a compression molding machine (GOTECH, GT-7014-A30). The compression condition was processed at a temperature of 150°C and a pressure of 130 MPa.

Properties Measurement. Cure characteristics were measured on a Moving Die Rheometer (GOTECH, M2000F) at a temperature of 150°C. Composite density after crosslinking was determined according to JIS K 6350 on Electronic Densimeter (MIRAGE, MD-200S). Tensile properties were tested according to ASTM D 412 on a Universal Testing Machine (INSTRON, 5565) at an extension rate of 500 mm/min and a gauge length of 33 mm. Hardness was determined according to ASTM D 1415 on Micro hardness (BAIEISS, DIGI TEST).

Results and Discussion

Cure characteristics. Table 2 shows the maximum torque (MH), minimum torque (ML), scorch time (t_{s1}), and cure time (t_{90}) of NR composites with different types of fillers. Maximum torque values increased with the addition of conventional reinforcing fillers (carbon black and $CaCO_3$) and sisal fiber. This indicated an increase in stiffness of the composites. The presence of fillers imparted limitation to the deformation of NR molecules so the composites became stronger, harder, and stiffer. The composites with alkali treated fiber exhibited higher maximum torque and minimum torque than the composites with untreated 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 similar observation was reported by Mathew and Joseph [10] in isora fiber/NR composites. No significant effect of alkalization on the scorch time and cure time of the composites was found. On the other hand, De et al. [9] found that cure time of alkali treated grass fiber filled NR composites was faster when compared to untreated grass fiber filled composites. This was attributed to the influence of pH on the fiber (8.1). The vulcanization reaction became faster in alkaline pH. In our study, the fibers were treated with NaOH solution, then the fibers were washed in distrilled water until pH of the fiber reached 7. Therefore, the alkalization did not affect on the cure time of the composites. Carbon black-NR composite showed faster scorch time and slower cure time than other fillers filled NR

composites. This may be because cure characteristics of rubber composites depend on properties of fillers such as surface area, surface activity, particle size, moisture content, and metal oxide content [3]. Egwaikhide et al. [11] found that the coconut fiber filled NR showed slower cure time in comparison to carbon black filled NR. This was due to the larger surface area of carbon black; thus, providing greater interaction between the filler and rubber matrix.

Density. Density of the composites increased with adding filler into NR as shown in Table 3. The density of carbon black-NR composite was higher than those of $CaCO_3$-NR and sisal fiber-NR composites. Density of carbon black, $CaCO_3$, and sisal fiber were 1.73, 1.58, and 1.43 g/cm^3, respectively. Among all filler filled NR, sisal fiber-NR composite showed the lowest density. This suggested that sisal fiber-NR composite is a potential material for light weight products. Alkalization showed insignificant effect on the density of the composites.

Table 1: NR formulations.

Materials	Formulation [phr]				
	Gum	UT	AT	$CaCO_3$	CB
NR	100	100	100	100	100
Sulfur	2.5	2.5	2.5	2.5	2.5
ZnO	5.0	5.0	5.0	5.0	5.0
Stearic acid	1.5	1.5	1.5	1.5	1.5
CBS	0.5	0.5	0.5	0.5	0.5
Sisal fiber	-	20	-	-	-
NaOH treated sisal fiber	-	-	20	-	-
Calcium carbonate	-	-	-	20	-
Carbon black	-	-	-	-	20

Table 2: Cure characteristic of NR composites.

Formulation	MH [lbf-in]	ML [lbf-in]	Time [min]	
			t_{s1}	t_{90}
Gum	13.54	2.76	4.07	8.23
UT	18.96	3.71	3.20	5.36
AT	20.33	3.87	3.30	5.45
$CaCO_3$	23.85	4.06	2.98	6.28
CB	34.75	4.18	2.57	6.34

Table 3: Density of NR composites.

Formulation	Density [g/cm^3]
Gum	0.906
UT	0.910
AT	0.912
$CaCO_3$	1.056
CB	1.089

Tensile properties. Tensile properties of the composites are shown in Fig. 1-4. Specific modulus (strength) is a ratio between the modulus (strength) and the density of composites. Modulus at 100% strain (M100), 300% strain (M300), and tensile strength increased with the addition of fillers into NR as shown in Fig. 1, 2, and 3, respectively. This was attributed to the improvement of stiffness of NR composites by fillers [12]. Among all filler filled NR, $CaCO_3$ filled NR showed the lowest modulus and tensile strength. This may be due to poor dispersion of $CaCO_3$ in rubber matrix. In case of sisal fiber-NR composites, it can be seen that both the composites with untreated sisal fiber and alkali treated sisal fiber had lower tensile strength than the composite with carbon black. This may be attributed to the larger particle size (lower surface area) of sisal fiber [13]. The composite with alkali treated fiber provided higher modulus and tensile strength than the composite with untreated fiber. This may be due to an enhancement in fiber-NR bonding through the mechanical interlocking mechanism [10]. According to Lopattananon et al. [4], the NR composite with alkali treated pineapple leaf fiber gave better modulus and tensile strength than untreated pineapple leaf fiber composite. This indicated an increase in effective surface area and roughness of the pineapple leaf fiber, which increased the interfacial bond strength through mechanical interlocking between the fiber and NR. Egwaikhide et al. [11] found that the improvement in mechanical properties of filled vulcanizates can be obtained by increasing surface area and surface activity of filler, filler dispersion and filler-rubber interaction. The composite filled with alkali treated sisal fiber gave better specific strength and specific modulus when compared with the composites filled with carbon black and $CaCO_3$. This suggested that alkali treated sisal fiber-NR composite would be beneficial for applications requiring weight reduction. The elongation at break of the composites decreased with the addition of fillers as shown in Fig. 4. Sisal fiber-NR composite showed higher elongation at break than carbon black filled NR. This was due to the fiber-rubber matrix adherence resulting from fiber-rubber interactions, which led to an increase in the strain under stress. Osabohien and Egboh [14] also observed the similar result with hemp fiber filled NR and carbon black filled NR.

Hardness. Adding fillers into the rubber compound enhanced hardness due to the reinforcing effect of filler as shown in Fig. 5. NR filled with sisal fiber gave higher hardness than that of NR filled with conventional reinforcing fillers (carbon black and $CaCO_3$). Osabohien and Egboh [14] reported that the NR composite with hemp fiber gave superior hardness level than the NR composite with carbon black at equal filler content. Alkalization can improve the adhesion between the fiber and rubber; thus, enhancing the hardness of the NR composites. Mathew and Joseph [10] reported similar observation in isora fiber-NR composites.

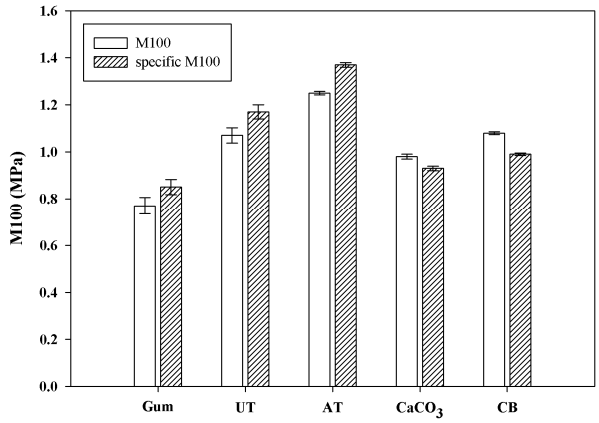

Fig. 1 Variation of modulus at 100% elongation with formulation.

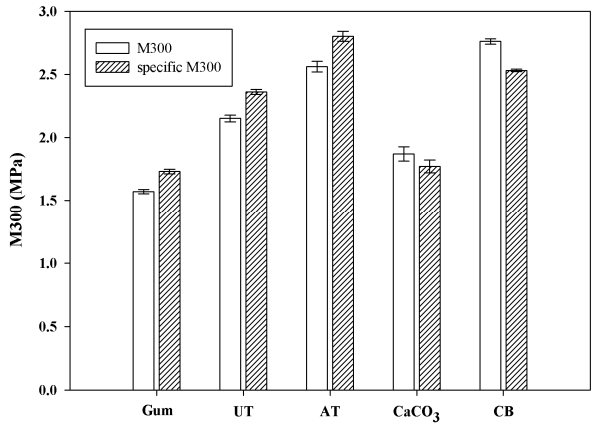

Fig. 2 Variation of modulus at 300% elongation with formulation.

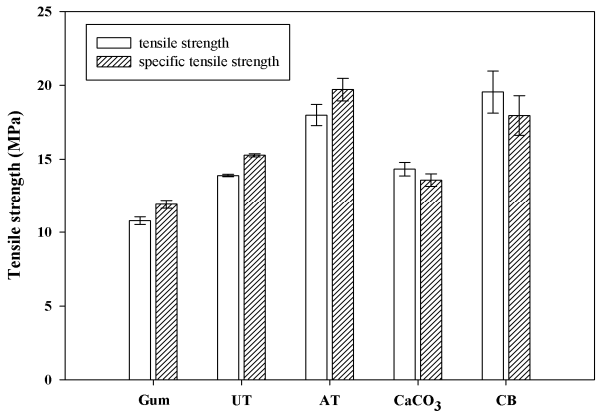

Fig. 3 Variation of tensile strength with formulation.

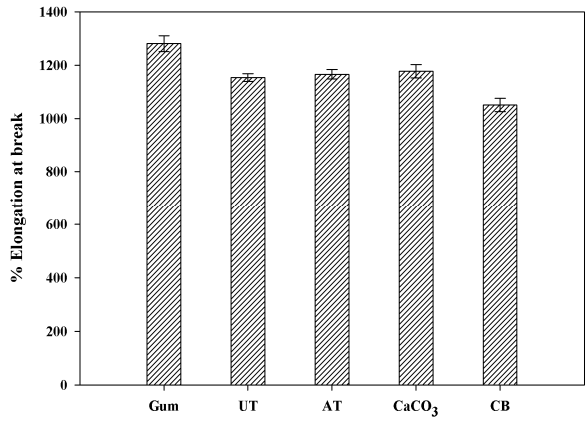

Fig. 4 Variation of % elongation at break with formulation.

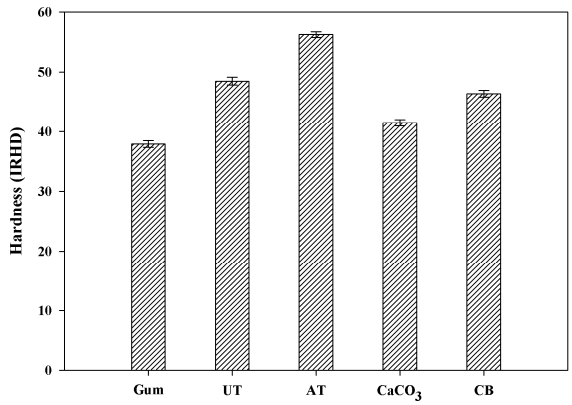

Fig. 5 Variation of hardness with formulation.

Conclusions

The addition of 20 phr filler to the rubber matrix increased maximum torque as well as mechanical properties such as modulus, tensile strength, and hardness compared to the unfilled NR. Scorch time and cure time of the NR composites decreased with the addition of fillers. Sisal fiber-NR composite showed good processing safety in terms of scorch time and torque as compared to $CaCO_3$-NR and carbon black-NR composites. Alkalization gave a positive impact on the tensile properties and hardness of the sisal fiber-NR composite. The sisal fiber filled NR had lower density than $CaCO_3$

and carbon black filled NR. Specific tensile strength and specific modulus of alkali treated sisal fiber filled NR were higher than those of NR filled with conventional fillers. Since alkali treated sisal fiber-NR composite provided high strength as well as low density, they could be useful in light weight products with high strength.

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