EFFECT OF THE PERCENTAGE OF RUBBER COMPOSED BY NATURAL RUBBER, RUBBERIZED FILLED WITH SOOT ON THEIR TENSILE PROPERTIES AND BAKING BEHAVIOR

EFEITO DA PORCENTAGEM DE BORRACHA COMPOSTA DE BORRACHA NATURAL, EMBORRACHADA COM FULIGEM NAS PROPRIEDADES DE TRAÇÃO E COMPORTAMENTO DE COZIMENTO

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Abstract: *The purpose of the present studies is to provide a perspective of the main factor of the soot reinforcement effect as filler in composite with the base of natural rubber, rubberized or limited tires. For this purpose, natural rubber composites (NR-CB) were extracted by Soxhlet and thermal extraction, finally, Soxhlet extraction with the longest operating time removed more untreated rubber than thermal extraction and performed more efficiently and optimally.Using Tg curves to extract Soxhlet for 120 hours and thermal extraction for 24 hours for NR-CB N550 composite, the results obtained from the extraction step were evaluated.To further investigate the difference of soot type in the reinforcement of the composite, the aforementioned methods were performed with two natural rubber composites with two types of soot N550 and N660 after determining the carbon content of the gel, the results were compared with the results obtained from the measurement of soot surface structure and the physical and rheological properties of the composite.*

Keywords: *Composite, Soot, Carbon gel. Extraction.*

Resumo: O objetivo dos presentes estudos é fornecer uma visão do principal fator do efeito de reforço de fuligem como material de enchimento em composição à base de borracha natural, pneus emborrachados ou pneus limitados. Para este propósito, a composição de borracha natural (NR-CB) foram extraídos por Soxhlet e extração térmica; finalmente, a extração de Soxhlet com maior tempo de operação removeu mais borracha não tratada do que a extração térmica e teve um desempenho mais eficiente e ideal.Utilizando curvas Tg para extrair Soxhlet por 120 horas e extração térmica por 24 horas para o composto NR-CB N550, foram avaliados os resultados obtidos na etapa de extração.Para investigar melhor a diferença do tipo de fuligem no reforço do compósito, os métodos mencionados foram realizados com dois compósitos de borracha natural com dois tipos de fuligem N550 e N660 após a determinação do teor de carbono do gel, os resultados foram comparados com os resultados obtidos na medição da estrutura da superfície da fuligem e das propriedades físicas e reológicas da composição.

Palavras-chave: Composto, Fuligem, Gel de carbono. Extração.

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Introduction

Rubber is considered an important economic and strategic material and rubber has become an industry. Due to its unique properties, rubber composites are used in many fields including medical, automotive, defense, sports, etc., especially for their durability, strength, light weight, design flexibility and flexibility. Many of them are used in modern electronic devices which are very useful in everyday life such as computers, modems, printers, mobile phones, etc.

On the other hand, unprepared tires have two disadvantages; they soften during heating and they become hard and solid in the cold, for this reason they are not used as raw material and they use a blending process to produce a rubber piece with the proper properties.

Blending is a process to obtain the best formulation for combining several different types of materials with a selective rubber (elastomer) to achieve the following parameters: Desirable properties, better and easier vulcanization, more convenient shaping methods, achieving a more reasonable price, This process consists of three steps: 1) primary baking: Manufacture of rubber compound according to specific process instructions and conditions 2) Shaping: by special devices 3) Secondary baking: the operation of vulcanization.

So far, many researchers have investigated how elastomer-soot bonds (rubberized or confined rubber) are formed under the influence of mixing conditions, type and structure of elastomer, filler morphology, and amount of addition and surface properties. They also examine how these bonds affect the baking behavior and ultimately the mechanical properties, however, since a rubber composite is quite a complex system, it is very difficult to analyze this structure, especially at the molecular scale. Another contributing factor to the analysis is that the amount of bonded rubber depends strongly on the extraction method used to determine the percentage of rubber, the operating conditions of the extraction method and the extraction time.

The importance of doing this research is that the present study provides an insight into the role of environmental conditions on the amount of soot bonded rubber that is responsible for improving the desirable properties of a natural rubber composite through two methods of Soxhlet extraction and thermal extraction. Soxhlet extraction is a more common method. This method can be used for a longer period because they continuously heat the sample with a fresh thermal solvent. However, the Soxhlet extraction period is longer than the thermal extraction method. Both extractions methods appear to be suitable for extracting residual rubber gels in a composite. However, as we show in this study, even thermal extraction and Soxhlet extraction are not sufficient for complete separation of physically absorbed rubber.

Bonded rubber in a composite is mainly classified into two components. One is a polymer phase that surrounds the carbon surface as a shell, called “strong bonding rubber”. Another is a “weak link rubber” whose chains interact with the filler through a strong rubber bonding shell and thus become much more mobile than the inner shell. The first interaction (strong bonding rubber) can be as the formation of chemical bonds between the polymer molecules and the carbon surface and the second interaction (poorly bonded rubber) as the formation of physical bonds between the inner shell and the carbon surface.

The purpose of this study was to investigate the effect of activated soot surface on the amount of rubber sealant and its effect on the physical and mechanical properties of NR based rubber compound. Percentage bonded rubber extraction was performed by both Soxhlet and Thermal methods, which showed better results due to the longer operating time. In this research project, we study the effect of rubber bonds created between soot and elastomer which can affect the functional properties of a rubber compound as filler. We also investigate the effect of these bonds on the curing behavior and finally determine its mechanical properties.

Background

First Research: Use of carbon black as reinforce for mechanical properties and fatigue strength in natural rubber composite- Butadiene and natural rubber- Butadiene rubber -Styrene butadiene rubber.

The research was published in 2019 in the RESM journal by Engin Burgaz, Okan Gencog and Mert Goksuzoglu.

The aim of this study was to investigate the effect of addition of BR elastomer on the reinforcing properties of carbon black (physical and mechanical properties) in two types of composites prepared from NR / BR and NR / SBR / BR mixtures which eventually came to the conclusion that the presence of BR elastomer due to its high viscosity with black crane in a suitable composite and will improve the reinforcing effect of Black Crane, including increasing fatigue resistance and reducing bending coefficient.

Second Research: Effect of Black Carbon Filler on Mechanical Properties in Composites Made of Natural Base Rubber

The research was published in 2018 at Iopscience Journals by R Ismail, Z-A Mahadi Ishak.

In this study, five formulations with different amounts of N660 black soot filler for each sample with NR base elastomer were used and their tensile, hardness and flexural properties were studied which eventually came to the conclusion that increasing the amount of soot will increase the amount of tensile strength and hardness and reduce the amount of flexibility.

Third Research: Effect of addition time and carbon structure on tensile properties of natural rubber composite

The study was published in 2017 in the Iopscience Journal by S Savetlana, Zulhendri Sukmana FA Saputra.

Adding carbon black to natural rubber is very important to increase the resistance of natural rubber. In this study, the tensile strength of composite was investigated by varying the percentage of black crane at the weight percentages of 20, 25 and 30 and the effects of the carbon structures of N220, N330, N550 and N660 series.

The results show that adding black crane increases the tensile strength and modulus of elasticity of natural rubber composite. Using SEM electron microscopy, it was observed that the aggregation of CB mixture increased with particle loading. With increases the aggregation, the tensile strength of the composite decreases. The results of this structural investigation showed that Black carbon composite from N220 series highest tensile stress, carbon black reinforced composite from N660 series and N220 series having similar properties and N330 and N550 will reduce stress.

Fourth Research: The effect of fillers on natural rubber composite and their effect on rubber compounds

The research was published in 2017 by ScienceDirect Procedia Engineering by Pajtášová, Zuzana Mičicová, Darina Ondrušová, Beáta Pecušová, Andrea.

In this study, natural rubber composite was prepared using N339 type carbon black filler And the performance of these fillers in rheological properties (minimum torque - ML, maximum torque MH), Ts process safety, optimum Tc (90) vulcanization time as well as physical and mechanical properties (tensile strength, tensile strength, hardness) and heat have been investigated. In many industrial applications, elastomers are mixed with fillers to improve their performance properties.

Methodology

Chemical materials

The materials used in this study are shown in Table 1.

Table 1: Materials used

Row	Material		Manufacturer	Country	Application
1	Natural rubber		Bandar-e Emam petroleum	Iran	Base tires
2	Black soot	N550 N660	Pars tire company from Iran	Iran	Filler
3	Argon oil		Vahdat chemical	Iran	Process Assistance
4	ZnO		Shokohieh	Iran	Accelerator actuator
5	Stearic acid		Acid Cheml	Malaysia	Baking activator
6	Sulfur		Tesdack	Iran	Create crosslinks
7	Paraffin wax		Iran chemical	Iran	Softener
8	HB		Nanjing	China	Antioxidants
9	TMQ		Pars tire	Iran	accelerator
10	CBS		Qilon chemical	China	Anti-Ozone
11	6PDD		Lanxess	Belgium	Antioxidants

Devices and equipment

The apparatus and equipment used in this study are listed in Table 2.

Composite preparation equipment	Extraction equipment	Physical and rheological tests	
		device name	The standard used
Two roller bar	Soxhlet extraction	Rheometer	ASTM D2084
Hydraulic Press	Thermal extraction	density	ASTM D792
Other equipment		Attraction	ASTM D412
Heat distribution analyzer		Abrasion Resistance	ASTM D5963
Soot particle level measuring device		Fatigue resistance	ISO 6943

Experiment plan

Rubber compound formulation

Table (3) shows the formulation of the two rubber compounds made in this study. The first is the N550 series soot filler and the second is the N660 soot filler.

Table 3: Formulation of blender

Weight (g)	The amount in the blender (%)	Formula
198.6	100	NR
99.3	50	CB
9	5	ZNO
4	2	St-A
2	1	TMQ
2.4	1.2	6PPD
4	2	P.Wax
19.9	10	Ar-29 Oil
340	171.2	Total
310	-	Master
1.4	0.8	CBS

2.4	2.4	S
313.8 g		Total

Extraction

Extracts used in this study:

Soxhlet extraction: Small composite fragments (50 g total) were heated in a Soxhlet extractor for 120 h with 100 g of toluene at a temperature of about 150 ° C and the residual solids (carbon gel) after filtration and solvent separation were dried overnight at room temperature and then in vacuo for one day (Fig. 1).

Figure 1: Soxhlet extractor

Thermal extraction: Small composite pieces (50 g total) were heated for 24 h with 100 g of toluene at temperatures slightly above 150 ° C, and then the resulting solution of small solid particles is dispersed in it the filter was dried for one night at room temperature and then vacuum for one day.

Considering that the elastoker weight ratio of soot is 2: 1, the percentage of residual rubber was calculated by Equations 2-3 and 1-1.

Equation (1-1):

Equation (2-1):

W_1: Sample weight 50gr

W_2: Weight of sample after extraction

W_R: Weight of remaining

R: The percent of rubber gel remaining

Introducing Devices and Equipment

Measurement of soot particle level ((BET)):

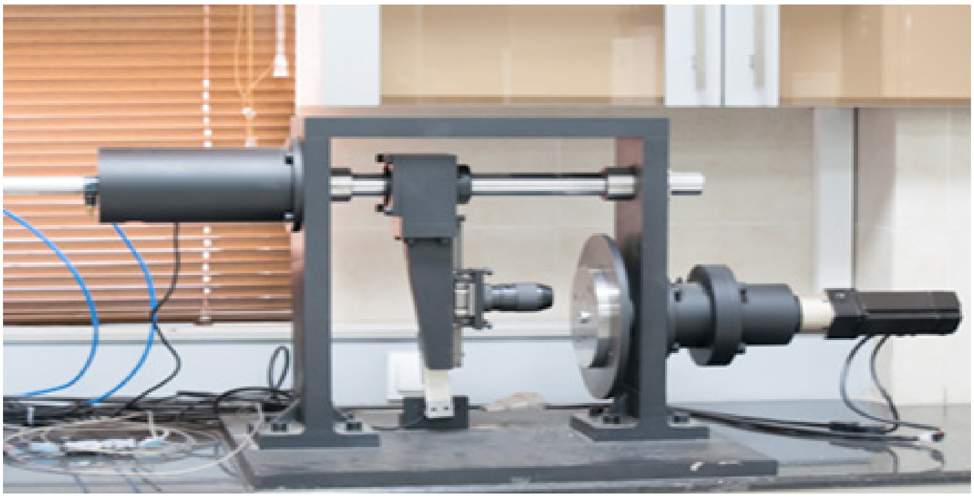
Specific Surface Measurement (BET) and porosity meter in many applications such as: Catalysts, nano-sorbents, compounds and additives, pharmaceuticals and the food industry as well as in nanostructures such as metal nanoparticles, nano-tubes, nano-fibers and so on are of great importance. Among the methods used to determine porosity, BET measurement using the adsorption-based Brunauer-Emmett--Teller theory has received much attention. In the BET method, a complete layer of molecules adsorbed onto the surface is formed. By knowing the average thickness of a molecule, one can calculate the area occupied by a molecule and measure the total surface area of the sample based on the amount of material absorbed. Effective surface area, physical adsorption, chemical adsorption and surface adsorption can be determined using BET and porosity meters.

Abrasion resistance measuring device:

Abrasion is the gradual disappearance of the surface of a tire in contact with a surface below and all abrasion tests are based on the relative motion between the tire and a surface below constant and clear force. There are various tests and methods for measuring tire abrasion but the major drawbacks of most of them are the inability to generalize the results from laboratory to practical. Often the abrasion test compares favorably between an ideal and a test case. The most common method of measuring wear is as a percentage of weight loss after the test. This means that a piece of a certain weight is subjected to an abrasion test and then weighed after testing.

This device tests the amount of abrasion resistance and uses a sliding pin motion on the disc to create abrasion. The common method is to make a disc made of steel (high hardness tool) and it is a piece of abrasion and the pin is the abrasive piece and the material being tested on the disk and on request the client can switch roles between the pin and disk. (Figure 2)

Figure 2: Abrasion resistance measuring device



The most common way to measure weight loss is after the test, this means that a piece of a certain weight is subjected to an abrasion test and then weighed after testing. Then the wear rate is calculated according to Equation 1-4.

Fatigue resistance measuring device

If the piece is affected by both thermal and mechanical stresses at the same time, its fatigue is called thermo mechanical fatigue. Thermo-mechanical fatigue plays an important role in the design of sensitive components and equipment that are subject to thermal and mechanical stresses.

The purpose of thermo mechanical fatigue test is to study the mechanisms of degradation due to thermal and mechanical cyclic stresses that are applied simultaneously to the part. One of the most important results of this test is the estimation of the lifetime of components such as gas turbine blades that are highly susceptible to this type of degradation. In this test, the stress (or strain) and temperature variables are controlled separately (Figure 3).

Figure 3: Fatigue resistance measuring device

Density test:

The density measurement is not specific to the raw blend, but is also used for the blended blend. Density is one of the simplest quantities to measure, while sometimes it contains valuable and useful information; all instruments used to measure density are based on the Archimedes principle.

Results

Measurement of physical and mechanical properties

Soot is widely used to produce tires for cars and trucks, because soot acts as a filler to enhance various mechanical properties such as tensile strength, tear strength and wear characteristics of tires. The soot reinforcing effect is mainly due to the strong interactions between the elastomer and the soot surface. Due to such strong interactions, when the elastomer and soot are mixed, some parts of the polymer cannot be soluble in the strongest solvents. This part of the insoluble polymer is called “limited and rubberized rubber”. Since the soot reinforcing effect on the composite is approximately correlated with the amount of rubberized rubber, its measurement is used as a screening test in the industry to evaluate the reinforcing effect. Structural analysis of the rubberized rubber is therefore crucial to understanding the reinforcing effect and further improving it. So far, many researchers have investigated how it is formed under various conditions, including elastomer-filler mixing, elastomer type and structure, filler morphology, amount of additives, and surface properties. They also examine how the rubberized rubber determines the final mechanical properties of the composite.

After preparation of composite specimens, the specimens were removed and tensile strength, longitudinal at rupture point, abrasion and fatigue tests were performed; the results are shown in Table 4. We will continue to investigate each.

Table 4: Results of mechanical tests

Tensile strength (Mpa)	Elongation at rupture point (%)	Abrasion (mm^3)Resistance	Density (g/cm^3)	Fatigue resistance (cycle)	Mix Test
20.01	612	258.12	1.11	259169	NR-CB 550
18.15	611	202.68	1.114	223568	NR-CB 660
ASTM D412	ASTM D412	ASTM D5963	ASTM D792	ISO 6943	test method

Tensile strength and longitudinal elongation at rupture point

In this study, two composites were tested for tensile strength. This test is one of the most important tests in the rubber industry, in that it strains the baking sample under stress to break it.

Tensile strength measurements are performed in accordance with ASTM D412. The specimen is positioned between the two jaws of the device and is inserted into the cross-section by a load cell embedded in force. The unit is the tensile strength of Mpa obtained from the force fraction in Newton to the cross-sectional area in mm.

For this test, in addition to the tensile strength values, the percentage of elongation at rupture point, tensile speed, and test temperature were also reported. These two parameters play an important role on the tensile properties and there will be a lot of changes with their rates. The results of this test are listed in Table 5.

Fig 4: Comparison of tensile strength of two composite types

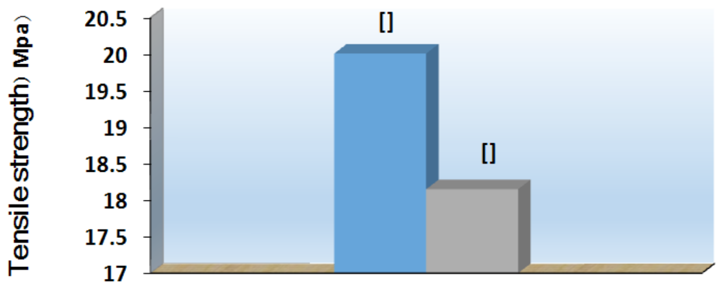
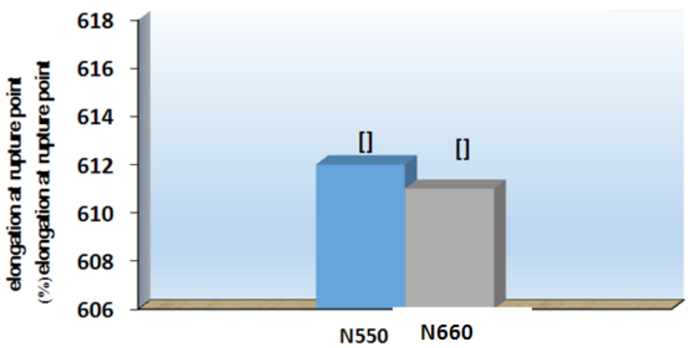


Fig 5: Comparison of rupture points in two composite types



Result: The tensile strength and tensile strength of NR-CB550 composite were higher than that of NR-CB660.

Interpretation: In this study, we investigated the effect of soot reinforcement on two properties of rubber soot and soot surface chemistry. Here's a look at the following about tensile strength:

Rubberized rubber: According to the results of the above table, the amount of residual rubber gel is higher in both Soxhlet extraction and thermal extraction in NR-CB N660 composite and in other words, the amount of rubberized rubber in this composite is lower.

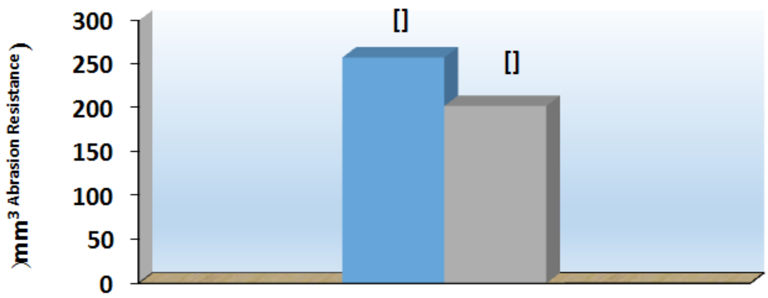
Returning to previous interpretations, it was stated that the amount of rubberized rubber used is a measure of the measured mechanical properties of the composite. For this reason, it can be concluded that the NR-CB550 composite is higher than the following: The N550 soot composite will be stronger due to the greater formation of strong elastomer-soot bonds.

Surface Chemistry: The higher the active surface area of the carbon particles and the smaller the particle size, the filler will be able to perform more and more effective interactions with the elastomer, which will strengthen the composite.

Abrasion Resistance

The abrasion resistance is measured according to ASTM D5963 (B). The results of this test are shown in the following diagram.

Fig 6: Comparison of abrasion resistance of two composites

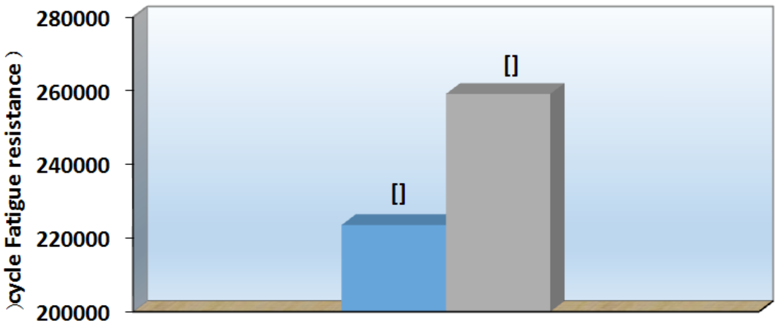


Fatigue resistance

Materials fail due to certain stresses. Materials under repetitive, oscillatory or rotational stresses due to shear or torsional stresses in the material, at a stress much less than the stress required to break a single load, they will fail which is called "fatigue failures", Different devices have been designed to study the fatigue phenomenon in different materials, depending on the material and the forces applied, capable of applying reciprocating or rotating cycles of over one million rounds and even some devices are capable of applying torsional torque to the piece.

In fatigue machines with rotary mechanisms, the stress mechanism is the weight applied to the alternating axial force for the rubber piece. In these devices, the number of cycles struck indicates the life-time of components before failure due to fatigue. The design of this test is in accordance with ISO 6943. The results of this test are listed below.

Figure (7): Comparison of fatigue strength of two composites



Result: Fatigue resistance in NR-CB550 composite is higher than NR-CB660. It should be noted that fatigue and wear resistance, as well as tensile strength and tearing properties, are among the composite strength indices and the commentary mentioned in the other section applies to the two.

Discussion

Investigation of extraction results

In this study, by calculating the residual gel fraction (rubberized), we conclude by using two types of Soxhlet and thermal extraction: Longer Soxhlet Extraction Performance Causes Removal of More Rubber Polymers with weaker Elastomer-Soot Bonds.

Evaluation of the results of BET measurements

From the surface chemistry point of view, the reason for the difference between the percentage of gel remaining in the two series of thermal extraction and Soxhlet can be expressed as follows: the higher the specific activity of the surface of the carbon particles, the stronger the bond between the soot and the elastomer, resulting in a higher percentage of rubber.

Evaluation of the results of thermal weighing analysis (TGA)

From the rapid weight loss in the temperature range of 300 to 500 ° C, the percentage of residual rubber gel from the Soxhlet extracted sample for 120 h and hot extracted sample for 24 h was calculated as 25 and 35%, respectively. These results corroborate the data obtained from the above extraction operations.

Evaluation of the effect of soot type on physical and mechanical properties

The main factor in the strength of the composite is the higher amount of strong rubber bonds than the weak rubber bonds in the composite. Therefore, the higher amount of rubberized rubber in an extraction, the stronger the result.

The effect of soot on baking time

In general, soot accelerates the speed of reactions leading to the vulcanization process and increases the speed of the baking process. The cooking speed variations will vary with the influence of the soot type associated with the surface chemistry changes and the amount of rubber produced. It is stated in Section 4.3 that: The higher the specific activated surface of the soot and the percentage of rubber impregnated, the composite will achieve its optimum properties by forming more elastomer-soot bonds faster during the curing process, the baking weight will be less.

Conclusion

In this research we tried to: Investigate the effect of soot as filler on the rate of composite reinforcement by examining the results of two types of thermal extraction and Soxhlet with both N660 and N550 soot.

Finally, we conclude that in a composite curing process, smaller particles and higher activated carbon surface area in the soot can be sintered faster by achieving higher percentages of rubberized rubber with stronger elastomer-soot bonds.

Another result is that the duration of operation is one of the most important factors in determining the extraction efficiency, which can be achieved by better extraction results.

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