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The effects of a silane coupling agent on properties of rice husk-filled maleic acid anhydride compatibilized natural rubber/low-density polyethylene blend

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Abstract Blends of natural rubber and low-density polyethylene were prepared in different weight compositions in presence of dicumyl peroxide and maleic acid anhydride. The effects of rice husk (RH) content and a silane coupling agent, that is, 3-aminopropyl triethoxy silane (3-APE, 1 wt% of filler content) on the physicomechanical properties and mass swell of the tested blend were investigated. The incorporation of untreated RH into the blend improved Young's modulus, hardness but decreased tensile strength, elongation at break, impact strength, and mass swell. Incorporation of 3-APE has produced composite with improved tensile strength, Young's modulus, hardness and impact strength with a sharp decrease in elongation, and better mass swell in comparison with untreated one. The effect of γ-irradiation doses on retained physicomechanical properties of RH (30 phr) filled blend samples before and after silane treatment was investigated at ambient temperature. The efficiency of silanized RH (30 phr) was also evaluated by the studies of the surface morphology (scanning electron microscopy) and thermal properties (thermal gravimetric analysis and thermal mechanical analysis).

Introduction

Thermoplastic elstomers (TPEs) is probably the fastest growing sector of polymer market [1]. They are obtained by blending a thermoplastic resin with an elastomer and are materials having the processing characteristics of thermoplastics and the technical properties of vulcanized elastomers [2]. Among the different types of TPEs, those prepared by the physical blending of a polyolefin and an elastomer have gained considerable attention due to the simple method of preparation and easy attainment of the required technical properties [2]. The study on thermoplastic elastomers with various types of elastomers and polyolefins has been reported by many researchers [3–6].

Of the various types of additives used to achieve different properties, fillers are added to TPEs either to improve properties or to further reduce the cost of the material or to achieve both objectives simultaneously [7, 8]. Coupled with the growing concern for disposal of agricultural and industrial waste and low-cost fillers, considerable studies have been carried out on the utilization of some of these materials as fillers with elastomers and plastics as well as with thermoplastic elastomers. Consequently, these environmentally problematic waste materials are converted to useful industrial materials. Recently the utilization of fillers (bio-fibers or powders) derived from agricultural sources such as banana, sisal, hemp, jute, pineapple, rubber wood powder, waste rubber particles, palm oil fruit bunch, bamboo, bagasse, wood chips flax, and rice husk (RH) has become a subject of interest in polymer composites and has become a strong competitor to inorganic fillers [9–12]. This mainly due to their low densities, very low cost, non-abrasiveness, high filling levels, recyclability, biodegradability, and renewable nature [13].

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RH is one of such major agro-waste products, which contains cellulose 32.7%, hemicellulose 20.5%, lignin 21.8%, silica 15.1%, solubles 2.8%, and moisture 7.5% [14]. These average values can change for different rice varieties. RH is a by-product of rice-milling process in rice-producing countries, especially in the Asian, Pacific, and United States regions. Most of this RH is used as a bedding material for animals; the rest is burned or used for landfilling. The industrial applications of this material are limited. Fortunately efforts are already underway for producing value-added products [15–17], such as composite materials, from this important bio-resource. The use of RH in making composite products is attracting much attention because of the potential for enormous gains in certain important properties of these products [9, 18]. These properties were further enhancing with the use of a coupling agent, which improved the interfacial adhesion between the polar hydrophilic RH and the non-polar hydrophobic plastic polymeric chains [11, 19, 20].

The aim of this study is to chemically modify RH surface, using a silane coupling agent, to improve its suitability as a reinforcing material. Besides the importance of property improvement, an additional incentive was responding to the social demand for the disposal of environmentally problematic agricultural waste.

Experimental

Materials

Natural rubber (NR) and low-density polyethylene (LDPE) were the blend components used in this study. NR was of the type (SMR-20) obtained from Transport and Engineering Co., Alexandria, Egypt, with a density of 0.913 g/cm^3 at 23 °C and Mooney viscosity ML $(1 + 4)$ at 100 °C $= 60$–90. LDPE was a heavy-duty grade, supplied by Dow Co., Midland, MI, USA, has a density of 0.919 g/cm^3, a melting temperature of 107 °C, and melt flow index of 0.26 g/10 min (190 °C, 2.16 kg). Dicumyl peroxide (DCP) and maleic acid anhydride (MAH) were used as a curing agent and a compatibilizer, respectively. DCP and MAH were pure grade, purchased from Sigma-Aldrich Co., Bellefonte, PA, USA, with melting points 39–41 and 54–56 °C, number-average molecular weights of 270.37 and 98.06 gmol^{-1}, respectively. RH was used as a filler and obtained from a local rice-milling plant. 3-aminopropyl triethoxy silane (3-APE) silane was used as a coupling agent, designated GS and supplied by Sigma-Aldrich Co. Esso extra motor oil and lockheed super 105 hydraulic brake fluid were used as swelling liquids.

Methods

Melt processing

NR/LDPE blends in various weight compositions (i.e., 100/0, 75/25, 50/50, 25/75, and 0/100) were prepared by melt mixing of the components in a Brabender Plasticorder (C.W.Bra. Instrument, Inc., 50 Hackensack, NJ, USA, 230 V, 40 A). At the start, LDPE was introduced into the Brabender, preheated to 155 °C for 5 min, and NR was added thereafter. The mixer was operated at 155 °C, maintaining the rotor speed at 60 rpm for 10 min. The molten mix was quickly removed from the Brabender and mixed with fixed contents of DCP and MAH in two roll-mill (outside D 470 mm and working distance 300 mm, speed of slow roll 24 rpm and fraction ratio of 1.4:1). The general formulations used in this study are given in Table 1. The formulated blend samples (25/75) were mixed with untreated RH in variable contents (viz., 10, 20, 30, 40, 50, and 60 phr) as well as with treated RH with 3-APE silane (1 wt% of RH filler) in two roll-mill. The samples were taken out and subsequently compressed in an electric-heated hydraulic press at about 150 °C for 7 min under 7 kN into thin sheets of about 2 mm thickness from which test specimens were prepared.

Filler treatment

The silane coupling agent, 3-APE, was diluted in ethanol in order to make a 20% solution. The filler was charged into a mixer and the silane solution (1 wt% of RH filler) was added slowly to ensure uniform distribution of the coupling agent. After completion of the silane addition, the filler was continuously mixed for another 30 min. The treated filler was then dried at 100 °C for about 5 h to allow complete evaporation of the ethanol.

Irradiation aging process

The RH (30 phr) filled NR/LDPE (25/75) samples before and after treatment with silane (1 wt% of RH filler) were irradiated at ambient temperature with γ-irradiation cell

Table 1 Formulations of NR/LDPE blends

Materials	1	2	3	4	5
NR	100	75	50	25	0
LDPE	0	25	50	75	100
DCP (phr)[a]	4	4	4	4	4
MAH (phr)[b]	4	4	4	4	4

[a] Dicumyl peroxide

[b] Maleic acid anhydride

(220 V) at different irradiation doses (viz., 200, 400, 600, and 800 kGy) and a dose rate of 1.4 Gy/s.

Techniques

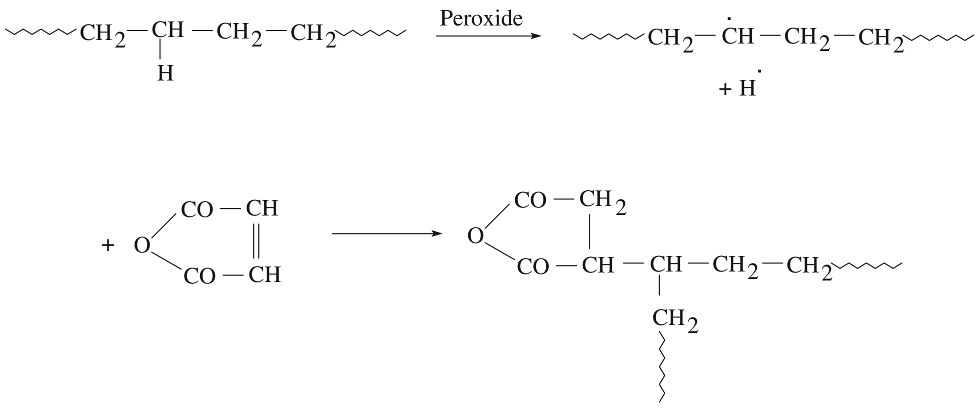
The tensile strength, elongation at break, and Young's modulus were determined with a Zwick 1425 testing machine, according to ASTM-D412-98a, 1998. The hardness of test specimens was measured with a shore A durometer according to ASTM-D2240-97, 1998. Impact strength was measured with a digital universal Froctoscope (Ceast) 6546/000 according to ASTM D256. Mass swell of the test pieces was carried out in the used oil according to ASTM D471-98, 1998. All these tests were performed at room temperature (25 ± 1 °C), and the reported results were averaged from a minimum of five specimens. The surface topography of the samples was analyzed by scanning electron microscopy (SEM). The samples were sputter coated with gold and examined in SEM-5600 (JEOL Co., Japan). The thermal studies were made with thermogravimetric analysis (TGA) and thermal mechanical analysis (TMA). TGA was conducted with a Shimadzo-TGA-50 system (Shimadzo, Japan). The studies were done in a nitrogen atmosphere at a flow rate of 10 mL/min and at a heating rate of 10 °C/min from 0 to 600 °C. TMA was performed with a Shimadzo-50 under air at a heating rate of 10 °C/min. The weight of the samples was 5 mg in all cases.

Results and discussion

NR/LDPE blends without RH

In a previous work [21], NR was blended with LLDPE by the ratio of 50:50 in the Brabender Premixer at 155 °C and rotor speed 60 rpm for 10 min, as the optimum conditions for the preparation of NR/LLDPE blends, which gave the most promising mechanical properties.

In this study, different blend ratios of NR and LDPE were prepared under the above-mentioned conditions and formulated with DCP and MAH as listed in Table 1. Physicomechanical properties (tensile strength σ_R, elongation at break ε_R, Young's modulus E, and hardness shore A) of the pure polymers and of the prepared blends were determined and the results are given in Table 2. It was noticed that the properties were improved with the increase in LDPE content. This could be attributed to the fact that DCP can crosslink both NR and LDPE phases. It has been also noticed that the mechanical properties of 25:75 blend ratio were higher than that for 50:50 blend ratio. The only way to explain this behavior may be attributed to the ability of DCP to crosslink LDPE more than NR [22]. It is believed that the DCP initiates radical formation on the LDPE backbone by hydrogen abstraction and chain scission. The radicals then react with MAH to from maleated polyethylene as follows:



Maleated LDPE

This indicated that MAH reacts with LDPE and modifies it, to be ready homogenized with NR. Thus, the data obtained revealed that the improvement of the physicomechanical properties NR/LDPE (25/75) blend samples was due to the enhancement of the adhesion between the two phases, suggesting the compatibility of the components [23].

Also, the effect of the blend compositions on mass swell percentages of NR/LDPE blends in the tested oil for 70 h was studied and listed in Table 2. It could be seen that the mass swell value decreased with increasing LDPE content in the composition. These results were in accordance with σ_R, E, and hardness values (see Table 2).

Table 2 Physicomechanical properties and mass swell of NR/LDPE blends without RH

Blend ratio		Physicomechanical properties				
NR	LDPE	σ_R (N/mm^2)	ε_R (%)	E (N/mm^2)	Hardness (shore A)	Mass swell (%)
100	0	12	400	140	48	45
75	25	15	500	180	55	40
50	50	19	650	250	65	37
25	75	25	800	400	83	30
0	100	18	700	320	88	35

σ_R tensile strength, ε_R elongation at break, E Young's modulus

NR/LDPE blend with RH

Figure 1a–f summarizes the effects of both filler content (viz., 10, 20, 30, 40, 50, 60 phr) and coupling agent (1 wt% of RH filler) on the physicomechanical properties as well as mass swell of NR/LDPE (25/75) blend.

The effects of filler content on physicomechanical properties and mass swell

Figure 1a shows the effect of RH content on tensile strength, σ_R, of blend samples. The effect was drastic at the 10 phr, a further addition of RH resulted in aproportionate decrease in σ_R. Thus, an increase in RH content (beyond 10 phr) would eventually reach a level whereby almost all filler particles or aggregates are no longer adequately separated or wetted by the matrix phases. Agglomeration of the RH and dewetting of the polymer at the interface aggravated the situation by creating stress-concentration points, which accounted for the weakness in the composite and the reduction of σ_R [24]. Other researchers [9, 25–27] observed a similar trend in other lignocellulosic-filled composites. For irregularly-shaped fillers such as RH, the strength of the composites frequently decreases due to the inability of the fillers to support stress transfers from the polymer matrix [28, 29]. Manson and Sperling [30] and Nielsen [31] reported that in the particulate-filled composites, due to poor stress transfer at the filler-polymer interface, discontinuity is created, which generates weak structure.

The effect of filler content on elongation at break, ε_R, of blend samples is shown in Fig. 1b. It could be seen that the incorporation of RH into the samples has resulted in a reduction of ε_R. This may be attributed to the decreased molecular mobility or deformability of a rigid interface between the filler and the matrix material. With increasing filler content ε_R showed a significant decrease. Since, at higher RH content, the domination of filler–matrix interaction can be expected to diminish and is being replaced by filler–filler interaction [9, 19]. This observation was also found for other lignocellulosic materials [32].

The effect of RH content on Young's modulus is shown in Fig. 1c. The addition of RH into blend samples was expected to increase the modulus resulting from the inclusion of rigid filler particles in the soft matrix. The increase in modulus with RH content clearly indicated that the addition of RH increased rigidity of blend samples through the introduction of restrictions in the mobility of the polymer molecules.

A similar observation can be seen in Fig. 1d for hardness test results. The hardness of a composite, which depends on the distribution of filler particles in the matrix, was increased with increasing filler contents. Incorporation of filler particles into the soft matrix reduced the elasticity or flexibility of polymer chains to a more rigid composite.

Impact strength is an indication of the tolerability for sudden impact when the composite is subjected to an impact and rapid crack propagation through the material. The crack propagation is usually initiated through RH particles in the filled composite. The filler is capable of absorbing the energy to stop the crack propagation if the filler–matrix interaction is strong enough [24].

The result, in Fig. 1e, shows that the impact strength of composites decreases with the addition of RH into the matrices and, therefore, indicates that the RH–matrix interaction is weak.

The effect of RH content on mass swell of blend samples in the tested oil is illustrated in Fig. 1f. It was found that the property decreased with increasing filler content. As the RH content increases within the blend samples, the blends will become stiffer, as shown in Fig. 1c and d, and less penetrable by the oil.

The effects of a coupling agent on physicomechanical properties and mass swell

In this study, as for other lignocellulosic filler/thermoplastic/rubber matrix combinations, the incompatibility of the hydrophilic RH and the hydrophobic matrix can actually lead to a reduction in the strength of the composite material as the volume fraction of RH increases, as we have seen before. Thus, although economical and environmental advantages can be achieved, application of these combinations is dramatically limited. Therefore, it is sensible to modify the surface of the RH in order to improve ease of adhesion with and dispersion within the matrix in order to realize the great potential of RH/NR/LDPE combination.

Considering different composite systems, one of the most successful and cost effective treatments includes the use of silane-based coupling agents for improving the adhesion between RH and polymer matrices.

Thus, throughout this study, the surface of RH was modified using an organofunctional silane (i.e., 3-APE). The importance of the treatment with a coupling agent could be assessed by comparing the results of treated and untreated composites.

Figure 1a, c, d, and e shows the effects of a coupling agent, 3-APE (1 wt% of filler content) on the variation of σ_R, E, hardness, and impact strength of RH-filled NR/LDPE (25/75) blend samples.

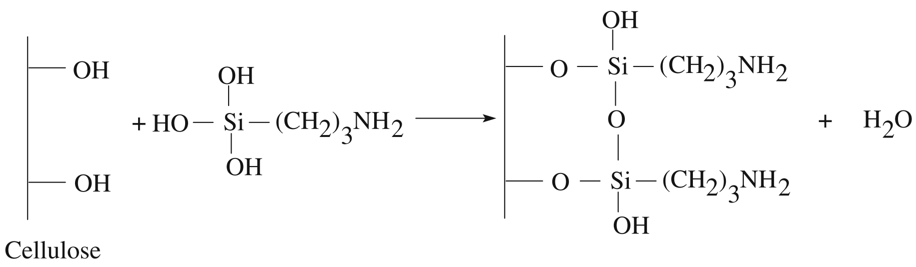
Figure 1a indicated that composites containing treated RH possessed a greater σ_R compared to untreated ones and also composites with silane-treated RH (30 phr) showed maximum σ_R. The increase in σ_R of the silanized composite was most likely caused by improved adhesion and effective wetting of the filler by the matrix [33]. The chemical bonding at the lignocellulosic filler matrix interfaces is critically important for understanding the interfacial strength in reinforced polymer composites.

In general, the reaction of silane with lignocellulosic materials could be described as follows [19, 20, 34–36].

Firstly, the silane used in this study was hydrolyzed in ethanol/water solutions to produce silanol and an alcohol [37, 38].

$$H_2N(CH_2)_3{-}Si(OCH_2CH_3)_3 \xrightarrow{\text{hydrolysis}} \underset{\text{Alkoxysilanol}}{H_2N(CH_2)_3Si(OH)_3} + \underset{\text{Ethanol}}{3CH_3CH_2OH}$$

Secondly, the hydrolyzed alkoxysilanol could be absorbed into the surface of the cellulosic components. Then, there is a possibility of a direct condensation reaction between the silanol group (Si–OH) and the hydroxyl group on the cellulosic component surface. This reaction would create a covalent bond through an ether linkage between the filler surface and the silanol group with the removal of water. The remaining silanol groups were capable of hydrogen bonding or condensing with adjacent silanol groups forming –Si–O–Si– network.



Finally, the hydrophobic part of the silane on the cellulosic filler surface could chemically bound through a covalent bond or interact through a hydrogen bond. Here the possibility of hydrogen bonding was greater due to the presence of nitrogen in amino group. Therefore, hydrophilic (RH) and hydrophobic materials (NR/LDPE) could be coupled together with the silane coupling agent (3-APE) acting as a bridge between them. Thus, improved adhesion between RH and NR/LDPE blends make it possible for stress transfer to occur from matrix to RH during loading, thereby improving the strength of the composite [19, 20, 34].

The variation of Young's modulus, E, with chemical treatment is presented in Fig. 1c. It was clear that the modulus of the chemically treated composites exhibited higher modulus values than untreated composites. This was due to the presence of a strong interface between the treated RH and matrix. Again, the load transfer between the RH and matrix occurred through the strong RH/matrix interface. Thus, the E value of a well-bonded composite is raised.

Figure 1d shows the variation of hardness with chemical treatment. It could be seen that the hardness of treated composites was higher than that of untreated ones. This could be attributed to a strong interface between the treated RH and matrix.

The effect of 3-APE silane on impact strength is shown in Fig. 1e. The data indicated that impact strength for treated composites was higher than that for untreated ones. This was due to improved adhesion between the treated RH and matrix. Thus, the silane coupling agent in the composite helps to reduce the crack propagation through well dispersal of the absorption energy.

The effects of 3-APE silane on elongation at break, ε_R, are shown in Fig. 1b. The results indicated that ε_R for treated composites was lower than that for untreated ones.

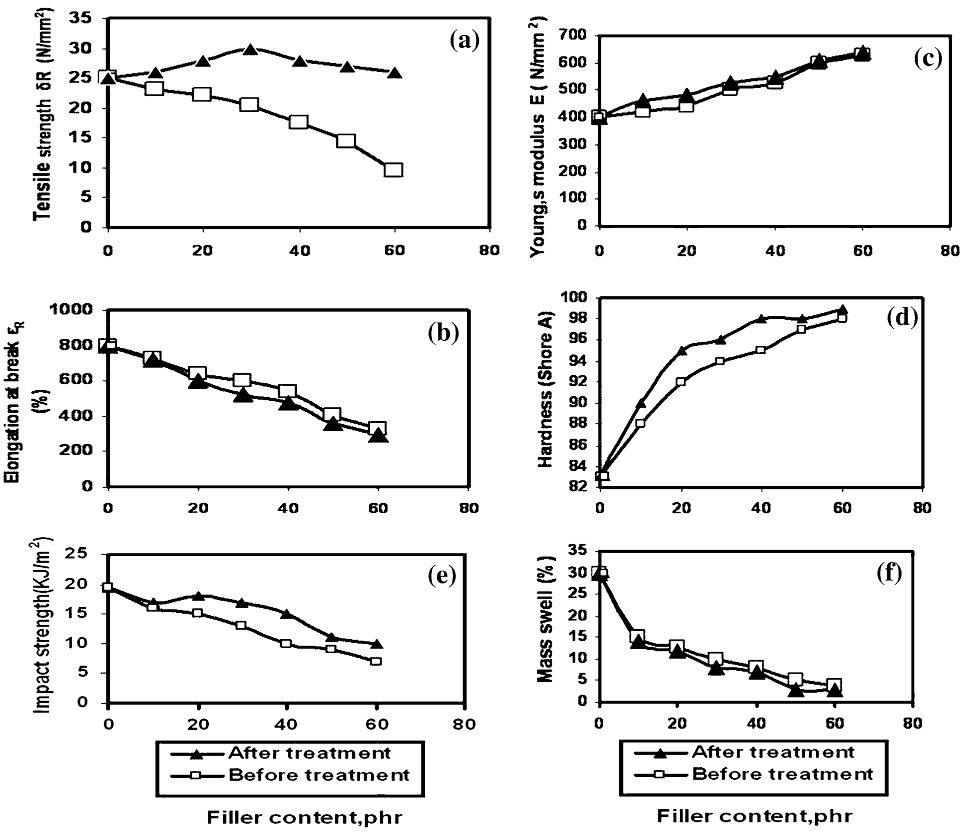


Fig. 1 Physicomechanical properties and mass swell of RH-filled NR/LDPE (25/75) blend before and after silane chemical treatment versus RH content **a** tensile strength (N/mm^2), **b** elongation at break (%), **c** Young's modulus (N/mm^2), **d** hardness (shore A), **e** impact strength (kJ/m^2), **f** mass swell (%)

This means that ε_R of the composites decreased, with presence of 3-APE. This could be attributed to better strength and stiffness achieved from strong adhesion between treated RH and matrix. Higher extension was obtained from weak interfacial adhesion as seen for untreated composition. This indicated that improvement in the filler–matrix adhesion by the use of 3-APE silane improved σ_R, E, hardness and impact strength but at the expense of ε_R. Other researchers also reported the similar observation [34, 39].

The effects of 3-APE silane on mass swell are shown in Fig. 1f. It was clear that at every hour the mass swell of RH filled blend samples treated with 3-APE was less than that of untreated ones. The better mass swell of treated RH samples may be attributed to the ability of the silane chemical to form a protective layer at the interface, which prevents the diffusion of oil molecules into RH.

The effect of irradiation aging

Figure 2a–c illustrates the effect of γ-irradiation aging on retained physicomechanical properties (viz., σ_R, E, and hardness) of RH (30 phr) filled NR/LDPE (25/75) blend samples before and after treatment with silane. By using different irradiation doses (i.e., 200, 400, 600, and 800 kGy) at a dose rate of 1.4 Gy/s, it could be concluded that all the above-mentioned retained properties of the tested samples decreased as the irradiation dose increased. Samples containing treated RH possessed greater retained properties compared to untreated ones (e.g., at 200 kGy there was an increase of 95, 94, and 96% of retained σ_R, E, and hardness, respectively, compared to those of 83, 85, and 88% of untreated ones). This observation was also found upon using 400, 600, and 800 kGy as listed in Fig. 2a–c. These results also confirmed the strong adhesion between the treated RH and the thermoplastic–rubber matrix.

Morphological study

The fractured surface of RH (30 phr) filled blend samples before and after silane treatment was observed by SEM in order to study the effects of treatment on the morphology. The fracture surface of the blend without RH possessed a fine morphology (Fig. 3a). The small domain size was an evidence of adhesion between the matrix (LDPE) and dispersed phase (NR), confirmed the compatibility of the two components with MAH compatibilizer. On the other hand, dewetting of filler by matrix as well as the formation of filler agglomeration could be seen clearly in Fig. 3b. The polar nature of RH and non-polar nature of the matrix could be cited as one of the possible causes for this

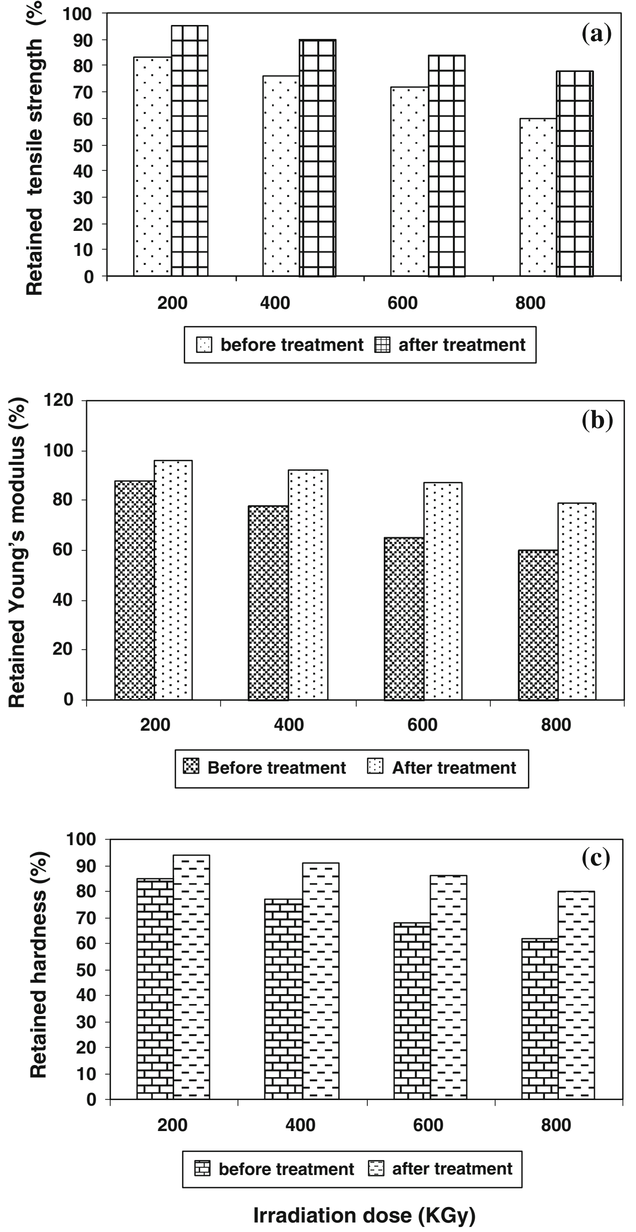


Fig. 2 Retained physicomechanical properties of RH (30 phr) filled NR/LDPE (25/75) blend before and after silane chemical treatment versus irradiation dose **a** retained tensile strength, **b** retained Young's modulus, and **c** retained hardness

observation. This supported our previous explanation that the interfacial interaction between filler and matrix was poor. Therefore, SEM observation of RH composites provides visual evidence for the deterioration of composites properties such as σ_R, ε_R and impact strength as we have seen before.

A better filler dispersion with smaller size of agglomeration could be seen in Fig. 3c. This may be due to the ability of 3-APE to reduce the filler–filler interaction through the reduction of surface energy and consequently resulted in better filler dispersion. The filler particles were coated and well bonded by the NR/LDPE matrix, which indicated better wetting and interaction at the filler–matrix

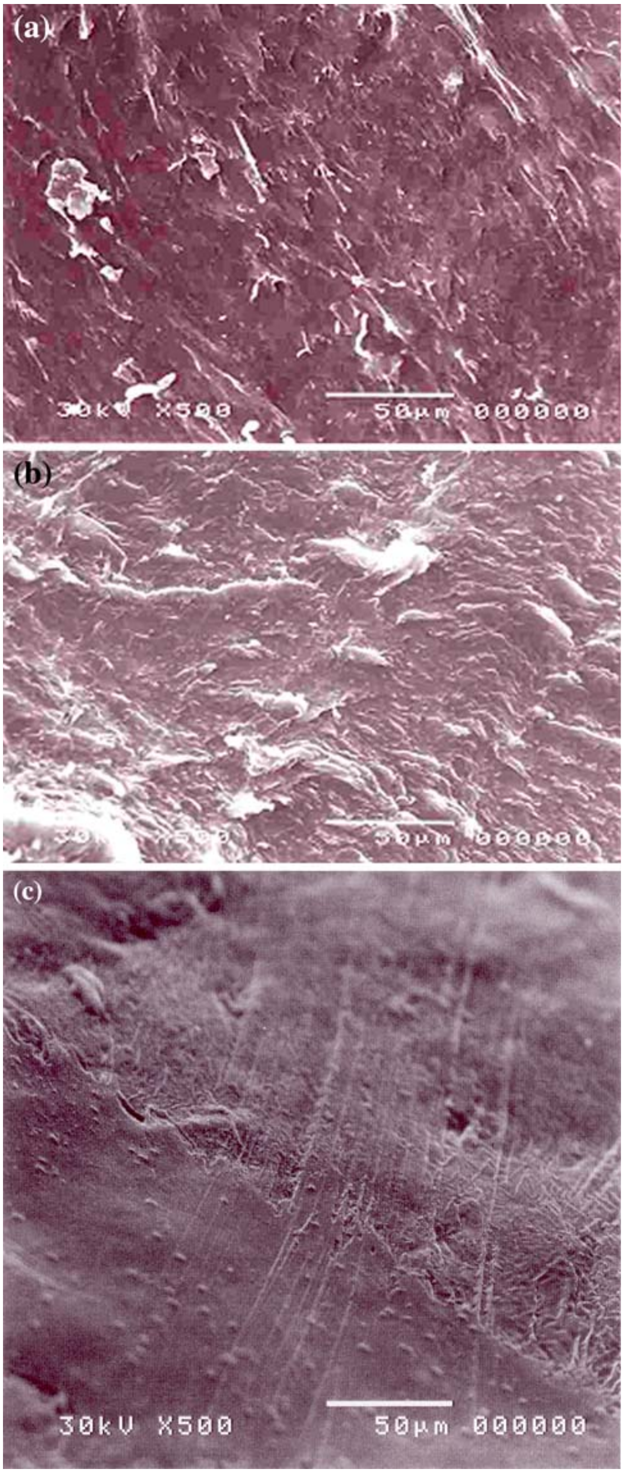


Fig. 3 SEM of different composition surfaces: **a** NR/LDPE (25/75) blend sample without RH, **b** blend sample with RH (30 phr) before silane chemical treatment, and **c** blend sample with RH (30 phr) after silane chemical treatment

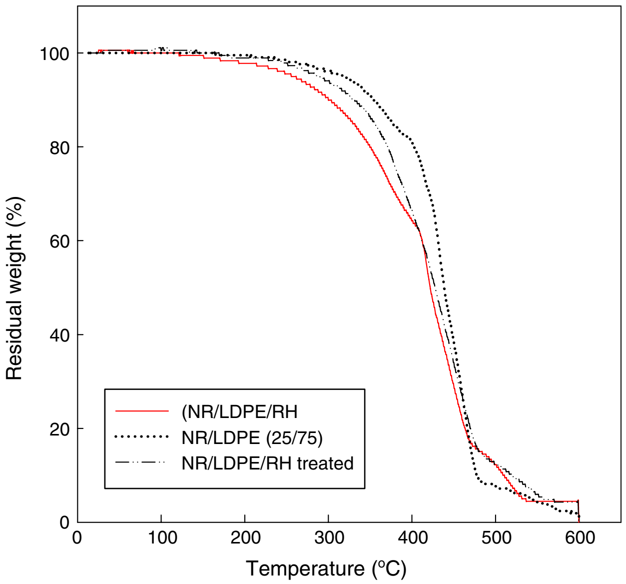


Fig. 4 TGA thermograms of NR/LDPE (25/75) blend sample without RH, and with RH (30 phr) before and after silane chemical treatment

Table 3 Thermal stability of NR/LDPE (25/75) blend samples without RH, and with RH (30 phr) before and after silane chemical treatment

Sample	Weight-loss temperatures (°C)					Residual (%)
	T_i	T_{25}	T_{50}	T_{75}	T_f	
NR/LDPE (25/75)	175	412	438	462	483	8.6
NR/LDPE/RH	121	365	421	455	471	16.7
NR/LDPE/RH treated	169	381	427	462	479	19.4

interface. This observation was consistent with the mechanical properties exhibited by silanized blends.

Thermal analyses

The TGA results of RH (30 phr) filled blend samples before and after silane treatment are shown in Fig. 4 and summarized in Table 3. From the TG curves of the samples, thermal stability is given by initial decomposition temperature (T_i), corresponding to ~5 wt% loss, after that degradation occurs rapidly (T_{25}, T_{50}, and T_{75}) the temperature at which 25, 50, and 75 wt% loss occurs, respectively, as well as final decomposition temperature (T_f) corresponding to the temperature of maximum weight loss, as listed in Table 3. It has been observed that T_i and T_f for RH-filled blends occured at lower temperature compared to those of blend samples without RH. The same trend was also observed for T_{25}, T_{50}, and T_{75}. This was a logical consequence of the lower thermal stability of the RH [17, 40].

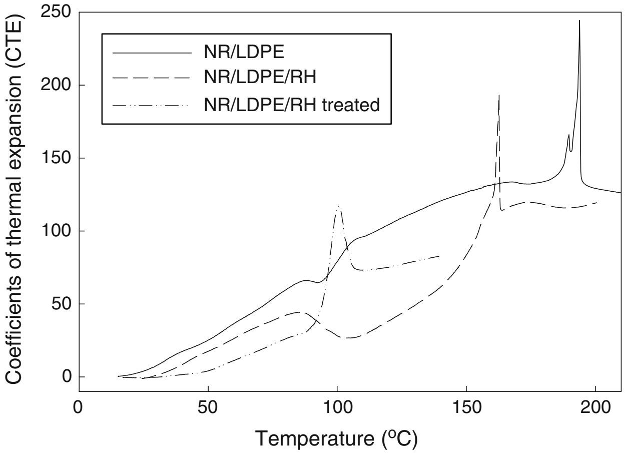


Fig. 5 TMA runs of NR/LDPE (25/75) blend sample without RH, and with RH (30 phr) before and after silane chemical treatment

It was also found that T_f for treated samples occurred at higher temperature with higher residual wt% compared with that of untreated ones. The same trend was also observed for T_i, T_{25}, T_{50}, and T_{75} as observed in Table 3. Thus, the treated samples displayed higher thermal resistance with higher residual wt% than untreated ones. This could be attributed to the fact that the development of organic functional groups on RH surfaces treated by coupling agent led to an increase in the crosslink density of the composites, resulting in enhancing final thermal stability [41].

The TMA thermograms from 23 to 200 °C for RH (30 phr) filled blend samples before and after silane treatment are shown in Fig. 5. It could be seen that incorporation of RH (30 phr) as a lignocellulosic filler into the tested blend reduced its coefficient of thermal expansion (CTE) compared to blend samples without RH. Other researchers [42, 43] observed a similar behavior in inorganic particulate-filled composites. A further decrease in CTE was observed upon using RH treated with (3-APE) silane at the least turning point (i.e., the transition point from expansion to contraction is at 100 °C). This suggested a strong adhesion between treated filler and matrix. This observation was in conformity with the physicomechanical measurements exhibited by the treated samples (i.e., an increase in σ_R, E, hardness, and impact strength values with a decrease in ε_R) compared to untreated ones.

Conclusions

From the above study it could be concluded that:

- The incorporation of RH in MAH compatibilized NR/LDPE (25/75) blends increased Young's modulus, hardness but decreased the tensile strength, elongation at break, impact strength, and mass swell.

- The RH-filled blends treated with 3-APE silane (1 wt% of RH content) showed better tensile strength, Young's modulus, hardness, and impact strength with a drop in elasticity or flexibility of the soft matrix, as well as improved mass swell compared to untreated samples.
- Silane treatment increased the resistance toward γ-irradiation compared to untreated samples.
- SEM micrographs of tensile fracture surfaces showed that the addition of 3-APE enhanced RH dispersion (30 phr) in matrix phase and facilitated the direct contact between RH and the matrix more than untreated samples did.
- The thermal stability of RH (30 phr) filled blend samples was lower than that of samples without RH. The samples treated by 3-APE silane showed superior crosslink density and thermal stability compared to untreated ones.
- Silane treatment reduced CTE of the tested samples compared to untreated ones.
- Silane treatment of the RH increased the RH–matrix interaction through a condensation reaction between hydrolyzed silane and hydroxyl groups of RH.
- On the basis of this, it could be concluded that silane coupling agents of RH can be utilized to develop more selective and effective fillers for different matrices.
- The validity of the use of these composites means the increase in the added value for RH, which is currently considered waste products and no industrial re-use possibilities.

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