Effect of nano silica on the mechanical properties of Styrene-butadiene rubber (SBR) composite

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Abstract: The aim of this research is to study the effect of nano silica which use as a filler with loading level (0.1, 0.3, 0.5, 1, 3, 5, 10, 20, 30, 40 pphr) on mechanical properties of SBR such as Tensile strength, elastic modulus, hardness, abrasion, fatigue and tear resistance. The results show that on addition of small quantities of nano silica an increase of mechanical properties occurs. While at high percent of nano silica aggregation in rubber matrix occurs and decrease themechanical properties.

Keywords: Nanocomposite, Mechanical Properties, Styrene-Butadiene Rubber, Nano-Silica

1. Introduction

Rubber nanocomposites have been widely discussed by scientists over the past few years by considering the number of potential nanoelements. A nanocomposite is termed when at least one of the phases within the composite has a size-scale of order of nanometers [1]. When nanoparticles incorporated in a soft matrix can lead to enhanced mechanical and electrical properties due to the large number of crosslinking sites at the nanoparticle-matrix interface [2]. In rubbery nanocomposites, due to interactions of matrix molecules with solid, the incorporation of filler particles in elastomers reduced chain mobility [3]. Increasing in the using of elastomeric materials in many commercial applications is, as a result of the strong influence of relatively small amounts of filler particles, on the mechanical properties of elastomers [4]. Rubber compounding is the art and science of selecting number of different compounding ingredients and their quantities to mix and produce a useful rubber formulation that is processable, meets or exceeds the customer’s final product requirements. Some of the ingredients such as vulcanizing or crosslinking agents, anti-degradants, plasticizers, processing aids and fillers, might modify the basic properties of the raw gum elastomer within the compound [5]. In the rubber industry, the tires industry consumes almost 70% of the total styrene butadiene rubber (SBR) production, since SBR has suitable properties of very good ageing resistance low temperature properties and good abrasion resistance. Styrene-butadiene rubber is a copolymer of styrene (\( \text{C}_6\text{H}_5\text{CH=CH}_2 \)) and butadiene (\( \text{CH}_2=\text{CH} - \text{CH}=\text{CH}_2 \)) with a Tg of approximately -55°C. SBR usually incorporates accelerators in its compounding formulations, its being an intrinsically slower curing material compared to natural rubber. The levels of reclaim rubber used are generally 15 to 25 phr, about half of which is rubber and the other half. The white color of silica and their small primary particle size, give rise to a remarkably high reinforcing efficiency and it provides the highest reinforcement to rubber products [9]. The main uses of silica are in the truck tires and treads of passenger for traction and low rolling resistance [6]. The effect of fillers on rubber reinforcement depends on the filler properties, e.g., particle size, shape and aspect ratio, dispersion of filler in the polymer matrix and interaction with polymer [10, 7]. Because of small particle size and complex aggregate structure of precipitated silica, it imparts the highest degree of reinforcement to rubber among all of the nonblack particulate fillers [6]. Nanofillers, which range from 1 to 100 nm, are able to significantly improve the mechanical performance of the rubber products and give good reinforcement [3]. The
The high surface area of nanoparticles leads to more polymer chains attached to the surface. This restricts the movement of polymer chains under loading, resulting in high reinforcement. Therefore, nanoparticles are profoundly important to the reinforcement of elastomers [3]. The improved mechanical properties of the rubber nanocomposites are not based on the incorporation of hard fillers only, but also on the interactions developed between the filler and the matrix. These interactions can vary from strong (chemical bonds) to intermediate (hydrogen bonding, dipole interactions) to weak (van der Waals) [11]. When silica is mixed with the commonly used rubbers, leads to mixing and reinforcement problems due to the difference in surface energy. Another major problem is that silica-to-silica interaction is high, resulting in the formation of large aggregates ultimately impeding the compound flow character [12]. In dispersive mixing the main goal is to breakdown agglomerates and aggregates of the original filler in to small particles to achieve the highest reinforcing effect [7, 13]. The profound effects of vulcanization, it transforms an elastomer particles to achieve the highest reinforcing effect [7, 13]. The mixing process continues for more time in order to get a good homogenization and decrease mill opening to (0.5-1 mm) for increasing the homogeneously and the efficiency of mixing.

2. Experimental Part

2.1. Materials

The rubber batch materials consist of silica, (laboratory prepared). Styrene –Butadiene Rubber (SBR 1712 and SBR 1502), Reclaim rubber, zinc oxide, stearic acid, tetramethylthiuram disulfide (TMTD), castor oil, sulfur, 2,2,4-trimethyl-1,2-dihydroquinoline (TMQ), Carbon black (N375), N-cyclohexyl-2-benzothiazole sulfonamide (CBS), N-(1,3-dimethylbutyl)-N-phenyl-p-phenylenediamine) (6PPD), which supplied by Babylon tire company.

2.2. Preparation Method

The master batch is prepared by mixing the materials by using the laboratory mill with rolls dimensions are: outside (150mm), working distance (300mm), speed of the slow roll 24 rpm. The roll mill has the facility of controlling the gap distance between the rolls. The ingredients were added according to the standard specifications (IT1060). The formulation of the rubber composites are given in Table1. The steps of processing recipe master batch with different mixing time are:

1. passing of (SBR 1502- SBR 1712) rubber between the two rolls for several times with decreasing the distance between the two rolls to the extent of (0.5-1) mm at temperture of laboratory.
2. Adding reclaim rubber to them, and banding for three minute.
3. During the process, rubber pieces are pass between rollers in horizontal and vertical state alternatively for several times for obtaining homogenous materials
4. Add the zinc oxide and repeat mixing for 2 minute, then addition of stearic acid.
5. Addition the carbon black and DOP oil alternatively and repeat mixing for 7 minutes.
6. Addition of accelerator CBS and repeat mixing for 5 minute, then addition of antioxidant.
7. Add the reinforcing fillers (SiO₂) blended with coupling agent, and repeat mixing.
8. Finally, adding the sulphur with pre-blending.
9. The mixing process continues for more time in order to get a good homogenization and decrease mill opening to (0.5-1 mm) for increasing the homogenously and the efficiency of mixing.

<table>
<thead>
<tr>
<th>compounding ingredients</th>
<th>ppbh</th>
</tr>
</thead>
<tbody>
<tr>
<td>SBR 1502</td>
<td>20</td>
</tr>
<tr>
<td>SBR 1712</td>
<td>80</td>
</tr>
<tr>
<td>Reclaim</td>
<td>4</td>
</tr>
<tr>
<td>Zinc oxide</td>
<td>1.5</td>
</tr>
<tr>
<td>Stearic acid</td>
<td>1</td>
</tr>
<tr>
<td>Paraffin wax</td>
<td>1</td>
</tr>
<tr>
<td>TMQ</td>
<td>0.5</td>
</tr>
<tr>
<td>6PPD</td>
<td>1</td>
</tr>
<tr>
<td>DOP</td>
<td>8.5</td>
</tr>
<tr>
<td>Sulfur</td>
<td>1.75</td>
</tr>
<tr>
<td>Carbon black (N-375)</td>
<td>60</td>
</tr>
<tr>
<td>CBS</td>
<td>1</td>
</tr>
<tr>
<td>Silica</td>
<td>0- 0.1- 0.3- 0.5- 1- 3- 5- 10- 20- 30- 40</td>
</tr>
</tbody>
</table>

2.3. Characterization and Testing

2.3.1. Mechanical Tests

To examine tensile and elongation at break and modulus of elasticity and tear resistance properties use a tensile Monsanto T10 Tensometer according to the ASTM D412. It consisting of two jaws, one fixed and the other moving quickly 500mm / min.

2.3.2. Hardness Test

The test for hardness samples carried on according the ASTM D 2240 by using a device (Shore A hardness) type (Th200). The examination occurs by work of five strikes at different places of the sample with thickness 6nm, and calculated the average value of hardness.

2.3.3. Fatigue Test

In this test sample is installed in the jaws of the device and then calculate the number of cycles during crack growth in
rubber until it is cut at 75 °C. The device used for this test type (Wallace-Dematta Flexing Machine). Testing is done according to the ASTM D430.

2.3.4. Abrasion Resistance Test
Testing is done by device type (wallace test equipment, HZ 50) as ASTM D2228. The device works with 250 Watts, where the sample is placed touching the lever, the lever spin 500 cycle for 5 min and then calculated the difference in weight before and after the test.

3. Results and Discussions
3.1. Mechanical Tests Results
3.1.1. Tensile Strength
Addition of small quantity of nano silica (figure1A) shows an increase of the tensile strength. As the concentration increases, tensile strength decrease (figure1B). Such behavior can be explain that in case of small quantities, particles will be fill the spaces between rubber chains, thus gave a rigid structure with better tensile strength (19.25 Mpa). While in high quantities of nano silica aggregate occurs in rubber matrix, this weakening the bond between chains and decrease the tensile strength. This result agree with references [3,4,16].

3.1.2. Elongation Property
The elongation property decreases with the addition of small quantities of nanosilica as Figure 2A, because the rubber is highly stretching so that when very fine particles fill the spaces, it will be restrict the movement of chains and then decrease elongation property. As an increase the quantities of nano silica as in Figure 2B increased the elongation property. This is due to the diffusion of very fine nano silica through the rubber chains and support the rubber chains so enhanced stretching which reflect on elongation. This agree with reference [16].

3.1.3. Modulus of Elasticity Property
As the inversely proportional of elastic modulus to the elongation property, Figure 3A shows an increase in the modulus of elasticity with small amounts of the nano silica. Due to the fineness of particles makes it easy to distributed between rubber chains, so increase the modulus of elasticity. But when an increases in quantities of the nano silica as Figure 3B decrease the modulus of elasticity. This is due to heterogeneous distribution leads to the agglomeration of particles between the chains, which agree with references [3,16].

3.1.4. Tear Resistance Property
This property related to the tensile property so that tear resistance increases with small quantities of the nano silica. as Figure 4A, it reaches to a maximum value of 9.05 Mpa at 1pphr of silica for the same reason as previously mentioned in the case of tensile property that the particles will be fill the spaces between rubber chains and increase the mechanical bond between them this lead to better tear resistance. In case of increase the quantity of nanosilica for 3pphr, tear resistance decreases to 7.05 Mpa, as shown in Figure 4B. The decreases of tear resistance due to irregular distribution of nanosilica particles in rubber matrix and aggregate between
chains. This agree with reference [3].

![Graph A](image1.png) ![Graph B](image2.png)

**Fig. 3A, B.** Shows the effect of small and high quantities of nano silica on Modulus of elasticity property of recipe.

![Graph A](image3.png) ![Graph B](image4.png)

**Fig. 4A, B.** Shows the effect of small and high quantities of nano silica on Tear resistance property of recipe.

### 3.2. Hardness Property

In Figure 5A shows hardness property increases with small quantities of the nano silica, it reach to the maximum resists value 63.44 at 1pphr of silica due to silica act as filler that it strengthening the recipe and makes it resistance the applied forces, which gives high hardness value and this agreed with the increase in modulus of elasticity. At 3 pphr of nanosilica as Figure 5B agglomerate will be take place and make pressure in structure of rubber matrix which enhanced tear process and decrease hardness property, and starts to rise at 5 pphr of silica. In general hardness increases, even at high amounts of nanoparticles [3,16].

![Graph A](image5.png) ![Graph B](image6.png)

**Fig. 5A, B.** Shows the effect of small and high quantities of nano silica on Hardness property of recipe.

### 3.3. Abrasion Resistance Property

Figure 6A shows that the addition of small amounts of silica e.g. 0.1 pphr, the weight loss of rubber increases due to silica acts as filler which not contact with abrasion load. Then weight loss of rubber begins to decrease until it reaches 0.41% at 1pphr of silica due to nano particles will be restrict weight loss. In case of increasing amount of nanosilica> 3 pphr as in Figure 6B weight loss increases weight loss, due to the agglomerate of particles and form excess fillers this weakening the link between molecular and also reduces tension and abrasion resistances.
3.4. Fatigue Property

This property represents the material resistant to cracking growth. At addition small amounts of nano silica, as shown in Figure 7A silica behave as filler and give a resistance to crack growth. As the amount of silica increases agglomerate will take place and inhomogenous distribution makes stress in recipe which discrete cracking as Figure 7B.

Fig. 6A, B. Shows the effect of small and high quantities of nano silica on abrasion resistance property of recipe.

Fig. 7A, B. Shows the effect of small and high quantities of nano silica on Fatigue property of recipe.

4. Conclusions

On addition of Small quantities of nano silica are improved the mechanical properties comparing with high quantities this is due to the fact that silica at low concentration act as a filler to fill the spaces in the recipe of SBR composite. At high quantities, nano-particles acts as micro-particles and aggregate between chains with irregular distribution in rubber matrix, so that reduce the properties. At 1 pphr of nano-silica is optimum percent which give better tensile strength, tear resistance, fatigue resistance, and abrasion loss give 19.25 Mpa, 9.05 Mpa, 1350 and 0.41% respectively. Any addition percent of nano-silica, result in increasing hardness. The elongation property decreases with the addition of small quantities of nanosilica. As the quantities increase the elongation reach 471% at 40 pphr of silica, this is due to the diffusion of very fine nano silica through the rubber chains and support the rubber bond so enhanced stretching.

References


