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The Effects of Extrusion Temperature and Sodium Bicarbonate Foaming Agent on Density and Tensile Properties of Kenaf Fibre-Reinforced Biocomposites

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Foaming composites reduce transportation costs and fuel usage by achieving weight reduction through the introduction of foaming gas. This study investigates the impact of extrusion temperature profiles and sodium bicarbonate ($NaHCO_3$) foaming agent loading (2 and 4%) on the density, morphological foam structure, and mechanical properties solid and foamed poly (lactic acid) (PLA) biocomposites reinforced with kenaf fibre. The biocomposites were prepared via melt-blending method. Two extrusion temperature profiles, set at 165-170-175-160 ºC (higher temperatures, HT) and 155-160-165- 150 ºC (lower temperatures, LT). Results indicate that, with an increase in $NaHCO_3$ loadings, a gradual decrease in density was observed, with LT extrusion showing a higher density reduction (up to 8%) compared to HT. Field emission scanning electron microscope observations revealed a more consistent foam cell size structure for LT-extruded samples. However, the tensile properties of the foamed biocomposites deteriorated, with approximately 23% decrease in tensile strength and 26-29% in elongation at break. In conclusion, a lower extrusion processing temperature (LT) was found to be optimal for producing uniform porous biocomposite foams with lower density and higher mechanical strength.

Keywords: Natural fibre polymer composites; sodium bicarbonate; cell morphology; mechanical properties

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Poly(lactic acid) (PLA) is an aliphatic polyester thermosplastic which can be obtained from renewable sources such as sugar, corn, wheat and sugarcane. PLA is environmentally friendly and fully biodegradable that can be decomposed into water and carbon dioxide by microbes [1]. By owing high strength, good biocompatibility and ease of processing, PLA is a biopolymer that has potential to be candidate for petroleum based polymer [2, 3]. Recently, natural fibres including hemp, jute and kenaf have received lots of attention to replace synthetic fibre in the production of biobased materials [4]. The incorporation of low cost natural fibres such as kenaf into PLA matrix was found to produce a biocomposite with better texture and improved properties [5]. Several researches related to PLA/kenaf fibre biocomposite showed the increase in mechanical strength and modulus with different filler loading. Tharaziet et al. 2017 [6] reported that the optimum tensile strength and modulus were achieved at 40 % long kenaf fibre, showing increment of 248 % and 650 %, respectively. In another study, the highest flexural and tensile properties were obtained at volume fraction of 70 % [7].

Nevertheless, in most cases, the inclusion of natural fibre will contribute to the increased final weight of biocomposite that impact on the fuel consumption and transportation cost [8]. To solve this issue, foaming is a viable method to fabricate lightweight composite material [9]. In the foaming technology, chemical foaming agent has been used in many researches where it involves the release of gas via chemical reaction under suitable conditions like temperature and pressure that allows its thermal decomposition [10]. Sodium bicarbonate ($NaHCO_3$) is a common endothermic foaming agent where it starts to decompose in the vicinity of 160 ºC and releases carbon dioxide gas during its decomposition [11]. Garbacz et al. 2020 [12] reported that the foaming of PLA with endothermic chemical foaming agent, i.e. Hydrocerol ITP810 (which contains a mixture of active substances including $NaHCO_3$ and citric acid [13]) was shown to significantly reduce the density of composite up to 24 % by using 3 % foaming agent, however, it also deteriorated the tensile properties by approximately 23 % [12]. Another similar finding was reported by Hussein et al. 2018 [14] whom worked on epoxy foam with $NaHCO_3$ loading up to 25 phr.

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By considering the thermal properties of $NaHCO_3$ (low decomposition temperature range), Luo et al. 2021 [15] have investigated the modified $NaHCO_3$ in the capsule form in order to improve the foaming quality of foamed polypropylene, which was fully proven in the study.

Numerous studies have been reported the foaming on pure PLA [11, 16] and conventional PLA biocomposites reinforced with a variety of natural fibre such as jute [17-19], wood flour [20-22], sugarcane bagasse [23] and etc. To our knowledge, the foaming of natural fibre reinforced PLA biocomposites with the aids of chemical foaming agent, specifically endothermic foaming agent, is not widely explored yet. This study aims to examine the effect of extrusion temperature profile and foaming agent content of $NaHCO_3$ on the density, foam morphology and tensile properties of PLA/kenaf fibre biocomposites.

EXPERIMENTAL

Chemicals and Materials

In this study, thermoplastic resin used was poly (lactic acid) (PLA) with the grade of Naturework IngeoTM Biopolymer 3251D was obtained from Unic Technology Ltd, China. It has a density of 1.24 g/cm^3, melt flow index (MFI) of 80 g/10 min (190˚C/2.16 kg) and melting temperature of 188-210 ºC. Kenaf fibre with the size of 100-150 µm was supplied from Kenaf and Tobacco State (LKTN). It was used as filler to reinforce PLA matrix. An endothermic foaming agent of sodium bicarbonate ($NaHCO_3$) and its modifier of citric acid ($C_6H_8O_7$) were purchased by Sigma Aldrich.

Preparation of PLA/Kenaf Fibre Biocomposite Foam

Prior to mixing, both PLA and kenaf fibre were oven-dried at 80 ºC overnight to remove moisture. To prepare extrudate (mixed compound), twin-screw co-rotating extruder (*Thermo Prism TSE 16PC*) was used with different extrusion temperature profile setup (from feeding to die zone barrel). Two temperature profiles were used for extrusion, which are higher temperature of 165-170-175-160 ºC and lower temperature of 155-160-165-150 ºC, namely HT and LT, respectively. The rotating speed was fixed at 40 rpm. The weight ratio of PLA/KF was fixed at 70:30, whereas the content of $NaHCO_3$ was varied at 2 and 4 %. The $NaHCO_3$: $C_6H_8O_7$ ratio was mixed at 1:1. The solid PLA/kenaf fibre biocomposites without presence of foaming agent, namely HT0 and LT0, were served as control sample, for each HT and LT systems. The details of investigation in this study are described in Table 1. The extrudate pellets were continued with hot and cold pressing using compression molding machine (LP50, *Labtech Engineering Company Ltd*) to form specimen panels. The compression molding was undergone at 190 ºC with a pressure of 6.9 MPa for 15 min. The process involved 3 min of pre-heating, 4 min of ventilation, full hot pressing of 5 min and 3 min of cold pressing. The pressed specimens were kept about 40 hours for stabilization before proceeding with characterization.

Characterization Methods

The density of unfoamed (HT0 and LT0) and foamed (HT2, HT4, LT2 and LT4) biocomposites was measured using gravimetric method (mass over volume calculation [24]). Five replicates for each sample were used to determine the average density value. For foam structure observation, the surface morphology of the tensile-fractured samples was analyzed by field emission scanning electron microscope (FESEM) (*model LEO 1450 VP*). To avoid electrostatic discharging, the sample surfaces were sputter-coated with a thin layer of gold prior to SEM observation.

Table 1. Parameters of extrusion temperature and $NaHCO_3$ loading for investigation.

Samples	Extrusion temperature profile	Composition		
		PLA (wt%)	KF (wt%)	$NaHCO_3$ (%)
HT0 (unfoamed)	HT	70	30	0
HT2	HT	70	30	2
HT4	HT	70	30	4
LT0 (unfoamed)	LT	70	30	0
LT2	LT	70	30	2
LT4	LT	70	30	4

Note: HT denotes high extrusion temperature profile of 165-170-175-160ºC; LT denotes low extrusion temperature profile of 155-160-165-150ºC.

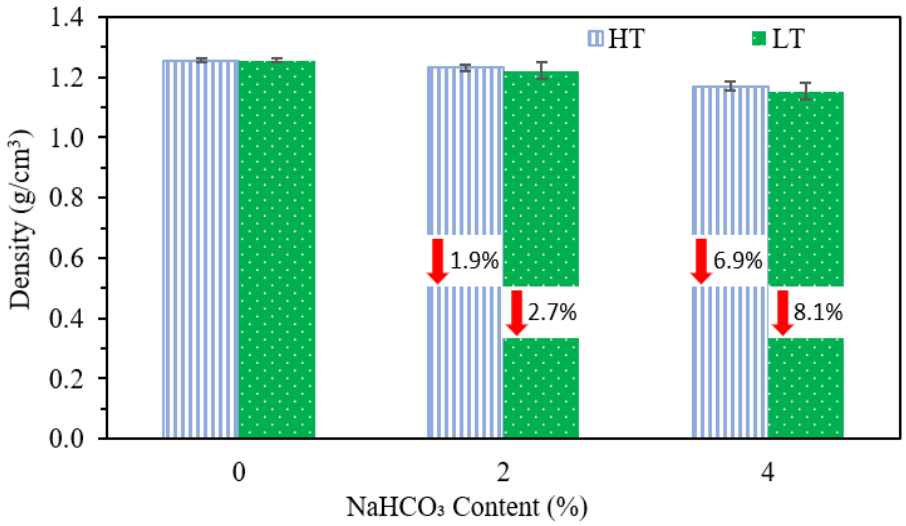

Figure 1. Effect of extrusion temperature profile and $NaHCO_3$ content on the density of PLA/kenaf fibre biocomposites. Note: HT means (165-170-175-160) ºC and LT means (155-160-165-150)ºC. Red arrow indicates the reduction percentage compared to solid biocomposite without $NaHCO_3$ (HT0 and LT0).

In the aspect of mechanical property examination, tensile test was conducted on the solid and foamed biocomposites using tensile machine (*model Testometric M500-50CT*) based on the standard of ASTM D638-03. A load cell of 10kN and a crosshead speed of 5 mm/min was used during tensile testing.

RESULTS AND DISCUSSION

Density Measurement

Figure 1 shows the influence of two extrusion temperature profiles and $NaHCO_3$ contents on the density of PLA/kenaf fibre biocomposites. Regardless of extrusion temperature profile, as $NaHCO_3$ contents increased, the density of biocomposite foam reduced. The unfoamed PLA/kenaf fibre biocomposite recorded a density of 1.26 g/cm^3. Inclusion of 4 % $NaHCO_3$ into biocomposite led to a reduction by 6.9 % and 8.1 % for HT4 and LT4, respectively. This could be correlated to the increased CO_2 gas release with the foaming agent content which caused to improve the microcellular structure of the samples [25], as observed in Figure 2. By comparison of extrusion temperature profile, when the low extrusion temperature (LT) was used, the foamed biocomposite exhibited lower density compared to high temperature profile (HT). This is attributed to the HT is much higher than the decomposition temperature of the modified $NaHCO_3$ with stearic acid, citric acid and etc., which is located at approximately ~130-166 ºC [15]. In this sense, the release of gas easily occurs, and the produced gases tend to escape from the extrudate surface before the completion of melting and compression process. The early decomposition of $NaHCO_3$ during extrusion results in the loss of gases for effective foaming process [10, 26], therefore biocomposite foams prepared by HT had higher density. The more effective foaming process with LT regardless of $NaHCO_3$ loading is supported by the FESEM morphological observation (more obviously observed with higher $NaHCO_3$ loading for HT4 and LT4 in Figure 2 (e) and (f)) with more consistent size and regular shape of foam cells.

Foam Morphology

The morphological microstructures of PLA/kenaf fibre biocomposite samples with various $NaHCO_3$ contents prepared by different extrusion temperatures are demonstrated in Figure 2. In the absence of foaming agent, Figure 2(a) and (b) show non-foaming morphological structure with uniform distribution of kenaf fibre within PLA matrix, for both biocomposite systems (HT0 and LT0). When compared to HT0, the biocomposite prepared by LT0 had a more compact structure with less interfacial gap between matrix and fibre. This supported by the previous study [27] whom stated that lower extrusion temperature would encourage continuous mixing and better melt flow in the extruder. This can be explained by the plasticizing effects of the foaming gas where the foamed bicomposites tend to operate under lower temperature when compared to the solid counterparts [28]. Additionally, it can be observed that the tension fracture of the LT biocomposite showed a certain strain effect, whereas the HT biocomposite showed a smoother broken surface. These phenomena suggest the greater tensile properties as in agreement with the results in Figure 3.

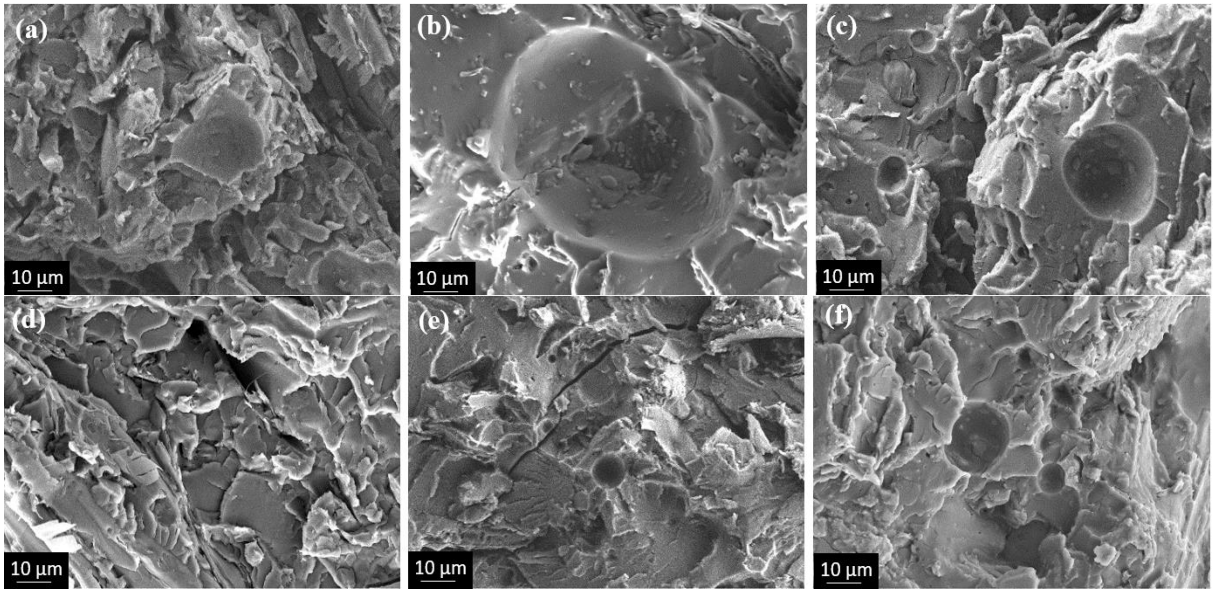

Figure 2. FESEM micrograph (magnification of 1000 x) of PLA/kenaf fibre biocomposites foamed with 0, 2 and 4 % $NaHCO_3$ (a, b and c) via HT condition and (d, e and f) via LT condition. Note: HT means (165-170-175-160) ºC and LT means (155-160-165-150) ºC.

For the case of biocomposite foams, LT system showed foam morphology that having several cells with average smaller cell size (LT2 and LT4 in Figure 2 (e) and (f)) in comparison to the larger cell size in HT system (HT2 and HT4 in Figure 2 (b) and (c)). This could be explained by Standau et al. 2019 [3] that the formation of stable foam structure is dependent upon processing temperature. In this case, the extrusion temperature which is nearer to the decomposition temperature of foaming agent (HT) would lead to premature release of gas and thus adversely affect the cell growth. Besides, the increased processing temperature may decrease the viscosity of polymer matrix. As a consequence, the polymer matrix cannot withstand the biaxial strain that occurs during cell growth where causes the collapse of the foam cells in the composite materials at high temperature [29]. By looking at the effect of $NaHCO_3$ content, it showed that the number of cells increased with the foaming agent loading as more CO_2 gas was produced.

Tensile Properties

Figure 3 displays the effect of extrusion temperature profile and $NaHCO_3$ content on the tensile properties of PLA/kenaf fibre biocomposites. Without presence of $NaHCO_3$, the PLA/kenaf fibre biocomposite (HT0) extruded under HT exhibited tensile strength and elongation at break at 14.7 MPa and 1.8 %, respectively. Meanwhile, for extrusion via LT condition, each tensile strength and elongation at break of the corresponding biocomposite (LT0) were recorded at 16.4 MPa and 2.1 %. The trend of higher values for LT was similarly reported in the foamed biocomposite system. This result can be correlated to the FESEM micrograph (Figure 2) where the foam produced via later condition was consistently smaller in size. Presence of big cell size would act as defect or crack point which consequently result in the premature failure and thus deterioration in tensile properties, as in agreement with Kord, 2013 [30].

As expected, the tensile strength (Figure 3 (a)) and elongation at break (Figure 3(b)) of the biocomposites were remarkably reduced by foaming, regardless of extrusion temperature profile. This trend is probably because of the increased bubble nucleation and cell coalescence that was induced by increasing the foaming agent contents [31]. Although a higher foaming agent content can result in increased foam cell formation leading to a lower composite density, it indeed affects the pressure transfer between the matrix and the composite filler [2]. This accounts for the gradual decrement of tensile strength as shown in Figurer 3(a). The presence of larger cells observed in Figure 2(b) and (c) with HT acted as defects, leading to premature failure and consequently resulting in lower elongation at break (Figure 3(b)). The lowest tensile strength and elongation at break were ~12 MPa and ~1.3 % for HT4 sample.

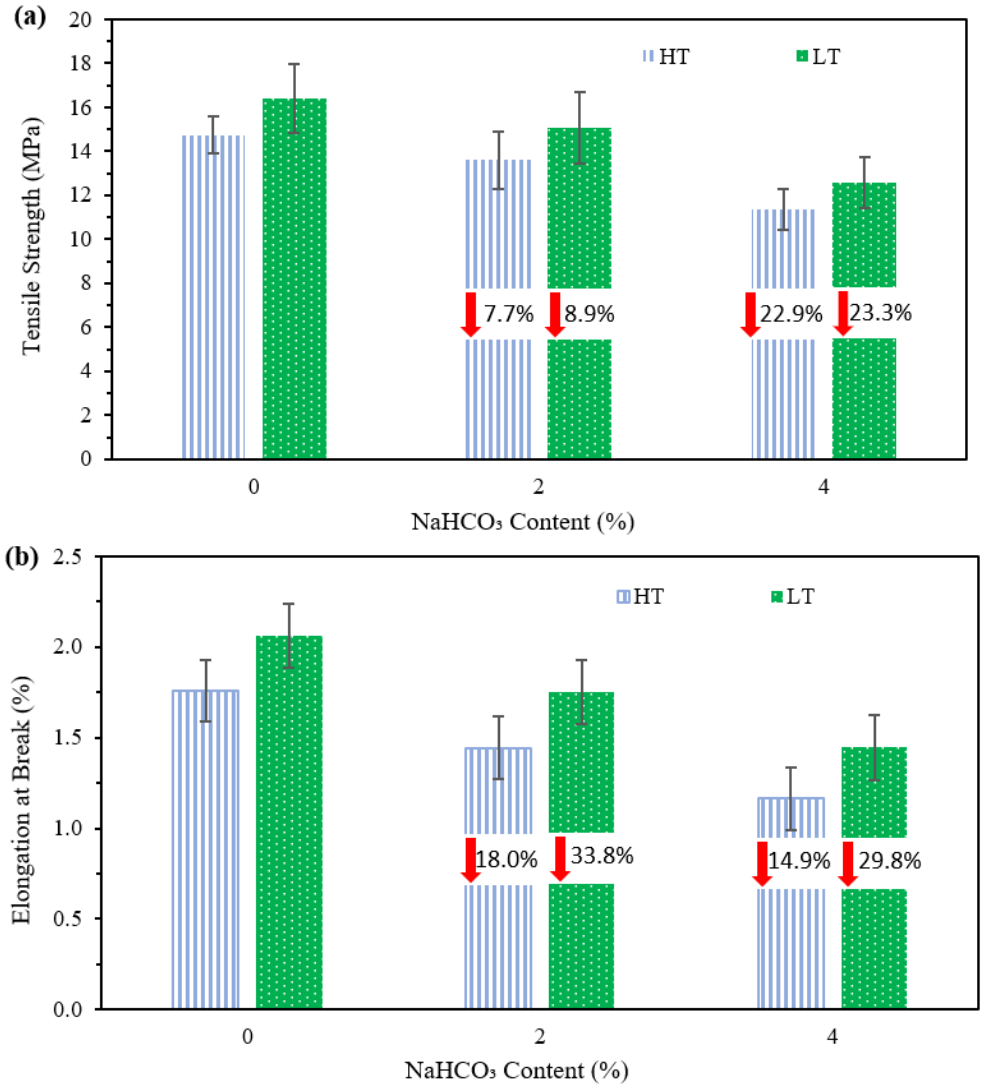

Figure 3. Effect of extrusion temperature profile and $NaHCO_3$ content on the (a) tensile strength and (b) elongation at break of PLA/kenaf fibre biocomposites. Note: HT means (165-170-175-160) ºC and LT means (155-160-165-150) ºC.

CONCLUSION

Kenaf fibre reinforced PLA biocomposites foams were prepared through extrusion and compression molding. The study investigated the impact of extrusion temperature profile and $NaHCO_3$ contents on the density, foam morphology and tensile properties. The results demonstrated substantial reductions in biocomposites density with increasing $NaHCO_3$ content up to 4 %, particularly noteworthy when employing a lower extrusion temperature profile (155-160-165-150 ºC) as opposed to a higher profile (165-170-175-160 ºC). The research findings highlight the potential of this biocomposite foam for practical applications, such as environmentally friendly packaging materials or light-weight components for the automotive industry. Further exploration of additional foaming parameters, different foaming agent and additive like stabilizer could further enhance the outstanding of optimizing biocomposite properties in future investigations.

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