

Water absorption and enzymatic degradation of poly(lactic acid)/rice starch composites

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Abstract

Poly(lactic acid) (PLA) composites consisting of PLA, rice starch (RS) (0–50 wt%) and epoxidised natural rubber (ENR50) were compounded by a twin-screw extruder and compression moulded into dumbbell specimens. Tensile tests were performed to characterize the mechanical properties of the PLA/RS composites. Morphological studies were done on the tensile fractured surface of the specimens by using scanning electron microscopy (SEM). Twenty weight percent of RS achieved a good balance of strength and stiffness. Beyond 20 wt% loading of RS, the tensile strength and elongation at break of PLA decreased drastically. This may be attributed to the agglomeration of RS, which could then act as stress concentrator. The incorporation of ENR50 increased the tensile strength and elongation at break of the PLA/RS composites remarkably, owing to the elastomeric behaviour and compatibilisation effects of ENR50. Interestingly, the morphology of PLA/RS composites transformed to a more ductile one with the addition of ENR. The kinetics of water absorption of the PLA/RS composites conforms to Fick's law. The M_m and D values are dependent on the RS and ENR concentrations. The tensile properties of the PLA/RS composites deteriorated after water absorption. The retention-ability and recoverability of the PLA/RS composites are relatively low, attributed to the hydrolysis of PLA, degradation of the PLA–RS interface and leaching of the RS particles. In addition, the tensile properties of PLA/RS composites decreased drastically upon exposure to enzymatic degradation. Extensive pinhole and surface erosion on the PLA/RS composites indicate high degree of hydrolysis. Whilst the addition of ENR leads to some improvements in tensile properties, nevertheless, it enhanced the biodegradability of the PLA/RS composites when exposed to water and α -amylase enzymatic treatments.

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1. Introduction

Advanced technology in the field of petrochemical polymers has brought many advantages to mankind. However, the ecosystems are now considerably disturbed

and are slowly being damaged as a result of the usage of non-degradable materials in disposable items. The development of biodegradable plastics can play a crucial role in helping to solve the waste disposal problems. Since waste reduction is important for future municipal solid waste, a number of important issues need to be addressed to ensure that new biodegradable plastics industry develops [1].

Poly(lactic acid) (PLA) is a degradable thermoplastic polymer with excellent mechanical properties which is now, beginning to be produced on a large scale from

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fermentation of corn to lactic acid and subsequent chemical polymerisation. Pure PLA can degrade to carbon dioxide, water and methane in the environment over a period of several months to 2 years, compared to other petroleum plastics which need 500–1000 years [2].

Starch is of interest as a naturally occurring biodegradable material due to its cost effectiveness, abundance, renewability, availability, thermo-processability using conventional plastic processing equipment (extrusion, injection moulding), and its adequate thermal stability for melt blending with synthetic plastics (i.e. aliphatic polyester, non-biodegradable thermoplastics) [3]. The incorporation of starch into plastics to enhance their fragmentation and degradability in the environment has generated considerable interest. The introduction of low-cost starch as a filler into PLA would result in a fully renewable biodegradable material [2].

Epoxidised natural rubber (ENR) is obtained from natural rubber by replacing some of the double bonds with epoxide units. ENR with 50 mol% epoxidation has been shown to be polar with properties similar to those of acrylonitrile butadiene rubber and butyl rubber [4]. It has been reported that compatibility of ENR increases as the ENR's polarity increases [5]. The presence of the oxirane groups in ENR introduced polarity and made the rubber more versatile for rubber–rubber or rubber–plastic blending [6]. However, ENR vulcanizates exhibit poor aging properties. The unstable nature of ENR is believed to be due to the residual acidity that was inherited from the natural rubber modification (epoxidation) with peroxide. In long-term exposure to ambient temperature (27–30 °C) in the laboratory, poly(vinyl chloride)/ENR blends deteriorated rather rapidly [7]. Addition of ENR into LLDPE/sago starch composites had achieved a comparable mechanical properties and remarkable degradability [8].

Water absorption behaviour of starch filled polylactic acid composites has been studied by some researchers [9–11]. The addition of starch increased the water absorption of the PLA [9]. According to Wang et al. [2], the higher water absorption after a long time would probably lead to enhanced biodegradability. According to Garlotta et al. [3], PLA is easily hydrolysed by moisture, and its hydrolysis products are non-toxic to human being. Water absorption may strongly affect the organic materials behaviour, by producing changes in its chemical and physical nature [11]. It is believed that, the hydrophilicity of PLA (ester groups), ENR (oxirane groups) and RS (hydroxyl groups) will increase the water uptake. Thus, water, may act as a plasticiser, and/or “hydrolysis agent” for the PLA matrix, and subsequently deteriorate the properties (e.g. mechanical properties) of PLA/RS composites. Therefore, the degradability of the PLA/RS composites might be accelerated in a moist environment by water during its service life.

α -Amylase are endoamylases catalysing the hydrolysis of internal α -1,4-glycosidic linkages in the starch in a random manner. The microbial α -amylase for industrial purposes are derived mainly from *Bacillus licheniformis*, *Bacillus amyloliquefaciens* and *Aspergillus oryzae* [12]. α -Amylase catalyses the hydrolysis of starch resulting in a significant release of maltose and leads to formation of smaller maltosaccharide fragments [13,14]. Starch susceptibility to enzyme attack is influenced by several factors, such as amylose and amylopectin content, crystalline structure, granular structure, particle size and the presence of enzyme inhibitors. Hydrolysis pattern is an important factor for susceptibility to α -amylase attack. Extensive surface erosion was shown to indicate a high degree of hydrolysis, whereas less surface erosion indicated less degradation [15]. Ishiaku et al. [1] demonstrated that extensive enzyme activity occurred on a polycaprolactone/sago starch film after being subjected to enzyme amylase. The majority of the sago starch granules were removed from the surface of the film and leave behind transparent voids. Danjaji et al. [16] reported that microbial activity occurred on the enzyme amylase-treated LLDPE/sago starch sample, which is indicated by the presence of holes and agglomerates on the sample surface. Araújo et al. [17] have investigated the enzymatic degradation of corn starch/poly(ethylene-vinyl alcohol) copolymer blends (SEVA-C), and interestingly, they have found that the action of α -amylase leads to the starch degradation, and part of the amorphous of SEVA-C blend was released and degraded.

This paper focuses on the study of PLA composites consisting of PLA, RS and ENR. The effect of RS loading and ENR on the mechanical properties and morphology of PLA/RS composites will be investigated. Whilst the addition of ENR could lead to some improvements of the mechanical properties of PLA/RS composites, nevertheless, it could enhance the biodegradability of the composites when the composites are being subjected to a moist environment (water). Moreover, emphasis was laid on the moisture absorption studies (i.e. water uptake) and enzymatic tests of PLA/RS composites. Effects of water absorption and enzymatic attack on both mechanical and morphological properties of PLA/RS composites were determined.

2. Experimental

2.1. Materials

Poly(lactic acid) (PLA) was supplied by Mitsui Chemicals Inc., Japan. Melt flow index (MFI) and density of PLA were 64.8 g/10 min (at 190 °C and 2.16 kg load) and 1.23 g/cm³, respectively. Rice starch was obtained from TIGA Gajah Cho Heng Sdn. Bhd.,

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