

Transforming fish scale waste into an efficient filler for starch foam

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ABSTRACT

In this work modified fish scale waste (FS) was used as a filler in order to improve the properties of starch foam prepared by a baking process. The FS was modified under two calcination conditions: at 500 °C for 5 h and at 700 °C for 3 h. The FS powder calcined at 700 °C for 3 h (FS700-3) had higher Ca content and lower protein content and was the filler chosen for experimental study of the effects of its addition on the properties of the starch foam. The addition of FS700-3 to the starch foam produced a more expanded structure due to FS700-3 efficiently supported the growth of existing cells during nucleation and bubble growth. Moreover, the flexural stress at maximum load of the starch foam increased from 1.03 MPa to 1.54 MPa after adding FS700-3 at 10 wt% because of the good adhesion between the two components.

1. Introduction

The environmental pollution problems presented by single-use foam trays made from polystyrene (PS) have focused attention on the environmental advantages of foam made from starch. Biodegradable foams can be made from various types of starch such as cassava, wheat, sago and corn, which are classical renewable materials (Glenn, Orts, & Nobes, 2001; Pimpa et al., 2007; Polat, Uslu, Aygün, & Certel, 2013; Pornsuksomboon, Hollo, Szecsenyi, & Kaewtatip, 2016). Starch foams can be prepared using extrusion, microwave heating and baking (Soykeabkaew, Thanomsilp, & Suwantong, 2015). The last method is widely used to produce starch foam in a two-step process: starch gelatinization and water evaporation. However, starch foam has a number of drawbacks including high water absorption, poor mechanical properties and poor thermal stability. Many published research works have described various modifications that attempted to overcome the limitations of starch foam. These modifications have included corn starch cross-linked with glyoxal, citric acid-modified cassava starch, starch acetate and oxidized starch (Polat et al., 2013; Pornsuksomboon et al., 2016; Xu & Hanna, 2005; Zhang et al., 2017). Although modification has produced starch foams with high water resistance and better mechanical properties, some preparations are expensive and use hazardous chemical reagents. Other studies have shown that plant proteins (gluten and zein), beeswax and palm oil added into starch foam improved the mechanical properties and water sensitivity as well as thermal stability (Kaisangsri, Kerdchoechuen, & Laohakunjit, 2014; Kasemsiri, Dulsang, Pongsa, Hiziroglu, & Chindaprasirt, 2017; Polat et al., 2013; Salgado, Schmidt, Molina Ortiz, Mauri, & Laurindo, 2008). However, some of

them increase the viscosity of the starch batter producing a high density starch foam with a dense structure.

Nowadays, research often focuses on polymer composites that use residual waste materials from agriculture (coir, corn cob and straw) (de Farias et al., 2017; Huang et al., 2017; Kalaoglu, Unlu, & Galioglu Atici, 2016), thermal power plants (fly ash) (Goh, Valavan, Low, & Tang, 2016; Kang et al., 2017) and the industry (pearl shell, shrimp shell, fish scale, eggshell and cuttlebone) (Boronat, Fombuena, Garcia-Sanoguera, Sanchez-Nacher, & Balart, 2015; Ji, Zhu, Jiang, Qi, & Zhang, 2009; Klungsuan, Jarerat, & Poompradub, 2012; Nourbakhsh, Ashori, & Kazemi Tabrizi, 2014). These materials have advantages of low cost, renewability, abundance and environmental friendliness and bio-calcium filler from shell waste is especially efficient at improving the properties of various polymers such as epoxy resin, polypropylene and natural rubber.

However, fish scale waste from the food industry is the largest source of bio-calcium and, according to Yang et al. (2014), most fish scales have similar constituent materials: calcium-deficient hydroxyapatite (HAP) and type I collagen fibrils. Nevertheless, there have been fewer reports of the use of fish scale waste as a filler for polymer composites.

Nourbakhsh et al. (2014) reported that fish scale waste could increase the biodegradation rate of polypropylene and our previous work also reported that the addition of fish scale waste could increase the tensile strength of a wheat gluten-based bioplastic but there have been no reports of the effect of fish scale waste on the properties of starch foam (Thammahiwes, Riyajan, & Kaewtatip, 2017a). Ji et al. (2009), reported that the biopolymeric materials from the surface of pearl shell

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negatively affected the mechanical properties of polymer composites. Proposing calcination as an effective method of eliminating these biopolymeric materials, they explained that the greatest improvement in mechanical properties was obtained in an epoxy resin composite prepared with pearl shell powder calcined at 700 °C for 3 h. This result was attributed to the complete degradation of the layered biopolymeric materials during calcination.

Therefore, this work comprises two objectives, the first of which is an evaluation of the effects of the different calcination conditions on the morphology, crystallinity, N and Ca content of the fish scale powder. The second objective is an investigation of the effect of calcined fish scale powder on the physical, mechanical and thermal properties of starch foam for comparison with the effects produced by a filler of crude fish scale powder.

2. Materials and methods

2.1. Materials

Native cassava starch (PD 10369), kindly supplied by Siam Modified Starch Co., Ltd was dried at 105 °C for 48 h in an oven and kept in a desiccator prior to use. Glycerol was from Ajax Finechem. Guar gum and magnesium stearate were purchased from Sigma-Aldrich, Inc and used as received. Fish scales were kindly supplied by Kingfisher Holding Ltd. (Songkhla, Thailand). The fish scales were washed in water to remove any impurities, then dried in a hot air oven at 70 °C for 24 h and ground in a mortar grinder, (FRITSCH, Germany). After grinding, the powder was dried in a hot air oven (105 °C) until a constant weight was achieved and kept in a desiccator prior to use.

2.2. Foam preparation

Native cassava starch, fillers (crude and calcined fish scale powder, 10 wt%), magnesium stearate (2 wt%) and guar gum (1 wt%) were mixed using a mixer (Kenwood, KM 262) at room temperature for 10 min. Distilled water (100 wt%) and glycerol (5 wt%) were added to the mixture, which was further mixed for 10 min. After that, 100 g of the batter was applied to a mold (15 cm × 10 cm × 4 mm) using a compression molding machine operating at 220 °C for 5 min. Before testing, the starch foams were stored for 14 days at 70.00 ± 2.07% Relative Humidity (RH) and 29 ± 1 °C.

2.3. Preparation of the calcined fish scale powder

The fish scale powder samples were calcined in a furnace under 2 different conditions: at 500 °C for 5 h (FS500-5; low temperature and long period of time); and at 700 °C for 3 h (FS700-3; high temperature and short period of time). The calcined fish scale powder was packed in polyethylene plastic bags and kept in a desiccator prior to use. The optimum calcination conditions for the removal of collagen and other organic compounds from the structure of the fish scale powders were investigated by determination of Ca and N contents.

2.4. Characterization

Wide angle X-ray diffraction (XRD) studies were carried out using a Phillips diffractometer (Model PW, 1830) with copper as the target material. The scanning regions of the diffraction angle 2θ were 10°–80°, which covered all the significant diffraction peaks of the fish scale powders.

The morphology of the crude fish scale powder, calcined fish scale powder and fractured surfaces of the starch foams was observed by scanning electron microscope (SEM) (Quanta 400, FEI). All specimens were first coated with a thin layer of gold.

Ca and N contents were determined using an ICP-OES (Method 985.01) (Optima 4300 DV, USA) and CN Analyzer (Method 993.13)

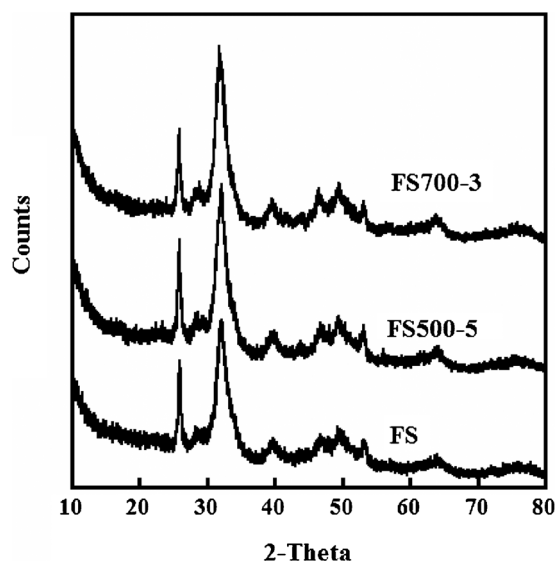


Fig. 1. Wide angle X-ray diffraction patterns of FS, FS500-5 and FS700-3.

(CN628, USA).

The density was calculated as the relationship between weight and volume.

Flexural tests were performed according to ASTM D790 on a universal testing machine (Instron model 3365) with a 100 N load cell and a crosshead speed of 2.5 mm/min. Dimensions of the specimens were 130 × 30 × 4 mm and the span length was fixed at 80 mm. Testing was performed at 25 ± 3 °C and 55 ± 2%RH. At least three samples were used to calculate the average and standard deviation.

The thermal decomposition temperatures of the samples were determined using a PerkinElmer TGA 7 operated at a heating rate of 10 °C/min from 50 to 1200 °C under a nitrogen atmosphere.

3. Results and discussion

3.1. Characterization of crude and calcined fish scale powder

The wide angle X-ray diffraction patterns of crude and calcined fish scale powder in the 2θ between 10°–80°, shown in Fig. 1, confirm the hydroxyapatite structure (PDF#01-086-0740; Hydroxyapatite; Calcium Phosphate Hydroxide, Ca₅(PO₄)₃(OH)) of crude fish scale powder (FS). FS500-5 and FS700-3 calcined fish scale powder both present similar XRD patterns to the crude fish scale powder. This behavior is in agreement with the results of the work of Chakraborty, Bepari, and Banerjee (2011) who presented XRD patterns of waste fish scale calcined at temperatures ranging from 600 to 1000 °C. The transformation of hydroxyapatite into β-Ca₃(PO₄)₂ only occurred after calcination at 900 °C or above. Therefore, the experimental calcination conditions used in this work did not change the hydroxyapatite structure of the fish scale.

It is well known that the microscopic surfaces of the filler are key factors that affect adhesion between the filler and the polymer matrix and, consequently, the final properties of the polymer composite (Raabe et al., 2015). SEM image showed microscopic fibrils on the surface of FS (Fig. 2(a)) and this characteristic is consistent with our previous work, which confirmed these fibrils as collagen by FTIR (Thammahiwee, Riyajan, & Kaewtatip, 2017b). The morphology of collagen fibrils in the fish scale structure has also been shown by Yang et al. (2014). However, in contrast to the crude fish scale powder, the microscopic surfaces of both the calcined fish scale powders (Fig. 2(b) and (c)) were cleaner and smoother. Thus, as was expected, the calcination process effectively removed the collagen and other organic compounds from the fish scale powder. The removal of the collagen and other organic compounds

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