PLA/LLDPE/Organo-Precipitated Calcium Carbonate Composites Containing LLDPE-g-OA Compatibilizers: Mechanical, Physical, Thermal, and Morphology

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Abstract: *A plastic composite consisting of polylactic acid (PLA), linear low-density polyethylene (LLDPE), oleic acid-grafted linear low-density polyethylene (LLDPE-g-OA) compatibilizer, and organo-precipitated calcium carbonate (O-PCC) have been successfully made in the molten state. This study aims to characterize the mechanical, physical, thermal, and morphological characteristics of the PLA/LLDPE/O-PCC plastic composite in the presence of an LLDPE-g-OA compatibilizer. The plastic composite was prepared by blending PLA, LLDPE, LLDPE-g-OA, and O-PCC using an internal mixer with a heating of 160 °C and a rotation of 100 rpm. LLDPE and LLDPE-g-OA are put together into the inner mixer chamber until melted, followed by PLA and O-PCC. The most optimum plastic composite composition is PLA/LLDPE/LLDPE-g-g-OA/O-PCC (67.5:22.5:5:5). The mechanical properties showed an increase in tensile strength of 9.78 MPa. The physical properties showed that the minimum water absorption was 0.74%, the biodegradation in humus soil showed a degradation rate of 0.09% per day, and the thermal properties showed better stability with a melting point of 146.5 °C. The FTIR spectrum is similar to the polymer blend without O-PCC. The morphology indicates that the composite is compatible and homogeneous. This semi-biodegradable plastic composite has significant implications for reducing the accumulation of plastic waste in the environment.*

Keywords: *PLA; LLDPE; O-PCC; LLDPE-g-OA; composite*

■ INTRODUCTION

Over the past decade, the demand for plastic has increased significantly, which has been widely applied to households, industry, electronics, food packaging, and various other sectors. Plastics that are quite popular for use by the public as food packaging are synthetic plastics such as Linear Low-Density Polyethylene (LLDPE). LLDPE is synthetic plastic obtained from non-renewable materials and difficult to degrade in the soil, which causes severe problems for the environment, such as the accumulation of plastic waste [1-3]. In addition, LLDPE has poor compatibility with other materials with different polarities. Efforts made to reduce the impact of using LLDPE on the environment are utilizing natural materials to produce biodegradable plastics or combining thermoplastic polymer materials with biopolymers [4-5].

Biodegradable plastic such as polylactic acid (PLA) is an alternative to eco-friendly packaging because PLA is easily degraded naturally into CO_2 and water by microorganisms. PLA is a type of biopolymer made from renewable resources via a starch fermentation process using lactic acid bacteria and chemical polymerization. PLA also has good biocompatibility with thermoplastic materials and can be fabricated. However, PLA also has weaknesses due to its brittle structure and high gas permeability [6-8]. The development of PLA is currently being carried out to produce better properties and compatibility by blending it with other materials.

Several studies on PLA/LLDPE blends have been performed, such as Engku Zawawi et al. [9] have blended PLA with LLDPE, showing that the two polymers are immiscible blends. Singh et al. [10] have prepared PLA/LLDPE blends with polyethylene-g-glycidyl methacrylate (PE-g-GMA) compatibilizers, in which LLDPE with a larger fraction has more prominent hardening characteristics. Bhasney et al. [11] have studied the influence of micro-cellulose fibers (MCC) of 5% on PLA/LLDPE blends, which resulted in relatively high crystallinity and decreased tensile strength values. The PLA/LLDPE blends are expected to produce semi-biodegradable plastic blends with good tensile mechanical properties, with most of the components of the plastic blends being degradable. However, the PLA/LLDPE blends will result in poor compatibility because the two have different polarities, which can be overcome by adding compatibilizers, such as LLDPE-g-oleic acid (LLDPE-g-OA). This LLDPE-g-OA compatibilizer was obtained from grafting OA monomer onto LLDPE in the presence of a BPO initiator via the molten state or solvent system. This compatibilizer is effectively used in LLDPE/Cyclic Natural Rubber (CNR) blends. LLDPE-g-AO is also believed to be effective in reducing the surface tension of the PLA/LLDPE blends, where the functional group of the OA monomer is compatible with the hydrophilic group of PLA from its carbonyl group. In contrast, the polymer part remains compatible with the LLDPE matrix of the C-H group, and it has also been applied to LLDPE/CNR blends [12]. Adding additives to polymer blends is required as fillers and co-compatibilizers, such as precipitated calcium carbonate (PCC). PCC is a natural material processing product that contains calcite with an amorphous crystal structure and low hardness. PCC has been widely used as a filler material in polymer composites. Modifying organic PCC with oleic acid to form organo-PCC (O-PCC) effectively reduces the surface tension and increases the fit in the polymer matrix [13-17].

This study used O-PCC as filler in the PLA/LLDPE blends containing LLDPE-g-OA to improve the mechanical, physical, thermal, and morphological properties. The novelty of this research is a semi-biodegradable PLA/LLDPE composite containing hydrophobic modified PCC with oleic acid as filler and LLDPE-g-OA graft copolymer as compatibilizers. This study aims to prepare a semi-biodegradable PLA/LLDPE/O-PCC plastic composite in the presence of LLDPE-g-OA compatibilizer via the blending method using an internal mixer in the molten state and characterize its mechanical, physical, thermal, and morphological.

■ EXPERIMENTAL SECTION

Materials

The materials used in this study were polylactic acid (3052D) with a melt flow rate (MFR) of 14 g/10 min (210 °C/2.16 kg) and a density of 1.24 g/cm^3 purchased from NatureWorks LLC, Minnesota, US. Linear low-density polyethylene (Asrene UF-1810MH) with MFR of 1.0 g/10 min (190 °C/2.16 kg) and density of 0.918 g/cm^3 was purchased from Chandra Asri Petrochemical Company, Jakarta, Indonesia. Oleic acid with a density of 0.895 g/mL was obtained from HK Chemical Company, Bekasi, Indonesia. Precipitated calcium carbonate (Schaefer Precarb 100) with a density of 0.0027 g/cm^3 was supplied by Schaefer Kalk, Kuala Lumpur, Malaysia. Benzoyl peroxide initiator, acetone, ethanol, methanol, and xylene were all chemical reagents obtained from Merck, Darmstadt, Germany.

Instrumentation

The PCC and O-PCC nanofillers were characterized using an XRF (PANalytical Epsilon-3). Tensile strength testing was determined using an Ultimate Testing Machine (GoTech AI-7000) at 10 mm/min speed with test specimens referring to ASTM D638-14 (type I). Thermal stability was characterized using TGA/DTA (Hitachi STA-7300) with a heating rate of 10 °C/min. Functional groups were analyzed using an FTIR spectrometer (Agilent Cary 630) at wave numbers 650–4000 cm^{-1}. Morphology was characterized using a Scanning Electron Microscope (ZEISS EVO@MA10).

Procedure

Surface modification of precipitated calcium carbonate with oleic acid

The oleic acid (OA) of 9 mL was put into the beaker glass containing 300 mL of o-xylene, stirred, and PCC nanoparticles of 6 g were added while still stirring and heating at 50 °C for 2 h. The blends were separated by centrifugation at 15000 rpm for 15 min, washed with toluene four times repetitions to remove unreacted OA, and dried at room temperature for 24 h. The O-PCC sample was weighed and characterized using XRF [17-18].

Grafting of an oleic acid monomer onto LLDPE

LLDPE of 30 g was put into the internal mixer at a heating of 160 °C and a speed of 100 rpm for 5 min. Next, 5.0 mL of OA and 1.5 g of BPO initiator were added, and the blending process was allowed for 15 min. The sample was removed from the internal mixer, left for 15 min, and cut into small pieces. The LLDPE/OA sample was put into a bottom flask containing 200 mL xylene, refluxed at 180 °C, and stirred at 60 rpm until a solution formed. The solution was added with 80 mL of acetone to separate the unreacted OA and the OA homopolymer. The formed LLDPE-g-OA precipitate was filtered, washed with 150 mL of methanol (2 times repetitions), dried in an oven at 85 °C for 6 h, cooled, removed, and stored in a desiccator for 24 h. The LLDPE-g-OA sample was weighed and analyzed using FTIR [12,19-20].

Synthesize of PLA/LLDPE/Organo-PCC composites containing LLDPE-g-OA

The LLDPE and LLDPE-OA were slowly put into the internal mixer at a heating 160 °C with a speed of 100 rpm for 5 min. Next, PLA was added into the LLDPE/LLDPE-g-OA blends for 5 min. After that, O-PCC was added to the PLA/LLDPE/LLDPE-g-OA (PLC) blend according to the variation in Table 1. The blending process was left for 10 min. The polymer composite was removed from the internal mixer, left for 10 min, and cut into small pieces [17,21]. The obtained PLA/LLDPE/LLDPE-g-OA/O-PCC (PLCO) composite was weighed and characterized.

Water absorption test of polymer composites

This water absorption test was conducted on polymer blends and composites. The test sample was prepared in a size of 20 mm^2 with a thickness of 3 mm. The sample is cleaned, weighed, and called dry mass (Md). After that, the sample is soaked in water for 24 h, removed, cleaned, and weighed, called the wet mass (Mw). Next, the percentage of water absorption is calculated [22].

Biodegradable test of polymer composites

Samples were prepared in a size of 20 mm^2 with a thickness of 3 mm. The tested sample is cleaned, weighed, and called the initial mass (Mi). Next, the samples were planted using humus soil as deep as 10 cm from the soil surface and monitored regularly for 15, 30, 45, and 60 d. After that, the sample is cleaned, washed, dried, and weighed to a constant weight called the final mass (Mf). The results obtained measured the value of

Table 1. Composition of PLA/LLDPE/ LLDPE-g-OA/O-PCC composites

Sample code	PLA (wt.%)	LLDPE (wt.%)	LLDPE-g-OA (wt.%)	O-PCC (wt.%)
PLCO-0	70.0	25.0	5.0	-
PLCO-1	69.5	24.5	5.0	1.0
PLCO-2	69.0	24.0	5.0	2.0
PLCO-3	68.5	23.5	5.0	3.0
PLCO-4	68.0	23.0	5.0	4.0
PLCO-5	67.5	22.5	5.0	5.0
PLCO-6	67.0	22.0	5.0	6.0
PLCO-7	66.5	21.5	5.0	7.0
PLCO-8	66.0	21.0	5.0	8.0

weight loss percentage. This biodegradation test was conducted in a room with a humidity of 45–65% RH [22].

■ RESULTS AND DISCUSSION

X-ray Fluorescence

PCC and O-PCC fillers were characterized by XRF (Table 2). In the PCC sample, the concentration of elemental Ca was 98.1%, and CaO was 97.8%. It is suitable in prior work [23-24]. Meanwhile, in the O-PCC, the concentration of elemental Ca is 98.0%, and CaO is 97.5%. These results indicate that after PCC was modified with OA, the concentration of elemental Ca was decreased by 0.1% and CaO by 0.3%.

Mechanical Properties

The polymer composite's mechanical properties were analyzed via a tensile strength test on all variations of the polymer composite samples. The test results obtained tensile strength values, elongation at break, and Young's modulus, as shown in Fig. 1. The PLCO-1 to PLCO-8 samples showed increased tensile strength, elongation at break, and Young's modulus compared to the PLCO-0 sample (absence of O-PCC filler). The surface interaction between the polymers and the O-PCC has caused the polymer blends to be adsorbed on the O-PCC surface, which causes an increase in mechanical properties. The even dispersion of the O-PCC in polymer blends occurred because it has the same polarity, which affects the increase in tensile strength, elongation at break, and Young's modulus [25-26]. The increasing number of O-PCC filler components added to polymer composites impacts the mechanical properties of polymer composites. However, a certain amount can weaken the chemical bonds in polymer composites, causing a decrease in their mechanical properties because the elasticity of the polymer blends is getting smaller. The optimal value for adding O-PCC to polymer composites is 5% by weight. The O-PCC filler evenly distributed in the polymer composite can also act as a co-compatibilizer because the mechanical properties of tensile strength, elongation at break, and Young's modulus increase significantly [17-18,27]. The increased elongation at break value after reaching the yield point indicates that the polymer composite is a hard and tough material [12]. Likewise, Young's modulus value increases with increasing tensile strength and elongation at break, indicating a strong bond between PLA and LLDPE in the presence of LLDPE-g-AO compatibilizer and O-PCC filler. The addition of the O-PCC concentration of above 5% will cause the elongation at break distance to decrease and reduce the bond strength, which means the optimum composition for variations of PLA/LLDPE/LLDPE-g-g-OA/O-PCC (67.5:22.5:5:5).

Physical Properties

The physical properties of polymer composites were determined by testing their water absorption. The water

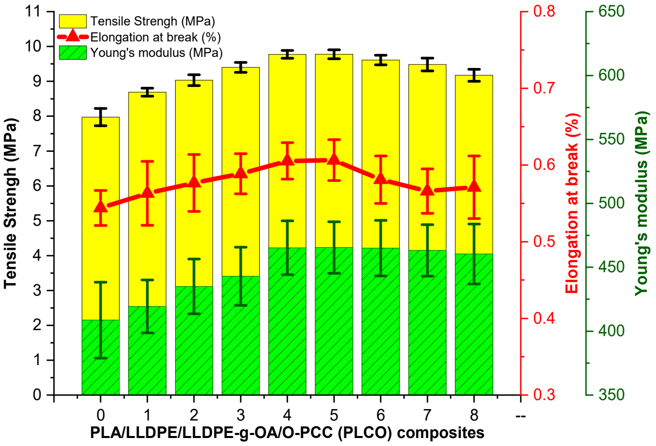


Fig 1. Tensile strength, elongation at break, and Young's modulus of polymer composites

Table 2. X-ray fluorescence of PCC and O-PCC

PCC				O-PCC			
Element	Conc (%)	Oxides	Conc (%)	Element	Conc (%)	Oxides	Conc (%)
Al	0.2	Al_2O_3	0.4	Al	0.3	Al_2O_3	0.4
P	0.4	P_2O_5	0.8	P	0.5	P_2O_5	1.1
Ca	98.1	CaO	97.8	Ca	98.0	CaO	97.5
Zn	0.5	ZnO	0.4	Zn	0.4	ZnO	0.3
Ag	0.7	Ag_2O	0.6	Ag	0.7	Ag_2O	0.6

absorption test aims to determine the ability limit of the polymer composite to absorb water up to a maximum limit for 24 h. The water absorption test result is presented in Fig. 2. The existence of O-PCC as fillers in the polymer composite dramatically influences the water absorption percentage. The more filler in the polymer composite, the higher the water absorption percentage was, indicating an interfacial bond between the polymer blends and the filler, which has been modified with oleic acid. This polymer composite has water-resistant physical properties, characterized by a decrease in the water absorption percentage as the number of fillers increases, the best variation being the PLCO-5 sample. However, when O-PCC is added again into the polymer composite, it causes an increase in the water absorption percentage. This condition can be caused by the uneven dispersion of the filler in the polymer composite or agglomeration [22,25]. In polymer composites with O-PCC concentrations above 5%, there is an increase in water absorption because O-PCC only acts as a filler and is no longer a co-compatibilizer in the PLA and LLDPE mixture so that water is more easily absorbed in the polymer composite, besides the uneven dispersion of the O-PCC also can cause that.

Biodegradation

The biodegradation test of polymer composites was carried out in humus soil with a depth of 10 cm from the surface of the tested soil for 15, 30, 45, and 60 d. The biodegradation test results for this polymer composite are presented in Fig. 3. PLA, which is biodegradable and more dominant in these polymer composites, dramatically influences the degraded polymer material. The biodegradation test results indicated that O-PCC in polymer composites would increase the degradation percentage of the sample. The higher the degradation percentage, the more O-PCC fraction was added into the polymer composite. It is suitable with prior studies [11,28], where the presence of inorganic materials as fillers in polymer composites positively affects the degradation percentage. The PLCO-5 sample showed a significant increase in degradation percentage compared to samples PLCO-4, which means that the optimum value for adding O-PCC is 5 wt.%. Whether the fraction of filler in the polymer composite is increased will certainly impact the decrease in its mechanical and physical properties [18]. In PLCO-5, the degradation percentage was 2.12% in 30 d, 3.24% in 45 d, and 5.54% in 60 d, with an average degradation rate of 0.092% per day for 60 d. The PLCO-6, PLCO-7, and PLCO-8, respectively, showed a non-significant increase in mass degradation rate per day of 0.093, 0.094, and 0.094%, and this means that the degradation process continues even though the duration of biodegradation time increases.

Thermal Analysis

Thermal analysis was performed to investigate the stability, degradation, and thermal properties using

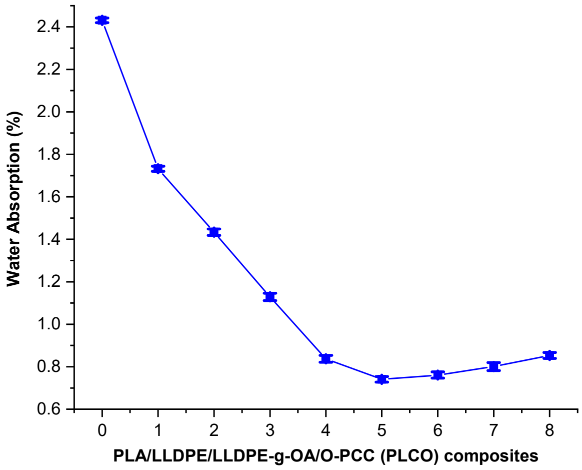


Fig 2. Water absorption of PLCO composites

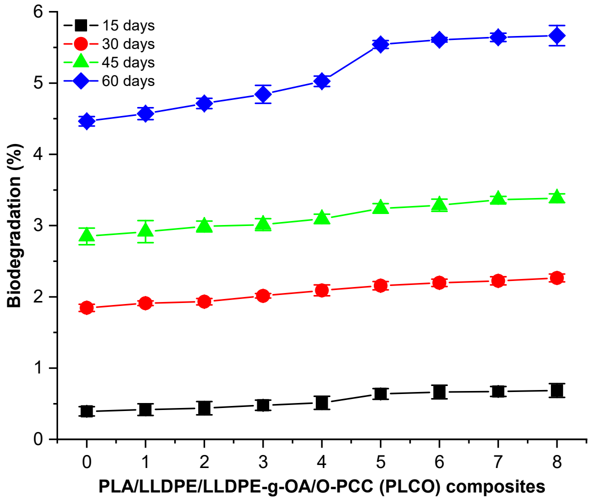


Fig 3. Biodegradation of PLCO composites

TGA-DTA on PLA/LLDPE (PL) blend (70:30), PLA/LLDPE/LLDPE-g-OA (PLC) blend (70:25:5), and PLA/LLDPE/LLDPE0g-OA/O-PCC (PLCO) composite (67.5:22.5:5:5). In the TGA and DTG curves (Fig. 4(a) and Fig. 4(b)), the mass degradation percentage of 1, 5, and 10%, and the maximum expressed as a function of temperature at $T_{1\%}$, $T_{5\%}$, $T_{10\%}$, and T_{max}. The existence of compatibilizers and fillers has increased the mass degradation temperature of the PLC blend and PLCO composite compared to the PL blend presented at $T_{1\%}$, $T_{5\%}$, and $T_{10\%}$. The O-PCC in the PLCO composite has significantly increased thermal stability. Meanwhile, the maximum mass degradation temperature (T_{max}) occurs twice in the PLC blend and PLCO composite, which means that the existence of PLA in the compatible blend or composite is degraded faster than LLDPE. The melting point and decomposition temperatures are expressed as Tm and Td in the DTA curve (Fig. 4(c)) and Table 3. The PL blend shows T_m at 106.3 and 141.3 °C, while the PLC blend and the PLCO composite have just one T_m. According to prior research, if the polymer blend has two T_m, it indicates that the polymer blend is incompatible. It demonstrates that the presence of compatibilizers and fillers has produced a compatible polymer blend or composite [21,29]. The PLC blend and PLCO composite are compatible but have two T_d, meaning the PLA has decomposed earlier than LLDPE. The presence of O-PCC filler can cause the Td in PLCO to increase compared to PLC, which means that O-PCC also acts as a co-compatibilizer in the PLA/LLDPE blend.

FTIR Analysis

FTIR spectra of LLDPE-g-OA, LLDPE, PLA/LLDPE (PL) blend, PLA/LLDPE/LLDPE-g-OA (PLC) blend, PLA/LLDPE/LLDPE-g-OA/O-PCC (PLCO) composite, PLA, and O-PCC presented in Fig. 5. All samples analyzed resulted in a band at 2914.7–2847.6 cm^{-1}, indicating

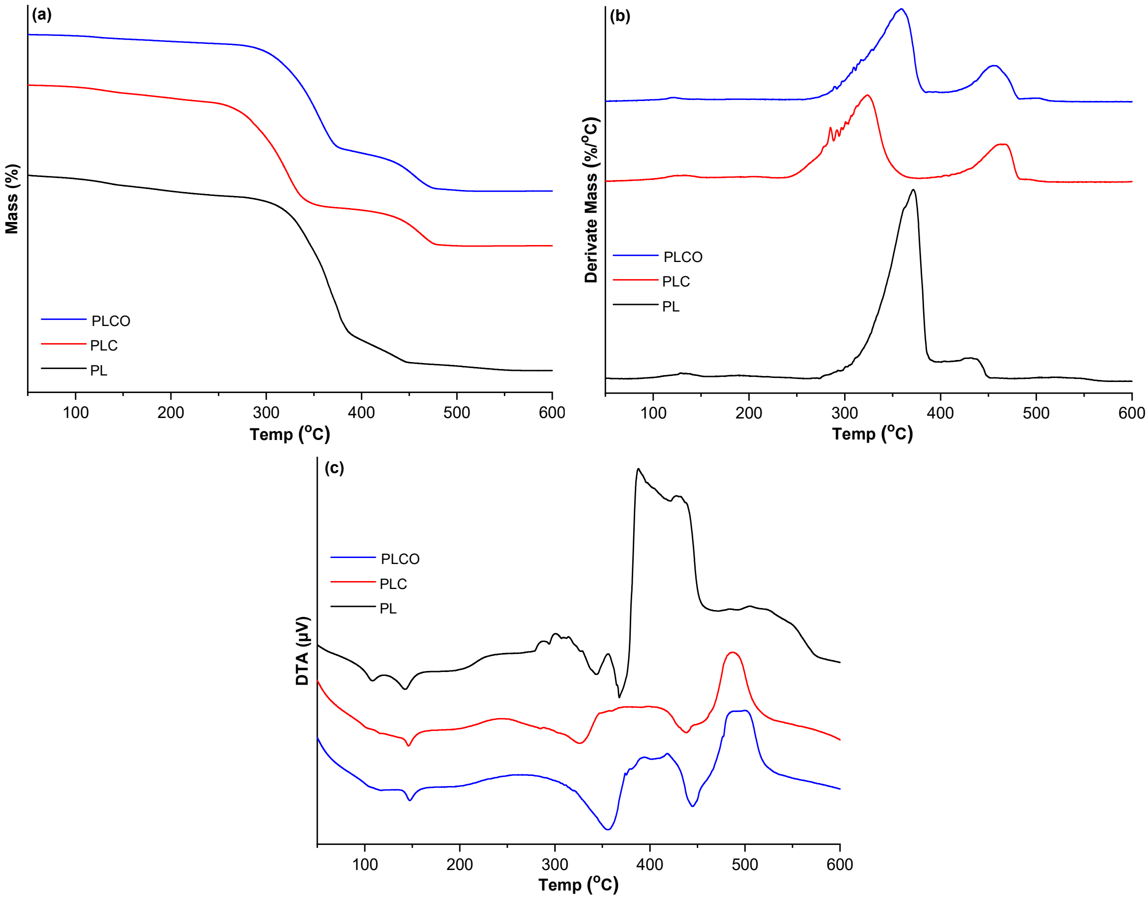


Fig 4. (a) TGA, (b) DTG, (c) DTA curves of PL, PLC, and PLCO

Table 3. Thermal analysis of PL, PLC, and PLCO

Sample	$T_{1\%}$ (°C)	$T_{5\%}$ (°C)	$T_{10\%}$ (°C)	T_{max} (°C)		T_m (°C)		T_d (°C)	
PL	47.3	132.5	216.3	371.9	-	106.3	141.3	343.4	-
PLC	79.8	148.2	233.0	323.1	454.0	-	146.4	324.9	438.1
PLCO	92.9	209.5	294.8	359.2	462.7	-	146.5	354.9	444.1

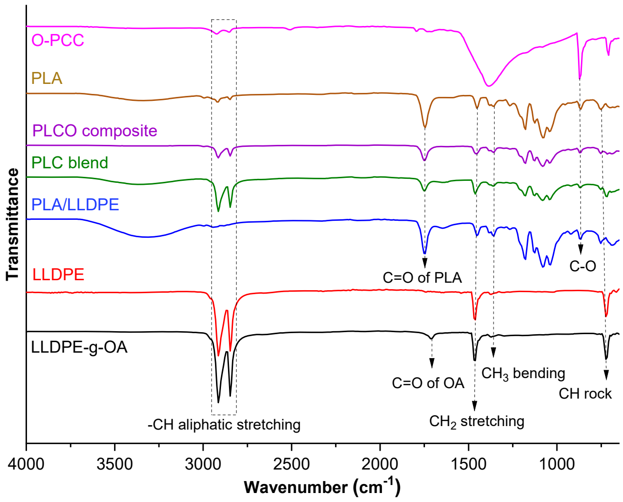
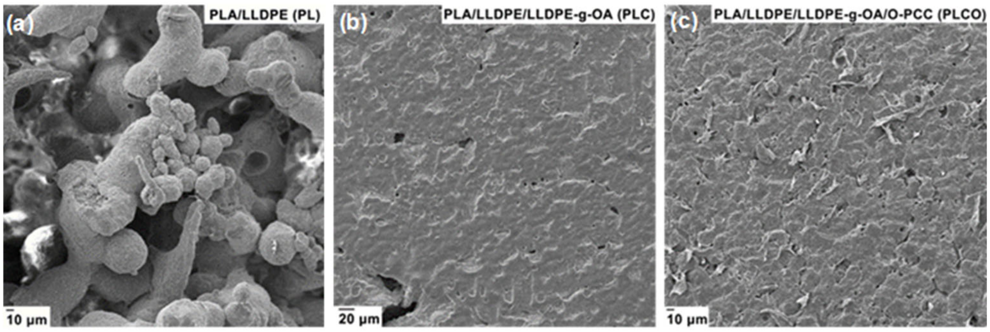


Fig 5. FTIR spectra of LLDPE-g-OA, LLDPE, PLA/LLDPE, PLA/LLDPE/LLDPE-g-OA (PLC) blend, PLA/LLDPE/LLDPE-g-OA/O-PCC (PLCO) composite, PLA, and O-PCC

saturated aliphatic hydrocarbon compounds. The PLA/LLDPE blend resulted in a small band in that area, which means that there is no bond between the two materials because what is visible is only the absorption band of PLA. The band at 1751.8 cm^{-1} indicates the presence of a carbonyl group from PLA, while the small band at 1707.1 cm^{-1} is a carbonyl group from oleic acid grafted onto LLDPE. The bands at 1461.1 and 1371.6 cm^{-1} indicate the presence of CH_2 and CH_3 vibrations. The band at 864.7 cm^{-1} shows the C–O vibration of PLA and O-PCC. A band at 752.9 cm^{-1} in the fingerprint area indicates a C-H rock vibration. The PLC blend and the PLCO composite had almost identical spectra, indicating that the two samples had chemical bonds between PLA and LLDPE in the presence of LLDPE-g-OA compatibilizers. The filler in the polymer composite has also resulted in a difference in the intensity of each absorption band from the two samples [18,25].

Morphological Analysis

Morphological analysis of the PL blend, PLC blend, and PLCO composite (Fig. 6) was performed to observe the surface structure of the samples. The PL blend surface morphology (Fig. 6(a) and Fig. 6(d)) shows a surface structure of an incompatible blend, where the blending of PLA and LLDPE is not homogeneous and appears to form aggregations. This is due to the difference in polarity between the two polymeric materials. The surface morphology of the PLC blend (Fig. 6(b) and Fig. 6(c)) shows a surface structure of a compatible blend, where the blending of PLA and LLDPE is homogeneous, where PLA and LLDPE appear to merge to form a compatible blend after adding the LLDPE-g-OA compatibilizer. The PLC blend surface structure also appears smooth with tiny pores. The surface morphology of the PLCO composite (Fig. 6(c) and Fig. 6(f)) also shows a surface structure of a compatible O-PCC filler, which is evenly dispersed even though there



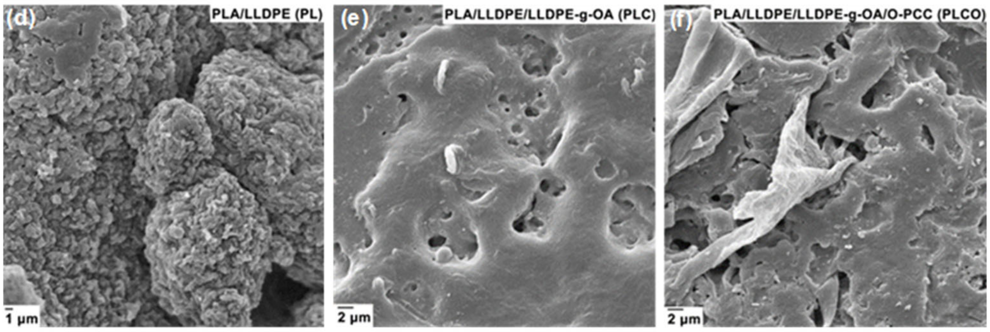


Fig 6. SEM micrographs of (a) PL, (b) PLC, (c) PLCO at 1000× magnification, and (d) PL, (e) PLC, (f) PLCO at 5000× magnification

are tiny pores formed in the composite. The presence of O-PCC in the composite functions as a filler and co-compatibilizer, making the composite more compatible with its constituent materials [17-18,30].

■ CONCLUSION

The semi-biodegradable PLA/LLDPE/O-PCC plastic composite in the presence of LLDPE-g-OA compatibilizer can be produced in a molten state at 160 °C using an internal mixer with the most optimum variation is PLA/LLDPE/LLDPE-g-g-OA/O-PCC (67.5:22.5:5:5). The mechanical properties of PLCO composites resulted in the most optimum tensile strength of 9.78 MPa, elongation at break of 0.61%, and Young's modulus of 465.48 MPa. The physical properties of PLCO composites resulted in a minimum water absorption of 0.74%. The biodegradation of PLCO composites in humus soil was 0.64% in 15 d, 2.12% in 30 d, 3.24% in 45 d, and 5.54% in 60 d, with a degradation rate of 0.09%/d, which is not significantly different from the variations above it. The thermal properties of PLCO composites resulted in better thermal stability than the PL and PLC blends, with a melting point temperature of 146.5 °C and a decomposition temperature of 444.1 °C. The FTIR spectrum of PLCO composites showed spectrum similarity with the PLC blend and had $CaCO_3$ characteristics at 864.7 and 1701.1 cm^{-1}, indicating the presence of O-PCC in the PLCO composite. The morphology of PLCO composites has resulted in a compatible, homogeneous surface structure with evenly dispersed O-PCC.

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■ AUTHOR CONTRIBUTIONS

Ahmad Hafizullah Ritonga, Barita Aritonang, and Debi Meilani conducted the experiment. Ahmad Hafizullah Ritonga, Gusliani Eka Putri, and Khairiah conducted the calculations and analysis. Ahmad Hafizullah Ritonga wrote the manuscript. Ahmad Hafizullah Ritonga, Gusliani Eka Putri, and Enzo Wiranta Battra Siahaan revised the manuscript. All authors agreed to the final version of this manuscript.

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