

Influence of MA-g-HDPE Compatibilizer Content on Quality and Adhesion/Bending Strength of Polyethylene/O-MMT Coating Films

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ABSTRACT: High density polyethylene/organo-modified montmorillonite composites with various concentrations of maleic anhydride grafted high density polyethylene (MA-g-HDPE) as compatibilizer (5–20 wt %) have been prepared by melt process. The extruded composite powders are applied on the treated steel surfaces using spray electrostatic powder technique, followed by oven curing at various temperatures (180°C–220°C) and times (15–45 min). The surface uniformity of produced coating films is studied by scanning electron microscopy. Comparison of micrographs of the coatings shows the composite coating films are measured using standard methods. The uniformity, adhesion, and bending strength of the coating films are compared to select high performance coatings. The results indicate that the presence of 15 wt % MA-g-HDPE in the coatings shows the highest properties (adhesion and bending strength) and more surface uniformity. © 2014 Wiley Periodicals, Inc. *J. Appl. Polym. Sci.* **2014**, *131*, 40926.

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INTRODUCTION

Solvent-based organic coatings are formed on substrates (metal, wood, plastic, . . .) by the application of liquid coaters at ambient temperature. During the drying process, solvents are released from such coatings which pollute the environment.¹ The need to reduce the amounts of volatile organic compound emitted into the atmosphere brings about the increasing use of powder coating as an alternative to the solvent coating technology.²

The polymer powder coatings market is growing considerably because it is environmentally friendly, economical, safe, and energy-saving.³ Powder coatings are usually applied by spray electrostatic, flame spraying, and fluidized-bed techniques. The first technique is one of the most widespread methods of powder coating. The electrostatic powder coating process is based on a very simple principle.⁴ This is to charge powder particles in an intense electric field generated by high voltages. Due to a similar polarity, charged particles repel each other, forming a cloud across the coating area.⁵ Particle size, spraying process, and curing conditions are critical variables that can influence uniformity of the continuous coating formation, mechanical properties, and performance of the coatings.^{5,6}

Polyolefins are among the common materials used for coating processes.^{7,8} Polyethylene is probably the most widely protective coating for steel substrates due to its excellent mechanical and

thermal properties, barrier against moisture, chemical and biological resistance, convenient process ability, and low cost.⁹ However, low surface energy, non-polar character, and the lack of reactive sites significantly limit its use in applications requiring good surface/interface properties. Blending of polyethylene with polar polymers and chemical modification of polyethylene have been carried out to improve the coating quality and reduce its defects.^{10–13} Experimental results have shown that olefin polymers containing polar functional groups in their composition can have sufficient adhesion strength to the steel surface.¹⁴ The protective properties of such thermoplastic coatings can also be enhanced by adjusting coating thickness and adding nano fillers to form particulate polymer composites.^{1,15,16} In the past decade, the literature has been shown many works which evaluate the use of layered silicate in the synthesis of a variety of particulate polyethylene composites. The formation of the particulate polyethylene composite requires the dispersion of the layered silicate and distribution of the silicate layers throughout the polyethylene matrix. Silicate layers are naturally hydrophilic while polyethylene is hydrophobic; thus, the surface energies between the two materials can be vastly different, prohibiting any significant degree of dispersion of silicate layers within the polyethylene matrix.³ It was found that the use of layered silicate with high hydrophilicity and the addition of polyethylene grafted with functional groups to the polyethylene matrix is a viable approach to the synthesis of a particulate

polyethylene composite. Recent studies have shown that by using MA-g-PE in the blends of polyethylene and modifying layered silicate, the compatibility, dispersion of silicate layers in the polyethylene matrix and subsequently, mechanical properties can be improved.^{17,18} The mechanism of improving dispersion of layered silicate within the polyethylene matrix indicates that MAH-g-PE is known to interact well with the layered silicate surface and also the polyethylene and MAH-g-PE interact well; then MAH-g-PE intercalation into the inter layers space of silicate facilitates further intercalation by polyethylene chains. This means that a considerable amount of polyethylene chains can diffuse and intercalate into the inter layers of silicate, resulting in maximum interfacial interaction between MAH-g-PE and silicate layers for the intercalated morphology.¹⁹ Application and properties of polyethylene/organo-modified montmorillonite (O-MMT) composites as coating materials on steel surface using spray electrostatic powder technique have not been reported; so it is the subject of the present study.

The aim of this article is to prepare high performance uniform HDPE/maleic anhydride grafted high density polyethylene (MA-g-HDPE)/CL15A composite coatings on carbon steel surface with sufficient adhesion/bending strength using spray electrostatic powder technique.

EXPERIMENTAL

Materials

High density polyethylene (HDPE) [HD-I3, density = 0.957 g/cm³, MFI = 8 g/10 min (190°C, 2.16 kg)] was supplied by Iranian petrochemical Complex. MA-g-HDPE as compatibilizer (Karabond[®]PEC) was obtained from Karanegin in Iran (density = 0.960 g/cm³, MFI = 1.3 g/10 min at 190°C, 2.16 kg and maleic anhydride content of 1 wt %). The particle size of HDPE powders is 125 μm and that of MA-g-HDPE is 105 μm. Commercial silicate layer (Cloisite[®]15A) was supplied by Southern Clay Products (CL15A). The average particle size of silicate (CL15A) powders is 2–13 μm. All chemicals were in the form of powder and used as received without further purification. The carbon steel plates (st37) of size 100 mm × 50 mm × 1 mm were supplied by Isfahan Steel Plant (Isfahan, Iran).

Composites Preparation

HDPE/MA-g-HDPE/CL15A composites were prepared by melt mixing of the components in two steps. In the first step, two master batches of MA-g-HDPE/CL15A (75/25 wt %) and HDPE/CL15A (90/10 wt %) were produced separately in a co-rotating twin screw extruder (Nanjing Shengchi, SHJ20, $D = 25$, $L/D = 32$). The temperature profile employed was 160°C–180°C (zone 2–zone 5) in the barrel of the extruder and 180°C at the die, at a screw velocity of 200 rpm, with a mass flow rate of 2 kg/h. In the second step, the calculated amount master batches and pure HDPE were melt mixed extruded with a temperature profile between 170°C–190°C (zone 2–zone 5) in the barrel of the extruder and 190°C at the die, screw velocity of 250 rpm and a mass flow rate of 2.5 kg/h. The prepared polyethylene composite samples were performed using the PC_X code. PC indicates polyethylene composite and X is the weight percentage of MA-g-HDPE in the composite ($X = 0, 5, 10, 15, \text{ and } 20$). The CL15A concentration in all samples was kept constant at 4 wt

%. The extruded composites were powdered with a grinder (Moulinex DPA1) and sieved through a 100 μm sieve. Before the extrusion process was carried out, MA-g-HDPE and CL15A were dried at 80°C for 24 h to remove any absorbed water. The CL15A powder was dry blended with MA-g-HDPE powder and then fed into the hopper of the extruder.

Coating Sample Preparation

To remove all surface oxides, debris, and greases, primary mechanical and chemical surface treatment was performed. The carbon steel surfaces were subjected to shot pinning as well as polishing on 60 grit size sand papers (using mechanical polishing equipment). After mechanical roughening, the following surface chemical cleaning was performed: (1) immersing in 50°C degreasing alkaline solution for 5 min; (2) washing in distilled water; (3) immersing in 5 vol % of sulfuric acid solution, at 40°C for 1 min; and (4) washing with plenty of distilled water and then drying by hot air. After surface preparations, the prepared surfaces were preheated at 100°C for 1 min to remove any trace of water molecules on the surface. Then, the polyethylene composite powders were sprayed electrostatically onto the metal substrate with a spraying gun at 60 kV and 1 bar air pressure (distance between gun and work piece is approximately 25 cm), and placed vertically in a vacuum oven (Heraeus LUT 6050) at 200°C for 30 min. The coated films were air-cooled at room temperature. The coating films with a thickness of 370 ± 5 μm were obtained and used for analysis. The obtained coatings were coded as PC_{XC}. Index XC indicates coatings with X wt % MA-g-HDPE ($X = 0, 5, 10, 15, \text{ and } 20$).

Measurements

Determination of Surface Morphology. The surface morphology of the prepared coatings was studied by scanning electron microscopy (SEM; Phillips Company, model XL30). After coating sample (10 mm × 10 mm × 1 mm) preparation in vacuum oven at 200°C for 30 min, the samples were coated with Au to increase their conductivity by a Bal-Tec SCD 004 sputter coater. The sputter coating was performed over a period of 1 min with cycles that involved a 4-s coating time and a 4-s resting time. Accelerating voltages of 15–20 kV were used during SEM observations. Optical microscope (Canon model IXUS 300HS) was also used to inspect the modes of failure of polyethylene composite coatings after carrying out the bending test.

Adhesion and Bending Strength Tests. Adhesion and bending strength tests were conducted according to ASTM-D4541²⁰ (pull-off) and ASTM-D522²¹ (cylindrical mandrel bend), respectively. A Hounsfield tensile testing instrument (Model-h50ks) was used for the pull-off method to measure the lift force required to pull a small area of coating film away from the base carbon steel. The composite powders were applied onto steel dollies and sheets. The metal surfaces coated with a layer thickness of 370 ± 5 μm were air-cooled at room temperature. Steel dollies and sheets were affixed by an adhesive against each other. The dollies and sheets were attached to the upper and lower jaw of the tensile test machine. Pull-off strength was measured at an extension rate of 50 mm/min. For adhesion strength test, the average and the standard deviation were calculated from at least five samples, and data points greater than

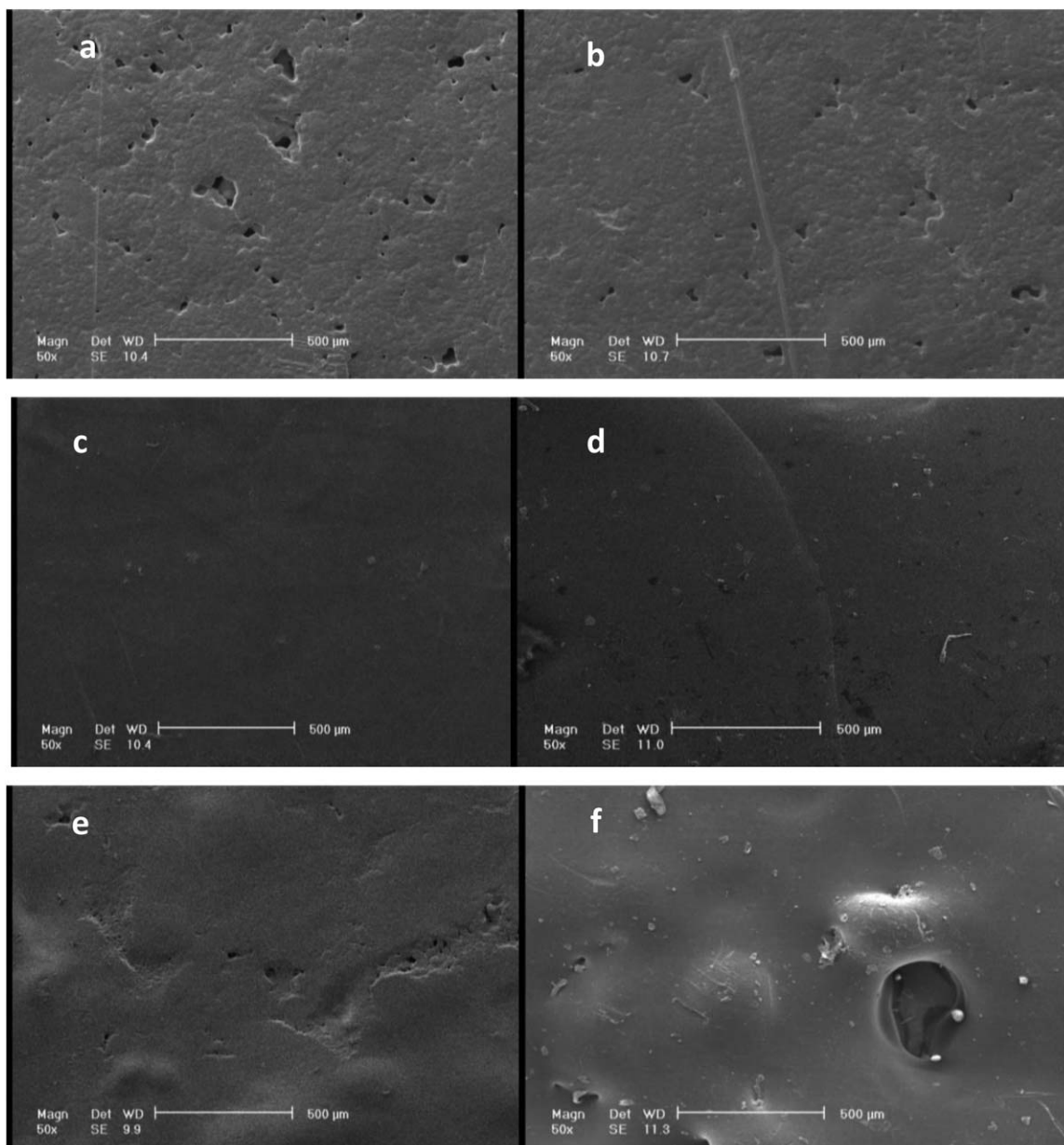


Figure 1. SEM micrographs of the PC₂₀ sample at various curing times/temperatures : (a) 180°C, 30 min; (b) 180°C, 45 min; (c) 200°C, 30 min; (d) 200°C, 45 min; (e) 220°C, 30 min; (f) 220°C, 45 min.

two standard deviations from the mean were removed. Cylindrical mandrel bend test describes the ductility of coating films on carbon steel substrates. The test plane was placed over a mandrel (diameter 9.5 mm) with uncoated side. A steady pressure of the fingers was applied so that, the plane could be bended approximately 90° and 180° around the mandrel at a speed of 1.3 mm/min. The surface of the film was observed to that whether cracking and discoloration have occurred. Typically five samples were used for each determination.

Composite Characterization. X-ray diffraction. X-ray diffraction was performed at room temperature by a X-Pert Pro Philips diffractometer (40 kV, 30 mA) using the Cu $k\alpha$ radiation at

the rate of 1° min⁻¹, in the range from 2° to 10°, in order to determine the extent of intercalation in the composites.

Gel content determination. The gel content of polyethylene composites was determined according to ASTM-D2765²² by an extraction of polyethylene matrixes from the polyethylene composites using hot solvent (xylene) in a soxhlet extractor. Approximately, 1.5 g of extruded composite was weighted and placed in a steel pouch. The steel pouch was immersed in boiling xylene (132°C) for 24 h under N₂. Then, the extract specimen was completely dried in a vacuum oven at 80°C for 24 h and reweighed. At least five measurements for each sample were done.

Melt behavior. The melt behavior of polyethylene composites was studied by a melt flow index tester using a melt flow

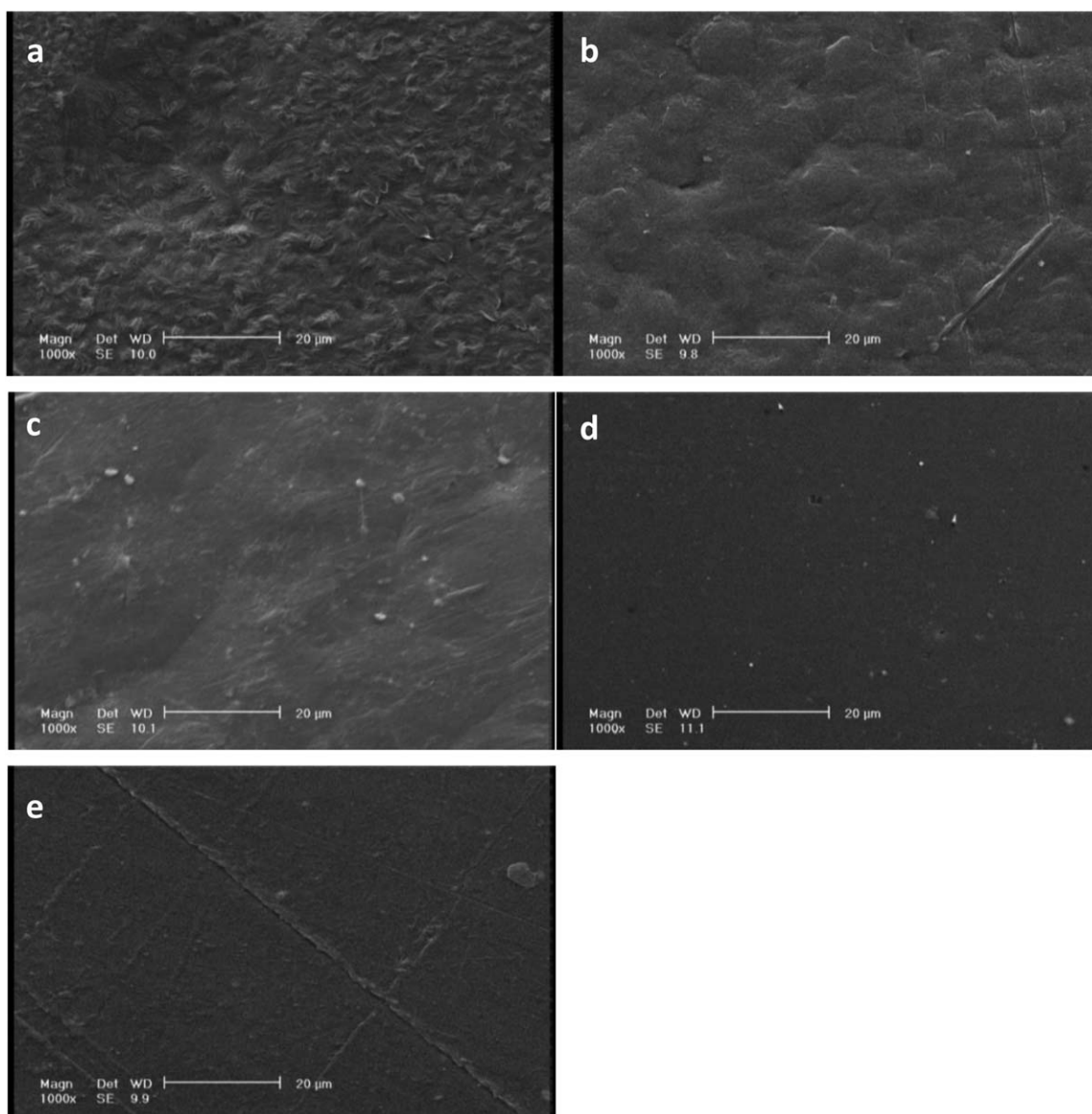


Figure 2. SEM images of the coating samples cured at 200°C for 30 min: (a) PC_{0c}; (b) PC_{5c}; (c) PC_{10c}; (d) PC_{15c}; (e) PC_{20c}.

Indexer according to the ASTM-D1238.²³ The average of five measurements was taken as the value for each sample.

RESULTS AND DISCUSSION

Quality of the Coatings

Figure 1(a–f) shows the SEM micrographs of the PC_{20c} coating sample with the highest melt viscosity cured versus temperatures and times. The effect of curing time and temperature on the uniformity of the coating film (PC_{20c}) was observed. Determination of these two parameters is important, because the quality and uniformity of the coating films on carbon steel surface depend on them.⁶ The best selection of both curing time and temperature was done by comparison of the SEM images of the coating sample cured at different times and temperatures. SEM micrograph in Figure 1a shows that at curing time/temperature of 30 min/180°C, a nonuniform coating is obtained.

The residence time at 180°C of the particles in the oven is increased to 45 min (Figure 1b), indicating that no acceptable quality of coating is obtained. It has been found that the density and size of coating voids could be decreased as time and/or temperature of curing are increased.⁶ SEM micrographs of coatings cured at 200°C and 220°C for various periods of times (30 min and 45 min) are shown in Figure 1(c–f). From Figure 1(c,d), it is seen that at curing times of 30 min and 45 min and curing temperature of 200°C, the density and size of voids on the coatings surface are decreased and their uniformity is increased. At curing times of 30 min and 45 min and curing temperature of 220°C, blistering of the coating films is observed (Figure 1e, f), which may be due to thermal oxidative degradation. Thus, the comparative analysis of obtained results demonstrates that the best quality coating is obtained at a curing time of 30 min and curing temperature of 200°C.

Table I. Melt Flow Index of the Polyethylene Compounds Extraction of the Composite-Coating Films

Measured properties	Samples				
	PC ₀	PC ₅	PC ₁₀	PC ₁₅	PC ₂₀
Melt flow index (g/10 min) (190°C, 2.16 kg)	7.2	6.2	4	2.5	1.48
Extraction (wt %)	100	98.1	96.8	94.9	94.8

The surface morphologies of the coating samples containing different concentrations of MA-g-HDPE are shown in Figure 2(a–e). The coatings were cured at 200°C for 30 min with the thickness of about $370 \pm 5 \mu\text{m}$. From Figure 2a, it can be clearly seen that the surface of coating (PC_{0c}) is completely non-uniform, revealing the presence of clusters formed by the CL15A particles. With the incorporation of 5 wt % MA-g-HDPE in the polyethylene composite system, the surface of coating (PC_{5c}) is uniform, as compared with the surface of PC_{0c} coating sample (Figure 2b). This can be attributed to deagglomeration of CL15A particles and distribution of the silicate layers throughout the polyethylene matrix. Figure 2c, clearly shows that the surface coating is free of heterogeneity and the uniformity of the coating is more distinct in PC_{10c} sample than PC_{5c} sample. From the Figure 2d, we can see that the surface of sample PC_{15c} is more uniform than other samples. This indicates that the surface roughness of the polyethylene composite coatings is lowered by the increase of the MA-g-HDPE content and the uniformity of polymer coatings is slightly improved. The uniformity of the surface of the sample PC_{20c} (Figure 2e) is slightly less than sample PC_{15c}. This may be due to the covering of the MA-g-HDPE on CL15A surface, which leads to a decrease in the dispersion of the CL15A layers in the polyethylene matrix. The comparative analysis of obtained surface micrographs demonstrates that the better quality coating is obtained for sample PC_{15c}. The results indicate that at curing time/temperature of 30 min/200°C, the increase of the MA-g-HDPE content has strong effects on improving the uniformity and decreasing the heterogeneity on the coating surface.

The MFI data of the polyethylene composite samples in Table I reveal that an increase in the melt viscosity during curing is achieved with the increase in MA-g-HDPE content. This is attributed to the formation of chemical interactions between the functional groups of MA-g-HDPE chains and hydroxyl groups of silicate layers.^{19,24,25} The formation of covalent bonds between the functional groups of MA-g-HDPE and hydrophilic groups of CL15A layers can be confirmed by the gel content of the composite samples shown in the Table I. Esterification reaction between hydroxyl groups in CL15A and the succinic anhydride groups of MA-g-HDPE has also been reported.^{26,27} As can be seen, the melt viscosity of the all composite samples, due to the high wt.% of polyethylene and the presence of CL15A in the polymer matrix, is sufficiently high (Table I). This can bring about enough coating strength on the carbone steel surfaces.

Adhesion and Bending Strength of the Coating

Table II shows the pull-off adhesion strength and mode of failure of polyethylene composite coating films with different MA-

Table II. Pull-off Adhesion Strength and Mode of Failure of Pure Polyethylene and Polyethylene Composite-Coating Films with Different MA-g-HDPE Content Produced on Plain Carbon Steel

Samples	Adhesion strength (KPa)	Mode of failure
HDPE	176.4	Cohesive
PC _{0c}	179.6	Cohesive
PC _{5c}	280.6	Adhesion/cohesive
PC _{10c}	411.3	Adhesion/cohesive
PC _{15c}	583.5	Adhesion/cohesive
PC _{20c}	556.7	Adhesion/cohesive

g-HDPE content, as compared with pure polyethylene coating. It is seen that the incorporation of CL15A particles in the polyethylene matrix and the adhesion strength of polyethylene composite coating sample (PC_{0c}) are the same as those of the pure polyethylene, ($\sim 180 \text{ kPa}$). It can be clearly seen that the adhesion strength of the polyethylene composite coating compatibilized with 5 wt % MA-g-HDPE (sample PC_{5c}) is higher than that of this sample (i.e., PC_{0c}). Polarity of MA-g-HDPE, improves the coating adhesion on the metal substrate.¹² Incorporation of 10 wt % MA-g-HDPE in the polyethylene composite coating (PC_{10c}) increases adhesion strength from 280.6 to 411.3 kPa. From Table II, it can be observed that PC_{15c} coating has the highest adhesion strength (583.5 kPa), as compared to other coating samples. The results indicate that adhesion of the composite coating films is increased with the increase of MA-g-HDPE content up to 15 wt %. The increase in adhesive strength with the increase in MA-g-HDPE content could be explained by available polar groups and the polar–polar interactions between the polyethylene composite coating films and metal substrate surface. Visual observations of the samples show cohesive failure in pure polyethylene and non-compatibilized composite sample (PC_{0c}). Here, the coating film is completely intact to the metal surface but there is no residual of polymer on the other metal surface. Adhesion/cohesive modes of failure are observed in the case of PC_{5c}, PC_{10c}, PC_{15c}, and PC_{20c}, where residual polymer coating is left on both substrate surfaces. This can be attributed to the competition between cohesive/adhesive properties within the polymer blend, where polyethylene provides tensile strength and MA-g-HDPE provides the functional groups necessary for polymer/metal adhesion.

Figure 3 shows the optical macroscopic graphs of polyethylene composite coating samples (PC_{0c}, PC_{5c}, PC_{10c}, PC_{15c}, and PC_{20c}) bended at 90° and 180°. Inspection by visual observation shows the differences between the coatings damages that occurred on the samples. Bending of the PC_{0c} and PC_{5c} samples at both 90° and 180° bend illustrates crack formation at various sizes. Less damage is observed on the PC_{10c} sample surface. However, bending of the PC_{15c}, PC_{20c} samples shows no damage on their surface. The results indicate that PC_{15c}, PC_{20c} samples are more flexible/ductile and they can release energy. It is reported that under a strain bend, failure (detachment of the filler particles from the polymer matrix) occurs in close proximity to the filler



Figure 3. Macroscopic graphs of the coating samples each bended at 90° and 180° (the code of each sample is left side its photograph). [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

particles and more cracks at the interface of the polymer and filler particles are evolved.²⁸ Many particles may agglomerate in these samples. The agglomerated particles will act as weak points in the obtained coating films and cause the fracture to occur at lower stresses.

XRD Patterns

Typical XRD patterns of the polyethylene composite samples with and without a compatibilizer (MA-g-HDPE) as well as pristine CL15A are shown in Figure 4. It can be observed that the XRD curve of pristine CL15A displays two main peaks, at 3.2° (≈ 2.7 nm) and 7.02° (≈ 1.2 nm). In the case of the polyethylene composite sample without compatibilizer (PC₀), two peaks are also

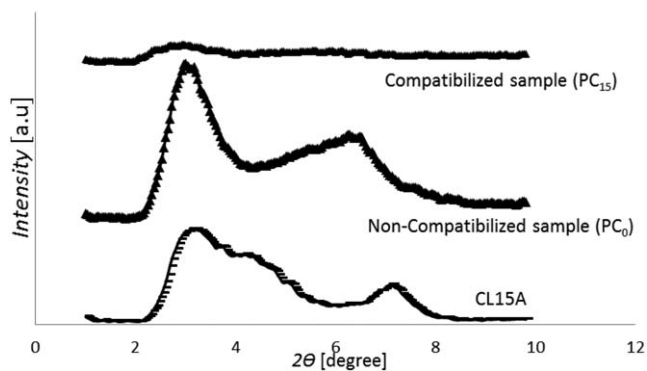


Figure 4. XRD patterns of CL15A and polyethylene composite samples with and without a compatibilizer (MA-g-HDPE).

observed. The main peak is at 3.18°, corresponding to a basal space of 2.7 nm and the second peak is at 5.6°, corresponding to a basal space of 1.5 nm. It can be seen that in the polyethylene composite sample without compatibilizer (PC₀), there is no indication that the main peak is dislocated. It is shown that polyethylene chains do not cause intercalation on pristine CL15A layers in non-compatibilized composite sample (i.e., a microcomposite is formed). It can be observed that in the polyethylene composite sample with compatibilizer (PC₁₅), the second peak was completely eliminated and the main peak value of 2θ was 2.8° (3.08 nm), which is higher than that of the CL15A (2.7 nm). This implies that intercalation of CL15A layers occurs. It can be seen that the CL15A layer spacing is also increased as the compatibilizer (MA-g-HDPE) content is increased.²⁹ In a similar work, Lee et al.³⁰ investigated the preparation of HDPE/MAH-g-HDPE/CL15A nanocomposites by melt processing. They reported that the interlayer space of CL15A was increased to 2.8 nm.

CONCLUSIONS

The plane surfaces were coated with polyethylene composites by an electrostatic powder spray technique. Our experimental results indicate that there are several variables that influence the coating uniformity. Uniformity of polyethylene composite coatings is a function of curing time/temperature and MA-g-HDPE content. The most uniform coating film is achieved at 200°C for 30 min curing temperature and time for the sample with the highest amount of compatibilizer as well as melt viscosity. The adhesion strength and bending results indicate that the coating sample with 15 wt % of MA-g-HDPE has the highest adhesive strength and ductility. As the increase in interfacial interactions resulted from the improved dispersion state, an enhancement in quality and adhesion/bending strength of polyethylene composite coating films can be seen when 15 wt % of MA-g-HDPE is added to the polyethylene composite. The XRD analyses indicate that incorporation of MAH-g-HDPE in the polyethylene composite coating, containing CL15A, results in more intercalation of the silicate layers, and subsequently, nanocomposite formation.

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