

# Application of Rice Husk Ash as Fillers in the Natural Rubber Industry

Wanvimon Arayapranee,<sup>1</sup> Nuchanat Na-Ranong,<sup>2</sup> Garry L. Rempel<sup>3</sup>

<sup>1</sup>Department of Chemical Engineering, Rangsit University, Phatum Thani, 12000, Thailand

<sup>2</sup>Post-harvest and Products Processing Research and Development Office, Department of Agriculture, Lad-yao Jatujak, Bangkok 10900, Thailand

<sup>3</sup>Department of Chemical Engineering, University of Waterloo, Ontario N2L 3G1, Canada

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**ABSTRACT:** Rice husk ash (RHA) obtained from agricultural waste, by using rice husk as a power source, is mainly composed of silica and carbon black. A two-stage conventional mixing procedure was used to incorporate rice husk ash into natural rubber. For comparison purposes, two commercial reinforcing fillers, silica and carbon black, were also used. The effect of these fillers on cure characteristics and mechanical properties of natural rubber materials at various loadings, ranging from 0 to 40 phr, was investigated. The results indicated that RHA filler resulted in lower Mooney viscosity and shorter cure time of the natural rubber materials. The incorporation of RHA into natural rubber improved hardness but decreased tensile strength and tear strength. Other properties, such as Young's modulus and

abrasion loss, show no significant change. However, RHA is characterized by a better resilience property than that of silica and carbon black. Scanning electron micrographs revealed that the dispersion of RHA filler in the rubber matrix is discontinuous, which in turn generates a weak structure compared with that of carbon black and silica. Overall results indicate that RHA can be used as a cheaper filler for natural rubber materials where improved mechanical properties are not critical. © 2005 Wiley Periodicals, Inc. *J Appl Polym Sci* 98: 34–41, 2005

**Key words:** rice husk ash (RHA); fillers; cure characteristics; mechanical properties; rubber

## INTRODUCTION

Rice husk, usually regarded as agricultural waste, can be used as a combustion source for electrical power plants, from which rice husk ash (RHA) is obtained. Rice husk ash consists primarily of amorphous silica and residual carbon black from incomplete combustion. The amount of silica and carbon black in the ash varies depending on the combustion conditions. Rubbers are rarely used in their unmodified form with respect to their applications: they are often mixed with fillers to improve their processability and mechanical strength and to reduce cost. Because carbon black and silica are known as commercial fillers,<sup>1–13</sup> the use of RHA as a filler in rubber compounding has attracted increasing interest because of its low cost, environmental preservation benefit, and an increased emphasis on the use of renewable resources. Many attempts have been made to use RHA as a filler for polymeric materials. Sae-oui et al.<sup>14</sup> investigated the effects of filler loading on the properties of RHA-filled natural rubber (NR) materials compared with those of com-

mercial fillers. They found that both grades of RHA, low- and high-carbon contents, provided inferior mechanical properties (tensile strength, modulus, hardness, abrasion resistance, and tear strength) compared with those of reinforcing filler such as silica and carbon black. Costa et al.<sup>15,16</sup> reported that milled husk ash (MHA)-filled vulcanizates with 20 phr, gave the best results, providing physical properties slightly inferior to those of commercial carbon black- or silica-filled compounds, except for abrasion resistance. For natural rubber compositions with treated husk ash, obtained by means of chemical treatment with NaOH and HCl, the value for tensile strength, considering the composition with 20 phr of filler, was slightly superior compared with that of commercial fillers. Fuad et al.<sup>17</sup> reported that incorporation of RHA into polypropylene led to an increased flexural modulus of the composites, whereas tensile strength, elongation at break, and Izod impact strength showed a decrease. Ismail et al.<sup>18–20</sup> studied the effect of dynamic vulcanization and influence of filler loading of white rice husk ash (WRHA)-filled ethylene-propylene-diene terpolymer/polypropylene blends. They reported that WRHA-filled blends could also be dynamically vulcanized to achieve enhanced properties in a manner similar to that of the unfilled blends. The influences of a compatibilizer [poly(propylene-ethylene acrylic

Correspondence to: G. Rempel (grempe@cpe.uwaterloo.ca).

**TABLE I**  
Physical Properties of the Rice Husk Ash,  
Silica, and Carbon Black

Filler	Mean particle size ( $\mu\text{m}$ )	Surface area ( $\text{m}^2/\text{g}$ )
RHA	36.6	68
Silica	18	165
Carbon black	20.2	84

acid)] and a silane coupling agent [3-aminopropyl-trithoxy silane (3-APF)] on mechanical properties of WRHA-filled polypropylene/natural rubber blends was also investigated.<sup>21</sup>

The mechanical properties of filled rubbers are usually described in terms of tensile and tear strength, Young's modulus, hardness, resilience, and abrasive loss. The mechanical properties of filled rubbers depend mainly on the type of filler, filler loading, filler dispersion, filler particle size, and the interaction between the filler and the rubber matrix. However, no serious attempt has been made to evaluate the use of RHA as a reinforcing filler for natural rubber. Because rice husk is readily available at extremely low cost as an unwanted by-product of rice mills, the finding of useful applications for the rice husk ash will certainly help to alleviate the problems related to the disposal of the waste husks. In this study, RHA was incorporated into natural rubber and the effect of filler loading compared to that of commercial fillers, silica, and carbon black was investigated by examining processability and mechanical properties as performance indicators.

## EXPERIMENTAL

All mixing ingredients were used as received. Natural rubber (STR 5 L) was supplied by Siamese Rubber Co., Ltd. (Kranthuban, Thailand). RHA was obtained from Biomass Power Co., Ltd. (Thailand). Hi-Sil 233s silica ( $\text{SiO}_2$ ) was manufactured by PPG-Siam Silica Co., Ltd. (Thailand) and N330 black (carbon black) was supplied by Thai Carbon Product Co., Ltd. (Bangkok, Thailand). Stearic acid, zinc oxide ( $\text{ZnO}$ ), and sulfur (S) were obtained from Imperial Corp., Ltd. (Thailand), Uni Thai Oxide Co. Ltd., and Siamchemie Co. Ltd., respectively. *N*-Cyclohexyl-2-benzothiazole-2-sulfenamide (CBS) was supplied by Flexsys Co. Ltd. (Bangkok, Thailand). Poly(ethylene glycol) (PEG) was supplied by Brand Co., Ltd. (Thailand).

Rice husk ash (RHA) was sieved on a 325-mesh sieve. The chemical composition was analyzed by X-ray fluorescence spectrometry. The particle size distribution was measured by using a particle size analyzer (Mastersizer-S, Malvern Instruments, Malvern, UK). The surface area of the fillers was determined by the

**TABLE II**  
Chemical Composition of Rice Husk Ash

Component	%
Loss on ignition at 800°C	4.00
Moisture content	4.98
Silica ( $\text{SiO}_2$ )	92.1
Alumina ( $\text{Al}_2\text{O}_3$ )	0.51
Iron oxide ( $\text{Fe}_2\text{O}_3$ )	0.40
Calcium oxide ( $\text{CaO}$ )	0.55
Magnesium oxide ( $\text{MgO}$ )	nd
Sodium oxide ( $\text{Na}_2\text{O}$ )	nd
Potassium oxide ( $\text{K}_2\text{O}$ )	1.53
Titanium dioxide ( $\text{TiO}_2$ )	0.02
Manganese oxide ( $\text{MnO}$ )	0.08
Phosphorus pentoxide ( $\text{P}_2\text{O}_5$ )	0.36
Sulfur trioxide ( $\text{SO}_3$ )	0.12

Brunauer–Emmett–Teller (BET) method on the BET ASAP 2000 accelerated surface area and porosimetry system. The physical and chemical properties of RHA, silica, and carbon black are presented in Tables I and II.

## Preparation of rubber compounds

Mixing was done in an internal mixer using conventional mixing procedures by two stages: in the first stage, the mixing was carried out in a dispersion kneader machine with a fill factor of 0.7, at a chamber temperature of 75°C and a rotor speed of 40 rpm. Natural rubber was initially masticated in the mixer for 3 min, followed by addition of the activators (zinc oxide, stearic acid), filler, and accelerators (CBS). In the second stage, complete vulcanized compounds were prepared by the addition of sulfur on a laboratory-sized two-roll mill at 70°C for 5 min. Table III shows the compound formulation used in this study.

## Cure characteristics

Cure characteristics were studied using a rheometer (TECH-PRO, Cuyahoga Falls, OH) according to ISO 3414 for 30 min at 150°C. The Mooney viscosity

**TABLE III**  
Formulation for Natural Rubber Material

Ingredient	phr <sup>a</sup>
NR (STR 5L)	100
Filler	Variable (0–40)
ZnO	5.0
Stearic acid	2.0
CBS	1.8
S	2.4
PEG <sup>b</sup>	2.0

<sup>a</sup> phr, parts per hundred of rubber.

<sup>b</sup> Used in batches having silica as a filler.

( $ML_{1+4}$  at  $100^\circ\text{C}$ ) was determined by using a Mooney viscometer (TECH-PRO). The testing procedure was conducted according to the method described in ISO 289-1.

### Vulcanization process

All compounds were compression molded at  $150^\circ\text{C}$  with force of 17.5 MPa using a hydraulic hot press according to their respective cure time,  $t_{90}$ , determined with the TECH-PRO. Vulcanizates were conditioned for 24 h before testing.

### Mechanical properties

Tensile properties were determined on an Instron UTM (Model 1011) using C-type dumbbell-specimen, according to ASTM D 412. The tear strength was measured using a Lloyd Instruments (Hampshire, UK) apparatus according to ASTM D 624 (die c). Hardness measurement of samples was done according to ASTM D 2240 (Shore A) using a Lever loader (Model 917). Resilience was studied using a Wallace Dunlop (Kingston, UK) tripsometer according to B.S. 903 Part A8. An abrasion test was carried out according to B.S. 903 Part A9 on a Wallace test apparatus.

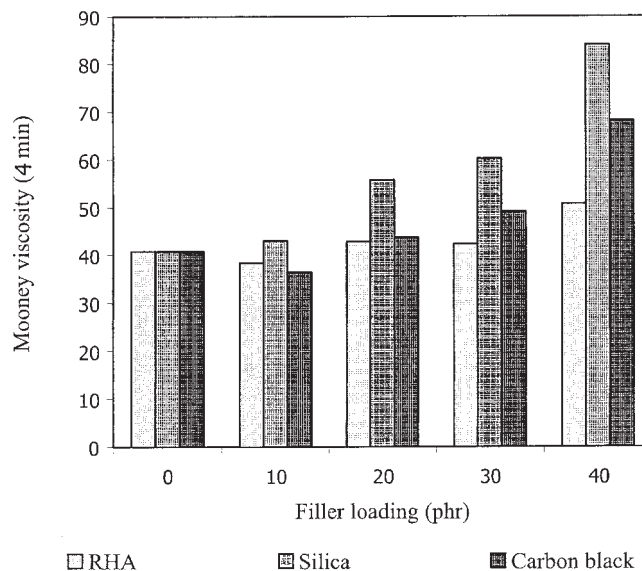
### Scanning electron microscopy (SEM)

SEM studies of the compounds' tensile fracture surfaces were carried out on gold-coated samples in a JSM 5600 LV microscope (JEOL, Tokyo, Japan) at a magnification of  $\times 2000$ . The objective was to gather information regarding filler (RHA, silica, and carbon black) dispersion and interfacial-bonding quality between filler and the rubber matrix.

## RESULTS AND DISCUSSION

### Processability and cure characteristics of natural rubber compounds

Various fillers were incorporated, with loading ranging from 0 to 40 phr (parts per hundred of rubber), in compounds to study the effect of filler loading on Mooney viscosity of natural rubber materials, as shown in Figure 1. It can be seen that for silica and carbon black, the Mooney viscosity substantially increases with increasing filler loading. For RHA, however, the change of this property with filler loading is negligible. Silica-filled natural rubber materials have the highest Mooney viscosity followed by carbon black and RHA-filled natural rubber materials. In the mixing state, large particle size (Table I) and low interaction between the fillers and rubber give rise to lower viscosity, compared to that of rubbers filled with reinforcing fillers. The presence of reinforcing



**Figure 1** Relationship between filler loading and Mooney viscosity ( $ML_{1+4}$  at  $100^\circ\text{C}$ ) of natural rubber materials filled with various fillers.

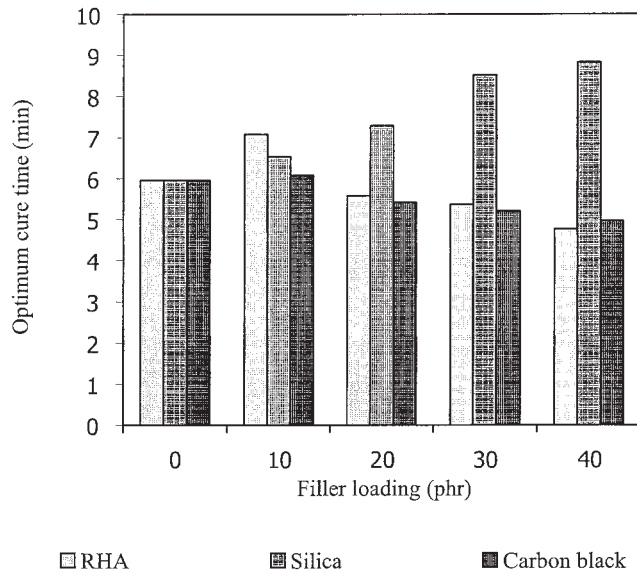
fillers in the rubber matrix reduces the mobility of the rubber's macromolecular chains. The high values of viscosity for silica- and carbon black-filled natural rubber materials indicate that there is a high restriction to the molecular motion of the macromolecules, probably caused by the greater interaction between both fillers and the rubber matrix. The lowest Mooney viscosity of RHA indicated that it could be processed more easily than silica- and carbon black-filled natural rubber compounds. Similar results were also reported by Sae-oui et al.<sup>14</sup>

The rubber compounds were vulcanized for their optimum cure time. Figure 2 represents the optimum cure time ( $t_{90}$ ) of various fillers. Increased loading of carbon black and RHA slightly decreased the optimum curing time. However, at a similar filler loading, carbon black-filled natural rubber materials exhibited the shortest  $t_{90}$  followed by RHA-filled natural rubber materials. For carbon black the reduction of  $t_{90}$  is attributed to the role of carbon black, which accelerates the vulcanization process. With silica, there is a different trend in cure characteristics: the curing time tends to increase with increasing silica content. This is commonly explained by the silica structure, which can absorb some accelerators into the porous structure, thus slowing down the vulcanization process. The cure characteristics achieved here are similar to those reported by other workers who carried out similar studies.<sup>14-16</sup>

### Mechanical properties of natural rubber compounds

Various fillers were incorporated in different ratios up to 40 phr in natural rubber to study the effect of filler

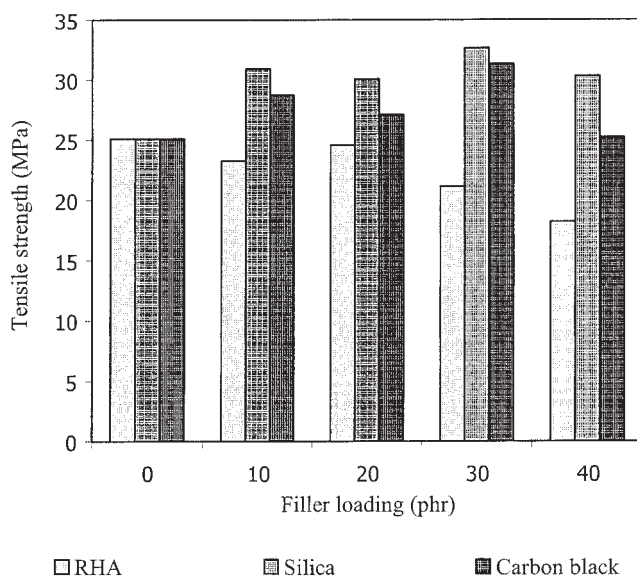




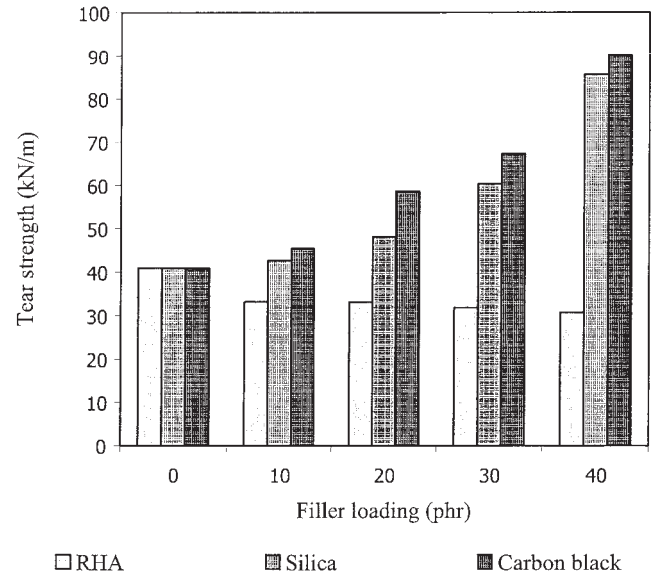
**Figure 2** Relationship between filler loading and optimum cure time of natural rubber materials filled with various fillers.

loading on the mechanical properties of the filled natural rubber. To facilitate understanding of such behavior, the effect of various fillers on each mechanical property will be discussed separately.

Figure 3 shows the effect of filler loading on tensile strength of natural rubber, which may increase or decrease with the incorporation of filler. It can be seen that, at a similar filler loading, silica-filled natural rubber gave the highest tensile strength, followed by carbon black-filled and then RHA-filled natural rubber. Both silica- and carbon black-filled natural rubber



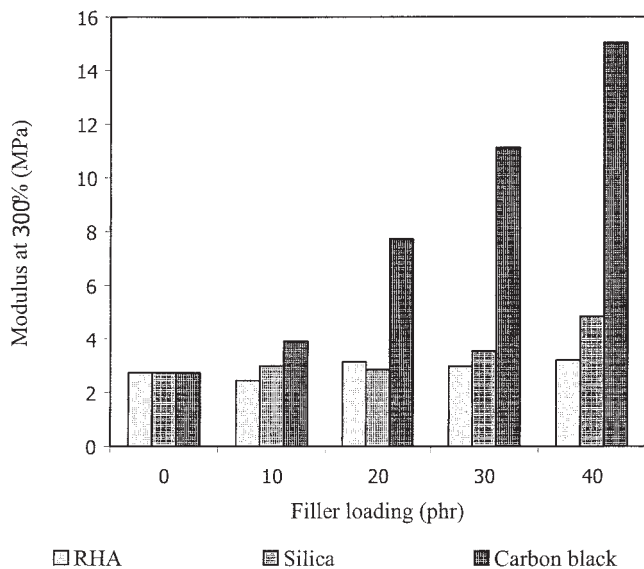
**Figure 3** Effect of filler loading on tensile strength of RHA, silica, and carbon black-filled natural rubber materials.



**Figure 4** Effect of filler loading on tear strength of RHA, silica, and carbon black-filled natural rubber materials.

compounds exhibited similar trends; the tensile strength of the filled natural rubber increases with filler loading until a maximum level is reached (at  $\sim 30$  phr) and then the property starts to decrease with increasing filler loading. Rigbi<sup>22</sup> reported that the reinforcing filler increases the tensile strength up to maximum filler loading of elastomer, after which the detrimental effect on tensile strength at high loadings may be attributed to the dilution effect or agglomeration of filler. On the other hand, a negative effect on the tensile strength was observed for RHA-filled natural rubber. Tensile strength of the filled natural rubber decreases with an increase in RHA loading because of the inability of filler to support stresses transferred from the polymer matrix. RHA-filled natural rubber materials exhibited the highest tensile strength, with the loading being 20 phr RHA compared with the other loadings investigated, as shown in Figure 3, considering the best formulation (20 phr) and lower strength values compared with those of unfilled natural rubber materials. It is believed that a smaller particle size (Table I) and uniform dispersion of both silica and carbon black in natural rubber materials contribute to better tensile strength. In addition, the nature of the RHA filler, given by the presence of both silica and carbon components, with different physical and chemical properties (Tables I and II), may also reduce the efficiency of this filler in strengthening the rubber matrix. Ismail and Mega<sup>21</sup> observed a similar trend in strength from their work on WRHA-filled polypropylene/natural rubber blend.

Figure 4 shows the tear strength of the filled rubber materials. Tear strength, like tensile strength, is affected by filler particle size and surface area. As can be



**Figure 5** Effect of filler loading on modulus values at 300% of RHA, silica, and carbon black-filled natural rubber materials.

seen, the addition of silica or carbon black substantially increases the tear strength of the rubber materials. This is mainly attributed to the reduction of stress concentration at the crack tips in the presence of fillers, particularly the fine-particle ones. In contrast, the addition of the RHA fillers slightly reduces the tear strength of the rubber materials. This may be attributed not only to their large particle size, but also to their low rubber-filler interaction. For this reason, both tensile and tear strengths are decreased with increased loading of the ashes.

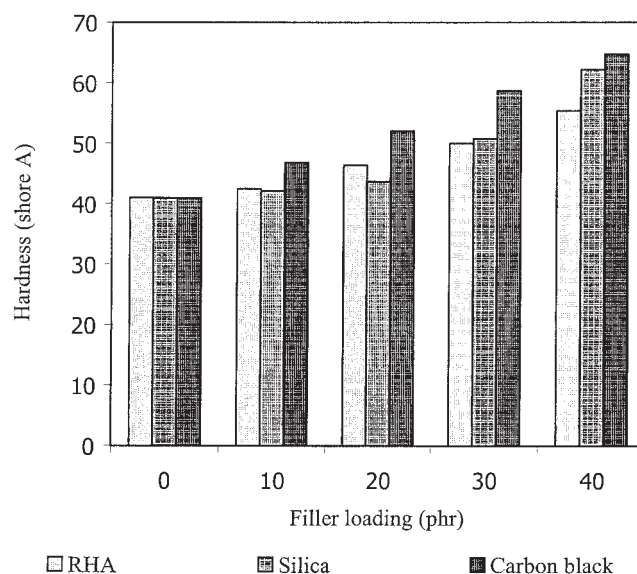
Modulus is an indication of the relative stiffness of the material. Figure 5 shows the effect of filler loading on modulus at 300% elongation of the rubber materials. It can be seen that carbon black improves the stiffness of natural rubber materials, whereas silica and RHA show a lesser effect. The sharp increasing trend for carbon black may be attributed to the good rubber-filler interaction of this filler. As more filler particles are introduced into the rubber, the elasticity of the rubber chains is reduced, resulting in higher stiffness properties. It can be observed that, at any particular loading, the silica-filled compound has higher tensile strength, whereas its modulus is substantially lower than that of carbon black-filled rubber materials. Sae-oui et al.<sup>14</sup> also reported that surface area is the most important factor controlling the tensile strength, whereas the surface activity (rubber-filler interaction) controls the modulus (i.e., natural rubber forms stronger adsorptive bonds with carbon black than with silica).

Figure 6 shows the effect of filler loading on hardness of rubber materials, which exhibit similar trends; hardness of filled rubber materials increases with filler

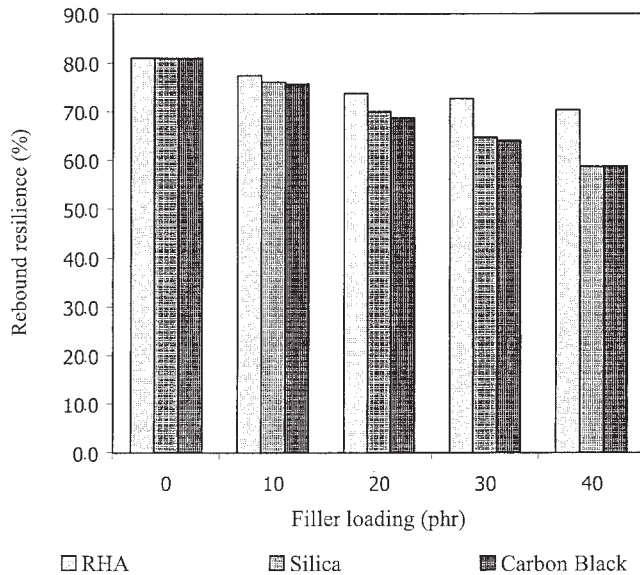
loading. This is simply explained by the fact that incorporation of more rigid filler particles into rubber reduces elasticity of the rubber chains, leading to more rigid rubber materials. As expected, silica- and RHA-filled rubber materials have lower modulus and hardness than do the corresponding rubber materials with carbon black. This may be caused by differences in filler properties. The effect of filler loading on hardness is even more pronounced in systems filled with carbon black, giving a harder compound than silica because of its stronger adsorptive bonds. Costa et al.<sup>15,16</sup> also reported similar filler-type influences on Young's modulus and hardness.

The effect of various filler loadings on resilience of the natural rubber materials is shown in Figure 7. It can be seen that for silica and carbon black, the resilience of the natural rubber materials drastically decreases with increasing filler loading. For RHA, however, the reduction of this property with filler loading is negligible. This observation may be attributed to poorer rubber-filler interaction of RHA. On the contrary, the sharp decreasing trend for silica and carbon black may be attributed to better rubber-filler interaction of these fillers. As more filler particles are introduced into the rubber, the elasticity of the rubber chains is reduced, resulting in lower resilience properties. The surface activity is an important factor, indicating the extent of rubber-filler interaction. According to Jacques<sup>23</sup> the incorporation of most of the particulate fillers into rubber leads to increase in hardness and a reduction in resilience, particularly with more reinforcing filler.

The incorporation of silica and carbon black substantially reduces the abrasion loss of the natural rubber materials, whereas RHA shows no effect with filler



**Figure 6** Effect of filler loading on hardness of RHA, silica, and carbon black-filled natural rubber materials.



**Figure 7** Effect of filler loading on resilience of RHA, silica, and carbon black-filled natural rubber materials.

loading, as shown in Figure 8. With respect to this property, the ashes confer a similar effect to nonreinforcement fillers. The abrasion loss shows no significant change with increase in loading. An inverse effect is found in the systems filled with silica and carbon black, in which the abrasion loss is found to decrease sharply with an increase of the filler loading up 20 phr after which the abrasion loss decreases only marginally. Reinforcing fillers, silica and carbon black, interact preferentially with the natural rubber phase, as shown by the higher reduction of abrasion loss in the compounds. This improvement is probably explained by the greater surface area and better filler-rubber interfacial adhesion resulting in an improved abrasion resistance. Fine particles actually reflect their greater interface between the filler and the rubber matrix, thus providing a better abrasion resistance than that of the coarse particles. Similar results were also reported by Sae-oui et al.<sup>14</sup>

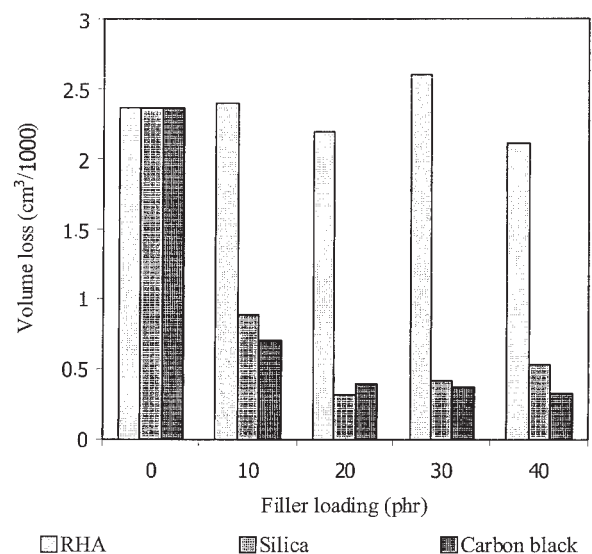
### Morphology

The morphology of natural rubber and the unfilled and filler-filled natural rubber compounds at 20 phr filler loading after tensile fracture is shown in Figure 9. The surface of the unfilled natural rubber materials is smooth [Fig. 9(a)]. For the fracture surface of RHA-filled natural rubber materials [Fig. 9(b)], the dispersion of RHA in the rubber matrix was not continuous where the formation of filler agglomeration started as a result of the interfacial interaction between filler and matrix, which led to void formation. The presence of many holes on the fracture surface may be explained by the weak interfacial interaction resulting in the

deterioration of compound properties such as tensile strength when compared with unfilled natural rubber materials. In the case of the incorporation of WRHA into polypropylene/natural rubber blend, Ismail and Mega<sup>21</sup> reported that agglomeration of the WRHA within the blend at the interphase aggravates the situation, which accounts for the reduction in tensile strength. Silica- and carbon black-filled natural rubber compounds are shown in Figure 9(c) and (d), respectively; it can be seen that the fracture surface of reinforcing filler shows a more homogeneous phase dispersion, justifying its higher tensile strength. Figure 9(c) shows a SEM photomicrograph of the fracture surface of silica-filled natural rubber materials. The rough surface with tear lines is a typical feature of ductile failure, which indicates the higher tensile strength of the silica-filled natural rubber materials. From the fracture surface in Figure 9(d) [compare to Fig. 9(c)], the incorporation of carbon black does not alter the ductile mode of failure of the materials, which resulted in brittle failure and thus the enhancement in the stiffness. The SEM photomicrographs shown in Figure 9 confirm that the smaller particle size of the filler provides a larger surface area for a better filler dispersion and interfacial bond between filler and matrix.

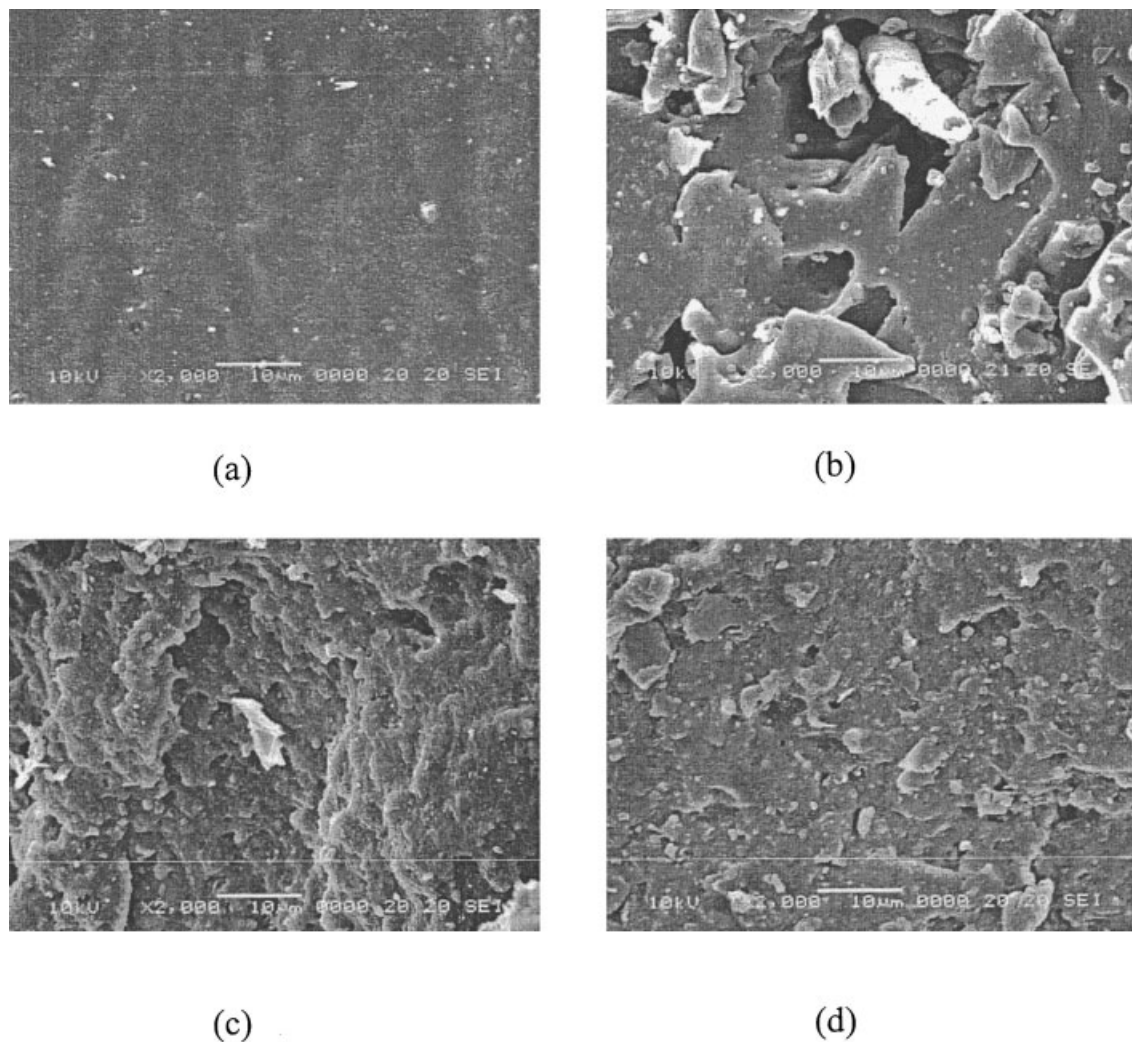
### CONCLUSIONS

The purpose of this study was to verify the possibility of using rice husk ash (RHA) as an alternative filler for natural rubber. The incorporation of fillers in natural rubber compounds decreases the cure time ( $t_{90}$ ) with increasing RHA and carbon black loading in natural



**Figure 8** Effect of filler loading on abrasion resistance of RHA, silica, and carbon black-filled natural rubber materials.





**Figure 9** SEM micrographs of (a) unfilled natural rubber material and (b) RHA, (c) silica, and (d) carbon black-filled natural rubber materials after tensile fracture at 20 phr filler loading ( $\times 2000$ ).

rubber compounds, whereas silica shows a different trend in cure time, tending to increase with increasing silica loading. At a similar filler loading, carbon black shows the shortest  $t_{90}$  followed by RHA and silica. Mooney viscosity increases with increase in silica and carbon black loading, whereas RHA shows only a negligible change in this property. At a similar filler loading, RHA shows the lowest viscosity followed by carbon black and silica. Filler loading and filler type influence the processability of the rubber materials in RHA, offering a better processing advantage over silica and carbon black. Changes (positive or negative) in tensile and tear strength, Young's modulus, hardness, resilience, and abrasion loss are quite significant with increasing loading of silica and carbon black in natural rubber materials. Tensile and tear strengths of the compounds decline slightly with increasing RHA loading, whereas the hardness of the compounds increases slightly. Young's modulus and abrasion loss show no significant changes. Silica and carbon black

provide better mechanical properties than the RHA, mainly because of their high surface area (small particle size) and surface activity. Scanning electron microscopy on fracture surface shows poor filler dispersion and weak filler-matrix interaction with the RHA-filled natural rubber materials. This explains the reduction of tensile and tear strength with RHA-filled natural rubber materials. Although, from overall mechanical properties, it could be suggested that RHA has a poorer reinforcing ability than that of silica and carbon black, one may still consider the use of RHA as a filler in the rubber industry for economic and ecological reasons. Suitable application for the RHA compound could be in components demanding high resilience and excellent dimension stability without significantly affecting the properties of interest in the compounds.

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