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# Studies on Ginger Powder and Crysnanoclay Filled Acrylonitrile-Butadiene Rubber/Styrene Butadiene Rubber (NBR/SBR) Green Composites

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**Abstract:** The development of commercially viable "green products" based on natural resources for both matrices and reinforcements for a wide range of applications is on the rise. This effort includes new pathways to produce natural polymers with better mechanical properties and thermal stability using nanotechnology and use of naturally occurring fillers such as lignocellulosic and hemicellulose to make biodegradable rubber composites. The blends of acrylonitrile butadiene rubber (NBR) and styrene butadiene rubber (SBR) with ginger powder (GP) have been prepared by using a compounding technique in presence of different amounts of crysnanoclay (CN). The physico-mechanical properties, such as tensile strength, modulus at 100% and 200% elongation at break and hardness of the composites before and after heat ageing have been studied. After heat ageing most of the properties improved. A markedly improvement or retain in tensile properties (strength and modulus) after incorporation of CN the water sorption tended to decrease. The dispersion of GP and CN in rubber matrix was analysed from scanning electron microscopy (SEM).

Keywords: NBR/SBR; crysnanoclay; ginger powder; mechanical properties; thermal stability.

## Introduction

In recent years there has been an extensive research work on development of newer materials with high performance at affordable costs. With growing environmental awareness, this search has particularly focused on eco-friendly materials, with terms such as "renewable", "recyclable", "sustainable" and "triggered biodegradable" becoming buzzwords. This underscores the emergence of a new type of materials, a change from non-renewable, but difficult to degrade or non-degradable, to renewable and easily degradable materials. The development or selection of a material to meet the desired structural and design requirements calls for a compromise between conflicting objectives. This can be overcome by resorting to multi objective optimization in material design and selection. Composite materials, which are prepared using natural or synthetic reinforcements and a variety of matrix materials, are included in this philosophy.

The objective of the incorporation of fillers in polymers is the improvement of physical, thermal and mechanical properties, as well as the modification of processing characteristics and cost reduction of the final product [1]. Some types of fillers and processing aids that are either synthetic or naturally occurred as minerals, metallic powders, organic or inorganic compounds, possessing a wide range of particle sizes and geometric shapes [2]. In the rubber industry the use of fillers alone or in combination, is widely accepted because of the several advantages they present. Carbon black (CB) is the main reinforcing agent for elastomeric vulcanizates and is also used in mixtures with other fillers [3]. The reinforcement given by CB brings a segmental immobilization of the polymer chains. The modification of the elastomer by means of reinforcing agents and vulcanization gives rise to three dimensional networks which then may turn a flexible, highly deformable rubber into a material with large elastic recovery [4].

Polymer composites reinforced with bio-based materials have raised great interest among material scientists and engineers due to the consideration of developing eco-friendly composites and partly replacing mineral fillers [5]. Natural fillers are widely used due to their inherent advantages such as low cost, low density, high stiffness and high specific strength [6]. Also, the demand for lignocellulosic sources with low cost is increasing in the composite industries which can decrease overall manufacturing costs and increase stiffness of the materials [7]. Some drawbacks, such as the incompatibility between hydrophilic natural filler and hydrophobic polymer matrices, the tendency to form aggregates during processing, poor resistance to moisture and reduction in mechanical properties of the composites with filler content [8, 9], reduce the use of natural fillers as reinforcements in hydrophobic polymers. Polymer/clay nanocomposites are of interest for both scientific challenges and industrial applications [10-12]. Polymer/nanoclay nano-composites with a relatively low loading of the filler have superior mechanical properties, thermal stability, flame retardancy and gas barrier properties [13-16]. These improvements in the properties are the result of the nanometer scale dispersion of clay minerals in the polymer matrix. Nanocomposites with completely exfoliated clay mineral particles are desired as the exfoliated particles exhibit the greatest reinforcement [17]. To make rubber compatible with fillers and obtain a high degree of dispersion, the clay minerals are modified with organic compounds. The SBR/NBR systems are widely used in automobile industries. Hence, in this research article an attempt has been made to utilize agro-industrial waste to prepare green composites which has little or no commercial value.

## **Experimental**

#### Materials

The elastomer used for preparing nanocomposites was SBR Kosyn 1502; styrene content is 23 wt. %; (random copolymer) and specific gravity is 0.945 was obtained from Kumpho Petrochemicals, Korea. The NBR – JSR N230 SL (acrylonitrile content ~ 32 % with specific gravity =  $1.17 \pm 0.005$ ) was obtained from JSR Co., Japan. Ginger powder (GP) is similar to hard woods in chemical composition though the lignin content is higher (35–45%) and the cellulose content is lower (23–43%). The GP (particle size: 30-40 µm) was obtained from local industry. The Crysnano 1030 is a natural montmorillonite mineral modified with quaternary ammonium salt. The typical properties of crysnano 1030 are; d-value - 19 nm and specific gravity - 1.97 at 25°C. It was obtained from Talegaon Dabhade, Pune, India. The NBR/SBR blends were formulated with dibenzothiazole disulfide (MBTS), zine dimethyl dithiocarbamate (ZMDC), zinc oxide, stearic acid, sulfur and carbon black (N330).

#### Fabrication of the rubber nanocomposites

Acrylonitrile butadiene rubber/styrene butadiene rubber (NBR/SBR)/GP/CN nanocomposites were compounded by mixing the rubber with zinc oxide (5 phr), stearic acid (2 phr), agerite stalite (1 phr), and clay and ginger powder in a Brabender mixer (Plasti-Corder PL 2000) for 10 min at room temperature. The SBR compound was then milled in a 2-roll mill (Farrel) at room temperature with the vulcanizing agents: sulfur (1.5 phr), MBTS (dibenzothiazole disulphide) (1 phr) and cummate (0.5 phr). The vulcanization was achieved by compression moulding for 20 min. The typical formulation along with sample code is given in Table 1.

#### **Mechanical properties**

Dumb-bell shaped samples were cut from the molded sheets and the tensile testing procedure was done according to ISO 37. The tensile test was performed at a crosshead speed of 500 mm/min using an Instron 3366. The hardness measurements of the samples were done according to ISO 48 using a Wallace dead load instrument with a hardness range from 30 to 85 IRHD (international rubber hardness degree).

	Sample code				
Formulation (phr)	Α	B	С	D	Ε
SBR+NBR (50/50)	100	100	100	100	100
Carbon black	40	40	40	40	40
Nanoclay	0	0	2.5	5	10
Ginger powder	0	5	5	5	5
Sulphur	1.5	1.5	1.5	1.5	1.5
MBTS	1.5	1.5	1.5	1.5	1.5
ZMDC	0.5	0.5	0.5	0.5	0.5
Zinc Oxide	5	5	5	5	5
Stearic acid	1.0	1.0	1.0	1.0	1.0
Anti oxidant	1.5	1.5	1.5	1.5	1.5

Table 1. Sample formulations and sample code for (NBR/SBR)/GP/CN nanocomposites

#### **Compression set measurement**

Compression set test (ASTM D395) was performed on standard test specimen of cylindrical shape of  $25 \pm 0.1$  mm diameter and  $12 \pm 0.5$  mm thicknesses, vulcanized by compression mold. The test specimen shall be placed between the plates of the compression device with the spacers on each side of it, allowing sufficient clearance for bulging of the rubber when compressed. The bolts shall be tightened so that the plates are drawn together uniformly until they are in contact with the spacers. The percentage of the compression employed shall be 25% of the original thickness. Then the assembled compression device shall be placed in oven at 70 °C for 22 h. After completion of the assembly and remain in dry air circulated oven for specific period at the test temperature, the device shall be taken out from the oven and the test specimen removed immediately and allowed to cool for 30 min, after this the final thickness shall be measured by an electronic digital caliber with 0.01 mm accuracy. The compression set is defined as:

$$C\% = \frac{t_0 - t_1}{t_0 - t_1} \times 100$$
(1)

where,  $t_0$  is the original thickness of the sample;  $t_1$  is the thickness of the sample after removed from the clamp; and  $t_s$  is the thickness of the spacer bar used.

#### Swelling behavior

Procedure to measure percent swelling of composites in organic solvents is briefly explained as follows: known weights  $(w_1)$  of dried nanocomposites were immersed in different solvents until a state of equilibrium was attained at room temperature. When material swells, weight of the swollen material is noted  $(w_2)$ . The percentage of swelling was calculated by the relation;

Percentage swelling = 
$$\frac{W_2 - W_1}{W_1} \times 100$$
 (2)

#### Fourier transforms infrared (FT-IR) spectroscopy

The FTIR spectra were obtained using Perkin–Elmer spectrum and the attenuated total reflection (ATR) technique was adopted. The selected spectrum resolution and the scanning range were 4 cm<sup>-1</sup> and 600–4000 cm<sup>-1</sup>, respectively.

#### Scanning electron microscopy (SEM)

Scanning electron microscopy (SEM), model Zeiss SUPRA 35VP was used to analyze the surface aspects concerning the quality of bonding and to detect the presence of micro-defects, if any. Samples were mounted on aluminium stubs and the surface sputter coated with a thin layer of gold, about 20 mm thick, prior to scanning to avoid electrostatic charging and poor resolution during examination.

## **Results and Discussion**

#### **Physico-mechanical properties**

Incompatible elastomeric blends generally exhibit poor mechanical properties [18] due to inhomogeneous mixing and phase separation. Figures 1 - 6 represent the physico-mechanical properties before and after heat aging such as tensile strength, elongation at break, Young's modulus (100 % elongation and 200 % elongation), density and hardness for NBR/SBR (50/50) blends without and with GP and crysnanoclay, respectively.

## Density

The (NBR/SBR)/GP/CN had an average density of 1.142 and 1.168 g/cc respectively. The measured density values for all (NBR/SBR)/GP/CN nanocomposites are shown in Figure 1. From the figure 1, it can be noticed that, the density of the CN filled (NBR/SBR)/GP/CN nanocomposites increases as increase in CN content as expected. This is due to increase in high dense filler in low dense NBR/SBR matrix.



Figure 1. The bar chart of density as a function of GP and nanofiller content for (NBR/SBR)/GP/CN nanocomposites

#### Surface hardness before and after heat ageing

Hardness is defined as the resistance of a material to deformation, particularly permanent deformation, indentation, or scratching. The experimentally obtained surface hardness values are shown in Figure 2. The surface hardness of CN filled (NBR/SBR)/GP/CN nanocomposites lies in the range 64 - 73 IRH. The surface hardness value for NBR/SBR blend is 64 IRH and for 10% CN filled (NBR/SBR)/GP/CN nanocomposite is 73 IRH. From the Figure 2, it can be seen that hardness of nanocomposites and green composite after heat ageing in air (120°C/72 h) increased around 3 and 5 units. It was observed that a significant improvement in surface hardness of the nanocomposites as increase in rigid CN content in rubber blend as expected. This is because crysnanoclay is inorganic and rigid filler. The incorporated filler interlocking between rubber networks and reduces the flexibility of the rubber, which led to improvement in dimensional stability of the composites. This result clearly indicates that the filled nanocomposite has good dimensional stability than the unfilled rubber. Finally, the hardness of (NBR/SBR)/GP/CN nanocomposites was much superior to the rubber blend as shown in Figure 2. This data is also slightly higher to the literature data reported by Wang and co-workers [19]. The effect of heat ageing on the surface hardness behaviors was also shown in Figure 2. From Figure 2 it was noticed that after ageing hardness of nanocomposites increased may be due to increase in physical interaction of the rubber.



Figure 2. Surface hardness results of NBR/SBR and its nanocomposites

#### **Tensile behavior**

Mechanical properties of composites play a pivotal role in most of the load bearing applications. In recent years, demand for materials that are stiffer and stronger, yet lighter is increasing which can be used in aeronautics, civil engineering and in various structural applications. Interest has grown in recent years to use agro residues for proactive system of land filling management, which offers an advantage of being lighter weight along with comparable mechanical properties. The measured tensile properties of (NBR/SBR)/ GP/ CN nanocomposites are presented in Figure 3. From the Figure 3, it was found that the tensile strength of NBR/SBR was 16.28 MPa and that of green composite and nanocomposites were lies in the range 15.26 - 16.0 MPa. The GP acts as flaws in elastomeric phase. The incorporation of GP leading to initial reduction followed by retains in tensile strength with the addition of nanoclay and similar properties have been reported in the literature [20]. An increase in property may be due to the better stress transformation between matrix and fillers. Poorly dispersed fillers can easily agglomerates and obstruct stress propagation. On the other hand, well dispersed fillers prevent the obstruction of stress propagation, thus allowing the stress to be well propagated.

The incorporation of crysnanoclay in NBR/SBR/GP improved the tensile properties, due to exfoliated dispersions and also due to Vander wall's force of attraction between rubber matrix and nanoclay. The (NBR/SBR)/GP/CN nanocomposites showed a noticeable reduction percentage elongation at break as in Figure 4. The steep decline in elongation on filler addition is obvious, because lignocellulosic fillers have low elongation at break and restrict the polymer molecules flowing past one another. This behavior is typical of reinforced thermoplastics and has been reported by many researchers [21].



Figure 3. Tensile strength values of the NBR/SBR (50/50) blend and (NBR/SBR)/GP/CN nanocomposites

Figures 5 - 6 demonstrated significant increase in modulus at 100% and 200% elongation as a function of clay content as compared to reference sample. Composites prepared with GP results the tensile strength however, enhances the tensile modulus to a larger extent. Many researchers have reported a similar kind of increase in tensile modulus with the increase in filler content [22]. The strong interaction between the crysnanoclay layers and rubber chains increased the constraint of the motion of the rubber chains.

As the clay content increases, the elongation appears to reach plateau clay. There is a strong matrixclay interaction; since the chemical bond of the matrix to the filler reduces chain slipping along the filler surface. The largely increased mechanical properties are caused by good dispersion of the nanoclay layers with the high aspect ratio and parallel orientation [23].



Figure 4. Elongation at break values of the NBR/SBR (50/50) blend and (NBR/SBR)/GP/CN nanocomposites

## Effect of heat ageing on tensile properties

The effect of heat aging at 100  $^{\circ}$ C for 72 h on the tensile properties of nanoclay and ginger powder filled NBR/SBR nanocomposites has been studied. Figures 5 and 6 showed the tensile modulus at 100 % elongation and tensile modulus at 200 % elongation increased after CN and GP loading into rubber matrix. But tensile strength of nanocomposites and green composite is almost same.



Figure 5. Young's modulus at 100 % elongation at break of the NBR/SBR (50/50) blend and (NBR/SBR)/GP/CN nanocomposites



Figure 6. Young's modulus at 200 % elongation at break of the NBR/SBR (50/50) blend and (NBR/SBR)/GP/CN nanocomposites



Figure 7. Modulus at 100 % elongation and modulus at 200 % elongation for (NBR/SBR)/GP/CN nanocomposites as a function of clay content

From these figures it was noticed that after heat aging there was a significant reduction in elongation at break for nanocomposites due to cross-linking effect. The % retain in elongation at break lies in the range 84.7 - 93.1. From Figure 3 and Figures 5-6 it was noticed that tensile strength and tensile modulus increases after heat aging. This is due to increase in crosslink density and increase in physical interaction between elastomer and CN. Figure 7 is revealed that the modulus at 200 % elongation is higher than the modulus at 100 % elongation may be due to cross-linking effect.

#### **Compression set study**

To determine the effect of GP and CN loading on the compression set behavior of NBR/SBR and (NBR/SBR)/GP/CN nanocomposites, the compression set test is carried out. The percentage of compression set of nanocomposites is shown in Figure 8. The compression set decreases after incorporation of GP. Furthermore, increase in nanoclay retains or improvement in compression set was noticed. From this figure it is clear that in the case of SN9 the compression set is high, at higher loading of nanoclay in rubber matrix. This is because as CN loading increases the physical interaction between rubber-filler increases and the mobility of the rubber chains decreases, consequently, induces the stiffness in the resultant compounds [24].



Figure 8. Compression set study of (NBR/SBR)/GP/CN nanocomposites

#### Fourier transforms infrared spectroscopy

The IR spectra of the NBR/SBR blend and (NBR/SBR)/GP/CN nanocomposites are shown in Figure 9. FTIR spectra indicates that, the peak at 3730–3400 cm<sup>-1</sup> which may be due to the stretching peak of -OH bond of stearic acid, but it has been become broad after incorporating GP and CN due to overlapping of the –OH groups of lignin, cellulose and Si- OH groups band. The broad and strong band ranging from 3000 to 3600 cm<sup>-1</sup> indicates the overlapping of Si–OH (silanol) and R–OH (hydroxyl) groups of GP stretching vibrations. The peak visible at 2913 cm<sup>-1</sup> is due to the symmetric stretching of the C–H band. The peaks observed at 2922 and 2851 cm<sup>-1</sup> can be assigned to asymmetric and symmetric – CH<sub>2</sub> groups, respectively. The peak at 2325 cm<sup>-1</sup> is due to the C–H band of SBR and at 2260–2240 cm<sup>-1</sup> is due to nitrile linkages of the blend. The peak at 1600 cm<sup>-1</sup> is due to the c = C band of butaliene in the system. The peak at 1636 cm<sup>-1</sup> corresponds to the C = C stretching that can be attributed to the aromatic group and the peak at 1021 cm<sup>-1</sup> is may be due to the styrene [25]. There is no peak at 1730 cm<sup>-1</sup> (> C=O stretching peak), which indicated that the oxidation of main polymeric chain did not occurred at the time of rubber milling with the help of cracker cum mixing mill at high temperature. The wave numbers from 1400 to 1600 cm<sup>-1</sup> are assigned to aromatic skeletal vibration.



Figure 9. FTIR spectra of NBR/SBR blend and its nanocomposites

#### Water sorption

The hydrophilic groups in the cellulose and hemicelluloses of the ginger powder might have been exposed, which resulted in an easy integration with water. From the Figure 10 it was noticed that NBR/SBR blend with along GP and CN has low water absorption value, because it is hydrophilic in nature. After, incorporation of 5 wt % GP water uptake by composites increases drastically as expected. Furthermore increase in nanoclay content in rubber matrix, a slight reduction in water uptake behaviour of composites was noticed. Therefore, moisture content of the specimen depends on the amount and nature of fillers present. Most natural materials are hydrophilic in nature which easily influences the material for water absorption. The low water absorption of rubber prevents wrinkling and distortion due to changes in humidity.



Figure 10. Effect of ginger powder and crysnanoclay on water absorption of (NBR/SBR)/GP/CN nanocomposites

#### Swelling behavior

The solvent uptake curves for different compositions of (NBR/SBR)/GP/CN composites in aliphatic solvents (CHCl<sub>3</sub>, CH<sub>2</sub>Cl<sub>2</sub>, and CCl<sub>4</sub>) and aromatic solvents (benzene, toluene and xylene) are given in Figure 11. All the diffusion curves are similar in nature. It is very clear that the rate of swelling is high at the initial stages. The mole percent solvent uptake increases linearly with time. From the figures it was observed that the percent of swelling is different for different solvents. It is interesting to note that the highest maximum uptake is shown by SN1 sample and the lowest by SN9 sample, as in the case of optimum cured samples. The swelling behaviors of aromatic hydrocarbons were different as compared to aliphatic hydrocarbons. The chemical resistance of NBR/SBR systems increases after incorporation of crysnanoclay. The order of swelling behavior of (NBR/SBR)/GP/CN nanocomposites is as follows; CHCl<sub>3</sub> > CH<sub>2</sub>Cl<sub>2</sub> > CCl<sub>4</sub> in aliphatic and in aromatic; benzene > toluene > xylene. This is due to polarity, solubility parameter, size and shape of penetrates, crosslink density and void content of the elastomeric membranes plays a important role in swelling characteristics.





Figure 11. Swelling behaviours of (NBR/SBR)/GP/CN nanocomposites in organic solvents

#### **Dispersion of fillers**

Figure 12 has been revealed that a grey scale image is obtained the light dots are associated with filler and agglomerates whereas the dark background is associated with the rubber matrix. This image is transformed by numerical treatment into a black and white image. Figure 12 revealed that SN1 has smooth surface (without fillers) and green composite and nanocomposites were exhibited two phase morphology; that is black phase in rubber and white phase is fillers. With the addition of nanoclay into rubber matrix the dispersion of fillers is homogenising.



Figure 12. Dispersion images of (a) NBR/SBR, (b) 5 % GP, (c) 2.5 CN, (d) 5 % CN and (e) 10 % CN of (NBR/SBR)/GP/CN nanocomposites

## Morphological behavior

Figure 13 shows the SEM image of NBR/SBR (50/50) (SN1) blend in absence of CN and GP. This micrograph indicates a loose matrix with an irregular domain size and shape. That means the investigated blend is incompatible blend, [26]. Figures 13 (b) - (e) shows the SEM photomicrographs of the NBR/SBR blend after incorporation of GP and CN. Figure SN2 indicates that poor interaction between hydrophilic GP to hydrophobic elastomeric phase. Also flaws, mircovoids and cracks in the continuous phase were noticed. It was clear that, the surface texture changed slightly. In other words, the addition of the crysnanoclay along with ginger powder has improved both the morphology and, consequently the compatibility of the blends due to the reduction of the interfacial tension between NBR and SBR. SEM images also revealed that fillers are distributed homogenously is continuous elastomeric phase. The domain size of the filler or second phase slightly increases with increase in filler loading. This is due to increase in filler – filler interaction.



Figure 13. SEM photomicrographs of (NBR/SBR)/GP/CN nanocomposites (SN1, SN6, SN7, SN8 and SN9)

SEM image support the tensile behavior of NBR/SBR/GP composites. After incorporation of nanoclay, the surface of composite is smooth and two phases was noticed. The finer and homogeneous dispersion of fillers (nanoclay, GP and carbon black) was noticed in all nanocomposites. Also micro/nano fillers are embedded in elastomeric phase. Domain size of second phase slightly increases with increase in nanoclay content. This is due to at higher filler loading filler-filler interaction is dominating than polymer-filler interaction. The observation made in SEM, supports the mechanical behaviours of the nanocomposites.

## Conclusions

Crysnanoclay was used as reinforcing filler at different ratios in NBR/SBR (50/50) blend. The role of the filler was investigated using physico-mechanical properties and scanning electron microscopy (SEM). From the tensile behaviour it was observed that the incorporation of GP component into blend matrix improves the surface hardness and tensile modulus of the resulting systems significantly. The compression set value increases with increasing CN loading for (NBR/SBR)/GP/CN nanocomposites.

The water uptake values of the nanocomposites decreases significantly with increase in nanoclay content. This result clearly indicates that the water uptake behaviour of the composites significantly depends on the filler content. Solvent resistance of the samples increased upon the addition of nanostructured silicates, due to the intercalation of rubber chain into the layers. It will result in nano metric level of dispersion of silicates into the polymer matrix. The swelling percentage decreases with increasing crysnanoclay loading for all nanocomposites. It was observed that as the size of the molecule increased, the solvent uptake of polymer decreased in the order; benzene > toluene > p-xylene. Use of cellulose with hydrophobic polymer matrix such as rubber deteriorates its resistance to water vapour permeation. The lower swelling percentage is due to close packing density of nanocomposites than blend matrix. SEM photomicrographs indicates that the homogeneous and finer dispersion of GP and CN fillers in rubber matrix. The optimised composition of this experimental investigation is the 10 wt. % of crysnanoclay loaded NBR/SBR blend which have a reasonable value of physico - mechanical, compression set and swelling percentage. GP could be utilized as biodegradable filler to minimize environmental pollution rather than strong reinforcing filler for elastomeric materials. Nevertheless, this problem could be reduced by modifying the filler surface using a compatibilizer or coupling agent.

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