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Novel synthesis of magnesium hydroxide nanoparticles modified with organic phosphate and their effect on the flammability of acrylonitrile-butadiene styrene nanocomposites

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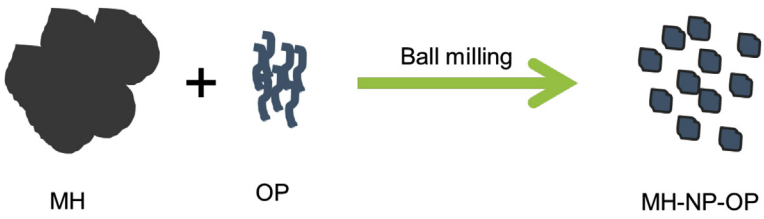
HIGHLIGHTS

- Novel and facile nanoparticles synthesis and modification have developed.
- Magnesium hydroxide nanoparticles size has tuned by the developed method.
- A well dispersed polymer nanocomposites has prepared.
- Flame retardancy of the new nanocomposite has significantly improved.
- Synergistic effect between nanoparticles and their organic shell has investigated.

GRAPHICAL ABSTRACT



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ABSTRACT

New and facile method for the synthesis and modification of magnesium hydroxide nanoparticles has been developed. The organic phosphate was used to facilitate the synthesis and wrapping of magnesium hydroxide nanoparticles with organic phosphate shell. The size of the nanoparticles wrapped with phosphate has an average diameter range from 46 to 125 nm. The preparation method has governed the nanoparticles diameter based on reaction time. Thermal stability and morphological properties of the new nanoparticles coated phosphates were investigated. The developed magnesium hydroxide nanoparticles-organic phosphate achieved a very good compatibility when dispersed in acrylonitrile-butadiene styrene polymer (ABS) produced dispersed nanocomposites. The flammability and thermal properties of the new polymer nanocomposites were studied. The rate of burning of the nanocomposites was reduced to 9.8 mm/min compared to 15, 21.9 and 42.5 mm/min for polymer-conventional magnesium hydroxide composite, polymer-conventional magnesium hydroxide-organic phosphate composite and virgin polymer, respectively. The peak heat release rate (PHRR) and total heat release (THR) of the new nanocomposites were recorded as 243.4 kW/m^2 and 19.2 MJ/m^2, respectively, achieved 71% reduction for PHRR and 55% for THR. The synergism between magnesium hydroxide nanoparticles and organic phosphates shell was also studied. The developed nanoparticles suppressed the emission of toxic gases. The different materials were characterized using thermal gravimetric analysis, fourier transform infrared spectroscopy, transmission electron microscopy. The flammability properties were evaluated

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using UL94 horizontal method and cone calorimeter. The dispersion of magnesium hydroxide nanoparticles-organic phosphate in ABS was studied using scanning electron microscope.

1. Introduction

Most of the organic polymers have low thermal stability and high flammability properties which shorten their use. Acrylonitrile–butadiene–styrene (ABS) is a thermoplastic polymer extensively used in various applications such as car industry, pipes, and telephone parts, but it is highly flammable polymer [1,2]. To improve the thermal stability and lower the flammability properties of ABS, halogenated flame retardants have been used [3,4]. However, due to their environmental pollution problems, halogenated flame retardants were prohibited to use [5,6]. Recently, the trend to use halogenated free flame retardants in polymers have been received a lot of attention [7–9]. Furthermore, various nanomaterials have been applied as fillers for improving thermal and mechanical properties, and reducing the flammability properties of the polymer nanocomposites due to their superior properties [1,10–13]. Metal hydroxides have been used widely as flame retardant fillers due to their cooling and dilution effects in addition to formation of metal oxide based char layer [14–17]. Magnesium hydroxide (MH) is a popular member of metal hydroxides, which is greatly used as flame retardant filler in different forms due to its properties and cost-effective price. Nevertheless, it has to be applied in high loading (60%) in polymer composites to achieve flame retardant efficiency. However, this high level of mass loading of MH to polymer attributed to aggregation and poor dispersion of MH. This is due to its inorganic nature and low compatibility with organic polymers. This resulted in reduction in mechanical properties of the final polymer composites [18–22]. There are many efforts have been carried out to improve the compatibility between MH and polymeric materials to enhance the dispersion of MH [23–25]. Recently, nanoparticles as a flame retardant fillers in polymer nanocomposites have been received attention, due to their effect in improving mechanical properties of polymer nanocomposites [26,27]. Various methods have been reported for the synthesis of MH nanoparticles [28–32]. It is pertinent to note that, our research group has long been involved in the study of fire resistant of various materials [1,33–37]. Recently, we have reported a significant reduction in flammability properties of ABS nanocomposites using halloysite nanotubes with intumescent flame retardant [1]. This is in conjunction with previous experience in processing of polymer nanocomposites with different nanomaterials [38–41]. Even though, the previous reported methods for the synthesis of MH nanoparticles produced nanoparticles with controlled size, but these techniques require prolonged synthesis time and difficult conditions which are hard to control in industrial scale. In this study, we reported for the first time the synthesis and modification of MH nanoparticles from conventional MH using a facile one step method. The MH nanoparticles (MH-NPs) with tunable size were developed from conventional one and modified with organic phosphate using wet ball milling method. The modified MH-NPs dispersed well in ABS matrix using solvent blending method forming well dispersed polymer nanocomposites. The thermal stability and flammability properties of ABS and their composites and nanocomposites were investigated. The synergistic effect of MH-NPs and modified phosphate layer was also studied.

2. Experimental

2.1. Materials

ABS suitable for injection moulding, with the trade name Terluran GP-22 was purchased from BASF, Ludwigshafen, Germany. Diethyl maleate was obtained from Merck, Schuchardt OHG 85662 hohenbrun, Germany. Magnesium Hydroxide 98% was supplied from Alpha Chemika, Mumbai, India. N, N-Dimethyl Formamide was provided from Sdfine-Chem limited, Mumbai, India. Dibenzoyl peroxide was obtained from Oxford laboratory, Mumbai, India. Acetone was purchased from El Nasr Pharmaceutical Chemicals Co., Egypt.

2.2. Preparation of organic phosphate flame retardants

2.2.1. Preparation of maleate di phosphate (DP)

Maleate diphosphate was prepared according to the previous method [35]. In details, in a dried round bottom flask phosphoric acid (2 mol) was mixed with diethyl maleate (1 mol) and then refluxed for 4 h at temperature 120 °C and afterwards viscous liquid was obtained.

2.2.2. Preparation of maleate mono phosphate (MP)

In a round bottom flask 1 mol of phosphoric acid was refluxed with 1 mol of diethyl maleate for 4 at 120 °C afterwards viscous liquid was obtained.

2.2.3. Preparation of polymaleate di phosphate (PDP)

Polymaleate di phosphate was prepared based on the previous report [42]. For the synthesis, in a dried round bottom flask 50 ml of DP was dissolved with 1.5 g of dibenzoyl peroxide in100 ml of N,N-Dimethyl formamide, then the mixture was refluxed for 4 h at 105 °C. Afterwards, solvent evaporated and polymaleate di phosphate was obtained.

2.2.4. Preparation of polymaleate mono phosphate (PMP)

In a round bottom flask contains 100 ml of N,N-Dimethyl formamide dissolved 50 ml of maleate mono phosphate and mixed with 1.5 g of dibenzoyl peroxide, after that the mixture was refluxed for 4 h at 105 °C. Afterwards, solvent evaporated and polymaleate mono phosphate was obtained.

2.2.5. Synthesis of modified magnesium hydroxide nanoparticles (MH-NPs)

First, conventional MH was dried at 110 °C for 4 h; after that quantized different mass ratios of dried MH were added individually in a ceramic capsule of ball milling machine contains zirconium oxide balls in acetone. Then different mass ratios of maleate diphosphate, maleate monophosphate, polymaleate diphosphate and polymaleate monophosphate were added to previous mixture, separately. Then the ball milling machine rotate at 300 rpm at interval times (12, 24 and 36 h). Afterwards, the produced mixtures were mechanically stirred for 1.5 h at 50 rpm.

2.2.6. Synthesis of ABS-MH-NP composites

In a beaker containing ABS solution dissolved in acetone,

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