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Thermal, morphological and mechanical properties of recycled Al film label /LLDPE compositesSirivan naknoy¹, Pollawat Charoeythornkhajhornchai^{2, 3} and Yanisa Laoong-u-thai^{1,*}¹ Department of Chemical Engineering, Faculty of Engineering, Burapha University, 20131, Thailand² Department of Advanced Materials Engineering, Faculty of Engineering, Burapha University, 20131, Thailand³ Department of Chemical Engineering, Faculty of Engineering, Srinakharinwirot University, 26120, Thailand

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Abstract

Plastic waste contaminated with aluminum film, particularly metallized film, was recognized as a severe environmental issue because it could not decompose naturally, leading to a long-lasting environmental presence with heavy metal contaminants. Therefore, effective management was required to address this problem. This study aimed to recycle metallized film into new materials to maximize its reusability by using linear low-density polyethylene (LLDPE) incorporated with metallized film at 5, 7.5, and 10 phr. The morphology, thermal properties, and thermal degradation of aluminum (Al) film were investigated. The results showed that the addition of aluminum film into the LLDPE matrix slightly increased the melting point due to the effect of the third polymer component and the presence of aluminum particles in the blend. The melt flow index (MFI) of LLDPE with 5, 7.5, and 10 phr aluminum film significantly decreased because the aluminum particles restricted molecular chain mobility, resulting in lower flowability. Furthermore, phase separation of the polymer was observed after the addition of aluminum film into the LLDPE matrix. In addition, the mechanical property results showed that the tensile strength and modulus decreased, whereas the elasticity increased due to the presence of the Al film layer within the LLDPE matrix.

Keywords: Linear low-density polyethylene, Metalized film, Thermal properties, Mechanical properties, Morphology

1. Introduction

Plastic waste represented one of the most critical environmental challenges in modern society, particularly waste that was generated from food packaging such as snack bags. These multilayer laminates, often consisted of plastic coated with aluminum film by a vapor deposition method, were designed to preserve food freshness but posed significant recycling difficulties due to the complexity of separating materials. [1-4]. Studies highlighted that laminated packaging contributed substantially to landfill waste, creating environmental and sustainability concerns. [5-7].

Recycling technologies for laminated materials were developed to address these issues, with a focus on transforming snack bag waste into valuable aluminum materials. In this context, a promising approach involved the use of hydrophobic deep eutectic solvents (HDES) to selectively delaminate multilayer packaging structures, such as PE/Al/PET composites. These composite films were widely used in snack bags and flexible food packaging owing to their excellent barrier properties and mechanical strength, yet their complex multilayer structure rendered them difficult to recycle using conventional methods [8]. The study explored the use of a hydrophobic deep eutectic solvent (HDES) system to dissolve adhesive layers or weaken interfacial bonding between layers, thereby enabling the mechanical separation of aluminum foil from the surrounding polymeric films, typically polyethylene (PE) and polyethylene terephthalate (PET) [9]. The choice of hydrophobic components in the HDES formulation effectively penetrated the lamination interfaces without being absorbed into the plastic layers, thus preserving the quality of the separated materials. The delaminated aluminum retained its

metallic integrity and purity, making it suitable for reuse or further refinement, while the separated plastic films could be processed for subsequent applications [10]. This method represented a milder and more sustainable alternative to traditional chemical or thermal delamination processes, which often involved the use of harsh reagents or required high energy input. [11]. Because of using large amount of solvent with high energy input in the process of this method thus the delamination process did not be chosen in this work. Furthermore, the HDES process was designed to be reusable and environmentally friendly, supporting a more circular and sustainable recycling model for flexible laminated packaging waste [12]. Such approaches aimed to reduce waste volume and greenhouse gas emissions while recovering resources from non-biodegradable materials. However, the direct reuse of recycled aluminum film in industrial applications remained challenging due to its unique physical and chemical properties compared to virgin aluminum [13]. Some researchers [14-15] studied recycling by incorporating metallized plastic waste fibers directly into cementitious materials, aiming to utilize construction materials as a means of resource recovery. These studies explored the feasibility of utilizing metallized plastic waste, such as foil-laminated packaging films, as a reinforcing component in concrete or mortar mixtures. The waste materials were typically cleaned, shredded, and cut into fibrous form before being incorporated into the cement matrix. The blending process often involved directly mixing the fibers with the dry components of concrete, such as cement, sand, and aggregates, prior to the addition of water. Alternatively, some studies [16] introduced the fibers during the wet mixing phase to enhance dispersion and prevent agglomeration. This direct blending approach aimed to improve mechanical properties, such as tensile and flexural strength, as well as to increase resistance to cracking and shrinkage. However, these efforts remained limited and faced several challenges. For instance, achieving uniform dispersion of the fibers, ensuring adequate bonding between the smooth plastic surface and the cement paste, and maintaining the workability of fresh concrete were key issues that had to be addressed. Nevertheless, this approach presented a promising pathway for both enhancing concrete performance and managing plastic waste in a sustainable manner. For this reason, the direct blending of recycled metallized film with polymers to enhance material performance was considered an interesting and viable approach. Some experimental results [17] indicated that the addition of this waste material could improve certain mechanical properties, such as tensile strength and toughness, to a certain extent. These improvements depended on various factors, including the proportion of the added waste, the size of the fibers, and the characteristics of the polymer matrix used.

Our promising approach involved blending recycled aluminum film with polymers, such as linear low-density polyethylene (LLDPE), to create composite materials. LLDPE, known for its excellent flexibility, toughness, and ease of processing [18-19], served as a suitable matrix for incorporating aluminum particles. The addition of aluminum film could potentially enhance the mechanical, thermal, and morphological properties of LLDPE, making it viable for diverse industrial applications. Moreover, the use of LLDPE/aluminum film composites aligned with sustainable practices by promoting the utilization of recycled materials and reducing dependence on virgin resources. Therefore, this study focused on the development of LLDPE-based composites by incorporating recycled aluminum film particles derived from the labels of plastic film packaging. The main objective of this work was to evaluate the properties and potential of these composites for use in further applications [20].

2. Materials and methods

2.1 Materials

Linear low-density polyethylene (LLDPE, injection grade) was purchased from PTT Polyethylene Co., Ltd., Thailand, and metallized film plastic, which was the label of the product, was supported by Sappe Public Co., Ltd., Thailand.

2.2 Processing method

The metallized film was cut into small pieces using scissors and then was mixed with LLDPE by a twin-screw extruder (CTE-D22L32, ChareonTut Co., Ltd., Thailand). The metallized film content in the mixture was adjusted to 5, 7.5, and 10 parts per hundred of resin (phr). The plastic mixture was processed twice to homogenize the polymer composition at a temperature of 170–180 °C with a screw speed of 30–45 rpm. The mixture then was pelletized into plastic pellets using a plastic pelletizer. Afterward, the plastic pellets were dried in a hot air oven at 80 °C overnight before fabrication and property measurements. For DSC, samples were cut from well-blended LLDPE/Al film pellets and were weighed (2–3 mg) into hermetically sealed aluminum pans. Measurements were carried out under a nitrogen atmosphere, heating from 25 °C to 350 °C at 10 °C/min. For TGA, samples were similarly cut from well-mixed pellets, weighed (2–3 mg) into graphite crucibles, and their initial weight was recorded. TGA was performed under a nitrogen atmosphere, heating from 25 °C to 650 °C at a rate of 10 °C/min, after which the analysis was terminated.

2.3 Sample preparation and characterization

The plastic pellets were injected into a dumbbell shape according to ASTM D638 Type I by an injection molding machine (INJ-20T, ChareonTut Co., Ltd., Thailand). The injection molding process was carried out at a temperature ranging from 220 to 275 °C. The cross-sectional area of the injected sample was analyzed morphologically to determine the distribution of the metalized film in the LLDPE matrix by scanning electron microscopy (LEO 1450 VP, ZEISS, USA) at 15 kV of accelerating voltage, and the elemental analysis was measured using an energy dispersive X-ray analysis system (EDS) to investigate the element composition in the polymer matrix. The phase separation of polymer blends was cut into small pieces for observation by optical microscopy (OM, Axiolab 5, ZEISS, Germany). The plastic flow properties (Melt Flow Index, MFI) were measured to assess the flow characteristics, which were a crucial factor for manufacturing processes such as injection molding and extrusion. The test was conducted at 190 °C for 10 minutes following the standard testing method. Thermal stability was measured using a thermogravimetric analyzer (TGA/DSC 3+, Mettler Toledo, Switzerland) under nitrogen gas with a flow rate of 50 mL/min and a heating rate of 10 °C/min, with a degradation temperature range from 25 °C to 650 °C. Additionally, the thermal properties of plastic pellets were analyzed using a differential scanning calorimeter (DSC 5+, Mettler Toledo, Switzerland) to determine the melting temperature within a range of 25 °C to 350 °C. The mechanical properties were assessed following ASTM D638 standards, with at least seven samples tested using a universal testing machine (NRI-TS500-50B, Narin Instrument Co., Ltd., Thailand) at a crosshead speed of 50 mm/min with a 10 kN load.

3. Results and discussion

3.1 Thermal Properties of LLDPE/Al film

The results for LLDPE and LLDPE/Al film blends at 0, 5, 7.5, and 10 phr of Al film composition in Figure 1 showed that the materials with different amounts of Al film exhibited low flow ability characteristics after melting. For example, in a comparison between virgin LLDPE and LLDPE/Al at 5 phr, the flow rate decreased from 2.1 to 1.6, a reduction of about 23%. As the proportion of Al film increased, the trend of the experiment continuously decreased due to the low flow ability of Al film caused by Al particles in its composition. This, in turn, affected the flow during the fabrication process in manufacturing.

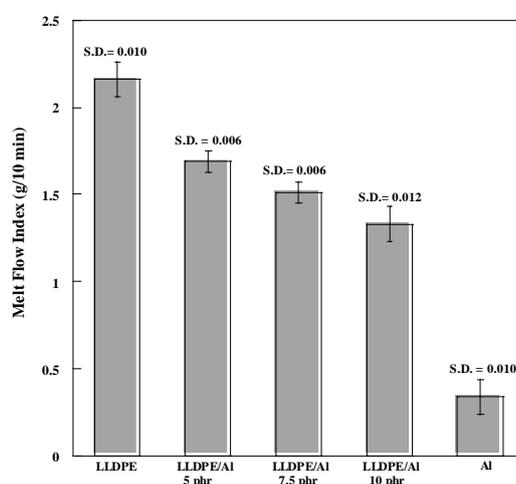


Figure 1 Melt flow index of LLDPE mixed with Al film with various compositions

From the results of the degradation property analysis of plastics containing aluminum, it was found that weight loss occurred between 430–500 °C, corresponding to the degradation temperature of LLDPE [21]. Similar characteristics were observed for three other samples, namely LLDPE/Al film 5 phr, LLDPE/Al 7.5 phr, and LLDPE/Al 10 phr. Additionally, it was found that Al film did not completely degrade but remained detectable in the results. The graph of Al film showed two degradation stages: one between 350–400 °C and another between 430–500 °C. This indicated that the polymer consisted of at least two types of materials. The DSC results showed differences in crystallization and melting behaviors of the materials, particularly the changes observed from mixing Al film with LLDPE. For virgin LLDPE, the first peak at 120 °C indicated the crystallization temperature (T_{cc}), which was the temperature at which LLDPE began to crystallize from the amorphous state. The second peak at 230 °C represented the melting temperature of LLDPE, indicating the temperature at which the crystalline

structure of LLDPE started to melt and transition to the liquid state. After mixing Al film with LLDPE in various amounts, such as 5 phr, 7.5 phr, and 10 phr, the results showed that the first peak at 120 °C still represented the crystallization temperature (T_{cc}) of LLDPE, but it changed slightly. However, the second peak of the LLDPE/Al film blend slightly increased as the Al film content increased. This might have been due to the restriction of polymer molecules in the LLDPE matrix, requiring more heat to melt the crystalline structure of LLDPE [22–23].

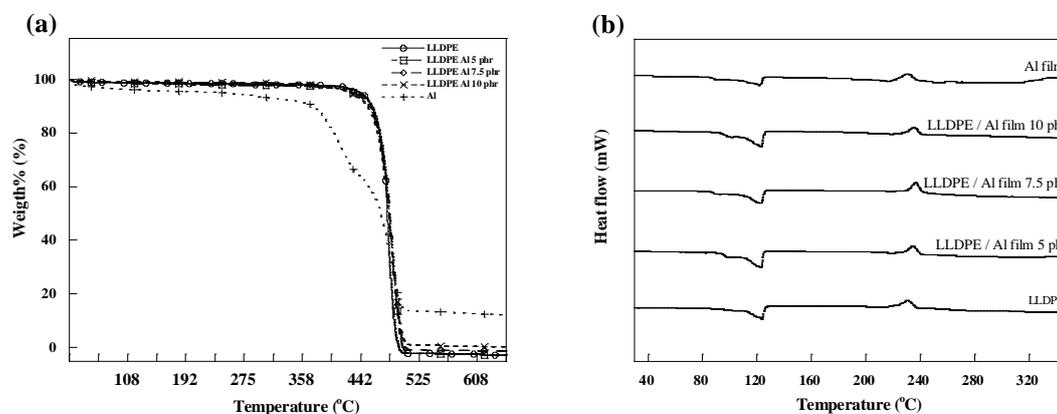


Figure 2 Thermogram of LLDPE/Al film with various composition; TGA (a) and DSC (b)

3.2 Mechanical properties

The tensile strength results for LLDPE/Al film composites at 0, 5, 7.5, and 10 phr are shown in Table 1. As the Al film content increased, tensile strength and modulus decreased, whereas elongation at break continuously increased. The presence of Al film affected the distribution of tensile stress and may have reduced stress concentration in the material, allowing greater stretching with higher Al film content. Additionally, aluminum films produced through multilayer processes often contained other polymer types, which could modify and enhance certain mechanical properties of the composites [24]. The mechanical performance, however, was not determined solely by composition but also was significantly influenced by morphology and microstructure, including filler dispersion, phase continuity, interfacial adhesion, and the size, shape, and orientation of Al film particles within the matrix. To provide context, previous studies on LLDPE composites incorporating recycled aluminum-plastic waste reported similar trends. The previous work observed that incorporating up to 20 wt% recycled aluminum from aseptic packaging reduced tensile strength slightly but it could be compensated with compatibilizers such as maleic anhydride-grafted polyethylene (MAPE), improving stress transfer between the polymer matrix and filler [25]. Another work reported that LLDPE/Al composites maintained tensile strength within acceptable ranges for industrial applications, despite increasing aluminum content [26]. These comparisons indicated that, although tensile strength decreased with higher Al film content, the obtained values in this study were comparable to or above those reported in prior work, demonstrating the composites' suitability for upcycling aluminum-plastic waste while maintaining mechanical performance.

Table 1 Mechanical properties of LLDPE and LLDPE filling Al film.

Sample	Ultimate tensile strength (MPa)	Elongation at break (%)	Young modulus (MPa)
LLDPE	29.30±0.42	337.01±8.43	3.03±0.06
LLDPE/Al5phr	27.64±0.52	323.52±16.62	2.97±0.13
LLDPE/Al7.5phr	26.00±0.26	394.69±13.21	1.92±0.13
LLDPE/Al10phr	25.64±0.17	404.27±27.40	1.25±0.06

3.3 Morphology of LLDPE/Al film

Figures 3a, b, c, d, and e demonstrated the surface structure and distribution of Al film particles. Figure 3a appeared smooth without any white spots, indicating that the polymer contained no added particles. In Figures 3b, c, and d, white particle spots were visible, with the number of spots increasing. These showed the distribution of Al film on the surface of LLDPE, while Figure 3e showed Al film clusters in the shape of fibers. To confirm the presence of Al film particles observed through optical microscopy, an analysis was conducted using SEM-EDX

to verify the elemental composition. The analysis was performed at two specific locations: the LLDPE matrix and the Al film particles (white spots in the images). The investigation covered samples with LLDPE and Al film at ratios of 5, 7.5, and 10 phr. The SEM-EDX analysis results of the LLDPE sample with 5 phr of aluminum (Al) film addition were compared between the left image (Figure 4a) and the right image (Figure 4b). It was found in Figure 4a that carbon, the major component of LLDPE, was at the highest proportion (96.56 wt%), with oxygen (O) and aluminum (Al) in low proportions (O = 3.23 wt% and Al = 0.23 wt%), indicating that the aluminum particles were not well dispersed in this area. In contrast, in Figure 4b, the aluminum (Al) content was significantly higher, reaching 56.68 wt%, suggesting that this area was where aluminum particles accumulated. The proportions of carbon and oxygen (O) decreased noticeably (C = 43.47 wt% and O = not detected). The analysis of LLDPE with 7.5 phr of aluminum (Al) film addition in the examined area (Figure 4c and d) also showed the same result, but the proportion of Al reached 64.63 wt%, while the carbon content decreased to 33.49%, suggesting that aluminum particles were concentrated in this area. The analysis of LLDPE with 10 phr of aluminum (Al) film addition also showed the same trend. In summary, the addition of Al caused more pronounced phase separation in LLDPE. The areas with aluminum accumulation appeared rough and dense. The distribution of Al film became more uneven, which could affect mechanical properties such as strength, elongation, and phase compatibility. This was the cause of low tensile strength and modulus with the increment of Al film content.

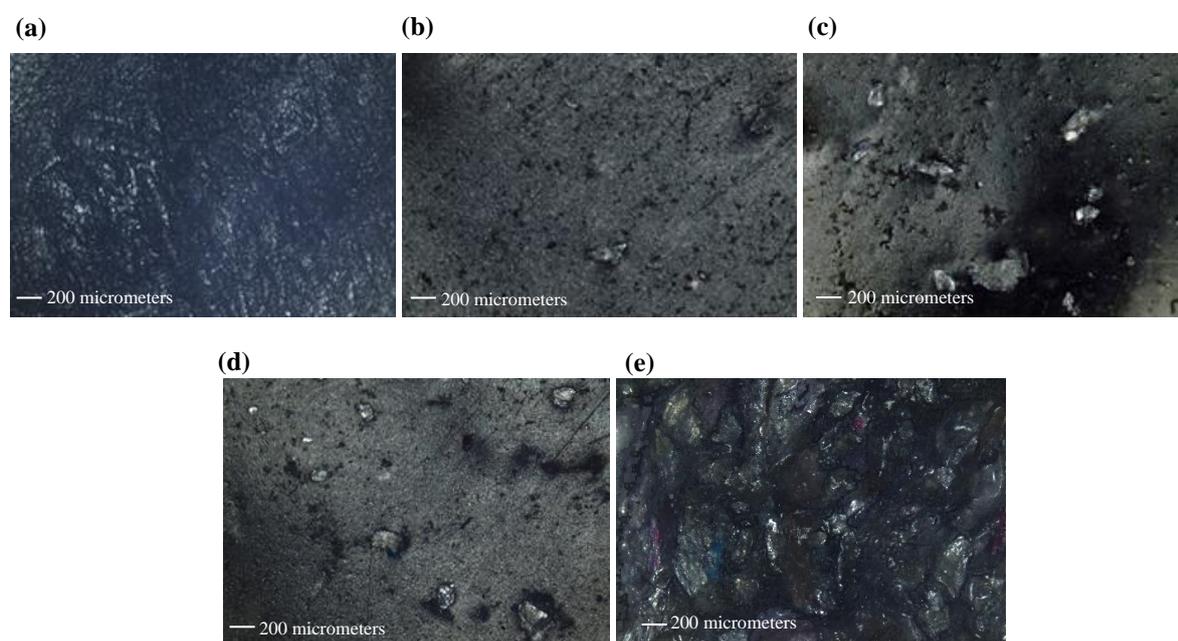


Figure 3 Optical microscopic image of (a) pure LLDPE (Linear Low-Density Polyethylene), (b) LLDPE blended with Al film at 5 phr, (c) LLDPE blended with Al film at 7.5 phr, (d) LLDPE blended with Al film at 10 phr, and (e) Surface structure of Al film alone

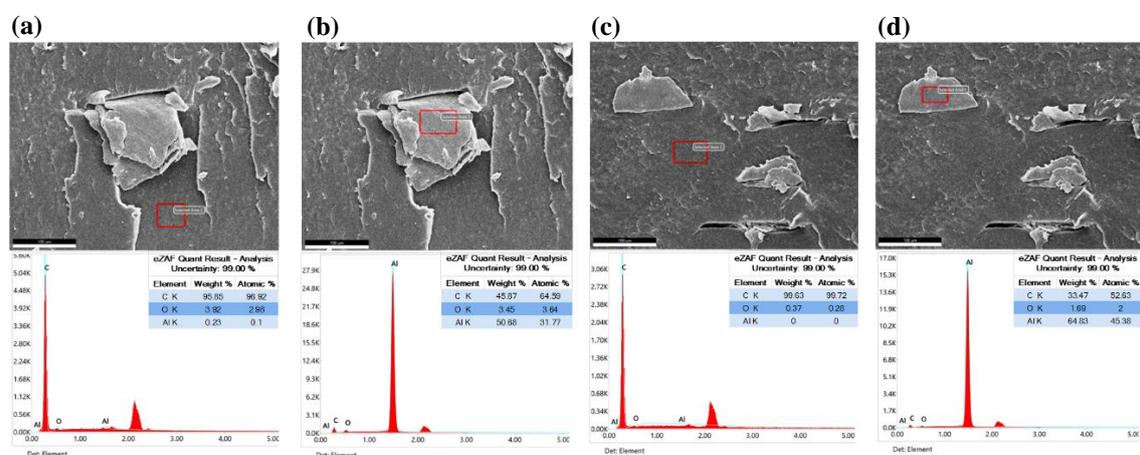


Figure 4 SEM-EDX of LLDPE/Al with different Al contents (5 phr and 7.5 phr) (a) Focus on the surface of LLDPE in the LLDPE/Al 5 phr sample (b) Focus on the Al film in the LLDPE/Al 5 phr sample (c) Focus on the surface of LLDPE in the LLDPE/Al 7.5 phr sample (d) Focus on the Al film in the LLDPE/Al 7.5 phr sample

4. Conclusion

The blending of Linear Low-Density Polyethylene (LLDPE) with aluminum film (Al film) at concentrations of 0, 5, 7.5, and 10 phr revealed that the addition of aluminum significantly influenced the thermal, mechanical, and morphological properties of the composite. The significance of these effects was statistically validated through analysis of variance (ANOVA), with a p-value below 0.05 considered indicative of a statistically significant difference among the measured parameters. In terms of thermal behavior, the Melt Flow Index (MFI) results indicated a pronounced reduction in flowability as the aluminum film content increased. Specifically, the MFI value decreased from 2.1 g/10 min for neat LLDPE to 1.4 g/10 min at 10 phr, representing a reduction of approximately 33%. This decline suggested that the aluminum film adversely affected the processability of the material. Thermogravimetric analysis (TGA) revealed that the onset temperature of degradation remained relatively stable within the range of 390–395 °C, indicating that the initial thermal stability of LLDPE was not significantly altered by the incorporation of aluminum. The onset degradation temperature remained nearly unchanged, indicating stable initial thermal behavior. In contrast, the final degradation temperature increased with Al film content, showing improved thermal resistance at later stages. However, the final degradation temperature exhibited a notable increase from 465 °C for the pure LLDPE to 478 °C at 10 phr, suggesting that the aluminum film contributed to enhanced thermal resistance during the later stages of decomposition. Differential Scanning Calorimetry (DSC) results showed only minor variations in the melting and crystallization behavior of the composites. The melting temperature (T_m) varied by less than ± 1 °C, while the degree of crystallinity (X_c) slightly decreased with increasing aluminum content, implying limited interference of aluminum with the crystalline phase formation of LLDPE. The incorporation of the aluminum film also influenced the mechanical performance of the material. The tensile strength declined from 18.5 MPa for neat LLDPE to 15.2 MPa at 10 phr, whereas the elongation at break increased from 120% to 158%, corresponding to an improvement of approximately 31%. The enhanced ductility could be attributed to the aluminum film facilitating stress redistribution and mitigating localized stress concentrations within the polymer matrix. Nevertheless, the reduction in tensile strength may have limited the suitability of these composites for applications requiring high mechanical integrity. Conversely, the improved flexibility and thermal stability could have been beneficial for products requiring superior deformability and thermal endurance, such as flexible packaging films or surface coatings. Morphological examination using Scanning Electron Microscopy (SEM) confirmed that the aluminum film particles were heterogeneously dispersed within the LLDPE matrix. Micrometer-scale agglomerates were observed at the interfacial regions, resulting in a rough surface texture and reduced interfacial adhesion. These morphological features likely contributed to the observed decrease in mechanical strength and overall phase compatibility of the composite system. The incorporation of high amounts of aluminum film (Al film) into the LLDPE matrix significantly resulted in lower thermal stability and a slightly shifted crystallization temperature, as indicated by TGA and DSC analyses. Tensile strength was slightly reduced, and Young's modulus decreased significantly, whereas elongation at break increased due to morphological changes with particle agglomeration observed in SEM images. Collectively, these findings confirmed that increasing Al film content altered the structure–property relationship of LLDPE composites toward lower strength and higher flexibility.

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6. Author Contributions

All authors contributed to the study's conception and experimental design; S.N., Y.L., and P.C. The data analysis and manuscript was written and discussed by S.N., and P.C. All authors approved the final manuscript.

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