A Study of The Effect of Carbon Black Powder on The Physical Properties of SBR/NR Blends Used In Passenger Tire Treads

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Abstract

The use of polymer blends is an effective method for altering the performance of polymer materials and is widely employed in engineering plastics, rubber, and fiber materials. Elastomeric blends are used for many reasons such as lowering the compound cost, for ease of fabrication and to improve the performance of the industrial rubber. In this study, 20 different rubber compounds were prepared by using SMR-20 type of natural rubber and SBR-1502 type of styrene–butadiene rubber in different levels (0, 25, 50, 75 and 100 pphr) and each recipe reinforced with carbon black at different ratio (20, 40, 60 and 80) pphr (part per hundred). The physical properties – such as thermal conductivity, thermal diffusivity, swelling and specific gravity were studied. The results show that the thermal conductivity and thermal diffusivity increased with increase of loading level of carbon black and these properties decreased with the increase of NR to SBR. The swelling property in this work measured by change in mass method by immersed the samples in oil and water. The swelling in water more than in oil, change in mass of the NR/SBR blends increased with immersing time increase and decreased with carbon black increment, and with the increment of NR percentages to SBR. The specific gravity increased with increase of carbon black loading for all recipes.

Keywords: Elastomer blend, carbon black, physical properties, swelling, thermal conductivity

دراسة تاثير اضافة الكربون الاسود على الخصائص الفيزيائية لخلائط المطاط المستخدمة في نفس الاطار (SBRINR)

الخلاصة

استخدام الخلائط البوليمرات هي طريقة فعالة لتعديل اداء المواد البوليمرية وكذلك انها تستخدم وعلى نطاق واسع في المواد الهندسية مثل (البلاستك, المطاط والالياف), ان خلائط المواد المطاطية المرنة تستخدم لاسباب عديدة مثل خفظ كلفة المركب, سهولة التصنيع وتحسين اداء المطاط المنتج. تم في هذا البحث تحضير (20) خلطة حضرت بواسطة استخدام المطاط الطبيعي نوع (SMR 20) ومطاط الستايرين بيوتادين نوع (1502) بنسب تحميل مختلفة (0, 25, 50, 75, 100 pphr) وكل خلطة مقواة بالكربون الاسود وبنسب تحميل مختلفة (20, 40, 60, 80 pphr) . تم أجراء العديد من الاختبارات الفيزيائية لغرض تحديد خواص المادة المتراكبة المحضرة, مثل (الموصلية الحرارية, الانتشارية الحرارية, الانتفاخية و الكثافة النوعية). حيث نلاحظ ان الموصلية والانتشارية الحرارية تزداد مع زيادة الكربون الاسود وانخفاض مستوى المطاط الطبيعي الى الصناعي, خاصية الانتفاخ في البحث تم حسابها من طريقة التغير في الوزن من خلال غمر النماذج في

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الزيت والماء. حيث لوحظ ان الانتفاخ في الماء اكثر منه بالزيت. و لوحظ ان التغير بالوزن لجميع الخلائط ازداد مع زيادة زمن الغمر وقلت مع ازدياد الكربون الاسود ومع زيادة نسبة المطاط الطبيعي الى الصناعي. وكذلك الوزن النوعي لجميع الخلائط ازداد مع زيادة الكربون الاسود .

Introduction

Blending of two or more types of polymer is a useful technique for preparation and developing materials with properties superior to those of individual constituents, this statement is also true with rubber blends, especially in tire manufacture [1, 2]. Styrene Butadiene Rubber (SBR) is now the most common synthetic rubber being used in tires. It is made by polymerizing Styrene and Butadiene together; it is also possible by changing Styrene content and polymerization process to make various types of SBR's with different characteristics. Natural rubber (NR) is known to exhibit numerous outstanding properties, such as good oil resistance, low gas permeability, improved wet grip and rolling resistance, coupled with high strength; having properties resembling those of synthetic rubbers. Natural rubber coming from latex is mostly polymerized isoprene with a small percentage of impurities in it. This will limit the range of properties available to it, although addition of sulfur and vulcanization is used to improve the mechanical and physical properties properties. Chemically, natural rubber is cis- 1,4-polyisoprene and occurs in hevea rubber trees [3, 4 and 5].To improve the mechanical and physical properties of vulcanized rubber compounds, carbon black has been traditionally used as a major reinforcing material together with a few minor reinforcing materials such as clay, calcium carbonate and silicates since the excellent reinforcement with carbon black for rubbers was elucidated in early twentieth century, of which the most important reinforcement, reduction in material costs, and improvements in processing. Reinforcement is primarily enhancement of strength and strength related properties such as abrasion resistance, hardness, and modulus. A wide variety of particulate fillers is used in the rubber industry for various purposes [6, 7, and 8].

The use of carbon black is synonymous with the history of tires. However, the primary properties of carb on blacks are normally controlled by particle size, surface area, structure and surface activity which in most cases are interrelated [9].

The idea of blending synthetic rubbers with natural rubber is certainly not a new one, but now it can be applied positively, by using new techniques developed over the last 5 years. These compounds are capable of forming a chemical link between these dissimilar rubbers to produce a technologically compatible blend. The blend vulcanizates produced exhibit enhanced physical properties by judicious selection of the SBR: NR ratio [2 and 10].

Blending of (SBR) and other types of rubber and its perforamance have been studied earlier [11, 12, 13 and 14], they have demonstrated that the

physical properties of such blends can be significantly improved by adding a suitable compatibiliser.

This work aims to improve the recipe properties of tire tread by adding the reinforcing fillers (carbon black) at different loading level in addition to the additive materials like (ZnO, steric acid, sulfur…etc) to elastomers styrene butadiene rubber SBR and natural rubber NR separately and blends of (SBR/NR) at different ratio.

To optimize:

- The effect of the blending and loading in phr of NR with SBR.
- The effect of loading level of carbon black (N220).

These properties are: thermal conductivity, thermal diffusivity, swelling (effect of liquid) and specific gravity.

Experimental

Materials

All materials used in this research come from al Dewania factory for tire manufacture, Iraq. The structure of these materials is as follows

- Styrene- Butadiene Rubber (SBR), with Styrene content 23.5%: Moony viscosity at 100 ^{0}C = 50; specific gravity 0.94 (g/cm^3); ash content 1%.
- Natural Rubber (NR), with specific gravity 0.934 (g/cm^3); ash content 1%.
- Carbon black (N220): Black granulated powder has Pour Density 345 ± 30, (kg/m^3). External Surface Area 111 ± 5 (m^2/g) DBP absorption number 114 ± 4 (cm^3/100 g).
- N-cyclohexyl-2-benzothiazole sulfonamide (Vulkacit CZ): pale grey; non hygroscopic powder; melting point 95–100 ^{0}C and specific gravity 1.27–1.31 (g/ cm^3).
- Antioxidant (TMQ): specific gravity 1.08 (g /cm^3)
- Antiozonant (6PPD) is a material of composition [N-(1, 3- dimethylbutyl)-N-phenyl-p-phenylenediamine]: specific gravity 1.0 (g / cm^3).
- Sulfur: Pale yellow powder of sulfur element; purity 99.9%; melting point 112 ^{0}C; specific gravity 2.04– 2.06 (g/ cm^3).
- Zinc oxide: fine powder; purity 99 %; specific gravity 5.6 (g/cm^3).
- Stearic acid: melting point 67–69 ^{o}C; specific gravity 0.838 (g /cm^3).

Recipes formulation used in this work

The rubber compounds used in this work were prepared from natural rubber (SMR-20) and styrene-butadiene rubber (SBR-1502). Twenty carbon black (N220) filled recipes with other compounding materials such as filler; vulcanizing agent (sulfur) and accelerator were prepared with the compound formulations given in table (1).

Mixing and mastication

Mixing and mastication are conveniently done using two roll mills (Calender) and or internal mixer called (Banbury); the first one is used in this work. The mixing operation was executed on two stages, as shown in table (2). The first one called ***master batch*** consist of rubbers (including reclaimed), activators, antioxidant / antiozonants, reinforcing agent carbon

black and processed oil. At the end of the first stage carbon black blended with process oil in order to have optimum dispersion and coupling with rubber. The second stage is called the ***final batch***. It consists of the previous master batch, curing agent (Sulfur), retarder and accelerators. These materials are added at the end of process to prevent pre-vulcanization which may occur due to the elevated temperatures, the test specimens were die cut from test slabs.

Physical Properties

Thermal conductivity

The measurements of the thermal conductivity (k) were performed by (Lee's disc apparatus) type (Griffin and George) and some accessories to measure the temperature of both sides of the rubber specimen in order to calculate the thermal conductivity. The specimens are cutting as disks with about 40-mm diameter and 4-mm thickness were prepared for thermal diffusivity measurements. The heater is switch on from the power supply with (V = 6 V and I = 0.2 A) to heat the brass disks (2, and 3) and the temperatures of the all disks increases in nonlinear relationships and at different rates with the time according to its position from the heat source. And the temperatures were recorded every (5 minutes) until reach to the equilibrium temperature of all disks. When a steady state of energy is obtained, i.e. when input energy equals output energy as represented in equation (1), where T_1, T_2, and T_3 are recorded temperatures.

Input energy = Output energy

$$\mathbf{i \times v = \Pi r^2}.e\,(T_1+T_3) + 2\Pi.r.e\,[\,d_1T_1+d\,(\frac{\mathbf{T_1+T_2}}{\mathbf{2}})+d_2\,T_2+d_3T_3] \quad \text{..... (1)}$$

From above equation (1), the value of (e) obtained is applied in equation (2) to compute the coefficient of thermal conductivity (k).

$$k\frac{(T_2-T_1)}{d}=e.[T_1+\frac{2}{r}(d_1+\frac{d}{2})T_1+\frac{1}{r}dT_2] \quad \text{..... (2)}$$

Where:
Q: heat flux (w/m^2).
k:Thermal conductivity coefficient($w/m.^oc$)
i: Electric current (A).
v: voltage (volt).
r: Radius of disk (mm).
e: Heat loss ($w/m^2.^oc$).
d: Thickness of disk (mm).

Thermal diffusivity

The measurements of the thermal diffusivity (α) were performed by a laser flash device (model TC-7000H / MELT Ulvac-Riko, Yokohama, Japan shown in figure (1). Disks with about 10-mm diameter and 2-mm thickness were prepared for thermal diffusivity measurements. The thermal diffusivity, α, is calculated from the following equation:

$$\alpha = \frac{\mathbf{1.37 * L^2}}{\boldsymbol{\pi^2 * t_{1/2}}} \quad \text{……. (3)}$$

Where:
L: Thickness of specimen (mm).
$t_{1/2}$ **:** Half rise time (sec).

Swelling

To estimate the percentage swelling (change in mass), specimens were cutting as shown in figure (2) and weighed accurately in air, and then half

of them immersed in water and the other half immersed in oil for (22, 46, 72, 166, 670 and 1006 hrs) at room temperature. This test is done according to ASTM D 471. The test specimens were then removed and dried by filter paper and the mass was determined. In this work, change in mass method is employed when immersed rubber specimens are under specific temperature and time. Calculation of this method is represented in the following equation.

$$\text{Change in Mass \%} = [(w_2 - w_1) / w_1] * 100 \quad \ldots\ldots. (4)$$

Where: -

w_1: Mass of specimen before immersion (g).

w_2: Mass of specimen after immersion (g).

Specific gravity

This test is carried out according to ASTM (792), samples prepared as shown in figure (3), the volume of each one are (2 cm^3). In this test the specimens are weighted in air and then weighted suspended in water. The obtained values are applied in the following equation.

Specific gravity of material = weight of material in air/ (weight of material in air – weight of body suspended in water) * Sp.Gr. of water …… (5)

Results and discussions

The variation of thermal conductivity with carbon black for SBR/NR composites with varying NR content is shown in figure (4). This figure shows that the thermal conductivity is directly proportional to the loading level of carbon black depending on the rule of mixture, so that the thermal conductivity of composite is governed by its component amounts and their properties. Carbon black improves the thermal conductivity of rubber composite due to the thermal conductivity of carbon black greater than the thermal conductivity of rubbers .In the dispersion system with low loading level of carbon black, few particles contribute to form conductive chains, and at this time the matrix polymer is almost continuous. Thus, the contribution of fillers to the thermal conductivity of a composite seems to be less than that of the matrix, so that the composites show low thermal conductivities, with the increasing of carbon black loading, many carbon black particles touch each other to begin to form carbon conductive chains, which greatly contribute to the thermal conductivities of composites [15, 16 and 17].

Figure (5) shows the thermal conductivity of SBR versus NR content. It can be seen from this figure that the thermal conductivity of SBR decreased with increasing of (NR) content. The decrease in thermal conductivity of SBR with the addition of NR may be attributed to

1. The blending of the rubbers having different thermal conductivity values (NR is less thermally conductive than SBR) the resulting superposition effect [18].
2. NR has more crosslinked than SBR so that the crosslinked of SBR increase with NR, the increase of crosslinked led to decrease of thermal conductivity this is due to decrease in phonon –phonon mean free path.

Figure (6) shows the values of the thermal diffusivity plotted again carbon black loading level (pphr) for all compounds (NR/SBR) used in this study. From this figure can be seen that the thermal diffusivity of all recipes increased with carbon black increment. This is due to the previous reasons mentioned above in thermal conductivity this is due to thermal diffusivity directly proportional to thermal conductivity. These values agreement with other work [19]. But the values in this work more than the values in work [19], this is due to the authors didn't use in his work carbon black. The values in this work ranged from (0.014 mm^2/sec at 100 NR loaded with 20 pphr carbon black) to(0.029 mm^2/sec) at 100 SBR loaded with 80 pphr carbon black) while as the values in the work [19] ranged between (0.08mm^2/sec) for NR and (0.012 mm^2/sec) for SBR. Figure (6) show thermal diffusivity of SBR verses the addition of NR content. And from this figure can be see that the thermal diffusivity of SBR decreases with NR increment. This is due to the same reasons which mentioned in thermal conductivity test, and these value agreements with other work [19].

Figures (7 and 8), (a, b, c, d and e) show the change in mass percentage plotted versus the immersing time (oil engine and water) for (100 SBR), (75 SBR/25 NR), (50 SBR/50 NR), (25 SBR/ 75 NR) and (100 NR) respectively at different loading level of carbon black.

This test is performed on 40 specimens, half of which are immersed into engine oil and the others are immersed into water because these are the most liquids in contact with tire during service. From these figures it can be noted clearly that the change in mass percentage for rubber composite shows a non-linear increment as the immersing time increase, and these figures shown similar patterns; the percentage of (oil and water) uptake was increased and relatively fast in the initial stage because of its great affinity to uptake oil or water. These figure show swelling in water more than in oil engine for all recipes, this due to water absorption is more than engine oil absorption that is due to the higher viscosity of oil compared with water. Also water swells through porosity of the rubber only, but engine oil swells by dissolving rubber and goes through, so that the change in the mass of SBR reinforced with 80 pphr of carbon black which was immersed in oil is (31.8%) but which was immersed in water is (37.1%). Whereas the change in the mass of NR reinforced with 80 pphr of carbon black which was immersed in oil is (11.3%) but which was immersed in water is (20.4%). Figures (10) and (10) show the swelling ratio verses carbon black loading of all (SBR/NR) recipes, in oil and water respectively. From these figures can be seen that the change in mass percentage for rubber composite decrease with loading level of carbon black increment, The change in swelling of compounds with the increase of carbon black loading could be explained as a consequence of the existing pressure involved between the rubber network and the liquid that act to expand or shrink the rubber network. In practice, the density of crosslinking in rubber compounds increases drastically with increasing carbon black content, resulting a rise of network elasticity contributions. These crosslinks restrict extensibility of the

rubber chains induced by swelling and make it more difficult for liquid to diffusion into the gaps between rubber molecules and decrease the swelling percentage and thus counter any tendency for dissolution. Thus the swelling reduces with the increase of network [20 and 21].

Figures (12) and (13) show the relationship between the change in mass percentage of SBR verses NR loading due to oil engine and water absorption respectively. These figures show that the change in mass of (SBR) rubber decrease with (NR) increment. This is may be due to the degree of the crosslink density increased with NR content [22], so that the change in mass decreased. A maximum value can be achieved with (SBR) at 20 pphr carbon black about (46.2 %) and (56.4 %) for oil engine and water absorption respectively. Compared with (NR) rubber at 20 pphr carbon black about (16.2%) and (25.3 %) for oil engine and water absorption respectively.The relationship between specific gravity and the loading level of carbon black is shown in Figure (14) for (100 SBR), (75 SBR/25 NR), (50 SBR/50 NR), (25 SBR/ 75 NR) and (100 NR) respectively. From this figure can be seen that the specific gravity shows a non-linear increment with the loading level of carbon black, this is because of the voids filling by fillers. These fillers have high specific gravity than rubber, this makes rubber composite denser per unit volume [23]. Figure (14) shows the specific gravity of SBR verses NR content. It can be seen that the specific gravity of SBR is not significantly affected by NR addition. It can be explained by the fact that the specific gravity of NR is close to that of SBR.

Conclusions

The addition of carbon black (N220) to the rubbers (SBR and NR) and their blends and addition NR to SBR leads to an improvement in the physical properties.

The conclusions are:

1. Thermal conductivity increased with the increase of loading level of carbon black for SBR / NR recipes, maximum thermal conductivity was (0.82158) w/m.oC of 100 SBR at 80 pphr carbon black and this property decreased with adding NR to SBR.
2. Thermal diffusivity of rubber composites increases with increase of loading level of carbon black and reaches a maximum value of (0.029 mm^2/sec) for SBR reinforced with 80 pphr carbon black. The thermal diffusivity of NR increase with addition of SBR.
3. The swelling in water more than in oil. Change in mass of the NR/SBR blends increased with immersing time increase and decreased with carbon black increment, and with the increment of NR percentages to SBR. Minimum value achieved was (11.3%) for 100 NR at 80 pphr carbon black.
4. Specific gravity of NR/SBR recipes increases with the addition of reinforcing filler (carbon black).

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Table (1) the compound formulations

Item	Material	Loading level (pphr)				
1	SBR	100	75	50	25	0
2	NR	0	25	50	75	100
3	Zinc oxide (activator)	5				
4	Stearic acid (activator)	2				
5	Antioxidant	1.5				
6	Antiozonant	1.5				
7	Process oil	5				
8	Carbon black (N220)	(20, 40, 60, and 80) pphr				
9	Sulphur	5				
10	Accelerator (TMTD)	2				
12	Reclaim	5				

Table (2) Mixing Step

Item No.	Description
Stage (1) Master Batch	
1	Passing rubbers through rolls several times with decreasing a mill roll opening to 0.5cm,at 70C^{o}.
2	During whole operation, cutting of milled rubber diagonally, rolled or spiraled ,and passed into nip in horizontal and vertical state alternatively several times for homogenization.
3	Adding reclaimed rubber to the rubber in step(1) then banding with mill opening 2.5cm to 0.5cm for several times, and repeat item (2).
4	Adding of Stearic acid and zinc oxide.
5	Adding of antidegredants, and repeat item (2).
6	Adding half of each carbon black and process oil, and repeat item (2).
7	Adding the other half of each carbon black and process oil, and repeat item (2).
Stage (2) Final Batch	
8	Cooling the master batch to the room temperature.
9	Adding the accelerator.
10	Adding the sulfur to the master batch.
11	Sheeting the batch to a thickness of (0.5) cm.
12	Cooling the batch to room temperature rapidly to prevent pre-vulcanization.

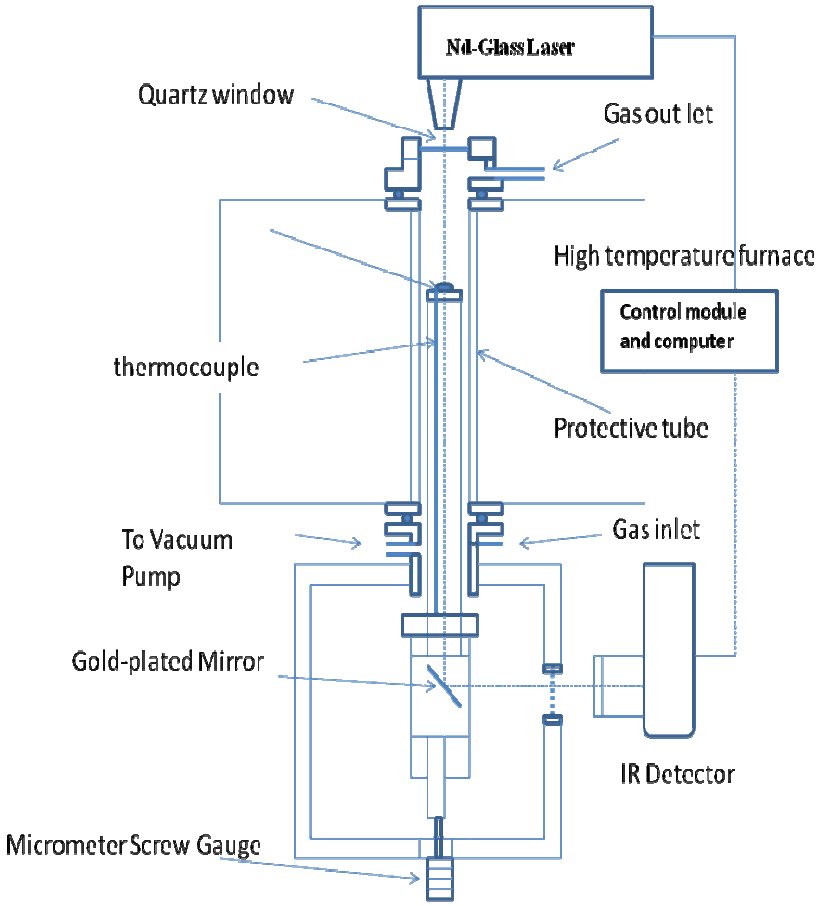


Figure (1): A Schematic Diagram of the Laser Flash Unit.

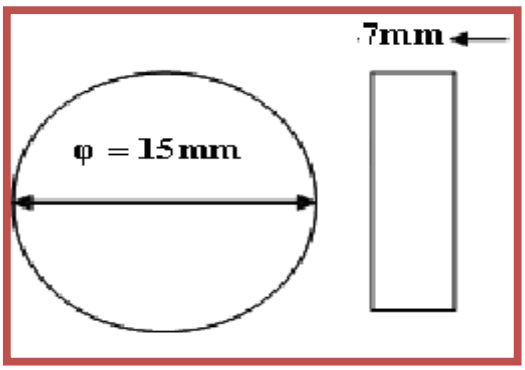


Figure (2) Swelling Test Specimens.

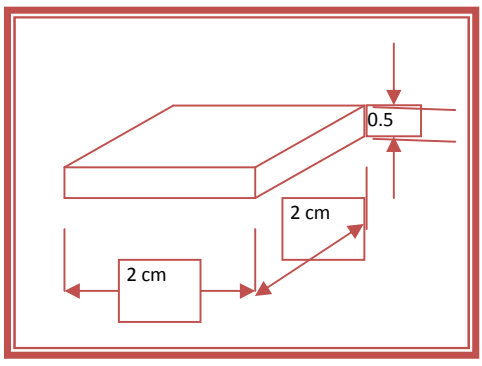


Figure (3) Specific Gravity Specimen.

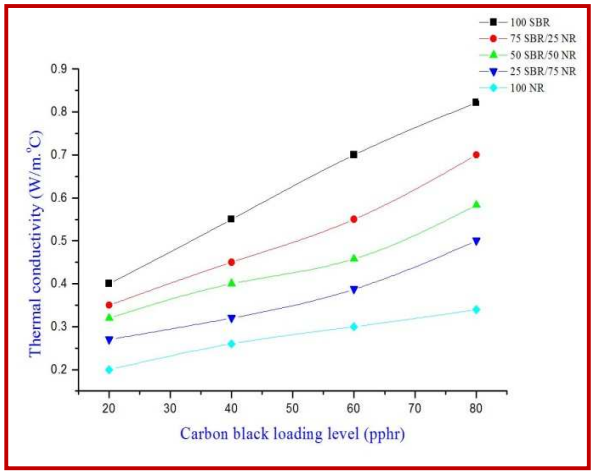


Figure (4) Thermal Conductivity Verses Loading Level of Carbon Black (N220) For SBR/NR

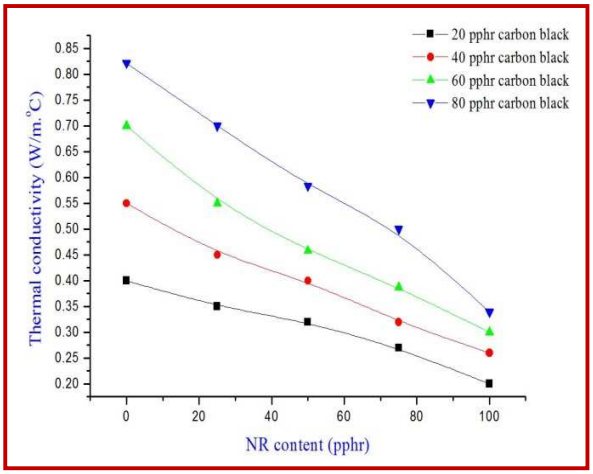


Figure (5) Thermal Conductivity Verses NR Content at Different pphr of Carbon Black.

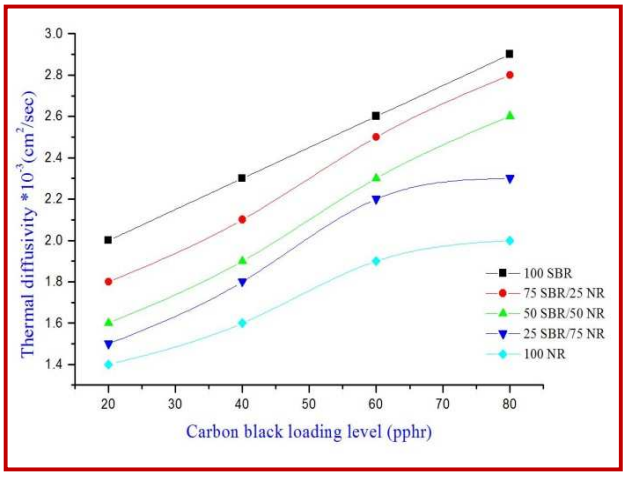


Figure (6) Thermal Diffusivity Verses Loading Level of Carbon Black (N220) For SBR/NR

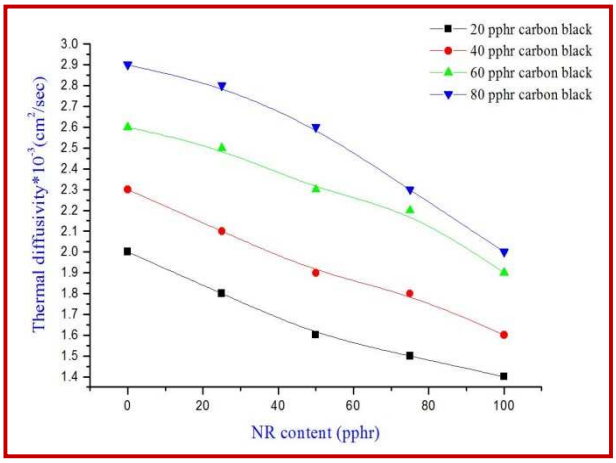


Figure (7) Thermal Diffusivity Verses NR Content at Different pphr Of Carbon Black.

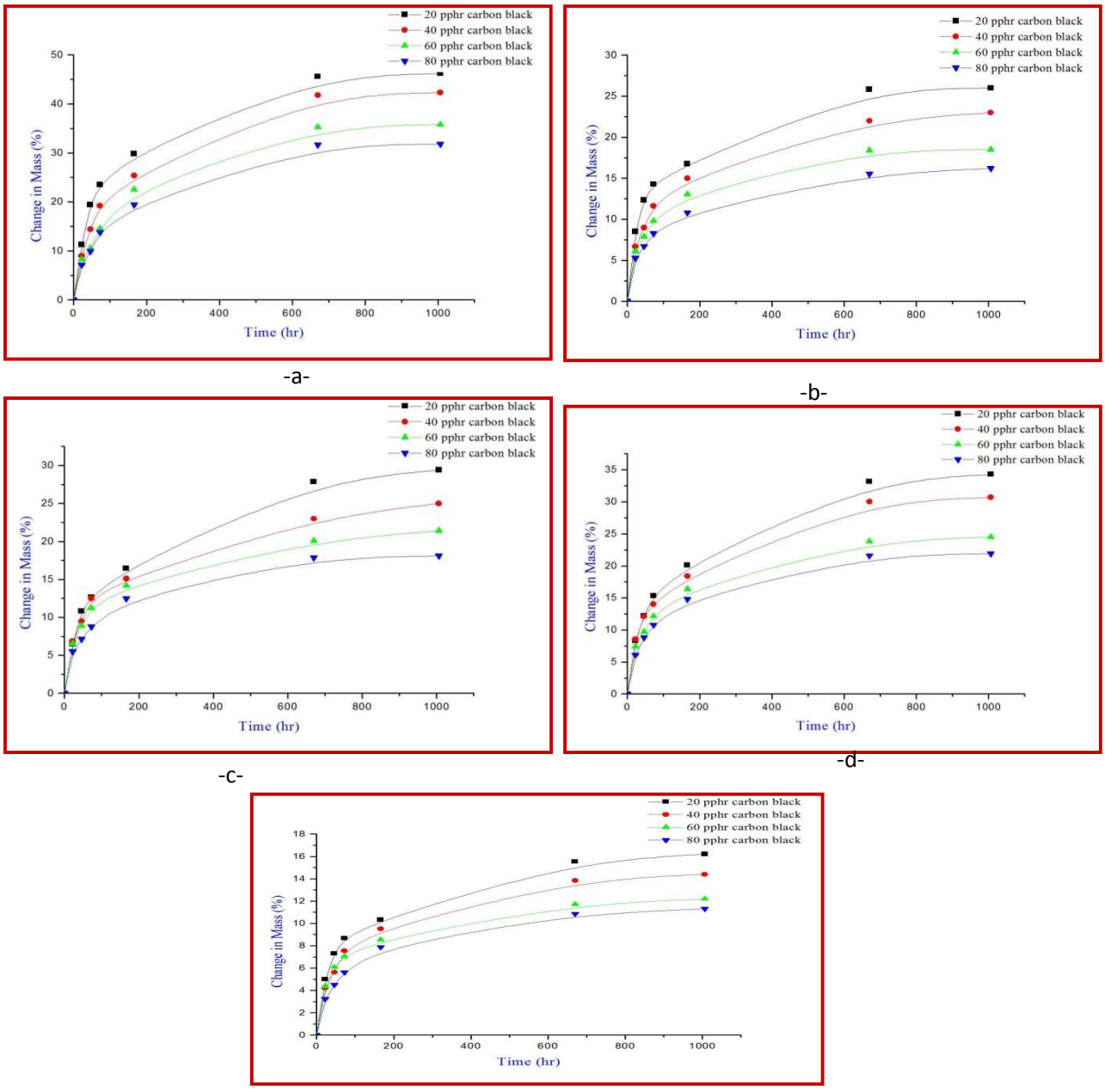
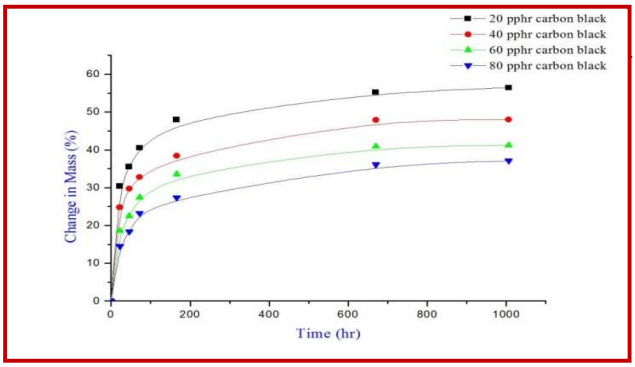
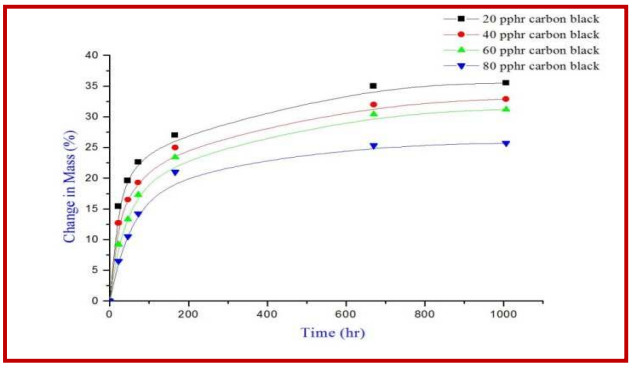


Figure (8) The Effect Of Carbon Black (N220) On Swelling Properties (Oil) Of Tire Tread Recipes (a) 100 SBR (b) 75 SBR/25 NR (c) 50 SBR/50 NR (d) 25 SBR/75 NR (e) 100 NR



(a)



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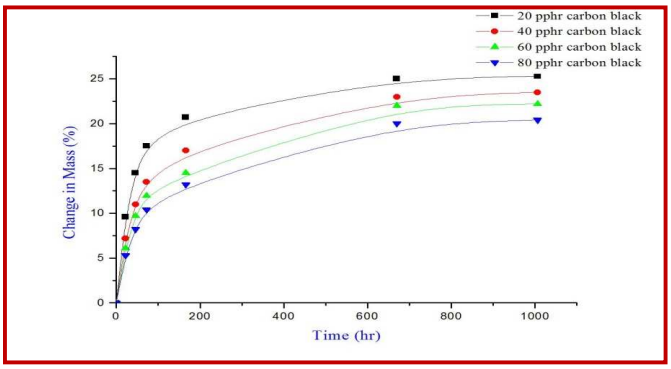
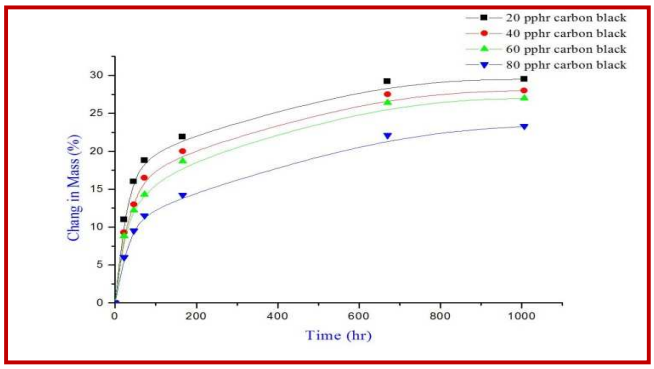
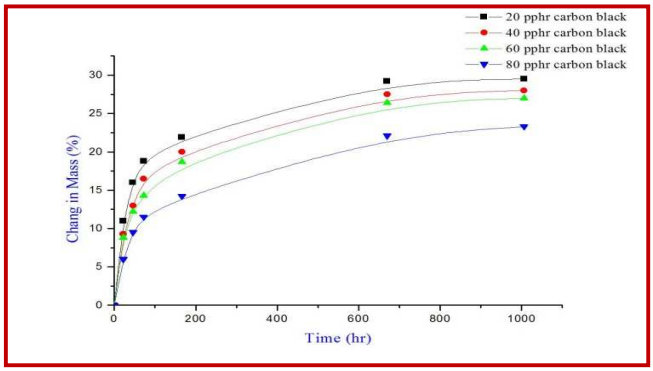


-c-



-d-



-e-

Figure (9) The Effect of Carbon Black (N220) on Swelling Properties (Water) of Tire Tread Recipes (a) 100 SBR (b) 75 SBR/25 NR (c) 50 SBR/50 NR (d) 25 SBR/75 NR (e) 100 NR

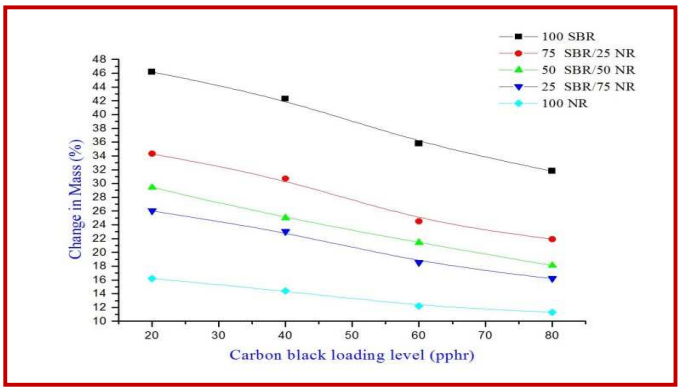


Figure (10) swelling properties (oil) verses loading level of carbon black (N220) for SBR/NR recipes.

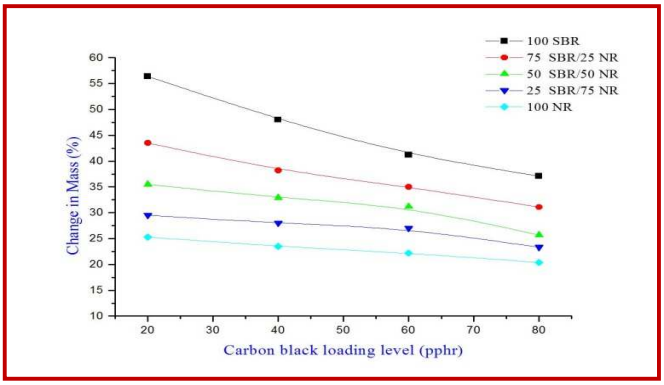


Figure (11) swelling properties (water) verses loading level of carbon black (N220) for SBR/NR

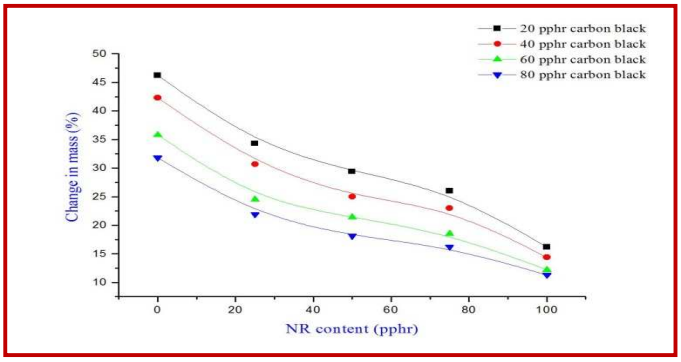


Figure (12) Swelling Properties in (Oil) of SBR Verses Loading Level of NR at Different pphr of Carbon Black.

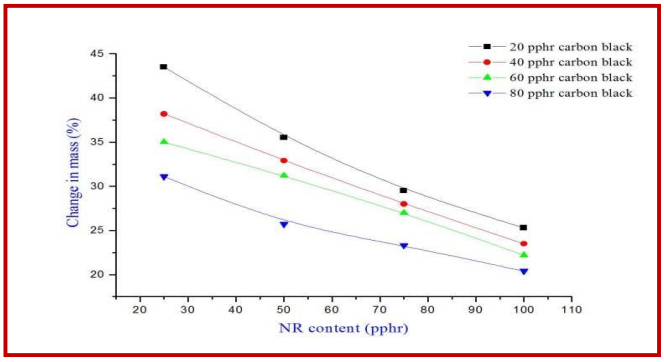


Figure (13) Swelling Properties in Water of SBR Verses Loading Level of NR at Different pphr of Carbon Black.

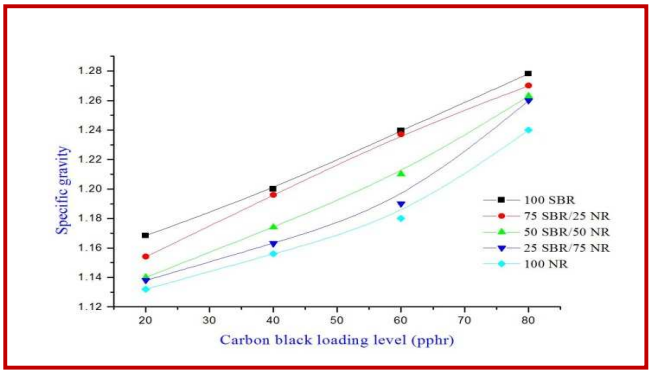


Figure (14) Specific Gravity Verses Loading Level of Carbon Black (N220) For SBR/NR

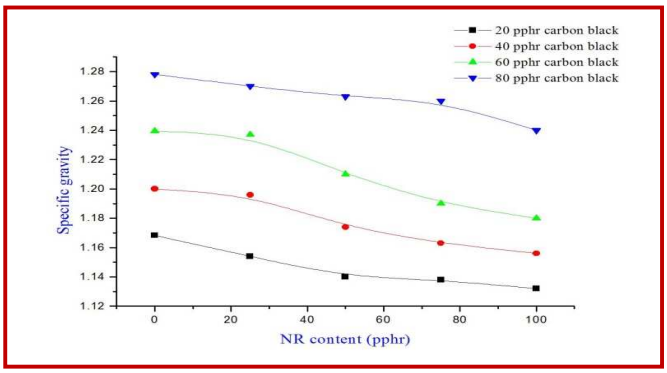


Figure (15) Specific Gravity of SBR Verses Loading Level of NR at Different pphr of Carbon Black.