



Novel toughened polylactic acid nanocomposite: Mechanical, thermal and morphological properties

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ABSTRACT

The objective of the study is to develop a novel toughened polylactic acid (PLA) nanocomposite. The effects of linear low density polyethylene (LLDPE) and organophilic modified montmorillonite (MMT) on mechanical, thermal and morphological properties of PLA were investigated. LLDPE toughened PLA nanocomposites consisting of PLA/LLDPE blends, of composition 100/0 and 90/10 with MMT content of 2 phr and 4 phr were prepared. The Young's and flexural modulus improved with increasing content of MMT indicating that MMT is effective in increasing stiffness of LLDPE toughened PLA nanocomposite even at low content. LLDPE improved the impact strength of PLA nanocomposites with a sacrifice of tensile and flexural strength. The tensile and flexural strength also decreased with increasing content of MMT in PLA/LLDPE nanocomposites. The impact strength and elongation at break of LLDPE toughened PLA nanocomposites also declined steadily with increasing loadings of MMT. The crystallization temperature and glass transition temperature dropped gradually while the thermal stability of PLA improved with addition of MMT in PLA/LLDPE nanocomposites. The storage modulus of PLA/LLDPE nanocomposites below glass transition temperature increased with increasing content of MMT. X-ray diffraction and transmission electron microscope studies revealed that an intercalated LLDPE toughened PLA nanocomposite was successfully prepared at 2 phr MMT content.

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1. Introduction

Biopolymers are expected to be an alternative for conventional plastics due to the limited resources and soaring petroleum price which will restrict the use of petroleum based plastics in the near future. PLA has attracted the attention of polymer scientist recently as a potential biopolymer to substitute the conventional petroleum based plastics. Apart from being in the category of biodegradable polymer, PLA has wide applications in biomedical field due to its biocompatibility characteristics. Recent studies and findings on PLA had proven that the biopolymer has good mechanical properties, thermal plasticity and biocompatibility, and is readily fabricated, thus being a promising polymer for various end-use applications [1]. However, PLA, similar to polystyrene, is a comparatively brittle and stiff polymer with low deformation at break [2]. One main task is to modify these properties in such a way that PLA is able to compete with other more flexible commodity polymers

such as polyethylene, polypropylene, polyethylene terephthalate or polyvinyl chloride.

Previous researchers have shown that addition of plasticizers such as polyethylene glycol (PEG), glucosemonoesters and partial fatty acid esters had successfully overcome the brittleness and widen PLA's application [2–4]. Jacobsen et al. [2] have revealed that plasticization had significantly improved the elongation at break and impact strength of PLA. However, plasticization of PLA had sacrificed the inherent stiffness of the material limiting its application for structural uses. Thus, the development of an impact modifier is significantly required to improve the toughness of the material without extensively forfeiting the stiffness of PLA.

Recent achievements in nanocomposite technology have fueled the need for new knowledge and findings in the field of polymer nanocomposites resulting in the development of respective polymer nanocomposites; polyamide6/polypropylene [5,6], polyamide 66 [7], polypropylene [8], polycaprolactone [9], polystyrene [10] and natural rubber [11]. For the past few years, the field of PLA nanocomposites [12,13] based on layered silicates, such as MMT, has gained its popularity among scientist and industrials. The nanoscale distribution of such high aspect ratio fillers brings up some large improvements to the polymer matrix in terms of

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mechanical, fire retardant, rheological, gas barrier and optical properties, especially at low clay content (as small as 1 wt.%) in comparison with conventional microcomposites (>30 wt.% of microfiller). In order to reach this nanoscale distribution, the naturally hydrophilic clay filler has to be organically modified to be more compatible with the organic polymer matrix.

Interestingly, the distribution of nanoclay in PLA is proven to be well dispersed without introduction of compatibilizing agents [13]. This is due to the interaction of hydrogen-bondings between ammonium groups in the organic “surfactant” of the organoclay with the carbonyl group of PLA chain segments contributes to this process [14]. There are also strong interactions between the PLA hydroxyl end groups and the MMT platelet surfaces or the hydroxyl groups of the ammonium surfactant in the organically modified MMT reported in previous study by Jiang et al. [15]. Regardless of the improvements achieved in the development of PLA nanocomposites, the polymer's brittleness had become more inherent. This had limited its application for structural applications.

Similar brittleness problems had been solved before by researchers with the approach of introduction of elastomeric materials into a brittle nanocomposite system. Wahit et al. [5] had examined the ethylene octene copolymer toughening of polyamide 6/polypropylene nanocomposites in terms of impact strength, ductile–brittle transition temperature, and tensile properties. Lim et al. [8] had successfully developed the toughened polypropylene nanocomposites with poly(ethylene-co-octene), PP/POE and studied its morphology, thermal and mechanical behaviour. However, only several studies had been conducted recently in the development of plasticized PLA nanocomposites [16–20] but, the mechanical properties of these nanocomposites were not studied in detail. Thellen et al. [18] had investigated the influence of MMT layered silicate on plasticized PLA blown films and concluded that the plasticized PLA/MMT nanocomposites did not see highly significant enhancements with addition of MMT but the toughness is at least maintained in the nanocomposites unlike other filled polymeric systems. They found that the plasticization effect reduced the brittleness of the nanocomposites and the breakthrough will widen PLA's applications.

Although the addition of plasticizers such as PEG will overcome PLA's brittleness, however, it comes with a sacrifice of stiffness which is also important for structural applications. Pluta et al. [19] showed that the addition of PEG into MMT filled PLA had significantly sacrificed the stiffness of the nanocomposite. The storage modulus (E') at room temperature, resembling the stiffness of PLA nanocomposite, had decreased from 700 to 450 MPa with addition of 20 wt.% PEG. Thus, a new toughener was needed to overcome the brittleness nevertheless significantly reducing the stiffness. LLDPE could become the best impact modifier for PLA as it is cost effective and easy processibility. The introduction of LLDPE as an impact modifier in polymers had been studied by previous researchers [21–23]. Previous study by Anderson et al. [21] who performed melt blending of PLA and LLDPE in an effort to toughen PLA showed that semicrystalline PLA has relatively good miscibility with LLDPE, hence these two materials are chosen as matrix and impact modifier. They also reported a significant increase of impact strength in semicrystalline PLA with the addition of LLDPE and stated that they had successfully toughen PLA with LLDPE. However, the researchers had only investigated the effect of 20 wt.% LLDPE with PLA and the study focused on the compatibility of LLDPE in semicrystalline PLA.

To the best of our knowledge, no systematic studies have been done so far to investigate the properties of LLDPE toughened PLA nanocomposites. Therefore, the objective of this paper is to investigate the mechanical, thermal and morphology properties of LLDPE toughened PLA nanocomposites with comparison to PLA nanocomposites with addition of MMT at various con-

tents. Although it seems that the use of LLDPE, a non-biodegradable polymer, as an impact modifier for PLA nanocomposites may defeat the purpose of developing a ‘green’ polymer, the potential application from this research is for utilization of PLA in structural applications especially when petroleum based commodity plastics becomes more scarce and expensive.

2. Experimental

2.1. Materials

Poly(lactic acid (PLA; molecular weight, Mw: 220 kDa; Mn: 101 kDa) was obtained from Biomer, Kraling, Germany (product name Biomer L9000). LLDPE of injection molding grade was obtained from Titan Polyethylene, Johor, Malaysia (Titanex® Li 5011, Melt Mass Flow Rate (MFR) of 50 g/10 min and density of 0.926 g/cm³), montmorillonite (MMT) obtained from Nanocer Inc. Arlington Heights IL, USA (Nanomer 1.30TC) organically modified with octadecylamine with mean dry particle size of 16–22 µm.

2.2. Nanocomposites preparations

PLA and LLDPE pellets were dried at 40 °C and MMT at 100 °C respectively for 24 h prior to compounding. The nanocomposites according to Table 1 were compounded by simultaneous adding of all components to Brabender Plasticoder PL 2000 counter-rotating twin-screw extruder. PLA and LLDPE blends were prepared with weight percentage while MMT was added as parts per hundred (phr) in the blends. The barrel temperature profile adopted during compounding of all blends was 190 °C at the feed section, decreasing to 175 °C at the die head. The screw rotation speed was fixed at 35 rpm. The extruded materials were injection molded into standard tensile, flexural and Izod impact specimens by using a JSW (Muroran, Japan) Model NIOOB II injection-moulding machine with a barrel temperature of 165–190 °C. All test specimens were allowed to condition under ambient conditions for at least 48 h prior to testing.

2.3. Mechanical analysis

Tensile test was carried out according to ASTM D638 using an Instron (Bucks, UK) 5567 under ambient conditions with crosshead speeds of 5 mm min⁻¹. Flexural test was done according to ASTM D790 by AG-5kNE Shimadzu universal testing machine under ambient conditions with crosshead speed of 3 mm min⁻¹. Izod impact tests were carried out on notched impact specimens according to ASTM 256, by using a Toyoseiki (Tokyo, Japan) impact testing machine under ambient conditions. Five specimens of each formulation were tested and the average values were reported.

2.4. Thermal analysis

The melting and crystallization behaviour of the blends and nanocomposites were studied under nitrogen atmosphere by differential scanning calorimetry (DSC) (Perkin–Elmer DSC-6), using

Table 1
Designations of material and their compositions.

Designations	Compositions	Parts
PLA	PLA	100
PLA/M2	PLA/MMT	100/2
PLA/M4	PLA/MMT	100/4
P90/L10	PLA/LLDPE	90/10
P90/L10/M2	PLA/LLDPE/MMT	90/10/2
P90/L10/M4	PLA/LLDPE/MMT	90/10/4

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