



Biocompatibility and characterization of renewable agricultural residues and polyester composites



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ABSTRACT

Composites of sesame husk and glycidyl methacrylate-grafted poly(trimethylene terephthalate) (PTT-g-GMA/SH) exhibit noticeably superior mechanical properties compared to PTT/SH composites due to greater compatibility between the two components. The dispersion of SH in the PTT-g-GMA matrix is highly homogeneous as a result of condensation reaction formations. Human lung fibroblasts (FBs) were seeded on these two series of composites to characterize the biocompatibility properties. In a time-dependent course, the FB proliferation results demonstrated higher performance from the PTT/SH series of composites than from the PTT-g-GMA/SH composites. In addition, collagen production by FBs present in the PTT/SH series was 20% higher than in regular culture-plates after 7 days of incubation. The water resistance of PTT-g-GMA/SH was higher than that of PTT/SH, although the weight loss of both composites buried in soil compost indicated that they were both biodegradable, especially at higher levels of SH substitution. The PTT/SH and PTT-g-GMA/SH composites were more biodegradable than pure PTT, implying a strong connection between SH content and biodegradability.

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1. Introduction

Biomedical materials can be approximately divided into three major types: metals, ceramics, and organic plastic matter. Their derived products are used for different clinical applications (Golish & Anderson, 2012; Lévesque, Hermawan, Dubé, & Mantovani, 2008; Nair & Laurencin, 2007; Xuanyong, Chu, & Ding, 2010). For example, the various surgical instruments and internal and external fixing devices used in orthopedics are mostly made of metal, while ceramic material is mostly applied as a bone filling material. Organic plastic materials have been extensively developed into diversified, expendable medical products for 50 years due to their easy processability and good mechanical properties. Organic plastic materials have high biocompatibility and can transmit cell recognition signals, as well as be used for cell adsorption, multiplication, and differentiation (Brocchini, 2001; Shruiti & John, 2011; You et al., 2010). Therefore, in addition to traditional medical appliances, organic plastic materials are presently being used in the development and application of tissue engineering materials. Tissue

engineering aims to replace body parts that have impaired function due to diseases or injuries; it also aims to assist in wound healing, enhance the functions of organs and tissues, and rehabilitate or replace defective and abnormal tissues and organs (Artemenko et al., 2012; Marquès, Méndez, Gironès, Ginebra, & Pèlach, 2009). Furthermore, biomedical materials are expected to be emphasized in regenerative medicine in the future, and today's biomedical materials are mostly composed of organic plastic. To provide plastics with multiple functions, developing high quality aliphatic polyester plastics is very important. However, due to their typically poor mechanical properties, aliphatic polyester plastics have thus far only been used to develop nonelastic products [e.g., polylactic acid (PLA), polyhydroxybutyrate (PHB), polycaprolacton (PCL)]; in contrast, aromatic polyester plastics [e.g., polyethylene terephthalate (PET), poly(trimethylene terephthalate) (PTT), polybutylene terephthalate (PBT)], with their good mechanical properties and processability, can be extensively used to develop products meeting the market demand in tissue engineering and regenerative medicine. For these reasons, in this study, we investigated aromatic polyester plastics (Liu et al., 2005; Safapour et al., 2010).

PTT is a thermoplastic organic plastic typically formed by the reaction of 1,3-propanediol (1,3-PDO) with *N,N*-dimethylaniline (DMA). This synthetic process saves more than 40% of the energy

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used in traditional petroleum extraction methods for PTT production, and the PTT material can be recovered completely, thus reducing the consumption of both raw materials and energy. In addition, the formation of the toxic by-product acrolein can be reduced by more than 95% compared to traditional PTT formation methods; hence, PTT today is regarded as an environmentally friendly, recycled material with zero pollution. PTT is characterized by elasticity, ductility, flexibility, and minimal deformability. It also has a positive effect on the fiber structure of green products through special processing, imparting such products with antistatic properties, ultraviolet resistance, chlorine resistance, and fouling resistance, without any chemical treatment (Gupta & Choudhary, 2012; Toshikazu, Barbara, & Akio, 2012). In the past decade, several companies have paid close attention to this topic and have investigated how to improve the durability and practicability of this polyester. Unfortunately, PTT does not typically provide economic benefits; it has a relatively high cost, and thus agricultural residues [e.g., rice husk, bamboo and sesame husk (SH)] have been added to the PTT plastic as a filling material to reduce its cost (Nyambo, Mohanty, & Misra, 2010; Sain & Panthapulakkal, 2006).

In this study, we evaluated sesame husk as filling material for PTT. Sesame husk is a type of vegetable fiber that contains large amounts of vitamins B₁, B₂, B₆, C, and E, as well as dietary fiber. The main constituent is vitamin E, which is regarded as an antiaging vitamin that can be effective for improving blood circulation and promoting metabolism. In addition to the above-mentioned nutrients, sesame husk contains vegetable compounds, including polyphenols such as anthocyanin, lignin, and vitamin B₁, all of which can improve immunity and anti-oxidation effects, inhibit cancer cell multiplication (e.g., mastocarcinoma and skin carcinoma), and enhance cell vitality, thus making cells unlikely to cancerate (Mohdaly, Smetanska, Ramadan, Sarhan, & Mahmoud, 2011; Sain & Panthapulakkal, 2006; Suja, Abraham, Thamizh, Jayalekshmy, & Arumughan, 2004; Suja, Jayalekshmy, & Arumughan, 2005).

This report describes a systematic investigation of the biocompatibility, mechanical and thermal properties, and biodegradability of SH composites with PTT and glycidyl methacrylate-grafted poly(trimethylene terephthalate) (PTT-g-GMA). The biocompatibility of composites was characterized using a cytotoxicity assay, cell surface adhesion, and the soluble collagen assay. The results of this study will be applicable to the development of biomedical materials and serve as a reference for future-developed biomedical composites.

2. Experimental

2.1. Materials

PTT was supplied by Shell Chemical Co. (Singapore); glycidyl methacrylate (GMA), benzoyl peroxide (BPO), and dimethyl sulfoxide (DMSO) were purchased from Sigma-Aldrich Chemical Inc. (St. Louis, MO, USA). 3-(4,5-Dimethylthiazol-2-yl)-2,5-diphenyltetrazoliumbromide (MTT) was obtained from Promega (Madison, WI, USA). Fetal bovine serum (FBS) and Dulbecco's modified Eagle's medium (DMEM) were purchased from Gibco-BRL (Gaithersburg, MD, USA). All buffers and other reagents were of the highest purity commercially available. SH was obtained from Tainan (Taiwan).

2.2. Preparation and testing of PTT/SH composites

2.2.1. PTT-g-GMA copolymer

The grafting reaction of GMA onto PTT is illustrated in Scheme 1(A). A mixture of GMA and benzoyl peroxide was added

in four equal portions at 2-min intervals to molten PTT to allow grafting to take place. The reactions were performed in a nitrogen (N₂) atmosphere at 45 ± 2 °C. Preliminary experiments showed that reaction equilibrium was attained in less than 12 h. Thus, reactions were allowed to progress for 12 h under stirring at 60 rpm. The product (4 g) was dissolved in 200 mL of refluxing phenol/tetrachloroethane solution (60:40 v/v) at 40 ± 2 °C, and then the solution was filtered through several layers of cheesecloth. The phenol/tetrachloroethane-soluble product in the filtrate was extracted five times using 600 mL of cold acetone per extraction. The GMA loading of the phenol/tetrachloroethane-soluble polymer was determined by titration and expressed as a grafting percentage as follows. Approximately 2 g of copolymer was heated for 2 h in 200 mL of refluxing phenol/tetrachloroethane solution. After the sample was completely dissolved, 0.8 mL of hydrogen chloride in 5 mL diethyl ether (1.0 M) was added to open the epoxide rings of PTT-g-GMA. Then, the excess hydrogen chloride (HCl) was titrated with 0.05 M methanolic sodium hydroxide (NaOH) solution using a phenolphthalein indicator. The grafting percentage was calculated using the following equations (Cho, Eom, Kim, & Park, 2008):

$$\text{Grafting percentage (wt\%)} = \frac{(0.6 \times 10^{-3} - 0.05 \times 10^{-3} V_s) \times 142.15 \text{ g/mol}}{2} \times 100, \quad (1)$$

where V_s is the volume of NaOH solution for titration (mL) and 142.15 g/mol is the molecular weight of GMA. The grafting yield was 2.11 wt% for BPO and GMA loadings of 0.3 and 10 wt%, respectively.

2.2.2. SH processing

SH was extracted as a by-product of sesame husk processing from 4 to 6 months and was supplied by Tainan of Taiwan. As shown in Scheme 1(B), purification consisted of immersing 60 g ground and dried SH in 1000 mL distilled water for 2 days to remove any water-soluble components. The product was then dried at 50–60 °C for 2 days under vacuum. The resulting brown fragments were 0.1–0.5 cm long. The fibers were dried, ground, and sorted. After grinding, the fiber mixture consisted of a fine brown powder. The samples were passed through 300-mesh and 400-mesh sieves, air-dried for 2 days at 50–60 °C, and vacuum-dried for at least 6 h at 100–110 °C until the moisture content fell to 5 ± 2%.

2.2.3. Composite preparation

Prior to composite fabrication, SH samples were cleaned with acetone and dried in an oven at 105 °C for 24 h. Composites were prepared in a Plastograph® 200-Nm mixer W50EHT with a blade rotor (Brabender, Dayton, OH, USA). The blends were mixed between 220 °C and 230 °C for 20 min at a rotor speed of 50 rpm. Composite samples were prepared with SH:PTT or SH:PTT-g-GMA mass ratios of 10/90, 20/80, 30/70, and 40/60. Residual GMA in the PTT-g-GMA reaction mixtures was removed via acetone extraction prior to the preparation of PTT-g-GMA/SH composites. After mixing, the composites were pressed into thin plates using a hot press and placed in a dryer for cooling. These thin plates were cut to standard sample dimensions for further characterization.

2.2.4. Characterization analyses

The composites were characterized using Fourier-transform infrared spectroscopy (FTIR) and ¹³C nuclear magnetic resonance (NMR) to identify bulk structural changes induced by the maleic anhydride moiety. Solid-state ¹³C NMR was performed using an AMX-400 NMR spectrometer (Bruker, Billerica, MA, USA) and was obtained at 100 MHz under cross-polarization while spinning at the magic angle. Power decoupling conditions were set with a 90° pulse and a 4-s cycle time. Infrared spectra of the samples were obtained using an FTS-7PC FTIR spectrophotometer (Bio-Rad, Hercules, CA,

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