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Red algae fibre/poly(butylene succinate) biocomposites: The effect of fibre content on their mechanical and thermal properties

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Abstract

The red algae (*Gelidium Elegance*) fibre was examined as a reinforcement of biocomposite. The extracting and bleaching process of the fibre from red algae were effective for both removal of mucilage materials and fiberization of red algae fibre. The bleached red algae fibre (BRAF) showed very similar crystallinity to the cellulose and also higher thermal stability with the maximum thermal decomposition temperature of 359.3 °C. Poly(butylene succinate) (PBS) biocomposites reinforced with BRAF have been fabricated with varying BRAF contents by a compression molding method and their mechanical and thermal properties have been studied. The storage modulus of PBS matrix and the thermomechanical stability are markedly improved with increasing the BRAF content, showing a maximum value of storage modulus and the least coefficient of thermal expansion value at 50 wt% of fibre loading. This work suggests that red algae fibre can be effectively used as a reinforcement for biocomposites and contribute to develop environmentally friendly biocomposites.

Keywords: A. Short fibre composite; B. Thermal Properties; B. Mechanical properties; B. Thermomechanical properties; D. Dynamic mechanical thermal analysis (DMTA)

1. Introduction

In recent years, with emphasis of growing environmental awareness, natural fibres have been increasingly used as reinforcing materials in biocomposites. Biocomposites utilize natural fibres as reinforcement and polymers as matrix for composites. Advantages of natural fibres over traditional reinforcing materials such as glass and carbon fibres are low cost, low density, renewable, biodegradability, etc. For this reason, biocomposites have several advantages such as eco-friendly, lightweight, energy saving and carbon dioxide reduction characteristics [1–6].

Natural fibres, generally plant-based fibres like flax, jute, sisal and kenaf have been more frequently utilized and studied so far and a large number of literatures have been reported on biocomposites based on these plant-based natural fibres earlier [7–11]. However, the use of marine-plant natural fibres such as red or green algae fibres as reinforcing materials for biocomposite has been rarely reported. The utilization of red algae has usually been suggested in the area of polyelectrolytes, pharmaceutical products, human nutrition, antimicrobial activity and polysaccharide production, etc. [12].

In the cells of red algae, there are large amount of mucilage materials such as agarose and carrageenan, which can be easily extracted out by hot water and weak, environment-friendly chemical treatment. The extracted mucilage materials have been used for food and other application; however, almost no attention was given to the remnant after extraction, algae fibre. Some commercial agar plants in Asia utilize the algae fibre as natural fertilizer. Therefore, utilizing the algae fibre as a reinforcing material is attractive for developing a new value-added material in terms of economics and eco-friendliness [13–15].

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Polybutylene succinate (PBS) is one of the aliphatic thermoplastic polyesters with a range of desirable properties including melt processability, and both thermal and chemical resistance. Besides, PBS can be naturally degraded into the environment by bacteria and fungi [16–18]. Therefore, the combination of algae fibre and PBS can produce the environmentally friendly biocomposite.

The objectives of this study are to fabricate biocomposite using the red algae fibre as reinforcing material of biocomposite and to investigate the effect of reinforcement contents on the thermal and mechanical properties of the biocomposite in terms of thermal stability, thermal expansion, dynamic mechanical properties, and flexural properties, including microscopic observations. Also, we examined a new manufacturing process to disperse of the BRAF in the biocomposite effectively.

2. Experimental

2.1. Materials

Poly(butylene succinate) (PBS, Enpol G-4500, Ire Chemical Ltd., Korea), biodegradable polymer, used as a matrix of biocomposites. It has a melt flow index of 29 g/10 min at 190 °C and a specific gravity of 1.22 g/cm^3 in the form of pellets with a melting point of 115 °C.

The BRAF was obtained by extracting and bleaching the red algae (Jeju Island, Korea, given a scientific name of *Gelidium Elegance*). The average length and diameter of red algae were 6–11 cm and 0.7–1.6 mm, respectively.

The crystalline cellulose powder (Aldrich Co., bulk density of 0.6 g/ml at 25 °C) was used as a reference to examine the crystalline and thermal properties of BRAF.

2.2. Extraction and fiberization of red algae fibre

The extracting of red algae was performed by boiling red algae in the water for 1 h and filtering the red algae. The filtered red algae were bleached with oxidizing agents such as 5% dioxide chlorine and 35% hydrogen peroxide both at 80 °C for 2 h [12]. Complete fiberization red algae to red algae fibers was accomplished during the bleaching process, and no further fiberization was necessary. The bleached fibre was washed with the excess water and dried in the oven at 105 °C for 24 h. The mucilage materials removed from the red algae easily with boiling water. The bleaching process with oxidizing agents was effective for the removal of remained mucilage from the extracted fibres. The fiberization of dried BRAF for biocomposite was performed using a high-speed pulverizer (Ultra centrifugal mill, Germany) with 6000 rpm for 40–60 s.

2.3. Fabrication of biocomposites

Both PBS and BRAF were dried at 80 °C for 24 h and the pulverized PBS powder was mechanically mixed with BRAF for 30 s using a kitchen mixer. The mixture of PBS and BRAF was placed in the steel mold and BRAF reinforced PBS biocomposites were fabricated by a compression molding technique. The molding compound was heated with heating rate of 2 °C/min. The pressure of 6.89 MPa applied to the compound at 130 °C and maintained for 10 min, then cooled down to room temperature by circulating cold water. For investigating the effect of fibre loading on the thermal and mechanical properties of the biocomposites, five levels of fibre loading were prepared; 20, 30, 40, 50, and 60 wt%.

2.4. Analysis

2.4.1. Optical microscopy

Differential dimension of the red algae fibre and the wood fibre was observed by Optical video microscope (Alphasystec, ICS-305B, Korea) of 12 V/100 W halogen lamp equipped with the 1/3 in. SONY SuperHAD CCD™ image sensor.

2.4.2. X-ray diffraction analysis

Crystalline structure of the red algae fiber was analyzed by X-ray diffractometer (Rigaku-D/MAX 2000,Ultima$^+$, Japan) with 5°/min scan speed. Ni-filtered Cu Kα radiation ($\lambda = 1.54$ Å) at 30 kV and 20 mA.

2.4.3. Thermal stability analysis

The thermal stability of the red algae fibre was analyzed up to 500 °C with a purging nitrogen gas of 100 ml/min using a thermogravimetric analyzer (TGA Q 500, TA Instruments). A heating rate of 10 °C/min was used. About 20 mg of each specimen was loaded for each measurement. Derivative thermogravimetric (DTG) curves were also recorded.

2.4.4. Dynamic mechanical analysis

The storage modulus and the glass transition temperature of each biocomposite were measured by a dynamic mechanical analyzer (DMA Q 800, TA Instruments) using the single cantilever mode at a fixed frequency of 1 Hz under the nitrogen atmosphere. The oscillating amplitude used was 0.2 mm and the heating rate was 5 °C/min. The glass transition temperature was determined from the peak temperature of each tan δ curve. Before each measurement, the instrument was calibrated to have the correct clamp position and compliance. The specimen dimensions were 30 mm × 10 mm × 1.8 mm.

2.4.5. Thermomechanical analysis

The coefficient of thermal expansion (CTE) was measured by heating the specimen from room temperature to 100 °C at a heating rate of 5 °C/min under the nitrogen atmosphere with a flow rate of 100 ml/min. The prove was applied 0.05 N loading and it measured strain and temperature of specimens. The coefficient of thermal expansion was taken as the linear slope of the strain–temperature curve with a thermomechanical analyzer (TMA Q 400,