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The Effect of Nano Loading and Ultrasonic Compounding of EVA/LDPE/Nano-magnesium Hydroxide on Mechanical Properties and Distribution of Nano Particles

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Abstract. Blends of Ethylene Vinyl Acetate (EVA) and Low-Density Polyethylene (LDPE) are promising composite which have good mechanical properties to environmental stress cracking. However, they lack fire resistant properties, which limits it usage in wire and cable industry. In order to improve flame retardancy ability, a range of nano-magnesium hydroxide (nano-MH) loading which is from 0 phr to maximum of 20 phr with ultrasonic extrusion 0-100 kHz frequencies have been introduced. Ultrasonic extrusion was used to improve the distribution of nano-MH. It was found that, 10 phr of nano loading with 100 kHz ultrasonic assisted has greater tensile strength compared to the nanocomposite without ultrasonication. Further increase of nano MH loading, will decrease the tensile properties. Better elongation at break was observed at10 phr nano-MH with the frequency of 50 kHz. The sample of 20 phr of nano-MH assisted with 50 kHz ultrasonic exhibits good flexural properties while 10 phr of nano-MH without the ultrasonic assisted demonstrates good in izod impact properties. From the evaluation of mechanical properties studied, it was found that 10 phr of nano-MH has shown the best performance among all the samples tested for EVA/LDPE/nano-MH composites. Transmission Electron Microscopy (TEM) has been conducted on 10 phr sample with different frequencies in order to observe the distribution of nano-MH particles. The sample with 100 kHz frequency shows more uniform dispersion of nano-MH in EVA/LDPE composites. This investigation indicates that the ultrasonic technology can enhance the mechanical properties studied as well as the dispersion of nano particles in the composite.

1. Introduction
The Polyolefins are the major group of commercial thermoplastics which have a wide range of applications due to their better combination of properties and low cost. Blends of Low-Density Polyethylene (LDPE) and Ethylene Vinyl Acetate (EVA) are widely used in consumer products such as cable and wire insulation [1][2]. It has good mechanical properties such as flexibility and toughness, good resistance to chemicals, and good process ability [3]. However, it has some drawbacks, such as low melting temperature, low thermal stability and high flammability, same goes with the ethylene vinyl acetate (EVA) applications that have been limited by its existing flammability. Therefore, flame retardants are introduced in order to improve the copolymer blends. Currently, the wire and cable industry are going through dramatic changes that require improvement in the design of wire and cable to compete with others in the industry. An alternative way to inhibit ignition and lower heat release rate during combustion is by adding additives such as nano magnesium hydroxide (nano-MH) [4]. The addition of these nanoparticle additives will affect the mechanical properties of the composite [5]. Apart from that, the success of nanoparticle applications depends on the ability of the nanoparticle to disperse evenly in the matrix. Since the powders are dry, the nanoparticle cannot disperse well in the copolymer

matrix which leads to agglomeration [6]. Hence, the ultrasonic has been introduced into the extrusion process to enhance the dispersion of the nano particle [7].

Therefore, the aims of this research are to study the mechanical properties of LDPE/EVA at different loading of nano-magnesium hydroxide and ultrasonic frequency. Subsequently, the effect of nanoparticle distribution will be studied by applying ultrasonic extrusion with varying frequencies on the optimum loading of nano-MH in EVA/LDPE blend.

2. Experimental Procedure

2.1. Material
LDPE was purchased from Asia Polymer Corporation, Taiwan with density 0.9185 g/cm3 and melt flow index 8.3 g/10 min, EVA (Exxon's Escorene UL02528) with melt flow index of 2.0g/10 min. Nano-MH was supplied by Sigma Aldrich, Switzerland, have an average thickness of 20 nm and length of 90 nm.

2.2. Composite preparation
In mixing process, the LDPE/EVA/nano-MH composites were prepared by using extruder with ultrasonic assisted. It had three controlled temperature zones which were set 170°C with die zone at 160°C and the extruder screw speed is 20 rpm. Ultrasonic was set between 0-100 kHz. The extruding ratio of the sample was based on the 100 phr of LDPE with 1:1 ratio of EVA and the nano MH was varied with 5 different ratios ranging from 0, 5, 10, 15 and 20 phr. The extruded samples were moulded in a compression mould at 170oC under pressure of 10 MPa for 6 minutes after 6 minutes. Then the mould was transferred to a cold press and then cooled at the same pressure. By using manual cutter machine, the rectangular sheet was being cut into required ASTM samples. 10 replicates for each formulation was being cut. The collected sample will undergo mechanical and dispersion testing.

2.3. Testing

2.3.1. Mechanical testing. The tensile strength elongation at break and young modulus, were measured with a universal testing machine from Instron Co. (Canton, MA) in accordance with ASTM D 638M (at a speed of 50 mm/min). The same machine with flexural tester was being used to obtain the flexural strength and flexural modulus with respect to the ASTM D790 (80 span mm, speed 50 mm/min). Izod impact (ASTM D256) strength was being measured by using Conventional Izod Impact Tester with an 4J impact load used. 2.54 mm notching depth have been notched at the sample. The sample was saoked in liquid nitrogen for 1 minute before being attach and tested at the machine. The purpose of soaking the sample in liquid nitrogen is to harden and toughen the sample in order to have an impact response.

2.3.2. Transmission electron microscope (TEM). TEM test was conducted to the LDPE/EVA/nano-MH samples to observe the nano magnesium hydroxide dispersion and intercalation in polymer matrix. 100kV of acceleration voltage was set at the TEM machine to observe the morphologies of the nano-MH particles in LDPE/EVA matrix. The sample was slice into ultrathin film form with a degradation temperature (Tg) of 55°C.

3. Results and discussion

3.1. Mechanical Properties

3.1.1. Tensile strength. Figure 1 shows the tensile strength for EVA/LDPE composites with different nano loading from 0, 5, 10, 15 and 20 phr at different frequency of ultrasonic. From the graph, it can be clearly seen that at 10 phr of nano loading with 100 kHz ultrasonic improved tensile strength compared to the nanocomposite without ultrasonication and 50 kHz of ultrasonic frequency. However, with further increased of nano MH, the tensile property decreased and reduced to 7MPa at 20phr. This is due to the decrease of interfacial interaction between nanoparticle and matrix [8]. Thus, deteriorate the tensile strength of the composite.

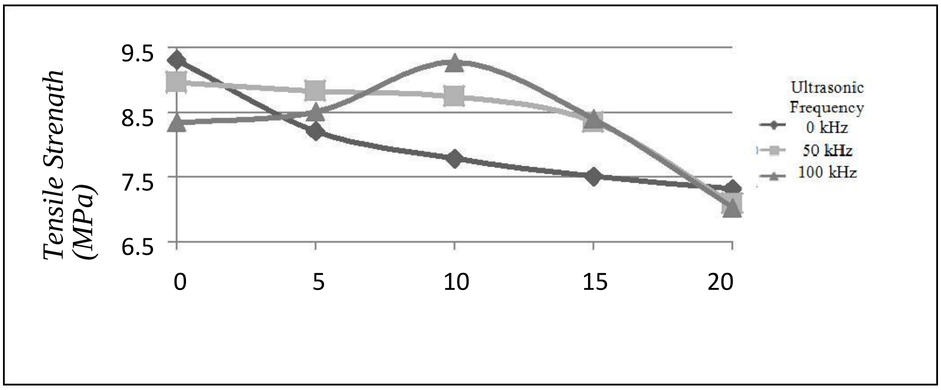

Figure 1. Tensile strength for varying ultrasonic frequency

3.1.2. Elongation at break. Figure 2 shows the flexural strength for EVA/LDPE composites with different nano loading from 0, 5, 10, 15 and 20 phr at different frequency of ultrasonic. From the graph, without ultrasonic, the elongation at break was slightly decreased when increased the nano loading from 5 phr to 20 phr. While for 50 kHz ultrasonic frequency, the elongation at break is higher at 10 phr of nano loading and decreased after the futher increased of nano-MH. Similar trend was observed at higher ultrasonic frequency (100kHz), but the elongation at break increment at 10phr of nano-MH loading is small. It was observed that, increasing the nano-MH loading from 10 to 20phr decrease the elongation ability of EVA-LDPE composite. This is due to the intercalation of EVA-LDPE matrix into the layer of nano-MH particles which will limit the mobility of the polymer chain to move freely Hence, lowering the elongation at break [9].

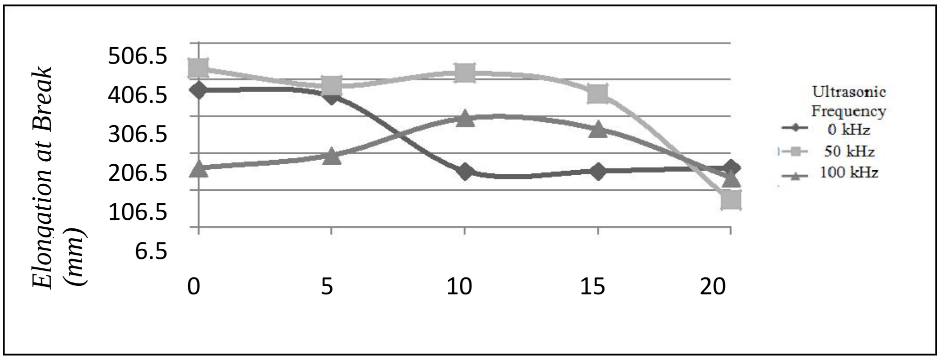

Figure 2. Elongation at break varying ultrasonic frequency

3.1.3. Youngs modulus. Figure 3 shows the Young Modulus for EVA/LDPE composites with different nano loading from 0, 5, 10, 15 and 20 phr with different frequency of ultrasonic.

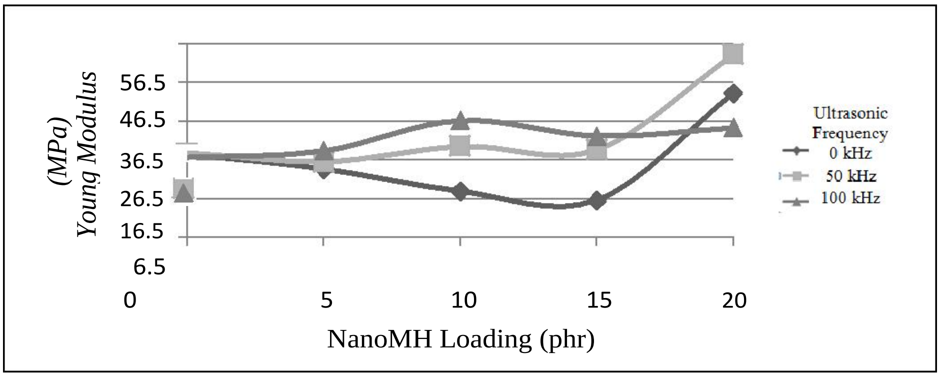

Figure 3. Young modulus for varying ultrasonic frequency

From the graph, it can be clearly seen that at 15 phr of nano loading with different frequency, the graph increased when reached at higher nano loading, 20 phr. However, with 50 kHz ultrasonic frequency, it shows the highest Young Modulus at 20 phr of nano loading. This indicates that, addition of nano-MH particle into EVA/LDPE matrix enhance the stiffness Young Modulus [9]. The increase in Youngs modulus with nano-MH loading suggests an efficient stress transfer between the nanoparticle and the matrix after treatment the fiber. Another reason is good dispersion or mixing between the nano-MH and the EVA/LDPE matrix [10].

3.1.4. Flexural strength. Figure 4 shows the flexural strength for EVA/LDPE composites with different nano loading from 0, 5, 10, 15 and 20 phr with different frequency of ultrasonic. From the graph, at 10phr with 100 kHz ultrasonic frequency, shows higher flexural strength (8.1 MPa). A similar trend is observed where, with increasing nano loading, the flexural strength increases. The flexural strength of the nanocomposite increases when 20phr nano-MH added due to compatibility factor which is affected from the better dispersion of nano particles.

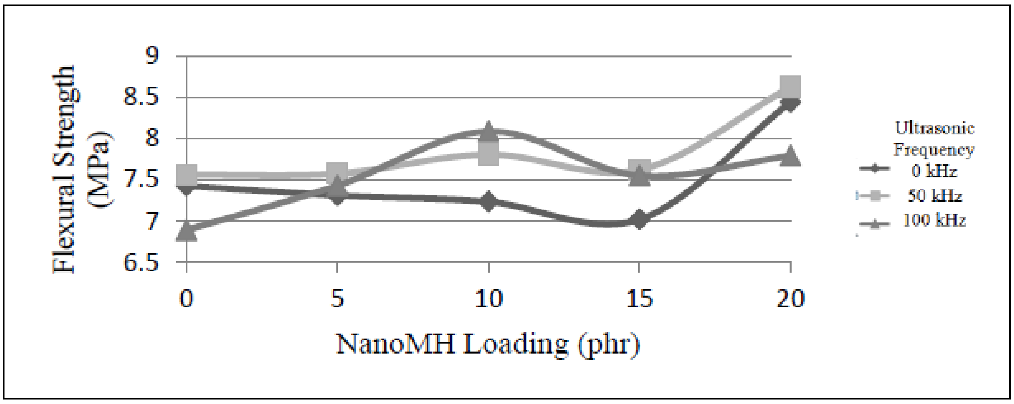

Figure 4. Flexural strength for varying ultrasonic frequency

3.1.5. Izod impact. Figure 5 shows the izod impact strength for EVA/LDPE composites with different nano loading from 0, 5, 10, 15 and 20 phr with different frequency of ultrasonic. From the graph, it can be clearly seen that at 10 phr of nano loading with 100 kHz ultrasonic assisted was the lowest in izod impact strength compared to the nanocomposite without ultrasonication and 50 kHz of ultrasonic frequency. Overall, at 5 phr of nano loading it can be clearly seen that with 50 kHz of ultrasonic frequency shows higher izod impact strength compared to others.

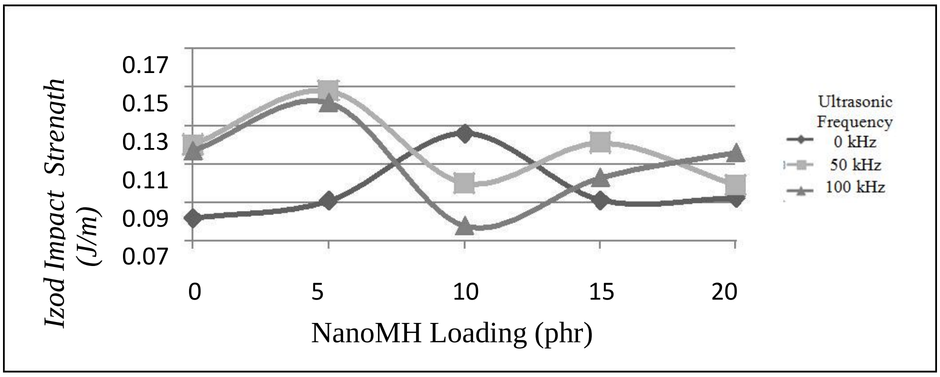

Figure 5. Izod impact strength varying ultrasonic frequency

3.2. Dispersion of nanoparticles

The distribution of the nanosized particles plays an important role in nanocomposite properties as nanocomposite properties are strongly dependant on the final morphology of the material. However, dispersion of the nanosized particles remains a challenge for nanocomposites due to the strong interaction of the nanoparticles.

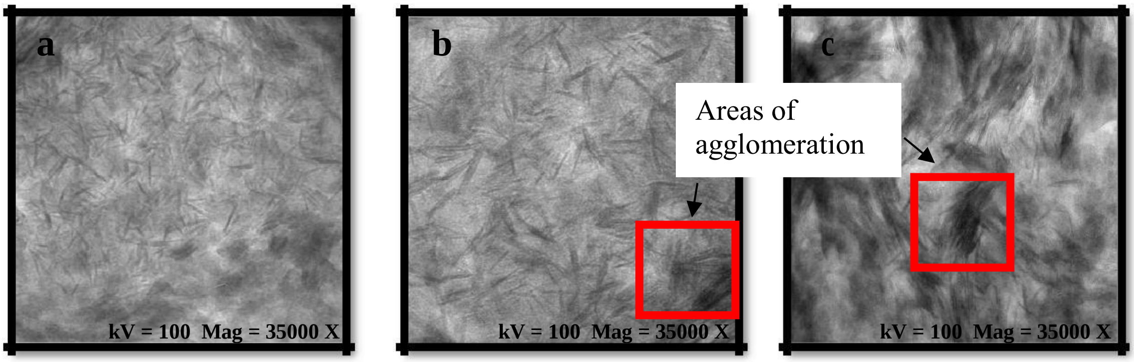

Figure 6. Dispersion of 10phr Nano-Magnesium Hydroxide with varying Ultrasonic Frequency (a)100 kHz, (b)50 kHz and (c) without ultrasonic with 100 kV acceleration of electron and 35000 X magnification.

The nano-MH is a good additive for promoting the flame retardancy ability of the nanocomposite. However, the distribution of the nano-MH is an important parameter that affects the final properties of the composite. A good distribution can be achieved by various methods, with the simplest method which is ultrasonication [11]. Therefore, ultrasonic assisted extrusion was conducted on the optimum blending which is 10phr of nano loading tested as shown in Figure 6(a), (b) and (c) with different ultrasonic frequencies. It can be clearly seen that, with 10 phr of nano loading at 100 kHz of ultrasonic frequency is sufficient to show a good distribution of nano-MH that contributes to excellent tensile strength and Youngs modulus properties of the composite which is verified by Transmission Electron Microscopy (TEM) images as seen in Figure 6(a). While for 10 phr nano loading with 50 kHz of ultrasonic frequency, the nanoparticles were still agglomerates in the copolymer matrix as can be seen in Figure 6(b). As shown in Figure 6(c), for 10 phr nano loading without ultrasonic, there was a bundle of agglomeration occurs which can be proven that the ultrasonic assisted is a better way to have a good distribution in nanoparticles [12]. When increased the nano loading, aggregation will start to occurs which leads to weakening of the property of the composite. The optimum properties will be obtained when the dispersion is uniform [13].

4. Conclusion

A rapid intercalation of nano composite inside a matrix was achieved through ultrasonic process. This can be seen in TEM analysis showing a better dispersion of nanoparticles. Enhanced dispersion will theoretically improve the efficiency of nano-MH as flame retardant. Nano-MH loading at 10phr has the highest tensile strength compared to other loadings. The elongation at break decreases as the increase of nano-MH added to the nanocomposite. However, Young Modulus, Flexural Strength and Izod Strength showing reverse result from tensile and elongation. It can be concluded that, 10 phr of nano-MH loading is sufficient in finding a balance of the various mechanical properties studied as well as cost efficiency.

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