

## INVESTIGATION INTO MORPHOLOGY, MICROSTRUCTURE AND PROPERTIES OF SBR/EPDM/ORGANO MONTMORILLONITE NANOCOMPOSITES

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**Abstract** Rubber compounds based on styrene-butadiene rubber/ethylene propylene diene monomer blends of different compositions (60/40, 70/30, 80/20, 90/10, 100/0) reinforced with 1 wt%, 3 wt%, 5 wt% and 7 wt% organoclay (Cloisite 20A) were prepared on a two roll mill via a vulcanization process and characterized by several techniques. Results of X-ray diffraction showed expansion of the inter-gallery distance, and transmission electron microscopy (TEM) micrographs confirmed that the prepared nanocomposite samples have intercalated and partially exfoliated structures. Cure characteristics showed that, organoclay not only accelerates the vulcanization reaction, but also gives rise to a marked increase of the torque, indicating crosslink density of the prepared compounds increases at the presence of organoclay. Mechanical properties of samples received markedly increase by clay loading due to the good interaction established between nanoclay particles and polymer matrix as it was evidenced by SEM photomicrographs. At the same time, rheological properties showed that addition of nanoclay could improve storage modulus as well as complex viscosity of SBR/EPDM samples. The results of ozone test revealed that the ozone resistance of samples significantly increases as nanoclay or EPDM content increases.

**Keywords:** Nanoclay; SBR/EPDM nanocomposite; Morphological properties; Mechanical and rheological measurements; TEM and SEM Photomicrographs.

### INTRODUCTION

In the last few decades the popularity of polymer composite materials has been increased tremendously in the materials industry<sup>[1, 2]</sup>. This is due to the remarkable properties, such as tensile strength, heat distortion temperature and modulus, exhibited by composites compared with unfilled or unreinforced polymers<sup>[3]</sup>. With these distinctive characteristics, it is not surprising that material scientists and polymer industries are still actively pursuing the related research interests today. Composite materials are basically made up of two distinct phases, the polymer matrix and the reinforcement. The reinforcement phase normally has a higher strength and modulus than the matrix, and is usually in the form of fibers or particles, which are also known as fillers. One of the most innovative and successful developments in composites is the discovery of nanocomposites<sup>[4–6]</sup>, in which at least one dimension of the dispersed particles is in the nanometer range<sup>[4]</sup>. These nano-sized reinforcing materials dramatically increase the surface area compared with conventional-sized reinforcement materials.

Polymer/clay nanocomposites were first discovered and developed by the Toyota research group<sup>[5]</sup>. For the inorganic filler phase, smectite clays are of particular interest as they exist in nature in abundance. The smectite clays are hydrophilic and can be broken down into platelets with a thickness of around 1 nm<sup>[5, 6]</sup>. The commonly used smectite clays are montmorillonite, hectorite and saponite, which belong to the family of structures known as 2:1 phyllosilicates (based on talc and mica).

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Smectite clays can also be modified with various organic chemicals to render them more hydrophobic, thus improving the compatibility between the clay and the hydrophobic polymers. In recent years, polymer/clay nanocomposites (PCNs), emerging as a new class of materials, have attracted unnegligible interest from academia and industry<sup>[7-34]</sup>. Compared with their relative micro and macro scale counterparts and their pristine polymer matrices, PCNs exhibit significantly improved performances such as tensile strength, modulus, heat resistance, gas barrier, swelling resistance and etc. All these properties could be achieved without a loss of clarity in the polymer. Since only a small amount of the silicate is needed to produce these desired characteristics, other than lightness, the PCNs can often be easily prepared using conventional processing methods. Very recently, increasing attention has been paid on rubber/clay nanocomposites (RCNs) as well as the development of nanocomposites using thermoplastics, the technology has been successfully applied to thermoset polymers<sup>[11, 12]</sup>. According to reports, several RCNs based on one or two elastomers have been prepared *via* solution intercalation, melt intercalation and rubber-latex compounding. For example, Alipour *et al.*<sup>[7, 10]</sup> showed that addition of nanoclay could significantly improve properties of NR/EPDM compounds. But to our knowledge, no studies have been published on (styrene-butadiene rubber/ethylene propylene diene monomer/organoclay) nanocomposites prepared by melt intercalation in the literature. Today, elastomer materials are popular in the polymer industry and are used widely, ranging from automotive to household applications. The development of rubber/clay nanocomposites would be a great advantage to the rubber industry due to the improved physical properties. Styrene butadiene rubber, a widely used rubber, is the first rubber considered for manufacturing rubber materials due to its low price. SBR, despite encompassing good mechanical properties especially abrasion resistance, is sensitive to the environmental factors such as ozone, light, moisture and heat due to the double bonds present in its molecular structure. This defect would be altered by blending SBR with highly saturated elastomers such as EPDM. Ethylene propylene diene monomer (EPDM) is an excellent all-purpose rubber that is useful in wide variety of applications. Due in part to the substantial absence of unsaturation in polymer backbone, EPDM rubbers exhibit superior oxidative and ozone resistance, weather resistance and heat resistance compared to conjugated diene rubbers. Nair *et al.*<sup>[22]</sup> showed that the highly ozone-resistant EPDM rubber effectively modified the weak SBR from ozone aging.

Furthermore, fillers are commonly added to rubber blends for their modifying effects on the tear and abrasion resistance, tensile strength and modulus. Size, structure and the aspect ratio of the filler affect processability and vulcanization of elastomers. SBR does not tolerate high levels of carbon black, showing a sharp decrease in its properties when even moderately high levels of filler are added to this elastomer<sup>[35]</sup>. So it is needed to reinforce SBR with low weight percentages of fillers. For this reason nanoclays are of interest, as they improve abrasion resistance, mechanical strength and heat distortion of rubbers even by the addition of just a low weight percentage (< 10 wt%). This is mainly attributed to the nanoscale dimension of silicate layers dispersed in the polymer matrix which causes a strong interfacial interaction between silicate layers and polymer chains, leading to a dramatic change in the thermal stability behavior, mechanical, dynamic-mechanical, barrier, optical properties and fire resistance compared to properties of their micro-counterparts or conventional filled polymers<sup>[10]</sup>.

In this study, poly(styrene-*co*-butadiene) rubber (SBR) and ethylene propylene diene monomer (EPDM)/clay nanocomposites were developed. It could be of great interest to use nanoparticles in SBR/EPDM blends and reinforce the compounds by addition low percentages of the filler. This involved the investigation of clay content and composition of matrix components on the cure characteristics, microstructure, mechanical and rheological properties of SBR/EPDM/Cloisite 20 A nanocomposites.

The blends of EPDM/SBR/organoclay developed by this study could be used in the automobile industry in the form of products such as windscreen channels, glass beadings, tire flaps, tire treads, and for electrical insulation.

## EXPERIMENTAL

### Material

SBR (1502), EPDM (KEP 270), nano-clay (Cloisite 20A) and the curing system (Zinc Oxide, Sulfur, Stearic Acid, MBTS and TMTD) were respectively supplied by Bandar Imam Petrochemical Complex (Iran), Korea Kumho Polychem, Southern Clay Ltd and Bayer Co., (see Table 1). The vulcanization ingredients including sulfur and mercapto-accelerators (MBTS and TMTD) are listed in Table 2.

**Table 1.** Material characteristics used in this research

Material	Characteristics		Units
SBR	Mooney viscosity ML (1 + 4) 100 °C	50	M
	Density	0.93	g/cm <sup>3</sup>
EPDM	Mooney viscosity ML (1 + 4) 125 °C	60	M
	Ethylene content	68	%
	Termonomer content	4.5	% ENB
	Density	0.86	g/cm <sup>3</sup>
Cloisite 20A	CEC	95	meq/100g
	Density	1.77	g/cm <sup>3</sup>

**Table 2.** The vulcanization ingredients

Ingredients	phr
Zinc oxide	4
Stearic acid	1.5
Sulfur	2.5
Tetramethyl thiuram disulfide(TMTD)	1
Benzothiazyl disulfide (MBTS)	1.25

### Preparation

Nanocomposite samples, after pre-drying of clay at 80 °C for 24 h, were prepared on a two-roll mill for 20 min, according to Table 3. First SBR, clay powder and EPDM were mixed and then curing system was added to the compound. The compositions of prepared samples are summarized in Table 3. Reference unfilled SBR/EPDM blend samples were also prepared for comparison purposes.

**Table 3.** Composition (wt%) and nomenclature of the nanocomposites and reference samples prepared

Sample code	SBR	EPDM	Clay	Sample code	SBR	EPDM	Clay
SEC <sub>1</sub>	60	40	1	SEC <sub>14</sub>	90	10	5
SEC <sub>2</sub>	70	30	1	SEC <sub>15</sub>	100	0	5
SEC <sub>3</sub>	80	20	1	SEC <sub>16</sub>	60	40	7
SEC <sub>4</sub>	90	10	1	SEC <sub>17</sub>	70	30	7
SEC <sub>5</sub>	100	0	1	SEC <sub>18</sub>	80	20	7
SEC <sub>6</sub>	60	40	3	SEC <sub>19</sub>	90	10	7
SEC <sub>7</sub>	70	30	3	SEC <sub>20</sub>	100	0	7
SEC <sub>8</sub>	80	20	3	Ref <sub>1</sub>	60	40	0
SEC <sub>9</sub>	90	10	3	Ref <sub>2</sub>	70	30	0
SEC <sub>10</sub>	100	0	3	Ref <sub>3</sub>	80	20	0
SEC <sub>11</sub>	60	40	5	Ref <sub>4</sub>	90	10	0
SEC <sub>12</sub>	70	30	5	Ref <sub>5</sub>	100	0	0
SEC <sub>13</sub>	80	20	5				

### Characterization

Curing process was carried out under hot press at 160 °C. Tensile properties of the compression molded samples were carried out according to ASTM D 412 by a Hiwa machine (Iran) at a cross head speed of 500 mm/min. The elongation at break, tensile strength and modulus are extracted from the stress-strain curves. Zwick Rheometer (ASTM D 2084-88) and Durometer (ASTM D 2240) were respectively used to study the cure characteristics and hardness of the compounds. Abrasion resistance tests of samples were performed by using a Zwick abrasion tester (ASTM D 5963). Resilience was studied using a Frank Tripsometer (France) according to ASTM D 1054.

X-ray diffraction patterns of the samples were recorded on a Philips model X'Pert (50 kV, 40 mA) by using Cu-K $\alpha$  radiation ( $\lambda = 0.1540598$  nm). The samples were sheets with a smooth surface and dimensions of 10 mm (width)  $\times$  10 mm (height)  $\times$  1 mm (thickness). The basal spacing of silicates was estimated and calculated on the basis of plane peak positions in the WAXD intensity profile using Bragg's law,  $d = \lambda / (2 \sin \theta_{\max})$ . Samples for SEM analysis were cryogenically fractured, then coated with gold and viewed by a TESCAN electron microscope. The nanostructure of the clay was observed by a transmission electron microscopy (Philips) with an accelerator voltage of 200 kV. A thin section of each specimen was prepared by using a cryogenic ultramicrotome. The rheological measurements were performed using a RPA 2000 oscillatory rheometer (Alpha Technology) at 80 °C and frequency of 0.01–1000 (rad/s). The gel content of the composites was determined by soxhlet extraction with toluene for 72 h. The extracted samples were vacuum dried to constant weight for 16 h. The gel content was calculated as the ratio of the weight of dried sample after extraction to the weight of the sample before extraction. Ozone resistance of the nano-blend vulcanizates was tested according to ASTM D 1149 using a Hampden (Northampton, England) model NN2 6EB ozone test chamber. The test specimens of 20 mm in width and 2 mm in thickness were clamped, elongated to 20% and tested under the following conditions: ozone concentration ( $50 \pm 5$ ) parts per hundred million, temperature ( $40 \pm 2$ ) °C, relative humidity ( $50 \pm 5$ )% for 96 h.

## RESULTS AND DISCUSSION

### Cure Characteristics

The curing characteristics, expressed in terms of the scorch time,  $t_5$ , optimum cure time,  $t_{90}$ , and torque value for the studied materials, are reported in Fig. 1, Fig. 2 and Table 4 respectively. It can be observed that the scorchtime and optimum cure time of SBR/EPDM samples were reduced when the organoclay was added to the elastomer, showing accelerated vulcanization with respect to that of pure SBR/EPDM. This can be attributed to the fact that the organoclay behaves as an effective vulcanizing agent for SBR/EPDM, leading a significant increase in the elastomer vulcanization rate<sup>[8, 10, 15]</sup>. In fact the amine groups present in the nanosilicate structure, which come from the organophilization of the clay, facilitate the curing reaction of nature rubber compound<sup>[16]</sup>. On the other hand, in a fixed clay content, cure time increases with EPDM content which could be ascribed to the lower double bond density in EPDM compared to SBR. This would lead to the longer vulcanization of EPDM with sulfur curing systems<sup>[8, 10]</sup>.

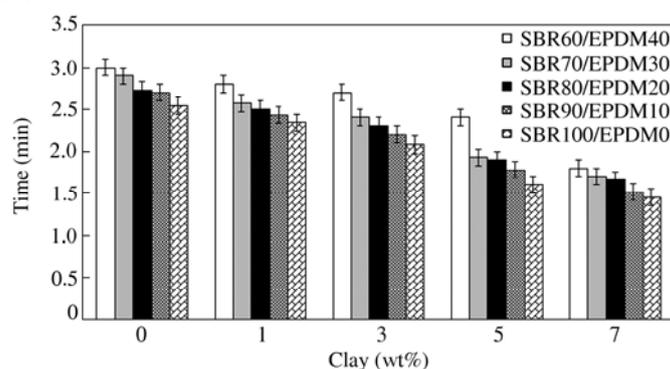


Fig. 1 Scorch time of the different compositions of SBR/EPDM containing nanoclay

Alongside this, the torque values, measured as the difference between the maximum and minimum torques were also evaluated as summarized in Table 4. It is of interest that the organoclay gives rise to a dramatic increase in the torque value of compounds when compared to pristine SBR/EPDM. Regarding the fact that the torque value is related to the crosslink density, it can be deduced that the organoclay certainly increases the crosslinking density of SBR/EPDM. These results are attributed to the intercalation of the elastomer chains between the silicate galleries, and consequently increasing the interlayer distance which facilitates the

incorporation of SBR/EPDM chains into the silicate galleries. This would provide a better interaction between filler and elastomer<sup>[7-10, 13, 15]</sup>. It is worth noting that in a fixed clay content, torque value increases by addition of EPDM to the compounds. In fact,  $M_H - M_L$  is a measure of the shear dynamic modulus. This may be attributed to the ethylene and propylene groups presented in EPDM<sup>[8]</sup>. Similar trends was observed by Muraleedharan Nair *et al*<sup>[23]</sup>.

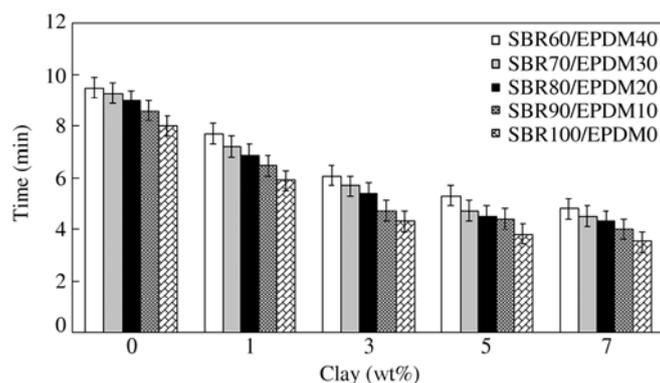


Fig. 2 Optimum cure time of the different compositions of SBR/EPDM containing nanoclay

Table 4. Torque values of the prepared samples

Sample code	Torque value = $M_H - M_L$ (Ibf.in)	Sample code	Torque value = $M_H - M_L$ (Ibf.in)
SEC <sub>1</sub>	65	SEC <sub>14</sub>	63
SEC <sub>2</sub>	60	SEC <sub>15</sub>	59
SEC <sub>3</sub>	57	SEC <sub>16</sub>	79
SEC <sub>4</sub>	56	SEC <sub>17</sub>	71
SEC <sub>5</sub>	53	SEC <sub>18</sub>	68
SEC <sub>6</sub>	70	SEC <sub>19</sub>	64
SEC <sub>7</sub>	65	SEC <sub>20</sub>	61
SEC <sub>8</sub>	63	Ref <sub>1</sub>	64
SEC <sub>9</sub>	61	Ref <sub>2</sub>	58
SEC <sub>10</sub>	58	Ref <sub>3</sub>	54
SEC <sub>11</sub>	75	Ref <sub>4</sub>	52
SEC <sub>12</sub>	69	Ref <sub>5</sub>	50
SEC <sub>13</sub>	66		

### Gel Content

In order to study the efficiency of nanoclay content on the compounds, the gel content of unfilled and filled SBR/EPDM (60/40) samples were determined, and the results are plotted in Fig. 3. The figure shows that the gel contents of SBR/EPDM samples change with the increase of clay content and slightly decrease. These results indicate that the presence of nanostructured silicate layers of Cloisite 20A in SBR/EPDM affect significantly the formation of the crosslinking network. The addition of clay results in the formation of some physical crosslinks which may increase the vulcanization rate and some mechanical properties but not contribute the gel content.

### XRD Results

XRD patterns of the SBR/EPDM (60/40) samples containing 1 wt%, 3 wt%, 5 wt% and 7 wt% organoclay are typically shown in Fig. 4. As we know, organoclay (Cloisite 20A) presents a peak at  $2\theta = 3.52^\circ$ , which as deduced by the Bragg equation corresponds to an interlayer distance of 2.42 nm. Shift of the organoclay characteristic peak to lower  $2\theta$  values (see Fig. 4) indicates that elastomer chains have intercalated between silicate layers. This clearly shows that inter lamellar spacing of the clay would enlarge after melt compounding. As seen, the intensity of the peaks increases by clay loading. Broad peaks with lower intensities are usually indicative of more intensive intercalations or even partial exfoliations<sup>[7, 8, 10]</sup>.

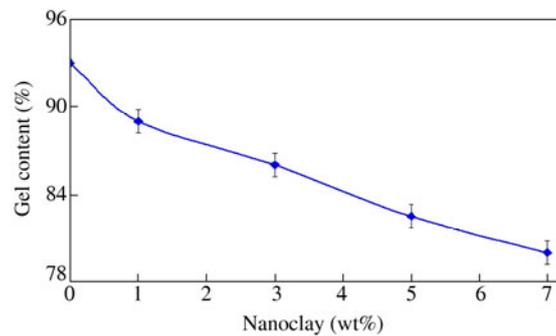


Fig. 3 Gel content values of SBR/EPDM (60/40) containing nanoclay

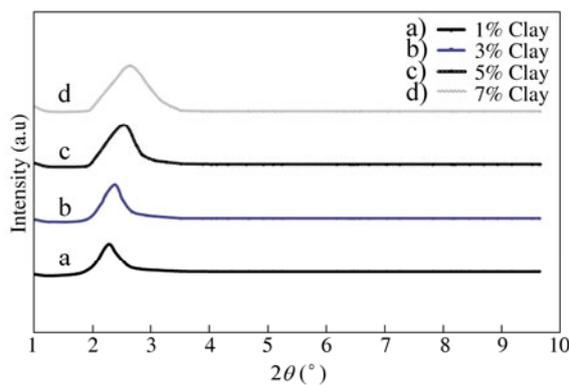


Fig. 4 XRD patterns of SBR 60/EPDM 40 containing 1 wt%, 3 wt%, 5 wt% and 7 wt% cloisite 20A

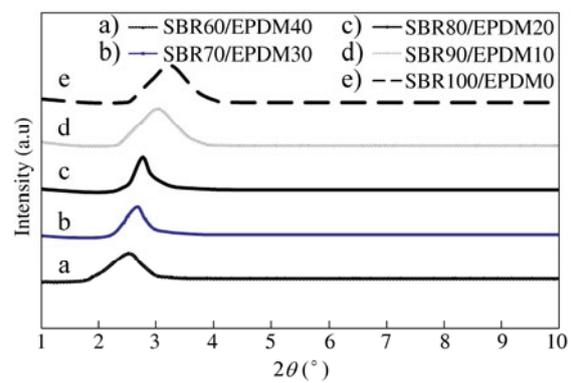


Fig. 5 XRD patterns of different compositions of SBR/EPDM containing 5 wt% nanoclay

Moreover, XRD results related to the different compositions of SBR/EPDM containing 5 wt% organoclay are shown in Fig. 5. It can be seen that as the EPDM content within the samples increases, the organoclay diffraction peak shifts toward lower angles along with the peak weakening. It can be attributed to the higher Mooney viscosity of EPDM resulting in a higher shear stress undergone by the matrix which might help the deagglomeration of the clay stocks within the elastomer matrix as evidenced by Alipour *et al.*<sup>[7, 10]</sup>.

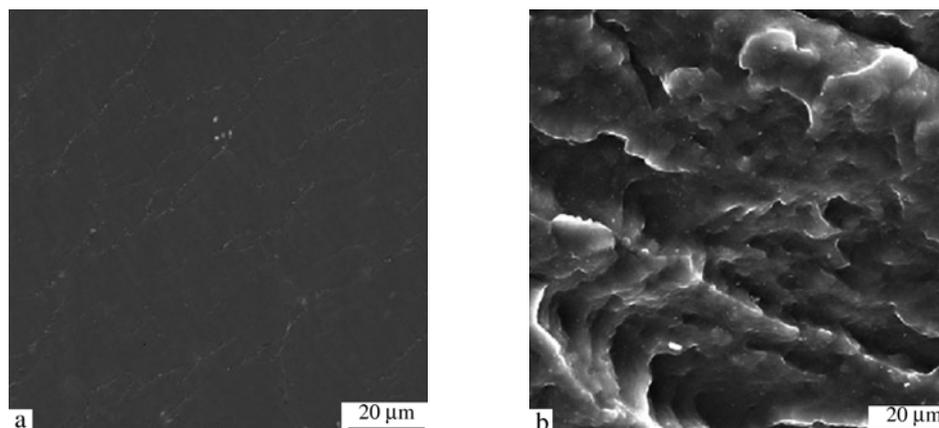


Fig. 6 SEM photomicrographs of SBR/EPDM (60/40): (a) without nanoclay and (b) 7 wt% nanoclay

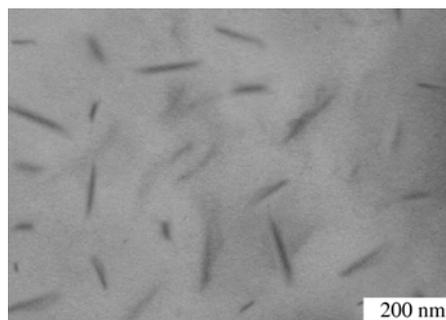
### Microscopy

Further insights on the morphology of these materials are found in Fig. 6, where SEM images of cryogenically fractured samples based on SBR/EPDM (60/40) with and without clay are reported. It is clearly seen that the

nanocomposite sample containing 7 wt% Cloisite 20A (Fig. 6b) compared to the pristine SBR/EPDM (60/40) (Fig. 6a) have a rough fracture surface implying a well established interaction between polymer chains and nanoclay particles<sup>[10]</sup>.

#### **TEM Results**

Figure 7 shows the TEM images of cryogenically fractured surfaces of SBR/EPDM (60/40) sample containing 5 wt% nanoclay in which dark lines represent the Cloisite layers dispersed within the matrix. The micrograph implies the intercalation and partial exfoliation of the clay through the polymer matrix, evidenced by XRD patterns.



**Fig. 7** TEM image of SBR 60/EPDM 40/nanoclay 5

#### **Mechanical Properties**

As a further characterization of the SBR/EPDM nanocomposites, mechanical properties of samples are reported in Table 5. Regarding to the obtained results, it can be deduced that nanoclay would enhance tensile strength, elongation at break, hardness and abrasion resistance of SBR/EPDM nanocomposites. Improvement of tensile strength and hardness in case of polymer-clay nanocomposites has also been reported by some researchers<sup>[10, 13–15]</sup>. They suggested that the increase of strength and hardness is directly related to the degree of dispersion of clay layers within the polymer matrix. The largely increased reinforcement of the nanocomposites should be ascribed to the dispersed structure of clay at the nano level, the high aspect ratio and the planar orientation of the silicate layers<sup>[18]</sup>. The reinforcement is also associated with the anisotropy and high aspect ratio of organoclay nanofillers, which act as short reinforcing fibers with nanoscale architecture. Clay with high aspect ratio is more efficient in restricting the rubber chains and in resisting the development of cracks than spherical fillers<sup>[10]</sup>.

Hardness is an indication of the relative stiffness of the material. Fillers are known to increase hardness provided the hardness of the filler is higher than that of the polymer matrix. 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 due to the chemical nature of the polymer. From Table 5 it can be seen that nanoclay improved the stiffness of the rubber blends. This could be ascribed to the huge surface area of clay dispersed at nanometer level and the largest aspect ratio of silicate layers, which results in the increased silicate layer networking<sup>[10]</sup>.

Also, incorporation of clay into the polymer matrix was found to enhance elongation at break of nanocomposite samples. Usually, at favorable matrix/organoclay interactions and relatively low organoclay contents, both tensile strength and elongation at break increase<sup>[10, 16–20]</sup>.

The effect of clay loading and matrix composition on the resilience of the blends is seen in Table 5. All of the SBR/EPDM filled samples exhibited similar trends; rebound resilience of the filled blends decreased with filler loading. The decreasing trend 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<sup>[23]</sup>. The surface activity is an important factor, indicating the extent of rubber-filler

interaction. According to Jacques<sup>[24]</sup> the incorporation of most of the particulate fillers into rubber leads to an increase in hardness and a reduction in resilience, particularly with more reinforcing filler.

**Table 5.** Mechanical properties of the prepared samples

Sample (SBR/EPDM/clay) dimensions	Tensile strength (MPa)	Elongation at break (%)	Hardness (Shore A)	Resilience (%)	Abrasion loss (mm <sup>3</sup> )
60/40/0	1.47 ± 0.0100	100 ± 2.20	62.5 ± 0.100	74.5 ± 0.0400	85.0 ± 1.20
70/30/0	1.33 ± 0.0100	75.0 ± 3.88	60.2 ± 0.100	71.6 ± 0.0100	90.0 ± 1.12
80/20/0	1.17 ± 0.0200	70.0 ± 1.55	58.7 ± 0.200	69.5 ± 0.0700	100 ± 2.64
90/10/0	1.01 ± 0.0100	58.0 ± 2.01	56.0 ± 0.100	67.5 ± 0.0200	120 ± 2.32
100/0/0	0.820 ± 0.0200	50.8 ± 1.24	55.8 ± 0.200	65.0 ± 0.0100	150 ± 2.12
60/40/1	1.72 ± 0.0200	105 ± 2.20	63.0 ± 0.100	74.2 ± 0.0300	75.0 ± 1.11
70/30/1	1.54 ± 0.0100	87.0 ± 4.81	61.0 ± 0.200	71.4 ± 0.0700	80.0 ± 1.33
80/20/1	1.39 ± 0.0200	80.0 ± 0.98	59.0 ± 0.100	69.2 ± 0.0100	90.0 ± 2.75
90/10/1	1.23 ± 0.0100	72.0 ± 2.69	56.3 ± 0.100	67.0 ± 0.0900	110 ± 2.65
100/0/1	1.08 ± 0.0200	56.0 ± 1.11	56.0 ± 0.100	64.5 ± 0.0800	140 ± 2.97
60/40/3	2.01 ± 0.0300	113 ± 5.2	63.2 ± 0.100	74.0 ± 0.0500	65.0 ± 1.98
70/30/3	1.80 ± 0.0300	99.0 ± 2.99	61.2 ± 0.100	71.2 ± .0200	72.0 ± 1.89
80/20/3	1.64 ± 0.0200	92.0 ± 2.18	59.5 ± 0.100	69.0 ± 0.0100	83.0 ± 1.31
90/10/3	1.40 ± 0.0100	79.0 ± 1.54	57.0 ± 0.200	66.0 ± 0.0100	95.0 ± 3.12
100/0/3	1.33 ± 0.0200	67.0 ± 1.43	56.5 ± 0.100	64.0 ± 0.0300	132 ± 3.22
60/40/5	2.34 ± 0.0100	130 ± 3.31	63.5 ± 0.100	73.0 ± 0.0500	60.0 ± 1.84
70/30/5	2.20 ± 0.0100	115 ± 2.51	61.5 ± 0.200	71.0 ± 0.0800	66.3 ± 1.07
80/20/5	1.85 ± 0.0100	105 ± 3.28	59.7 ± 0.200	68.0 ± 0.0200	77.1 ± 1.11
90/10/5	1.60 ± 0.0200	86.0 ± 1.98	57.0 ± 0.100	65.5 ± 0.0300	85.0 ± 1.10
100/0/5	1.50 ± 0.0200	72.0 ± 5.45	56.8 ± 0.100	63.0 ± .0100	120 ± 3.20
60/40/7	2.51 ± 0.0200	153 ± 3.57	64.0 ± 0.200	71.0 ± 0.0800	57.0 ± 1.02
70/30/7	2.35 ± 0.0200	132 ± 2.23	62.0 ± 0.200	69.0 ± 0.0400	61.2 ± 1.01
80/20/7	2.20 ± 0.0300	115 ± 3.33	60.0 ± 0.200	67.0 ± 0.0700	70.0 ± 1.02
90/10/7	1.98 ± 0.0100	100 ± 2.74	58.0 ± 0.200	65.0 ± 0.0600	80.0 ± 1.03
100/0/7	1.80 ± 0.0200	85.0 ± 3.28	57.0 ± 0.200	62.0 ± 0.0500	105 ± 3.33

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 a rubbing, scraping, or erosive nature. The incorporation of nanoclay reduced the abrasion loss of SBR/EPDM samples notably. Reinforcing fillers, such as nanoclay, interact preferentially with the rubber phase, as shown by the higher reduction of abrasion loss in the samples. This improvement is probably due to the greater surface area and better filler-rubber interfacial adhesion resulting in an improved abrasion resistance<sup>[10]</sup>. Fine particles actually reflect their greater interface between the filler and the rubber matrix and, hence, provide a better abrasion resistance and adhesion than the coarse ones. Similar results were also reported by other researchers<sup>[10, 26, 27]</sup>.

At fixed clay contents, tensile strength is observed to increase with EPDM. This behavior might be caused by the orientation of crystalline regions of EPDM under the applied stress<sup>[22]</sup>. Furthermore, hardness, elongation at break and abrasion resistance of the prepared samples increase with EPDM content. In this study, the obtained results are in concordance with the general tendency reported in the literatures<sup>[22]</sup>.

### Rheological Properties

Figure 8 shows the elastic modulus ( $G'$ ) and complex viscosities ( $\eta^*$ ) of SBR/EPDM (60/40) containing 0 wt%, 1 wt%, 3 wt%, 5 wt% and 7 wt% nanoclay as a function of angular frequency. It can be seen that there is a noticeable improvement in rheological properties of the polymer with addition of clay. In other words, incorporation of nanoclay results in the formation of network structure and establishment of good interaction between polymer/clay which is the main reason for enhanced complex viscosity and elastic modulus<sup>[9, 10]</sup>. In addition, viscosity of samples declines as angular frequency increases. It can be attributed to the pseudoplastic nature and shear-thinning behavior of the prepared sample<sup>[28]</sup>. The remarkable rise of complex viscosity at low

frequencies could be compared to the materials exhibiting a yield stress indicating that a yield stress may be present for nanocomposites compared to the pure matrix<sup>[26]</sup>.

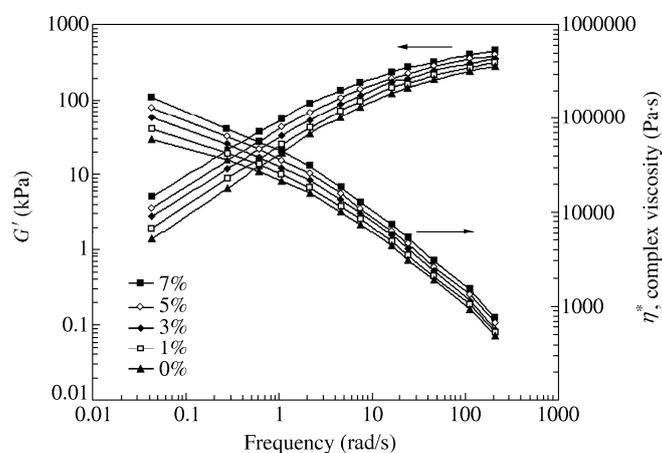


Fig. 8 Rheological properties of SBR/EPDM (60/40) nanocomposite samples

### Ozone Resistance

The interaction of rubber with ozone is best noted when the rubber is stressed or stretched in use. The ozone resistance times of the nanocomposite samples, defined as the duration until the first cracks are observed manually in the sample, are reported in Table 6. It can be seen that the ozone resistance of the samples are improved by increasing nanoclay content. Clay platelets, hindering the penetration of environmental gasses into the polymer bulk would increase the ozone resistance of nanocomposite samples<sup>[10]</sup>. Generally, the incorporation of clay into the polymer matrix is found to enhance thermal stability by acting as a superior insulator and mass transport barrier to the volatile products generated during decomposition. It is worth noting that it is necessary to add just 5 wt% nanoclay to the SBR to reach the ozone resistance even more than that of induced by addition of 40 wt% EPDM. (Compare samples (60/40/0 and 100/0/5) (SBR/EPDM/nanoclay)).

Table 6. Effect of EPDM and nanoclay contents on the ozone resistance of SBR/EPDM/nanoclay nanocomposites

Composition (SBR/EPDM/nanoclay)	Ozone resistance time (h)	Composition (SBR/EPDM/nanoclay)	Ozone resistance time (h)
60/40/0	46	100/0/5	48
60/40/1	49	90/10/5	52
60/40/3	55	80/20/5	63
60/40/5	No crack after 72 h	70/30/5	68
60/40/7	No crack after 72 h		

### CONCLUSIONS

In this article SBR/EPDM/Cloisite 20A nanocomposites were prepared by two-roll mill. The XRD patterns and TEM photomicrographs showed that polymer chains could penetrate into silicate layers and form an intercalated and partially exfoliated structure. The cure characteristics confirmed that nanoclay acted as an accelerating and reinforcing agent for SBR/EPDM chains. Also, an increase in torque value was obtained for the organoclay nanocomposites, indicating a higher number of crosslinks formed.

From obtained results it was noticed that strong interactions between polymer/nanoclay resulted in significantly improved mechanical and rheological properties. There were noticeable enhancements in tensile strength, hardness, abrasion resistance as well as complex viscosity and storage modulus. The incorporation of clay into the polymer matrix was found to enhance ozone resistance. Furthermore, it was concluded that matrix composition has great effect on the properties of samples. In a fixed nanoclay content, the more EPDM content led to an increase in scorch time and optimum cure time. Moreover the torque values, tensile strength, hardness,

elongation at break and ozone resistance of the samples received marked enhancement as EPDM content of samples increased. The novel nanocomposites prepared in this study exhibited excellent properties. The results and findings obtained, provide some useful achievements. Based on the noticed results, it is needed to use just a low weight percentage of nanoclay to reinforce SBR/EPDM blends. The results of this work indicate that the use of organoclay as reinforcing agent to the SBR/EPDM results in improved properties for potential use in wide range of applications.

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