Effect of Removing Sucrose and Moisture in Bagasse Fibers on Improvement of Limit of Processing Temperature in Bagasse-Polypropylene Composites

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This study focused on the increase of processing temperature of sugarcane bagasse fiber and polypropylene composites by removing sucrose and moisture in bagasse fibers. The relationship between the number of washing times and remaining sucrose in bagasse fiber were measured by high-performance liquid chromatography (HPLC). The analysis showed that original bagasse fibers, which had been obtained from a sugar cane mill, contained 4.0 wt% sucrose. To clarify the effect of the remaining sucrose and moisture on the limit of processing temperature in bagasse composites, the sucrose removed bagasse (40 wt%) or the original bagasse (40 wt%) was mixed with polypropylene (60 wt%), respectively. Then, the composite specimens were prepared with hot-press forming at various temperature. The observations of the composites appearances and their flexural tests were carried out. The results showed that the limit of processing temperature in the removing sucrose and moisture of bagasse composites was dramatically improved. The flexural properties in the sucrose and moisture removed bagasse composites did not decrease until 260 °C, while that in the original bagasse composites decreased at 240 °C.

Keywords: Bagasse fiber; Polypropylene; Natural fiber composite; Processing temperature

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INTRODUCTION

Natural fibers have been widely studied as a potential substitute for synthetic fibers due to their low cost, high Yong's modulus, recyclability, *etc.* (Goda and Cao 2007; Kalia *et al*. 2009; Ahmad *et al*. 2014; Sanjay *et al*. 2018). The addition of natural fibers in synthetic polymer matrices can be an interesting method to reduce the petrochemical resources. Some key features of naturally produced cellulosic fibers are their abundance in nature, biodegradability, light weight, low abrasion resistance, low thermal expansion, and high toughness. Combining natural fibers in a synthetic polymer as biocomposites has promising potential for obtaining optimal utility or minimizing the use of synthetics (Shubhra *et al*. 2011).

There are important improvements needed for biocomposites, *i.e*., the lower heat resistance, larger amount of fillers, low impact strength, poor compatibility between fiber and matrix. For instance, Ashori and Nourbakhsh (2008) investigated the effect of polybutadiene isocyanate treatment on bagasse-PP composites. The composites treated with this isocyanate agent showed superior tensile and impact properties compared to those without treatment. Anggono *et al*. (2017) comparatively studied calcium hydroxide and sodium hydroxide on sugarcane bagasse-PP composites to improve the interface of fibers

and matrix.

The deterioration in mechanical properties was observed by Bourmaud *et al.* (2016) experimentally after fibers were exposed to temperatures above 170 °C. This deterioration was due to the beginning of thermal degradation by chemical interactions among water molecules, sucrose, pectin, hemicellulose, and non-crystalline cellulose. This is the reason why most biocomposites were fabricated below 200 °C to avoid the deterioration. However, sucrose and water, which may affect thermal degradation of natural fibers, were not removed completely in the above-mentioned papers in the preparation of natural fiber composites. As described in the results section, this was attributed to the fact that relatively low amounts of moisture absorption accelerated thermal degradation in bagasse composites and decreased the limit of processing temperature.

The limit of processing temperature 200 °C in natural fibers has been a constraint in the selection of polymer matrices, especially for thermoelastic matrices (Joseph *et al.*1999; Shibata *et al.* 2005; Ashori and Nourbakhsh 2008; Lee *et al.* 2009). For example, the range of injection molding processing temperature is 220-240 °C for ABS, 250-280 °C for PET, and 240-280 °C for PA6. Even for PP, which is often used as the matrix with grass fibers, its range is 200-230 °C (Fambri e*t al.* 2013). Azwa and Yousif (2013) investigated the thermal degradation in kenaf-epoxy composites by TG and DTA analysis for 1 to 3 h heating. These results are suggestive discussion for the thermal degradation process of natural fiber. However, the experiments were not conducted with a practical length of processing time such as 1 to 20 min, and the report did not connect the relationship between thermal degradation temperature and its mechanical properties. Paul *et al.* (2008) examined the effect of chemical treatment such as alkali, on the thermal conductivity and thermal diffusivity in banana fiber composites. The thermal conductivity declined with fiber loading after treating banana fibers with different NaOH concentration.

Though the increase of the limit of processing temperature in natural fibers in the practical temperature range and duration of processing is quite important to extend the range of natural fibers used as biocomposites, the related research has been quite few. The objective of the present work is to increase processing temperature in natural fiber composites up to 300 °C in order to be able to use it in ABS, Nylon, and polycarbonate as matrixes with natural fibers. However, this temperature of 300 °C is likely too high for natural fibers at the present time. Hence, as a first step, in this study our objective is to increase the limit of processing temperature in bagasse PP composites with a simple method such as desugaring and dehydration. As a result of the removal of sucrose and moisture in bagasse fibers before fabrication, the thermal degradation in the composites in molding processing was shifted to higher temperatures by approximately 20 to 40 °C.

EXPERIMENTAL

Materials and Methods

Sugarcane bagasse, hereafter "bagasse", fibers were supplied by a local sugar cane mill, and this bagasse was sieved to the range 5 to 20 mm in order to remove long fibers, pith, and particles, as shown in Fig. 1. The bagasse was not chemically treated; it was crushed, squeezed, and washed by hot water in the sugarcane mill. The average fiber length and diameter of 500 samples were 10 mm and 0.39 mm, respectively. Polypropylene (PP, AZ864-N, Sumitomo Chemical Co., Ltd, Osaka Japan, M_w 162000, M_w/M_n 6.1) was used as a matrix. The MAPP (maleic acid grafted polypropylene), which included 0.5 wt%

maleic acid, was used at 5 wt% to improve the surface adhesion between polypropylene and bagasse fibers. The mechanical properties of these fibers and PP have been measured in previous reports (Shibata *et al.* 2006) as shown in Table 1.

Table 1. Mechanical Properties of Bagasse and Polypropylene

Sample	Young's Modulus (MPa)	Tensile Strength (MPa)	Specific Gravity (kg/m^3)	Diameter (mm)
Bagasse	4517	89	344	0.39
Polypropylene	1315	24	902	-

Two types of bagasse fibers were prepared. One type was washed at various temperatures by hot water and dried at 80 °C for 24 h, and the other was original fibers used as supplied. Composite pellets were prepared as follows. Bagasse fibers and the PP were mixed with a single extruder at 190 °C as shown in Fig. 2. The material composition was 40 wt% bagasse fiber and 60 wt% polypropylene. During compounding, the extruded strand was cut into 10 mm long and 5 mm diameter as composites pellets.

Fig. 1. Bagasse fibers

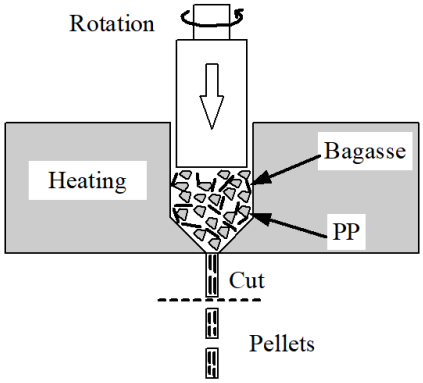

Fig. 2. Single extruder for composite pellets

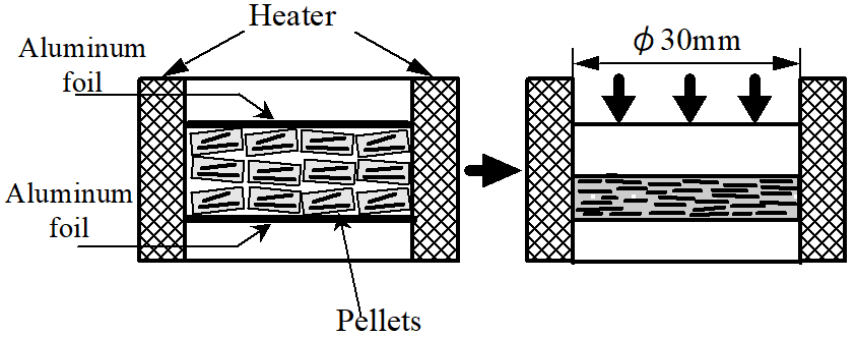

Fig. 3. Hot-press moulding for bagasse-PP composites specimen

Press forming

The disc-shaped specimen was fabricated by hot-press forming. Figure 3 shows the fabrication process. At first, the cylinder and piston were heated to the desired temperature. After that, the pellets were placed in the cylinder and pressed for 5 minutes, and the metal mold was cooled by water. The obtained composite specimens were circular shaped (30 mm in diameter, 1.5 to 2.0 mm in thickness). The hot-press molding was performed at various temperature of 200 °C, 220 °C, 240 °C, and 260 °C in order to observe the limit of processing temperature in bagasse composites at a pressure of 100 kgf/cm^2 for 5 min.

Flexural tests

The flexural test was carried out in accordance with ISO 178 at each condition with five specimens using a universal testing machine (Orientec Co. Ltd. STA-1225, Tokyo, Japan). The flexural strength and flexural modulus were determined by averaging the values of specimens generated from the three-point bending test. The test used a span length of 22 mm and a crosshead speed of 1 mm/min. The specimens used in the flexural test were 30 mm in length, 15 mm in width, and 1.5 to 2.0 mm in thickness. The flexural modulus was determined by the initial derivative line of the load-displacement curve. The derivative part was defined as being between 0 and 15% of the maximum load. The flexural strength was calculated using the maximum load.

HPLC analysis

The concentration of sucrose in the original bagasse was measured to determine the desugaring process condition. Bagasse 10 g and water 800 mL was put in a 1.0 L flask and kept at 80 °C with a heater for 1 h. After heating, this wash water was replaced with new water, and the bagasse was washed again. In each washing, the wash water was supplied for concentration analysis by high-performance liquid chromatography (HPLC) with a refractive index detector. The weight ratio of sucrose was calculated against the initial weight of bagasse fiber.

Dehydration of bagasse and bagasse's pellets

Dehydration treatment of bagasse was performed in order to decrease chemical interactions and increase the temperature of the thermal degradation. Bagasse PP pellets had been under vacuum at 1 torr for 24 h. The pellets were taken out of vacuum chamber and immediately hot-pressed at 260 °C. To clarify the dehydration effect, the dehydrated pellets and bagasse fibers were exposed to room atmosphere at 25 °C and a relative humidity of 70% for 0, 20, 60, and 180 sec, respectively. Also time-dependent moisture absorption of the bagasse-PP pellets and bagasse fiber itself were measured. The weight change of the pellets and the fibers was measured immediately after being taken out of vacuum oven under an atmosphere of 20 °C and 70% relative humidity.

RESULTS AND DISCUSSION

Figure 4 shows the relationship between the number of washing times and the concentration of sucrose by HPLC analysis.

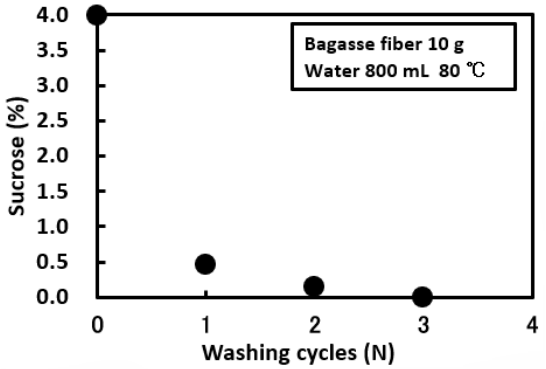

Fig. 4. Relationship betweem the number of washing times and sucrose

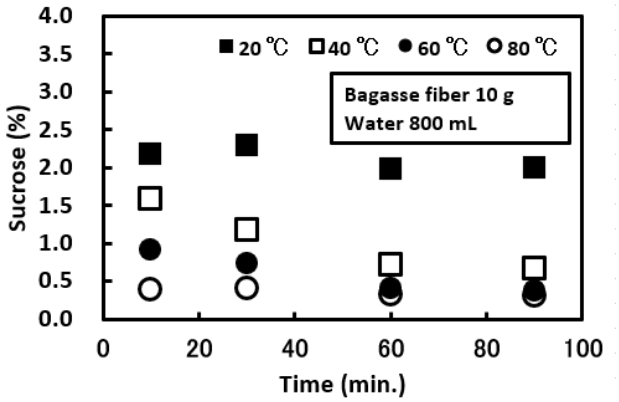

Fig. 5. Relationship between length of washing time and sucrose in each water temperature

It was found that the original bagasse contained 4.0 wt% sucrose, and three times washing was required to remove sucrose completely in the original bagasse fibers. Figure 5 shows the relationship between the length of washing time and sucrose in the original bagasse with different washing temperature. Judging from the figure, washing water more than 60 ºC was found to be efficient and desirable to remove sucrose completely in bagasse fibers.

Figure 6 shows the appearances of hot-press (200 °C) specimens of bagasse-polypropylene composites. Figures 6 (a) and (c) are with and without desugaring treatment of bagasse, respectively. All the bagasse in the composites were washed three times at 80 °C and dried for 24 h before mixing with polypropylene. As can be seen, the disc composite in Fig. 6 (a) with desugaring was not charred. This result indicates that sucrose included in bagasse interacts with one another or among water, pectin, hemicellulose, and non-crystalline cellulose as suggested by Bourmaud *et al.* (2016). Also, as described below, the deterioration in mechanical properties occurred in the charred black composites as shown in Fig. 6 (c). Thus, the removal of sucrose in bagasse was found to increase the limit of the processing temperature in the composites. Besides, Fig. 6 (a) also showed the surface of aluminum foil, which had been between the composites and the mold during hot-press, and was not stained with tar deposit compared to Fig. 6 (d). This tar deposit tended to stick to the mold surface; this can be an inconvenience for continuous processing method such as an injection molding.

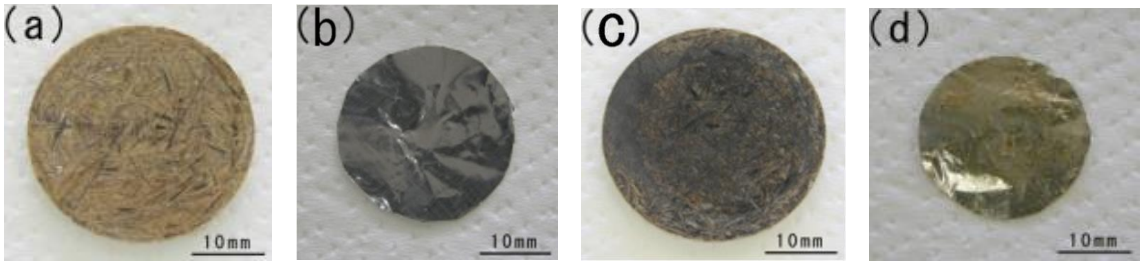

Fig. 6. Apearances of bagasse composites with (a) Desugared bagasse PP composite, (b) its attached foil, (c) original bagasse PP composite, and (d) its attached foil

Next, the effect of hot-pressed temperature on the thermal degradation in the composites was examined. Figures 7 (a)-(d) shows the appearances of bagasse composites by desugared bagasse at 200 °C, 220 °C, 240 °C, and 240 °C. Especially, item (d) was charred black and found to be brittle. The results indicated that the limit of processing

temperature in the composites could be around 260 °C. Though, even with the removal of sucrose, bagasse still contains water and other molecules with a hydroxyl group such hemicellulose, non-crystalline cellulose. Thus, these chemicals would react with one another and can result in the charred black as shown in Fig. 7 (d).

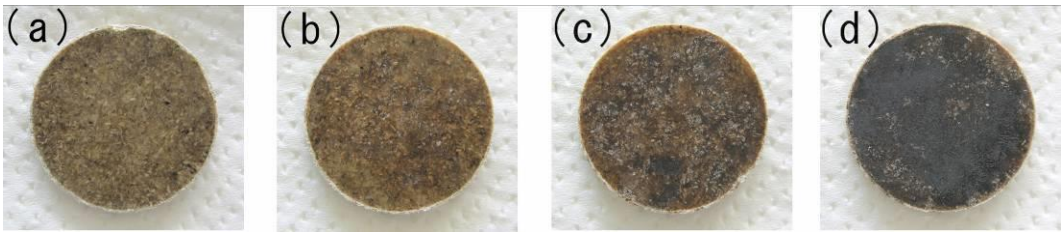

Fig. 7. Appearances of bagasse PP composites with desugared bagasse,
(a) 200 °C, (b) 220 °C, (c) 240 °C and (d) 260 °C

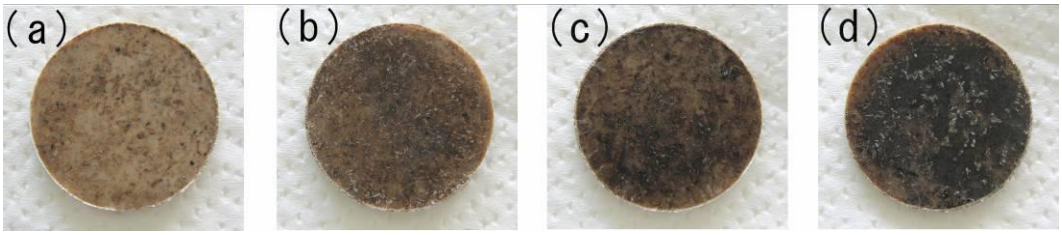

Fig. 8. Appearances of bagasse PP composites with desugared bagasse and dehydrated pellets (a) 0 sec, (b) 20 sec, (c) 60 sec and (d) 180 sec at 260 °C

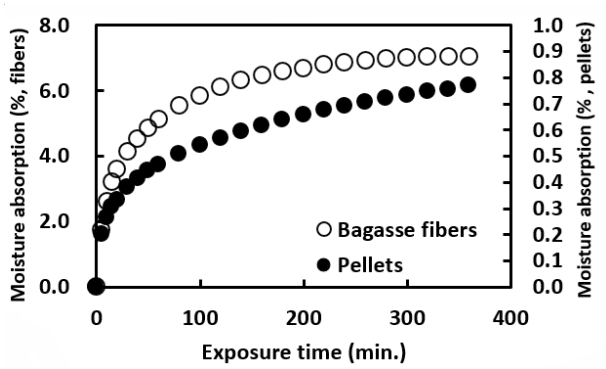

Fig. 9. Moisture absorption of fibers and composites pellets at different time exposure

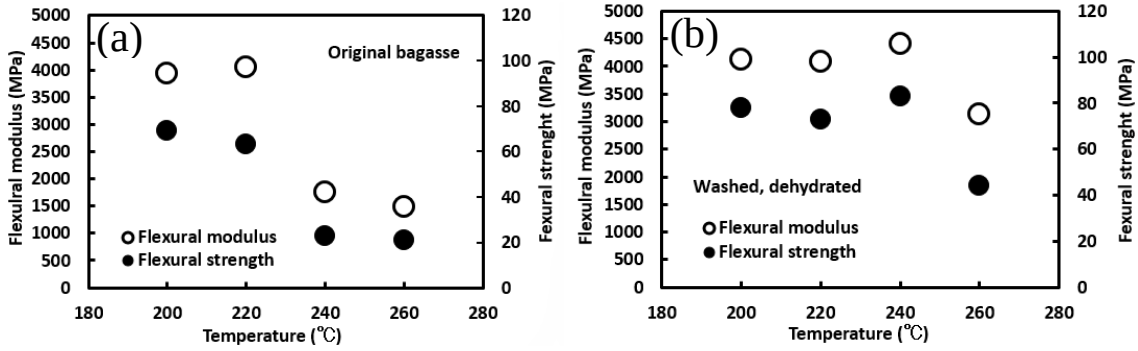

Fig. 10. Relationship between flexural properties and hot-pressed temperature of bagasse PP composites ((a) original bagasse; (b) washed and dehydrated)

Figures 8 (a)-(d) show the appearances of the composites with the desugared and dehydrated bagasse, as described in the experimental methods section. The appearance of the composites turned from light blown to charred black with exposure time increasing. Figure 9 shows the relationship between bagasse, 40 wt% bagasse-PP pellets and moisture absorption just after these materials taking out of vacuum chamber. Moisture absorption in percentage was measured using an electric balance. Moisture absorption of bagasse was progressing rapidly approximately for 10 to 30 sec. The absorption was saturated to 7 wt% for bagasse, 0.8 wt% for the pellets. Considering of bagasse percentage, the pellets should absorb 2.8 wt%; however, the moisture absorption could be considered to occur in the vicinity of the surface. Figure 10 (a) shows the relationship between the flexural modulus and strength in composites formed from the original bagasse, which is without washing. It was found that the both flexural properties decreased dramatically above 240 °C. The composite specimens above 240 °C were obviously charred black and brittle. On the other hand, Fig. 10 (b) shows the same relationship in the case of the composites with the above desugared and dehydrated treatment. The flexural properties in the composites did not decrease until 240 °C. Also, the appearance was still light brown at that 240 °C.

Hence, the desugaring and the dehydrate treatment was found to be effective for improvement of the limit of the processing temperature in bagasse composites. Especially, these treatments were practically helpful to avoid charred products in the injection molding using bagasse and polypropylene composites for automobile parts, in our experience. Also further research in this treatment is expected to widen the selection range of polymer matrix and processing conditions in biocomposites.

CONCLUSIONS

1. The effect of removing sucrose in bagasse fibers on the thermal degradation by heating was examined. It was found that the original bagasse fibers contained 4.0 wt% sucrose, and this sucrose could be removed by three-times washing with water that had been heated above 60 °C. The limit of processing temperature in the sucrose-removed bagasse PP composites was increased. The color in the original bagasse PP composites turned into charred black by heating at 200 °C, while that in the sucrose removed bagasse PP composites remained light brown. In addition to the desugaring treatment, the dehydrated bagasse PP composites showed improvement in the processing temperature also. The color in the composites remained light brown at a press temperature of 260 °C.
2. Flexural modulus and strength in the desugared and dehydrate bagasse PP composites did not decrease until 240 °C, while those in the original bagasse PP composites did at 240 °C. The removal of sucrose and moisture in the original bagasse improved the limit of processing temperature in the bagasse PP composites due to a decrease of chemical interactions among water, sucrose, hemicellulose, and non-crystalline cellulose.

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