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# The Effect of the Nanoparticle SiO<sub>2</sub> on the Mechanical and Physical Properties of Rubber Composites

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**ABSTRACT.** This paper investigates the effect of the nanoparticle SiO<sub>2</sub> on the mechanical and physical properties of a rubber blend of natural rubber (NR) and styrene-butadiene rubber (SBR). The proportions of the nanoparticle that are added are 0, 0.2, 0.4, 0.8, 1, 1.5, 2, 2.5 and 3 pphr to a fixed percentage of rubber blend (30% NR/70% SBR), which has been prepared with vulcanisation factors, accelerators and activators using a two-roll mill laboratory, moulds and a thermal piston; the pressure, temperature and time difference test conditions according to standards. The results of the laborator tests show an increase in the mechanical properties (i.e. tensile strength, modulus of elasticity, resilience and tear resistance) with increases in the percentage load (pphr) until 0.8 pphr, which is when there is a slight decrease; the properties of hardness, fatigue and compression set also increase. While the results for the physical properties show an increase in viscosity, the greater torque, scorch and cure times decrease with increases in the proportion of SiO<sub>2</sub> in the rubber recipe.

## 1. Introduction

Non-reinforced rubber composites have only a few use cases. The addition of ceramic materials such as silica is needed to enhance the rubber's mechanical and physical properties and to minimise the cost of production. Accordingly, studying the mechanical and physical properties of different engineering materials is important because of their engineering applications. The increased use of rubber in engineering has been the result of its unique properties, which include high extensibility, high strength, high energy absorption, high fatigue resistance, strong eco-resistance and high resilience [1]. Rubber by itself has poor properties and no advantage over other materials; in order to improve its performance, it needs to be vulcanised. Vulcanisation, named after Vulcan the Roman God of Fire, describes the process of transforming mechanically brittle raw rubber materials into durable goods. One of the most effective ways of producing high-quality rubber is to strengthen the rigid entities that enhance the mechanical and other properties of the rubber depending on the nature of the reinforcement, whether it is black carbon, silica, oxygen or clay. Integrating fillers into elastomer materials typically leads to a substantial improvement in their mechanical properties, resulting in increased abrasion and tear resistance. The interactions between the rubber fillers are an important parameter affecting these properties [2]. The term 'rubber blend' refers to a combination of two or more different rubbers resulting in a material with a range of properties that do not originate from either one of the components [3]. The elastomer original is NR, which acts as an imperative position among elastomers. NR is extracted as latex from the Pará rubber tree [4]. NR has a moderate teragram



(around  $-73\text{ }^{\circ}\text{C}$ ), which is accompanied by flexible behaviour, especially when it contains a high level of cis, because the presence of methyl groups on the same side of the rubber chain prevents NR from being properly arranged, causing spaces between the particles to form and dissipate the passing wave vibration energy. However, due to the regularity of the molecular structure, NR can easily crystallise at low ambient temperatures, as well as increased in hysteresis. NR products exhibit high tensile strength and high resistance to tearing, cutting and abrasion [5]. NR also has very strong tolerance for low temperatures, down to  $-57\text{ }^{\circ}\text{C}$ , at which point it becomes substantially stiffer. NR has a low glass transition temperature (around  $-73\text{ }^{\circ}\text{C}$ ), which allows it to exist at lower temperatures than most other rubbers [6]. In addition to being used in tires and tubes, NR is used in conveyor belts, engine supports, structural bearing pads, flags, rubber bands, vehicle helmets, heels and gaskets [7].

The synthetic rubber SBR, which is made by polymerising styrene and butadiene, is the most common substitute for natural rubber [8]. Production-based polymerisation reactions can be divided into two forms: hot polymerisation and cold polymerisation. In hot polymerisation, the rubber comprises several branches in the chain and has greater dimensional stability and stronger extrusion compared to the rubber resulting from cold polymerisation. What is generated is linear chains with strong vulcanisation properties and with a low styrene content of up to 20–30% polymer [4]. Particulate fillers can make the strength of the rubber over ten times that of amorphous rubber. To trigger this effect, the fillers must have a highly specific surface area, i.e. the particles must be small. Small particles have a wide surface area to interact with the compound's rubber and similar particle-to-particle spacing. The two types of fillers that are most important for rubber reinforcement are black carbon and silica [9]. Nano-silica consists of spherical particles with a diameter of less than 100 nm. The hydrophilic character of silica also influences the cure properties of rubber compounds; vulcanised rubber properties and even compatible with non-polar rubber such as NR [10]. Silica retards vulcanisation as it reacts to the complex the zinc-accelerator - the sulphur [11]. The most widely used silane coupling agent is 3-(Trimethoxysilyl)propyl methacrylate (TMSPM). A silane coupling agent is a bi-functional molecule: one end has the ability to react chemically with silica, forming an intermediate silica-silane coupling agent, and the other end with rubber. The intermediate silica-silane coupling agent participates in the vulcanisation process, which improves the degree of compatibility between silica and non-polar rubber, thus increasing the reinforcing efficiency of the silica [10]. Interactions between rubber and silica can either happen through physical adsorption or chemical bonding.

This paragraph provides an overview of scientific studies in this area. Sun *et al.* [12] studied the improved performance of nano-silica-filled SBR composites through the surface modification of nano-silica using silane coupling agents. Nano-silica dispersion in SBR and the bonding power of nano-silica with SBR have been significantly improved, and the physical and mechanical properties of vulcanised rubber have also been greatly improved. Sombutosompop *et al.* [13] investigated the effect of untreated silica, precipitated silica (PSi) and fly ash silica (FASi) as fillers on the properties of NR and SBR. It was found that the cure properties at NR loading 30–75 pphr increased with PSi and decreased with FASi at the same rate. All properties of filled SBR were similar to filled NR except for tear strength and tensile, which makes NR the superior rubber. Salimi *et al.* [14] studied the effect of silica enhancements on NR and on NR/SBR compounds. To chemically bind the silica to the rubber, the silica surfaces were treated with a silane-coupling compound (bis(triethoxysilylpropyl)tetrasulfide). The tests of the NR/SBR compound with an optimal amount of silane demonstrate that the cure time decreases as the NR content increases. (The NR/SBR's tensile properties combine with rising SBR material). Mayyadaha [15] studied two types of rubbers (SBR and NR) combined with two types of binding fillers ( $\text{Al}_2\text{O}_3$  and  $\text{SiO}_2$ ). Her findings revealed that, rather than alumina fillers, silica fillers improved their properties. She also found that, with the introduction of all forms of reinforcing fillers and the improvement of their loading speed, their mechanical properties and even durability improved. SBR coated with 25 pphr of silica has the highest hardness qualities and lowest abrasion wear ratings. This research studies the impact of variable amounts of silica on different properties of rubber blends.

## 2. Materials and Method

### 2.1 Used Materials

The materials used in this research are natural rubber (SMR20) and synthetic rubber (SBR 1500), and the materials used in preparing the rubber recipe are CBS, zinc oxide (activated), stearic acid (activated), anox (HB), antiozone (WAX A 111), aromatic oil, carbon black (GPF 660), sulphur and IPPD (imported by the Diwanayah tire company) and nanoparticle material (SiO<sub>2</sub>).

### 2.2 Method of Preparation

The formulation of the rubber composites are given in Table 1.

**Table 1. The composition of the rubber composites.**

Item no.	Recipe ingredients	Loading level (pphr)									
1	SMR20	30									
2	SBR 1500	70									
3	Activator (zinc oxide)	4									
4	Sulphur	2									
5	CBS	0.5									
6	Anox (HB)	1									
7	IPPD	1.75									
8	Antiozone (WAX A 111)	2									
9	Activator (stearic acid)	2									
10	Aromatic oil	8.5									
11	Carbon black (GPF 660)	51									
12	SiO <sub>2</sub>	0	0.2	0.4	0.8	1	1.5	2	2.5	3	

Mixing was done in a two-roll mill (Figure 1); kneading and mixing operations were conducted on this machine as per ASTM D 15. The ingredients were added as follows, as per ASTM D3187-89:

1. Pass SMR20 and SBR 1500 between the two rolls several times with a reduction of 0.5–1 mm between the two rolls.
2. Add stearic acid and zinc oxide and then blend for material homogenisation.
3. Add half of the carbon black with operating oil evenly across the mill at a uniform rate.
4. When the carbon black has been completely incorporated, open the rolls to 1.65 mm.
5. Add the remaining carbon black and oil evenly across the rolls at a uniform rate.
6. Add reinforcement materials (nano-silica) while continuously mixing to start homogenising; the mixing process should continue until excellent homogenisation is achieved.
7. Add sulphur and mix to achieve the homogenisation of the materials under room temperature and then add the accelerators and sulfur.
8. Set the rolls to 0.8 mm and then combine the materials for homogenisation.
9. Open the mill to provide a minimum batch thickness of 6 mm and continue the mixing process until premature homogenisation is achieved.

After preparing the rubber composite recipes, test models were prepared to provide mouldings and state of test, such as temperature, pressure and time. Following that, according to the ASTM, which is a method of thermal piston vulcanisation, the temperature and time of the vulcanisation process variable according to examination.



**Figure 1.** The mixing machine.

### **3. Characterisation and Testing**

#### *3.1 Mechanical Properties*

A tensile test was carried out on the samples prepared from the vulcanised slice (i.e. by cutting three dumbbell specimen). The cutting was done by hand press and by a Monsanto Tensometer T10 in conjunction with the ASTM d412-88, using special tensile test sample holding jaws that are mobile at a speed of 500 mm/min. A tearing examination was conducted in compliance with the standard ASTM D624-54 by using a system tensile (Tansometer10) and speed (500 mm/min) after model testing (dumbbell) by cutting machine models and by cutting machine (Nicking cutter) form (Wallace) incision work in the narrow area of the unit. A Tansometer10 was used to measure the results in N/mm. A hardness test was carried out using a durometer Hardness shore A according to the standard ASTM-D2240. Standardised hardness-measuring equipment was applied directly to the surface of the specimens using a sharp needle to test the intensity. Six different positions were averaged. Resilience was tested using a Wallace Dunlop Tripsometer according to the specifications in ASTM D1054, and the data was linked to the digital computer information, Wprpra-wallace Rebound Resilience Data File, which records the percentage of resilience. The compression set test (ASTM D395) was conducted on ordinary  $29 \pm 0.5$  mm diameter and  $12.5 \pm 0.5$  mm thick cylindrical test specimens. Using a compression set tool, the samples were compressed to 40 percentage of their thickness for 72 hours and then left for 30 minutes until the final sample thickness was taken. Fatigue tests were carried out for the prepared samples in compliance with ASTM D430 using a flexibility fatigue instrument (Wallace-Dematta Flexing Machine), which included a monitor for the number of cycles at which a crack occurs.

#### *3.2 Physical Properties Test*

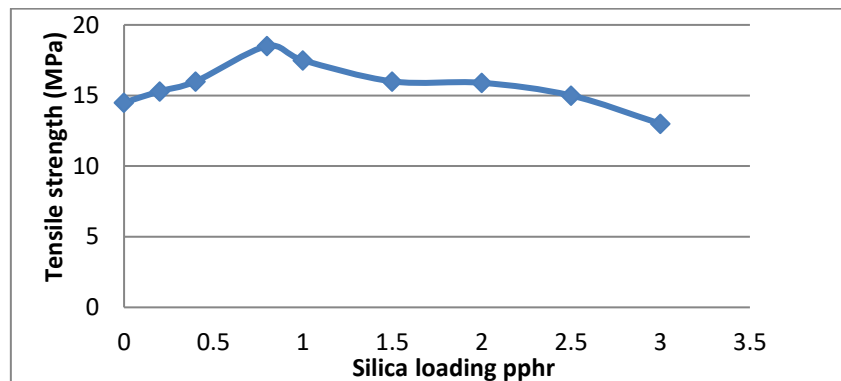
The viscosity and curing characteristics are conducted in compliance with ASTM D2084-89 using an oscillating disk rheometer (ODR) for the prepared samples (i.e. cut from the rubber composite recipes), in which the upper and lower jaw, restraint clutches for six minutes below 20 Pa and 185 °C. As shown by the following results (max. torque), min. torque, scorch time, cure time (min) and viscosity), the results are shown in a graph of torque vs. vulcanised time. The rehometer was connected to a machine with a precision control system.

### **4. Results and Discussions**

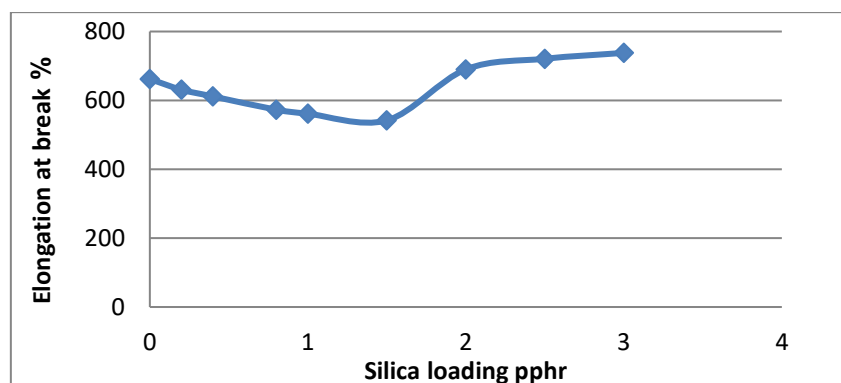
#### *4.1 Mechanical Properties*

The load is increase from zero and even the breakdown happens rapidly (500 mm/min), to establish the properties of the material under the influence of axial tension load in one direction of the sample

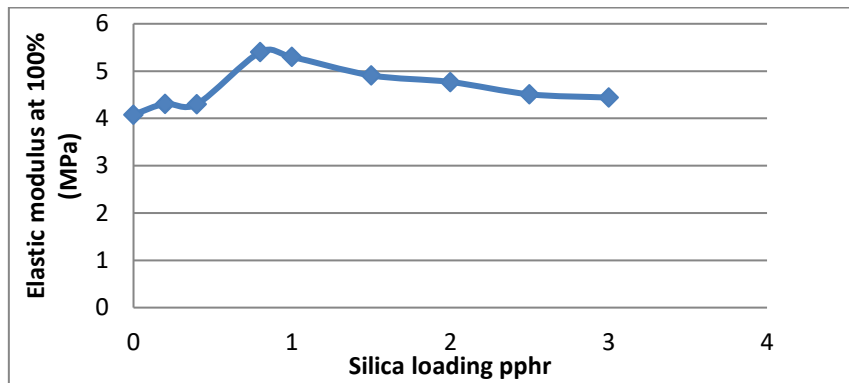
(longitudinal axis). Several models were tested, and an average was taken to achieve the optimal outcome for get rid of human errors and to reduce the rate of error from the samples' heterogeneity. Figure 2 shows an increase in tensile strength with an increase in the percentage of silica in the rubber recipe; however, when the ratio of silica reaches a certain percentage, there is a noticeable decrease in tensile strength due to the aggregation of the granules as a result of pressure and compression. The particles filled the spaces between the rubber chains and hardened the structure, while a large amount will result in an agglomeration in the compound material, which leads to defects in the formation of cracks that lead to weakened tension (10 pphr). This result is consistent with other research [16,17,18]. Figure 3 shows that, when a small amount of nano-silica is added to the rubber recipe, the elongation property of the rubber decreases, because the rubber extends when the fine particles fill the spaces, which reduces the elongation property by limiting the movement of the chains. On the other hand, when the quantity of nano-silica is increased, the elongation property also increases due to the fact that the rubber recipe does not absorb a high amount of filling; therefore, the accumulation of nanoparticles between the chains occurs and leads to the formation of cracks, thus reducing tensile strength and increasing the elongation property. This result is consistent with other research [17]. Figure 4 shows an inverse relationship between the modulus of elasticity and elongation: an increase in the modulus of elasticity was observed with only a small amount of modified nanoparticles. The distribution between the rubber chains facilitates the precision of the particles, and thus the elastic modulus increases. However, an increase in the amount of nanoparticles leads to a decrease in the modulus of elasticity; this is due to the heterogeneous distribution, which leads to the clumping of molecules between the chains. The entanglement then becomes weak, and thus cracks are generated, which reduces the modulus of elasticity. This result is consistent with other research [16,17,19].



**Figure 2.** Effect of nano-silica (pphr) on the tensile strength of the NR/SBR composite.

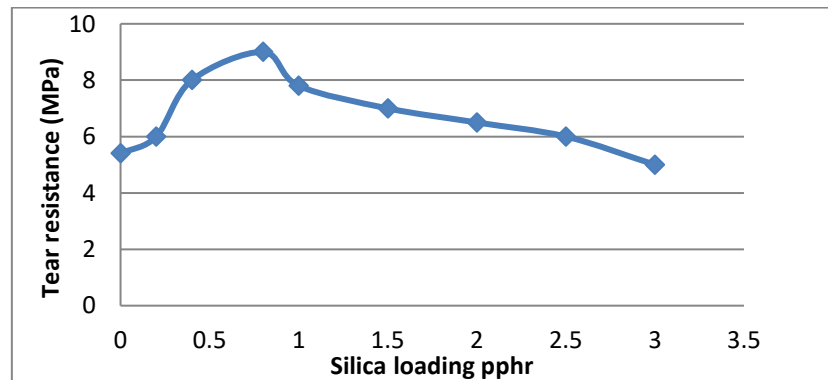


**Figure 3.** Effect of nano-silica (pphr) on the elongation of the NR/SBR composite.



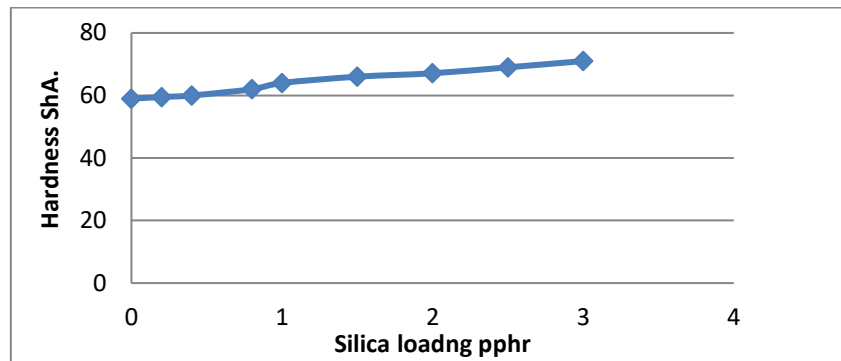
**Figure 4.** Effect of nano-silica (pphr) on the modulus of the NR/SBR composite.

Figure 5 shows the effect of additional materials on tear resistance, which is measured in N/mm. tear indicates the materials' resistance to the growth of any things when under stress. This feature is related to the tension feature. Accordingly, resistance increases when a small amount of silica is added. The maximum value of resistance is 8.2 MPa at 0.8 pphr of silica, because the particles fill the spaces between the rubber chains and increase the mechanical bond between them (i.e. the same reason as mentioned earlier in the case of the tensile property). This leads to the best resistance to tearing. In the case of increasing the amount of modified nano-silica to 3 pphr, resistance decreases to 7 MPa; the resistance decreases due to the irregular distribution of nano-silica particles in the rubber and the assemblies between the chains, as mentioned in the case of the tensile property. This result is consistent with other research [20].



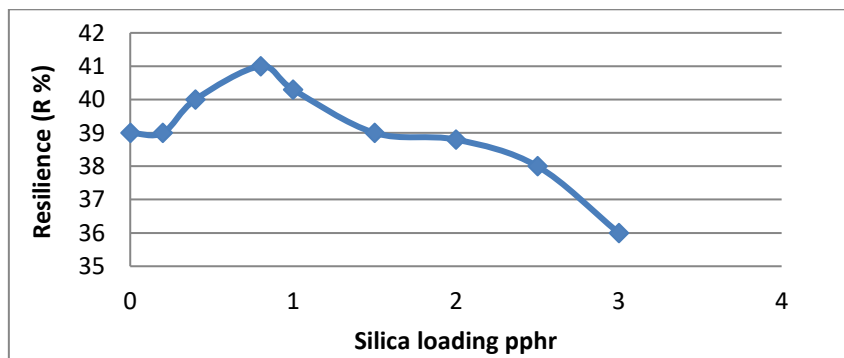
**Figure 5.** Effect of nano-silica (pphr) on the tear of the NR/SBR composite.

Test relies on testing the penetration of the rigid ball in a rubber sample, and releasing the force during load. The hardness was measured from the centre and sides for the samples, which were prepared with a cylindrical form, and took the range to get the maximum result and prevent errors. Figure 6 shows that, when adding nano-silica particles containing a particle size of 65–85 nm to the rubber in different proportions (i.e. 0, 0.2, 0.4, 0.8, 1, 1.5, 2, 2.5 and 3 pphr) in the rubber recipe, there is an increase in hardness, because silica particles have an active and irregular surface area which increases penetration between themselves and the rubber chains upon contact with the rubber, which increases the compound's outer body's resistance to penetration due to increased surface tension. The result is increased rigidity. This result is consistent with other research [19].



**Figure 6.** Effect of nano-silica (pphr) on the hardness of the NR/SBR composite.

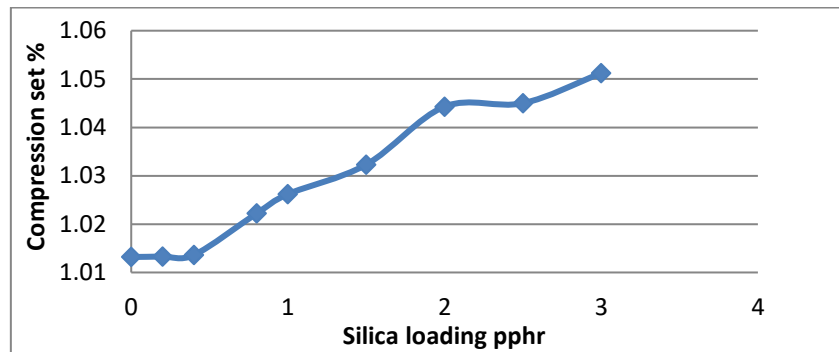
Figure 7 shows an increase in the rebound; at 0.8 pphr, there is a jump in the values, because the nanoparticles act as fillers in the spaces and do not absorb energy. Then, there is a decline, even as the proportion of nanoparticles increases. At 3 pphr, the rebound elasticity increases due to particle aggregation and pressure in the compound rubber structure, which promotes the growth of cracks and reduces the hardness property. After that, agglomeration occurs, and a substance forms that absorbs and dissipates energy as heat, thereby reducing the strength of elasticity. Resilience is related to hardness and density.



**Figure 7.** Effect of nano-silica (pphr) on the resilience of the NR/SBR composite.

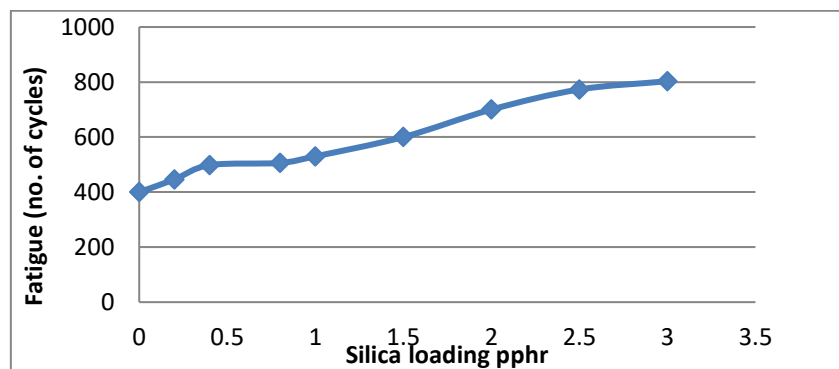
Figure 8 shows that the compression set increases the ratio of silica; this is due to an increase in the surface area as a result of the interference between silica granules and rubber and an increase in hardness, i.e. increased crosslinking and reduced movement of chains, thus reducing Recoverability of the rubber compound. This result is consistent with other research [21].





**Figure 8.** Effect of nano-silica (pphr) on the compression set of the NR/SBR composite.

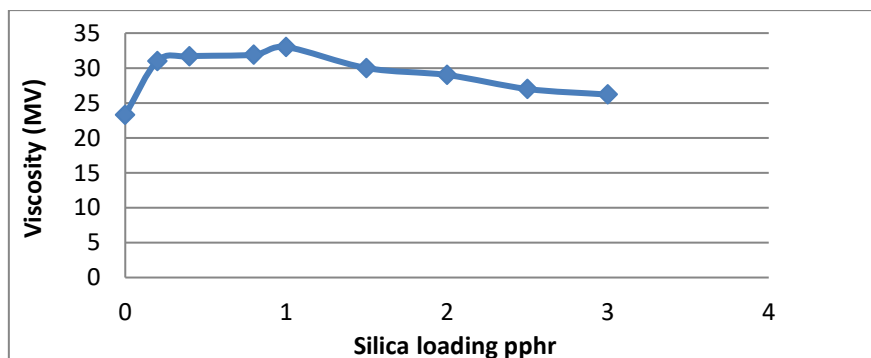
Fatigue strength is represented by the materials that are resistant to the growth of cracks. Figure 9 shows that, in small quantities, silica provides resistance to crack growth. The higher the amount of nanoparticles, The greater the heterogeneous agglomeration and the distribution will make fatigue in the sample making the cracks invisible.



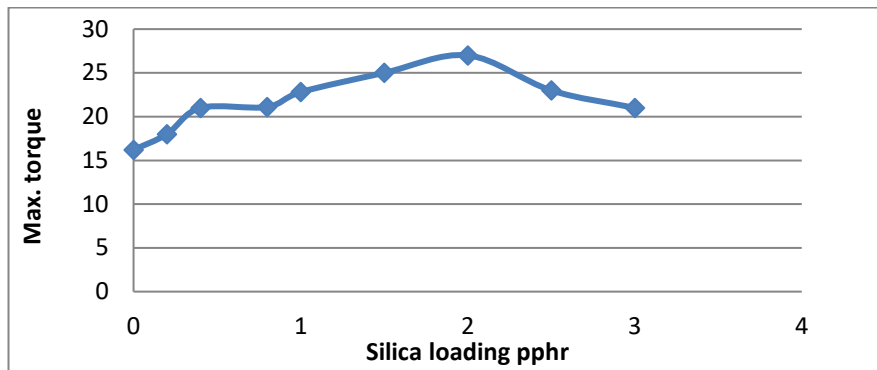
**Figure 9.** Effect of small quantities of nano-silica (pphr) on the fatigue of the NR/SBR composite.

#### 4.2 Physical Properties

Addition nano-silica with limited percentage increases the force of the elastic mixture, indicating an increase in torque and viscosity. Figures 10 and 11 show that the presence of nano-particles means that the mobility of rubber chains is limited; therefore, these properties decrease. This result is consistent with other research [22].

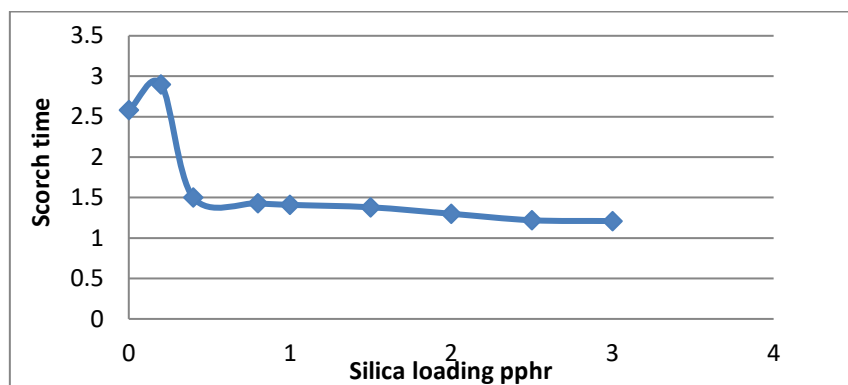


**Figure 10.** Effect of nano-silica (pphr) on the viscosity of the NR/SBR composite.

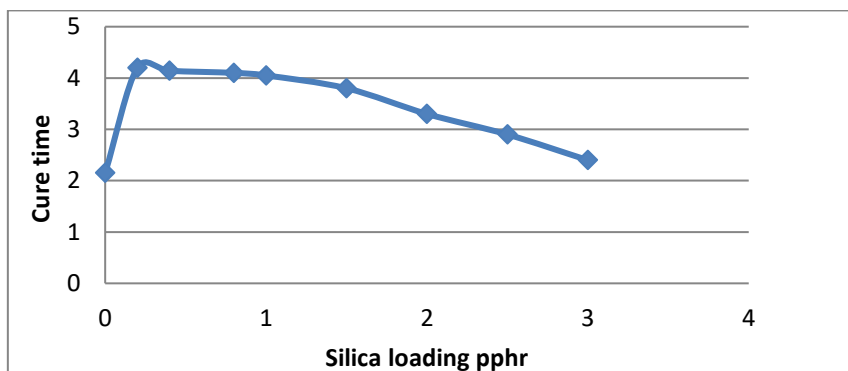


**Figure 11.** Effect of nano-silica (pphr) on the maximum torque of the NR/SBR composite.

Figures 12 and 13 show that, at low percentages, the modified nano-silica increases scorch and cure times. This is due to the fact that silica makes a coat on the rubber molecules, which allows the reaction with sulphur partially. However, there is no subsequent effect on these times as the nano particles increase.



**Figure 12.** Effect of nano-silica (pphr) on the scorch time of the NR/SBR composite.



**Figure 13.** Effect of nano-silica (pphr) on the cure time of the NR/SBR composite.

## 5. CONCLUSIONS

These results show that increasing the silica content in a rubber blend of NR and SBR increases the rubber's tensile strength, modulus of elasticity, hardness, compression set and fatigue. Properties such as tear resistance and resilience decrease with increases in the silica. While the results for the physical properties of the rubber show an increase in viscosity, the greater torque, scorch time and cure time decrease when the proportion of SiO<sub>2</sub> in the rubber recipe is increased. The ratio of 0.8 pphr of nano silica achieved the best result in most of the tests. It is possible to take advantage of this new rubber compound in engineering applications that require the aforementioned advantages.

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