

Preparation and Characterization of Polyethylene-based Hybrid Particles by an Environmentally-friendly and Aqueous Solvent Evaporation Method

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Abstract

The present paper reports preparation procedure and characterization of micrometer-sized polyethylene (PE)-based hybrid particles containing various amounts of Mg(OH)₂ powder treated with different amounts of methylhydrogen polysiloxane (MHS). The PE-based hybrid particles were fabricated by an environmentally-friendly and aqueous solvent evaporation method by employing different kinds of surfactants. The shape, microstructure and other properties of the resultant PE-based hybrid particles were dependent markedly on the changes in composition of raw materials, especially for the amount of MHS used for the treatment of Mg(OH)₂ and the kind of surfactant. The particles fabricated by using 1 wt% MHS-treated Mg(OH)₂ (ST-1) powder and polyoxyethylene (10) octylphenyl ether (Triton X-100) as a surfactant showed spherical shape and their primary particle sizes were about 4 - 10 μm, irrespective of the additive amount of ST-1 powder. These particles showed superior properties in terms of the actual content of MHS-treated Mg(OH)₂ powder incorporated inside the hybrid particles, particle size distribution and particle shape, in comparison with other particles fabricated by using 5 wt% MHS-treated Mg(OH)₂ (ST-5) powder and polyoxyethylene (8) octylphenyl ether (Triton X-114) as a surfactant. This is due to good affinity (average contact angle was 19.4°) between the ST-1 powder and the aqueous phase, i.e. a continuous phase, dissolving Triton X-100. Furthermore, a composite fabricated by employing these PE-based hybrid particles showed uniform and homogeneous distribution of ST-1 powder in the PE matrix.

Keywords: Polyethylene; Solvent evaporation method; Hybrid particle; Environment; Methylhydrogen polysiloxane

1. Introduction

Polyethylene (PE) is one of important polymer materials used in various fields, such as electrical and agricultural fields, for the fabrication of household, packing and insulating products. Depending upon its application fields, PE-based composites containing certain functional fillers have been put into practical use. Mechanical mixing by an electrothermal roll, a hydraulic roll or a twin screw extruder is the simplest and commonest method [1-2] for the preparation of the PE-based composites. After mechanical mixing, the resultant PE-based composites were formed into pellets of a few millimeters in size for easy-handling in the subsequent process. However, conventional mechanical mixing is rather a time-consuming and skilled process for achieving homogenous dispersion of fillers. For example, segregation of functional fillers inside of the composites due to insufficient mixing may lead to poor mechanical properties, i.e. decreases in both of the coefficient of elongation and the rupture stress. Thus, a key point to achieve homogenous filler dispersion in final products within a short time is to use micrometer-sized PE-based hybrid particles containing functional fillers as a raw material. This method also enables us to make a complex shape and a tailored design and/or control of properties of products by changing the kinds and contents of functional fillers.

Therefore, such polymer-based hybrid particles containing homogeneously dispersed functional fillers such as metal and ceramics, are highly requested in various fields such as recording and display, beauty aids, fragrance, food and drug delivery [3-8]. Consequently, special attentions have been focused on developing new preparation conditions of such polymer-based hybrid particles so as to fabricate final products with excellent properties [9-14]. Among several techniques and methods so far reported for the conventional polymer-based hybrid particles, a solvent evaporation method is of interest from the viewpoint of simplicity of the preparation process [15-18]. To our knowledge, however, the solvent evaporation method was not a relevant method for certain polymers such as PE and polypropylene, because of limitation of environmentally-friendly solvents capable of dissolving these polymers completely. Therefore, this method has not been studied well for the fabrication of PE-based hybrid particles.

Among several functional properties of final PE-based composite products, our final target is improvement of their flame retardancy [19]. In the present study, therefore, our efforts have been directed to establishing a fabrication process of PE-based hybrid particles containing $Mg(OH)_2$ powder modified with methylhydrogen polysiloxane (MHS) by an environmentally-friendly aqueous solvent evaporation method. Namely, a mixture of cyclohexane and heptane was used to dissolve PE at the first step and then distilled water containing only a surfactant was employed as a continuous phase when organic solvents were evaporated from PE-based hybrid droplets. The

effects of the amounts of both the MHS to be used for the treatment of $\text{Mg}(\text{OH})_2$ powder and the MHS-treated $\text{Mg}(\text{OH})_2$ powder to be added to the PE matrix, and the kind and amount of surfactants in the continuous phase on the properties of resulting PE-based hybrid particles have been investigated.

2. Experimental

2.1 Chemicals and preparation of PE-based hybrid particles

2.1.1 Surface modification of $\text{Mg}(\text{OH})_2$ powder with methylhydrogen polysiloxane

$\text{Mg}(\text{OH})_2$ powder (Konoshima Chemical Co., Ltd.) with a mean particle size of ca. 0.6 μm was used as a filler for the PE-based flame retardant hybrid particles. Prior to fabricating hybrid particles, the surface of $\text{Mg}(\text{OH})_2$ powder was treated with methylhydrogen polysiloxane (MHS, Dow Corning Toray Co., Ltd.) in the following manner, in order to change the surface property from hydrophilic to hydrophobic so as to achieve homogeneous distribution in the PE matrix and then to enhance the affinity of the MHS-treated $\text{Mg}(\text{OH})_2$ powder with the PE matrix. MHS was mixed with isopropyl alcohol (IPA) in a weight ratio of 1 : 5, and the mixture was sprayed to the $\text{Mg}(\text{OH})_2$ powder in a general mixer while stirring at 1740 rpm for 0.9 ks. The amounts of MHS used for the surface modification was controlled to be 1 or 5 wt% of $\text{Mg}(\text{OH})_2$ powder by adjusting the amount of the mixture sprayed. Then, the resultant powders were subjected to the heat treatment at 423 K for 7.2 ks under vacuum, in order to remove IPA and proceed a chemical reaction between $\text{Mg}(\text{OH})_2$ powder and MHS. These hydrophobic MHS-treated $\text{Mg}(\text{OH})_2$ powders will be referred to as ST-1 and ST-5 powders, depending upon the amount of MHS used, 1 wt% and 5 wt%, respectively.

2.1.2 Fabrication of PE-based particles by using a solvent evaporation method

A solvent evaporation method (SE method) was adopted to fabricate PE-based hybrid particles containing MHS-treated $\text{Mg}(\text{OH})_2$ powder. First, 2 g of PE powder (UF-80, Sumitomo Seika Chemicals, Co., Ltd.) was dissolved in 20 g of cyclohexane-heptane (volume ratio of 1 : 1) mixed solvent at 353 K. Then, a given amount of ST-1 or ST-5 powder was added to the mixture. Thereafter, the resulting mixture containing ST-1 or ST-5 powder was gradually dropped into 900 ml of the continuous phase, i.e. the deionized water dissolving Triton X-114 ($4-(\text{C}_8\text{H}_{17})\text{C}_6\text{H}_4(\text{OCH}_2\text{CH}_2)_8\text{OH}$, hydrophile-lypophile balance (HLB) value = 12.4, Wako Pure Chemical Industries, Ltd.) or Triton X-100 ($4-(\text{C}_8\text{H}_{17})\text{C}_6\text{H}_4(\text{OCH}_2\text{CH}_2)_{10}\text{OH}$, HLB value = 13.5, Wako Pure Chemical Industries, Ltd.) kept around 348 K, which was higher than the cloud points of

these two surfactants. Namely, the composition of raw chemicals was controlled to be 45000 parts of deionized water, 225 or 450 parts of Triton X-114 or Triton X-100, 1000 parts of a cyclohexane-heptane mixed solvent, x ($x = 0, 10, 30, 50$ and 70) parts of ST-1 or ST-5 powder, and 100 parts of PE powder on a basis of weight of chemicals. Small droplets of the raw mixture in the continuous phase will be referred to as green PE-based hybrid droplets or green PE-based droplets, whether the MHS-treated $\text{Mg}(\text{OH})_2$ (ST-1 or ST-5) powder was involved or not, respectively. The detailed composition of each sample is summarized in Table 1 along with the sample number: the first numeral expresses the kind of filler (zero: no addition), the second the amount of the filler added, the third the kind of surfactant (A: Triton X-114 and B: Triton X-100) and the fourth the amount of surfactant used for fabricating the sample. The resulting oil-in-water (O/W) emulsion was suspended by using a homomixer at a rotation speed of 4000 rpm for 0.6 ks and then was subjected to evaporation of the organic solvent at 353 K for 14.4 ks while stirring at a speed of 2.5 rps. After the evaporation process, the resultant PE-based hybrid particles were collected by double filtrations (1 and 7 μm) under reduced pressure, washed sufficiently with deionized water and dried at 353 K for 7.2 ks.

2.2 Fabrication of composites from PE-based hybrid particles

Two kinds of PE-based hybrid particles, 1-7-B-4.5 and 1-7-B-9, were formed into a cylinder shape (10 mm in diameter and 2 mm in height) by uniaxial pressing, and heated at 423 K for 7.2 ks, in order to confirm the dispersion behavior of ST-1 powder in the PE-based composites. For comparative purpose, a PE-based composite was also fabricated under the same condition by employing a mixture of PE and ST-1 powders (weigh ratio of 2 : 1).

2.3 Characterization of PE-based hybrid particles and composites

Affinities of untreated $\text{Mg}(\text{OH})_2$, ST-1 and ST-5 powders to deionized water and the continuous phase, i.e. the deionized water dissolving Triton X-114 or Triton X-100 at different concentrations were evaluated by measuring the contact angles of droplets of these aqueous phases kept around 348 K, which was higher than the cloud points of both surfactants, on the tablets of those powders with a $\theta/2$ method. Concentration of the surfactants in deionized water was controlled to be the same as that for the continuous phase used for the SE method (see Table 1).

The shapes of the PE-based hybrid particles fabricated were observed by scanning electron microscopy (SEM, S-2250N, Hitachi High-Technologies Corp.). Untreated $\text{Mg}(\text{OH})_2$ powder, ST-1 powder, raw PE powder and 1- x -B-9.0 series samples including 0-0-B-9.0 were examined with a Fourier transform infrared (FT-IR) spectrometer (FT/IR-680, JASCO Corp.) at room temperature

using a KBr method at 4 cm^{-1} resolution. All PE-based hybrid particles were fired at 1273 K at a heating rate of 6.7 deg/min under flowing air (flow rate: 100 ml/min) and the amounts of MgO left behind were measured to confirm actual contents of ST-1 or ST-5 powder in the hybrid particles. The particle size distributions of 1-x-B-4.5 series samples including 0-0-B-4.5 and 1-x-A-9.0 series samples including 0-0-A-9.0 were measured with a laser particle size analyzer (SALD-3000s, Shimadzu). The measurement was done under the conditions where they were dispersed into deionized water with an alcohol ethoxy sulfate.

Fracture surfaces of the PE-based composites were observed by field-emission scanning electron microscopy (FE-SEM, JSM-7500FA, JEOL Ltd.). In addition, energy dispersion X-ray spectrometer (EDS, JSM-7500FA, JEOL Ltd.) analysis was performed to observe the dispersion state of ST-1 powder inside the composites.

3. Results and discussion

3.1 Properties of MHS-treated $\text{Mg}(\text{OH})_2$ powder

In general, the surface of untreated $\text{Mg}(\text{OH})_2$ powder is covered with hydroxyl groups and therefore shows a hydrophilic nature. This results in a poor affinity with a PE matrix. Therefore, the surface of $\text{Mg}(\text{OH})_2$ powder was treated with MHS so as to change to a hydrophobic nature and to guarantee a good affinity with the PE matrix. During the surface treatment, the hydroxyl groups on the surface of $\text{Mg}(\text{OH})_2$ powder was expected to react with MHS. Figure 1 shows FT-IR spectra of (a) untreated $\text{Mg}(\text{OH})_2$ and (b) ST-1 powders. The FT-IR spectrum of ST-5 powder was almost the same as that for the ST-1 powder. The intensity of a peak centered at 3696 cm^{-1} , assignable to O-H stretching, remained almost unchanged even after the reaction with MHS. This may be because the amount of OH groups reacted is small, in comparison with the total amount of OH groups on the surface. However, the IR spectrum shown in Fig. 1(b) revealed Si-H stretching between 2250 and 2100 cm^{-1} , Si-CH₃ bending between 1280 and 1255 cm^{-1} and Si-O stretching between 1110 and 1000 cm^{-1} . The chemical reaction between hydroxyl groups and MHS results in the formation of a new Si-O bond accompanied by evolution of gaseous H₂. Although the new Si-O bond could not be distinguished from the bonds originally existing in the MHS molecule in the FT-IR measurement, formation of gaseous H₂ was confirmed during the reaction. Therefore, it is obvious that the chemical reaction between $\text{Mg}(\text{OH})_2$ and MHS has occurred under the conditions employed in the present study, but the surface of $\text{Mg}(\text{OH})_2$ powder is also expected to be covered with physically adsorbed MHS from the appearance of peaks ascribable to Si-H stretching.

3.2 Properties of PE-based particles fabricated by using a solvent evaporation method

Spherical PE particles with a smooth surface and a particle size of ca. 60 μm could be fabricated by the present SE method by employing chlorobenzene as an organic solvent to dissolve PE (the data is not shown for the sake of simplicity). However, the use of chlorobenzene leads to serious environmental problems, and therefore, our efforts were first directed to finding an alternative environmentally-friendly organic solvent. Among the environmentally-friendly organic solvents tested, a mixture of cyclohexane and heptane (volume ratio of 1 : 1) was found to be most suitable for dissolving PE. Therefore, the results obtained with the cyclohexane-heptane solvent are reported below. To improve dispersion behavior of the O/W emulsion in the subsequent process, the effect of surfactants to be added to deionized water was evaluated from the shapes of resulting PE particles, i.e. under the conditions of no ST-1 or ST-5 powder addition.

Figure 2 shows SEM images of the resulting four kinds of PE particles, 0-0-A-4.5, 0-0-A-9.0, 0-0-B-4.5 and 0-0-B-9.0, shown in Table 1. The 0-0-A-4.5 and 0-0-A-9.0 samples fabricated by employing the Triton X-114 resulted in agglomerated large particles with undesirable shapes, irrespective of the additive amount of Triton X-114, as shown in Figs. 2(a) and 2(b). As a result, when Triton X-114 was used, spherical PE particles with a smooth surface could not be fabricated.

In contrast, the use of Triton X-100, instead of Triton X-114, led to small, spherical and well-dispersed PE particles with a smooth surface, irrespective of the additive amount of Triton X-100, as shown in Figs. 2(c) and 2(d). However, when the additive amount of PE increased by a factor of 3, the surface of the resulting PE particles became rough, even if Triton X-100 was used (the data is not shown). This is due to the increased viscosity of the green PE-based droplets.

When green PE-based hybrid droplets were used, the resulting PE-based hybrid particles also agglomerated to large particles with a rough surface, as far as Triton X-114 was used as a surfactant to be added to the continuous phase, irrespective of the additive amount of Triton X-114. However, Triton X-100 was found to be effective for fabricating small, spherical and well-dispersed PE-based hybrid particles, whereas particle distributions were influenced by the kind of MHS-treated $\text{Mg}(\text{OH})_2$ powders.

Figures 3 and 4 show SEM images of 1-x-B-4.5 and 1-x-B-9.0 series samples fabricated under the conditions indicated in Table 1, respectively. It is obvious that these hybrid particles are sphere in shape with a smooth surface and a primary particle size of about 5 - 10 μm , especially when the mixed content of ST-1 powder is 10 phr. However, the surfaces of these hybrid particles became rough with increasing the additive amount of ST-1 powder. Especially, under higher Triton X-100 concentration and higher ST-1 powder content conditions, surface roughness seems to originate from adhesion of smaller PE particles and ST-1 powder on large PE-based hybrid particles, as shown in Figs. 3(h) and 4(h), rather than the uneven surfaces of large PE-based hybrid particles themselves. EDS analysis has relieved that ST-1 powder is not distributed homogeneously inside the PE-based

hybrid particles and tends to gather in the surface region of the PE-based hybrid particles, especially for higher ST-1 powder contents, as shown in Fig. 5. Similar tendency was also found more or less for all series of samples. This phenomenon may be explained by either one of the following two factors or by both: an increase in surface to volume ratio by decreasing the size of green PE-based hybrid droplets during the mixing in the SE method and better affinity of ST-1 powder to the continuous phase than the PE matrix. Besides the primary particles, both the secondary particles in the size of 4 - 10 μm and the agglomerated particles in the size of ca. 100 μm were observed by SEM and an optical microscope, respectively, for 1-x-B-4.5 and 1-x-B-9.0 series samples.

When ST-5 powder was employed, segregation of ST-5 powder in the surface region of PE-based hybrid particles became significant, and beside particles in 4 - 10 μm size, formation of agglomerated particles was pronounced, irrespective of the amount of Triton X-100 in the continuous phase, though the data is not shown here. Therefore, ST-1 powder was found to be superior to ST-5 powder as a filler for the PE-based hybrid particles.

Figure 6 shows the IR spectra of raw PE powder and 1-x-B-9.0 series samples including 0-0-B-9.0. All the IR spectra revealed the existence of C-H asymmetrical stretching at 2926 cm^{-1} , C-H symmetrical stretching at 2853 cm^{-1} , C-H asymmetrical bending at 1465 cm^{-1} and C-H symmetrical bending at 1375 cm^{-1} originating from PE. Besides these absorption peaks, an additional peak attributed to O-H stretching were observed at 3696 cm^{-1} in the IR spectra of Figs. 6(c) - (f). These results also confirm the existence of ST-1 powder in the PE matrix, i.e. the formation of PE-based hybrid particles by the present method. The peaks attributed to O-H stretching were also observed in 5-x-B-9.0 series samples, although the data is not shown here.

Relationships between mixed and observed MHS-treated $\text{Mg}(\text{OH})_2$ powder contents of 1-x-A-4.5, 1-x-A-9.0, 5-x-A-4.5 and 5-x-A-9.0 series samples and 1-x-B-4.5, 1-x-B-9.0, 5-x-B-4.5 and 5-x-B-9.0 series samples are shown in Figs. 7 and 8, respectively. When Triton X-114 was employed, the actual contents of MHS-treated $\text{Mg}(\text{OH})_2$ powder incorporated in all the PE-based hybrid particles were far less than the mixed values. Especially, the observed contents of 1-x-A-9.0 and 5-x-A-4.5 series samples were less than 4 phr over the whole mixed contents even at 70 phr, while the observed contents of 1-7-A-4.5 and 5-7-A-9.0 samples increased to ca. 20 phr, as shown in Fig. 7. These results indicate that the almost all ST-1 or ST-5 powder involved in green PE-based hybrid droplets was extracted into the continuous phase during the second stage of the present SE method, namely during the evaporation of organic solvents from the droplets in the continuous phase. In contrast, when Triton X-100 was used, i.e. in the cases of 1-x-B-4.5, 1-x-B-9.0, 5-x-B-4.5 and 5-x-B-9.0 series samples, the observed content of MHS-treated $\text{Mg}(\text{OH})_2$ powder increased almost linearly with an increase in the mixed contents, although they showed 30 - 40% loss from the mixed contents, with only one exception observed for the 5-7-B-4.5 sample, as shown in Fig. 8. These results indicate that Triton X-100 is useful for limiting the extraction of ST-1 or ST-5 powder from

the droplets in the continuous phase and then for increasing the contents of the ST-1 or ST-5 powder in the resulting PE-based hybrid particles. Such phenomena undoubtedly arise from differences in nature of the continuous phases dissolving the surfactants, namely, the affinity of the ST-1 or ST-5 powder with the continuous phase dissolving Triton X-100 is anticipated to be smaller than that dissolving Triton X-114 which shows more oily nature than Triton X-100. Such a tendency is more pronounced in the case of ST-1 powder, than ST-5 powder.

Figures 9(a) and 9(b) show the particle size distributions of the 1-x-B-4.5 series samples including 0-0-B-4.5 and 1-x-B-9.0 series samples including 0-0-B-9.0, respectively. It is found that the particle size of both series samples distributes over a wide range of ca. 1.0 - 127.0 μm . This result also confirms that these samples are a mixture of primary and secondary particles, as was observed by SEM and an optical microscope. In addition, no remarkable change in particle size distribution was observed between 1-x-B-4.5 and 1x-B-9.0 series samples. This fact implies that the additive amount of Triton X-100 exhibits little influence on the number and size of green PE-based hybrid droplets in the continuous phase.

As mentioned above, microstructure of resultant PE-based hybrid particles was markedly affected by the kind of surfactant dissolving in the continuous phase and the amount of MHS used for the treatment of $\text{Mg}(\text{OH})_2$ powder. In order to find possible reasons to explain such phenomena, affinity of the continuous phase dissolving Triton X-114 or Triton X-100 to ST-1 and ST-5 powders was estimated from the contact angles of droplets of the continuous phases on the tablet of ST-1 and ST-5 powders. This is because most of ST-1 and ST-5 powders are anticipated to gather in the surface region of green PE-based hybrid droplets and therefore affect the behavior of green PE-based hybrid droplets in the continuous phase.

Figure 10 shows external views and contact angles of droplets of the continuous phases on the tablets of ST-1 and ST-5 powders. For comparative purpose, the contact angles of droplets of the continuous phases and deionized water on a tablet of untreated $\text{Mg}(\text{OH})_2$ powder were also measured. As expected, the contact angle of zero, i.e. fairly good affinity to every continuous phase, including deionized water, was confirmed with untreated $\text{Mg}(\text{OH})_2$ powder. On the other hand, contact angle values between the droplets of deionized water and the tablets of ST-1 and ST-5 powders were 120.7° and 123.9° , respectively, confirming the change from the hydrophilic to the hydrophobic nature by the MHS treatment. The contact angle values between the droplets of the continuous phase dissolving Triton X-114 and the tablets of ST-1 and ST-5 powders were lower than those of the continuous phase dissolving Triton X-100, when the comparison was made at the same concentration of surfactants. This indicates better affinity of the continuous phase dissolving Triton X-114 to MHS-treated $\text{Mg}(\text{OH})_2$ powders than that dissolving Triton X-100. Such phenomena is suggested to arise from higher lipophilic nature of Triton X-114 than Triton X-100, as is expected from its functional groups involved in its molecule. An increase in surfactant concentration

resulted in a lower contact angle and then a further increase in affinity. The better affinity is considered to lead to extraction of ST-1 or ST-5 powder from the surface of green PE-based hybrid droplets into the continuous phase. Therefore, this may be the main reason for the lower $\text{Mg}(\text{OH})_2$ contents observed for 1-x-A-4.5, 1-x-A-9.0, 5-x-A-4.5 and 5-x-A-9.0 series samples, than those observed for 1-x-B-4.5, 1-x-B-9.0, 5-x-B-4.5 and 5-x-B-9.0 series samples (see Figs. 6 and 7).

When comparison was made between ST-1 and ST-5 powders, a larger contact angle was obtained with ST-5 powder for every continuous phase. This implies poor affinity of ST-5 powder to the continuous phase, in comparison with ST-1 powder, and predicts less extraction of ST-5 powder from the green PE-based hybrid droplets. However, results shown in Figs. 7 and 8 cannot be explained by the changes in contact angle. The affinity between surfactants and ST-5 powder or the affinity among ST-5 powders may also affect the content of MHS-treated $\text{Mg}(\text{OH})_2$ powder and the shapes of the resulting particles, though the details are not clear at present.

3.3 Properties of composites fabricated from the hybrid particles

To confirm the usefulness of PE-based hybrid particles, dispersion states of ST-1 powder in the composites fabricated with 1-7-B-4.5, 1-7-B-9.0 particles and a mixture of PE and ST-1 powders were compared. The fabricated composites could maintain their cylindrical shape after the molding without the addition of other binders, and no cracks were observed after their heat treatment. Fig. 11 shows SEM and EDS results ($\text{Mg-K}\alpha$) of the fracture surfaces of these composites. The EDS analysis shown in Fig 11(b) indicates insufficient distribution of ST-1 powder in the PE matrix, i.e. two different regions, high ST-1 powder content and no ST-1 powder regions, were clearly observed. In contrast, more uniform and homogeneous distribution of ST-1 powder in the PE matrix was confirmed at the fracture surfaces of the composites fabricated from the PE-based hybrid particles. This result revealed an advantage of the PE-based hybrid particles to achieve sufficient dispersion of $\text{Mg}(\text{OH})_2$ powder in the final composites, even though $\text{Mg}(\text{OH})_2$ powder was rather gathered in the surface region of the PE-based hybrid particles. The small particle size of the resulting PE-based hybrid particles was anticipated to be useful for achieving uniform and homogeneous distribution of ST-1 powder in the PE matrix in the final products with a short processing time.

4. Conclusions

PE spherical particles and PE-based hybrid particles with different additive amounts of $\text{Mg}(\text{OH})_2$ powder were fabricated by an environmentally-friendly and aqueous solvent evaporation method by employing different kinds of surfactants. The shape, microstructure and other properties of the resultant PE-based hybrid particles were dependent markedly on the amount of MHS used for the

treatment of $\text{Mg}(\text{OH})_2$ and the kind of surfactant. The particles fabricated by using 1 wt% MHS-treated $\text{Mg}(\text{OH})_2$ (ST-1) powder and polyoxyethylene (10) octylphenyl ether (Triton X-100) as a surfactant showed spherical shape, comparatively high values of observed MHS-treated $\text{Mg}(\text{OH})_2$ powder contents and their primary particle sizes were about 4 - 10 μm , irrespective of the additive amount of ST-1 powder. These particles showed superior properties in terms of the actual contents of MHS-treated $\text{Mg}(\text{OH})_2$ powder incorporated inside the hybrid particles, particle size distributions and particle shape, in comparison with other particles fabricated by using 5 wt% MHS-treated $\text{Mg}(\text{OH})_2$ (ST-5) powder and polyoxyethylene (8) octylphenyl ether (Triton X-114) as a surfactant. This is due to good affinity (average contact angle was 19.4°) between the ST-1 powder and the aqueous phase, i.e. a continuous phase, dissolving Triton X-100. Furthermore, a composite fabricated by employing these PE-based hybrid particles showed uniform and homogeneous distribution of ST-1 powder in the PE matrix. These results also provide useful information on the fabrication of PE-based hybrid particles mixed with other functional fillers such as lubricants and antioxidants. These PE-based hybrid particles have a potential for a wide variety of application fields such as household, packing and electrical insulating material.

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Figure captions and tables

Fig. 1 FT-IR spectra of (a) untreated $Mg(OH)_2$ and (b) ST-1 powders.

Fig. 2 SEM images of the PE particles without fillers. (a) 0-0-A-4.5, (b) 0-0-A-9.0, (c) 0-0-B-4.5 and (d) 0-0-B-9.0.

Fig. 3 SEM images of the PE-based hybrid particles based on ST-1 powder and 4.5 g of Triton X-100. (a) and (b) 1-1-B-4.5, (c) and (d) 1-3-B-4.5, (e) and (f) 1-5-B-4.5, (g) and (h) 1-7-B-4.5.

Fig. 4 SEM images of the PE-based hybrid particles based on ST-1 powder and 9.0 g of Triton X-100. (a) and (b) 1-1-B-9.0, (c) and (d) 1-3-B-9.0, (e) and (f) 1-5-B-9.0, (g) and (h) 1-7-B-9.0.

Fig. 5 EDS data measured for a 1-7-B-9.0 particle based on 70 phr of ST-1 powder and 9.0 g of Triton X-100. (a) SEI, (b) C-K α image, (c) Mg-K α image and (d) O-K α image.

Fig. 6 FT-IR spectra of (a) raw PE powder, (b) 0-0-B-9.0, (c) 1-1-B-9.0, (d) 1-3-B-9.0, (e) 1-5-B-9.0 and (f) 1-7-B-9.0.

Fig. 7 Relationship between mixed and observed MHS-treated $Mg(OH)_2$ powder contents in the 1-x-A-4.5, 1-x-A-9.0, 5-x-A-4.5 and 5-x-A-9.0 series samples.

Fig. 8 Relationship between mixed and observed MHS-treated $Mg(OH)_2$ powder contents in the 1-x-B-4.5, 1-x-B-9.0, 5-x-B-4.5 and 5-x-B-9.0 series samples.

Fig. 9 Particle size distributions of (a) 1-x-B-4.5 series samples including 0-0-B-4.5 and (b) 1-x-B-9.0 series samples including 0-0-B-9.0.

Fig. 10 External views and contact angles of droplets of the continuous phases on tablets of ST-1 and ST-5 powders.

Fig. 11 SEM images and EDS analysis results (Mg-K α) of fracture surfaces of the composites fabricated from (a) and (b) a mixture of PE and ST-1 powders, (c) and (d) 1-7-B-4.5 based on 70 phr of ST-1 powder and 4.5 g of Triton X-100, and (e) and (f) 1-7-B-9.0 based on 70 phr of ST-1 powder and 9.0 g of Triton X-100. (a), (c) and (e) SEM images and (b), (d) and (f) corresponding EDS images.

Table 1 Preparation conditions of PE particles and PE-based hybrid particles.