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Evaluation of Kenaf Fibres Reinforced Starch Based Biocomposite Film through Water Absorption and Biodegradation Properties

Zuraida Ahmad, Nur Humairah Abdul Razak, Nur Suraya Mohd Roslan and Nazariah Mosman

Department of Manufacturing and Materials Engineering,
Kulliyyah of Engineering, International Islamic University Malaysia,
P.O. Box 10, 50728 Kuala Lumpur, Malaysia

*Corresponding author: nurhumairah87@gmail.com

Abstract: *The effect of kenaf short fibres (KF) on thermoplastic sago starch (TPSS) film was investigated for its water absorbency, biodegradability and structure morphology. The TPSS reinforced KF biocomposite film was prepared via casting method at varying TPSS/KF weight percentage of 100/0, 95/05, 90/10, 85/15 and 80/20, while the plasticiser was at a constant amount of 30%, relative to the weight of starch. The result of water absorbency tended to increase for both pure TPSS (100/0) and biocomposite film with prolonged testing days and that the incorporation of KF improves the water resistance of the biocomposite film. Moreover, biocomposite film at 15 wt% of KF content demonstrated the highest weight loss of 39% after 40 days of soil burial for biodegradability test. Scanning Electron Microscopy (SEM) analysed the morphology of the film and revealed good wetting of matrix over the fibres that lead to better adhesion of fibres with the TPSS matrix.*

Keywords: Sago starch, kenaf fibres, biocomposite film, thermoplastic starch, biodegradability, structure morphology

1. INTRODUCTION

Despite being a very convenient and durable material, plastics generate numerous environmental problems. Over 200 billion pounds of plastics are produced each year with much of them used for packaging,[1] consequently leading the nondegradable plastics to end up as waste. Depletion of petroleum resources, landfill problem and pollutions add to the list of plastic drawbacks. This has led to an increasing interest in the production of bio-based polymers which are independent of fossil fuels and can help sustain the environment. One alternative is by blending the plastics with natural polymers such as starch.

Research on thermoplastic from various starches has been widely studied such as potato, corn, wheat and cassava starch.[2] Sago starch (*Metroxylon sagu*) from the trunk of sago palm is also a natural carbohydrate semicrystalline

polymer with polysaccharide chains consisting of 27% linear chained amylose and 73% branched chain amylopectin.[3,4] These chains facilitate the formation of sago starch into a thermoplastic with the addition of a plasticiser through the disruption and gelatinisation of starch polymer chains at a specific critical temperature.[5] The possibility of converting sago into thermoplastic starch has gained considerable interest due to sago's unique characteristics such as having a satisfactory thermal stability, easy to gelatinise and moulded, and possess high viscosity.[6]

Despite being an environmental friendly material, thermoplastic starch, however has poor performance in water resistance and poor mechanical properties. In order to improve these properties whilst upgrading biodegradability, thermoplastic starch can be reinforced with natural fibres.[7] Natural fibres have been replacing synthetic fibres in various composite applications due to their lower weight and production cost, higher processing and operational safety, enhanced biodegradability and biocompatibility, higher strength and stiffness as well as ability to improve product's acoustic effect.[8,9]

Kenaf reinforced composites are one of the most current biocomposites researched areas. Kenaf fibre (KF) or *Hibiscus cannabinus* is a natural plant fibre that consists of stiff fillers called cellulose fibres that can replace wood. It grows rapidly under a variety of soils and weathers up to 3 m in height in approximately 5 months.[10,11] The hemicellulose and lignin within the KF contribute to the biodegradation, moisture absorption and thermal stability of the fibre.[7] KF also possesses excellent toughness as well as high aspect ratio, tensile strength and modulus.[7] Normally used in paper production, textiles and ropes, the versatility of KF has made it an attention as a reinforcement in thermoplastic composites.[7,11] It is reported that composites could attained a 50% increment in tensile and flexural strength when reinforced with KF.[12]

Therefore, the aim of the present work is to produce TPSS reinforced with KF in the form of biocomposite film whilst investigating the effect of the KF on the water absorption, biodegradability and morphology of the biocomposite film.

2. EXPERIMENTAL

2.1 Materials

Sago starch powder was purchased from Hup Seng Heng Sdn. Bhd., Malaysia. The particle size of the starch was approximately 50 μm with density of 0.577 g cm^{-3} and 13% moisture. The non-chemically treated kenaf bast fibres

were supplied by Institute Tropical Forestry and Forest Product (INTROP), Malaysia and were cut into an average length of 0.5 mm. The non-volatile glycerol, with 95% purity and density of 1.261 g cm^{-3} was supplied by Merck KGaA. Distilled water in this experiment acted as co-plasticiser.

2.2 Preparation of Biocomposite Film

The biocomposite film was fabricated by film casting method with TPSS/KF weight percentage of 100:0, 95:5, 90:10, 85:15 and 80:20. Glycerol was at constant amount of 30 wt%, relative to the weight of dry starch. The starch was first dried in a vacuum oven at 80°C for 24 h. Starch, glycerol and 150 ml of distilled water were mixed until starch gelatinisation at 80°C to form TPSS and KF were added to the solution. The solution was continuously stirred for 15 min and casted onto a glass plate to form a flat film. The composites film was subjected to dryness at 60°C for 24 h to obtain a dry film. The film sample was kept in the dessicator for 72 h prior to testing.

2.3 Characterisation

2.3.1 Water absorption test

The water absorption test was performed in accordance to ASTM D 570–98 where the film samples were first dried for 24 h at 50ºC, cooled in a desiccator and immediately weighed. The films were then immersed in distilled water at ambient temperature. After specific immersion period, the samples were removed from the water, gently wiped off all surface water with dry cloth and weighed to the nearest 0.001 g. The differences in initial and final masses for five replicates of sample for 3, 6, 9, 12, 15, 18 and 21 days were calculated using Equation 1.

$$\text{Water absorption (\%)} = [\,(M_{final} - M_{initial}) / M_{initial}\,] \times 100 \quad (1)$$

where, M_{final} = weight of the sample after immersion in water (g) and $M_{initial}$ = weight of the sample before immersion in water (g).

2.3.2 Biodegradability test

Film of 30 mm × 40 mm in size was buried in mineral soil at 25 ± 2ºC and 60 ± 5% relative humidity. The sample was buried at a depth of approximately 3 cm. In every specific time interval, the film was removed and rinsed gently with distilled water to remove the soil. The weight loss (%) due to degradation was determined every 10 days and calculated using Equation 2.

$$\text{Weight loss (\%)} = [\,(M_{initial} - M_{final}) / M_{final}\,] \times 100 \quad (2)$$

where $M_{initial}$ = weight of the sample before soil burial and M_{final} = weight of the sample after soil burial.

2.3.3 Scanning electron microscopy (SEM)

The surface structure or morphology of the non-reinforced and reinforced kenaf biocomposite films was visualised by SEM (Quanta 200 MK2) at 5 kV. The samples were sputter coated with gold layer to avoid electrostatic charging during examination.

3. RESULTS AND DISCUSSION

Biocomposite film performance is influenced by the composite absorption behaviour as well as the resilience and stability of the natural fibre under water. The water absorption of biocomposite film at different immersion time for various TPSS/KF ratios is presented in Figure 1 which shows that the unreinforced film (100% TPSS) has higher water absorption rate compared to the biocomposite film. This is in agreement with report elsewhere.[13] The unreinforced film absorbed 40% of water after three days of immersion.

Meanwhile, approximate reductions of 5%, 9%, 11% and 17% of water absorption were obtained with the increment of 5 wt%, 10 wt%, 15 wt% and 20 wt% of fibre content, respectively, as measured for the first three absorption days. During film immersion in water, the amylase and amylopectin structures in starch as well as the hydrogen and oxygen in both starch and glycerol formed bonds with water. Due to this, the unreinforced film with the highest starch content experienced the highest water absorption. In addition, the hydrophilic characteristic of fibre is less than that of starch[14] and the higher KF content formed a rigid network that hinders water diffusion during matrix swelling[13,15] resulting in lesser water absorption for the KF reinforced composites.

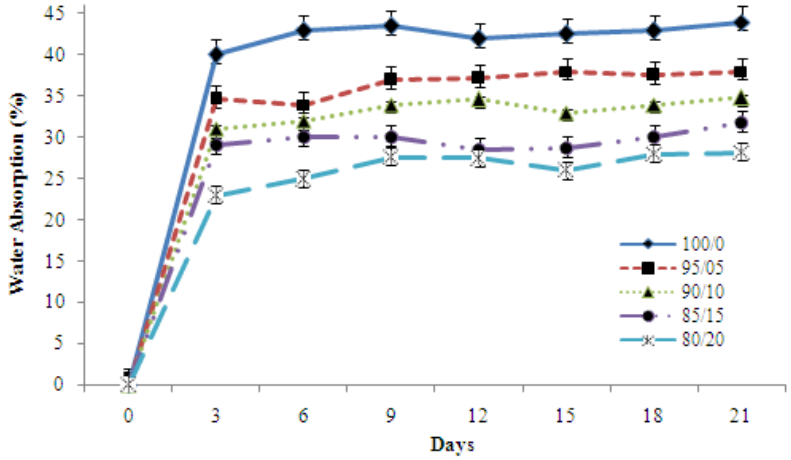

Figure 1: The water absorption of pure TPSS and biocomposite film at different KF loading at different immersion time

There is also an upward trend of water absorbency as a function of time. From Figure 1, all samples show an increment of water absorption with prolonged immersion time. A sharp absorption rate, however, occurred at the early immersion indicating a rapid water penetration into the films and a slower rate after day nine. The absorption later reached an equilibrium, demonstrating a Fickian's behaviour.[16] The rapid absorption is attributed to the water penetrating and capillary action phenomenon. The intermolecular forces between water and KF had caused the water to actively penetrate into the swelling KF[16] while the slower rate was induced by the fully occupied hydroxyl groups in the biocomposites molecular structure in the beginning of immersion days.

A soil burial test had been carried out on a laboratory scale to examine the film biodegradability in terms of weight loss. Biodegradable is used to describe materials that can be broken down or degraded by enzymatic action of natural living organisms. Figure 2 shows an increment of weight loss with burial period for the TPSS and biocomposites film.

At the beginning of the burial stage, all samples experienced an average weight loss of 24%. The diffusion of water from the soil that had caused swelling effect in the polymer boosted microorganism attack[17] and enhanced microbial activity. Microbial growth is dependent on the presence of water and oxygen. After 40 days, the percentage weight loss had increased between 34% to 39% for all the samples, with 15 wt% KF content biocomposites film experienced the highest weight loss. There was also an enhancement of biodegradability with the addition of KF content as cellulosic materials were favourly attacked by

microorganism in the soil environment.[18] The samples were hard, fragile and discoloured after 40 days of test, approving the degradation criterion, as shown in Figure 3.

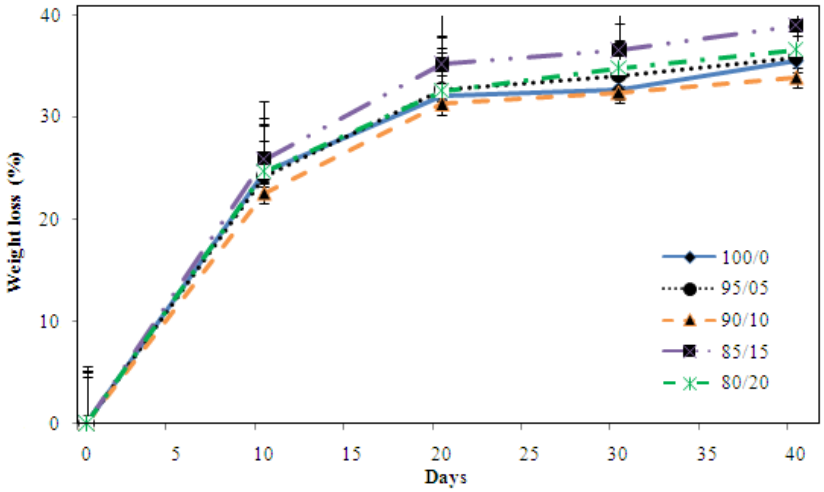

Figure 2: The biodegradability test of pure TPSS and biocomposite film at different KF loading at different burial period.

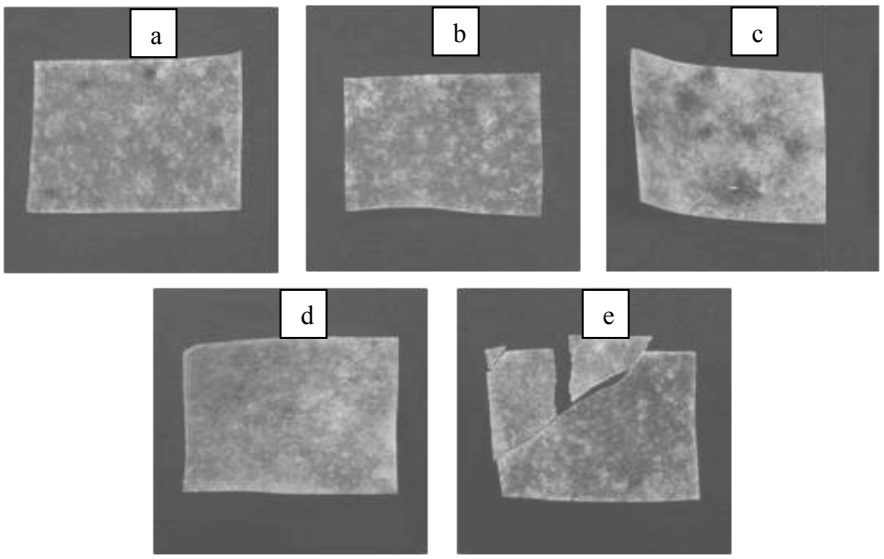

Figure 3: Films condition after 40 days of soil burial for TPSS/KF at (a) 100/0 wt%, (b) 95/05 wt%, (c) 90/10 wt%, (d) 85/15 wt%, and (e) 80/20 wt%.

The living microorganism succumb water from the swelling film for survivability which resulted in hard, fragile and brittle sample. There were also black spots on the surface of the samples indicating microorganism invasion. Furthermore, the biocomposite film with 20 wt% KF content had cracks and disintegrated, also presenting a sign of degradation.

Figure 4 presents the SEM micrograph of unreinforced and reinforced TPSS at 500× magnification. Pure TPSS film, as shown in Figure 4(a) shows a smooth and continuous surface indicating miscibility between the starch and the plasticisers while Figure 4(b) and (c) reveal the dispersion of KF in the TPSS matrix in a randomly oriented manner. At lower fibre content of 10 wt%, the surface is smooth, unlike the biocomposites film at 20 wt% of KF content. The KF are well coated with the TPSS matrix signifying a good adhesion of the fibres with the matrix. It can be postulated that the biocomposite film experienced good dispersion efficiency as there is no agglomeration of fibres observed. Furthermore, there is no phase separation in the polymer due to the same polarity of starch, plasticisers and fibres.

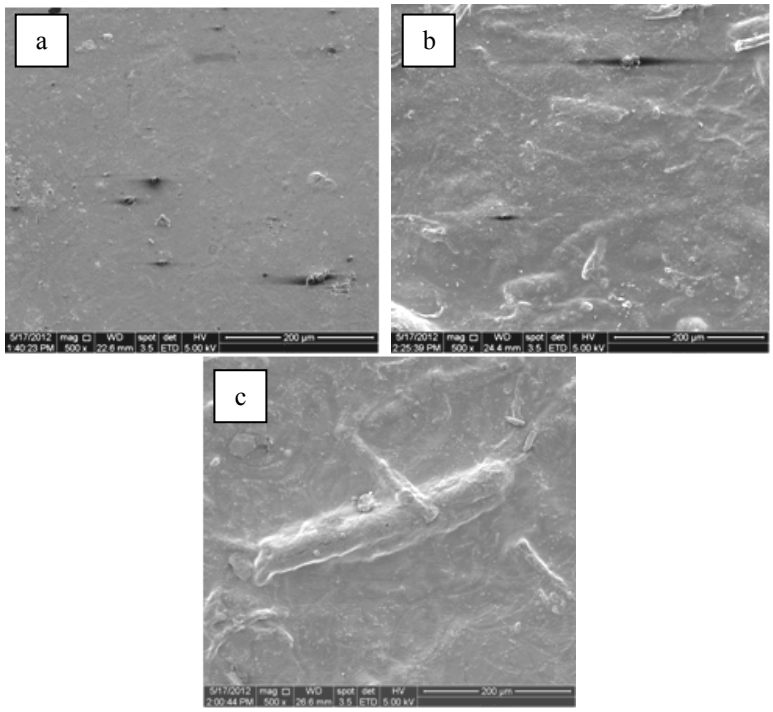

Figure 4: SEM micrographs of TPSS/KF at (a) 100/0, (b) 90/10 and (c) 80/20 wt%.

4. CONCLUSION

The reinforcement of KF with TPSS to produce biocomposite film significantly improves the water absorption, biodegradability as well as adhesion of fibre and matrix of the composites. The increment of KF content from 5 wt% to 20 wt% caused a reduction of 5% to 17% of water absorption as higher amount of fibre formed a rigid network that hindered the water diffusion into the matrix. Biodegradation of the biocomposites was contributed by the presence of KF that supported microorganism attack. The weight loss due to degradation had increased between 34% to 39% for all the unreinforced and reinforced samples after 40 days of soil burial. The micrographs results from SEM indicate good fibre-matrix affinity and unseparated phase in the polymer due to the same polarity of TPSS matrix and KF.

5. ACKNOWLEDGEMENT

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