

A Comparison of the Properties of Rice Husk Ash, Silica, and Calcium Carbonate Filled 75 : 25 NR/EPDM Blends

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ABSTRACT: The effects of incorporation of three different fillers, i.e., rice husk ash (RHA), silica, and calcium carbonate (CaCO_3), over a loading range of 0–60 phr on the curing characteristics, processability, mechanical properties, and morphology of 75 : 25 natural rubber (NR)/ethylene-propylene-diene monomer (EPDM) blends were studied using a conventional vulcanization system. Filler loading and type influence the processability of the blends in which RHA and CaCO_3 offer better processing advantage over silica. The best improvement in the tensile and tear strength and abrasion resistance of the 75 : 25 NR/EPDM blends with additional fillers was achieved when filled with silica. However, RHA and CaCO_3 were better in resilience prop-

erty compared to that of silica. The RHA filled blends showed higher failure properties and abrasion resistance but lower ozone resistance than that containing CaCO_3 . Scanning electron micrographs revealed that the morphology of the blend filled with silica is finer and more homogeneous compared to the blend filled with RHA and CaCO_3 . According to these observations, RHA can be used as a cheaper filler to replace CaCO_3 in rubber blends where improved mechanical properties are not so critical. © 2008 Wiley Periodicals, Inc. *J Appl Polym Sci* 110: 1165–1174, 2008

Key words: natural rubber; EPDM; blend; rice husk ash; silica; calcium carbonate; filler

INTRODUCTION

Natural rubber (NR) is a natural biosynthesis polymer having an attractive range of properties, possessing excellent mechanical properties, and good processing characteristics. However, NR is highly susceptible to degradation because of the presence of double bonds in the main chain. Degradation of NR is accelerated mainly by heat, humidity, light, ozone, radiation, etc. In general, improvement in the poor ozone resistance of NR can be achieved by blending it with low unsaturated rubbers such as ethylene-propylene-diene rubber (EPDM) obtained by polymerizing ethylene and propylene with a small amount of a nonconjugated diene, which imparts usually good ageing characteristics, good resistance to weathering oxidation, and chemical resistance.¹ Blending with EPDM has been reported to be effective in improving the ozone resistance of NR.²

The influence of fillers is important because the elastomers are seldom used without fillers, and in some cases, blends of two or more elastomers are preferred to achieve the desired properties. When

NR is blended with EPDM, it is often found necessary to modify it with fillers to improve the modulus, failure properties (tensile and tear strength), and abrasion resistance and reduce unit manufacturing costs while maintaining a high level of product quality and performance. Many studies on the mechanical properties of various rubber compounds have been reported, including the effect of the type and the amount of rubber and filler, and the interaction between rubbers and fillers.^{3–11} Filler dispersion also plays an important role in reinforcement. The effect of fillers on mechanical and other properties of the blends depends strongly on their shape, size and size distribution of the primary particles and their aggregates, surface characteristics, and degree of dispersion and distribution.¹² A variety of black and nonblack fillers of the reinforcing and nonreinforcing types cater to this need. One of the most important fillers, which is added to rubber, is calcium carbonate (CaCO_3). Much work^{13–15} has been done to study the use of CaCO_3 as a filler for polymeric materials. It not only lowers the cost but also improves mechanical properties such as modulus, hardness, toughness, dimensional stability, thermal resistance, and flame retardancy. Among several fillers, silica is the most important reinforcing agent used in compounding recipes. In spite of being well known for its strong interaction with rubbers, this filler is relatively expensive when compared with natural fillers.

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TABLE I
Ingredients Used in the Present Study

Ingredient	Amount (phr) ^a	Grade/supplier
NR	75	STR XL/Thailand
EPDM	25	DuPont's Nordel [®] IP 4570/DuPont Dow Elastomers L.L.C./Wilmington, DE, USA
Paraffin oil	4	Flexon 845/Exxon, Thailand
Zinc oxide (ZnO)	2.0	Commercial/Gradmann, Thailand
Stearic acid	2.0	Commercial/P. T. Cisadaneraya Chemical, Indonesia
Tetramethylthiuram bisulfide (TMTD)	0.15	Flexsys, Germany
Mercaptobenzothiazole disulfide (MBTS)	1	Flexsys, Belgium
Sulfur (S)	2	Commercial/Chemmin, Thailand
Filler		
CaCO ₃	Variable (0–60)	Quality Minerals Public Company Limited, Thailand
Silica (SiO ₂)	Variable (0–60)	Hi-Sil 233s/PPG-Siam Silica, Thailand
Rice husk ash (RHA)	Variable (0–60)	Ooncharoensap Rice Mill, Thailand

^a phr, parts per hundred of rubber.

The application of fillers derived from agricultural wastes has attracted interest because of their low cost, renewability, and environment friendly nature. Nowadays rice husk ash (RHA) is used in plastic, rubber, and thermoplastic elastomers (TPEs) because of various advantages, such as ease of processing, availability, economic considerations, environmental preservation, and an increased emphasis on the use of renewable resources.^{16–24}

In this study, a NR/EPDM weight ratio corresponding to 75 : 25 has been chosen because at this composition, it is possible to maintain the good mechanical properties of natural rubber while improving the ozone resistance of the rubber material. RHA was introduced into 75 : 25 NR/EPDM blends and the effects of filler loading compared to that of the commercial fillers, silica and CaCO₃, were investigated. This study evaluated the processability, mechanical properties, abrasion and ozone resistance, and morphology studies of the tensile fracture surfaces of filler filled 75 : 25 NR/EPDM blends.

EXPERIMENTAL

Materials

Details of blending ingredients and rubber formulas used are shown in Table I. The particle size distribution was measured by using a particle size analyzer (Mastersizer-S, Malvern Instruments, England). The surface area of the fillers was determined by the Brunauer-Emmett-Teller (BET) method using an ASAP 2000 surface area analyzer (Micromeritics Instrument Corp., Norcross, GA).

When rice husk is burnt about 20 wt % of the husk remains as ash. The majority of the substance in RHA is silicon dioxide (=78.33%) together with an amount of residual carbon (=17.05%) and other oxides such as Al₂O₃, Fe₂O₃, CaO, MgO, Na₂O, K₂O, and P₂O₅ in small amounts (=4.62%). The particles

show an irregular form with a minimum size of 0.49 μm and a maximum size of 45 μm. The average particle size of RHA was 36.6 μm. The physical properties of RHA, silica, and CaCO₃ are presented in Table II.

Preparation of NR/EPDM blends

Formulations of the blends are provided in Table I. NR/EPDM (75 : 25 wt %) blends were prepared in an internal mixer using conventional mixing procedures involving two stages. In the first stage, the blends were prepared in a dispersion kneader machine (Kneader Machinery, Tainan Shiann, Taiwan) with a fill factor of 0.7, at a chamber temperature of 75°C, and a rotor speed of 40 rpm. NR was initially masticated in the mixer for 3 min and blended with the EPDM component, followed by the other ingredients in this order: plasticizer (paraffin oil), activators (ZnO and stearic acid), filler and accelerators (MBTS and TMTD). In the second stage, complete vulcanized compounds were prepared by the addition of sulfur using a laboratory-sized two-roll mill (Kodaira Seisakusho, Tokyo, Japan) at 70°C for 3 min.

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 (ML₁₊₄ at 100°C) was determined by using a

TABLE II
Physical Properties of the RHA, Silica, and CaCO₃

Filler	Mean particle size (μm)	Surface area (m ² /g)
RHA	36.6	17
Silica	18	165
CaCO ₃	5.3	2.8

Mooney viscometer (TECH-PRO, Cuyahoga Falls, OH). The testing procedure was conducted according to the method described in ISO 289-1.

Vulcanization process

A conventional vulcanization system (CV) was used for curing. All blends were compression molded at 150°C with a force of 17.5 MPa using a hydraulic hot press (Power Drive System, Thailand) according to their respective cure time, t_{90} , determined with a TECH-PRO instrument.

Mechanical properties

Tensile properties were determined on an Instron Testing (Model 1011; Norwood, MA) using C-type Dumbbell-specimen, according to ASTM D412. The specimens having the geometry of a dumbbell shape with 2 mm in thickness, 25 mm in width, and 115 mm in gauge length were cut into standard test pieces using a Wallace die cutter. The cross-head speed was set at 500 mm/min. At least six specimens were tested for each blend. Average value and the standard deviation (SD) of the data around the average value were then calculated and reported in each case.

The tear strength is a measure of the resistance of a material to tear forces. The tear strength of a notched specimen is calculated by dividing the maximum load by the thickness of the specimen. The tear strength was measured in a Lloyd Instrument according to ASTM D624 (Die C) using unnotched 90° test pieces. The tear properties were tested at a speed of 500 mm/min. At least five specimens were tested for each blend. The average value and SD of the data around the average value were then calculated and reported in each case.

Five test pieces were fractured and the tearing energies were calculated from eq. (1):

$$T = 2F/t \quad (1)$$

where F is the force and t the thickness of the test piece.

Resilience was studied using a Wallace Dunlop Tripsometer (England) according to B.S. 903 Part A8. Rebound resilience was calculated according to the following equation:

$$\text{Percentage resilience} = \{(1 - \cos\theta)/(1 - \cos\phi)\} \times 100 \quad (2)$$

where ϕ is the initial angle (45°) and θ is the maximum rebound angle. Three specimens were used and the average value and SD of the data around

the average value were then calculated and reported in each case.

An abrasion test was carried out according to B.S. 903 Part A9 using a Wallace Test Equipment (England) and expressed as abrasion loss, which is the volume in cm³ abraded from a test piece per 1000 cycles. The average value of two tests for each sample was carried out to ensure consistency in the results.

Ozone ageing

Ozone ageing studies under static conditions were conducted according to ISO 1431/1-1980 (E) in a Hampden (Northampton, England) Model 703 ozone test chamber at 40°C. Ozone concentration in the chamber was adjusted to 50 parts per hundred million (pphm). The ozonized oxygen generated in the ozonizer by a UV quartz lamp was collected in a separate chamber where the two specimens having 20% strain were exposed.

Photographs were taken using an optical microscope Carl Zeiss Stemi 2000 C (Werk Göttingen, Germany) with a magnification of 50 times.

Scanning electron microscopy

Scanning electron microscopic studies of the compounds' tensile fracture surfaces were carried out on gold-coated samples using a Joel Microscope (model JSM 5600 LV; Tokyo, Japan) at a magnification of 3000.

RESULTS AND DISCUSSION

Cure characteristics and processability of 75 : 25 NR/EPDM blends

Various fillers were incorporated with loading ranging from 0 to 60 phr (parts per hundred of rubber) at 15 phr intervals in 75 : 25 NR/EPDM blends to study the effect of filler loading on the cure properties of the blends as shown in Figure 1. The blends were vulcanized for their optimum cure time. For the optimum cure time, it can be observed that compared to the unfilled blend, an increase in loading of RHA and CaCO₃ decreased the optimum curing time. However, at a similar filler loading, RHA filled 75 : 25 NR/EPDM blends exhibit the shortest t_{90} followed by CaCO₃ and silica filled 75 : 25 NR/EPDM blends. For silica, there is a different trend in cure characteristics. The curing time tends to increase with increasing silica content. The retardation effect found in the silica vulcanizate can be attributed to a silica-accelerator system interaction. This filler reacts with ZnO and subsequently reduces the zinc available, thus slowing down the vulcanization process.

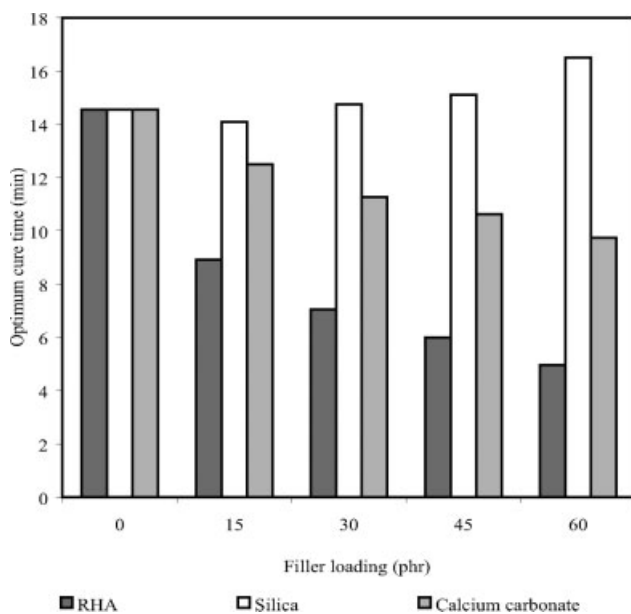


Figure 1 Relationship between filler loading and optimum cure time (ML_{1+4} at $100^{\circ}C$) of 75 : 25 NR/EPDM blends filled with various fillers.

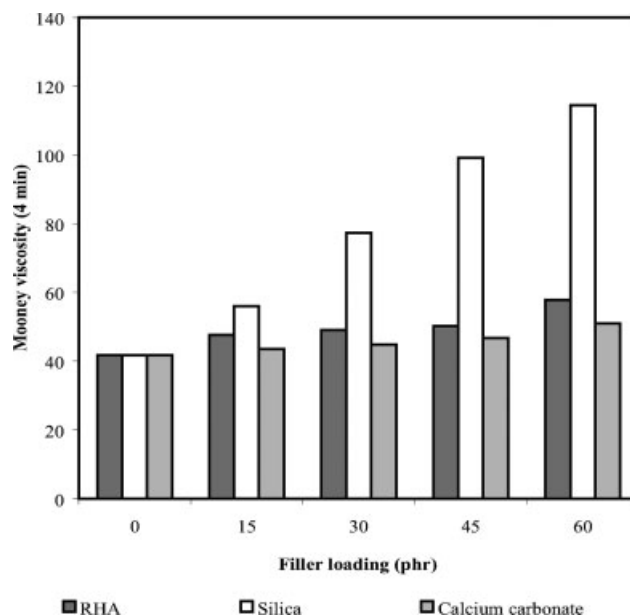


Figure 2 Relationship between filler loading and Mooney viscosity of 75 : 25 NR/EPDM blends filled with various fillers.

Other researchers¹¹ observed a similar trend in other RHA filled natural rubber compounds.

The effect of filler loading on the Mooney viscosity of the blends is shown in Figure 2. It can be seen that for silica, Mooney viscosity increases significantly with an increase in filler loading. However, for RHA and $CaCO_3$, the change of this property with filler loading is relatively small. Silica filled 75 : 25 NR/EPDM blends have the highest Mooney viscosity followed by RHA and $CaCO_3$ filled 75 : 25 NR/EPDM blends. During the mixing state, low interaction between the fillers and rubber gave rise to lower viscosity, as compared to blends filled with reinforcing fillers. Polymer–filler interaction leads to immobilization of chain segments on the filler surface whose mobility is reduced with regard to that of the polymer matrix. The presence of reinforcing fillers in the rubber matrix reduced the mobility of the rubber's macromolecular chains. The lower Mooney viscosity of RHA and $CaCO_3$ indicated that it could be processed more easily than silica filled 75 : 25 NR/EPDM blends.

Mechanical properties of 75 : 25 NR/EPDM blends

Tensile properties

Tensile strength is an important characteristic of polymeric materials because it indicates the limit of final stress in most applications. Various fillers were incorporated at different ratios up to 60 phr in 75 : 25 NR/EPDM blends to study the effect of filler loading on the mechanical properties of the filled

75 : 25 NR/EPDM blends. Figure 3 shows the tensile strength of 75 : 25 NR/EPDM blends as a function of filler loadings. The effect of filler loading on tensile strength of 75 : 25 NR/EPDM blends may increase or decrease with the incorporation of filler. It can be seen that at a similar filler loading, silica filled 75 : 25 NR/EPDM blends gave the highest tensile strength, followed by RHA and then $CaCO_3$ filled 75 : 25 NR/EPDM blends. Tensile strength of

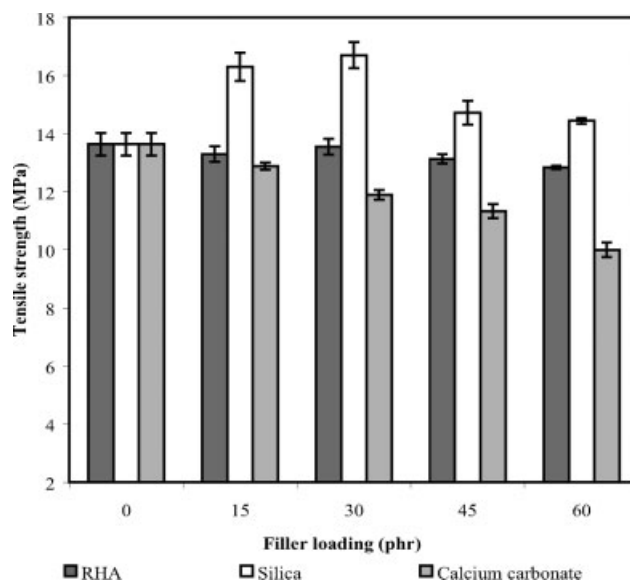


Figure 3 The effect of filler loading on tensile strength of RHA, silica, and $CaCO_3$ filled 75 : 25 NR/EPDM blends.

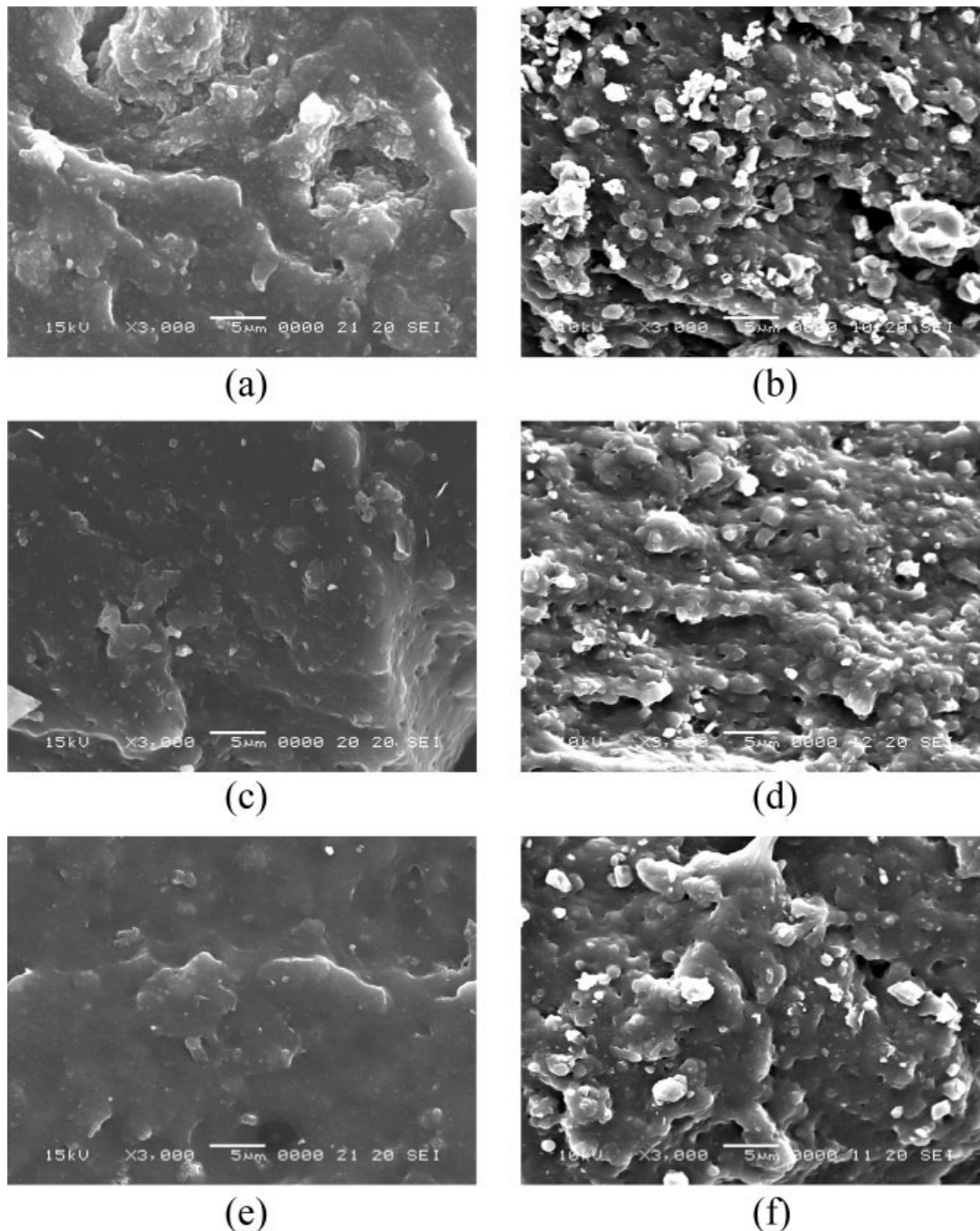


Figure 4 SEM micrographs of 75 : 25 NR/EPDM blends with (a) 15 phr of RHA, (b) 60 phr of RHA, (c) 15 phr of silica, (d) 60 phr of silica, (e) 15 phr of CaCO_3 , and (f) 60 phr of CaCO_3 .

the silica filled 75 : 25 NR/EPDM blends increased with filler loading until a maximum level was reached (at approximately 30 phr) and then the property started to decrease with an increase in filler loading. The detrimental effect on tensile strength at high silica loadings may be attributed to a dilution effect or to agglomeration of the filler, as shown in Figure 4(b). Another researcher²⁵ also reported a similar finding. On the other hand, a negative effect on the tensile strength was observed both for RHA and CaCO_3 filled 75 : 25 NR/EPDM blends exhibit-

ing similar trends; tensile strength of the filled 75 : 25 NR/EPDM blends decreased with an increase in filler loading because of the inability of the filler to support stresses transferred from the rubber matrix. On the other hand, the agglomeration of the filler particles at high loading may form a domain that could act as a foreign particle leading to localized stress concentration during deformation and finally resulting in premature failure of the blends [Fig. 4(d,f)].

Figure 5 shows the effect of filler loading on modulus at 100% elongation of the rubber blends, which

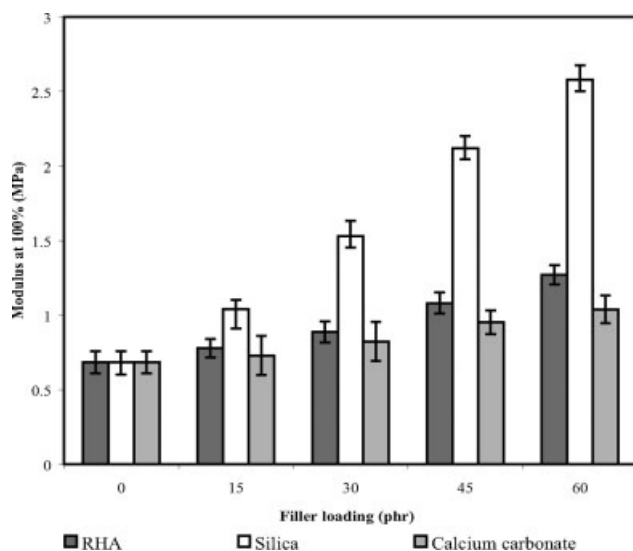


Figure 5 The effect of filler loading on modulus values at 100% of RHA, silica, and CaCO_3 filled 75 : 25 NR/EPDM blends.

is an indication of material stiffness for filler filled 75 : 25 NR/EPDM blends. Fillers are known to increase modulus provided the modulus of the filler is higher than that of the polymer matrix. Modulus of the filled blends increased with an increase in filler loading. It can be seen that silica improved the stiffness of the rubber blends whereas RHA and CaCO_3 showed a small increase in this property. The most important contribution to the elastic modulus arises from polymer–filler interactions which can be increased if a good dispersion of the filler is characteristic of the particles and also on the chemical nature of the polymer. As more filler particles are introduced into the rubber, the elasticity of the rubber chains is reduced, resulting in higher stiffness properties. Sae-Oui et al.¹¹ also reported that surface area is the most important factor controlling the tensile strength, whereas the surface activity controls the modulus.

Tear strength

Figure 6 shows the tear strength of the filled rubber materials. Tear strength, like tensile strength, is affected by surface area. At a similar filler loading, silica filled 75 : 25 NR/EPDM blends gave the highest tear strength, followed by RHA and then CaCO_3 filled 75 : 25 NR/EPDM blends. As can be seen, the addition of silica increased considerably 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 high surface area. In contrast, the addition of the RHA and CaCO_3 fillers reduced somewhat the tear strength of the

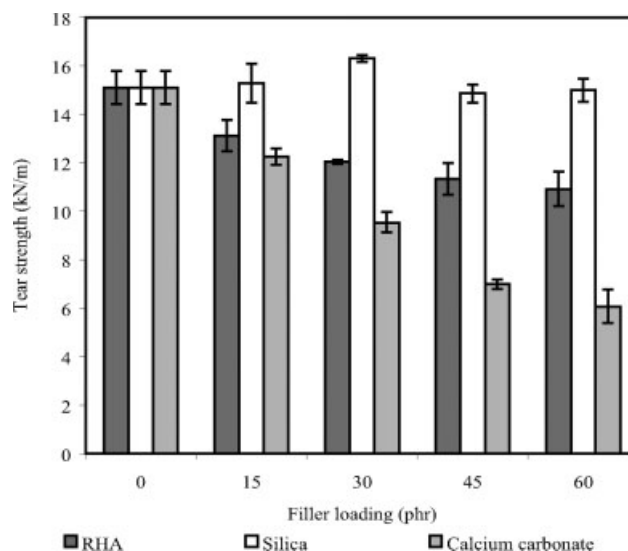


Figure 6 The effect of filler loading on tear strength of RHA, silica, and CaCO_3 filled 75 : 25 NR/EPDM blends.

rubber materials. This may be attributed to their low surface area and low rubber–filler interaction. For this reason, both tensile and tear strengths were decreased with increased loading of the fillers.

Rebound resilience

The effect of various fillers and loading on resilience of the rubber blends is shown in Figure 7. It can be

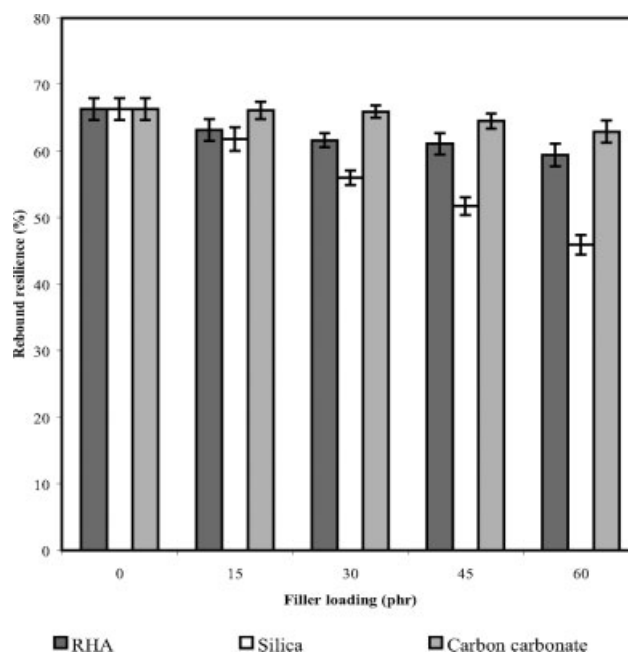


Figure 7 The effect of filler loading on resilience of RHA, silica, and CaCO_3 filled 75 : 25 NR/EPDM blends.

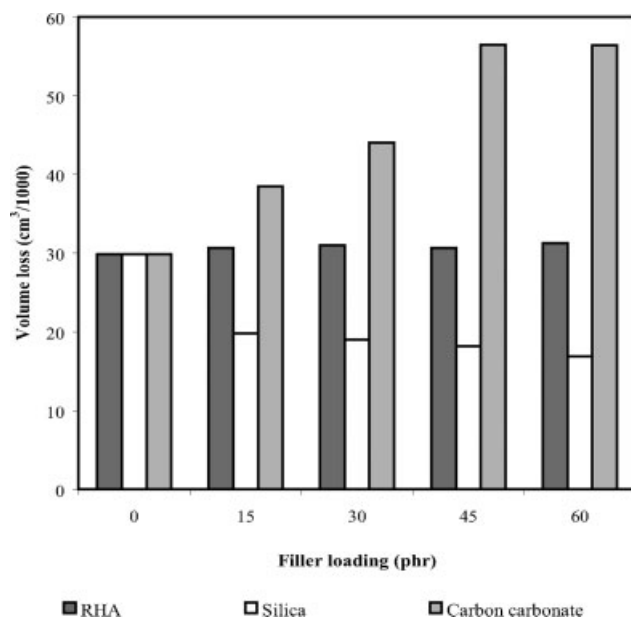


Figure 8 The effect of filler loading on abrasion resistance of RHA, silica, and CaCO₃ filled 75 : 25 NR/EPDM blends.

seen that at a similar filler loading, CaCO₃ filled 75 : 25 NR/EPDM blends gave the highest resilience, followed by RHA and then silica filled 75 : 25 NR/EPDM blends. All of the filler filled 75 : 25 NR/EPDM blends exhibited similar trends; rebound resilience of the filled blends decreased with filler loading. The resilience of the rubber blends decreased drastically with an increase in silica loading, whereas RHA and CaCO₃ showed a small change in this property. This observation may be attributed to a poorer rubber–filler interaction of RHA and CaCO₃. However, the sharp decreasing trend for silica may be attributed to better 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 lower resilience properties. The surface activity is an important factor, indicating the extent of rubber–filler interaction.

Abrasion resistance

The abrasion resistance of a solid body is defined as its ability to withstand the progressive removal of material from its surface, as the result of mechanical action of rubbing, scraping, or of an erosive nature. The effect of various fillers and loading on abrasion loss of the 75 : 25 NR/EPDM blends is shown in Figure 8. The incorporation of silica reduced the abrasion loss of the 75 : 25 NR/EPDM blends notably, whereas CaCO₃ showed a different trend in abrasion loss tending to increase it with an increase in CaCO₃ loading. However, RHA showed less of an effect

with filler loading. Figure 8 also shows that at a similar filler loading, silica filled 75 : 25 NR/EPDM blends had the lowest abrasion loss followed by RHA and CaCO₃ filled 75 : 25 NR/EPDM blends. As far as this property is concerned, the RHA gave a similar effect to nonreinforcement fillers. The abrasion loss with the RHA filler essentially showed no effect with an increase in loading. An inverse effect is found in the systems filled with silica in which, the abrasion loss is found to decrease sharply with an increase of the filler loading at 15 phr and then the abrasion loss decreased marginally with increased loading. Silica interacted preferentially with the rubber phase, as shown by the higher reduction of abrasion loss in the blends. This improvement is probably due to the greater surface area and better filler–rubber interfacial adhesion resulting in an improved abrasion resistance. High surface area particles actually reflect their greater interface between the filler and the rubber matrix and, hence, provide a better abrasion resistance and adhesion than the low surface area particles. Similar results were also reported by Sae-Oui et al.¹¹

Ozone ageing

The interaction of rubber with ozone is best noted when the rubber is stressed or stretched in use. To assess the ozone resistance of the filler filled blends, test samples having 20% strain were exposed to ozonized air of 50 ppm ozone concentration for 72 h at 40°C in the dark. The nature and intensity of cracks due to ozone attack are different for various fillers.

Optical photographs of the surfaces of the unfilled and filled blends at 30 phr filler loading of ozone exposed samples are presented in Figure 9. The photographs clearly show that the unfilled NR/EPDM blend showed fine cracks represented by the horizontal lines [Fig. 9(a)] while the highest level of cracks developed were deep, wide, and continuous in RHA [Fig. 9(b)]. However, the CaCO₃ filled blends [Fig. 9(d)] showed longer cracks than unfilled blends. The silica filled NR rubber blends showed no cracks as seen in Figure 9(c), which confirms that crack growth was stopped more effectively by the finely dispersed filler particles in the rubber matrix and a better filler–matrix interaction was achieved with the silica filled NR/EPDM blends. This shows that the ozone resistance of the blends containing reinforcing fillers is superior to the blends containing unfilled and other filled blends. Results revealed that when a reinforcing filler is added to a blend, it has a higher affinity to go more into the polymer phase. That is, better filler–matrix interaction prohibits the growth of ozone cracks initiated in the rubber matrix before the cracks grow over the critical length

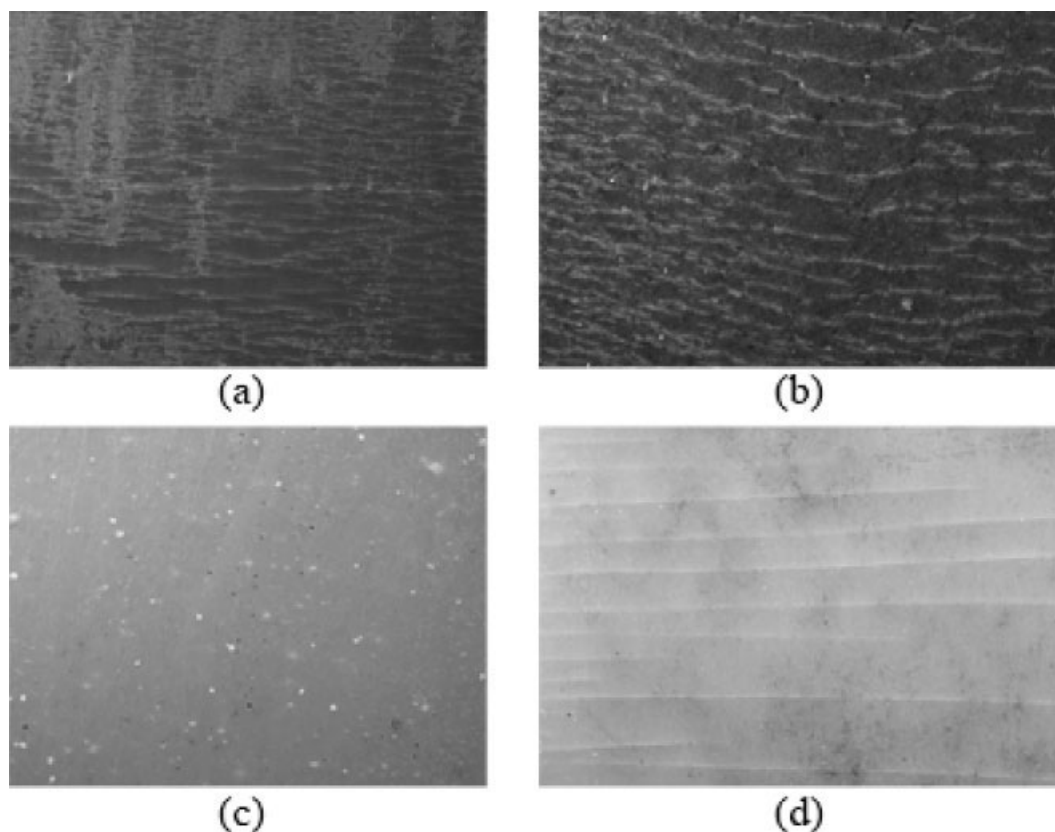


Figure 9 Optical photographs of exposed (a) unfilled 75 : 25 NR/EPDM blends and (b) RHA, (c) silica, and (d) CaCO_3 filled 75 : 25 NR/EPDM blends after tensile fracture at 30 phr filler loading ($\times 50$).

for failure. Thus, the ozone resistance of the silica filled blends improved remarkably. The use of CaCO_3 as an additional filler provided a slight improvement in the ozone resistance of the 75 : 25 NR/EPDM blends. In the case of RHA filled blends, the ozone resistance of the blends was inferior to those obtained for the unfilled blend. This may be due to the poor adhesion and nonuniform dispersion of the discrete phase in the matrix.

Morphology

The morphology of unfilled and filler filled 75 : 25 NR/EPDM blends at 30 phr filler loading after tensile fracture is shown in Figure 10. The surface of the unfilled 75 : 25 NR/EPDM blends is smooth [Fig. 10(a)]. For the fracture surface of RHA and CaCO_3 filled 75 : 25 NR/EPDM blends [Fig. 10(b,d)], the dispersion of filler in the rubber matrix was not continuous as a result of filler agglomeration because of a lack of interfacial interaction between the filler and the matrix, which led to void formation. The presence of many holes on the fracture surface may be due to the weak interfacial interaction resulting in the deterioration of blend properties such as tensile and tear strength when compared with unfilled 75 : 25 NR/EPDM blends. For the

silica filled 75 : 25 NR/EPDM blend as shown in Figure 10(c), it can be seen that the fracture surface of the reinforcing filler shows a more homogenous phase dispersion, resulting in a higher tensile strength. The SEM photomicrographs shown in Figure 10 confirm that a larger surface area of the filler provides a better filler dispersion and interfacial bond between filler and rubber matrix.

CONCLUSIONS

We studied binary blends of NR and EPDM with a fixed 75 : 25 blend ratio (wt %) to investigate the effects of three different types of fillers on the processability and mechanical properties. Silica, RHA, and CaCO_3 were used as filler. The incorporation of fillers in 75 : 25 NR/EPDM blends decreased the cure time t_{90} with an increase in RHA and CaCO_3 loading whereas silica showed a different trend in cure time tending to increase with an increase in silica loading. At a similar filler loading, RHA showed the shortest t_{90} followed by CaCO_3 and silica. Mooney viscosity increased with an increase in silica loading, whereas RHA and CaCO_3 showed a small change in this property. At a similar filler loading, CaCO_3 showed the lowest viscosity followed by RHA and silica. Filler loading and filler

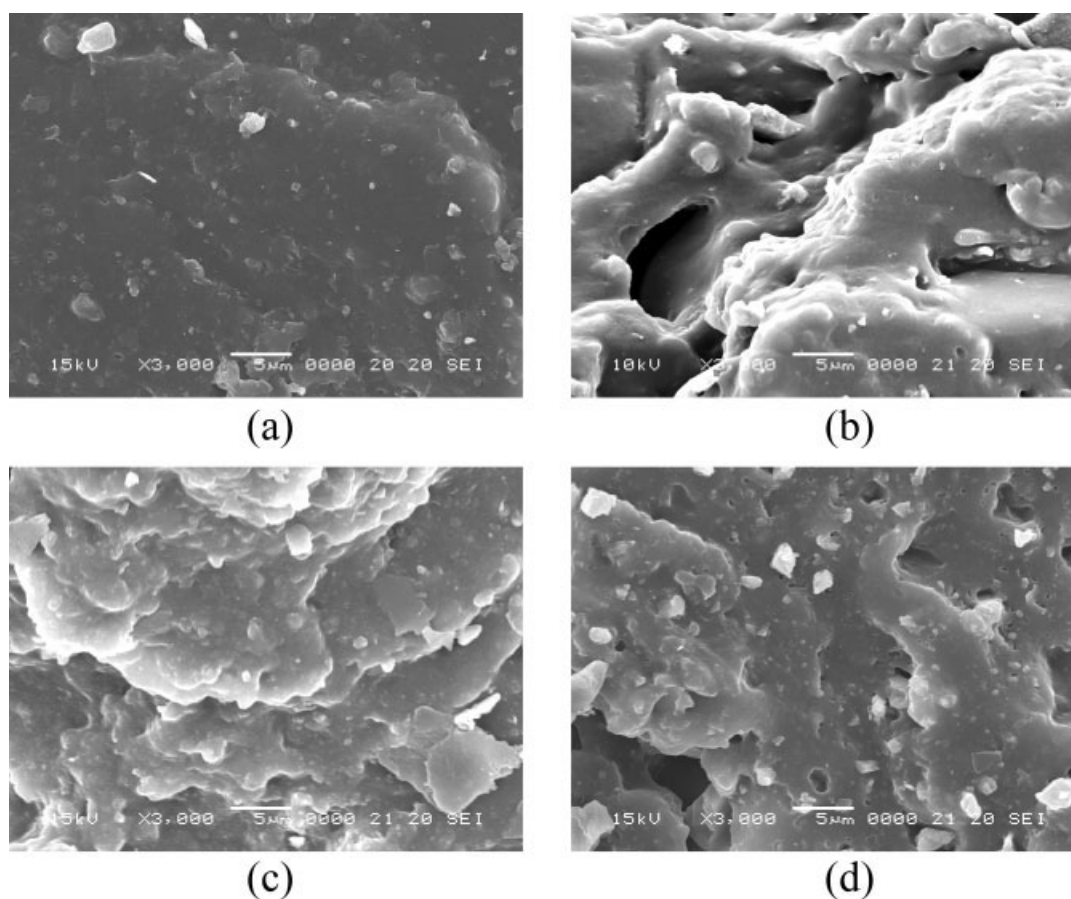


Figure 10 SEM micrographs of (a) unfilled 75 : 25 NR/EPDM blends and (b) RHA, (c) silica, and (d) CaCO_3 filled 75 : 25 NR/EPDM blends after tensile fracture at 30 phr filler loading ($\times 3000$).

type influence the processability of the rubber blends with RHA, offering a better processing advantage over silica and CaCO_3 . Additions of silica gave rise to marked improvements in the tensile and tear strength and abrasion resistance of 75 : 25 NR and EPDM blends. Tensile and tear strength and abrasion resistance of the materials reduced with increasing CaCO_3 loading, whereas RHA showed a small change in these properties. Young's modulus increased with increasing filler loading while resilience is often reduced. At a similar filler loading, CaCO_3 showed the highest resilience followed by RHA and silica. Optical photographs of ozone exposed samples showed that no cracks were found in all of the silica filled blends. The ozone resistance of the blends showed no significant improvement with the addition of the CaCO_3 filler while this property of the blends was inferior to those obtained from the addition of the RHA filler. Scanning electron microscopy on fracture surfaces shows poor filler dispersion and weak filler–matrix interaction with the RHA and CaCO_3 filled NR/EPDM blends. This explains the reduction of tensile strength with RHA and CaCO_3 filled NR/EPDM blends. Although, from overall mechanical properties, it

could be suggested that RHA has poorer reinforcing ability than silica, one may still consider the utilization of RHA as a filler in the rubber industry for economic and ecological reasons. Furthermore, RHA provides better mechanical properties than the CaCO_3 . Overall results indicate that RHA could be used as a nonreinforcing filler for the 75 : 25 NR/EPDM blend to replace CaCO_3 in rubber blends.

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References

1. Botros, S. H. *Polym Plast Technol Eng* 2002, 41, 341.
2. Ghosh, A. K.; Debnath, S. C.; Naskar, N.; Basu, D. K. *J Appl Polym Sci* 2001, 81, 800.
3. Ono, S.; Kiuchi, Y.; Sawanoburi, J.; Ito, M. *Polym Int* 1999, 48, 1035.
4. Manna, A. K.; De, P. P.; Tripathy, D. K.; De, S. K.; Peiffer, D. G. *J Appl Polym Sci* 1999, 74, 389.
5. George, K. M.; Varkey, J. K.; Thomas, K. T.; Mathew, N. M. *J Appl Polym Sci* 2002, 85, 292.

6. Zhang, J.; Yao, Y.-Z.; Li, L.; Zhou, N.-L.; Huang, Y.-X.; Ma, Z.-M.; Shen, J. *J Appl Polym Sci* 2007, 103, 2395.
7. Chiu, H.-T.; Chiu, W.-M. *J Appl Polym Sci* 1996, 61, 607.
8. Nakajima, N.; Yamaguchi, Y. *J Appl Polym Sci* 1997, 66, 1445.
9. Sukumar, R.; Menon, A. R. R. *J Appl Polym Sci* 2007, 107, 3476.
10. Mathew, S.; Varghese, S.; Rajammal, G.; Thomas, P. C. *J Appl Polym Sci* 2007, 104, 58.
11. Sae-Oui, P.; Rakdee, C.; Thanmathorn, P. *J Appl Polym Sci* 2002, 83, 2485.
12. Chan, C.-M.; Wu, J.; Li, J.-X.; Cheung, Y.-K. *Polymer* 2002, 43, 2981.
13. Atikler, U.; Basalp, D.; Tihminlioglu, F. *J Appl Polym Sci* 2006, 102, 4460.
14. Shah, G. B.; Fuzail, M. *J Appl Polym Sci* 1928, 2006, 99.
15. Ismail, H.; Nordin, R.; Noor, A. M. *Polym Plast Technol Eng* 2002, 41, 847.
16. Arayaprane, W.; Na-Ranong, N.; Rempel, G. L. *J Appl Polym Sci* 2005, 98, 34.
17. Da Costa, H. M.; Visconte, L. L. Y.; Nunes, R. C. R.; Furtado, C. R. G. *J Appl Polym Sci* 2000, 76, 1019.
18. Da Costa, H. M.; Visconte, L. L. Y.; Nunes, R. C. R.; Furtado, C. R. G. *J Appl Polym Sci* 2002, 83, 2331.
19. Da Costa, H. M.; Visconte, L. L. Y.; Nunes, R. C. R.; Furtado, C. R. G. *J Appl Polym Sci* 2003, 87, 1194.
20. Da Costa, H. M.; Visconte, L. L. Y.; Nunes, R. C. R.; Furtado, C. R. G. *J Appl Polym Sci* 2003, 87, 1405.
21. Da Costa, H. M.; Visconte, L. L. Y.; Nunes, R. C. R.; Furtado, C. R. G. *J Appl Polym Sci* 2003, 90, 1519.
22. Siriwardena, S.; Ismail, H.; Ishiaku, U. S. *Polym Plast Technol Eng* 2001, 40, 519.
23. Siriwardena, S.; Ismail, H.; Ishiaku, U. S. *Polym-Plast Technol Eng* 2002, 41, 419.
24. Siriwardena, S.; Ismail, H.; Ishiaku, U. S.; Perera, M. C. S. *J Appl Polym Sci* 2002, 85, 438.
25. Rigbi, Z. *Rubber Chem Technol* 1982, 55, 1180.