



Investigation of an Insulating Waste Conscious Material For Sustainable Building Application

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Abstract

One of the risks that threaten future generations is depletion of resources. Therefore, the exploitation of waste is one of the most successful ways to preserve resources and produce multi-use building materials. Calcium carbonate (CaCO₃)/blend rubber (NR/SBR) and (CaCO₃)/foamed rubber blend were prepared by mastication -vulcanization technique. Specific gravity for all samples were determined while ranged between (0.42-1.4) for (CaCO₃)/foamed rubber blend and between (1.003 – 2.9) for unfoamed rubber blend. Scanning electron microscope (SEM) for samples 150 phr of foamed and unfoamed rubber was done to compare the types of matrices. Thermal conductivity of the two groups were measured. The samples with concentrations till 150 phr CaCO₃ foamed rubber lies in the range of thermal insulating materials while the samples of unfoamed rubber have higher thermal conductivity values. Water absorption test was conducted. The water absorption coefficient values of the foamed rubber are lower than that of unfoamed rubber. Mechanical measurements and swelling properties were investigated for all samples. The sample with 150 phr foamed rubber blend considered the optimum sample that achieve all desired thermal and moisture insulating properties.

Keywords: Foaming agent; rubber blend; thermal insulating; water proofing; thermal comfort

Introduction

Green technology is also called “clean technology” is an important technology that deals with any tactic conserves the earth and uses its resources. It is beneficial to curb the bad human impacts, so using ecofriendly products one of the most important aims of green technology applications. Feasible management of waste is significant for sustainable building and livable cities, but remains confrontation for many developing countries and cities. Addition of frugal and abundant waste as a substituent to carbon black loaded rubber produces an economic product used for many construction applications. In construction industry calcium carbonate can play an important role as cement ingredients (bonding bricks, stones, concrete, blocks, shingles and rubber compounds), roads constructions, and masonry materials or as starting builders lime preparation materials. Reusing waste calcium carbonate in construction industry cuts both ways, as the environment gets rid of waste while producing

multifunctional construction product [1]. Water absorption test, water tightness, thermal stability, UV weathering, hardness and mechanical measurements were investigated for natural rubber loaded with calcium hydroxide (CaOH), calcium carbonate (CaCO₃), waste rubber and fly ash (class F) as industrial by products. The tests results showed that rubber loaded with waste rubber, fly ash and calcium carbonate are suitable for marketing as a waterproofing membranes [2]. An economical construction material may be obtained by natural rubber (NR) clay composite with desirable thermal, mechanical and waterproofing properties. With the increase of clay content tensile strength increased, while swelling and swelling index decrease. The thermal conductivity of the sample loaded 60 phr lies in thermal insulation range for construction materials while reasonable values of water absorption coefficient were obtained for all samples [3]. Cement kiln dust (CKD) was loaded NR/SBR blend. The thermal conductivity of the blend was increased with

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the increase of CKD while swelling parameters decreased. Mechanical compression set, hardness and tensile strength slightly improved with CKD increase [4]. Waste rubber loaded natural rubber to obtain frugal thermal insulating material for masonry applications. Thermal and mechanical properties were studied and proved that the waste rubber/natural rubber composite is worthy composite for thermal insulating building applications [5].

This paper aims to produce frugal waste conscious thermal and moisture insulating material used for sustainable building applications.

1. Experimental work

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

Natural Rubber (NR), with specific gravity 0.934 g/cm³

Zinc oxide (ZnO) as activators with specific gravity at 15 oC is 5.55–5.61 g/cm³

Stearic acid: melting point 67–69 oC; specific gravity 0.838 g/cm³

Tetramethyl Thiuram disulfide (TMTD) as accelerator with specific gravity 1.29–1.31 g/cm³, melting point 1485 oC and order less powder Antioxidant N-isopropyl N'-phenyl-1,4 phenylenediamine (IPPD): purple gray flakes have density 1.17 g/cm³.

Elemental sulfur (S) with fine pale yellow powder and specific gravity 2.04–2.06 g/cm³.

Naphthenic oil, with specific gravity 0.94–0.96 g/cm³ at 15 oC, viscosity 80–90 poise at 100 oC and deep green viscous oil.

Foaming agent (Azodicarbonamide) is a synthetic chemical with the molecular formula C₂H₄O₂N₄. It is a yellow to orange red, odorless, crystalline powder. Used to reduce thermal conductivity and density of rubber to obtain low cost thermal insulating material.

Waste CaCO₃ were collected from a factory by-product.

The samples used to perform experimental tests were prepared with the recipe presented in Tables 1 and 2, The vulcanization of the rubber composites was carried out in a hydraulic press under a pressure of about 150 bar, temperature 150 o C and time of 45min [2].

Table (1): Shows the composition of prepared samples.

Ingredients (phr [*])	Samples					
	50	50	50	50	50	50
NR	50	50	50	50	50	50
SBR	50	50	50	50	50	50
Stearic acid	2	2	2	2	2	2
Zinc oxide	5	5	5	5	5	5
Processing Oil	10	10	10	10	10	10
TMTD	2	2	2	2	2	2
IPPD(4020)	1	1	1	1	1	1
Sulphur	2.5	2.5	2.5	2.5	2.5	2.5
CaCO ₃	0	100	200	300	400	500

* Part per hundred parts of rubber.

Table (2): Shows the composition of prepared samples

Ingredients (phr [*])	Samples			
	50	50	50	50
NR	50	50	50	50
SBR	50	50	50	50
Stearic acid	2	2	2	2
Zinc oxide	5	5	5	5
Foaming agent	8	8	8	8
Processing Oil	10	10	10	10
TMTD	2	2	2	2
IPPD(4020)	1	1	1	1
Sulphur	2.5	2.5	2.5	2.5
CaCO ₃	50	100	150	200

Thermal Conductivity test conducted using KD2 – Pro portable thermal conductivity meter according to transient method techniques. This test is done on cubic samples of side length 5 cm at different temperatures

according to the standard. Specific gravity test was conducted according to [ASTM D792] [6]. Samples were cut with dimensions (100 x 100x2) mm, soaked in water bath for 24h while weighed before and after

soaking. Dry weight and suspended in water weight were recorded. Water absorption test was conducted according to [ASTM D570] [7, 8]. Water tightness test was conducted according to [EN: 1928] [9]. Mechanical measurements (Tensile strength, young's modulus and elongation at break) were measured in tensile testing machine (SHIMADZU AG-X) according to [ASTM D-412-06] [10]. The hardness was determined using Shore A durometer in accordance with [ASTM D 2240-07] [11]. Sample was weighed by a digital balance with sensitivity 0.1 mg and then inserted in a test tube containing solvent. The blends were removed from the solvent and blotted with filter paper to remove excess solvent on the sample surface. At a given time and fixed temperature (30 °C) the blends were weighed again to calculate the different parameters of swelling measurement. Inspect S Scanning electron microscopy (SEM) with magnification up to 150000 X and resolution of 4 nm was used to indicate SEM micrographs of the polymeric blends.

2. Results and discussion

3.1. Specific gravity

Specific gravity is a sign for the formation process, change in microstructure, composition [12, 13], that's effect the quality of the final product. A noticeable decrease in specific gravity indicates pores existence within the product.

The specific gravity (ρ) was calculated according to Eq. (1) [14]

$$\rho = \frac{w_a}{w_a - w_s} \times \text{water specific gravity} \quad (1)$$

W_a is dry weight, and W_s is suspended in water weight.

Figure (1) shows the variation of specific gravity as a function of CaCO₃ loading foamed and unfoamed rubber blend. The specific gravity has approximately the same trend with fitting equations and coefficient of determination value (R^2) shown on figure. The specific gravity of CaCO₃ loading foamed rubber is less than that of unfoamed rubber. The density of 150 phr CaCO₃ for foamed rubber is reduced by about 50% of its original value. While, the specific gravity of 200 phr for foamed NR becomes non applicable because the foaming agent has slight effect on its value compared to that of unfoamed rubber at the same concentration.

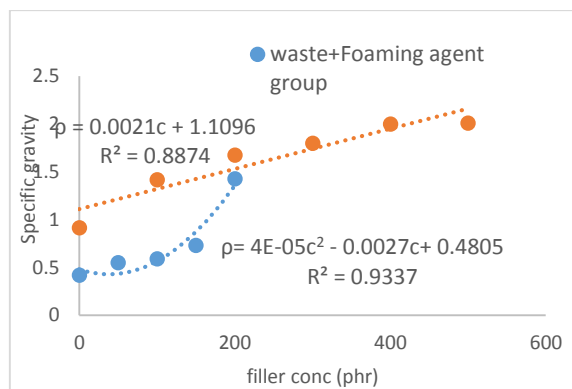


Fig. (1) Specific gravity dependence for CaCO₃ loading foamed and unfoamed rubber blend

3.2. Microstructure measurements

Figure (2) shows the SEM photographs for CaCO₃ (150phr) loading (a) foamed rubber blend, (b) unfoamed rubber blend. There is a good dispersion of the filler in the matrix. Closed air pores in the foamed rubber appeared which may affect thermal, moisture insulation and mechanical properties.

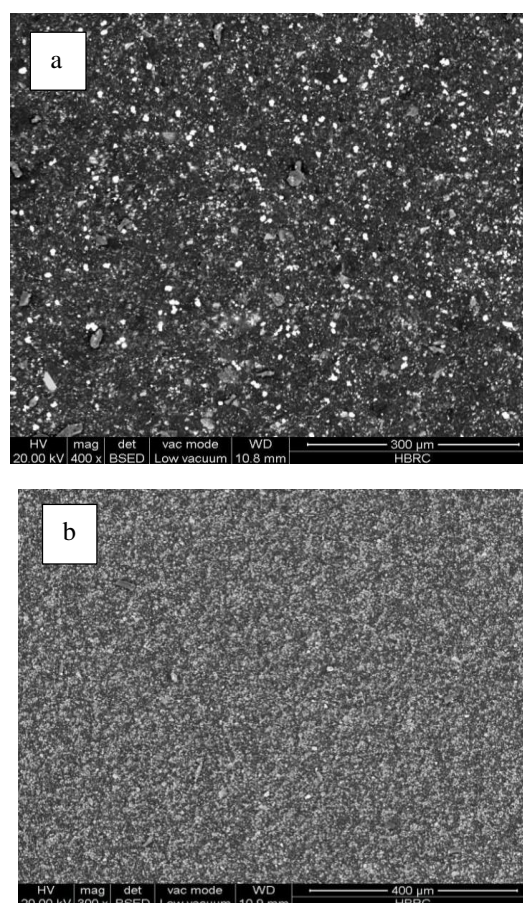


Fig. (2) SEM photographs for CaCO₃ loading (a) foamed rubber blend, (b) unfoamed rubber blend.

3.3. Thermal conductivity measurements

More energy efficient buildings reduce the quantities of fuel consumed consequently reduce environmental pollution. Insulation reduces unwanted heat loss or gain and can decrease energy requirements of heating and cooling systems [15]. Thermal comfort for building occupants can be achieved by using materials with convenient thermal conductivity values as it is considered as an indication of how well a material insulates [16]. Fig. 3(a, b) shows the variation of thermal conductivity with CaCO_3 loading in foamed and unfoamed rubber blend at different temperatures. All samples have thermal stability at different temperatures, CaCO_3 loading foamed rubber blend samples have lower values of thermal conductivity than these of unfoamed rubber blend. In sample with 150 phr CaCO_3 the foaming agent has no effect on thermal conductivity values. K values for CaCO_3 loading foamed rubber blend till 150 phr lying in the range of thermal insulating materials and lower than those of ordinary building materials [4]. Fig. 4 shows a comparison between the thermal conductivities of the two groups. The thermal conductivity increases linearly with unfoamed rubber and polynomial with foamed rubber as CaCO_3 content increases. The fitting equations which give the highest coefficient of determination value (R^2) are shown in the figure. From this comparison, The thermal conductivity at 150 phr of CaCO_3 / unfoamed rubber blend (0.36 W/m.K), which is out of range of the thermal insulating materials [4], and that of the same concentration for foamed rubber blend (0.187 W/m.K). Such comparison indicates that, for the mentioned concentration of CaCO_3 , the addition of foam reduces the thermal conductivity by about 43% lower than that of unfoamed (which emphasizes the specific gravity values) and its value is in the range of insulating masonry materials. Then, the thermal conductivity values start to increase and become non applicable for building insulation.

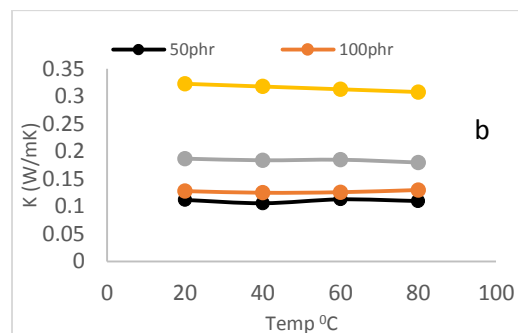
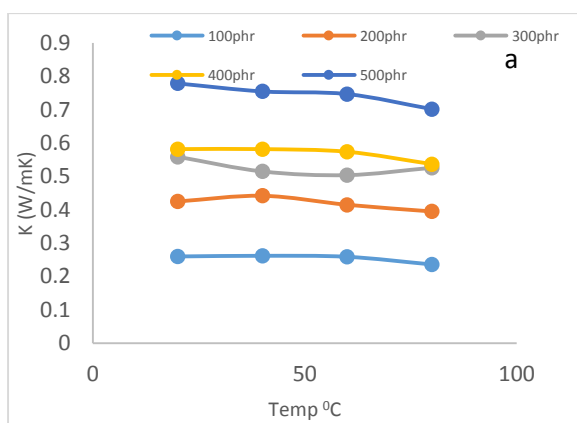


Fig.(3) Shows the variation of thermal conductivity for CaCO_3 loading (a) rubber blend (b) foamed rubber blend

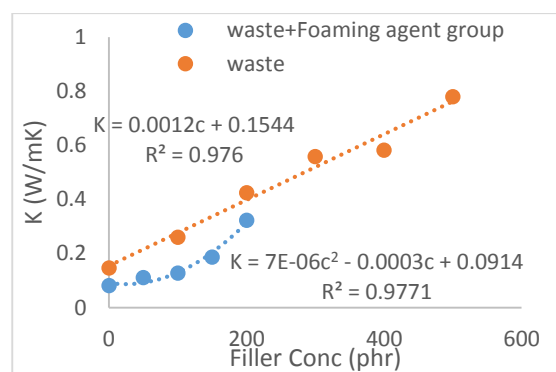


Fig.(4) A comparison between CaCO_3 loading foamed and unfoamed rubber blend

From Fig. (4) There is a correlation between the thermal conductivity and the specific gravity shown in Fig (5) and is given by the following relation [17]:

$$K = A \exp^{B\rho} \quad (2).$$

Where K is thermal conductivity, A and B are constants and ρ is specific gravity.

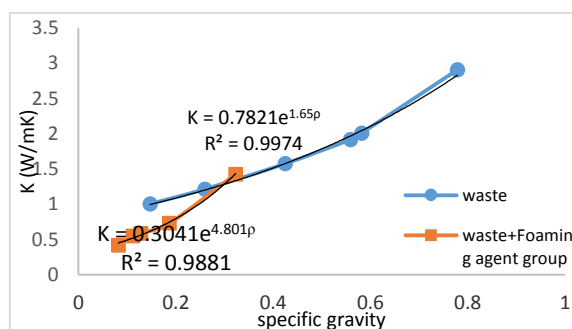


Fig. (5) Thermal conductivity versus the specific gravity for CaCO_3 loading foamed and unfoamed rubber blend

3.4. water absorption:

Water absorption percent for any construction material is a substantial factor to determine its moisture insulating quality. The samples were immersed in water for 24h, the weights before and after immersion were recorded. Water absorption present was calculated according to eq. (3 according to ASTM (D570) [8].

$$\text{water absorption \%} = \frac{m_t - m_0}{m_0} \times 100 \quad (3)$$

Where, m_0 is the sample mass before immersion, m_t is the sample mass after 24h immersion.

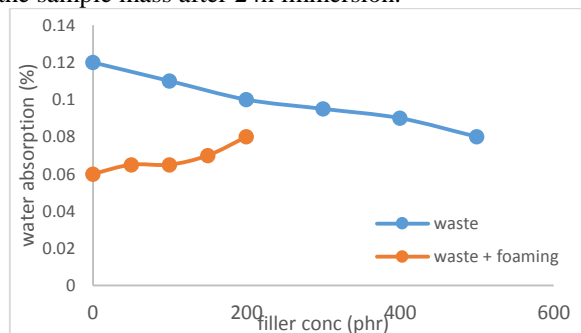


Fig. (6) The variation of water absorption with CaCO₃ loading foamed and unfoamed rubber blend.

From fig (6) The values of water absorption for foamed rubber blend is lower than that of unfoamed

blend this may be attributed to the existence of closed air pores in foamed rubber that proved by SEM and specific gravity values.

3.5. Water tightness measurements:

All samples are connected to the water tightness system, at pressure of (0.6, 2, 4, 6 bars) without any infiltration or cracking, all samples passed the test. The sample considered to be accepted at 0.6 bar according to EN standard [EN: 1928] [9].

3.6. Swelling properties

Diffusion mechanism in rubber is essentially connected with the ability of polymer to provide pathways for the solvent to progress in the form of randomly generated voids. As the void formation decreases with filler loading, the solvent uptake also decreases. The degree of swelling (Q%) of the samples was calculated as follows

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

Where M_t is the solvent absorbed (benzene) at time t , M_e polymer mass 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 square root of time $t^{1/2}$.

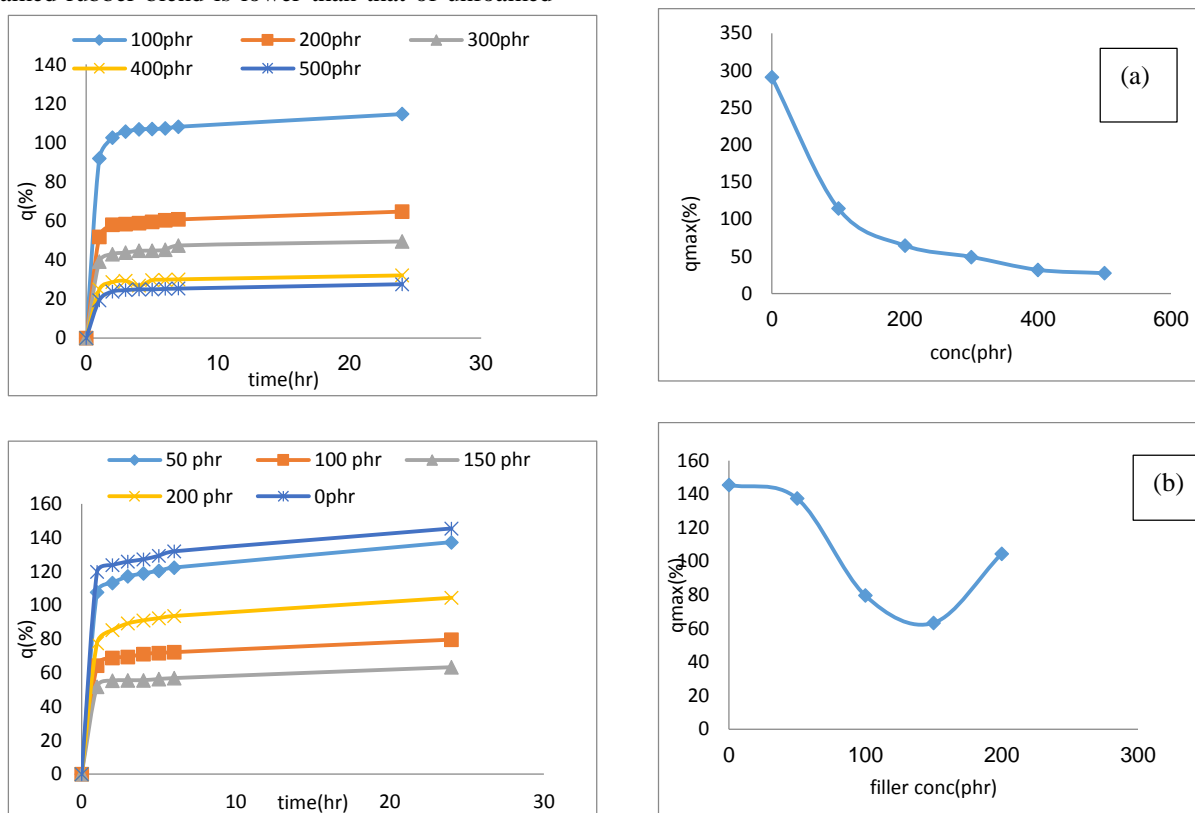


Fig. (7): Sorption curves for (a) unfoamed, (b) foamed NR/SBR blend loaded with different CaCO₃ concentration.

The diffusion coefficient in the unfoamed and foamed rubber blend loaded with CaCO_3 is determined, in Fig. (7). All the sorption processes are similar in nature, on the contrary of expectation for foamed rubber, the solvent uptake decreases. The diffusion coefficient decreases for both groups as CaCO_3 content increases. In the case of foamed rubber blend, the concentration 150phr CaCO_3 has minimum value of diffusion coefficient. The diffusion coefficient of foamed rubber loaded with CaCO_3 is lower than that of the unfoamed one due to the contribution of closed pores during vulcanization process.

Swelling index was calculated using the following equation [18].

$$q - 1 = \left[\left(\frac{W}{W_0} \right) - 1 \right] \frac{\rho_c}{\rho_s} \times 100 \quad (5)$$

(5)

Where q is the ratio of swollen volume to original unswollen volume of samples, $q - 1$ is swelling index, W_0 is the mass of sample before swelling, W is the mass of sample after swelling for 24hr. ρ_c and ρ_s are the densities of the specimen and the solvent, respectively.

Evidently, for a given solvent, the higher the crosslink density of the rubber the lower the degree of swelling is obtained, while 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 [19, 20]:

$$M_c = - \frac{\rho_p V_s V_r^{1/3}}{\ln(1-V_r) + V_r + \chi V_r^2} \quad (6)$$

Where ρ_p is the density of polymer; $\rho_p(\text{NR/SBR}) = 0.913 \text{ g/cm}^3$, V_s is the molar volume of the solvent (benzene) = $89 \text{ cm}^3/\text{mol}$, χ is the interaction parameter V_r is the volume fraction of swollen rubber and given by the following relation

$$V_r = \frac{1}{1+Q} \quad (7)$$

Where Q is defined as grams of solvent per gram of rubber hydrocarbon and calculated by

$$Q = (W - W_d) / W_d \quad (8)$$

Where W_d is the deswollen weight of the sample. 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:

$$v_e = \rho_p N_A / M_c \quad (9)$$

Where: N_A is the Avogadro number.

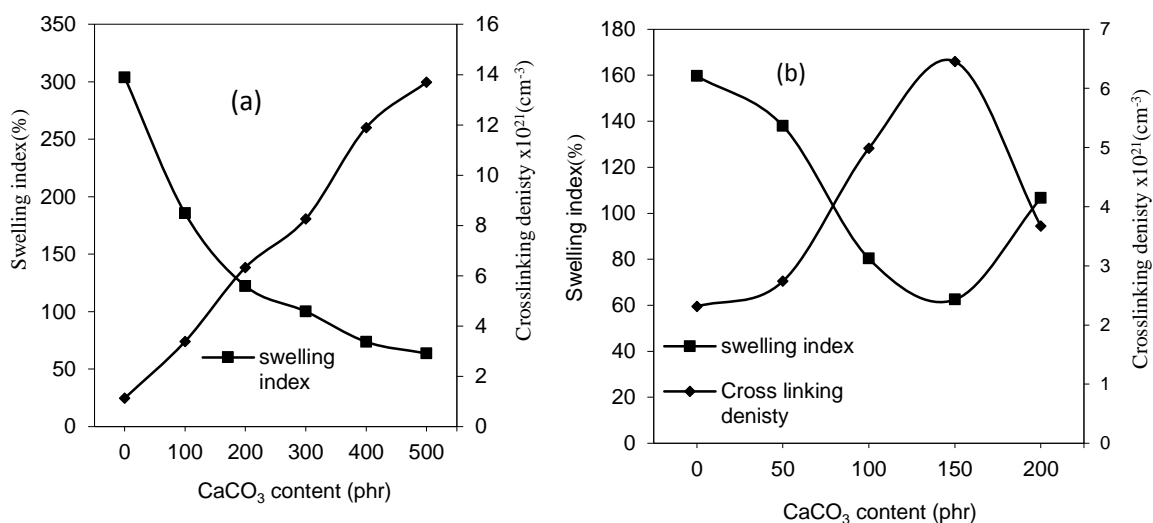


Fig. (8): The variation of the crosslink density and swelling index as a function of CaCO_3 content a) unfoamed rubber b) foamed rubber

The dependence of the crosslinking density and swelling index on variation CaCO_3 content for unfoamed and foamed rubber blend are shown in fig. (8). The same trend occurred for the swelling index and crosslink density values without the emergence of maximum or minimum behavior. Fig. (8) shows that the optimum concentration for CaCO_3 /foamed rubber group is 150 phr. At this concentration, the crosslinking density is higher than that of without foam

whereas the swelling index is lower. From all previous swelling measurement, crosslinks of the filler form a barrier and a substantially reduces diffusion length of the solvent molecules. Thus, the absorption values decrease gradually with the increase in filler level. The increase in crosslinking density in the case of foaming agent may be a result of the presence of the closed pores.

Filler–Polymer Interaction

Another model used for investigating the extent of interaction between rubber and filler can be assessed using Kraus equation [21] which is given by

$$\frac{V_{ro}}{V_{rf}} = 1 - m \left(\frac{\phi}{1-\phi} \right) \tag{10}$$

Where V_{ro} and V_{rf} are the volume fraction of rubber in the solvent-swollen gum and filled vulcanizates, respectively, ϕ is the volume fraction of filler and m is the filler-polymer interaction parameter. V_{rf} is given by the following equation:

$$V_{rf} = \frac{(W_d - FW_o)\rho_p^{-1}}{(W_d - FW_o)\rho_p^{-1} + (W - W_o)\rho_s^{-1}} \tag{11}$$

Where F is the weight fraction of the insoluble components in the specimen and $(W - W_o)$ is the amount of solvent absorbed.

Since equation (10) has the general form of an equation of a straight line, a plot of V_{ro}/V_{rf} as a function of $\phi/(1-\phi)$ should give a straight line with a negative slope. The parameter m obtained from the slope of this plot describes how much swelling is restricted for a given volume fraction of filler; it is basically a measure of the filler-polymer interaction during the swelling process. According to Kraus theory, the parameter (m) will be a direct measure of the reinforcing ability of the filler used. This means that the more the reinforcing ability of the filler, the more the swelling resistance caused by that filler. The plots of Kraus equation for waste CaCO_3 filled unfoamed or foamed rubber are shown in Figure (9). Figure (9) shows that the higher negative slope for waste CaCO_3 filled rubber foam than these of waste CaCO_3 /rubber indicating a better reinforcing effect due to the effect of closed air pores which may contribute to higher reinforcement besides CaCO_3 reinforcing filler.

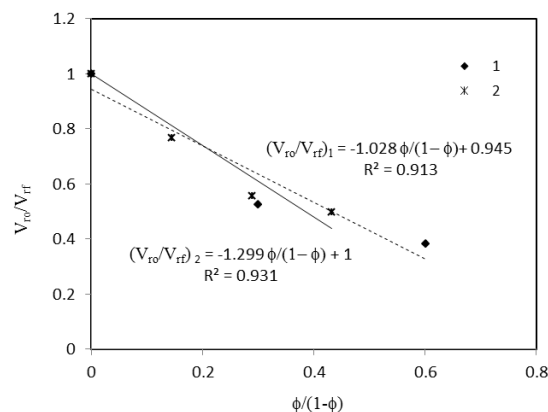


Fig. (9) V_{ro}/V_{rf} versus $\phi/(1-\phi)$ for samples in benzene 1) CaCO_3 /rubber blend 2) CaCO_3 / foamed rubber composites.

3.7. Mechanical measurements:

Natural rubber inherently possesses high strength due to strain-induced crystallization [22]. When filler (CaCO_3) is incorporated into rubber blend, the regular arrangement of rubber molecules is disrupted, hence the ability for crystallization is lost. This is the reason why filler reinforced rubber composites possess lower tensile strength than gum compounds Fig. (10) The tensile strength for CaCO_3 / foamed rubber group increases as CaCO_3 increases and such values lying below pure unfoamed rubber which may be attributed to existence of the air pores which soften these samples.

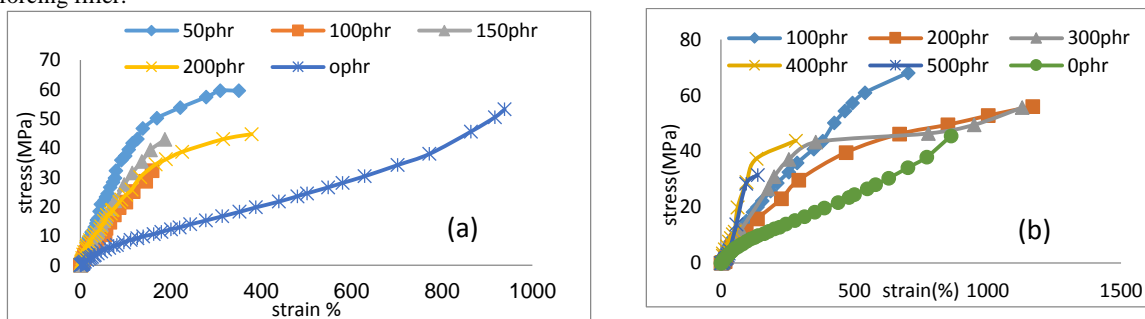
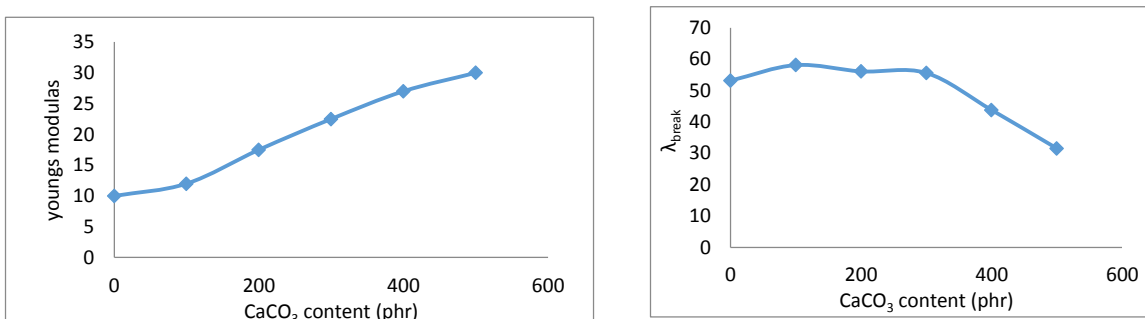


Fig. (10); Stress – Strain relationship for CaCO_3 loading (a) foamed rubber (b) unfoamed rubber



g (11): The variation of youngs modulus and λ_{break} as function of concentration of CaCO_3 loaded unfoamed rubber blend

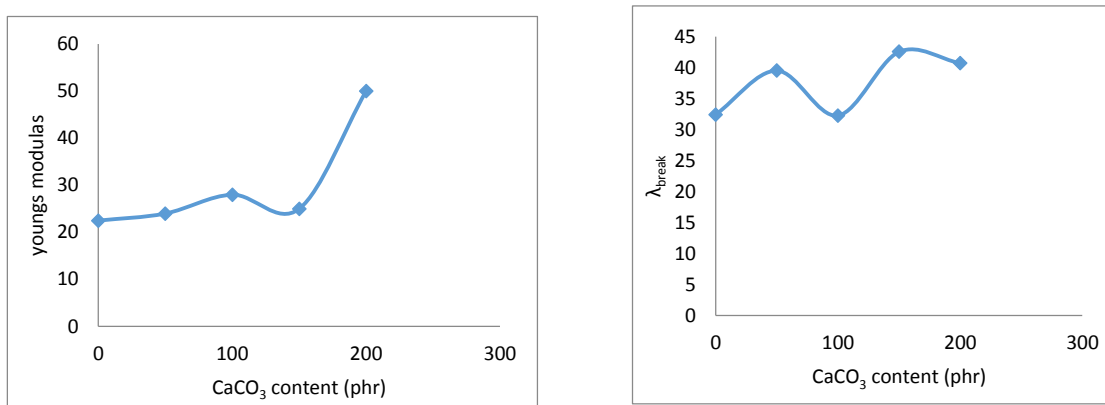


Fig. (12): The variation of youngs modulus and λ_{break} as function of concentration of CaCO_3 loaded foamed rubber blend

Figures (11, 12) show the dependence of tensile modulus and elongation at break on the variation of CaCO_3 concentration for unfoamed and foamed rubber respectively. It can be seen that, as the CaCO_3 loading increases, the tensile modulus also increases and elongation decreases. From figures (11, 12) the Young's modulus of CaCO_3 /foamed NR is greater than the unfoamed samples at the same concentration while the elongation at break is lower due to the contribution of closed pores in enhancement the reinforcement. The optimum concentration for CaCO_3 /foamed rubber group is 150 phr. The reason of reduction in tensile modulus after optimum concentration is due to agglomeration of the filler particles to form a domain that acts as a foreign body

or as a result of physical contacts between adjacent aggregates [23].

The crosslinking densities were determined from the elastic rubbery moduli of the samples according to the rubber elasticity theory modified by Nielsen [24], from eq. (12) Where ν represents the crosslinking density which is defined as the number of moles of chains per cm^3 , R is the gas constant, T is the temperature in Kelvin (300K) and E is the elastic modulus obtained from the stress-stain curves. The obtained values is represented in table (3), this values are emphasized by the calculated values of the crosslinking densities calculated from swelling measurements. It was found that they have the same trend and order of magnitude.

$$\nu = \frac{E}{3RT} \quad (12)$$

Table (3): The variation of crosslinking densities as a function of concentration of CaCO_3 loaded foamed and unfoamed rubber blend

CaCO_3 loaded rubber blend	$\nu \times 10^{21} (\text{cm}^{-3})$	CaCO_3 loaded foamed rubber	$\nu \times 10^{21} (\text{cm}^{-3})$
0	0.84	0	1.65
100	1.13	50	2.98
200	0.99	100	2.50
300	1.31	150	2.79
400	2.17	200	2.33
500	0.92		

3.8. Hardness measurements:

Shore hardness is the resistance of the material to permanent deformation induced by mechanical indentation or abrasion. Fig (13) shows the variation of hardness as a function of CaCO_3 concentration in

foamed and unfoamed rubber. From figure (13) the values of the hardness for foamed rubber are less than that of unfoamed rubber due to the contribution of air pores in the matrix which cause soften of the samples, except for 200 phr as the foaming agent effect reduced.

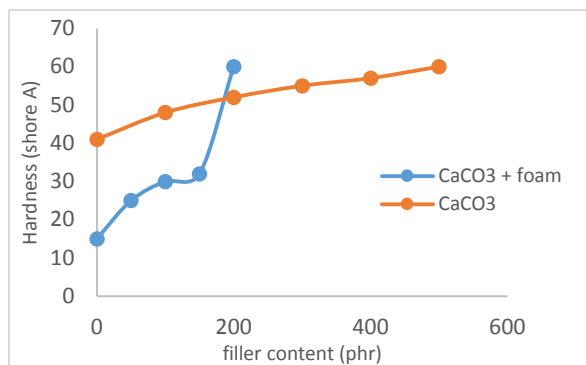


Fig (13): The variation of hardness as a function of CaCO_3 concentration in foamed and unfoamed rubber.

3. Conclusion

Waste management can reduce the undesirable human remnants, so a new frugal material can be produced for many application: The thermal conductivity increased with increasing CaCO_3 for unfoamed and foamed rubber composites, but thermal conductivity values of reinforced CaCO_3 foamed rubber composites until 150phr lying in the range of thermally insulating material. The density had the same trend as the thermal conductivity. The diffusion coefficient and swelling index exhibited the decrease trend until the (150phr) and then increase. The crosslinking density exhibits an increase behavior till 150 phr then decrease. The elastic modulus had the same trend as the crosslinking density. The elongation at break has the opposite trend as the elastic modulus. The crosslinking density is calculated using Nielsen relation and it gives the same trend and order of magnitude as these calculated in the swelling measurement. Samples of CaCO_3 loading rubber blend can be applied as a sustainable low cost waterproofing membranes with good efficiency and these of CaCO_3 loading foamed rubber blend till 150 phr can be applied as a multifunctional sustainable building materials as they has efficient thermal insulating and waterproofing properties

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