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Reinforcement the Mechanical Properties of (NR50/SBRs50/OSP) Composites with Oyster Shell Powder and Carbon Black

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Abstract. In this study, the rubber batches composites were prepared using Natural rubber (NR) and synthetic rubber (SBR) as model materials. The composites were reinforced using oyster shell powder (OSP) with particle size less than (225 μ m). Two groups of samples were prepared with (0, 5, 10, 15, 20, 25, 30, 40 and 60 pphr) with loading ratio of OSP and (CB-10pphr) of CB, whereas other composites were without CB. Density, viscosity and multi ultrasound velocity wave devices were applied to characterize the density according to the specification (ASTM-D1817-66) (Densitron), rheological properties with specification (ASTM-D2084-89) standard and mechanical properties, respectively. The result showed significant influence of the reinforcements (OSP) material and (CB) on some of these properties that exhibited an increasing with the increasing of loading ratio of (OSP) and (CB), such as density, relaxation amplitude and compressibility, but the other properties were decreasing like viscosity, relaxation time, velocity waves, bulk modulus and specific acoustic impedance.

Keywords: rubber-based composites, reinforcement, oyster shell, Carbon black, rheological properties, mechanical properties, ultrasonic technique.

1. Introduction

Composites materials have been attracted many researchers and engineering for over many years due to its easy to process with incredible properties in comparison with original components that lead to valuable and interchangeable industrial applications (1). However, it has gained a prestigious position among the different engineering materials. New properties depend on the processes it passes during its preparation as well as on its precise composition (2). Composite materials are defined as the collection of two materials with different mechanical and physical properties, where the purpose of this collection is to develop new properties that are not available in the original materials (1,3).

The composite material contains two phases:

1. Matrix material.
2. Reinforcement material.

Polymer Matrix composite (PMCs) is one of the combined composite materials, where the polymer after basic material contains fillers, such as carbon black the base material can be from thermoplastic, thermosetting, elastomer and these materials (4–6). It is the one of the best filler because of their mechanical properties are high relative to density as well as ease of manufacture (7,8), as the polymeric – based materials is one the most combined types of materials has increased interest in these materials, it has been used in many application starting from the manufacture of aircraft parts and many others by low



density, high strength and resistance that has leads to ideal materials with low cost and low consumption (9–11). As a result reinforced resins have gained wide fame as a modern material in the industry and several alternatives to traditional materials or alloys in many applications because polymers are low density but lack strength and resistance, some other composites are added to the homogeneous polymer and the composition of the copolymers in order to change some of its properties and introduce new attributes (12–15).

2. Experimental

2.1. Materials

NR (50 pphr) of MSR 20 was used that was supplied by the Perlis, Malaysia. SBR (type SBR1502) contains a 23.5% styrene content and butadiene with specific gravity (0.95 g/cm³), and supplied by the Petkim, Turkey, it used to prepared the samples. Also, (50 pphr) of it was used. Carbon black N375 was supplied by Doudah, Iran. It is examined in accordance with the DBP absorption (ASTM D136) and Iodine absorption (ASTM D135). (CB-10 pphr) of N375 were used.

Zinc oxide (97%) and stearic acid (99.4%) were supplied by Durham, the UK. 6PPD N-(1, 3-Dimethyl butyl)-N-Phenyl-Para-Phenylenediamine (98%) was supplied by Flexsys, Belgium. MBS N-oxydiethylenebenzothiazole. 2-sulfonamide (98.2%) was supplied by ITT, India. processing oil. Sulfur was

Oyster shell powder: It is a powder containing 82.4% of the calcium carbonate (CaCO₃) that was carried out at the standardization and quality control center in Baghdad, which has a high mechanical properties, lightweight and ease of manufacture. It is obtained from the banks of the rivers that was well cleaned and grinding before transfer to powder to obtain a particle size equal to or less than (225 μm). It was added to the basic materials with different loading ratio (0, 5, 10, 15, 20, 25, 30, 40 and 60) pphr to obtain (NR50/SPR50/OSP) in group (A) and using the same loading ration of carbon black (CB-10 pphr) another samples were prepared to get overlays on boats (NR50 / SBR50/ OSP/CB) in group (B). That were summarized in Tables (1) and (2).

In this sample , natural rubber (NR) was used as a base material of (50 pphr) with synthetic rubber (SBR) of (50 pphr) and vulcanization materials, including (zinc oxide , citric acid as activator and (MBS) as accelerator and sulfur as vulcanization material and drops from oil as plasticizers material) see table (1), the batch is then supported by oyster shell powder (OSP) and by the proportion of the substrate (OSP) at different loading ration to obtain (NR50/SBR50/OSR)

Table 1. summarized the compounding ingredients of the composites.

<i>Compounding ingredients</i>	<i>PPhr</i>
NR	50
SBR	50
Stearic Acid	2
Zinc Oxide	3
MBS	1
Sulfur	2
6PPD	1
Process oil	3

Table 2. summarized the loading ratio of the components of group (A) composites.

Material	A1	A2	A3	A4	A5	A6	A7	A8	A9
NR	50	50	50	50	50	50	50	50	50
SBR	50	50	50	50	50	50	50	50	50
ZnO	3	3	3	3	3	3	3	3	3
Stearic acid	2	2	2	2	2	2	2	2	2
MBS	1	1	1	1	1	1	1	1	1
6PPD	1	1	1	1	1	1	1	1	1
Carbon black	0	0	0	0	0	0	0	0	0
Process oil	3	3	3	3	3	3	3	3	3
Oyster Shell Powder	0	5	10	15	20	25	30	40	60
Sulfur	2	2	2	2	2	2	2	2	2

Table 3. summarized the loading ratio of the components of group (B) composites.

Material	B1	B2	B3	B4	B5	B6	B7	B8	B9
NR	50	50	50	50	50	50	50	50	50
SBR	50	50	50	50	50	50	50	50	50
ZnO	3	3	3	3	3	3	3	3	3
Stearic acid	2	2	2	2	2	2	2	2	2
MBS	1	1	1	1	1	1	1	1	1
6PPD	1	1	1	1	1	1	1	1	1
Carbon black	10	10	10	10	10	10	10	10	10
Process oil	3	3	3	3	3	3	3	3	3
Oyester Shell Powder	0	5	10	15	20	25	30	40	60
Sulfur	2	2	2	2	2	2	2	2	2

2.2. Ultrasonic measurement

Ultrasonic measurement is made by pulse technique of sender-receiver type (SV-DH-7A/SVX-7 velocity of sound instrument) with frequency (30 KHz), the receiver quartz crystal mounted on a digital vernier scale of slow motion, the receiver crystal could be displaced parallel to the sender and the sample placed between and receiver. The sender and receiver pulses (waves) were displaced as two traces of cathodes ray oscilloscope and the digital delay time (t) of receiver pulses were recorded with respect to the thickness of the samples (x). The pulses height on oscilloscope (CH1) represents incident ultrasonic wave's amplitude (A_0) and pulses height on oscilloscope (CH2) represents the received ultrasonic wave's amplitude (A).

2.3. Composite preparation

The composites were prepared by mixing the materials using the rubber batch and applying (Comerio Ercole Busto Avsizo, Italy), which is consist of (2-Rolls) laboratory. The diameter is 150 mm per roll and the length is 300 mm. Well mixing were carried out according to ASTN1 D15, temperature was controlled at (50 ± 5) °C and the sequence of adding materials to the architecture and the time required of each material and for all types of batches.

2.4. Calculation

The rheological and mechanical properties were studied for the all prepared composites, where the viscosity was calculated using the equation 1 (16).

$$\text{Viscosity} = ML * 2.7 \dots\dots\dots (1)$$

Were ML = Minimum Torque

Specific density (ρ) obtained from equation 6 were carried out by dentition device according to Archimedes principle (10).

$$\text{density} = \frac{\text{weight body in air}}{\text{weight body in air} - \text{weight in liqued}} * \text{density of liqued} \dots\dots (2)$$

Ultrasonic wave's velocity values were obtained from the following equation

$$v = x/t \dots\dots\dots (3)$$

The velocity can be defined to equal the ratio between the distances of ultrasonic waves that travelled through the sample to the delay time.

v = velocity, x = thickness and t = delay time of velocity ultrasonic through the material

The relaxation time (τ) was calculated from equation 4 (17).

$$\tau = 4 \eta s / 3 \rho v^3 \dots\dots\dots (4)$$

ηs = shear viscosity

The relaxation amplitude of ultrasonic wave was calculated from the following equation, where (f) is the frequency and (α) means the absorption coefficient (2).

$$D = \alpha / f^2 \dots\dots\dots (5)$$

The acoustic impedance of a medium (Z) of the material property was calculated by equation 6, where (ρ) means the density (17).

$$Z = \rho v \dots\dots\dots (6)$$

Compressibility (β) is a measure of the relative volume change of a fluid or solid as a response to a pressure (or mean stress) change, it was applied to calculate the β by equation 7 (2):

$$\beta = (\rho v^2)^{-1} \dots\dots\dots (7)$$

The bulk modulus (K) of a substance measures and the substance's resistance to uniform compression that is defined as the pressure increasing to decrease the volume. It was calculated by following equation 8 (17).

$$K = \rho v^2 \dots\dots\dots (8)$$

3. Results and Discussion

Figure (1) shows the effect of oyster shell powder (OSP) on the viscosity of the prepared rubber (NR50/SBR50/OSP) composites that were measured using the equation (1). The results exhibited a decreasing in the viscosity values with the increasing of the loading ratio of (OSP) and the addition of the carbon black (CB-10 pphr). This behavior was attributed the increasing the entanglement between the polymer chains by vulcanization, which reduces the flow of polymer chains over each other. These fillers reduced the intermolecular spaces in these chains and restricted their movement thereby increasing fraction force between the molecules. This made a difficult movement of the polymeric chains which agreed with finding by other researchers (2,17).

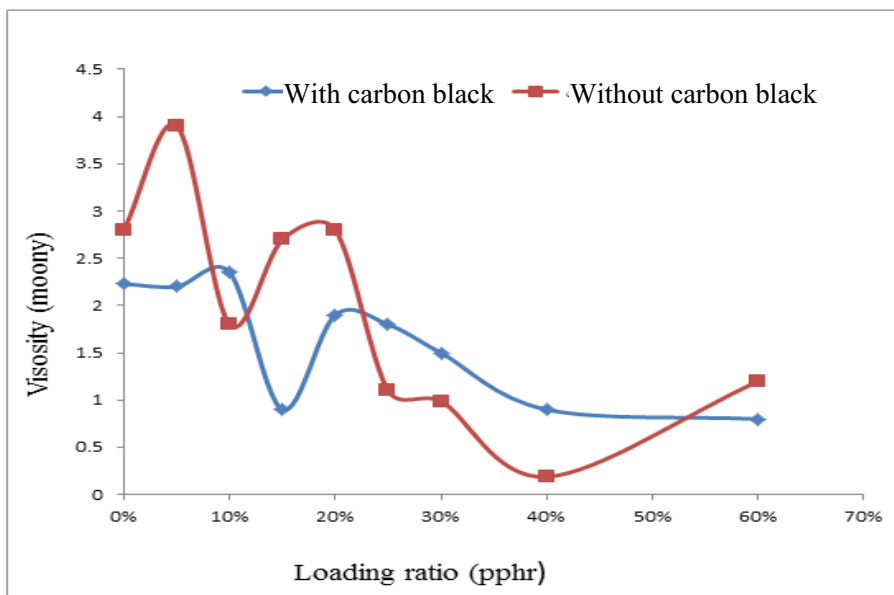


Figure 1. the viscosity of the sample with different loading ratio of A and B composites.

Figure (2) shows density, which was obtained in brackets by (desitorn) and according to archimedes principle of buoyancy and according to equation (2), when applied the oyster shell powder (OSP) in addition to loading the carbon black (CB-10 pphr) that caused increase in the density of the rubber composite (NR50/ SBR50/OSP). This increasing may resulted from the increasing the crosslinking in polymeric chains in addition to attached this fillers to polymer molecules in the base material in agreement with other researcher finding (10,17).

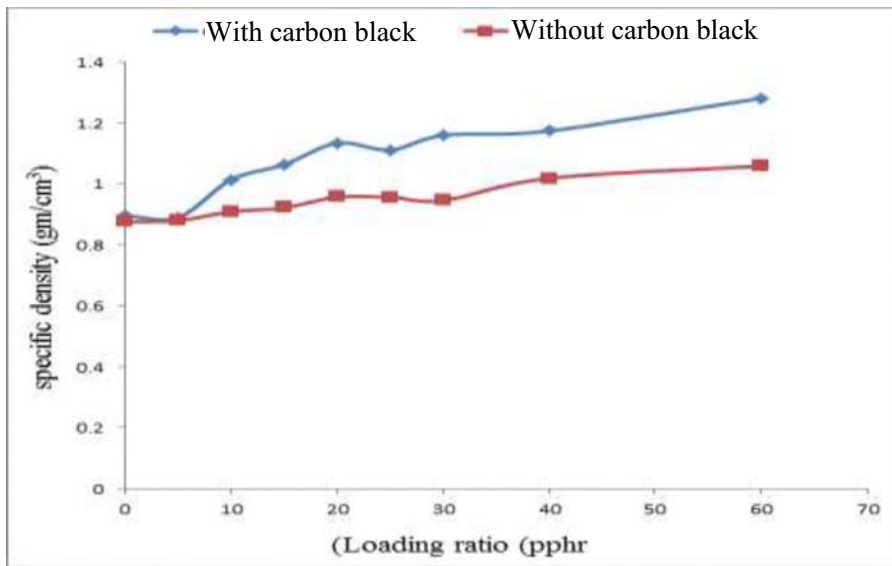


Figure 2. the specific density of the sample with different loading ratio of A and B composites.

Figure (3) shows the velocity of ultrasonic waves was calculated from equation (1), the velocity of oyster shell powder (OSP) without carbon black (CB) and with the addition of carbon black (CB-10 pphr) to the rubber compounds (NR50/SBR50/OSP) for the same loading ratio, as well as when adding carbon black. The velocity of ultrasonic waves, where passing through the composites exhibited a reduction due to the increasing the cross-linking within the material and restrict the movement of the molecules. Moreover, this results was supported by the result of the viscosity, where it is the direct factor influence the wave’s velocity inside the material, the decrease of viscosity led to reduce the areas of dislocation and compression. This increasing the dissipation energy of the ultrasonic within the composite rubber material (2,10,18).

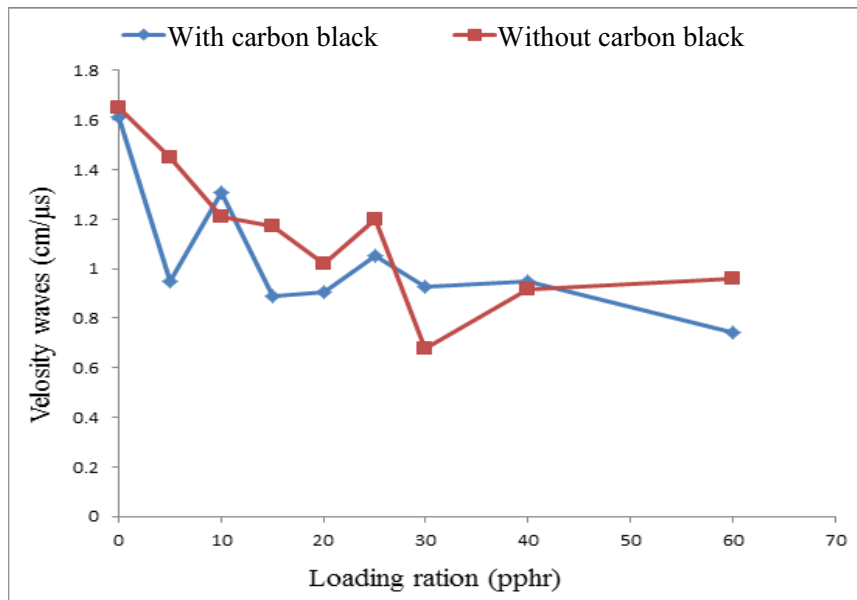


Figure 3. the ultrasound velocity waves with the loading ratio material of composites.

The relaxation time values were calculated for the ultrasonic waves passing through the rubber composite using the equation (2), where noticed from figure (4) that the relaxation time values decreased when the oyster shell powder loading ratio (OSP) increased, as well as when black carbon (CB-10 pphr) was added for rubber compounds (NR50/ SBR50/ OSP) for the same loading ratios. The results exhibited a decreasing in relaxation time that could related to increase in the crosslinking between polymeric chains by vulcanization process, in addition to increase in molecules size which led to difficulty movement and less fluctuate of these molecules around their position. The time of returning these molecules to their original positions decreased in agreement with other researchers found (2,10,17,19).

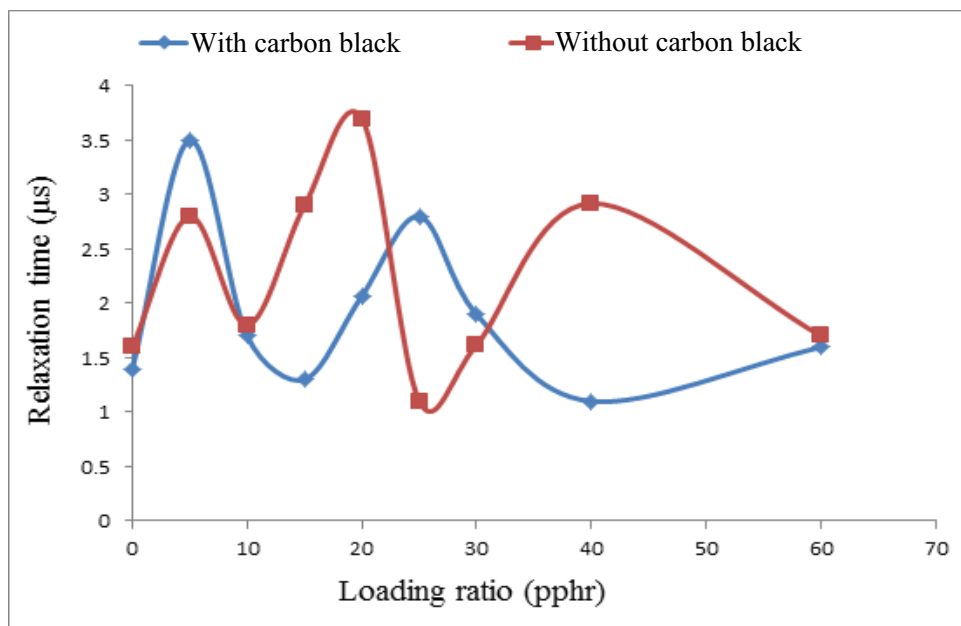


Figure 4. the relaxation time with the loading ratio of rubber composite (NR50/SBR50/OSP) with carbon black and without carbon black (CB-10pphr).

The values of the relaxation amplitude were calculated from equation (3). It was clear shown in Figure (4). That exhibited an increasing in the relaxation amplitude values by adding oyster shell powder (OSP) in addition of carbon black (CB-10 pphr) to the rubber compounds (NR50/SBR50). In the same loading ratio, the relaxation amplitude was directly proportional to the increasing in the absorption coefficient of the frequency constant, it was expected that the relaxation amplitude increased with the increasing in the absorption coefficient in agreement with literature (2,6,10,17).

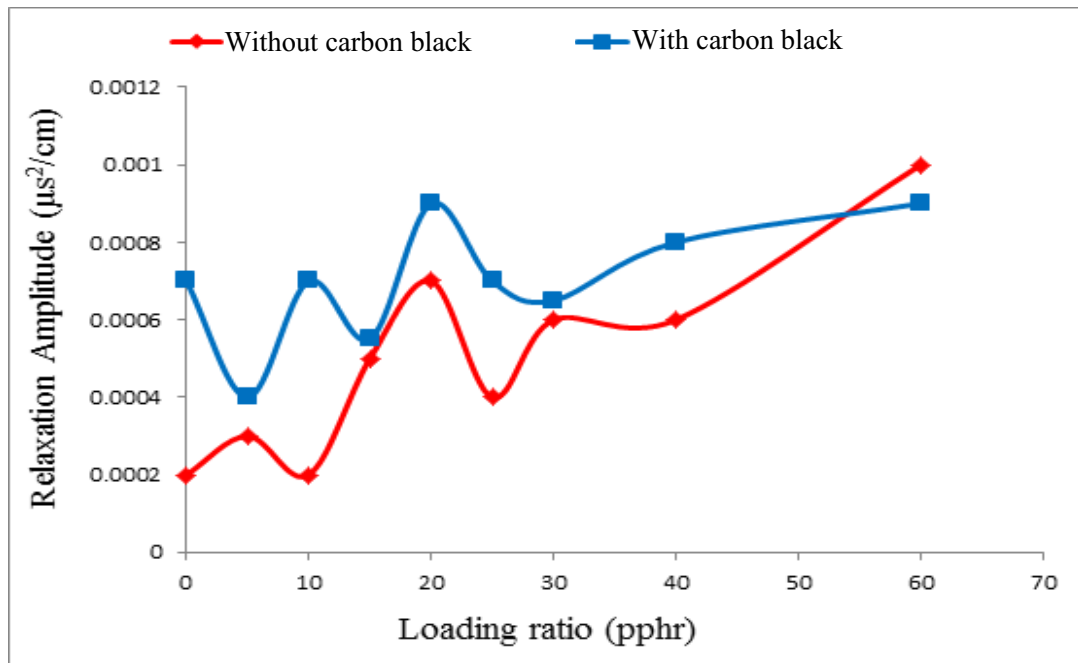


Figure 5. the relaxation amplitude with the loading ratio of rubber composites (NR50/SBR50/OSP) with carbon black and without carbon black (CB-10pphr).

Acoustic impedance values were obtained from equation (4) and Figure (5) that shows the decreasing of the acoustic impedance values with the addition of the oyster shell powder (OSP), as well as the addition of carbon black (CB-10 pphr) for rubber compounds (NR50/SBR50/OSP) at the same loading ratio. The acoustic impedance depends on the speed of ultrasound values, therefore the acoustic impedance values were expected to decrease when the ultrasonic velocity decreased within the rubber compounds (7,10,17).

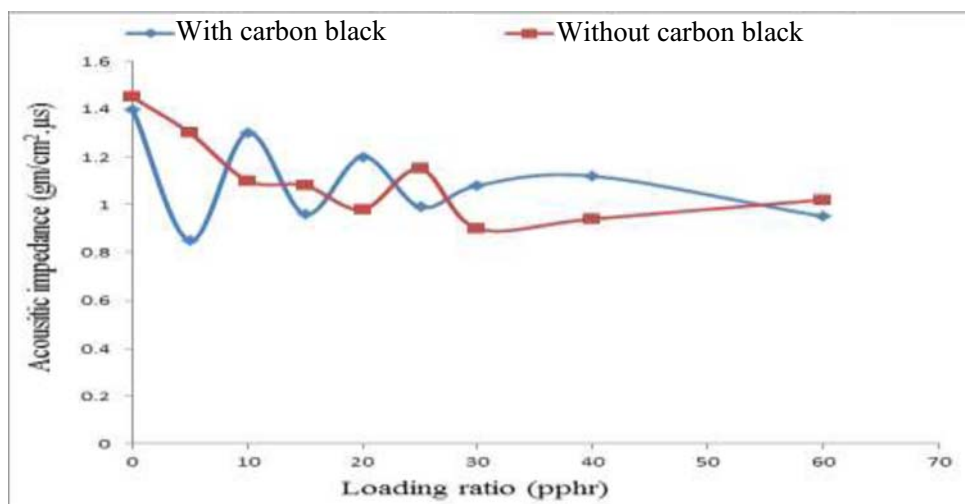


Figure 6. the acoustic impedance with the loading ratio of rubber composites (NR50/SBR50/OSP) with carbon black and without carbon black (CB-10pphr).

The compressibility values were obtained from the relationship (5) since the compressibility is inversely proportional to the square of the ultrasonic velocity, there the decreasing of velocity led to increase the compressibility values. Figure (7) illustrates the increasing of the compressibility of compounds rubber. Also, with the adding of carbon black (CB-10 pphr) to rubber compounds (NR50/SBR50/OSP) at the same loading ratios. This due to the increasing of the density as well as the increasing of the crosslinking between polymer chains by vulcanization process, which match other researcher finding (2,7) .

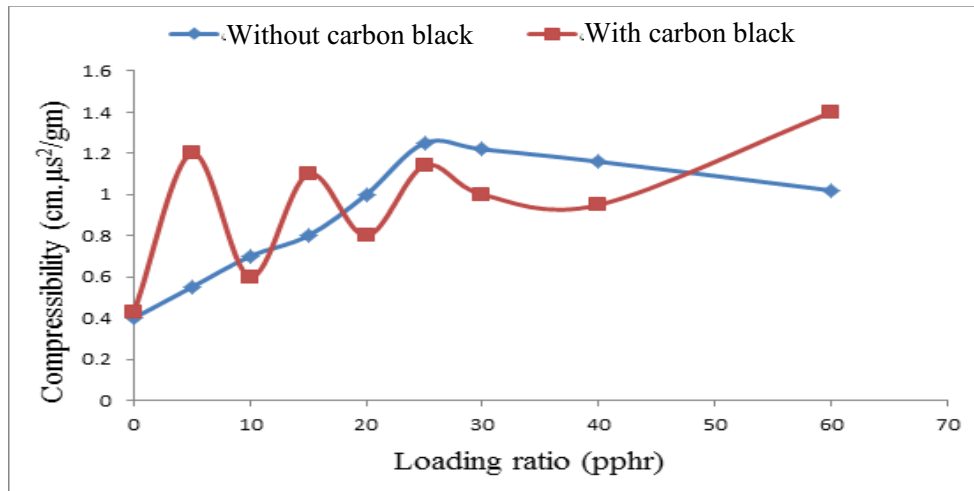


Figure 7. the compressibility with loading ratio of rubber composites (NR50/SBR50/OSP) with carbon black and without carbon black (CB-10pphr).

The bulk modulus values were calculated from equation (6) and Figure (7) that shows a decreasing in bulk modulus of rubber composites by increasing the oyster shell powder loading ratio (OSP) as well as with the addition of carbon black (CB-10 pphr) to rubber compounds (NR50/SBR50/OSP) at the same loading ratios. The bulk modulus was decreased by increasing the loading ratio of the oyster shell powder (OSP) as well as increasing of loading ratio the addition of carbon black (CB-10 pphr), which much what other researcher found (2,10). In addition to the velocity of ultrasonic is the main factor that effect on the bulk modulus, where that was another evidence to explain the bulk modulus behavior which was the similar to the ultrasonic velocity behavior.

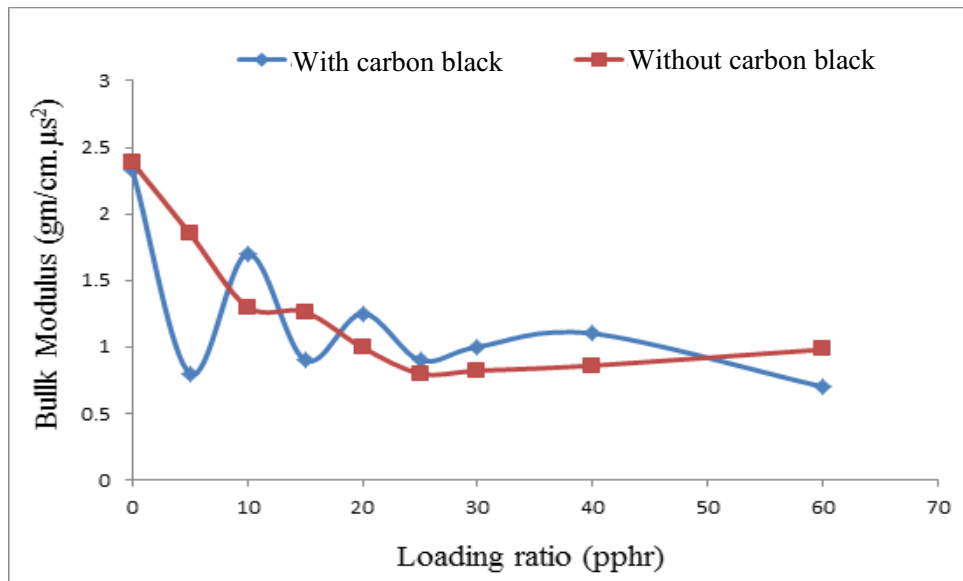


Figure (8) the bulk modulus with the loading ratio of rubber composites (NR50/SBR50/OSP) with carbon black and without carbon black (CB-10pphr).

4. Conclusion

The method was successfully prepared the rubber composites with different loading ratio of OSP and CB-10pphr. The results showed a significant effect of the increasing the loading ratio of (OSP) and the carbon black (CB-10pphr) that caused a reduction in the velocity of ultrasonic, relaxation time, acoustic impedance and bulk modulus. Additionally, these lodgings at the same loading ratio led to essential increasing the relaxation amplitude and compressibility.

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