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Effect of duration of sonication during gelatinization on properties of tapioca starch water hyacinth fiber biocomposite

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ABSTRACT

This paper characterizes properties of biocomposite sonicated during gelatinization. The biocomposite consisted of tapioca starch based plastic reinforced by 10% volume fraction of water hyacinth fiber (WHF). During gelatinization, the biocomposite was poured into a rectangular glass mold then vibrated in an ultrasonic bath using 40 kHz, 250 W for varying durations (0, 15, 30, and 60 min). The resulting biocomposite was then dried in a drying oven at 50 °C for 20 h. The results of this study indicate that a biocomposite with optimal properties can be produced using tapioca starch and WHF if the gelatinizing mixture is exposed to ultrasound vibration for 30 min. After this vibration duration, tensile strength (TS) and tensile modulus (TM) increased 83% and 108%. A further 60 min vibration only increased the TS at 13% and TM at 23%. Moisture resistance of the biocomposite after vibration increased by around 25% reaching a maximal level after 30 min. Thermal resistance of the vibrated biocomposites was also increased.

1. Introduction

Degradable bioplastic materials show potential as an environmental pollution-reducing replacement for synthetic polymers. Polysaccharide based bioplastics provide promise for food protection and preservation applications [1]. The use of biopolymers based on polysaccharides substance provides promise for innovative applications in food protection and preservation [2]. However, starch based thermoplastic has low strength and thermal resistance, high moisture absorption, low resistance to microbial activity [3–5]. Many attempts to overcome these weaknesses have been trialed. Mixing starch with ramie crystallites and cellulose nanofibers from cassava bagasse has been found to improve the mechanical properties and moisture resistance of biocomposites [6,7]. When mixed with plant fibers the resulting biocomposite could provide an attractive substitute for many synthetic polymers [8]. The fiber of the water hyacinth plant with its high cellulose content is one attractive reinforcement material for starch biocomposites, as it is fast growing and abundantly available [9,10]. Some previous reports have used these fibers with polyester to produce composites with viable mechanical properties [10–13].

The introduction of air bubbles and agglomeration in the starch matrix during pouring decreased mechanical properties of the biocomposite [14–16]. Last works reported that applying of ultrasound during preparing sample has created good filler dispersion and minimized agglomeration in matrix [15,17,18]. Previous studies have shown that applying ultrasound during sample preparation improved performance of the biocomposite, because agglomeration could be minimized and good filler dispersion created [19,20]. Ultrasound process was found superior in terms of its efficiency of breaking the strong agglomerates of MWCNTs along with their homogeneous distribution in the epoxy matrix [20]. Ultrasonication created good dispersion of cellulose nanofibers in polyvinyl alcohol, yielding high mechanical properties of the composite [17,21]. A previous study reported that SEM fractured surface was smooth which signals good dispersion and homogeneity in the composite [21]. Even an increase in concentration of the nanofibers up to 10% did not lead to the emergence of signs of aggregation of fibers in a micron-scale. In all cases, no signs of fiber pull-out could be spotted.

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Gelatinization during the manufacture of starch based plastic is achieved by heating and gently stirring the starch dispersion in excess water, and is characterized by hydration, swelling, and loss of birefringence and crystallinity of the starch chains [22,23]. However, undamaged amylopectin-rich fractions often remain resulting in incomplete starch solubility. These insoluble fractions, known as ghosts, are dispersed throughout the film's microstructure in a similar way to micro-filler. Cheng et al. [24] studied the impact of ultrasonic treatment on properties of starch film-forming dispersion and the resulting film. Ultrasound treatment ruptured the swelling starch granules disintegrated all the ghosts in gelatinized maize starch dispersions increased the amount of solubilized amylose and enhanced free mobility of polymers in the starch dispersions, resulting in a drastic decrease in apparent viscosity and increase in solubility even for a 10% starch dispersion. This process yielded films with good transparency, improved tensile strength and moisture resistance, and stronger structure [24].

An ultrasonic bath generates mechanical forces as a result of cavitation. The collapse of the microbubbles created by cavitation produces sound waves which propagate through the solution resulting in strong shear forces [5,18,25,26]. Several works have reported the effect of intensity and time of ultrasound during formation on the performance of a range of composites [5,21,23,27–32]. However, no investigations of the impact of ultrasound duration in a starch based biocomposite sonicated during gelatinization exist. There is insufficient information on the effect of vibration of gelatinized biocomposites. Therefore, in this present research, the effect of various vibration durations during gelatinized on the properties of a 10% WHF pulp and tapioca starch biocomposite was studied. The benefit of this study is to discover the duration of 40 kHz, 250-W ultrasonic bath radiation necessary to optimize both the mechanical properties of gelatinized 10% WHF pulp and tapioca starch, and energy consumption. The properties of the biocomposite measured were tensile testing, thermal stability, and moisture absorption. Meanwhile, X-ray diffraction (XRD), Fourier transform infrared spectroscopy (FTIR), SEM photograph of the surface was used to identify chemical features, interactions of all functional groups, and defects in the biocomposite.

2. Materials and methods

2.1. Extraction of WHF

WHF obtained from a swamp in 50 Kota district, Indonesia was cut into 1 cm lengths and cleaned in fresh water. These were then placed in a ventilated drying space with plastic roof and walls exposed to sunlight for 5 days until dry. The dried WHF samples were treated with 25% NaOH solution in a digester for 6 h at about 130 °C, and 2 bar. Finally, the pulp was washed until the pH was neutral, and blended for 10 min using an electric mixer. The pulp was screened by using a printing screen with T61 mesh, then dried using sunlight.

2.2. Preparation of biocomposite

Local commercially produced *Cap Pak Tani* brand tapioca powder was used as a matrix for the biocomposite. This tapioca powder consists of 19% water, 15% amylose, and 85% amylopectin. 1 g WHF was mixed with 140 ml of the water. 10 g tapioca powder and 3 ml of glycerol were poured into the mixture which was then mixed with a (Daihan HG 15D-Set A) homogenizer for 5 min at 5000 rpm. The solution was gelatinized using a magnetic stirrer at 150 °C, 500 rpm for 18 min. Finally, the gelatinizing biocomposite was poured into a rectangular glass mold and placed in the ultrasonic bath (model number PS-70AL, 40 kHz and 420 W). The mold was only vibrated at 250 W, 40 kHz, and 100% amplitude with various duration for 15, 30, 60 min respectively. For duration 30 and 60 min, water in the bath was replaced every 15 min to keep the water temperature lower than 40 °C. A Universal oven (Memmert UN-55) was used to dry the samples at 50 °C for 20 h.

2.3. Moisture absorption

A piece of the biocomposite sample was dried in oven drying until a constant weight was achieved. It was weighed using precision balance (Kenko) with 0.1 mg accuracy then stored in a closed chamber with 99% relative humidity (RH) and 25 °C for 30 min, taken out and weighed again. Moisture absorption of the sample was calculated by using [33]:

$$\textit{Moisture absorption}\,(\%) = \frac{(w_h - w_o)}{w_o} \times 100 \quad (1)$$

where w_h is weight of moist sample and w_0 initial weight of dried sample.

2.4. Tensile testing

Tensile strength of the biocomposite was measured at room temperature using a Com-Ten testing machine (95T Series) with tensile speed of 3 mm/min. Five biocomposite samples from each vibration duration were measured according to standard ASTM D638 Type I. Thickness and width of the film were measured using a dial micrometer to 1 μm accuracy. The tensile strain was measured at the point of fracture of the sample.

2.5. Thermal gravimetric analyzing (TGA/DTG), differential scanning calorimetry (DSC)

TGA and DTG measurement of samples were performed using a thermal analyzer (TGA Q500 V20.6, USA). The samples were weighed between 4 and 5 mg. The scans were run from room temperature up to 600 °C at a heating rate of 10 °C/min under nitrogen flow of 25 ml/min. DSC was measured using TA Instrument (Model Q20) from room temperature up to 280 °C at 10 °C/min with nitrogen flow rate of 50 ml/min. The weight of each sample was 5–10 mg.

2.6. FTIR characterization

FTIR characterization was performed by using a Perkin Elmer Frontier spectrometer to determine the functional groups of the TSB and TSB/WHF composite. Samples were formed in the form of thin sheets and scanned in the spectral range 4000–600 cm^{-1}.

2.7. SEM observation

SEM photography was performed using a Hitachi 3400 N series SEM to scan the fracture surface of the tensile sample.

2.8. X-ray diffraction

X-ray diffraction (XRD– PANalytical Xpert PRO) measurement was performed using CuKα radiation (λ = 0.1542 nm) scanning from 10 to 100° with a generator setting of 40 kV and 30 mA. Percentage of crystallinity index (CI) was measured as follows [34]:

$$CI\,(\%) = \frac{(I_{200} - I_{am})}{I_{200}} \times 100 \quad (2)$$

where I_{200} is intensity of the peak corresponding to cellulose I, and I_{am} is intensity of the peak of the amorphous fraction.