Processing and Characterization of Surface Treated Chicken Eggshell and Calcium Carbonate Particles Filled High-Density Polyethylene Composites

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The use of the biowaste chicken eggshell (ES) particles as an alternative filler for the inorganic calcium carbonate (CC) in polymeric materials was investigated in this study. ES and CC were chemically surface treated with stearic acid by dry and wet methods. Composites of ES and CC filled high-density polyethylene (HDPE) was prepared via melt mixing and their thermal-mechanical- and flow behavior was characterized. FTIR results showed some changes in the chemical structure of SA treated ES and CC. The mechanical properties of the composites fluctuated in their values based on the SA treatment method. The wet-treated HDPE/ES composites showed higher tensile and flexural strengths compared to that of the untreated ones which were also higher than their counterparts of HDPE/CC composites. The treated-ES particles motivated the crystallization process of HDPE compared to the untreated-ES and better flow properties was reported for composites with higher crystallinity and for HDPE/CC composites. Morphological observations showed particles agglomeration and void existence which led to deterioration of the mechanical properties.

Keywords: polymer composites, surface modification, chemical analysis, thermal analysis, mechanical testing, microscopy.

1. Introduction

The use of inorganic calcium carbonate (CC) as a filler is common in the production of polymer products. In recent years, there was also an interest in utilizing chicken eggshell (ES) as a bio-filler in polymeric materials and this was initially pronounced by Toro et al. The high bio-calcium carbonate percentage of 95% in the ES composition revealed to the researchers to use it as a filler in polymeric materials products instead of using the common inorganic CC filler. A recent review conducted by Owuamanam and Cree to follow the progress of using the bio-filler ES waste in polymer composites has emphasized the importance of its use in the production of new thermoset and thermoplastic polymer composites. The polymer/ES similar to polymer/CC composites have been produced by different techniques such as: injection molding, extrusion/compression molding, film-casting, solution mixing, and hand-layup technique. In general, the use of such fillers either CC or ES has its advantages and disadvantages. The main advantage of using these fillers in compositing the polymeric materials is to reduce the cost by replacing a weight percentage of the relatively expensive polymeric materials with a cheaper filler material and filling load up to 40 wt.% with ES or CC was reported in the literature. Moreover, these composites are more environmentally friendly. On the other hand, the degradation in the mechanical properties of neat polymeric materials when incorporating ES or CC was an important issue to be taken into consideration by the researchers. The values of tensile, flexural, and impact strengths of these composites might be improved only at low filler loading, while their stiffness could be significantly improved with the increase in the filler content. Teixeira et al. showed that the addition of 2.5-4 µm untreated CC particles to HDPE provoked a significant decrease in its impact strength at 5 wt.% CC and a slight decrease in the tensile strength compared to that of the neat HDPE. A comparison carried out by Azman et al. between the mechanical properties of HDPE/untreated ES and HDPE/untreated CC with 15 wt.% filler content showed a similar trend of both types of composites in decreasing the tensile strength of about 23% compared to the pure HDPE. Their flexural strength also decreased but its value was higher for ES composites compared to CC ones which was attributed to higher tendency of agglomeration of the smaller CC particles. Khalaf in his investigation on the mechanical properties of untreated CC filled HDPE observed that the tensile strength increased at 5 wt.% CC by about 19% compared to pure HDPE and its value decreased, thereafter, by about 20% at 20 wt.% compared to that of the matrix. Wahab et al. showed a decrease in the impact strength of HDPE/untreated CC composites at all filling loads. Shuhadah and Supri showed a decrease of 50% in tensile strength of low-density polyethylene composites combined with 20 wt.% of NaOH-treated ES of size 63 µm while the tensile modulus increased by more than 100%. Boronat et al. showed a decrease of 15% for composites of HDPE and 20 wt.% ES of size 25 µm and a slight increase in the tensile and flexural modulus. The researchers attributed the decrease in the mechanical properties to the lack in processing and characterization of surface treated chicken eggshell.
adhesion bonding between the fillers and the matrix as well as the bad dispersion of the fillers. Therefore, previous studies in different directions have been carried out where some researchers have used nano-sized fillers to enhance the mechanical properties of the composites. Due to the matrix hydrophobic nature and the fillers hydrophilic nature, other researchers have implemented different filler surface modification methods using different coupling agents to enhance the compatibility between the filler and the matrix as well as the filler dispersion in the matrix. Hydrogen bonding between the polymer and the filler which may be promoted by amine, carboxylic, and hydroxyl groups could improve the bonding between them. Inexpensive fatty acids such as stearic acid was also extensively used to improve the hydrophopocity of CC. Wet and dry coating methods of CC with a hydrophobic layer of SA was reported. ES was also treated by stearic acid and observations of better dispersion and reduction in ES particles were reported by Shah et al., who also observed that a 22% increment in the elongation of epoxy/ES composites compared to neat epoxy and attributed that to the hydrogen bonding between the treated particles and matrix.

Finally, the motivation is still present among researchers to conduct further work in the processing of the polymer/ES composites and to better understanding their mechanical-thermal behavior as well as their other properties and compare them with that of the conventional mineral calcium carbonate CC composites. In this study, HDPE/ES and HDPE/CC composites were prepared via melt mixing and compression molding technique. The choice of HDPE as a matrix was due to that HDPE was and still one of the most consumed semicrystalline thermoplastics with increased demand market for many engineering applications and it is commonly reinforced with CC. HDPE composites were prepared at a filler concentration of 20 wt.% and it was reported in previous studies that CC content higher than this ratio resulted in highly deterioration in the mechanical properties of the composites with a more brittle-like failure manner. The surface treatment of CC particles with stearic acid which was suggested by the researchers in the literature, using dry and wet methods, were also applied to ES particles in this study.

2. Experimental

2.1. Materials

Chicken eggshell (ES) was collected from the local market in Jordan and it was washed and dried at 80 °C for overnight. The dried ES was then grinded into particles and sieved to obtain a size passed through 65-mesh and its density ranged between 2.5 and 2.6 g/cm³. Laboratory reagent calcium carbonate (CC) with a fine average particle size of less than 1 µm was supplied by SDFCL-sd Fine Chem. Ltd., India. HDPE in granular form was supplied by SABIC, Saudi Arabia, with a density of 0.953 g/cm³. Chemicals also provided for chemical treatment of ES and CC particles such as sodium hydroxide (NaOH) solid which was supplied by Unichem Co., Ltd. (Mumbai, India).

2.2. Chemical surface treatment of ES and CC particles

A chemical surface treatment of ES and CC particles with SA was carried out by three different methods: (i) direct melt treatment method where ES or CC particles were manually dry mixed with SA before melt compounded with HDPE at 170 °C using two-roll mill; (ii) one-suspension method where a suspension of ES or CC, SA, and NaOH mixed with distilled water in a beaker and stirred for 10 min at 75 °C and the mixture was then washed, filtered and oven-dried; (iii) two-suspension method where two separate suspensions (one suspension contained SA dissolved in NaOH at 75 °C and the other suspension contained of ES or CC in distilled water at 75 °C) were mixed and stirred at 75 °C for 15 min and then washed, filtered and oven-dried. Similar procedure for the last two methods was suggested by Cao et al. in their preparation and characterization of HDPE/CC composites.

2.3. Preparation of HDPE/ES and HDPE/CC composites

The composites of HDPE/ES or HDPE/CC were prepared by initially melting the HDPE granules using Brabender two-roll mill (C.W. Brabender Instruments, Inc. (for 2 min at 170 °C and a rotating speed of 50 rpm). The untreated or treated particles of ES or CC were then added to the molten HDPE and mixing process of all components was continued for another 8 min. The resulting compounds were then hot pressed into square mold of 3 mm thickness at a temperature of 170 °C and a pressure of 15 KPa for 10 min. Samples for impact, tensile, and flexural tests were prepared according to different ASTM standards. Eight types of composites were prepared with 80 to 20 weight ratio of HDPE to ES or HDPE to CC and compared with the neat HDPE and they were given the designation shown in Table 1.

2.4. Characterization of composites

A Fourier transform infrared (FTIR) spectrophotometer (model- Bruker Tensor 27 SN_3880, Germany) was utilized to identify the functional groups. KBr is used as a carrier for the sample in IR spectrum scanned from 400 to 4000 cm⁻¹ with a resolution of 4 cm⁻¹. A differential scanning calorimeter (DSC, Perkin Elmer7) was used to investigate the thermal behavior of the composites and it carried out using oxygen purging at a rate of 50 mL/min. Aluminum pans containing 6 to 7 mg of composite material were used and the specimens

| Sample Type                  | Designation | Filler-to-PP weight ratio (wt.%) |
|------------------------------|-------------|----------------------------------|
| HDPE/ES (untreated)          | HDPE/UES    | 20:80                            |
| HDPE/ES (direct melt mixing) | HDPE/TES1    | 20:80                            |
| HDPE/ES (one-suspension)     | HDPE/TES2    | 20:80                            |
| HDPE/ES (two-suspensions)    | HDPE/TES3    | 20:80                            |
| HDPE/CC (untreated)          | HDPE/UCC     | 20:80                            |
| HDPE/CC (direct melt mixing) | HDPE/TCC1    | 20:80                            |
| HDPE/CC (one-suspension)     | HDPE/TCC2    | 20:80                            |
| HDPE/CC (two-suspensions)    | HDPE/TCC3    | 20:80                            |
were heated to 200 °C, and then cooled to room temperature at a heating and cooling rates of 10 °C/min. Tensile and flexural test of the composites were carried out according ASTM D638 and ASTM D790 standard respectively using universal testing machine (HOYTOM, Model DI-922 FL, Spain) at 2 and 1 mm/min using a load-cell with the capacity of 50 kN. Rectangular specimens were prepared of dimensions (165 × 13 × 3.2 mm) mm for tensile test and of dimensions (50.8 × 12.7 × 3.2 mm) for flexural test. The Izod notched impact test was also carried out according to ASTM D265 standard using a Ceast pendulum impact tester and rectangular specimens (64 × 12.7 × 3.2 mm). The melt flow index (MFI) of the composites was determined according to ISO1133 using Ceast melt flow tester measured at 190 °C/2.16 kg. Finally, the fractured surface of the composites and the distribution of particles throughout the matrix were examined by scanning electron microscopy (FEI Quanta 600 FEG-SEM). The specimens were coated with Platinum/Palladium Pt/Pd. The average value of three specimens of each type was considered in the mechanical testing.

3. Results and Discussion

3.1. FTIR results

The results of FTIR analysis for ES, CC, HDPE, HDPE/ES, and HDPE/CC are shown in Figures 1 and 2. The FTIR spectra of the untreated ES (Figure 1a) showed three characteristic peaks located at 1421, 875, and 687 cm⁻¹, which represented the stretch and bending vibrations of CO₃²⁻ ion. Between 3000 and 2500 cm⁻¹, the absorption bands were identified as organic matter (C-H stretching) and an absorption band at 3421 cm⁻¹ was related to the stretching absorption of H₂O. Similar characteristics to ES structure were observed for CC from FTIR spectra shown in Figure 1b at slightly different positions for the peaks. FTIR spectra of SA in Figure 1a and 1b showed adsorption peaks at about 2921 and 2843 cm⁻¹ which are attributed to stretching vibrations of –CH₂ groups. In the low-frequency region, the peak at 1703 cm⁻¹ was attributed to carbonyl (–COOH) stretch and there were different peaks located in the range 500 to 1500 cm⁻¹. In general, after surface treatment the particles with SA, a chemical reaction of SA with ES or with CC and a formation of a hydrophobic monolayer film might be expected and this can be noticed in FTIR analysis usually by the appearance of new absorption peaks or the absence of old ones, a change in their intensities, and a broadening in peaks or shifting in their positions. The surface treatment of ES with SA resulted in the appearance of new characteristic peaks at 1578, 1539, and 1468 cm⁻¹ for one-suspension treated ES and at 1585 and 1539 cm⁻¹ for two-suspension treated ES indicating the reaction of CO₃²⁻ existed on the surface of ES particles with SA. On the other hand, the increase of the intensities of C–H vibration bands between 3000 and 2500 indicated that the surface of ES was coated by SA, so that the hydroxyl number was reduced. The peaks identified as organic matter stretching increased in treated ES which may point out to a chemical reaction of SA with ES particles.

The chemical structure of HDPE and HDPE/ES and HDPE/CC was examined by FTIR as shown in Figure 2. A stretching vibrations of –CH₃ appeared at a frequency range of 2921-2843 cm⁻¹, a bending and a rocking vibration of –CH₃ appeared at 1468 cm⁻¹ and at 703 cm⁻¹ respectively corresponded to the native bonds present in HDPE. The weak absorption
band at 1078 and 875 cm⁻¹ can be assigned to the vinyl groups according to Apone et al. The small absorption bands appeared in the range 1625-2000 cm⁻¹ which related to carbonyl groups appearance. According to the FTIR spectra, all the peaks of the HDPE/ES and HDPE/CC composites are in conformity with those of HDPE. This indicates that the main chemical structure of the composites is HDPE and it is an indication to bad compatibility. Azman et al reported that HDPE composites showed, to some extent, similar spectra in the presence of ES or CC. Basilia et al reported that the changes in the transmittance shown on FTIR of PE, PP, and PVC by the addition of CC particles into them indicating that there was a direct binding between the fillers and matrix.

3.2. DSC and thermal properties

The DSC heating and cooling curves of untreated and treated HDPE/ES and HDPE/CC composites are shown in Figure 3 and the thermal parameters obtained from DSC measurements are tabulated in Table 2. The crystallinity percentage ($X_c$) was calculated according to following equation:

$$X_c = \frac{\Delta H_m}{w_p \times \Delta H_{100}} \times 100\%$$

The $\Delta H_m$ is the melting heat in J/g, $\Delta H_{100}$ is the melting heat for 100% crystalline HDPE which is equal to 293.6 J/g according to Cao et al., and $w_p$ is the weight fraction of HDPE in the sample. As can be observed from Table 1, the melting temperature ($T_m$) decreased by 3 to 7 °C for the composites compared to that of the neat HDPE, while there was a slight increment by up to 2 °C in the recrystallisation temperature ($T_r$) of the composites. The decrease in $T_m$ was also observed by Cao et al. and due to the resulted thinner lamellar crystallites in the composites compared to that of the pure HDPE. Bartczak et al. observed in a previous study an increase in $T_r$ up to 1 °C or lower which was dependent on the particle size and indicated to some effect on nucleation of HDPE crystallites. The significant decrease in the melting heat ($\Delta H_m$) and the degree of crystallinity of the composites compared to that of the neat HDPE indicated a lower nucleating ability of the particles. This was more pronounced in case of HDPE/ES composites. The decrease in $\Delta H_m$ clearly decreased the amount of energy required to break the solid for of the HDPE. It was reported by Lazzeri et al. that untreated CC particles as well the SA treated ones have a small nucleating effect on HDPE which can be observed in this study by the small fluctuation of the crystallinity values of HDPE/CC composites about that of the neat HDPE. The improvement in both $\Delta H_m$ and $X_c$ was clear in treated HDPE/ES composites compared to that of untreated ones induced a higher heterogeneous nucleating and a higher crystallinity of HDPE. The HDPE/ES composites treated by two-suspensions method showed slightly better results than the ones treated by one-suspension method and significantly better results than direct-melting treated composites. Similar trend was observed for HDPE/CC composites regarding to melting heat but a slight increase in the degree crystallinity of treated composites was found compared to that of untreated composites and neat HDPE. Saeedi and Sharahi reported that incorporating CC particles played a role in the arrangement of the chain molecules of HDPE and acting as nucleating agent which makes HDPE crystallize heterogeneously. Other researchers found that the excess or insufficient surfactant of such particles diminished the nucleating ability. Higher $\Delta H_m$ and $X_c$ indicated a better surfactant of ES and CC. The change in the crystallinity was only supported by a slight change in the recrystallization temperature ($T_r$). The increase in $T_r$ meant the acceleration of crystallization during cooling.

3.3. Melt flow index

MFI plays a curial role in controlling the processability of HDPE composites. The melt flow of the roll-milled HDPE and its composites were tested and the results are shown in Figure 4. It was observed that there was a decrease in the MFI by the addition of the 20 wt.% UES to HDPE, while no significant change occurred on MFI by the addition of UCC compared to that of the pure HDPE. Sahai et al. reported that the decrease in MFI is attributed to the agglomeration of the particles which results in restriction of flow for untreated CC filled polyphenylene oxide composites. They also found
that the treated CC composites improved adhesion between the constituents and lowered the agglomeration but the good adhesion also contributed to reduce the MFI at a filler content less than 10wt.%. Jang\(^4\) in his study of polycarbonate (PC) composites explained the reduction of MFI by the large size of talc particles which act as flaws and their tendency to slide due to the flatly shape specially at higher concentrations. In Figure 4, the treated composites showed an improved melt flow properties compared to that of the untreated ones, especially for HDPE/CC composites. Adeosun et al.\(^4\) indicated that CC is capable to improve the processability and the plasticity of the polymer. Apone et al.\(^3\) claimed that chain scissions and chain branching occurred during the degradation of HDPE control the MFI and its value, on average, increases or decreases based on the predominant factor. Ertürk and Şahin\(^4\) found that the addition of CC up to 20 wt% to HDPE has no influence on MFI, while higher filler loadings increased MFI of the composites.

### 3.4. Mechanical properties

The investigation of the mechanical properties of the composites is important where any improvement occurs in them reflects the ability of the matrix to transfer stress to the filler which depends on the adhesion properties, particle size, and filler loading.\(^4\) The tensile, impact, and flexural strength for HDPE and its composites are shown in in Figures 5 and 6. A reduction of about 40% was observed in the value of the tensile strength as HDPE filled by 20wt.% untreated ES. This deterioration in tensile strength was expected and reported by many researchers, e.g. Shuhadah and Supri\(^1\) reported a reduction of 50% in the tensile strength of low density polyethylene (LDPE) filled with 20 wt.% of untreated ES having a particle size of 63 µm. Hayeemasae et al.\(^3\) reported a 44% reduction in the tensile strength of polystyrene (PS)/20wt.% untreated ES composites with ES particle size less than 75 µm and Murugan et al.\(^2\) reported a decrease of 18% for HDPE/22.5 wt.% ES (7 µm particle size). They attributed the decrease in tensile strength to the poor interfacial adhesion between the components as well as the agglomeration of the...
particles. But compared to untreated HDPE/ES composites in this study, a clear improvement of about 25% occurred in the value of tensile strength of treated ES by one- or two-suspension methods (HDPE/TES2 and HDPE/TES3). Cao et al.\textsuperscript{12} found that the wet treated processes was more effective in improving the tensile strength due to the better interfacial adhesion between the filler and the matrix which could be observed from morphological changes shown in section 3.5. The treated HDPE/CC composites showed no improvement in tensile strength compared to untreated ones and some improvement was noticed for two-suspension HDPE/TES3 composites compared to the other treated ones. Mohan et al.\textsuperscript{22} attributed the increase and decrease in the tensile strength of nano-EG filled epoxy composites to the action of the particles or agglomerates as a barrier to the crack propagation resulting in an increased strength and, on the other side, the action of particles or agglomerates as a stress concentration which aid the crack propagation and reduce strength. According to the flexural strength, the treated HDPE/ES composites showed a strength value near to that of neat HDPE and higher than that of untreated ones. The increase in the crystallinity of treated HDPE/ES composites compared to untreated ones may have a positive impact on the increase in their flexural strength values. Melo et al.\textsuperscript{23} in their study of the mechanical properties of tianatified mollusk shell-waste filled HDPE composites pointed out to the enhancement in the flexural strength of the composites by their higher crystallinity compared to the neat HDPE which might overcome the negative impact of the particle agglomeration. The trend of flexural strength for HDPE/CC composites is similar to that of tensile strength except and increase of the strength value was observed for treated HDPE/CC composites by one-suspension compared to other types of treated ones. Regarding to the impact properties of the composites which is shown in Figure 5, a decrease in the impact strength of ~35% and 54% was reported for HDPE/UES and HDPE/UCC respectively. Similar trend was reported by Buakaew\textsuperscript{24} in his investigations of the mechanical properties of HDPE/ES composites who found that the impact strength of these composites at 20 wt.% ES reduced by ~55% compared to the neat HDPE and the presence of particles had negative impact on the ability of composite to absorb energy before rupture. He also found that the smaller the particle size the higher the impact strength due to lower voids content which act as stress concentrator. Bonse and Molina\textsuperscript{25} found that the impact strength of polyamide 6/20wt.% CC composites decreased by ~50% compared to the neat matrix regardless the size and the type of the CC fillers. It was observed from the impact results in Figure 5 that there was no clear improvement in the impact strength appeared for treated composites compared to untreated ones in all types of composites. The impact strength for treated ES composites was almost similar to that of untreated ones as shown in Figure 6. For silane treated ES/HDPE composites compared to that of neat matrix. They attributed the reduction in the ductility by that the fillers usually restrain the movement of polymer chains during tension resulting in a strain localization which leads to a stiffer and a more brittle material.\textsuperscript{34,55} While the further decrease in the ductility of silane treated ES/ HDPE was related to the better adhesion which requires higher stress for small deformation. Shah et al.\textsuperscript{30} reported a reduction of ~25% in the ductility of epoxy/20w.%UES and a rise of ~22% for SA treated ES composites at same filler content and the explained the improvement in the ductility by the lower protein content in ES, and more carbonyl groups in TES and the better dispersion of the particles, as well as the reduction in the crystal size and introducing ore hydrogen bonding. The ductility of HDPE/CC composites reduced further for treated composites compared to that of untreated ones as shown in Figure 6.

3.5. Morphological observations

The dispersion of ES and CC particles and their compatibility with the matrix were investigated from the tensile fracture surface analysis of the specimens and their SEM micrographs are shown in Figures 7 and 8 at different magnifications for each type of composites. The ES particles appeared to have a combination of fine and coarse plate-like and sphere-like irregular morphologies while the CC particles had finer regular sphere-like morphologies. The SEM images of HDPE/UES showed a more flattened shaped particles which were not or partially wetted with the matrix and some agglomerates were found. The bad adhesion evidenced by the clear and smooth surfaces of the particles and their detachment during fracture. The agglomeration of the particles might contribute to the formation of some voids and the porosity appearance led to increase their content on the surfaces. The fracture surface of HDPE/UCC composites appeared with smaller particle size and higher surface area and slightly better embedded particles in the matrix which reflected a higher tensile strength and more ductility compared to HDPE/UES composites, but agglomerates and voids still existed. On the other hand, preparing the composites by directly adding the dry SA and filler particles to the molten HDPE showed no improvement in the particles dispersion and adhesion with higher voids and porosity appearance which resulted in a further deterioration in the mechanical properties compared to untreated composites. Regarding to the wet treated HDPE/ES and HDPE/CC composites, the particles agglomeration and the void existence were also found. The wet treated ES particles were better embedded in the matrix and this was more pronounced in the case of two-suspension treated method. Moreover, the reducing in the treated ES particle size and the more their spherical morphology contributes to improve mechanical properties of the composites. Owuamanma\textsuperscript{26} found that the SA treated ES had a narrower particle size distribution compared to untreated particles and reduced the agglomerate size which might be due to the removal of bioorganic impurities. Better continuity of the matrix phase is also shown for two-suspension treated HDPE/TES3 and HDPE/TCC3 composites. More void contents and pulled out particles were noticed in HDPE/TCC2 composites. The plate-like shape of ES particles allowed better interfacial adhesion with the matrix at their edges compared to sphere-like particles.
Figure 7. SEM images of tensile fracture surface of HDPE/ES composites.

Figure 8. SEM images of tensile surface fracture of HDPE/CC composites.
It is also known that the finer the particles the more sever is the problem of its dispersion in the matrix such as in case of HDPE/CC composites. On the other hand, Murugan et al. showed that the lower surface area of the large ES particles leads to their agglomeration. The rough surface topology of the composites indicated that they still ruptured in a ductile manner which might be due to more crack initiation and propagation sites.

4. Conclusion

In this study, the influence of the addition of ES as a filler to HDPE and their effect on its thermal, melt flow, and mechanical properties was investigated and compared to the conventional HDPE/CC composites. The chemical surface treatment of the particles with SA was also investigated for both types of fillers and their composites and the FTIR results showed some chemical structural changes of the ES and CC particles after the acidic treatment indicating the reaction of the SA with the particles. It is worth to mention that an optimization process of the concentration of SA, temperature, and time is needed in the future to further investigate the formation of a sufficient coating monolayer of SA on ES or CC surface. The change in the thermal properties of the neat HDPE occurred by incorporating ES or CC particles and the nucleating ability of ES or CC was investigated and the treated composites showed higher crystallinity values compared to the untreated ones mainly for HDPE/ES composites. The crystallinity and the flow properties of HDPE/CC composites were better enhanced compared to HDPE/ES composites. A slight change occurred in the melting points of the composites. The improvement in the mechanical properties of treated HDPE/ES composites was clear compared to that of untreated ones and it was more pronounced than that of HDPE/CC composites. This motivates us to use the ES powder as an alternative to CC for the purpose of comparison, the combination of both fillers at once in the matrix, and the adoption of more efficient processing technique are also some recommended work for future studies in this area.

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