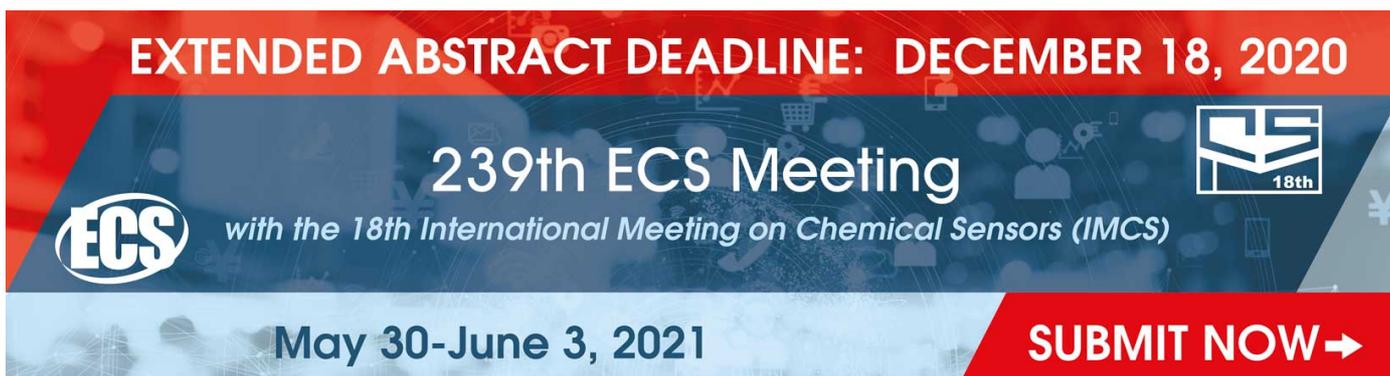


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The Effects Of Phthalic Anhydride On R-Hdpe / Eva / Cff Composites: Tensile And Physical Properties

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Abstract. Phthalic anhydride is one of the compromise coupling agents that enhance the dispersion and hydrophobicity of fiber which consequently improved the interfacial adhesion of composites. This article is concerned with the mechanical properties, morphology, and functional groups towards recycled high density polyethylene (r-HDPE)/ethylene vinyl acetate (EVA)/chicken feather fiber (CFF)/phthalic anhydride (PAH) composites. In this study, the r-HDPE/EVA/CFF/PAH composites with varies loading of phthalic anhydride (2, 4, 6 and 8 phr) were moulded into compression moulding machine and tested using conventional universal testing machine. Morphology and functional groups properties were characterized using field emission microscope (FESEM) and Fourier transform infrared spectroscopy (FTIR), respectively. The results showed that the tensile and physical properties (water absorption and oven aging) of r-HDPE/EVA/CFF/PAH composites were slightly affected by the phthalic anhydride loading.

Keywords: Natural filler reinforced composites, phthalic anhydride, mechanical properties, morphology, functional groups

1. Introduction

Generally, coupling agents comprise bonding agents and surfactants (surface-active agents), including compatibilizers and dispersing agents [1]. Bonding agents act as bridges that link fibers and thermoplastic polymers by one or more of the following mechanisms: covalent bonding, polymer chain entanglement, and strong secondary interactions as in the case of hydrogen bonding [2], [3]. Compatibilizers are used to provide compatibility between otherwise immiscible polymers through reduction of the interfacial tension. Some compatibilizers, such as acetic anhydride and methyl isocyanate, are monofunctional reactants. They lower the surface energy of the fiber, and make it non-polar, more similar to the plastic matrix.



Phthalic anhydride is a cyclic dicarboxylic anhydride that transform from phthalic acid to phthalic anhydride. According to Zhang and Zhang (2015) [4], phthalic anhydride act as compatibilizer that cause better dispersion of cellulose nanocrystal (CNC) and improve the hydrophobicity of CNC which obviously improved in interfacial adhesion and tensile strength of PBSA/CNC composites. Moreover, the result shown the increasing in phthalic anhydride in composites reduced the water uptake of all the composites which is due to the reaction between of phthalic anhydride with the hydroxyl groups of PBSA and CN. Besides that, Hanif and co-researchers (2015) [5], stated that the compatibilized r-HDPE/WF/PAH composites achieved higher tensile strength and modulus of elasticity than uncompatibilized composites. The presence of PAH which enhanced the interaction r-HDPE/WF/PAH composites effects of the stress transfer, hence reduced the chance of interfacial de-bonding and lead to improve their properties.

However, even there were many studies on the effect of coupling agents towards natural fiber reinforced thermoplastic composites, only few studies have been conducted on phthalic anhydride specifically for chicken feather as reinforcement fiber in r-HDPE/EVA/CFF/PAH composites. Hence, in this research, phthalic anhydride has been utilized as compatibilizer in r-HDPE/EVA/CFF/PAH composites. As to achieve the main goal in this research, phthalic anhydride has been compounded together with r-HDPE/EVA/CFF composites with varies ratio. Meanwhile, the morphology and functional groups properties of these composites was characterized using field emission scanning electron microscopy (FESEM) and Fourier transforms infrared (FT-IR), respectively.

2. Experimental Part

2.1 Materials

Recycled high density polyethylene (r-HDPE) and ethylene vinyl acetate (EVA) were the polymer matrix of the composites in this work. Recycled high density polyethylene (r-HDPE) pellets were obtained from Mega Makmur Sdn. Bhd. Penang, Malaysia. The material has a melt flow index of 0.7 g/10 min at 190 °C, a density of 0.948 g/cm³, and melting temperature range from 110 °C to 140 °C with a peak at 131.5 °C with a heating rate of 10 °C/min. The EVA contains 18.1 wt% vinyl acetate (VA), with melt flow index of 2.5 g/ 10 min at 80 °C and a density of 0.925 g/cm³ are used. EVA was obtained from A. R. Alatan Sdn. Bhd., Kedah Darul Aman, Malaysia. The chicken feather was obtained from morning market, Kangar, Perlis. Sodium hydroxide (NaOH with molar mass = 40 g/mol) was supplied by A.R. Alatan Sdn. Bhd., Kedah Darul Aman, Malaysia. Phthalic anhydride (PAH) with chemical formula (C₆H₄(CO)₂O and molar mass=148.12 g/mol was used as compatibilizer and obtained from AR Alatan Sdn. Bhd., Alor Setar, Kedah, Malaysia.

2.2 Alkaline treatment

First, chicken feather was immersed in 1 mol of sodium hydroxide for 24 hours at 25 °C followed by washing with distilled water. The chicken feather was dried in oven at 80 °C for 24 hours. Then, the dried chicken feather was crushed using a laboratory mixer and sieved to obtain average fiber size 0.4 cm to 1 cm.

2.3 Composite preparation

The composites were prepared using z-blade mixer at temperature 180 °C and rotor speed of 30 rpm for 18 minutes until the mixture homogenous Recycled high density polyethylene (r-HDPE) and ethylene vinyl acetate (EVA) were mixed and loaded into mixing chamber for melt mixing. The blend of r-HDPE (70 wt.%) and EVA (30 wt.%) is preheated for 10 minutes then the chicken feather fibers (10 wt.%) only added into the soften mixture together with phthalic anhydride at varies loading (2.0, 4.0, 6.0, and 8.0 phr). The soften composites was discharged from mixing chamber and ready for next process.

2.4 Compression molding

Hydraulic hot press machine model GT 7014A was used to produce the composite in compressed sheet form. The temperature was set as 180 °C for both top and bottom plate. The empty mould was heated for 2 minutes. Then the compounds were put into the mould, preheat for 1 minutes followed by 3 minutes partially compression and fully compression for 2 minutes once the compound started become soften. After compression, the samples were cooled for 4 minutes. The moulded composites then ready for testing.

2.5 Mechanical testing

2.5.1 Tensile testing

The composites plate was cut into dumbbell shape with 50 mm length and 4 mm width by using Wallace die cutter for tensile testing. The Shimadzu Tensile Testing Machine model AG-XD plus was used for tensile test. According to ASTM D638, the crosshead speed is set as 50 mm/min. Five dumbbell shaped samples for each formulation were tested. The tensile strength, modulus of elasticity and elongation at break of samples were obtained from this tensile test.

2.6 Characterization

2.6.1 Morphology

The morphology of the tensile fracture on the samples were observed by using field emission scanning electron microscope (FESEM) model Supra 35-VP at School of Material and Mineral Resources, USM Engineering Campus. First, the fracture surface was mounted on aluminum stage to undergo sputtering coating. The samples were coated with 20 nm thin layer gold by using Auto Fine Coater and then observed under microscope.

2.6.2 Functional groups

Fourier transform infrared spectroscopy (FTIR) characterization was used to characterize the chemical structure or functional group of the composites. The spectra were obtained from Perkin-Elmer Spectrum One Series equipment. The sample was cut into small rectangular shape. The scanning range for selected spectrum resolution and wavelength are 4 cm⁻¹ and 600-4000 cm⁻¹.

2.7 Physical testing

2.7.1 Water absorption

Three samples with dimension (20 x 10 x 1) mm for each formulation were used for water absorption test according to ASTM D570 - 98. First, the samples were weighted and totally immersed in distilled water for two months until the weight of samples were constant. After immersion, all samples were removed and wiped by filler paper to remove surface water. The amount of water absorbed was measured every week for two months. The final weight of samples was observed with 0.1 mg precision. The calculation for the percentage of water absorption is shown below:

$$\% \text{ Mass absorb} = \frac{W_2 - W_1}{W_1} \times 100\% \quad (1)$$

Where W_1 is the weight of dried sample and W_2 is the weight of the wet sample.

2.7.2 Oven aging

Five samples with dimension (20 x 10 x 1) mm for each formulation were used for oven aging test according to ASTM D3045 - 92. First, the samples were weighted and dried in oven at 80 °C for 48 hours. After that, all samples were removed and measured. The final weight of samples was observed with 0.1 mg precision. The calculation for the percentage of water retention is shown below:

$$\% \text{ Mass retention} = \frac{W_1 - W_2}{W_2} \times 100\% \quad (2)$$

Where W1 is the original weight of sample and W2 is the final weight of the sample.

3. Results and discussion

3.1 Tensile properties

Figure 1 exhibited the effect of PAH on the tensile strength of r-HDPE/EVA/CFF/PAH composites. The r-HDPE/EVA/CFF/PAH composites with 4 phr PAH loading demonstrated the highest tensile strength among all the composites due to good interfacial adhesion between r-HDPE/EVA matrix. The PAH acts as compatibilizer agent which promoted the compatibility of r-HDPE/EVA/CFF composites and enhanced the mechanical strength of compatibilized composites. This proved that PAH with 4 phr is the optimum ratio to achieve its function as compatibilizer. However, PAH was overloaded beyond optimum value deteriorated the tensile strength of composites. This mainly caused by the high PAH content which caused agglomeration of PAH in the composites promoted the stress concentration that weakened the polymer matrix -filler interaction and eventually broke fairly when stress was applied and reduction in tensile strength. The interfacial adhesion between r-HDPE and EVA are improved by carbonyl group in PAH that reacted with the ester group of EVA to form covalent bonds to strengthen the stress transmit from r-HDPE to EVA matrix [6]. PAH acts as compatibilizer that cause better dispersion of cellulose nanocrystal (CNC) and improved the hydrophobicity of CNC which obviously improvement of the mechanical properties of PBSA/CNC composites [4].

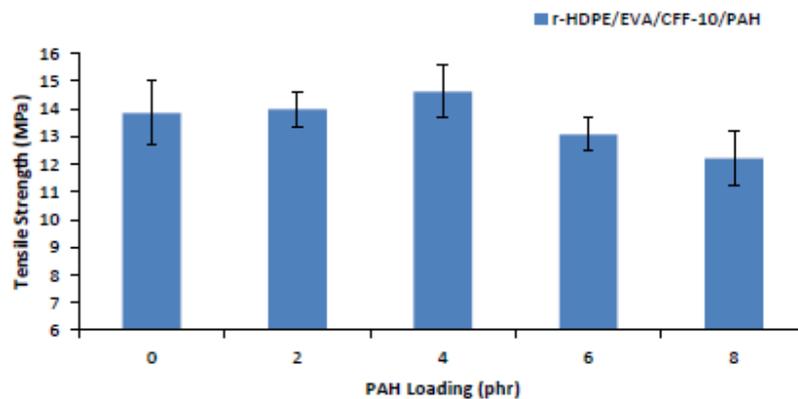


Figure 1. Effect of PAH loading on the tensile strength of r-HDPE/EVA/CFF composites

3.2 Modulus elasticity

Figure 2 exhibited the effect of PAH loading in the modulus of elasticity of r-HDPE/EVA/CFF composites. The result presented the compatibilized composites with 4 phr phthalic anhydride achieved the highest young modulus among all the r-HDPE/EVA/CFF/PAH composites. The interfacial tension between r-HDPE and EVA had decreased by the addition of PAH as compatibilizer to enhance the compatibility, stiffness and young modulus of r-HDPE/EVA/CFF composites. According to Hanif and co-researchers (2015), they revealed that r-HDPE/WF/PAH composites achieved higher tensile properties than uncompatibilized composites due to the presence of PAH which enhanced the interaction between the r-HDPE and wood fiber and effects of the stress transfer, hence reduced the chance of interfacial debonding and led to improve their properties. Supri and co-researchers (2015) [7], revealed that PE-g-MAH

transformed the surface of r-HDPE/EVA blends to become rough surface which indicated that PE-g-MAH linked the interface between r-HDPE and EVA polymers and lowered the interfacial tension then lead to modulus of elasticity. Akhlaghi and co-researchers (2012) [8] investigated the existence of benzyl urea (BU) improved the interaction of PE/EVA matrix with filler due to BU acts as compatibilizer which improved the interface of two polymers.

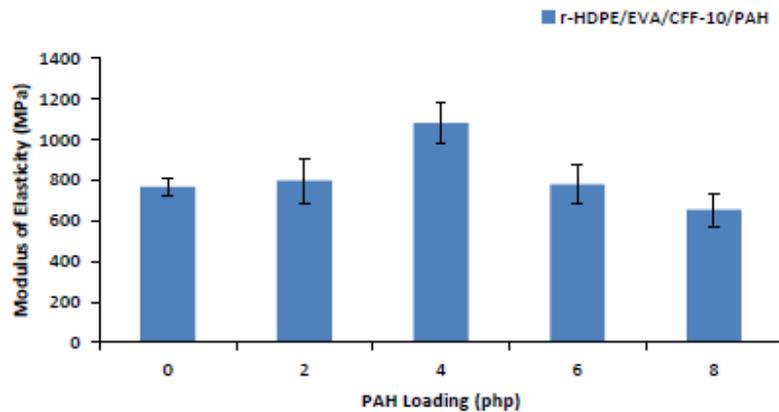


Figure 2. Effect of PAH loading on the modulus of elasticity of r-HDPE/EVA/CFF composites

3.3 Elongation at break

Figure 3 presented the effect of PAH loading on the elongation at break of r-HDPE/EVA/CFF/PAH composites. As the PAH loading increased in r-HDPE/EVA/CFF/PAH composites, the elongation of break of composites decreased gradually. The interfacial tension between r-HDPE and EVA had decreased by the addition of PAH as compatibilizer to enhance the compatibility and stiffness a of r-HDPE/EVA/CFF composites. This caused decrement in ductility of composites and eventually led to reduction in toughness and elongation at break of r-HDPE/EVA/CFF composites. However, the PAH content increases beyond the optimum value obviously increased in the elongation of break of composite. The agglomeration of PAH in the composites significantly reduced the strain hardening and enhanced the elongation at break of the composites [9-11].

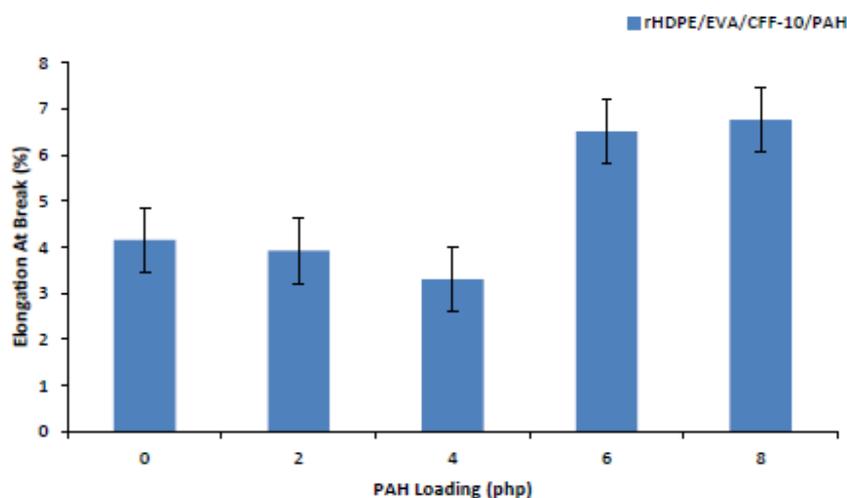


Figure 3. Effect of PAH loading on the elongation at break of r-HDPE/EVA/CFF composites

3.4 Morphology analysis

The scanning electron microscopic morphology of tensile fractured surface of r-HDPE/EVA/CFF and r-HDPE/EVA/CFF/PAH composites were examined in Figure 4. Scanning electron microscopic micrograph of r-HDPE/EVA/CFF/PAH composites in Figure 4 b illustrated many tear lines on the surface of composites and rougher surface than r-HDPE/EVA/CFF composites in Figure 4 a which proved that the composites has strong interfacial adhesion between the interface of matrix and fiber with the presence of phthalic anhydride as compatibilizer. This morphology analysis corresponded to the high tensile strength in Figure 1 due to compatibilized composites exhibit strong polymer matrix -filler interaction when stress was applied. However, the agglomeration can be clearly seen in Figure 4 c indicated that PAH content was overloaded which developed as stress concentration that reduced the fiber/matrix interaction and reduction in tensile strength and young modulus. The rough surface and better distribution of filler in r-HDPE/EVA/CaCO₃ composites which proved that benzyl urea as compatibilizer that reduced the interfacial tension and enhanced the interfacial adhesion of r-HDPE/EVA matrix and CaCO₃ fiber [7].

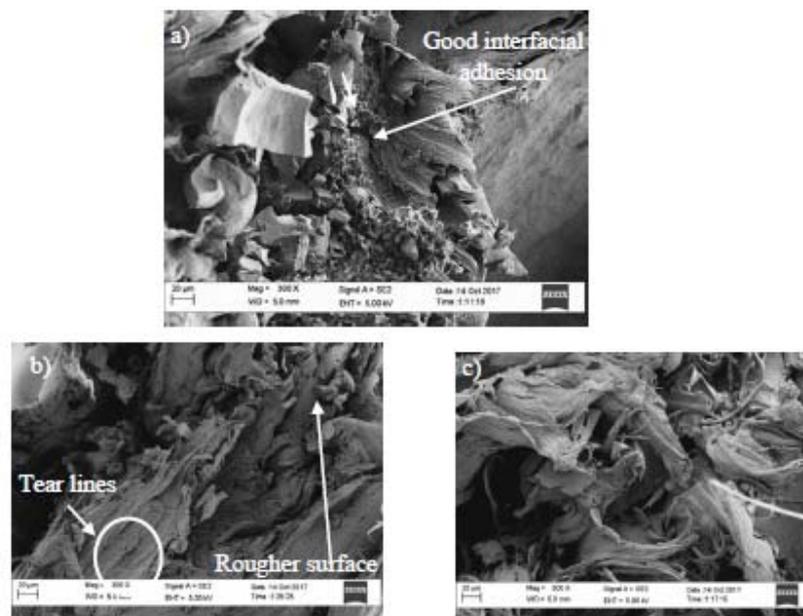


Figure 4. Scanning Electron Microscopy micrographs of tensile fractured surfaces of a) r-HDPE/EVA/CFF-10 composite b) r-HDPE/EVA/CFF/PAH-4 composite c) r-HDPE/EVA/CFF/PAH-8 composite

3.5 Functional group analysis

Figure 5 displayed the infrared spectrum of a) r-HDPE, b) r-HDPE/EVA blends and c) r-HDPE/EVA/CFF composites and d) r-HDPE/EVA/CFF/PAH composites. The intensity at the 1738.53 cm⁻¹ and 1239.45 cm⁻¹ were increased which indicated that the C=O carbonyl group and O-C ester group content increased in r-HDPE/EVA/CFF/PAH composites compared to r-HDPE/EVA/CFF composites. This indicated that there was some interaction between the ester group on the surface of EVA and anhydride group (-CO-O-OC-) on PAH. The intensity of peaks at 2914.94, 2848.10, 1464.68, and 718.09 cm⁻¹ were increased which indicated that the CH₂ stretching increased in r-HDPE/EVA/CFF/PAH composites compared to r-HDPE/EVA/CFF composites. This was due to the methyl group was formed from the chemical reaction

between EVA and PAH. The mechanism of chemical reaction between r-HDPE and EVA matrix with PAH and r-HDPE, EVA matrix and CFF fiber with PAH were shown in Figure 6 and Figure 7, respectively.

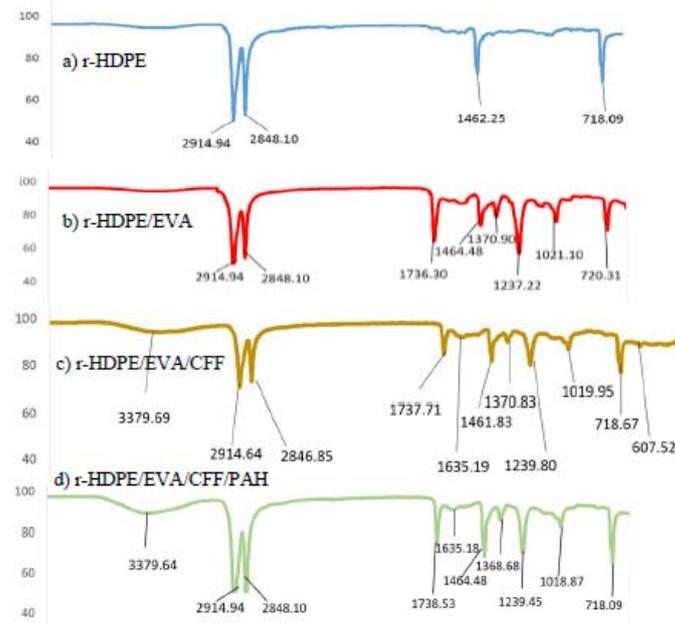


Figure 5. The FTIR spectra of a) r-HDPE b) r-HDPE/EVA blends c) r-HDPE/EVA/CFF composites d) r-HDPE/EVA/CFF/PAH composites

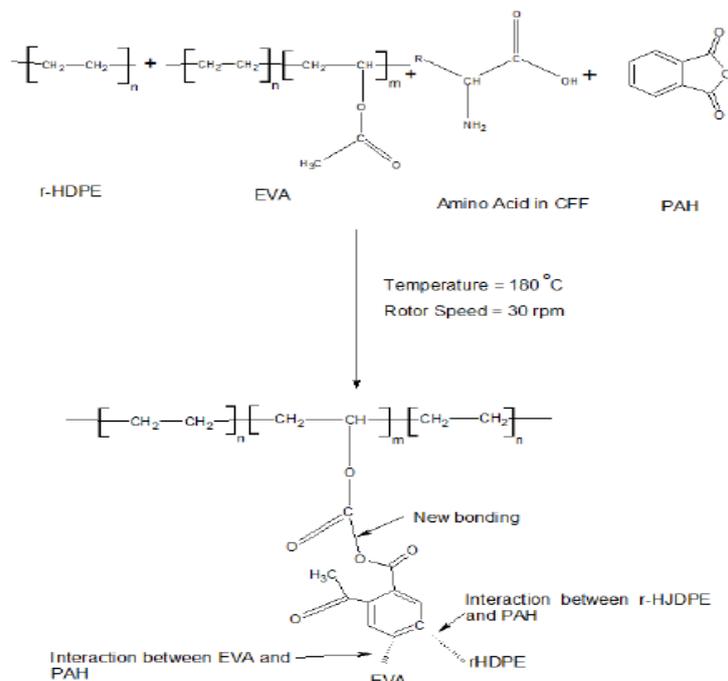


Figure 6. The mechanism of chemical reaction between r-HDPE and EVA matrix with PAH

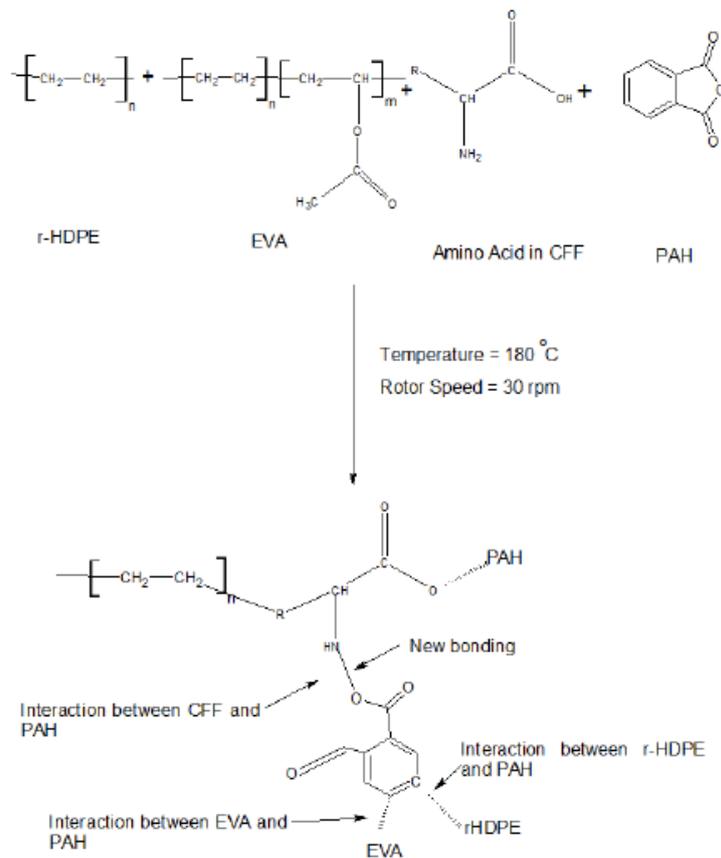


Figure 7. The mechanism of chemical reaction between r-HDPE, EVA matrix and CFF fiber with PAH

3.6 Weight loss analysis

Figure 8 demonstrated the oven aging properties of r-HDPE/EVA/CFF/PAH composites. The weight loss achieved the lowest percentage at 4 phr PAH in r-HDPE/EVA/CFF/PAH composites. As the PAH is added in the composites, the rougher surface than r-HDPE/EVA/CFF composites which proved that the composites has strong interfacial adhesion between the interface of matrix and fiber with the presence of phthalic anhydride as compatibilizer which can be observed in Figure 8 b. This hindered the moisture content in r-HDPE/EVA/CFF/PAH composites. Thus, this prevent the oxidation and the evaporation of moisture in compatibilized r-HDPE/EVA/CFF composites and lower the weight loss during elevated temperature.

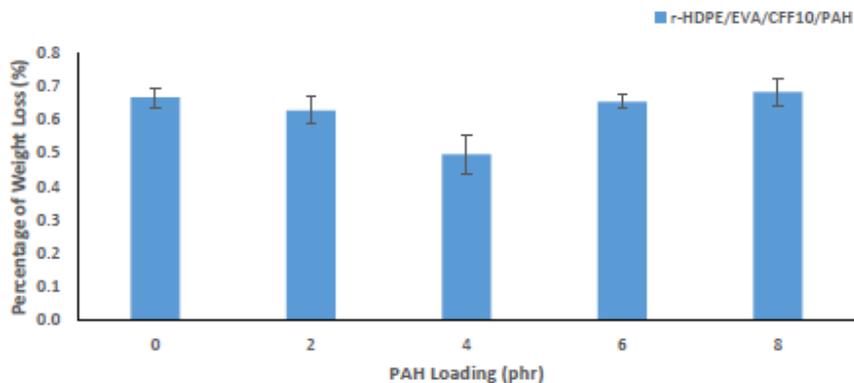


Figure 8. Effect of PAH loading on the weight loss properties of r-HDPE/EVA/CFF composites

3.7 Water absorption analysis

The percentage of water absorption of r-HDPE/EVA/CFF/PAH composites with different amount of phthalic anhydride as compatibilizer was demonstrated in Figure 9. The results revealed that the percentage water absorption of compatibilized composites was lower than uncompatibilized composites. The saturation point for water absorption was highest in uncompatibilized composites and decreased as phthalic anhydride increased in the composites. This indicated good interfacial adhesion in compatibilized composites reduced the interfacial voids in composites which prevent the water accumulation in the voids. Moreover, the result shown the increasing in phthalic anhydride in composites reduced the water uptake of all the composites which is due to the reaction between of phthalic anhydride with the hydroxyl groups of PBSA and CN [4]. Supri and co-researchers (2015) proved that r-HDPE/EVA/TP/PE-g-MAH composites has lower water absorption ability compared to r-HDPE/EVA/TP composites due to PE-g-MAH compatibilized the composites that restricted the water uptake into the composites.

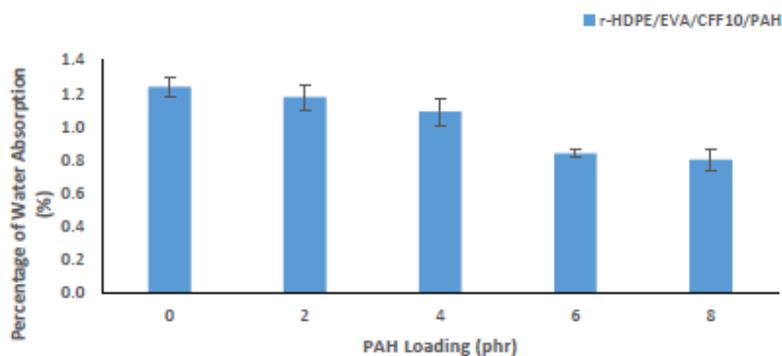


Figure 9. Effect of PAH loading on the water absorption properties of r-HDPE/EVA/CFF composites

4. Conclusion

1. The mechanical performance such as tensile strength, tensile modulus, and elongation at break of r-HDPE/EVA/CFF/PAH composites were slightly increased with increasing phthalic anhydride (PAH) loading up to 4 phr.
2. The addition of PAH in r-HDPE/EVA/CFF composites was proved to enhance the compatibility and adhesion between chicken feather and r-HDPE matrix, where all the studied properties show positive increment as the PAH loading increase.

3. Phthalic anhydride will be an alternative coupling agent towards natural fiber reinforced composites in the future as it is successfully improving the performance of the studied composites by enhance the compatibility between fiber and polymer matrix specifically for thermoplastic polymer.

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