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On The Potential of Organoclay with Respect to Conventional Fillers (Carbon Black, Silica) for Epoxidized Natural Rubber Compatibilized Natural Rubber Vulcanizates

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ABSTRACT: Onium modified montmorillonite (organoclay) was compounded with natural rubber (NR) in an internal mixer and cured by using a conventional sulfuric system. Epoxidized natural rubber with 50 mol % epoxidation (ENR 50) in 10 parts per hundred rubber (phr) was used as a compatibilizer in this study. For comparison purposes, two commercial fillers: carbon black (grade N330) and silica (grade vulcasil-S) were used. Cure characteristics were carried out on a Monsanto MDR2000 Rheometer. Organoclay filled vulcanizate showed the lowest values of torque maximum, torque minimum, scorch, and cure times. The kinetics of cure reaction showed organoclay could behave as a cocuring agent. The mechanical testing of the vulcanizates involved the determination of tensile and tear properties. The improvement of tensile strength, elongation at break, and

tear properties in organoclay filled vulcanizate were significantly higher compared to silica and carbon black filled vulcanizates. In terms of reinforcing efficiency (*RE*), organoclay exhibited the highest stiffness followed by silica and carbon black filled vulcanizates. Scanning electron microscopy revealed that incorporation of various types of fillers has transformed the failure mechanism of the resulting NR vulcanizates compared to the gum vulcanizates. Dynamic mechanical thermal analysis (DMTA) revealed that the stiffness and molecular relaxation of NR vulcanizates are strongly affected by the filler-rubber interactions. © 2004 Wiley Periodicals, Inc. *J Appl Polym Sci* 94: 2438–2445, 2004

Key words: nanocomposites; organoclay; natural rubber; epoxidized natural rubber; silica; carbon black

INTRODUCTION

Polymer/organoclay nanocomposites are a new class of materials that consist of two or more materials with one having a dimension in the nanometer range i.e., one billionth of a meter. These materials give a combination of properties that are not available in any of the individual components. Reinforcement is possible at very low loading of fillers (< 10 wt %) compared to conventional filler composites, which require much larger quantities.¹ Organoclay is a filler containing layered silicates that are normally occupied by cations such as Na⁺ and Ca⁺, which are usually replaced by organic cations such as alkyl ammonium cations via ion exchange reaction to obtain hydrophobic surfaces.² The crystal structure of a layered silicate (about 1 nm thick) such as montmorillonite (MMT) contains

octahedral alumina or magnesia sandwiched in between two tetrahedral silica sheets.³

The most important consideration to achieve successful nanocomposite formation is the dispersion of organoclay particles in polymer matrix. In the use of such organoclay as fillers, generally, three types of composite materials may be obtained: conventional composites consisting of clay tactoids having stacked layers; this type of reinforcement is similar to conventional fillers such as carbon black and silica; intercalated nanocomposites, whereby polymer chains are intercalated into silicate layers, and exfoliated nanocomposites, involving delamination of silicate layers and good dispersion in the polymer matrix. From all three types, the exfoliated clay is the best situation in terms of polymer reinforcement.⁴ According to previous research, exfoliated clay further enhanced the modulus, thermal expansion coefficient, swelling resistance, and ionic conductivity compared to the pristine polymers.^{5–9}

So far, the biggest challenge has been to find alternative “nonblack” fillers to replace carbon black in

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TABLE I
Physical Properties of Organoclay, Silica, and Carbon Black

	Organoclay	Silica	Carbon black
Mean particle size (μm)	4.43	7.17	9.14
Specific surface area (m^2/g)	0.6775	0.4184	0.3281
Density (g/cm^3)	2.00	2.00	2.00

rubber stocks.¹⁰ For this reason, organoclays will be compared with the most established fillers such as carbon black and silica. The potential of organoclay as a filler in ENR 50 compatibilized NR-clay nanocomposites will be investigated in this study. In a previous study on the natural rubber-organoclay nanocomposites,¹¹ the effect of epoxidized natural rubber as a compatibilizer on the cure and physical properties was reported. The tensile and tear strength of the ENR 50 containing nanocomposites were superior to the ENR 25 compatibilizer systems. X-ray diffraction (XRD) and transmission electron microscopy (TEM) results revealed that the organoclay was mostly intercalated in the ENR 50 compatibilized NR-clay nanocomposites.

For comparison purposes two commercial fillers, carbon black and silica, were used. Filler loadings at 50 phr for carbon black, 30 phr for silica, and 2 phr for organoclay were used, being the optimum levels of reinforcement reported in previous studies.^{12,13} In our previous work, 2 phr of organoclay has been determined as the optimum filler content, which achieved the best overall physical properties.¹¹ Earlier work by Boochathum et al.¹⁴ indicated that vulcanization using conventional vulcanization, "CV system," with a high concentration of sulfur yielded a higher degree of vulcanization and crosslinking in NR vulcanizates, so the CV system was used in this study.

EXPERIMENTAL

Materials

The elastomers used were natural rubber (SMR L) and epoxidized natural rubber (ENR) with Mooney viscosities of ML (1 + 4) 100°C = 78, ML (1 + 4) = 140, respectively. Both were purchased from Kumpulan Guthrie Sdn. Bhd., Seremban, Malaysia. The ENR contained 50 mol % epoxidation (ENR 50). The onium ion modified montmorillonite-based organoclay (Nanomer L28E; intercalant type, containing octadecyltrimethylamine compound) was purchased from Nanocor Inc., USA. Carbon black (grade N330), precipitated silica (grade Vulcasil S), and other compounding ingredients: sulfur, zinc oxide, stearic acid, *N*-isopropyl-*N'*-phenyl-*p*-phenylenediamine (IPPD), and *N*-cyclohexyl-2-benzothiazyl sulfenamide (CBS) were pur-

chased from Bayer (M) Sdn. Bhd Malaysia. The physical properties of organoclay, carbon black, and silica are given in Table I.

Compounding

A conventional vulcanization system was used for compounding. Table II shows the composition of the rubber stocks. The master batch (MB) preparation was done by using a Banbury internal mixer model BR1600. The rubber was masticated for 30 s and all ingredients, except curatives, were added and mixing continued for another 2.5 min. The compounding was done at 80°C and 100 rpm for 3 min. Curatives were mixed into the MB by using a two-roll mill sized (160 × 320 mm) according to ASTM designation D 3184-80 at 70 ± 5°C compounds for 2 min.

Cure characterization

Compound cure characteristics were measured using a Monsanto Moving Die Rheometer, model MDR 2000, at 150°C according to ASTM 2240-93.

Compression molding

The vulcanizates were prepared by curing the rubber sheets under pressure of 10 MPa at 150°C in a KAO Tech hot press for the respective cure times, t_{90} determined from the MDR 2000 tests. Vulcanized sheets of 2 mm thickness were produced.

Tensile properties

Dumbbell-shaped samples were cut from the molded sheets according to ASTM D412-93. Tensile properties were determined on a Monsanto Tensometer M500 with the crosshead speed of 500 mm/min.

TABLE II
Composition of the Rubber Compounding

Ingredient	Content (phr) ^a		
Natural rubber (SMR L)	100	100	100
ENR 50	10	10	10
Organoclay	2	—	—
Carbon black	—	50	—
Silica	—	—	30
Sulfur	2.5	2.5	2.5
Zinc oxide	5.0	5.0	5.0
IPPD	2.0	2.0	2.0
Stearic acid	1.0	1.0	1.0
CBS	0.5	0.5	0.5

^a Parts per hundred parts of rubber.

ENR, epoxidized natural rubber; IPPD, *N*-isopropyl-*N'*-phenyl-*p*-phenylenediamine; CBS, *N*-cyclohexyl-2-benzothiazyl sulfenamide.

TABLE III
Cure Characteristics of NR Vulcanizates Unfilled and Filled of Organoclay, Carbon Black, and Silica

Parameters	Gum	Organoclay	Carbon Black	Silica
Scorch time (t_2), (min)	5.40	1.49	2.20	4.53
Cure time (t_{90}) (min)	10.38	6.70	11.25	22.01
Minimum torque (M_L) (dNm)	0.97	1.33	1.87	2.02
Maximum torque (M_H) (dNm)	6.89	8.41	13.84	14.07
Max-min torque ($M_H - M_L$) (dNm)	5.92	7.08	11.97	12.05
Cure rate	0.112	0.145	0.088	0.073

Tear properties

Crescent-shaped samples, type die B, were stamp from the molded sheets according to ASTM D624-9. A nick (0.50 ± 0.05 mm) in depth was produced by using a sharp razor blade. Tear strength was determined on a Monsanto Tensometer M500 with the crosshead speed of 500 mm/min.

Swelling index

Swelling index was determined in toluene according to ASTM D471-79. Specimens of dimension $30 \times 5 \times 2$ mm³ were weighed. The test specimens were immersed in toluene at room temperature for 72 h. The swelling index was calculated as follows:

$$\text{Swelling index} = \frac{\text{Swollen mass}}{\text{Original mass}} \times 100 \quad (1)$$

Scanning electron microscopy (SEM)

The fracture surface of the vulcanizates was studied with a Leica Cambridge S-360 scanning electron microscope. The fracture surfaces were coated with gold to prevent electrostatic charging during examination.

Dynamic mechanical thermal analysis (DMTA)

The complex modulus (E^*), its storage (E'), loss (E''), and the mechanical loss factor ($\tan \delta = E''/E'$), as a function of temperature (T), were assessed by DMTA using an Eplexor 25N device of Gabo Qualimeter, Germany. DMTA spectra were taken on rectangular specimens (length \times width \times thickness) at 10 Hz frequency in a broad temperature range ($T = -100$ to $+100^\circ\text{C}$). The DMTA device operated under load control by setting 50 N as static and 50 N as dynamic load.

RESULTS AND DISCUSSION

Cure characterization

Table III shows the cure characteristics of the NR gum vulcanizate and vulcanizates filled with organoclay, carbon black, and silica.

It can be seen that each compound exhibits different cure characteristics, which may be attributed to the fact that each type of filler possesses different properties, such as particle size, structure, and aspect ratio.¹⁵ Organoclay filled vulcanizate exhibits the shortest scorch time, t_2 , and cure time, t_{90} , compared with gum vulcanizate and carbon black, silica filled vulcanizates. This may be related to a transition complex formation with amines and sulfur, which accelerates the curing process. This has also been observed for other rubbers.¹⁶ A similar effect was earlier reported by Mousa and Karger-Kocsis¹⁰ and Varghese et al.¹⁷ for SBR and ENR 50, respectively. The cure retardation displayed by the silica filled system as indicated by the highest t_2 and t_{90} values can be attributed to a silica-accelerator interaction. The silica reacts with stearic acid, zinc oxide, and subsequently reduces the number of active sulfurating agents, slowing down the sulfur reaction and increasing the t_2 and t_{90} of the vulcanizates.¹⁸

Silica filled vulcanizates show the highest torque maximum (M_H) and minimum (M_L) torque, followed by carbon black, and organoclay. The difference in the torque values may be related to the particle size of the fillers (Table I). It is believed that, the smaller the particle size is, the larger the surface area is, which implies the greater rubber-filler interaction. This tends to impose extra resistance to flow as a higher restriction to molecular motion of the macromolecules.¹⁹ However, it is interesting to note that, despite having the smallest particle size, the organoclay system recorded relatively lower M_H and M_L values compared to those of silica and carbon black systems. This may be attributed to the low organoclay loading (2 phr) compared to silica (30 phr) and carbon black (50 phr). The kinetics of the cure reaction is summarized as eq. (2):

$$dx/dt = k(a - x)^n \quad (2)$$

where k is the reaction rate constant, a is the initial reactant concentration, x is the consumed reactant at time t , and n is the reaction order. Equation (2) is analyzed by considering the equation for the kinetics of a first order chemical reaction:

$$\ln(a - x) = -kt + \ln a \quad (3)$$

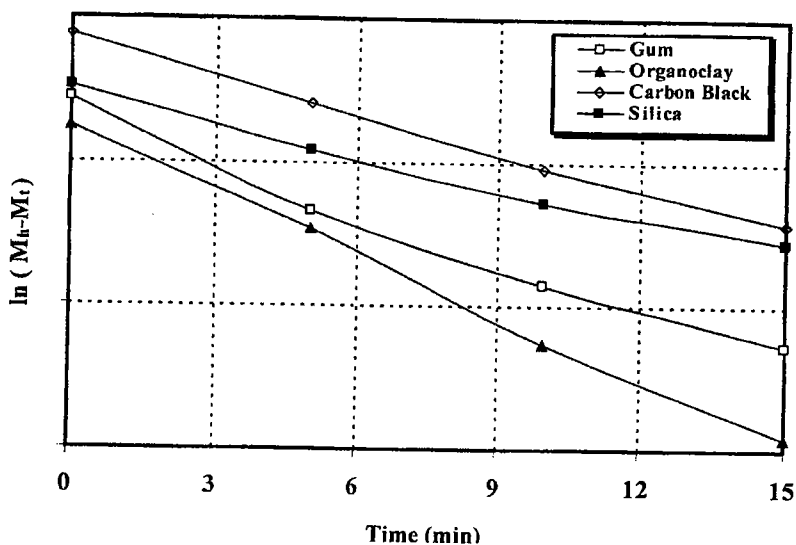


Figure 1 Kinetic plot for the cure reaction of gum and filled vulcanizates of organoclay, silica, and carbon black.

The rate of crosslinking is usually obtained by measuring the torque developed during vulcanization. If the change in a physical property, such as modulus, is measured rather than the change in reactant concentration, the following substitution can be made:²⁰

$$(a - x) = M_H - M_t \quad (4)$$

$$a = M_H - M_L \quad (5)$$

where M_H is the maximum torque, M_t is the torque at time t , and M_L is the minimum torque. Figure 1 shows the kinetic plot for the cure reaction of gum and filled vulcanizates with organoclay, silica, and carbon black. The cure rate can be obtained, based on the slope of the kinetic plot, and is shown in Table III. Organoclay filled vulcanizate displayed the highest cure rate among the others. These data are consistent with the cure and scorch time. This indicates that organoclay is not just a filler but also behaves as a cocuring agent, which could facilitate the curing reaction.

Mechanical properties

Figure 2 shows tensile strength and elongation at break of NR gum and vulcanizates filled with organoclay, carbon black, and silica. Organoclay filled NR vulcanizate showed the highest tensile strength followed by silica and then carbon black filled vulcanizates. The better reinforcement efficiency of organoclay may be attributed to the strong interaction between organoclay and NR.⁴ The smaller particle size provides a larger surface area for the interaction between the filler and rubber matrix. Silica has a larger surface area than carbon black (Table I), so it shows

better tensile strength than carbon black. On the other hand, the better tensile properties of organoclay could be due to its high aspect ratio and platelet structure. Elongation at break for organoclay filled NR vulcanizate shows the highest value followed by silica and carbon black filled. Organoclay shows higher elongation at break than NR gum vulcanizate, as reported earlier.¹⁰ This is believed to be related to the interca-

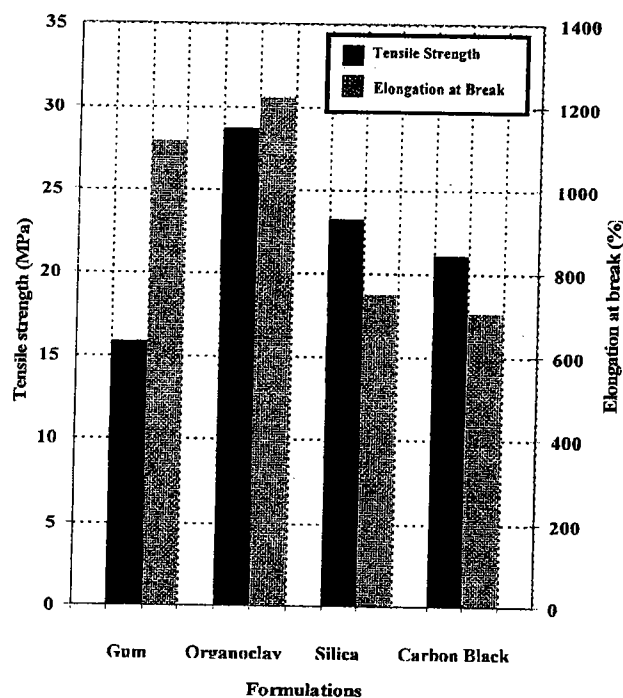


Figure 2 Tensile strength and elongation at break of NR gum, organoclay, silica, and carbon black filled vulcanizates.

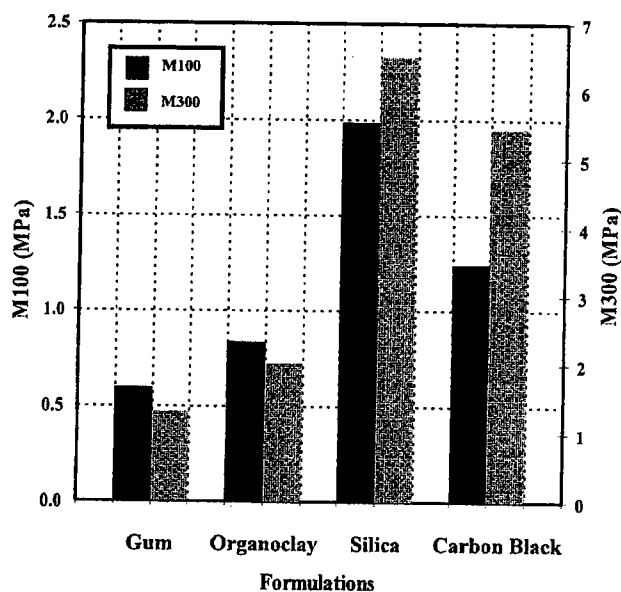


Figure 3 Modulus at 100% (M100) and modulus at 300% (M300) of NR gum, organoclay, silica, and carbon black filled vulcanizates.

lation/exfoliation process, which resulted in high strength reinforcement at very low filler loading. The elongation of the rubbers is largely retained due to the very low concentration organoclay. The same reason could be used to account for the very low modulus of the organoclay filled vulcanizate.¹⁰

The moduli at 100% elongation (M100) and 300% elongation (M300) are shown in Figure 3. Although silica filled vulcanizate shows the highest M100 and M300 among all these fillers, in terms of reinforcing efficiency (RE), as shown in Table IV, organoclay exhibits higher stiffness than silica and carbon black filled vulcanizates. RE is determined as the change in M100 per wt % of filler in the matrix:²¹

$$RE = (M_{100, \text{filled}} - M_{100, \text{gum}}) / \text{wt \% filler} \quad (6)$$

The RE values could be compared relative to each other. The stiffness or modulus is affected by surface reactivity of fillers, particle size, and filler-rubber interaction.²² For organoclay, as mentioned before, the filler-rubber interaction is stronger than that of silica

TABLE IV
Reinforcing Efficiency of Organoclay, Silica, and Carbon Black Filled Vulcanizates

	M ₁₀₀ (MPa)	Filler (wt %)	RE ^a
Organoclay	0.83	1.62	0.142
Silica	1.98	19.87	0.069
Carbon black	1.24	29.24	0.022

^a Reinforcing efficiency calculated by eq. (6).

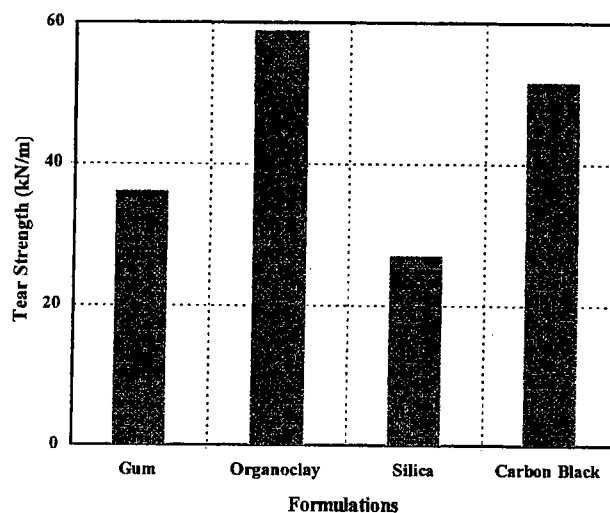


Figure 4 Tear strength of NR gum, organoclay, silica, and carbon black filled vulcanizates.

and carbon black. It also can be attributed to the high aspect ratio of organoclay, which allows better interaction with the rubber matrix, thus its RE is the highest followed by silica and carbon black.

Tear strength for organoclay filled vulcanizate shows the highest value followed by silica and carbon black filled vulcanizates (Fig. 4). The high tear strength of the organoclay vulcanizate is related to the intercalation/exfoliation phenomenon and also the higher RE, as explained in the mechanical properties above, as well for silica and carbon black filled vulcanizates.

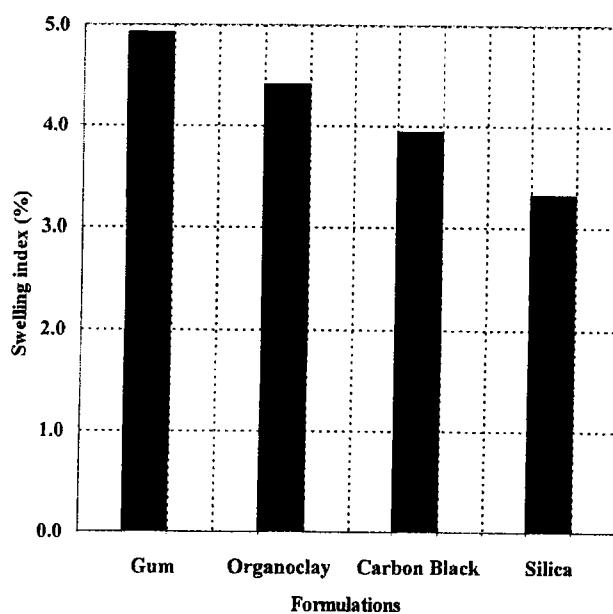


Figure 5 Swelling index of NR gum, organoclay, silica, and carbon black filled vulcanizates.

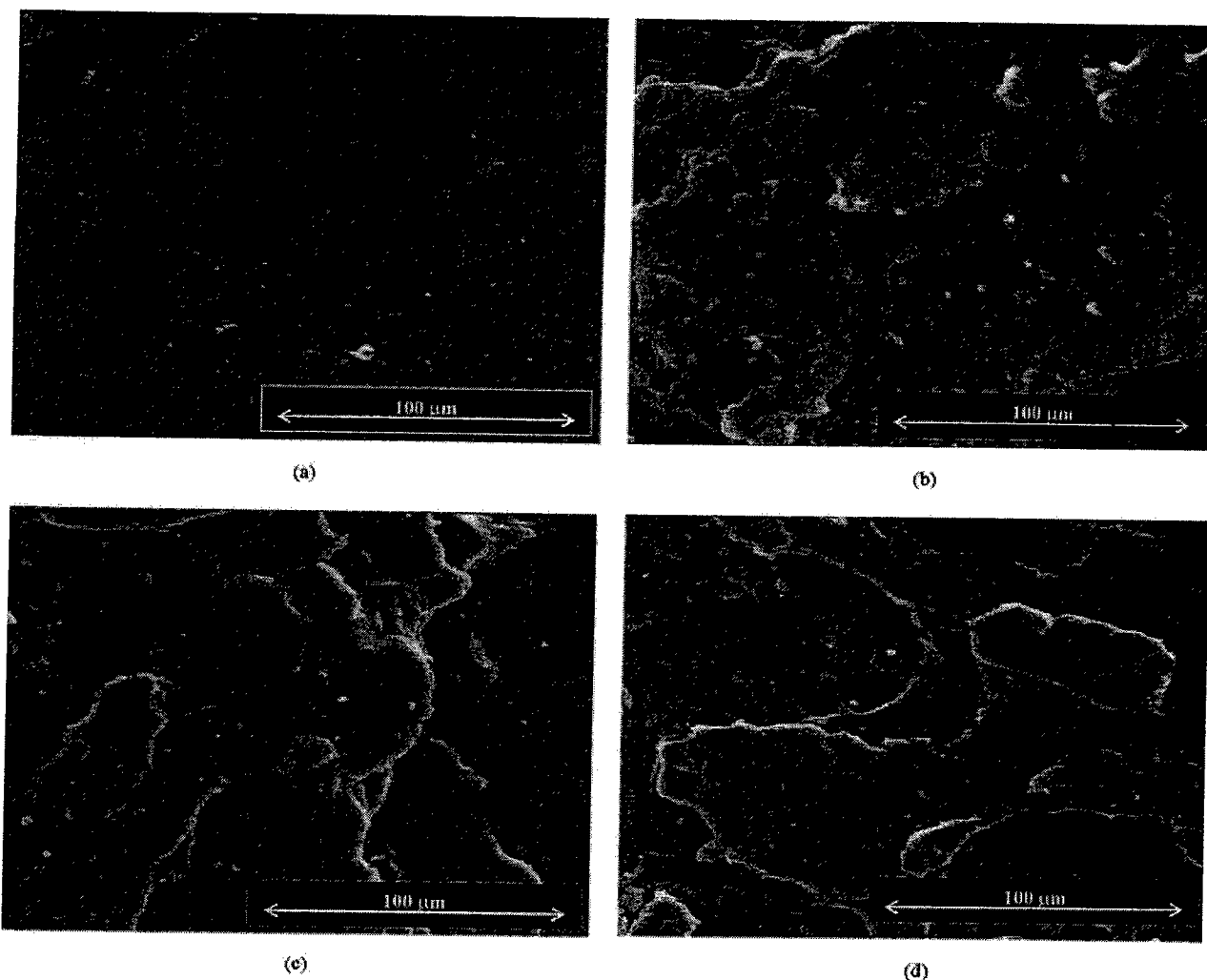


Figure 6 (a) SEM micrograph showing tensile fracture surface of the NR gum vulcanizate. (b) SEM micrograph showing tensile fracture surface of the organoclay filled NR vulcanizate. (c) SEM micrograph showing tensile fracture surface of the silica filled NR vulcanizate. (d) SEM micrograph showing tensile fracture surface of the carbon black filled NR vulcanizate.

Swelling index

Figure 5 shows swelling indices of NR gum, organoclay, silica, and carbon black filled vulcanizates. The silica filled vulcanizate shows the lowest value of swelling index, followed by carbon black and organoclay. The study by Mousa et al.²³ reported a similar trend. This indicates that the silica filled vulcanizate is stiffer and less penetrable by the solvent due to the high concentration of filler compared to the organoclay filled vulcanizate.

Scanning electron microscopy

Figures 6(a)–(d) show the SEM micrographs of gum, organoclay, silica, and carbon black filled NR vulcanizates. Filled NR vulcanizates [Figs. 6(b)–(d)] exhibit rougher fracture surface compared with the gum NR vulcanizate shown in Figure 6(a). This indicates that

the failure mechanism is strongly related to the reinforcement of filler–rubber matrix interaction. There is no significant difference in the morphological appearance between the organoclay filled vulcanizate [Fig. 6(b)] compared to the silica [Fig. 6(c)] and carbon black [Fig. 6(d)] filled vulcanizates. However, according to Costa et al.,²⁴ the larger specific surface area and smaller particle size of organoclay is believed to provide better filler–rubber interaction. This perhaps explains why organoclay filled vulcanizate shows better mechanical properties than the conventional fillers (cf. Fig. 1).

Dynamic mechanical thermal analysis

The dynamic storage modulus of gum NR and the filled vulcanizates as a function of temperature is shown in Figure 7(a). All of the filled vulcanizates

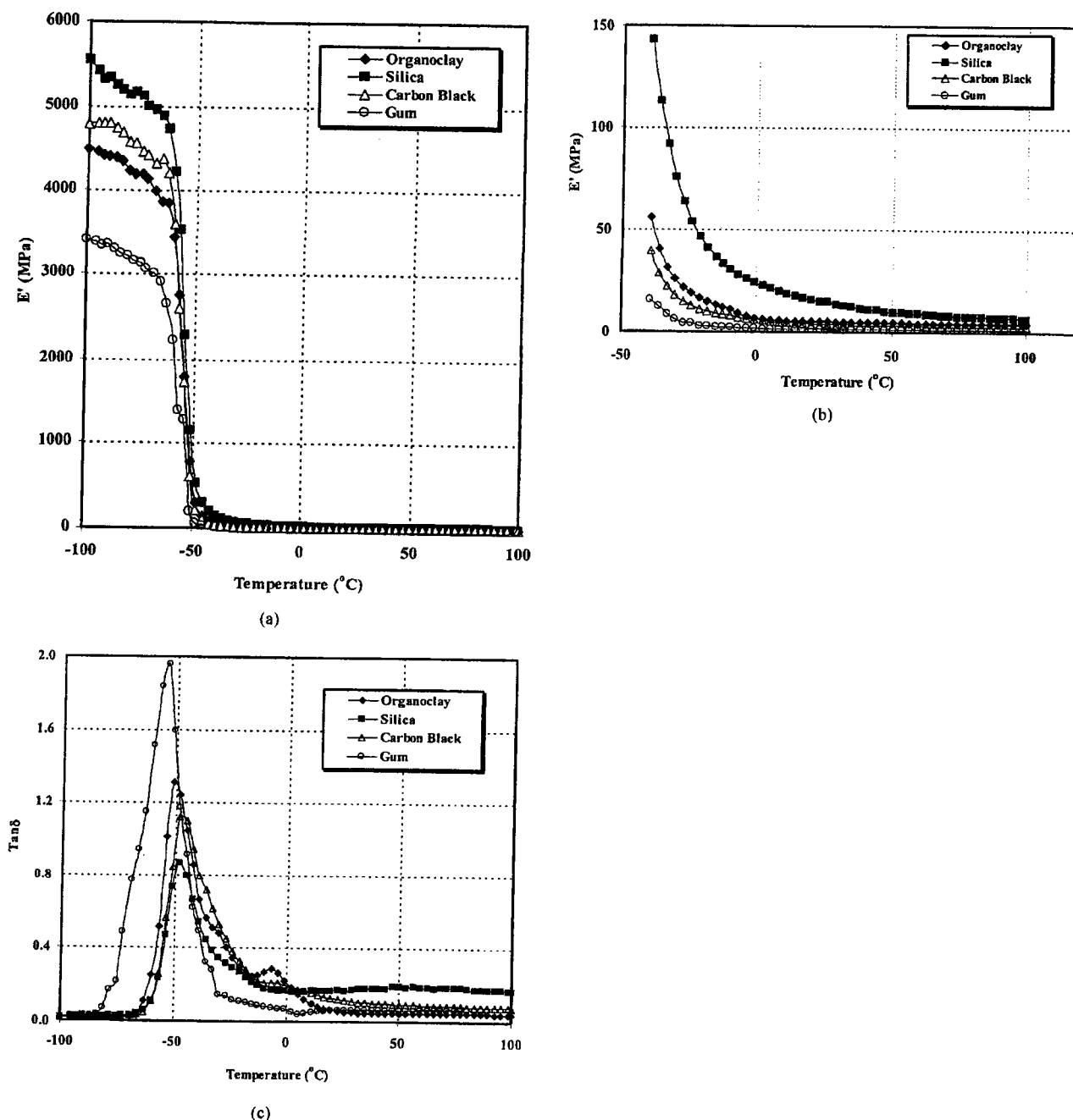


Figure 7 (a) Storage modulus (E') as a function of temperature for NR gum, organoclay, silica, and carbon black filled vulcanizates. (b) Storage modulus (E') as a function of temperature after the T_g for NR gum, organoclay, silica, and carbon black filled vulcanizates. (c) Loss factor ($\tan\delta$) as a function of temperature for NR gum, organoclay, silica, and carbon black filled vulcanizates.

show higher storage modulus than the gum vulcanizate; the ranking of fillers, corresponding to the stiffness below glass transition temperature (T_g), is: silica > carbon black > organoclay. Figure 7(b), which is an enlargement of Figure 7(a), i.e., for temperatures ranging from -100 $^{\circ}\text{C}$ to -50 $^{\circ}\text{C}$, indicates that silica filled vulcanizate still shows the highest modulus value

compared to carbon black and organoclay filled vulcanizates. However, it is interesting to note that the modulus of the organoclay vulcanizate is comparable to the carbon black filled vulcanizate. These results are similar with the M100 and M300 data shown in Figure 3. Similar observations were reported by Jimenez et al.²⁵ and Hambir et al.²⁶ for other rubber vulcanizates.

Figure 7(c) shows the variation of $\tan\delta$ as a function of temperature for NR gum, organoclay, silica, and carbon black filled vulcanizates. The height of the T_g peaks in the $\tan\delta$ curve for the vulcanizates are lower than those of the gum vulcanizate. This suggests a strong interaction between the fillers and the NR matrix as the molecular relaxation of the latter is hampered. Silica filled vulcanizates show the lowest damping peak followed by carbon black and organoclay. All the filled vulcanizates exhibit higher T_g compared to gum vulcanizate. This may again be attributed to the strong filler-rubber interaction and improved chain flexibility.²⁷ It can also be seen that there is a secondary peak observed at -6.0°C for the organoclay filled vulcanizate. A similar observation was also reported by Varghese and Karger-Kocsis²⁸ and Kumnuantip and Sombatsompop²⁹. This might be attributed to the part of the rubber that is intercalated in between the silicate layers.

CONCLUSION

Based on this work, devoted to study the effect of epoxidized natural rubber as a compatibilizer on the properties of natural rubber vulcanizates based on organoclay, silica, and carbon black, the following conclusions can be drawn:

Curing characteristics

The different cure characteristics displayed by organoclay, carbon black, and silica NR vulcanizates could be related not only to the fillers' characteristics, i.e., particle size, structure, and aspect ratio, but also to the filler-rubber interaction. The kinetics of cure reaction study revealed that organoclay could behave as a cocuring agent by facilitating the curing reactions.

Mechanical properties

Incorporation of 2 phr of organoclay and 10 phr of ENR 50, as reinforcement and compatibilizer, respectively, resulted in a significant enhancement of tensile and tear properties of the vulcanizates. This was attributed to the high aspect ratio of the filler and to the strong filler-rubber interaction. Further evidence of the interaction was derived from the relaxation behavior of the filled vulcanizates from the dynamic thermal mechanical analysis.

Fracture morphology

Morphological evidence from SEM revealed that the filler-matrix interaction played a strong role in determining the failure mechanism of the NR vulcanizates.

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