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Evaluation of thermal insulation and mechanical properties of waste rubber/natural rubber composite

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Abstract The influences of waste rubber loading on mechanical and thermal conductivity properties were investigated for NR composite. An experimental investigation was carried out to obtain low cost construction material with desirable mechanical and thermal insulation properties. Natural rubber was loaded with different concentrations of waste rubber (200, 400, 600, 800, and 1000) phr. The addition of waste rubber leads to a slight increase in thermal conductivity values of composites but it still lies around range of thermal insulating materials. Also addition of waste rubber leads to improvement of mechanical properties of composites. The crosslink density of NR composite increases with the increase of waste rubber loading until 600 phr and after that it decreases due to the stronger the rubber–filler interaction. This leads to the decrease of the swelling index that has the opposite trend of crosslink density. So, the sample with 600 phr waste rubber is considered the optimum concentration from the swelling measurement. Filler loading results in pronounced increase in the tensile modulus and decease in the elongation at fracture which reflects the reinforcement effect of the filler. The yield stress increases with waste rubber loading increment. This delays the permanent disruption of matrix morphology. So, the optimum concentration which is 600 phr waste rubber loading agrees with the swelling and mechanical measurements which has desirable thermal insulation and high mechanical properties and decreases the cost of materials to 82% of the NR cost.

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Introduction

The amount of waste rubber increases every year. Therefore, it contributes to environmental pollution because it cannot be easily decomposed, the exact time needed for its biodegradation being unknown. So, the disposal of used rubber tires from vehicles creates a significant environmental problem, especially

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when discarded indiscriminately, as ecological damage and risks to public health [1–3].

Efforts on a large scale have been made by the polymer industry to develop cost effective techniques to convert waste and used rubber into processable forms. Reutilization of waste rubber materials as building materials appears to be a viable solution not only to such pollution problem but also to the problem of economical design of buildings. The increase in the popularity of using environmentally friendly, low cost and lightweight construction materials in building industry brings the need for searching more innovative, flexible and versatile composites. This has set increased demands on both thermal and mechanical performances of new building products integrated with various waste and used rubbers.

Thermal Insulation in buildings is an important factor to achieving thermal comfort for its occupants. Insulation reduces unwanted heat loss or gain and can decrease the energy demands of heating and cooling systems. It does not necessarily deal with issues of adequate ventilation and may or may not affect the level of sound insulation [4].

This paper focuses on the experimental studies related to the development of low-cost insolation material by reusing waste rubber as potential filler in natural rubber (NR). In this series of experiments, the effect of rubber waste concentration on different physical, mechanical, physicochemical properties were studied.

Experimental work

Test samples

All materials used in this research come from Alexandria factory for tire manufacture, Egypt. The structure of these materials is as follows:

- Natural rubber (NR), with specific gravity 0.934.
- Zinc oxide (ZnO) as activators with specific gravity at 15 °C of 5.55–5.61.
- Stearic acid: melting point 67–69 °C; specific gravity 0.838.
- Tetramethyl thiuran disulfide (TMTD) as accelerator with specific gravity 1.29–1.31, melting point 1485 °C and order less powder.
- Antioxidant *N*-isopropyl *N*0-cyclohexyl paraphenylene diamine (IPPD): purple gray flakes have density 1.17 g/cm^3.
- Elemental sulfur (*S*) with fine pale yellow powder and specific.
- Gravity = 2.04–2.06.
- Naphthenic oil, with specific gravity 0.94–0.96 at 15 °C, viscosity 80–90 poise at 100 °C.
- Waste rubber were obtained from automobile tire pieces which can easily be obtained from the environment with almost no cost, are shredded and added into natural rubber from.

Techniques of characterization

Rubber mixing and preparing

The samples used to perform experimental tests used in this work were prepared according to [ASTM C 11361;99] with the recipes presented in Table 1 and used large amount of processed oil, 10 phr to assimilate the high waste rubber content and have optimum dispersion and coupling with rubber. The vulcanization of the rubber composites was carried out in a hydraulic press (Mackey Bowley,) under a pressure of about 150 bar, temperature 150 °C for a dwell time of 45 min.

Table 1 Formulation of the recipes.

Ingridents (phr[a])	Samples					
	W_1	W_2	W_3	W_4	W_5	W_6
NR	100	100	100	100	100	100
Stearic acid	2	2	2	2	2	2
Zinc oxide	5	5	5	5	5	5
Waste rubber	0	200	400	600	800	1000
Processing oil	10	10	10	10	10	10
TMTD[b]	2	2	2	2	2	2
IPPD(4020)[c]	1	1	1	1	1	1
Sulfur	2.5	2.5	2.5	2.5	2.5	2.5

[a] Part per hundred parts of rubber.
[b] Tetramethyl thiuram disulfide.
[c] *N*-isopropyl-*N*0-phenyl-*p*-phenylene diamine.

Thermal conductivity measurements

KD2–Pro portable thermal conductivity meter was used to measure the thermal properties of the polymeric composites according to transient method techniques as thermal conductivity, specific heat and thermal diffusivity, This test is done according to the stander [ASTM D 5334; 08] on cubic samples of side length 5 cm at different temperature as each sample was cured for 24 h at each temperature and then measured.

Specific gravity

This test is carried out according to [ASTM D792-08], samples prepared with dimensions $2 \times 2 \times 0.2$ cm. In this test the specimens are weighted in air and then weighed while suspended in water. The obtained values are applied in the following equation [5].

$$Sp.Gr.ofmaterial = \frac{W_a}{W_a - W_s} \times sp.Gr.ofwater \quad (1)$$

W_a is weight of sample in air, and W_s is weight of sample suspended in water.

Swelling measurements

Diffusion measurements

The test sample in the form of a Square of side length 0.1 cm and 0.2 cm thickness was cut from the rubber sheets. Each sample was weighted to an accuracy of 0.1 mg (using digital balance) and then soaked in benzene at room temperature. The sample was removed from the solvent after different specific time. The excess solvent on the surface of the test sample was removed by blotting liquid with filter paper then the sample was weighed again. The weightings were continued till equilibrium swelling was attained.

Crosslink density measurement

Swelling measurements were determined according to ASTM D471-10. Each specimen was weighed in a weighing bottle, which was covered with benzene for 24 h so that the state of equilibrium swelling could be reached. The swollen samples were weighed and then dried in an oven to a constant weight. The last weight was taken as the correct weight of the sample free from dissolved matter.

The diffusion mechanism in rubbers is essentially connected with the ability of the polymer to provide pathways for the solvent to progress in the form of randomly generated voids. Diffusion coefficient (D) of the solvents was calculated using the simplified expression given below [6].

$$\frac{M_t}{M_e} = \frac{4}{d}\left(\frac{Dt}{\pi}\right)^{\frac{1}{2}} \tag{2}$$

where M_t and M_e are the weight fraction of the solvent absorbed at time t and at equilibrium swelling respectively and d is the initial sample thickness. Thus, D can be calculated from the initial slope of the linear portion of a sorption curve obtained by plotting M_t/M_e versus $t^{1/2}$.

Volume swelling in percentage (swelling index) was calculated using the following equation [7].

$$q - 1 = \frac{[(\frac{w_2}{w_1}) - 1]\rho_c}{\rho_s} \tag{3}$$

where q is ratio of swollen volume to original unswollen volume of samples, $q - 1$ is swelling index, w_1 is the weight of dry sample, w_2 is the weight of sample after swelling for 24 h and ρ_c and ρ_s are the densities of the specimen and the test solvent respectively.

Evidently, for a given solvent, the higher the crosslink density of the rubber the lower the swelling, and conversely, for a given degree of crosslink density, a more powerful solvent will give a higher degree of swelling. Then the swelling ratio is a direct measurement of the degree of crosslinking. The formation of such cross-linking is achieved by determining its density from equilibrium swelling measurements through the average molecular weight of the polymer between cross-links (M_c) according to Flory–Rehner relation [8–10].

$$M_c = \frac{-\rho_p V_s V_r^{\frac{1}{3}}}{\ln(1 - V_r) + V_r + \aleph V_r^2} \tag{4}$$

where ρ_p is the density of polymer ρ_p (NR) = 0.913 g/cm^3, V_s is the molar volume of the solvent (benzene) = 89 cm^3/mol, χ is the interaction parameter of (NR) = 0.393, V_r is the volume fraction of swollen rubber and can be obtained from the masses and densities of rubber sample and the solvent [11].

$$V_r = \frac{1}{1 + Q} \tag{5}$$

where Q is defined as grams of solvent per gram of rubber hydrocarbon which is calculated by [12]:

$$Q = \frac{M_s - M_d}{M_d} \tag{6}$$

where M_s is the swollen weight and M_d is the dried weight. The crosslink density v_e is defined for a perfect network as the number of elastically active network chains per unit volume and is given by [13]:

$$v_e = \rho_p N_A / M_c \tag{7}$$

where N_A is the Avogadro number.

Mechanical properties measurements

Dumbbell shape samples prepared with dimensions 5 cm working length, 2 mm thickness and 7 mm width were used for stress–strain measurements.

Hardness measurements

The hardness was determined using Shore A durometer according to [ASTM D2240-85].

Results and discussion

Thermal conductivity

The effect of rubber waste addition to the thermal conductivity and density of the NR composites at room temperature are shown in Fig. 1. Initially, the increment of conductivity exhibits a linear portion with the loading level of waste rubber followed by a plateau region at higher concentration. Waste rubber increases the thermal conductivity of rubber composite due to the thermal conductivity of waste rubber greater than the thermal conductivity of NR. Because of rubber waste contains filler such as carbon black or Silica according to waste type so with the increasing of waste rubber loading, many filler particles touch each other to begin to form filler conductive chains, which greatly contribute to the thermal conductivities of composites. The nearly higher thermally conductive sample was at 600 phr content equal 0.25(W/m K) which is comparably less than the thermal conductivity of clay bricks [9], the wood concrete [14]and concrete containing waste rubber [15] .The values of conductivities of the samples were nearly in the range of insulating materials. The density increases also with the same trend as shown in Fig. 1. This is agreeing well with the direct proportionality between the thermal conductivity and the density in which the density values are in range of

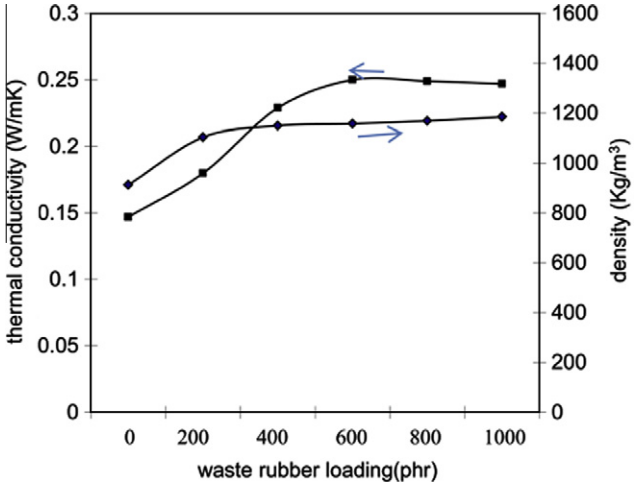

Fig. 1 Dependence of the thermal conductivity and density on waste rubber content of NR vulcanizate.

light weight materials. Figure 2 depicts the variation of thermal conductivity with the temperature change at different waste concentration. This figure investigates that the thermal conductivity is independent of the temperature change, i.e., the thermal insulation property of the samples isn't affected by the variation of temperature, (this will be a good application for the climate variation).

Figure 3 shows the plot of M_t/M_e against $t^{1/2}$(square root of the time) for pure NR and NR loaded with waste rubber. In this figure, all the sorption processes are similar in nature, and have sigmoidal-shaped profiles. Figure 3 clearly manifests that the NR vulcanisate exhibits the highest solvent uptake. Among the filled samples, NR loaded with 600 and 800 phr absorb the lowest amount of solvent. So the solvent uptake decreases because the void formation decreases with waste rubber addition. Influence of changing the waste content on the diffusion coefficient of benzene in the rubber matrix was represented in the Fig. 4. The diffusion coefficient decrease with waste rubber loading increase and has lowest value at 600 phr waste rubber content.

Figure 5 shows the swelling index and crosslink density values at different waste rubber loadings for all the proposed rubber specimens used in this study. This Figure shows that the swelling index decreases while the degree of crosslink density increases as the waste rubber content increases, it is clear that the lowest value of swelling index and the highest value of crosslink density at 600 phr waste rubber sample, which is considered as the optimum concentration obtained from swelling measurement.

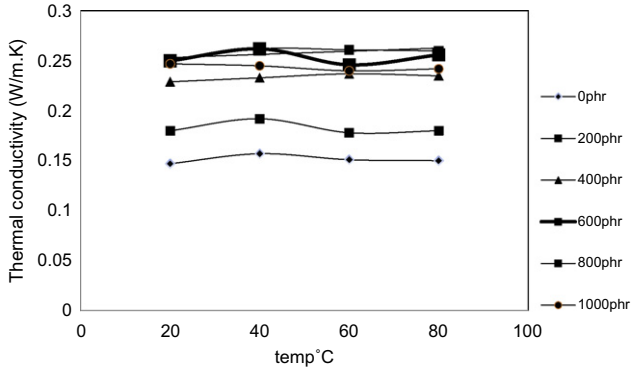

Fig. 2 Dependence of the thermal conductivity of waste rubber content of NR vulcanizate on temperature.

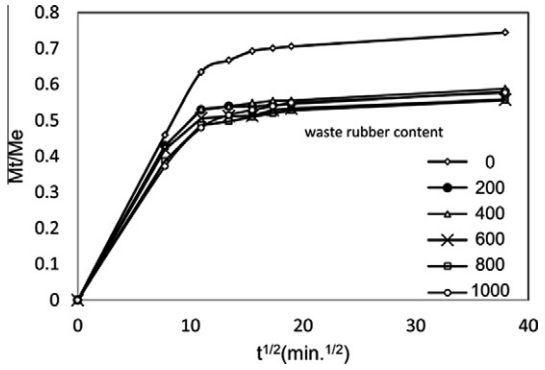

Fig. 3 Variation of M_t/M_e with $t^{1/2}$ for pure NR and NR with waste rubber.

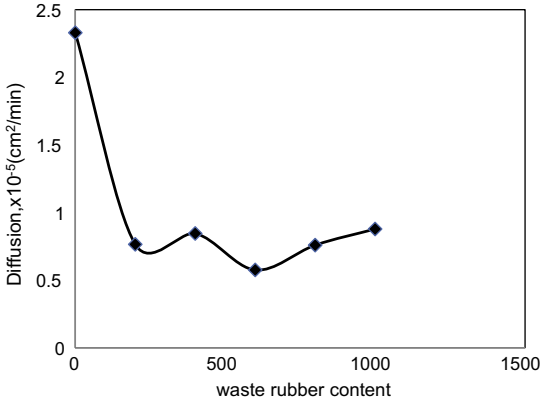

Fig. 4 Variation of diffusion coefficient with concentration of waste rubber concentration.

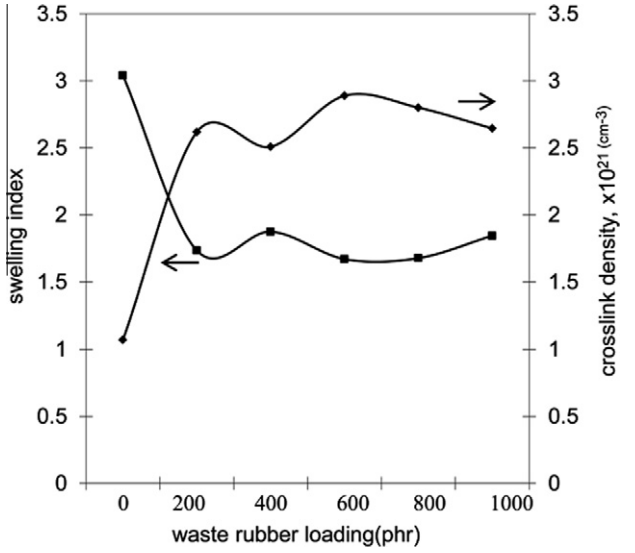

Fig. 5 Variation of the swelling index, crosslink density with waste rubber content.

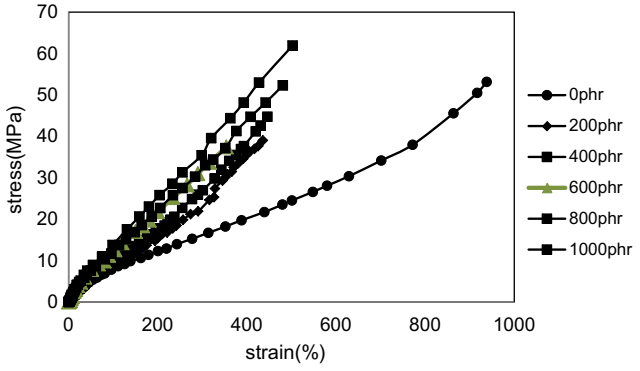

Fig. 6 Stress–strain relation of different concentrations.

Mechanical measurement (tensile properties)

The stress–strain relationship for waste rubber/NR is shown in Fig. 6. From this Figure we can obtain mechanical parameters such as tensile modulus, yield strength, elongation at break. tensile modulus was determined from the linear portion of the stress–strain curve. From this Figure it is clear that tensile strength of Natural rubber has the maximum value. Natural rubber inherently possesses high strength due to strain-induced crystallization. When waste rubber is incorporated into NR, the regular arrangement of rubber molecules is disrupted and hence the ability for crystallization is lost. This is the reason why waste rubber reinforced natural rubber composites possess lower tensile strength than gum compounds. But tensile strength increases with the waste rubber increment due to increasing crosslink density.

Figure 7 represents the variation of tensile modulus and elongation at fracture versus the waste rubber loading. From this Figure it can be seen that, as the waste rubber loading increases, the tensile modulus increases until 600 phr after that it decreases. This decrease of tensile modulus is due to aggregate of filler at high concentration. The tensile modulus has the same trend as crosslink density that measured from swelling

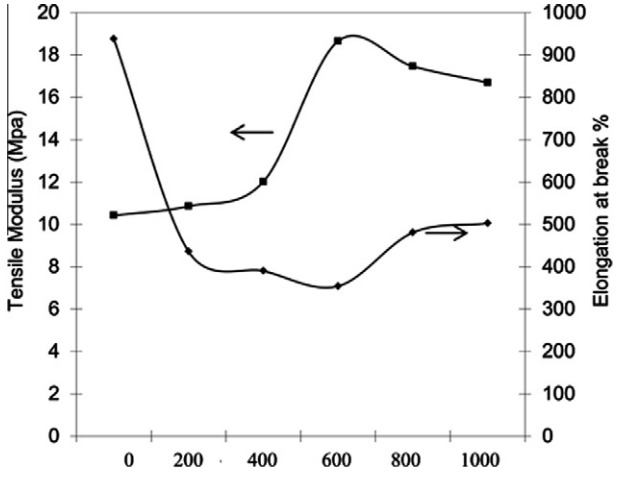

Fig. 7 Variation of the tensile modulus and the elongation at fracture, with waste rubber content.

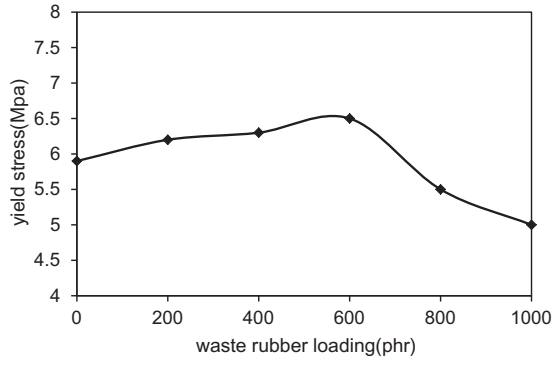

Fig. 8 Variation of yield stress with waste rubber.

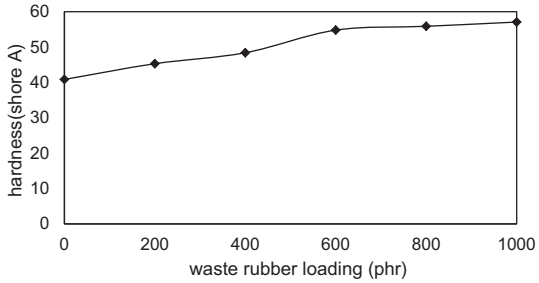

Fig. 9 Variation of hardness with waste rubber content.

measurement. However, strong matrix–filler interaction due to the increase of the number of crosslinks between polymer chains causes a loss of flexibility of the polymer composite so the value of elongation at break shows a reduction with increasing waste rubber loading and gives the opposite trend of tensile modulus resulted in composites becoming stiffer and harder. The increasing of waste rubber increases the yield stress as shown in Fig. 8. This may be due to the susceptibility of waste rubber to deformation. Thus the waste rubber is deformed along the matrix upon application of stress. These delays the permanent disruption of matrix morphology. Figure 9 presents the variation of hardness with waste rubber loading. As waste rubber content increases, hardness is seen to progressively increase because of the stiffness imparted by the waste rubber. From pervious measurement, increased waste loading in the rubber matrix resulted in composites becoming stiffer and harder. This will reduce composite's resilience and flexibility and lead to higher tensile modulus and lower elongation at break. The highest value of modulus and minimum value of elongation at break is at 600 phr which is considered optimum concentration in the mechanical measurement which agreed well with the previously shown by the swelling measurements.

Conclusion

It may be concluded from previous measurements of this study that:

1. Increasing of the crosslink density of NR composite due to addition of waste rubber makes chains unable to move relative to each other as easily. So the tensile increases and elongation at break decreases.
2. Filler loading results in pronounced increase in the tensile modulus which reflects the reinforcement effect of the filler and agree with swelling measurement.
3. The optimum concentration. which is 600 phr waste rubber loading has desirable thermal insulation and high mechanical properties and decreases the cost of building materials to 82% of the NR cost.

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