



The Effects of Silica/Carbon Black Ratio on the Dynamic Properties of the Tread compounds in Truck Tires

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Abstract: NR is the major constituent in the rubber compound used for the tread on the truck tires. A general compound formulation of the tire tread includes NR and BR as polymer base and carbon black as the reinforcing filler, and curative components. In this paper the effects of dual filler system (carbon black and precipitated silica) on the dynamic properties of tire treat has been studied. The results show by increasing of precipitated silica, significant improvement was observed in fatigue resistance, rolling resistance and heat buildup of the tire. Tensile strength and modulus and wet grip of tire tread decrease with increasing of silica in rubber compound formulation.

Keywords: Tire Tread, Dynamic properties, Fatigue resistance, Silica, Carbon black, NR.

Introduction

Regarding its vast improvement in the last decades, tire industry has always won the first place considering the volume of the studies done on the properties of rubber. Many researchers, from all over the world, are trying to present new products with higher capabilities and efficacy. From among these studies, the most has been dedicated to the tread of the tire for this is the part of the tire which has the greatest impact on the way and the type of use¹. Atashi and Shiva² have studied the optimization of failure properties of the Passenger Tire Tread Compound made of a compound of SBR with NR and BR, by altering curing condition and modeling rubber's behavior. Ismail Hanafi *et al.*³ studied the blends of two types of SBR with NR and the inherent properties of rubbers on the properties of the final compound. The results showed an increase in Mooney viscosity and scorch time as the SBR increased in the compound. Kaushik *et al.*⁴ studied the compound of NR/BR/HSR with the presence of different amounts of carbon black and found that the samples gained a very

good abrasion resistance and in addition, as the size of carbon black particles decreased and became close to nano size, the strength of the samples increased.

Since tire is a composite structure made of substances with various properties, choosing each of these substances and the amount of each of them requires a rich knowledge. One of the substances which are added to the rubber compounds are reinforced fillers and have an important effects on the properties of the final product. Carbon black is one of the reinforces that frequently used in tire industry and many researchers have studied its effect on mechanical and dynamic properties of tires. The size of carbon black particles and its specific surface are the most important factors affecting on tires' eventual properties. Today, substituting carbon black with precipitated, shapeless silica as a reinforcing additive is becoming popular in tire industry, particularly in formulating the compound for the tread of Passenger and truck tires. Silica's particular surface, as compared to that of carbon black, results in different dynamic properties when it reacts with elastomers. On the other hand, using silica alone increases the viscosity of the compound significantly, and hence, results in problem in processing and curing the rubber compounds. Therefore, using silica as filler has always been favorable alongside with using fluidity factors in order for improving process ability and reinforcing properties⁵⁻⁹. Nowadays, in order to achieve a balanced condition between the properties of rolling resistance, wet grip and abrasion resistance in tires, silica-filled compounds are frequently used in tire industries¹⁰⁻¹¹. For the elements which are under the effect of dynamic forces, it is very important to have fatigue life, and this should be taken into account when making compounds for such elements, for if a crack is created and developed in such samples results in their premature breakdown. Tires, too, are constantly under the effect of dynamic forces and hence, investigating the factors affecting their fatigue resistance is of a great importance. In this paper the effect of carbon black – silica ratio on the dynamic properties of the tread compounds of truck tires based on NR/BR has been studied.

Experimental

In this study, BR rubber made by Arak refinery, NR rubber (SMR 20) made in Malaysia, N375 carbon black (with the specific surface of 95-105 m²/g) made by Iran Carbon Co., CS180 silica (with the specific surface of 134.63 m²/g, and a mean particle size of 440 nm) made by Anhui Co., Si69 coupling agent made by Deggusa Co., aromatic oil No 290 made by Behran Oil Co., zinc oxide made by Pars Oxide Co., 95%-pure Stearic acid made by G&N SDN. BHD, Sulfur made by Iran Tesdak Co., Sulfonamide accelerator by the formula of *N*-cyclohexile 2-Benzotiazole Sulfonamide (CBS) made by Lanxess Co., anti oxidation and ozone 6PPD made by Duslo and TMQ anti oxidizer made by Flexys Co. were used to create a compound based on general formula of the tread of tire trucks⁷. The total amount of filler was the same for all the compounds, equal to 60 phr. We started with 60 parts of carbon black, and gradually replaced some part of carbon black with silica. The amount of substances used is represented in Table 1. The changes made in the properties of the cured tire tread were studied based on the amount of silica added and the carbon black replaced.

Instruments

The compounding formulations which were represented in Table 1, mixed in a two roll mill (Hiva Machinery Co., Iran) at a friction ratio of 1:2 following standard mixing sequence. The ingredients were added in four stages. The reinforcing fillers (carbon black and silica) were added along with the process oil and paraffinic wax in stage one. CBS and zinc oxide were added in stages two and three, respectively, and sulphur and antioxidant in the final stage.

The resultant rubber compounds were then compression-molded to a 90% cure with a hydraulic press (Iran co.) at 15 MPa, using a 160 °C cure temperature to produce vulcanized sample.

Table 1. Compound Formulations (Unit: phr).

Ingredients	Sample Code			
	NRS0	NRS5	NRS10	NRS20
NR	75	75	75	75
BR	25	25	25	25
Carbon Black	60	55	50	40
Silica	0	5	10	20
Silan	0	0.4	0.8	1.6
Aromatic oil	10	10	10	10
St. acid	2	2	2	2
ZnO	4	4	4	4
Wax	2	2	2	2
Sulfur	1.5	1.5	1.5	1.5
CBS	0.75	0.75	0.75	0.75
TMQ	1	1	1	1
6PPD	1.5	1.5	1.5	1.5

Rheometric properties of the samples were measured using MDR Rheometer 900 made by Hiwa Company, based on ASTM D5289 standard and in the temperature of 160 °C. Dumbbell-shaped samples were taken from vulcanized plate and tensile test was done based on ASTM D 412 standard, using Hiwa Mechanic Test Device. The state of fillers dispersion were analyzed by cryogenically fracturing (liquid nitrogen) the samples and observing them in a scanning electron microscope (SEM, Hitachi x-659) after coating with gold. Based on ISO 6943 dumbbell-shaped samples were subjected to fatigue test. A HIWA 600 made by Hiwa Co. with the cycles of up to 350000 was used to execute this test. Hardness test was done using a Hardnessmeter 49038 Shore A, made by Bareiss Co., based on ASTM D2240 standard, abrasion resistance of the samples was measured using an Abrasionmeter made by Bareiss Co., on pill-shaped samples. DMTA test was performed using a Tritec-2000 made in England and based on ASTM E 1640, in order to measure dynamic features of the samples, and heat generation test was done by means of Goodrich Flexometer based on ISO 4666. All the tests were performed on at least three samples and statistic calculations proved their accuracy to be 90% or more.

Results and Discussion

Cure characteristics

Figures 1-4 represent the tests done on the samples in which carbon black is gradually replaced with silica, using rheometer. Increased scorch time can be due to an increase of active surface of silica, and consequently, an increase in the adsorption of catalysts by silanol groups on silica surface. This adsorption is increases as silica increases by percentage, which results in scorch time to increase. In order to gain an optimum condition, the coupling agent (Si69) can be increased relative to the increase of silica content, in order to prevent the increase of this parameter¹². On the other hand, hysteresis can also be studied as the silica increases, because of the increase in the viscosity in minimum and maximum torque graphs, which confirms the increase of scorch time as the percentage of silica is increased.

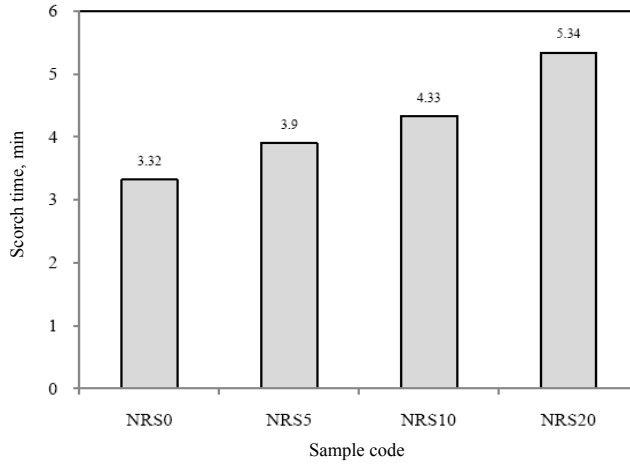


Figure 1. Scorch time of samples at various silica ratios.

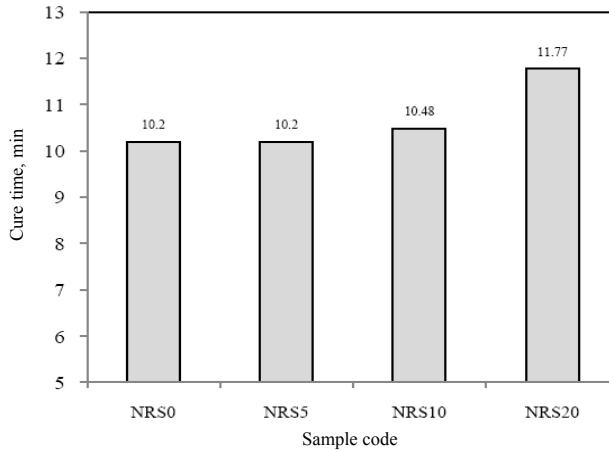


Figure 2. Cure time of samples at various silica ratios.

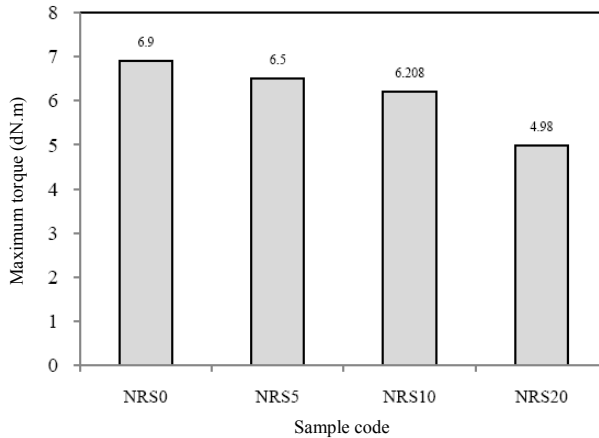


Figure 3. Maximum torque of samples at various silica ratios.

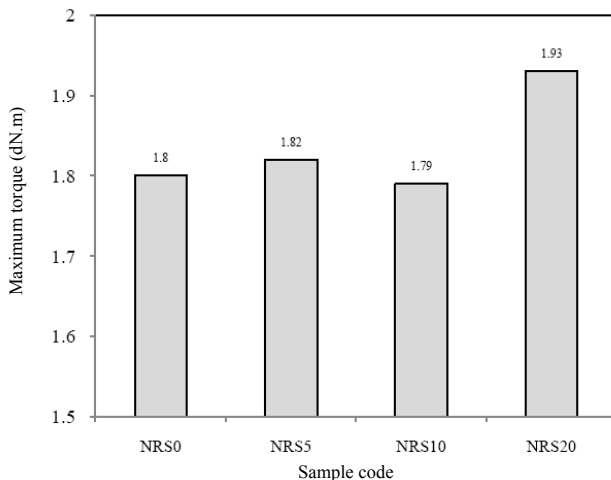


Figure 4. Minimum torque of samples at various silica ratios.

Mechanical properties

The results for tensile strength and elongation at break are represented in Figure 5 and 6, respectively by Increasing of silica content. The tensile strength of rubber compounds has been decreased and elongation at break increase. This behavior indicates the decrease of crosslink density in compounds. On the other hand, the decrease in crosslink density results in the decline of rubber's modulus. This is presented in Figure 7. Figure 8(a)-8(b) represents the SEM photographs of the fractured samples. It can be seen clearly from Figure 8(b) that silica distribution is uniform. As the total amount of the fillers is not changed in the compounds, it is expected that the hardness of the samples does not change significantly after being cured. As shown in Figure 9 however, the hardness of the samples decreases gradually as the percentage of the silica is increased. This decrease of hardness can be mainly because of the decrease in crosslink density in the samples which happens as the active surface of silica in contact with the factors creating crosslink is increased.

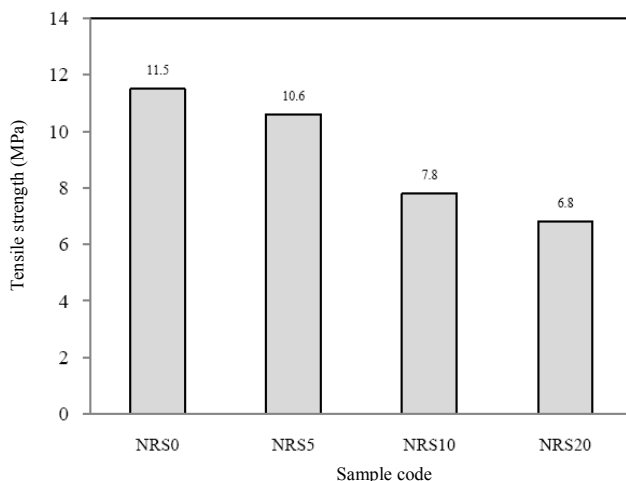


Figure 5. Tensile strength of samples at various silica ratios.

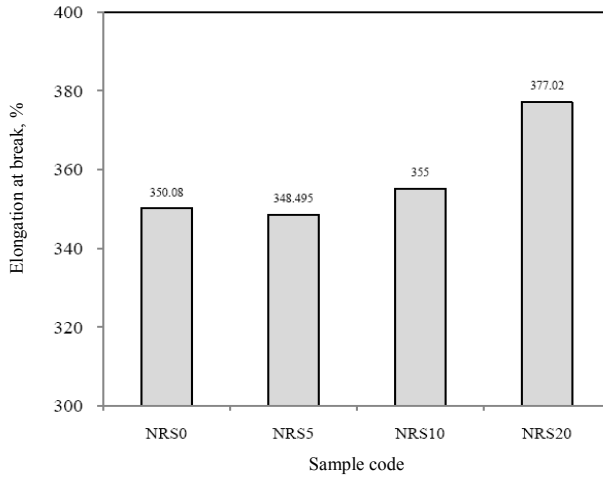


Figure 6. Elongation at break of samples at various silica ratios.

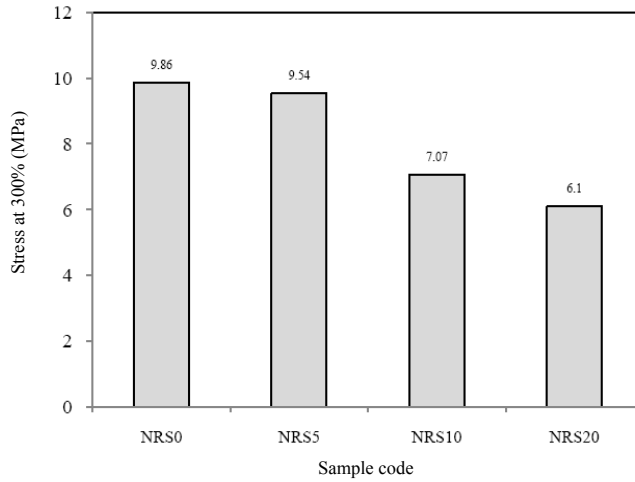


Figure 7. Stress at 300% of samples at various silica ratios.

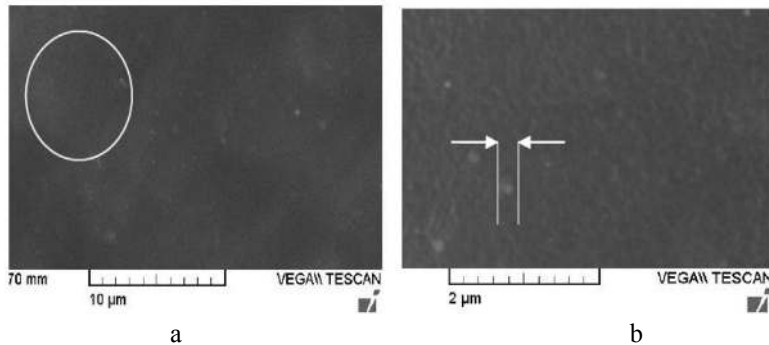


Figure 8. SEM photographs of cryogenically fractured surface of NRS20 sample: (a) 5.0 kx; (b) 25.0 kx.

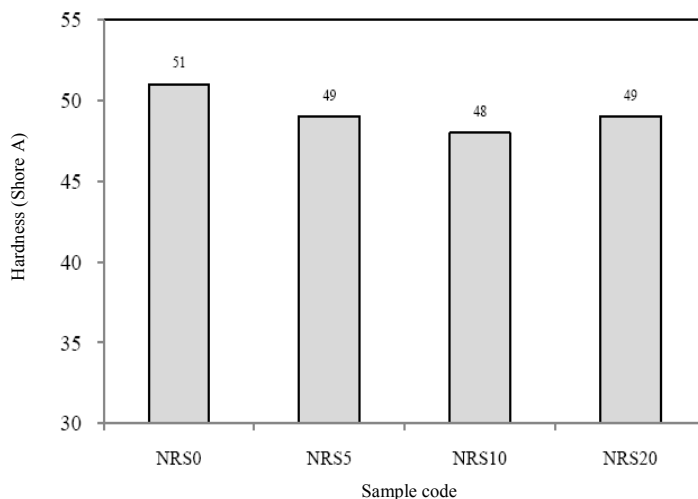


Figure 9. Hardness of samples at various silica ratios.

Abrasion resistance

The compounds made by the two-phase combination of carbon black and silica included 60 parts of filler in general, in which some percent of silica substitutes carbon black. Figure 10 illustrates the changes in abrasion properties of the samples as silica is increased. Two important factors in studying abrasion behaviors of rubbers are modulus and crosslink density. High modulus and crosslink density lead to the enhancement of abrasion strength of rubber compounds.

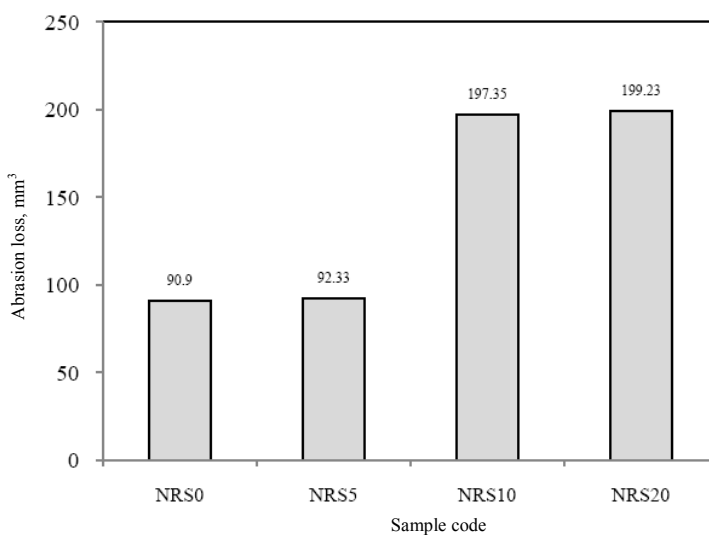


Figure 10. Abrasion loss of samples at various silica ratios.

Fatigue life

One of the factors affecting fatigue resistance of rubber is modulus in low tensions¹³⁻¹⁴. As the modulus decreases in low tensions, the cracks made by tension will also decrease and the control over the growth of cracks and consequently the breakdown of the sample is increased. The results of fatigue test represented in Figure 11 shows a significant improvement in fatigue resistance as the silica content increases in the rubber compounds. This shows the improvement in properties against dynamic tensions created in rubber compounds.

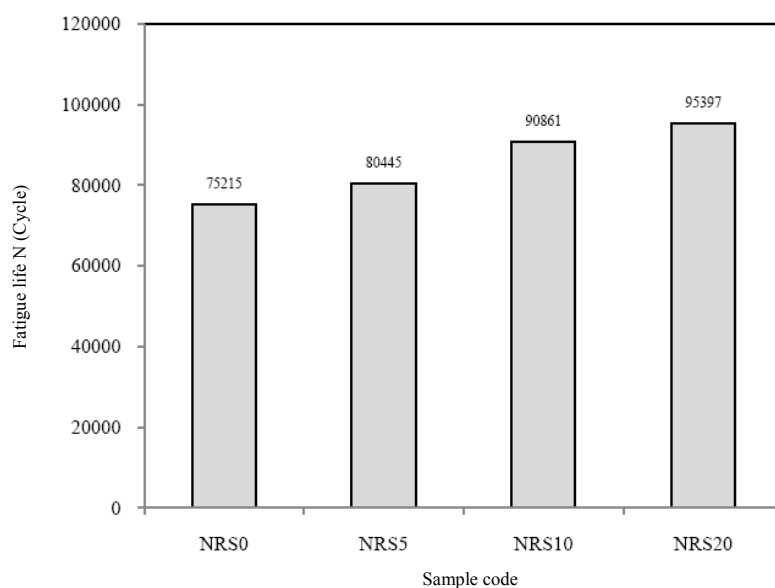


Figure 11. Fatigue life of samples at various silica ratios.

Dynamic properties

Figure 12 show the DMTA graph of samples under dynamic forces from -90 to 90 °C. The value of $\tan\delta$, within the temperature range of 50 to 80 °C is a criterion for measuring rolling resistance¹⁵ which is represented in Figure 13. The value of $\tan\delta$ within the range mentioned is decreased as the silica content is increased, which indicates that rolling resistance is decreased. On the other hand, decreasing T_g in the compound results in the decrease of rolling resistance, figure 14 shows the maximum decrease resulted from using 20 and 5 percent substitute silica. Figure 15 shows the changes of $\tan\delta$ within the temperature range of -10 to +10 °C, which are used as a criterion for evaluating the property of slippage on wet surface (wet grip). Within this range, the more the value of $\tan\delta$, the more appropriate the slippage of the sample, *i.e.* the safer the tire. Results show a decrease in this property as compared to when silica is not used. The phenomenon of heat buildup is a criterion showing the distribution of the heat resulted from the cyclic transformation of rubber and also the waste modulus (E'') of the rubber, which is related to $\tan\delta$ via the following equation:

$$\tan\delta = \frac{E''}{E'}$$

In this equation, (E') is the storage modulus and the result of this equation shows the changes of tanδ. The results of Goodrich Flexometer which are represented in Figure 16 shows a decrease in heat buildup of the samples as compared with the samples filled with carbon black.

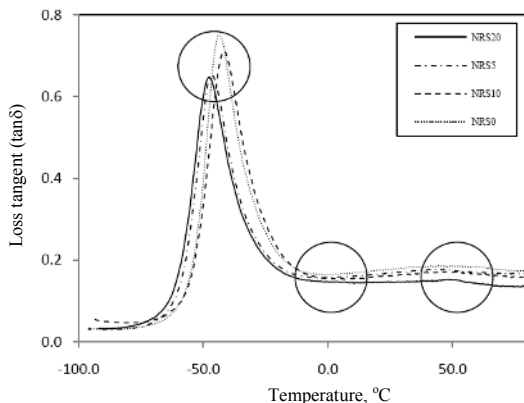


Figure 12. DMTA graph of samples at various silica ratios.

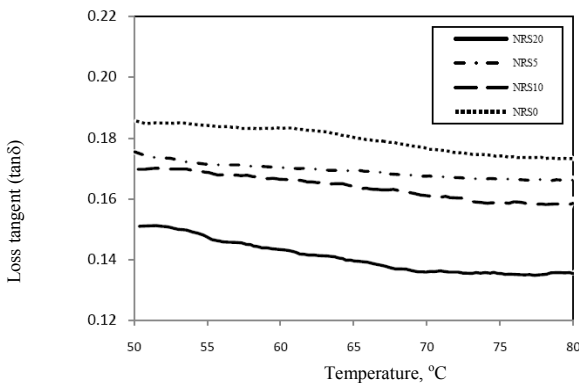


Figure 13. Range of temperature that criterion for measuring rolling resistance of samples at various silica ratios.

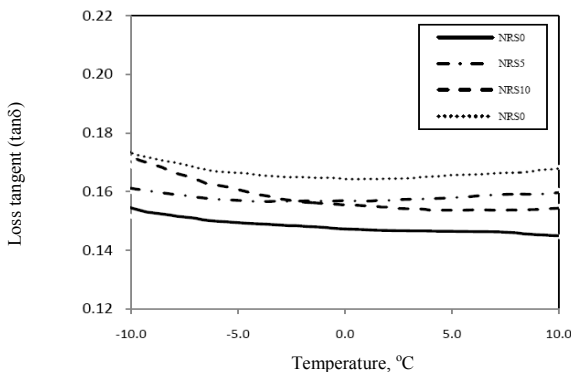


Figure 14. Range of temperature that criterion for measuring wet grip of samples at various silica ratios.

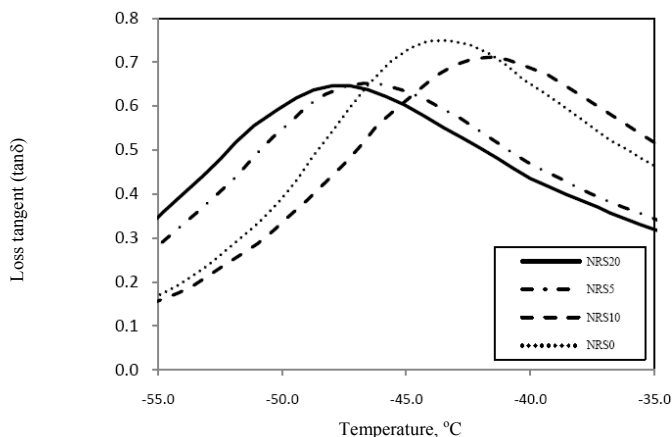


Figure 15. Tg of samples at various silica ratios.

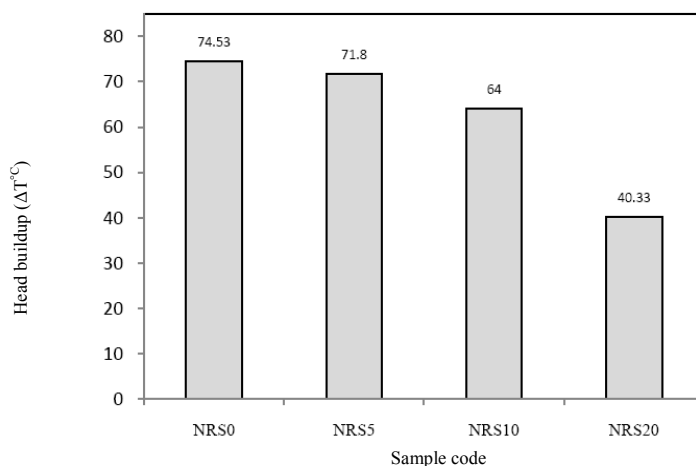


Figure 16. Heat buildup of samples at various silica ratios.

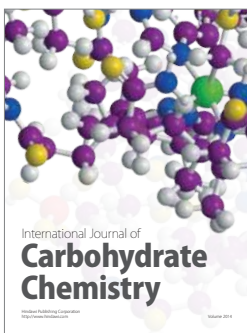
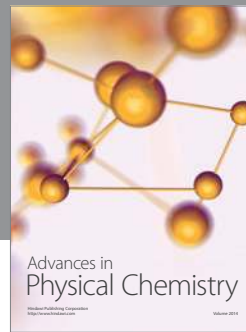
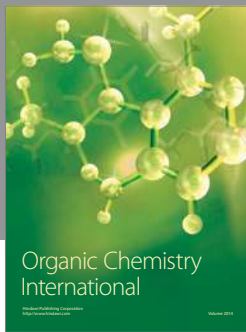
Conclusion

Using silica as a substitute for carbon black in the compounds of truck tires, at high amounts results in the decline of abrasion properties and consequently, the useful life of the tire. However, the significant improvement in its fatigue resistance increases fatigue life of the tire. Improved rolling resistance and decreased heat buildup, which are gained as the amount of silica increases, are the ultimate goals in designing tires. However, the decrease in the property of wet grip should be taken into account too. It should also be mentioned that increasing silica by high values will result in the decline of rubber's modulus and elasticity, which will impose its effect on the ultimate function of the tire.

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