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Properties of biodegradable foam composites made from coconut residue as a function of the reinforcing phase of cassava starch

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Abstract

The large amount of coconut residue produced by coconut producers and manufacturers has harmed the environment. Coconut waste was converted into coconut residue flour (CRF), which was used as a reinforcing material for biodegradable packaging development in order to decrease waste problems and increase the value of the circular economy. The effects of CRF content, ranging from 0 to 25 wt%, on the properties of foam-type composites made from cassava starch containing 10 wt% spent coffee grounds (SCG) were investigated. Flexural strength was raised to 2.53 MPa by adding CRF at 25 wt%. A thicker outer layer and denser foam structure were observed when the proportion of CRF in the samples was increased. The density of all samples was found to be 0.34-0.41 g/cm^3, which was in the same range as other starch-based foam composites. The water resistance of the samples was marginally enhanced by adding more CRF. The water absorption index (WAI) and water solubility index (WSI) of the samples were in the ranges of 5.10–8.95% and 2.16–5.05%, respectively. The soil burial test at 30 days showed that the weight loss of the samples increased from 74.88 to 100% with increasing CRF due to the high susceptibility of CRF to microbial spoilage. Based on the findings of this study, CRF could be a promising and environmentally friendly reinforcing material for biodegradable foam composites that could be used to replace expanded polystyrene in dry food packaging. This would be a sustainable solution to waste problems while increasing financial gains for coconut producers and manufacturers.

Keywords: Foam composite, Biodegradable packaging, Soil burial, Coconut residue, Cassava starch

1. Introduction

Expanded polystyrene (EPS) foam is a non-biodegradable synthetic polymer-based material that has been widely consumed in single-use packaging, particularly for fresh food and fast-food delivery. This is due to its ease of use, low cost, and excellent mechanical and thermal properties, as well as good resistance against moisture [1]. It is expected that global production of EPS will reach 11 Mt by 2023, resulting in a significant amount of post-consumer waste with limited opportunity for recycling [2]. Most of the non-biodegradable waste is burned or discarded in landfills or oceans, where it is slowly degrading into fragments of microplastics [3]. Eco-friendly and biodegradable packaging materials are being taken into consideration as competitive alternatives to non-biodegradable polymer foams as health and environmental concerns become more significant [4].

Starch-based materials have attracted interest and undergone extensive research to develop biodegradable packaging to replace conventional EPS foam materials since they are renewable, abundant, non-toxic, compostable, and reasonably priced [5]. Starch-based foam packing is created by mixing starch with water and other additives, then molding it under heat and pressure [6]. Starch-based foams have some disadvantages, namely a high sensitivity to moisture due to the hydrophilic characteristic of starch molecules, as well as poor mechanical and thermal properties [7]. Various additives, such as reinforcing materials derived from agro-industrial waste, have been added to the polymer matrix to improve these properties of the foam. According to Spada et al. [8], the incorporation of rice husk enhanced the overall mechanical properties of the foam samples while decreasing their density and water absorption in comparison to the starch foam without rice husk. Kasemsiri et al. [9] found that the water resistance of starch foam was significantly improved by adding 5–15 wt% of palm oil. Janaum et al. [10] reported that incorporating SCG and ZnO into starch-based foam composites improved their mechanical properties and water resistance. The samples had higher densities when more SCG was added, primarily because the mixture became less expandable and more viscous. Trongchuen et al. [11] also discovered that foam composites made from cassava starch, SCG, extracted SCG, and oregano essential oil demonstrated improved water resistance and a synergistic effect on antibacterial activity. The SCG contained important phenolic compounds like chlorogenic acid and caffeic acid. Based on the literature, SCG is an inexpensive reinforcing phase with advantageous properties for starch foam preparation. The SCG content in polymer composites was

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suggested at 10 wt%. Sugarcane bagasse and asparagus peel, both cellulose fibers derived from industrial waste, could be used as reinforcing materials to improve the properties of sweet potato starch-based foam trays [12]. In Engel et al.'s study [13], a foam composite based on cassava starch, a starch residue from cassava inner bark and grape stalks was successfully prepared. It showed better potential for use as single-use packaging and the container of low-moisture goods. This is one of the more sustainable methods of reducing waste and environmental issues.

Coconut residue is a fiber-rich byproduct from the extraction of coconut milk. It is estimated that at least 5 Mt of residue will be produced per year [14]. The substantial amount of such residue causes a major disposal problem creating a significant environmental issue [15]. This has inspired studies to explore the applications and use this waste by converting it into a value-added product. Coconut residue has been underutilized. It is used to a limited extent as a low-cost fertilizer and animal feed [16]. The use of coconut residue flour (CRF), which is made from dried and finely ground coconut waste, in food production has grown [17]. CRF can be used in place of or in combination with regular flour [18] in food products such as pasta [19], bread [20], and biscuits [21] since it is a rich source of protein, fat, carbohydrates, fiber, and minerals [22]. However, there has not been much research done on using CRF as a reinforcing material in the development of biodegradable foam packaging. Therefore, the objective of this research was to investigate the physical, mechanical, as well as biodegradation characteristics of foam composites based on cassava starch incorporated with CRF at levels ranging from 0 to 25 wt%, in order to use CRF more efficiently to produce value-added materials and increase the value of the circular economy.

2. Materials and methods

2.1 Materials

Cassava starch was purchased from Bangkok Interfood Company Limited, Thailand. Coconut residue after the extraction of coconut milk was supplied from a local coconut manufacturer in Prachuap Khiri Khan Province, Thailand. The residue was dried in the sunlight for several days and then further dried in an air-circulating oven at 60 °C for 24 h. After milling, CRF was sieved, resulting in sizes ranging from 180 to 250 µm. The chemical composition of the material includes 42.38% fat, 35.51% carbohydrates, 12.16% fiber, 5.87% protein, 2.49% moisture, and 1.59% ash, according to Association of Official Analytical Chemists' (AOAC, 2000) method. The SCG was supplied by a local coffee store in Prachuap Khiri Khan Province, Thailand. It was dried in an air-circulating oven at a temperature of 80 °C for 24 h. The dried powder was then sieved, resulting in particle sizes in the range of 180–250 µm. Palm oil was purchased from Lam Soon Public Company Limited in Thailand.

2.2 Preparation of starch-based foam composites

Cassava starch was combined with CRF at levels of 5, 15, and 25 wt%. Cassava starch and CRF were mixed homogeneously in a bowl with 10 wt% SCG for 3 min before water was added and mixed properly using a KitchenAid mixer, Electrolux EHM2000, at a temperature of 90 °C for 5 min to achieve gelatinization of starch. The dough obtained was mixed with 10 wt% palm oil for another 5 min. The compound obtained was placed in the mold and baked with a compression molding machine using a pressure of 4.82 MPa at a temperature of 190 °C for 8 min.

2.3 Characterization of starch-based foam composites

2.3.1 Density and morphology of the samples

The density of the foam composite samples was calculated from the relationship between mass and volume. All specimens were weighed carefully to an accuracy of 0.01 g. The volume of the specimen was calculated by multiplying the length, width, and thickness. The dimensions of the specimen were 130 mm x 30 mm x 7 mm. The average values were determined from ten replicates.

The morphology of starch-based foam composites was observed using a scanning electron microscope (SEM, JEOL JSM-6510LV) at an acceleration voltage of 20 kV. Before the SEM examination, all samples were fractured to obtain cross-sections and then coated with Au-Pd using a quorum sputter coater.

2.3.2 Water resistance of the samples

The water resistance of starch-based foam composites, in terms of water absorption index (WAI) and water solubility index (WSI), was investigated using a procedure similar to that described in the previous work [10]. A sample of about 1 g was added to 15 ml of distilled water in a pre-weighed centrifuge tube. The tube was then placed in a shaker at a temperature of 30 °C for 30 min. The sample was centrifuged for 30 min at a speed of 3000 rpm. The supernatant was poured into a pre-weighed aluminum dish and dried at a temperature of 105 °C for 24 h. The dry solid in the separated supernatant and the remaining sediment were weighed and used to determine WAI and WSI using Eqs. (1) and (2):

$$WAI = \frac{W_1 - W_2}{W} \times 100 \quad (1)$$

where W_1 and W_2 are the weights of the tube with the remaining sediment and the weight of the tube, respectively. W is the weight of the sample before testing:

$$WSI = \frac{W_{A1} - W_{A2}}{W} \times 100 \quad (2)$$

where W_{A1} and W_{A2} are the weights of the aluminum dish with the dry solid and the weight of the aluminum dish, respectively. W is the initial dry weight of the sample.

2.3.3 Mechanical properties of the samples

The flexural strength of starch-based foam composites was investigated using a Universal Testing Machine, UTM, Zwick/Roell Z020, equipped with a 1 kN load cell according to the ASTM D790 standard. The test was performed in a 3-point bending mode with a span length of 100 mm and a crosshead speed of 2.67 mm/min. Five replicates of each type of sample with dimensions of 130 mm by 30 mm were tested for bending characteristics.

2.3.4 Soil burial degradation of the samples

The degradation of starch-based foam composites was investigated by burial in soil. The specimen size was 40 mm × 20 mm. The samples were buried at 10 cm depth in a sand and soil mixture with a ratio of 1:1 at 30 °C and a water content of 35 ± 5%. After 7 and 30 days, the samples were collected and washed with distilled water before drying in the oven at a temperature of 105 °C for 24 h. The weight loss of the samples was calculated using Eq. (3).

$$\% \text{ Weight loss} = \frac{W_i - W_f}{W_i} \times 100 \quad (3)$$

where W_i is the initial dry weight of the sample and W_f is the weight of the sample after the soil burial.

3. Results and discussion

3.1 Morphology of starch-based foam composites

Figure 1 depicts the visual aspect of foam composites based on cassava starch containing different contents of CRF and 10 wt% SCG. Surface irregularities and porosity were more pronounced in the samples with higher CRF contents. The lower starch concentration in foam composites caused a lack of adhesion between cassava starch and CRF, which hindered starch gelatinization and led to discontinuities in network formation on the foam surface [23]. Color differences between regions were also observed in the samples with higher CRF content. This might be because the dough had a higher viscosity after the pre-gelatinization process, which had an effect on the rheological properties of the composite batter and decreased its expansion capacity [24]. The cross-section of foam composites with varying CRF contents is illustrated in Figure 2. All samples have a sandwich-like structure. The region near the surface was denser with small cells, while the inner area contained larger cells with thin walls. This could be mainly because the water acted as a blowing agent during the mold compression process and became vapor bubbles that migrated to the outside of the mold when the temperature was higher than the boiling point, leading to the expansion of the polymer matrix [25]. In the outer area, the starch was close to the mold, so it gelatinized and dried rapidly, hindering an expansion of the polymer matrix [23]. In the interior of the foam, evaporation of a significant volume of water and leakage through the mold could occur, forming larger cells and a more open structure [26]. The SEM micrographs of the cross-section of all samples are illustrated in Figure 3. Compared to the structure of foam without CRF, a smaller number and size of voids inside the foam as well as a denser and thicker outer layer, were observed with increasing amounts of CRF. Such an observation could be due to a lower starch concentration and an increase in batter viscosity, resulting in lower expandability of the samples. This is consistent with a previous study by Phiriyawirut et al. [27], who found that the viscosity of foam composites made from tapioca starch mixed with octenyl succinate starch increased with increasing the proportion of chitin, leading to higher surface tension and resisting the expansion of water vapor in the foam.

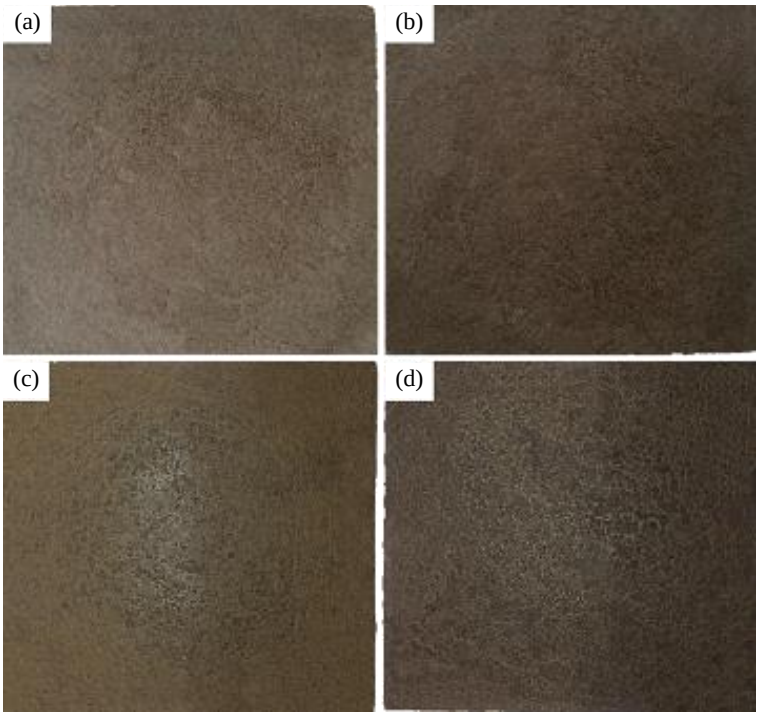

Figure 1 Visual aspect of starch-based foam composites with 10 wt% SCG and varying CRF amounts: (a) 0 wt%, (b) 5 wt%, (c) 15 wt%, and (d) 25 wt%

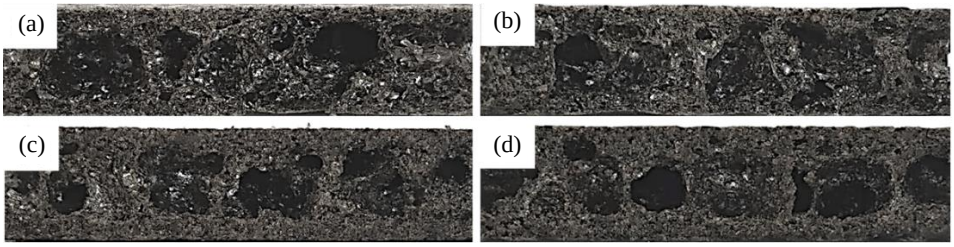

Figure 2 The cross-section of starch-based foam composites with 10 wt% SCG and varying CRF amounts: (a) 0 wt%, (b) 5 wt%, (c) 15 wt%, and (d) 25 wt%

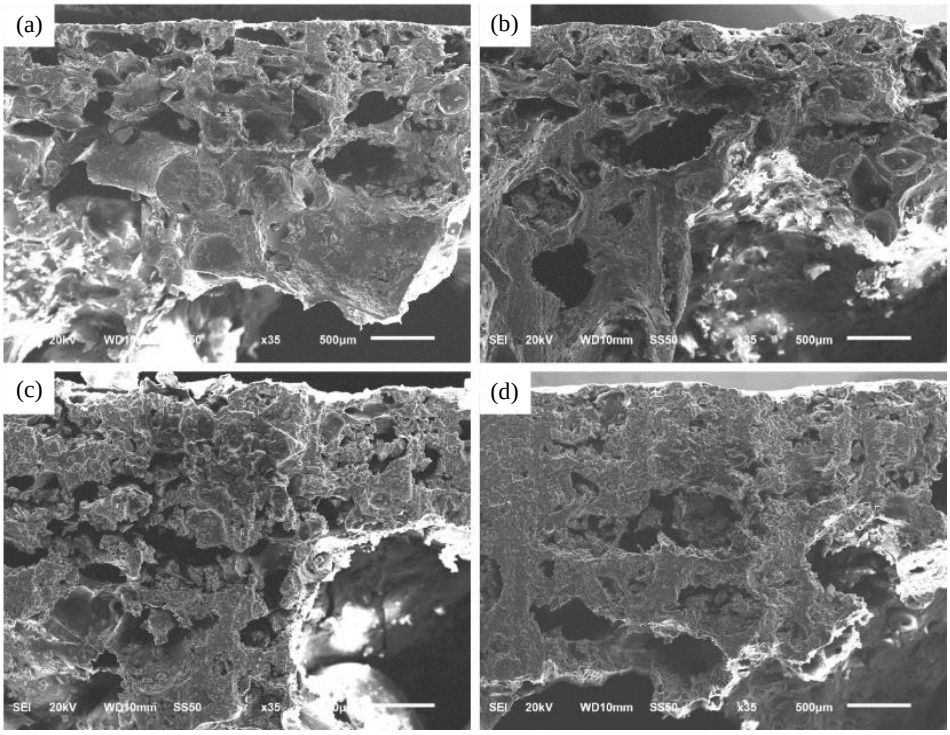

Figure 3 SEM images of starch-based foam composite samples with 10 wt% SCG and varying CRF amounts: (a) 0 wt%, (b) 5 wt%, (c) 15 wt%, and (d) 25 wt%

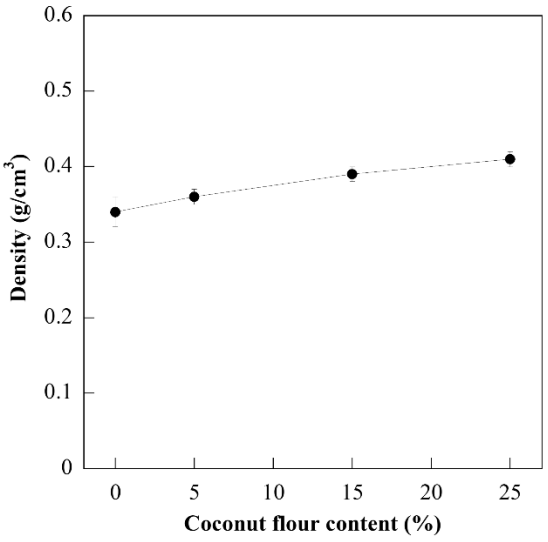

Figure 4 Density of starch-based foam composites with 10 wt% SCG and varying CRF amounts.

3.2 Density of starch-based foam composite samples

Density is an important property with an impact on the starch foam applications; generally, low density is preferred [28]. Figure 4 displays the density of foam composites containing 10 wt% SCG and various amounts of CRF. The density of foam composite samples

slightly increased from 0.34 to 0.41 g/cm^3 with increasing amounts of CRF at 0–25 wt%, as expected considering the morphological characteristics of the samples. The increased density of CRF-containing samples could be attributed to their lower starch content and higher viscosity, which hindered the expansion of water vapor bubbles during the foaming process, resulting in less expandable starch. It appears that the samples had smaller cells and higher densities. This result agrees with Machado et al. [29], who reported that the foam composites made from tapioca flour mixed with more broken rice had a lower expansion ability due to the lower starch content in their formulation, resulting in a higher density. The density of foam composites containing broken rice at 10–30 wt% was in the range of 0.3–0.53 g/cm^3. Chaireh et al. [30] found that increasing the amount of water hyacinth powder with a 250-µm particle size at 0–10 wt% increased the density of the foam composites from 0.32 to 0.43 g/cm^3. The expansion of steam bubbles during the foaming process was inhibited by the increased viscosity of the starch batter as the water hyacinth powder concentration increased, resulting in a higher density. Low-density foams are typically preferred, so CRF concentrations of less than 15 wt% should be used in foam composite formulations.

3.3 Water resistance of starch-based foam composites

The water-resistant behavior of starch-based foam samples is an important variable that greatly influences their functional properties and their application as food packaging and containers [31]. It is a well-known fact that starch-based materials are highly hydrophilic, absorbing a large quantity of water and weakening the hydrogen bonds of starch, decreasing their functional properties [32]. Figure 5 shows the water adsorption and solubility of foam composites made from cassava starch with varying CRF amounts and containing 10 wt% SCG in terms of WAI and WSI, respectively. It was found that the WAI and WSI of samples decreased slightly, from 8.95 to 5.10 and 5.05 to 3.16, as the CRF amount increased, respectively. This is primarily because the addition of CRF affected the higher viscosity of the composite batter, causing water molecules to be less expandable during the foaming process. This led to the foam composite having a denser outer wall and smaller porous cells, as shown by the cross-sectional morphology of samples observed with a scanning electron microscope. Therefore, it was rather difficult for water to enter the foam, leading to reduced WAI. As a result of fewer water molecules penetrating samples, there was less damage to the starch molecules, which could cause the decline in WSI. According to Kaisangsri et al. [33], the reduction in WAI and WSI could be attributed to the addition of kraft fiber into the starch matrix, which resulted in a dense outer skin that decreased the penetration of water molecules. Similar behavior was observed by Janaum et al. [10] in bioactive foam composites containing SCG, ZnO, and an oregano essential oil/palm oil mixture. They reported that starch foams with fillers had lower WAI and WSI due to thicker and denser outer skins with closed cells and tortuous paths for water molecules in the foam structure. CRF is a fiber-rich residue composed of hemicellulose, cellulose, and lignin, with a high concentration of insoluble fiber and trace amounts of soluble fiber [34]. It also contains significant amounts of fat and protein that cannot be removed completely from the coconut kernel [21]. Increasing the amount of CRF in the samples led to higher fat, insoluble fiber, and protein concentrations, which enhanced the water sensitivity of foam composites. This might be another explanation for the improvement in the water resistance of foam composites. This agrees with Machado et al. [35], who claimed that higher lipid, fiber, and protein levels in starch-based foams with more sesame cake residue led to greater water resistance.

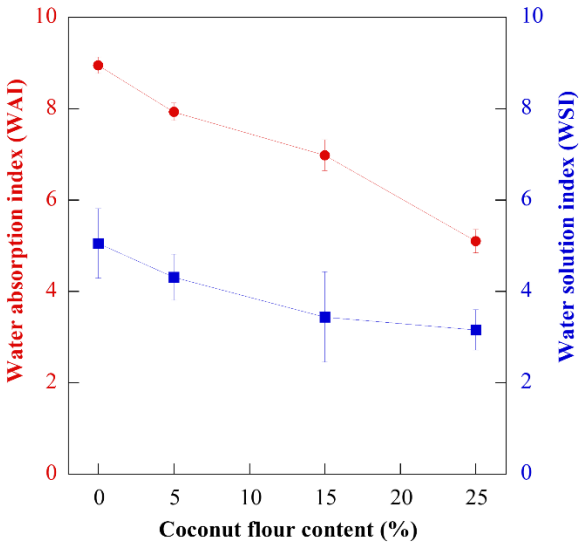

Figure 5 Water resistance of starch-based foam composites with varying levels of CRF content

3.4 Flexural strength of starch-based foam composite samples

The flexural strengths of starch-based foam composites with 0–25 wt% of CRF are shown in Figure 6. The flexural strength values were in the range of 0.53–2.53 MPa. The flexural strength gradually increased as the amount of CRF increased. This result could be attributed to the formation of a denser and thicker outer layer of starch-based foam composites containing higher levels of CRF. Machado et al. [36] mentioned that the denser outer layer with a closed-cell structure and thicker cell wall of the starch-based foam gave it higher mechanical strength because of its resistance to deformation. According to Spada et al. [8], although the filler encouraged discontinuities in the starch matrix, the samples with filler had a denser structure due to a higher concentration of solids per unit of volume, which could improve mechanical properties. The flexural strength of foam composites developed in this work was in the same range as those of other starch-based foam composites listed in Table 1 and was higher than that of expanded polystyrene foam (0.55 MPa) [5].

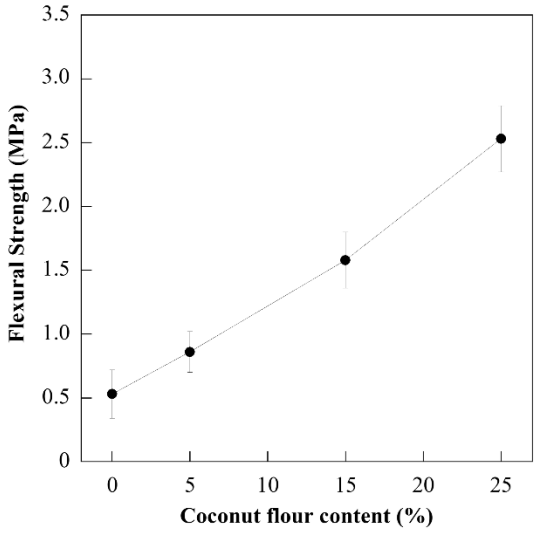

Figure 6 Flexural Strength of starch-based foam composites with 10 wt% SCG and varying CRF amounts.

Table 1 Flexural strength of various starch-based foam composites.

Starch foam	Additives	Flexural strength (MPa)
Cassava starch [11]	10 wt% spent coffee grounds, extracted spent coffee grounds, and 0-10 wt% oregano essential oil	0.30-1.22
Cassava starch [30]	0-5 wt% water hyacinth powder	2.95-3.42
Cassava starch and cassava inner bark [13]	7.2 wt% grape stalks with and without treatment	1.2-2.5
Potato starch [5]	11.2 wt% glycerol, 0.45 wt% maleic anhydride, 9 wt% PVA, 0.45 wt% nanoclay, 30 wt% PLA, 2 wt% azodicarbonamide and 2 wt% nisin	1.22-1.40
Potato starch [37]	20-80 wt% of brewer's spent grains	1.51-2.62

3.5 Biodegradability of starch-based foam composite samples

Figure 7 shows the weight loss of starch-based foam composites after a soil burial test for 7 and 30 days. It was observed that the weight loss of the samples increased as the percentage of CRF increased, ranging between 15.26–41.46% and 74.88–100% during periods of 7 and 30 days, respectively. The sample containing 25 wt% CRF was completely biodegradable in soil after 30 days. It was suggested that the addition of CRF to the polymer matrix improved the biodegradation of foam composites in soil in comparison to foam without CRF. This might be related to the high susceptibility to microbial spoilage of CRF in soil. Jongyingcharoen et al. [15] mentioned that microbial, yeast, and mold growths were encouraged by the presence of a high content of water in coconut residue, promoting a great biodegradation process. Nansu et al. [38] also reported a similar result in composites made from cassava starch and polyvinyl alcohol in a 70/30 dry weight ratio, with 0.1 wt% boric acid and varying amounts of coconut residue fiber. After 30 days, the biodegradation of the samples increased from 24.39% to 60.44% as coconut residue fiber content increased by 2–8 wt%. This is due to the fact that coconut residue fiber is easily degraded by bacterial and fungal enzymes due to its high oil and moisture content.

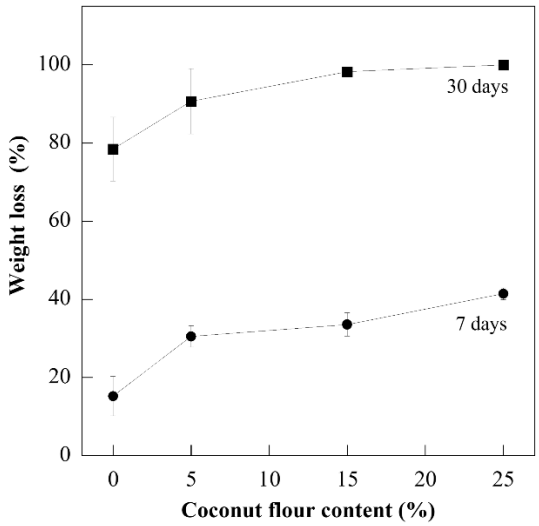

Figure 7 Weight loss of starch-based foam composites after a soil burial test.

4. Conclusions

Based on the findings of this work, it has been demonstrated that CRF made from coconut residue added to a polymer matrix could have the potential to be considered in the manufacturing of foam composites to be used for biodegradable packaging. By adding CRF at levels ranging from 0 to 25 wt% and 10 wt% SCG, starch-based foam composites were successfully formed using compression molding. As the amounts of CRF increased, cell expansion was inhibited during the foaming process, resulting in a denser and thicker outer layer and fewer and smaller voids within the foams. Samples with higher percentages of CRF showed higher values of density and flexural strength. The water-resistant properties of foam composites were also slightly improved by adding more CRF due to the tortuous water pathway that is likely present in denser foam as well as the high fat and insoluble fiber content of CRF. After soil burial tests, the weight loss of the samples increased as CRF contents increased due to the high susceptibility of CRF to microbial spoilage. The starch-based foam composites with 25 wt% CRF and 10 wt% SCG were completely buried in soil after 30 days. As a result, CRF can be viewed as a promising, cost-effective, and environmentally friendly alternative reinforcing material for foam composite packaging, providing financial benefits to regional coconut producers and manufacturers. Although further study is necessary to enhance the properties of foam composites, particularly their water resistance, the flexural strength values of the foam composites developed in this work were higher than those of conventional EPS foam. These biodegradable foam composites could thus be used in place of non-renewable and non-biodegradable foam in single-use packaging for dry and low-moisture food products.

5. Acknowledgements

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