Manifestation of Waste Silicate Type Additives and Electron Beam Irradiation on Properties of Sbr/devulcanized Waste Tire Rubber Composites for Floor Tiles Applications

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Research Article

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Manifestation of waste silicate type additives and electron beam irradiation on properties of SBR/devulcanized waste tire rubber composites for floor tiles applications

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Abstract

Virgin styrene-butadiene rubber (SBR) was replaced by devulcanized waste tire rubber (DWR) 50/50 and used as a rubber base for preparing composites to depend on different silicate types at fixed content 40 phr (part per hundred part of rubber). All composites were mixed on a rubber roll mill and then subjected to electron beam irradiation to induce cross-linking at a dose of 100 kGy. Different silicate fillers were used in this study like precipitated silica (PS) 40 phr, waste glass window (WG) - PS 20/20 phr, fly ash (FA)-PS 20/20 phr, and micaosilica (MS)-PS 20/20 phr. Waste silicate was treated with (3-aminopropyl)trimethoxysilane (APTMS) and blended with PS. Mechanical properties were investigated for composites like tensile strength, elongation at break, tensile modulus, and calculation of cross-link density from mechanical. As well as, application for floor tiles included compression set and abrasion resistance measurements. All results indicated an enhancement in tensile strength, modulus, and cross-link density by adding silicate fillers and more enhanced in presence of radiation. For the application of floor tiles, the MS filler gave a good compression set and abrasion resistance followed by other silicate fillers (PS, FA), except WG.

Keywords: styrene-butadiene rubber, devulcanized waste tire rubber, silicate fillers, mechanical properties, floor tiles, electron beam irradiation.

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Introduction

The significance of polymer reprocessing has seriously increased through the latest years. Eco-friendly and cost-effective strategies have been utilized for
waste management. Rubber is a significant constituent in the civic solid wastes. It has to be reclaimed after discarding [1]. The blending of reclaimed rubber with virgin polymer matrix diminishes the expense of the final product and supports in lessening the environmental contamination. Numerous techniques [2] were tried to lessen this ecological burden by providing well-organized procedures for waste rubber (WR) reprocessing. The presence of worn-out tires in landfills acts as nourishing media for insects to spread diseases. Besides, tire scarps can produce poisonous fumes upon ignition which leads to environmental pollution [3]. Discarded tire rubber as a vulcanized polymer is one of the solid waste pollutants that cannot be disposed of easily with its cross-linked arrangement. Sulfur is one of the used chemicals for rubber vulcanization through composing both S–S and C–S bonds creating a cross-linked matrix. Upon devulcanization, the S–S and C–S bonds are predictable to separate; nonetheless, this does not occur perfectly. For a fruitful devulcanization step, the cross-linked structure needs to separate without chain scission. Devulcanization techniques incorporate mechano-chemical, reclaiming, grinding, devulcanization microwave, and ultrasonic processes. Among these techniques, rubber mechano-chemical devulcanization is utilized. This is achieved by converting the vulcanized or scrap waste rubber mechanically and chemically at high temperatures into a beneficial material ready to be devulcanized and processed [4].

A wide assortment of particulate fillers is utilized in the rubber industry to revise and improve the physical properties of elastomeric materials. The addition of filler frequently prompts an expansion in modulus and significant abrasion and tear resistance. Although the mechanisms of reinforcement are not completely understood, there is an overall agreement about the essential process adding to the stress-strain conduct of the filled vulcanizate [5]. Numerous kinds of fillers are utilized for the improvement of mechanical properties. Carbon black is the most extensively utilized filler in rubber industries owing to its particulate nature and higher surface area, which grants high reinforcing capacity. Nonetheless, these days, more consideration is being paid to nonblack mineral fillers such as silica, owing to the monotonous color and the high costs of carbon black. Looking for lessening the cost of composite, new materials are being considered to supplant absolutely or somewhat the customary ones as a simple economical measure or to grant some required properties. Numerous efforts have been made to utilize silica
from the characteristic assets as substitute reinforcing filler in synthetic and natural rubbers in light of cost investments, better dimensional dependability, excessive mechanical properties, and environmental issue. Silica has various hydroxyl groups on the surface, which results in strong filler interaction and absorption of polar materials by hydrogen bonding. In the meantime intermolecular hydrogen bonding between hydroxyl groups on the surface of silica is very strong; it can form tight aggregates [6, 7]. The best approach to keep away aggregation is to enhance the compatibility between rubber and silica to lessen the silica migration. Thus, hydrated silica confers better physical properties to polar synthetic rubbers than it doesn't to nonpolar rubbers such as styrene-butadiene rubber (SBR), natural rubber, and so on and is utilized to create colored articles that need high strength properties [8].

New methodologies were explored by researchers to replace carbon black to diminish the utilization of petroleum. This prompts the institution of inorganic fillers such as silica [9] into the rubber, which assists with creating colored products. Silica is the most widely recognized inorganic filler utilized in the rubber industry for reinforcement [10, 11], conversely, the isolated silanol groups and hydrogen-bonded silanol groups on the surface of silica decrease its compatibility with non-polar rubber [8]. So, silica was revised with silane coupling agents [11, 12] to make it compatible with non-polar rubbers and hybrid filler systems [13] were likewise utilized nowadays.

The reason for consolidating inorganic filler into the polymer network isn't just to accomplish exceptional properties of polymer composites but also to diminish the expense. For ecological protection and sustainable improvement, people have required an excessive effort to manufacture polymer/FA composites that come across the necessity of practical application [14, 15].

Blending has to turn out to be the effective technique for growing the utilization of unique materials, and of accomplishing certain necessary properties, by joining the good original properties of individual parts. Rubber blends have established increasing consideration from numerous researchers around the world [16]. Silica (silicon dioxide) is the most bountiful mineral on the Earth [17]. Silica can be utilized as filler for rubber [18], which is utilized in the manufacture of numerous products, like tires and other industrial materials. Its numerous benefits
include: improving the mechanical durability, shrinkage, heat resistance, thermal extension, and stress of the rubber-established composite. These merits are owing to the substitution of a soft matrix by hard inorganic filler. The silica was improved with silane coupling agents, preceding its application as reinforcement filler for the NR/SBR blend. The alteration prompted the improvement of the mechanical properties and thermal degradation. Composites dependent on the silica and modified-silica filled rubber [19], have been broadly studied.

Fly ash is unavoidably created as waste after burning. About 750 million tons of fly ash is produced yearly, nonetheless, just 39 % can be reused in the U.S [20]. If fly ash is discharged to the environment via air and wind, a severe contamination problem would be made with the hazard of pneumonic diseases. Recycling fly ash is an effective method to avoid such contamination. Comparable carbon black (CB) and precipitated silica, fly ash can likewise go about as reinforcing filler to enhance the mechanical properties of rubber compounds. Palm-constructed fly ash can be utilized to modify the mechanical properties of thermoplastic materials [21]. Glass waste that got from the metropolitan district can use as a brick added substance [22].

Everywhere in the world, the issue of disposing of waste tires is increasing day by day due to the growth in the number of vehicles on the road. The burning of tires and direct landfilling strategies prompts environmental degradation. Because of the non-biodegradable nature, the decomposition of wastes in the environment takes much time [23].

Upon manufacturing polymer blends or composites, gamma irradiation can be utilized to improve the compatibility between the ingredients by achieving the polymerization method. Besides, better interaction initiates with enhanced interfacial adhesion between composite components at low irradiation doses promoting improved properties without degrading the polymeric network [24].

In this study, we prepared styrene-butadiene rubber (SBR)/devulcanized waste tire as a rubber base for preparing composites depend on different silicate types and afterward exposed them to electron beam irradiation to induce crosslinking.

2 Materials and techniques
2.1 Materials

SBR.Europrene® 1502, SBR virgin rubber acquired by cold polymerization, Mooney viscosity ML 1 + 4(100 °C) = 52, bound styrene, 23.5 wt %, provided by Versalis S.p.A. company, Piazza Boldrini, San Donato Milanese (MI), Italy.

- TMTD, a reclaiming agent of the molecular formula C\textsubscript{6}H\textsubscript{12}N\textsubscript{2}S\textsubscript{4}, molecular weight 240.43 g/mol and density 1.43 g/cm\textsuperscript{3}, provided by Zhedong Rubber Auxiliary Co., Ltd., China.

- Stearic acid, M.wt. = 284.49, molecular formula: CH\textsubscript{3}(CH\textsubscript{2})\textsubscript{16}COOH, melting point: 54 °C, PROLABO, France.

- Zinc oxide (ZnO), a white to yellow tined powder, 98.0 % purity commercial-grade, contains lead sulfate 0.25 % maximum and calcium residue 0.15% maximum, El Nasr Chemical Co. Egypt.

- 1,2-dihydro-2,2,4-trimethyl quinolone (TMQ) as an antioxidant in the form of brown granular beads with the chemical formula (C\textsubscript{12}H\textsubscript{15}N)n, n = 2–4, specific gravity: 1.05 g/cm\textsuperscript{3}, provided by Alfa Aesar GmbH, Germany.

- (3-Aminopropyl) trimethoxysilane (APTMS): Mw 179.20 g/mol, molecular formula C\textsubscript{6}H\textsubscript{17}NO\textsubscript{3}Si and density 0.903 g/ cm\textsuperscript{3}, was supplied by Sigma Aldrich., Germany.

- Silicate additives are four different types of silica-containing fillers additives. Waste-Glass (WG), commercial soda-lime-silica glass, was utilized with the chemical compositions of WG particles used in this study are listed in Table I. 

Silica bypass form ferromanganese byproduct named, Micaosilica (MS), that provided from Egyptian Ferroalloys Co., Egypt) has the next chemical composition presented in Table 1.

Fly Ash (FA), having a density of 2.33 g/cm\textsuperscript{3} and total evaporable moisture content of 1.54 % was formed from the ignition of coal as a byproduct. The particle size of FA falls in the range of 63 μm. The chemical composition of the FA (utilized in this study) was characterized by Energy dispersed X-ray analysis (EDX) as presented in Table 1.
Precipitated silica, SUPERSIL-140, was provided from Supersil Chemical Company, Mumbai, Maharashtra, India. SUPERSIL-140, having a bulk density of 0.235 g/cm³, has a surface area of 140-170 m²/gm, contains 89% SiO₂, 16-18 μm particle size, L.O.I at 1000 °C is 10 wt. %.

Table 1: Chemical composition of WG, MS and FA particles

| Chemical oxides | Content % | Chemical oxides | Content % | Chemical oxides | Content % |
|-----------------|-----------|-----------------|-----------|-----------------|-----------|
| SiO₂            | 71.9      | SiO₂            | 90        | SiO₂            | 59.23     |
| Fe₂O₃           | 0.08      | Fe₂O₃           | 4         | Fe₂O₃           | 5.03      |
| Al₂O₃           | 0.66      | Al₂O₃           | 1.5       | Al₂O₃           | 32.16     |
| CaO             | 8.6       | CaO             | 1         | CaO             | 0.99      |
| MgO             | 4.05      | MgO             | 1.5       | TiO₂            | 2.59      |
| K₂O             | 0.037     | K₂O             | 1.5       |                  |           |
| Na₂O            | 14.25     | Na₂O            | 0.5       |                  |           |
| TiO₂            | 0.025     | L.O.I. at 750 °C| 2         |                  |           |
| SO₃             | 0.32      |                  |           |                  |           |
| Impurities      | 0.078     |                  |           |                  |           |

2.2. Modification of waste silicate additives by APTMS silane

100 g of silicates types fillers were mixed with 100 mL 5wt% solution of APTMS in ethanol/water (80/20 v/v) in a polyethylene beaker under constant stirring by a Teflon rod, followed by removal of solvent via evaporation for 30 min in an oven overnight at 50 °C, the dry fillers were then ground into fine powders at 64 μm particle size.

2.3. Preparation of rubber composites

The SBR/DWR blends at 50/50 wt. %, the method for preparation of these blends was presented in previous work [25], and this composition was taken in this
study, according to superlative mechanical properties gained for this blend. Composites of this blend and other additives were prepared in an open mill as stated in Table 2. Subsequent to mixing, the samples were hot-pressed at 160 °C under 10 MPa for 5 min into sheets of fitting thickness and size for examination.

Table 2: Formulations for prepared SBR/ DWR composites utilizing different silica additives.

| Ingredients            | Blank sample(phr) | Composites (phr) |
|------------------------|-------------------|------------------|
| 50/50 SBR/DWR (wt. %)  | 100               | 100              |
| ZnO                    | 5                 | 5                |
| Stearic acid           | 2                 | 2                |
| TMQ                    | 1                 | 1                |
| Precipitated silica (PS)| 0                | 40               |
| FA : PS                | 0                 | 20:20            |
| GW : PS                | 0                 | 20:20            |
| MS : PS                | 0                 | 20:20            |

2.4 Electron beam irradiation of prepared blends

Irradiation of samples was done by utilizing electron beam accelerator (Energy 3 MeV, power 90 kW, Beam current 30 mA, conveyer speed 16m/min (50HZ) and scan width variable up to 90 cm) at the National Center for Radiation Research and Technology (NCRRT), Cairo, Egypt. The rubber blends were exposed to electron beam irradiation at ambient conditions for 100kGy. Note: this dose of radiation 100 kGy was acquired, as it is appropriate dose for good mechanical properties of SBR/DWR 50/50 wt. % blends [25].

2.5 Mechanical measurements

The tensile properties of the dumbbell-shaped samples were estimated by utilizing a Universal mechanical computer-aided testing machine, V1.6 Model HDB6004B-S, made by Haida International equipment LTD Company, Daojiao town, Dongguan city, Guangdong province, China with calibrated load cell, 5000 N. The ASTM D 412-16 techniques were followed in determining the next
mechanical properties: Elongation at break, tensile strength, modulus at 100 % and 300 % strain respectively. The average value of all mechanical parameters had been estimated for the prepared samples measured using at least three identical samples. A crosshead speed of 500 mm/min was utilized and the experiments were implemented at 25 °C. Shore hardness (Shore A) has been estimated for all samples utilizing Shore hardness A according to ASTM.

The compression set was done out as indicated by ASTM D 395-85 on a standard test specimen of the cylindrical shape of 16 ± 0.1 mm diameter and 6 ± 0.5 mm thicknesses. The test sample was located between the plates of the compression gadget with the spacers on each side of it, permitting enough clearance for expanding the rubber when compacted. The bolts were squeezed so the plates are drawn together consistently until they are in contact with the spacers. The assembled compression device was then placed at room temperature 20 °C for 22 h. The test sample was lastly removed and permitted to cool for 30 min, and the final thickness could be estimated by an electronic digital caliper with 0.01 mm accuracy. The compression set is defined as:

\[ C\% = \left( \frac{t_o - t_1}{t_o - t_s} \right) \times 100 \]  

Where \( t_o \) is the original thickness of the sample; \( t_1 \) is the thickness of the sample after removal from the clamp, and \( t_s \) is the thickness of the spacer bar utilized. Abrasion resistance tests were made by utilizing an abrasion tester type AP.40 (Maschinebau GmbH Rauenste in Thuringen, Germany). The loss in the mass percent was determined by ASTM D 3389-75 (1982), by equation (2):

\[ \text{Mass loss (mg) per revolution} = \left( \frac{W_i - W_f}{n} \right) \times 1000 \]  

Where \( W_i \) is the initial mass of sample (g), \( W_f \) is the final mass of sample (g), and \( n \) is the number of revolutions, 84 revolutions.
2.6 Determination of cross-link density of rubber networks determined from mechanical data

Measurement of stress-strain response gives a simple technique for assessment of the cross-link density of polymer networks. As indicated by the classical kinetic theory of rubber elasticity was initially developed by Wall, Flory, James and Guth [26]. They ascribed the high elasticity of a cross-linked rubber to the variation of the conformational entropy of long flexible molecular chains. The theory predicts the following relation in simple extension; equation 3:

\[ \sigma = A \Phi v_e KT (\lambda^2 - \lambda^{-1}) \]  

Where \( \sigma \) is the true stress, the force per unit area measured in the strained state, \( v_e \) is the number of effective plastic chains per unit volume, \( K \) is Boltzmann’s constant, \( T \) is the absolute temperature, and \( \lambda \) is the extension ratio; \( A \Phi \) is a pre-factor depending on the considered model. The elasticity of natural and SBR rubbers in simple extension at a constant strain rate was studied [27]. They plotted the true stress as a function of \( (\lambda^2 - \lambda^{-1}) \) as recommended by the molecular theory. They acquired a series of straight lines which don’t go through the origin. In contrast, rubber elasticity theory predicts that the relation between the tensile strength and the elongation ratio, \( \lambda \), is illustrated in equation 4:

\[ \sigma = \sigma_o(\lambda) + E(\lambda^2 - \lambda^{-1}) \]  

Where \( E \) is the modulus of elasticity and \( \lambda \) is the extension ratio of the strain that happened because of the applied stress. The relation between \( (\lambda^2 - \lambda^{-1}) \) and stress \( (\sigma) \) for rubber blends or composites were drawn in figures. From these figures, it has been calculated the slope of the lines represent these relations, and then tried to calculate the average molecular weight \( M_c \) between cross-links from the value (G) as shown in equation 5:

\[ G = 3E = A \Phi \rho RT/M_c \]  

Where \( G \) is the shear modulus, \( \rho \) is the density of the rubber and \( R \) the gas constant, the value of \( M_c \), the molecular weight between two cross-links can be determined and then the cross-link density \( \nu \) (CD) can be determined from equation 6:

\[ Crosslink\ density\ (\nu) = 1/2M_c \]  

Thus cross-link density is contrarily proportioned with double the molecular weight between two cross-links. Consequently, cross-link density was established
to be directly proportional to the true tensile modulus as indicated by equations 5 and 6.

2.7. FTIR investigation.

The chemical structure of the various types of silica untreated and treated with silane was analyzed by ATR-FTIR spectroscopy, Bruker Optik GmbH, Ettlingen, Germany, over the wavenumbers range 4000-500 cm\(^{-1}\).

3- Results and discussion
3.1. Fourier Transform Infrared Spectrometry (FTIR)

FTIR is utilized to study the chemical interaction between silane and silica particles. Figure 1a shows a typical FTIR spectrum for micaosilica (SiO\(_2\)~90\%) and its silane treatment. From the spectrum, the bands at 797 and 477 cm\(^{-1}\) are the characteristic bands of amorphous silica [28]. The strong broad band at approximately 1100 cm\(^{-1}\) corresponds to the frequency range of the siloxane group (Si–O–Si) vibration in silica [29]. The characteristic band located at 1620–1650 cm\(^{-1}\) was assigned to bending vibrations of aliphatic amine (N–H) groups. The surface of silica will usually contain an appreciable concentration of hydroxyl groups (–OH), the broad absorption band at 3300–3730 cm\(^{-1}\) is due to the stretching vibration silanol groups (Si-OH), hydroxyl groups (water or alcohol) stretching by hydrogen bonding [30]. When the MS particles are treated silane, a new two bands appeared at 2853 and 2923 cm\(^{-1}\) characteristic to asymmetric and symmetric stretching vibrations of –CH\(_2\) indicating grafting of APTMS on the surface of MS [31], the intensity of OH peak at 3300–3730 cm\(^{-1}\) due to hydrogen bonding, besides stretching vibrations of aliphatic amine (N–H) groups for silane was not affected, despite, the involvement of the OH groups during the reaction of silane with silanol group of silica, as shown in figure 2 [31]. Furthermore, for the fly ash (SiO\(_2\)~59.23\%), In raw fly ash (FA), the broad peak at 3412 cm\(^{-1}\) was ascribed to the stretching and deformation vibrations of OH and H-O-H groups from the water molecules, the presence of silica and alumina induced different linkages which have different vibration modes for identification. High intensity of Si–O–Si at 1435 cm\(^{-1}\) and Si–O–Al at 870 cm\(^{-1}\) [32] as shown in figure 1b. FA treated by silane shows the change in the intensity of the OH groups in silica, which resulted in lower reactivity of FA for silane than in the case of MS. FTIR spectra GW (SiO\(_2\)~71.9\%) untreated with silane showed the same characteristic peaks of silica and after treated with silane the characteristic peak of silane is
present with little change in the intensity of the OH groups in silica, which resulted in a lower quantity of hydrogen bond than in the case of MS as shown in Figure 1c.

**Figure 1:** FTIR of (a) Micaosilica, (b) Fly ash, and (c) Glass waste untreated and treated with silane, respectively.
Figure 2: The scheme represents the possible reaction between the silane coupling agent and silicate surfaces.
3.2 Mechanical properties

3.2.1 Tensile strength

Figure 3 represents the tensile strength (TS) for SBR/DWR 50/50 composites that unirradiated and irradiated at 100 kGy, also loaded with different silicate fillers with fixed concentrations namely, 40 phr. For unirradiated composites, the TS increases by addition silicate fillers; whatever it is, when compared with control samples; not contain fillers, and composites loaded by PS-MS 20/20 provided the highest values for TS, followed by PS, PS-MS 20/20, and finally PS-GW 20/20. The noticeable increase in TS of composites by PS-MS, due to MS contain high content of SiO\textsubscript{2} more than 90\%, and by treated it by silane, it adhered by creating chemical bonds by silanol groups on the surface of silica, in addition to forming hydrogen bonds between rubber matrix and PS. Alternatively, irradiated composites provided the highest values for TS, when compared with unirradiated ones for the same silicate filler, and irradiated SBR/DWR 50/50 composites that loaded with PS-MS 20/20 gave the highest values for TS as discussed before, as well as increasing cross-link density caused by electron beam radiation that formed free radicals and produces link macromolecules of rubber with each other form one hand, and like of macroradicals of rubber with filler on the other hand. It is observed that irradiated composites loaded with PS-GW 20/20, PS, and PS-FA 20/20 gave comparable values for TS, due to the lower content of silica in it, when compared with MS. Also the increases of TS by radiation due to physical bonds formed between rubber matrix and silicate fillers by radiation.
Figure 3: Tensile strength for SBR/DWR 50/50 composites loaded with 40 phr of different silicate fillers, unirradiated and irradiated at 100 kGy.

3.2.2. Elongation at break

Figure 4 exhibits the $E_b\%$ for SBR/DWR 50/50 composites that unirradiated and irradiated at 100 kGy, also loaded with different silicate fillers with fixed concentration namely, 40 phr. Generally, for unirradiated composites, the $E_b$ decreases with the addition of silicate fillers, due to the filler restrict stretching. On the other hand, irradiated composites i.e blank samples, and also loaded ones, the $E_b$ increases by radiation for the same composites. For blank samples increases $E_b$ by radiation owing to liberation of carbon black in DWR which resulted in a decrease in $E_b$. On the other hand, the $E_b$ increases by radiation for all silicate fillers owing to cross-linking not enough to retard elongation due to SBR rubber contain phenyl ring that dissipates the energy of electron beam irradiation, besides, it has low $G(X)$ (cross-link/100 eV) about 0.3 when compared with other rubbers [33].
3.2.3. Tensile modulus at 100 and 300% elongation

Figure 5 A and B shows $M_{100}$ and $M_{300}$ respectively for SBR/DWR 50/50 composites that unirradiated and irradiated at 100 kGy, likewise loaded with different silicate fillers with fixed concentration namely, 40 phr. For unirradiated composites the $M_{100}$ and $M_{300}$ increase with all types of silicate fillers, and PS-MS 20/20 filler gave the highest values for modulus, meanwhile PS, PS-FA 20/20 silicate fillers presented comparable values. On the other hand, PS-GW 20/20 filler provided the lowest values for modulus when compared with other silicate fillers type, owing to the non-reactivity of GW, however, it was treated by a silane coupling agent. Irradiated composites were denoted the same behavior, but they have the highest values for modulus when compared with unirradiated ones. Furthermore, silicate filler remain has the highest values for $M_{100}$ and $M_{300}$, it will be recognized that modulus at certain elongation related directly proportion to cross-link density, and composites loaded by PS-MS 20/20 have the highest cross-
link density owing to the reactivity of MS filler to interact with SBR/DWR 50/50 in the presence of electron beam irradiation.

Figure 5: (A) $M_{100}$ and (B) $M_{300}$ for SBR/DWR 50/50 composites loaded with 40 phr of different silicate fillers, unirradiated and irradiated at 100 kGy.
3.2.4. Cross-link density

Figure 6 represents the variation of the cross-link density (CD) for SBR/DWR 50/50 composites which are unirradiated and irradiated at 100 kGy, also loaded with various silicate fillers with fixed concentration namely, 40 phr. Generally, the CD in unirradiated composites increases with the addition of all types of silicate fillers owing to interfacial adhesion between fillers and rubber matrix. Alternatively, irradiated and loaded composites presented comparable values for CD for all forms of silicate fillers, but MS remains has the highest value for CD owing to increasing interfacial adhesion and reactivity of this silicate filler.

![Cross-linking density for SBR/DWR 50/50 composites loaded with 40 phr of different silicate fillers unirradiated and irradiated at 100 kGy.](image)

**Figure 6**: Cross-linking density for SBR/DWR 50/50 composites loaded with 40 phr of different silicate fillers unirradiated and irradiated at 100 kGy.

3.3. Compression set

Figure 7 elaborates the compression set for SBR/DWR 50/50 composites that unirradiated and irradiated at 100 kGy, also loaded with different silicate fillers with fixed concentration namely, 40 phr. For unirradiated composites, it can
be observed that the blank SBR/DWR 50/50 samples have the highest compression set when compared with filled composite. At the same time, this performance was acquired in irradiated composites, but the values of compression set were decreased for the same type of silicate filler when compared with unirradiated one, owing to increased cross-link density which inversely proportional to cross-link density [34, 35]. Besides, it can be seen that the SBR/DWR 50/50 composites loaded by PS-MS 20/20 gave the lowest values for compression set, and this was achieved for unirradiated and irradiated ones, owing to the increased cross-link density of these composites.

![Figure 7: Compression set for SBR/DWR 50/50 composites loaded with 40 phr of different silicate fillers, unirradiated and irradiated at 100 kGy.](image)

**3.4. Hardness**

Figure 8 illustrates the hardness for SBR/DWR 50/50 composites that unirradiated and irradiated at 100 kGy, besides loaded with different silicate fillers with fixed concentration namely, 40 phr. For unirradiated composites, it can be
observed that the blank SBR/DWR 50/50 samples have the lowest values for hardness for all unirradiated and irradiated composites; meanwhile, SBR/DWR 50/50 composites that loaded by PS-MS 20/20 provided the highest values for unirradiated and irradiated composites, owing to increased cross-link density. The composites loaded by PS, PS-MS 20/20, and PS-WG 20/20 provided comparable values for hardness and these for not irradiated composites and irradiated at 100 kGy.

![Figure 8](image)

**Figure 8**: Hardness shore A for SBR/DWR 50/50 composites loaded with 40 phr of different silicate fillers, unirradiated and irradiated at 100 kGy.

### 3.5. Abrasion resistance

Figure 9 demonstrates the abrasion loss for SBR/DWR 50/50 composites that unirradiated and irradiated at 100 kGy, also loaded with various silicate fillers with fixed concentration namely, 40 phr. It can be observed that, for unirradiated and irradiated SBR/DWR 50/50 composites, the abrasion resistance improved as follows; PS-MS 20/20 > PS, PS-FA 20/20 > PS-WG 20/20 > blank. These achieved
results for abrasion confirmed by data discussed before in mechanical properties and cross-link density. Depending on these results of abrasion resistance, the SBR/DWR 50/50 composites loaded with silicate type PS-MS 20/20 were applicable in flooring tiles.

![Figure 9: Abrasion loss for SBR/DWR 50/50 composites loaded with 40 phr of different silicate fillers, unirradiated and irradiated at 100 kGy.](image)

The properties of obtained materials have been compared with commercially available floor tiles, as presented in Table 3. It can be seen that, SBR/DWR 50/50 composites loaded with silicate type PS-MS 20/20 at irradiated at 100 kGy were applicable in flooring tiles, the composites gave good mechanical properties when compared with others of commercially available.
Table 3: Comparative properties of obtained applicable rubber flooring tiles with commercially available in market

| properties                  | Waste natural rubber | Coarse-grained colored EPDM granules bonded with Polyurethane elastomer. | SBR/DWR 50/50 loaded by PS-MS 20/20 |
|-----------------------------|----------------------|------------------------------------------------------------------------|-----------------------------------|
| Tensile Strength, MPa       | 3                    | 0.3                                                                    | 3.25                              |
| Elongation, %               | 200                  | 40                                                                     | 900                               |
| Hardness, Shore A           | 65±5                 | 60 ± 5                                                                 | 55±5                              |
| Thickness                   | 3-8mm                | 4 mm (± 0.3 mm)                                                        | 4 mm                              |
| Compression set, %          |                      | 15                                                                     | 40                                |
| Supplier                    | QINGDAO BOTHWIN CO., LTD., China | Kroiburg Relastec GmbH&Co.KG. Germany                                 | Our study                         |

Conclusion

Irradiated composites loaded by PS-MS 20/20 provided the highest values for TS, followed by PS, PS-FA 20/20, and finally PS-GW 20/20, meanwhile unirradiated composites provided lower values than irradiated composites for the same type of silicate filler. In general, for unirradiated composites the $E_b$ decreases with the addition of silicate fillers, owing to the filler restrict stretching. $E_b$ increases by radiation for all silicate fillers due to cross-linking not sufficient to retard elongation. For unirradiated and irradiated composites the $M_{100}$ and $M_{300}$ increase with all types of silicate fillers, and PS-MS 20/20 filler provided the highest values for modulus, meanwhile PS, PS-MS 20/20 silicate fillers presented comparable values. Unirradiated and irradiated composites but a loaded composite provided comparable value for CD for all types of silicate fillers, but MS remains has the highest value for CD. SBR/DWR 50/50 composites loaded by PS-MS 20/20 provided the lowest values for the compression set. The composites loaded by PS, PS-FA 20/20, and PS-WG 20/20 presented comparable values for hardness and these for not irradiated composites and irradiated at 100 kGy. The abrasion
resistance enhanced as follows; PS-MS 20/20> PS, PS-FA 20/20> PS-WG 20/20 > blank.

**Ethical approval**
Not applicable

**Consent to participate**
Not applicable.

**Consent to publish**
Not applicable. All authors approved for publication.

**Author's contributions**

*Khaled F. El-Nemr* was responsible for the conception and design, testing, data acquisition, writing - review, and editing. *Magdy A. Ali* was responsible for analysis and data interpretation, writing - review, and editing manuscript. All authors read, revise, and approved the final manuscript. *Yasser H.Gad* was responsible for analysis and data interpretation, writing - review, and editing manuscript. All authors read, revise, and approved the final manuscript.

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**Availability of data and materials**
The authors declare that all data supporting the findings and materials of this study are available within the paper

**Conflict of interest**
The authors declare that they have no competing interests and non-financial competing interests.

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**Declarations**
The authors declare no competing interests.

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