Effect of Carbon Black on Mechanical and Physical Properties of Acrylonitrile Butadiene Rubber (NBR) Composite

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ABSTRACT

This research aims to study the effect of addition carbon black material (type N-326) on the mechanical and physical properties of nitrile rubber (NBR) and the proportion of addition carbon black material is (0, 20, 40, 60, 80 pphr (part per hundred rubber)) to afixed percentage of nitrile rubber (100%) have been prepared with (factors of vulcanization, accelerators and activators using two-roll mill laboratory, molds, thermal piston and difference of examination conditions from pressure, temperature and time according to the ASTM), all tests were conducted in the laboratories of the College of materials Engineering - University of Babylon and laboratories of babylon tire company. Then carrying out the process of scheduling the results of laboratory testing and study its diagrams.

Where the results showed the mechanical properties of an increase in (the properties of tensile strength, hardness, modulus of elasticity, compression set, and tear resistance increase with the increasing percentages load until a certain levels of load bearing (60 pphr) and then decreases slightly, and a decrease in properties (elongation at break and resilience).

While the results of the physical properties show increase in viscosity, the greatest torque twisting, but scorch time and cure time decreases with the increasing the proportion of carbon black in the rubber recipe. specific gravity also increase with increasing carbon black material.

Keywords: Nitrile rubber (NBR), Carbon black, Mechanical and Physical Properties

INTRODUCTION

Rubber without reinforcement has limited applications, so that be added polymeric materials including carbon black to the rubber to reduce the cost and improve the mechanical and physical properties, that meaning the study of mechanical and physical properties of various engineering materials is considered important because of its engineering applications.

The growing use of rubber in engineering applications results from its unique properties that include high extensibility, high strength, high-energy absorption, and high resistance to fatigue. Other attributes are good environmental resistance and high resilience [1].

Rubber in its nature has weak properties and it has no benefit at all therefore in order to get high performance it must be vulcanized. Vulcanization, named after Vulcan, the Roman God of Fire, describes the process by which physically soft, compounded rubber materials are converted into high-quality products. One of the most important ways to

get high-quality rubber is reinforcement by rigid entities which improves the mechanical properties and other properties depending on the nature of the reinforcement such as carbon black, silica, oxides, clays. The incorporation of fillers into elastomer matrices usually conduces to a significant increase in their mechanical properties, to an improved abrasion and tear resistance. The rubber-filler interactions can be considered as an important parameter affecting such properties [2].

Nitrile Rubber (NBR) is commonly considered the workhorse of the industrial and automotive rubber products industries. NBR is actually a complex family of unsaturated copolymers of acrylonitrile and butadiene. By selecting an elastomer with the appropriate acrylonitrile content in balance with other properties, the rubber compounder can use NBR in a wide variety of application areas requiring oil, fuel, and chemical resistance. In the automotive area, NBR is used in fuel and oil handling hose, seals and grommets, and water handling applications [3].

With a temperature range of -40° C to $+125^{\circ}$ C, NBR materials can withstand all but the most severe automotive applications. On the industrial side NBR finds uses in roll covers, hydraulic hoses, conveyor belting, graphic arts, oil field packers, and seals for all kinds of plumbing and appliance applications. Worldwide consumption of NBR is expected to reach 368,000 metric tons annually by the year 2005 [3].

NBR is produced in an emulsion polymerization system [4].

Carbon black is the most widely used reinforcing filler in the rubber industry. carbon black is prepared by incomplete combustion or by thermal cracking of hydrocarbons. Carbon black from incomplete burning of natural gas is acidic on the surface. carbon black can also be produced by thermal cracking of hydrocarbons in the absence of oxygen and they have relatively low specific surface area (6-15 m2/g) [5].

Carbon black particles typically have their sized ranging from 20 to 300 nm [5]. There are three morphological forms of carbon black existing in rubber composites: particle, aggregate, and agglomerate. The sizes of these morphological forms have the following order: particle < aggregate < agglomerate.

Reinforcement of rubber by carbon black depends on many variables: surface area, structure, surface activity, dispersion, and loading. Surface area and surface activity of carbon black play an important role in its interaction with rubber chains. The high surface area means the small size of the carbon black particles in the same weight unit. It is also known that the higher the surface area the harder the even dispersion of carbon black into the rubber matrix [6]. The interactions between rubber and carbon black can be through either physical adsorption or chemical bonding. It is scientific studies in this area are as follows:

Mostafa et al.2009 [7]. The researchers used carbon black (CB) to enhance the mechanical properties of styrene butadiene (SBR) and nitrile butadiene (NBR) rubber compounds compared with unfilled compounds. In this study, the effect of carbon black on the swelling and compression set behavior of SBR and NBR compounds was investigated.

The obtained results of five different compositions for SBR and NBR with 0, 20, 30, 50 and 70 pphr of CB were compared. From the current investigation it was found that the increase in carbon black loading inherent with drastically reduction in swelling percentage while the compression set increases in both SBR-filled compounds and NBR-filled compounds, This behavior is more pronounced in NBR filled compounds than SBR filled compounds. This investigation clearly distinguishes the influence of vconditioning environment to the properties under study.

Muhannad K. D. 2007[8]. The researcher studied reinforcement technique –with copolymer matrix Elastomer (Nitrile butadiene rubber), and carbon black as follows:

With the addition of carbon black on the expense of silica percent in a balance of 50% (carbon + silica) we get the best mechanical properties ratio with % carbon +% silica .The hardness, tensile, impact and wear resistance are 100 IRHD, 22.865 Mpa , 18.1 R% , 0.003 gm respectively which are agreed with the required properties from the plant.The best type of the rubber composite is selected according to the rubber mechanical and physical properties.

AIM OF RESEARCH

The aim of this research to study the effect of adding variable amounts of carbon black on various properties of nitrile rubber.

MATERIALS AND METHOD

Materials Used

Nitrile rubber (NBR), Zinc oxide, Stearic acid, Tetramethylthiuramdisulfide (TMTD), Mercaptobenzothiazoldisulfide (MBTS), Castor oil, Sulfur, 2,2,4-trimethyl-1,2-dihydroquinoline (TMQ) and Carbon black (N 326) used in this study were supplied by Babylon tire company.

Preparation Method

The formulation of the rubber composites are given in Tables (1).

| Item No. | Recipe Ingredients | Loading level (pphr) | | | | |
|-------------|--------------------------|----------------------|-----|-----|-----|-----|
| | | (1) | (2) | (3) | (4) | (5) |
| 1 | NBR | 100 | 100 | 100 | 100 | 100 |
| 2 | Activator (zinc oxide) | 3 | 3 | 3 | 3 | 3 |
| 3 | Activator (stearic acid) | 1 | 1 | 1 | 1 | 1 |
| 4 | Carbon black N-326 | 0 | 20 | 40 | 60 | 80 |
| 5 | Castor oil | 10 | 10 | 10 | 10 | 10 |
| 6 | Anti-Oxidant (TMQ) | 3 | 3 | 3 | 3 | 3 |
| 7 | Accelerator (TMTD) | 2 | 2 | 2 | 2 | 2 |
| 8 | Accelerator (MBTS) | 1 | 1 | 1 | 1 | 1 |
| 9 | Sulfur | 1.5 | 1.5 | 1.5 | 1.5 | 1.5 |

Table 1. The composition of the rubber composites

Mixing was carried out in a two-roll mill, operations of kneading and mixing took place on this Equipment according to ASTM D 15.

The ingredients were added according to ASTM D3187-89 as follows:

- 1. Passing NBR between the two rolls several times with decreasing the distance between the two rolls to the extent of (0.5-1) mm.
- 2. Addition of sulfur follow by mixing for homogenization of the materials under room temperature.
- 3. Add stearic acid, zinc oxide, with the remaining quantity from sulfur step (2) together, follow by mixing for homogenization of the materials.

- 4. Add half of the carbon black with operating oil evenly across the mill at a uniform rate.
- 5. When this portion of the carbon black has been completely incorporated, open the rolls to 1.65 mm (0.065 in.).
- 6. Add the remaining carbon black and oil evenly across the rolls at a uniform rate.
- 7. Addition of accelerator TMTD and MBTS With the addition of antioxidant TMQ This is followed by mixing materials for the purpose of homogenization.
- 8. Set the rolls at 0.8 mm (0.032 in.), follow by mixing for homogenization of the materials.
- 9. Open the mill to give a minimum batch thickness of 6 mm (0.25 in.) and The mixing process continues for more time in order to get agood homogenization.

After preparation of recipes rubber composite by two-roll mill laboratory, examination models are prepared to provide (moldings and condition of examination such as temperature, pressure and time) then a process of vulcanization by the thermal piston, according to the ASTM.

CHARACTERIZATION AND TESTING

Mechanical Properties Test

Tensile, elongation, and modulus Tests

Tests are carried on samples which are prepared (Cutting three dumbbell specimen) from the vulcanized slice. the cutting take place by hand press.and according to the ASTM d412-88 by Monsanto T10 tensometer by using special jaws for holding tensile test sample which is movable by speed of 500 mm /min.

Tear Resistance Test

Examination carried out according to standard ASTM D624-54, by a device tensile (Tansometer10) and speed (500 mm/min) after setting the model examination (dumbbell) by cutting machine models and the work of the incision in the narrow area of the model by cutting machine (Nicking cutter) type (Wallace). Then examination by a Tansometer10 to give the results (N / mm).

Hardness Test

The test was carried out under ASTM-D2240 Standard using durometer (Hardness shore A). standardized hardness-measuring equipment using a sharp needle was applied directly on to the surface of specimens to measure hardness. data were averaged over six different positions.

Specific gravity Test

The density tester machine is used . the specification of this machine were digital accuracy = 0.0001 g/cm^3 , type GP-120 S, (Matsu Haku, China). first weight the sample (hardness sample) in air then in water at 23 °C, by using the equation (Specific gravity = [sample weight in air/(sample weight in air – sample weight in water)] * Specific gravity of liquid)

Resilience Test

Resiliency is checked according to specification (ASTM D1054) using device (Wallace Dunlop Tripsometer) and linked to a digital computer information "Wrpra-wallace Rebound Resilience Data Computer" records the percentage of resilient.

Adhesion Test

Tests are carried on prepared samples according to the ASTM 2229 by tensometer T10 in which we exchange the special jaws for adhesive test which is movable with speed of 50 mm /min for measurement the adhesion with wires and with textile .

Compression Set Test

Compression set test (ASTM D395) was performed on standard test specimen of cylindrical shape of 29 ± 0.5 mm diameter and 12.5 ± 0.5 mm thickness. Samples were compressed to 0.40 of its thickness for 72 hours in compression set device, then it is left for 30 min before taking the final thickness of the samples.

Physical Properties Test

Viscosity and Curing Characteristics Test

The test carry on according to ASTM D2084-89 by using Oscillating dick Rheometer (ODR) for samples prepared (cutting from recipes rubber composite) in which its control by upper and lower jaws clutch under 20 Pa and 185 °C for 6 min.

The result draw as a graph of Torque vs. Vulcanized time as pointed out the following data (max. torque (1b-in), min. torque (1b-in), scorch time (min), cure time (min), viscosity (ML^{*} 2.71)). The rehometer links to a computer with a precision control program.

RESULTS AND DISCUSSIONS

Discuss the results of the mechanical properties Test

Tensile (Mpa), elongation (%, and modulus (100% Mpa) Tests.

To determine properties of the material under the influence of axial tension load in one direction of the sample (longitudinal axis) the pregnancy is gradual from zero and even the failure occurs quickly (500mm / min).

Several models are examined and taking the average to get the desired result for the disposal of individual differences and reduce the error rate resulting from the heterogeneity of the sample.

In figure 1 shows high tensile strength with a high proportion of carbon black in rubber recipes because of the large surface area and zigzag for carbon black and at its contact with the rubber (NBR) creates a strong network structures which increase the mechanical properties of the new compound, and make composite materials resistant to flatten and drag, and continue this resistance with increasing carbon black, but when highest load percentages (60 pphr) and above) begin tensile strength decreases due to the increased fragility of rubber compound, and this result is corresponds to the results of other researchers [9,10].

While in figure 2 shows the rate of elongation decreases with increasing ratio of carbon black and the reason for that is the increase in carbon black which reduces the amount of the foundation rubber material surrounding carbon black granules which increases the fragility and hardening rubber compound and then lower elongation at break, and this result is corresponds to the results of other researchers [9,11].

The modulus of elasticity shown in Figure 3 increased with the increase of carbon black for the same reasons, to increase the tensile strength above, and this result is corresponds to the results of other researchers [9,11].



Figure 1. Effect of CB. (pphr) on the tensile strength of NBR matrix composite.



Figure 2. Effect of CB. (pphr) on the Elongation of NBR matrix composite.



Figure3. Effect of CB. (pphr) on the Elastic Modulus of NBR matrix composite.

Tear resistance Test (Mpa)

Through figure 4 observes the effect of adding carbon black to resist rupture (tear resistance), which is measured in units of (N/mm). Where we find that recipes rubber increase the tear resistance with the increasing percentages load until a certain levels of load bearing (60 pphr) and then decreases slightly.



Figure 4. Effect of CB. (pphr) on the Tear resistance of NBR matrix composite.

This is because of the increased intricacies chains but after percentage (60 pphr) and increased fragility of composite, a few decrease occurred in tear resistance. *Hardness Test* (ShA)

Examination depends on measuring the penetration of the rigid ball in a sample of rubber and the force be shed by pregnancy. The hardness is measured for samples prepared previously cylindrical shape from the middle and sides and took the rate to reach the best result and avoid mistakes.

When adding the type of carbon black (N-326), which has the particle size (350) Micron to synthetic rubber (nitrile rubber) in different proportions (0,20,40,60,80 pphr), note an increase in the hardness values, because the granules carbon black have a surface area of active and zigzagging which increases entanglements between them and the rubber chains when contact with the rubber, which increases resistance to penetrate the outer body of compound, as a result of increased surface tension, as shown in Figure 5, and this result is corresponds to the results of other researchers [9,10,8].



Figure 5. Effect of CB. (pphr) on the Hardness of NBR matrix composite.

Specific gravity Test

Carbon black increases the specific gravity of synthetic rubber (NBR) and as shown in figure 6, and this is attributed to the carbon black which has a specific gravity range (1.8 - 2.1), the highest of nitrile rubber, which has specific gravity(1.09 g/cm^3).



Figure 6. Effect of CB. (pphr) on the Specific gravity of NBR matrix composite.

Resilience Test

Note in Figure 7 decreases in the values of resilience and the reason is due to the increase in the hardness of the material rubber compound with an increased proportion of carbon black,

and this gives greater portability rubber material to absorb kinetic energy and converts it into thermal energy and portability to less traceable, and this result is consistent with previous research [9].



Figure 7. Effect of CB. (pphr) on the (Resilience) of NBR matrix composite.

Compression set Test (%)

The compression set increases with the increasing carbon black this is because of the increase in the density of entanglements (the same reasons for the increase of tensile strength above) and a decrease in the movement of rubber chains, and increase the rigidity or stiffness compound rubber, and these entanglements reduce portability retrieval of compound rubber, and this result corresponds to the two searches of former [12,7].





Discuss the Results of the Physical Properties Test

Viscosity (Mooney) & Max. Torque (MH lb*inch).

Figure 9 shows an increase in the values of the viscosity (which are an indication of the plasticity product rubber and its ability to manufacturing) of the composite rubber material with the increasing in carbon black, due to increased linkages within the chains and decrease the free movement of rubber chains, namely that the cohesion between the grains of carbon and rubber chains will be larger.

The same figure 9 shows the high torque and continuously with the increasing carbon black, for the same reasons, increase the viscosity above, and these results correspond with the results of a previous search [9].



Figure 9. Effect of CB. (pphr) on the (Viscosity, Max. Torque) of NBR composite.

Scorch Time (TS5 min) & Cure Time (TC90 min).

Figure 10 shows a decrease in the scorch time to increase carbon black (increasing the surface area of the granules of carbon due to meandering in the surface of carbon), this means that carbon black is working to deliver the heat needed to complete the process of vulcanization of the internal structures of recipes rubber, due to its interaction with the crosslinks interrelation, as well as the cure time decreases with the increasing the proportion of carbon black in the rubber recipe, where the carbon black enters in interactions with crosslinks interrelation and thus causing reaching to the case of early ripening [9].



Figure 10. Effect of CB. (pphr) on the (Scorch Time, Cure Time) of NBR composite.

CONCLUSIONS

- 1. New polymer composites were prepared from the mixing carbon black with (NBR).
- 2. Tensile strength increase (from 1.830 Mpa to 16.406 Mpa) with increasing the carbon black content.
- 3. Elongation at break decrease (from 814 % to 681 %) with increasing the carbon black content .
- 4. modulus of elasticty increase (from 0.88 Mpa to 1.59 Mpa) with increasing the carbon black content .
- 5. Hardness increase (from 52.5 to 69.6 ShA) with increasing the novolac content.

- 6. Other prperties such as Tear resistance, Compression set and Specific gravity increase, but resilince decrease with increasing the carbon black content.
- 7. Physical Properties such as Viscosity, Max.Torque, Scorch Time, Cure Time, its values volatile with increasing the carbon black content.

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