Effect of Time-dependent Moisture Absorption on Surface Roughness of Bagasse and Oil Palm Fibers / Polypropylene Composites

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The effect of time-dependent moisture absorption on the surface roughness in natural fiber/polypropylene composites after a hot-press molding was studied. The results showed that the moisture absorption in both bagasse and oil palm natural fiber composites correlated closely with time-dependent surface roughness in the composites. The surface roughness in all natural fiber composites increased with an increase of moisture absorption up to 50 d. The fibers absorbed moisture and swelled due to hydroxyl groups of celluloses in the natural fibers, which caused an increase in surface roughness. Time-dependent tests found that the surface roughness in long fiber composites was larger than that in short fiber composites due to inhomogeneous dispersion of long fibers in the vicinity of the surface of composites. The increase in surface roughness of oil palm composites was 55% that of the bagasse composites due to the lower temperature of thermal decomposition in oil palm fibers than in bagasse fibers. Thermal decomposition decreased the number of hydroxyl groups in fibers during heating and resulted in a decrease in moisture absorption in the palm fibers. Furthermore, the effect of the carbodiimide treatment on bagasse fibers was confirmed to reduce moisture absorption for both the fibers and the composites.

Keywords: Bagasse; Oil palm; Natural fiber; Polypropylene; Composites; Surface roughness; Thermal decomposition

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INTRODUCTION

It is desirable that environmentally friendly, low-density, natural fiber composites replace conventional fiber-reinforced plastics for effective utilization of resources (Mohanty *et al.* 2005; Abral *et al.* 2012a;). To achieve this objective, many studies on these composites have been reported. The natural fibers include bagasse, oil palm, bamboo, hemp, and sisal. The natural fiber composites are expected to give plastics high rigidity and strength (Chawla 1998; Khalil *et al.* 2001; Huda *et al.* 2005; Luz *et al.* 2007). To improve mechanical properties, the alkali treatment of fibers and maleic anhydride-modified polypropylene matrix have been incorporated for rigidity and strength by better interfacial bonding between hydrophilic natural fibers and hydrophobic plastics (Gassan and Bledzki 1999; Cao *et al.* 2006; Das and Chakraborty 2009).

Bagasse polypropylene composites have a problem unique to natural fiber composites: the surface roughness increases gradually with time after processing. This problem decreases the dimensional stability and external appearance of the molded part and needs to be resolved.

Plant fibers and animal fibers are hygroscopic because of their hydroxyl (-OH), amino (-NH2), and carboxyl (-COOH) hydrophilic groups (Abral *et al.* 2012b). The main component of plant fibers is cellulose, which is made up of hydroxyl groups that absorb water (Mohanty *et al.* 2004). This water absorption was decreased by alkali treatment (Ramandevi *et al.* 2012). Moisture absorption was revealed to depend on the chemical and the physical structures in fibers (Robertson *et al.* 2013). Also, moisture absorption makes debonding fiber-matrix in natural fiber composites (Dhakal *et al.* 2007). Thus, moisture absorption decreases mechanical properties in composites (Shahzad 2011).

However no article has examined the effect of time-dependent moisture absorption on the surface roughness in the natural fiber composites.

Bagasse fibers are heated, compressed, and deformed during hot-press molding. Generally, both bagasse fibers and polypropylene are dried to avoid thermal pyrolysis during molding (Shibata *et al.* 2010). The obtained composite gradually absorbs moisture from the air by diffusion after it is removed from the mold. This moisture diffusion occurs in the hydroxyl groups of the cellulose in bagasse fibers, not the polypropylene, because moisture absorption into natural fibers is approximately 7 to 9%, whereas it is only 0.02% in polypropylene. With moisture absorption, the hydrogen bonds are cut. The compressed fibers in the composites recoil with the release of elastic energy that is stored in microfibrils. In this process, the fibers swell and the surface roughness increases.

In this study, bagasse and oil palm fibers were used to fabricate polypropylene composites, and the relationship between surface roughness and time-dependent moisture absorption was observed. Mill-ground short fibers were used to improve the surface roughness after molding.

EXPERIMENTAL

Materials

Bagasse fibers are a by-product of sugar cane following the extraction of juice. Mesocarp fibers are a by-product of oil extraction from oil palm fruits. The fibers were mill ground and placed in a sieve to separate the long and short fibers. The characteristics of the long fibers and short fibers of the bagasse and palm are summarized in Table 1.

Both bagasse and palm fibers were washed twice with hot water at 80 °C for 10 min (95 wt% water and 5 wt% fibers). After the washing process, both fiber types were dried under a vacuum of 1 torr at 100 °C for 24 h in a vacuum furnace. The various fibers are shown in Fig. 1.

The polypropylene matrix was Noblen, AZ864 (Sumitomo Chemical Co. Ltd., Japan), with a molecular weight of 162,000 Da and a MW/Mn:1.4. The dried fibers (40 wt%) and polypropylene (60 wt%) were formed into pellets with a single extruder at 210 °C, as shown in Fig. 2.

Table 1. Fibers Used for Polypropylene Composites

Fiber type	Bagasse long	Bagasse short	Palm long	Palm short
Average length (mm)	7.7	0.45	3.4	0.20
Average diameter (mm)	0.53	0.15	0.6	0.10

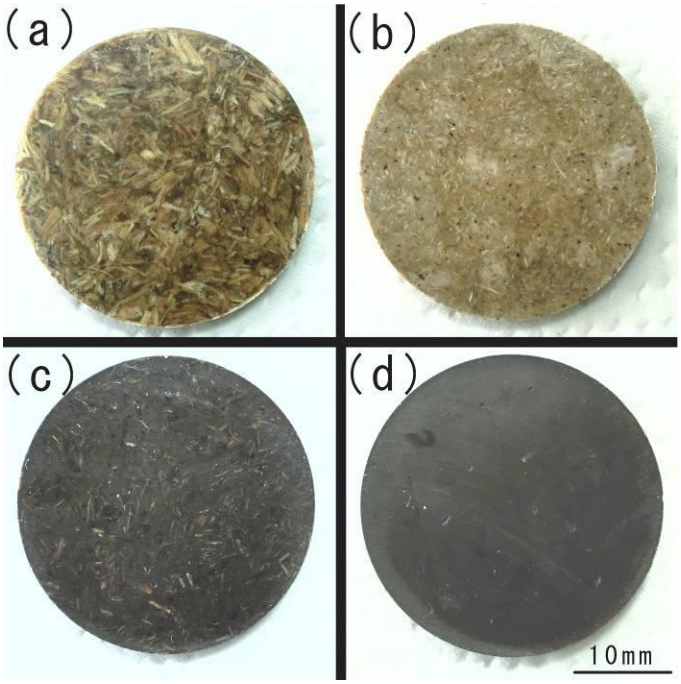

Fig. 1. The physical appearance of (a) bagasse long fibers, (b) bagasse short fibers, (c) palm long fibers, and (d) palm short fibers

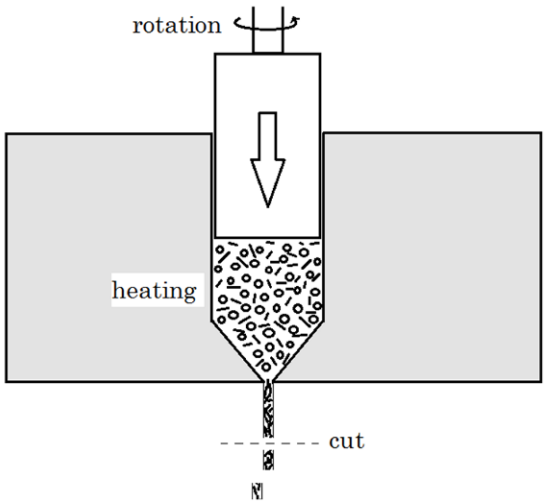

Fig. 2. The fabrication method for natural fiber composite pellets with a single screw extruder

Methods

Fabrication of composites

The composites were fabricated by a hot-press molding method (Shibata *et al.* 2005). The pellets from natural fibers and polypropylene were made with a single extruder at 200 °C as shown Fig. 2. The natural fibers were homogeneously mixed with polypropylene through two times passing with the extruder. The hot press machine consists of a metal cylinder and piston by 1.5 g of the pellets. The resulting composites were was circular shaped (30 mm in diameter, 1.5 to 2.0 mm in thickness). The hot-press molding was performed at a temperature of 200 °C and a pressure of 100 kgf/cm^2 for 5 min. The cylinder and piston were cooled by water for 5 min, after which the composite was removed from the cylinder.

Surface roughness testing

The surface roughness and the moisture absorption of the disc-shaped composite were measured for 50 d, and the relationship between them was investigated. The moisture absorption was defined as weight change percentage compared with original weight just after the disc shaped composites made. The average surface roughness of the three same composites was adopted as surface roughness. The surface roughness was measured with a Surftest SJ310, Mitsutoyo Co. Ltd. Japan. The average surface roughness was assumed the average of 8 random sites measuring 7.2 mm for each composite, and three same composites was used for one average data. R_z was adopted as the roughness parameter.

Scanning electron microscopy

The surface of composites were examined on a JEOL JSM-6510 scanning electron microscope (SEM; Japan) at an accelerating voltage of 10 kV or 20 kV.

Thermogravimetric measurement

Thermogravimetry (TG) was measured as the mass of fibers as a function of the time while constantly increasing the temperature of the fibers, at 10 °C/min up to 350 °C. The measurement machine used was Rigaku 8120 Themo plus TG, Rigaku. Co. Ltd. Japan. Samples were prepared with natural fibers milled and were dried under vacuum before test. The powder-like sample (10 mg) was used for the measurement.

Moisture absorption measurement

The sample fibers (1 g) were heated up to intended temperature and dried by a vacuum oven at 100 °C and 1 torr for 24 h. Then, the weight change of the fibers was measured immediately after being taken out of the oven using an electronic balance under an atmosphere of 20 °C and 50% relative humidity.

Carbodiimide treatment

The bagasse long fibers were dried in an oven under a 1-torr vacuum at 100 °C for 6 h. Carbodiimide (10 to 50 wt%), toluene, and stannous chloride catalyst (0.2 wt%) were stirred in a flask for 5 min to completely dissolve the carbodiimide. The carbodiimide was provided by Nissinbo Chemical Co. Ltd. (Japan) under the product name

Carbodiimide V-05. Carbodiimide V-05 was a liquid like carbodiimide modified isocyanate with no solvent. The fibers were immersed in the solution (20 x mass of fibers) for 10 min and removed from the flask to volatilize the solvent. The dried fibers were heated at 85 °C for 12 h in a furnace for the cross-linking reaction.

RESULTS AND DISCUSSION

Effect of Moisture Absorption on Time-dependent Surface Roughness in the Bagasse and Palm Fiber Composites

Figures 3a and 3b show the surface roughness profiles in the long and short bagasse/polypropylene composites immediately after formation and after 50 d, respectively. The surface roughness in the long fiber composites was found to be higher than that in the short fiber composites immediately after formation. Large mountain parts correspond to the fibers. In the case of long fiber composites after 50 d, the surface roughness was higher than that at the initial day. In the case of short fiber composites, it was not clear that the surface roughness increased.

Figures 4a and 4b show the relationship between elapsed time, surface roughness, R_z, and moisture absorption in the bagasse/polypropylene and palm/polypropylene composites, respectively. In all composites, the surface roughness and moisture absorption increased after formation. In the case of the bagasse long fiber, the surface roughness (R_z) was 4 μm just after molding, and the roughness increased with elapsed time up to 11.5 μm at 1350 h (55 d). The initial surface roughness is considered to be the initial recoil of fibers by removal of the compressive pressure of the mold. The surface roughness, R_z, of the metal mold was 0.1 μm. The moisture absorption increased from 0% to 3.1% in the same time period. Both surface roughness and moisture absorption increased rapidly immediately just after formation and increased slightly with elapsed time. After molding, the surface roughness and moisture absorption increased until an elapsed time of 750 h (30 d), after which the slope increased slightly. Thus in the case of the long bagasse fibers, the surface roughness and moisture absorption clearly correlated.

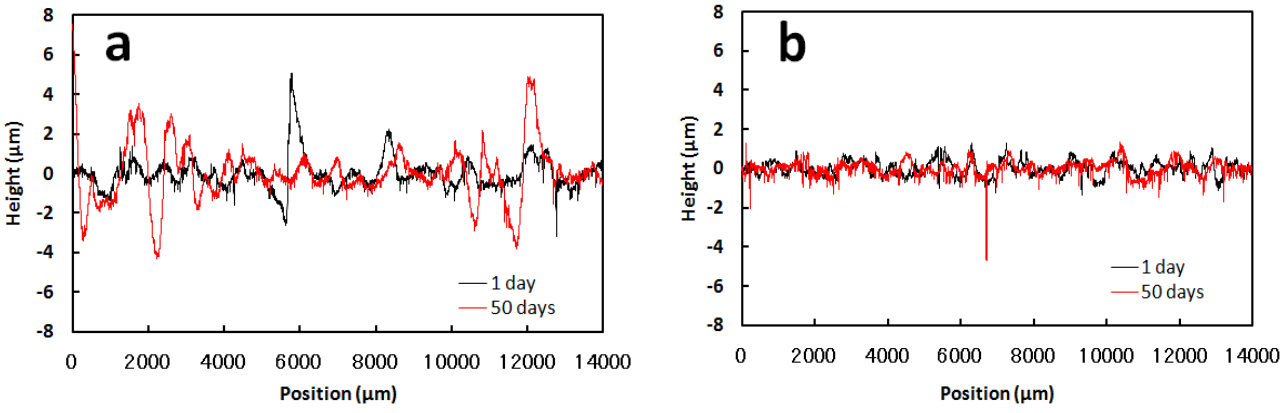

Fig. 3. Surface roughness profiles of (a) bagasse/polypropylene long fiber composites and (b) bagasse/polypropylene short fiber composites

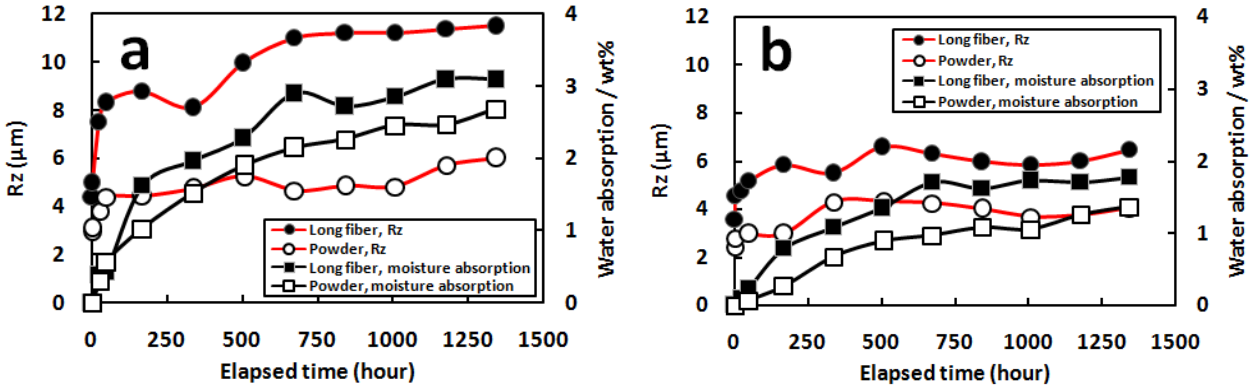

Fig. 4. Relationship between elapsed time and surface roughness, Rz, of (a) bagasse/polypropylene and (b) palm/polypropylene

Figure 5 shows the relationship between elapsed time and moisture absorption in vacuum dried bagasse fibers. As shown, the mass of bagasse fibers increased rapidly for 60 min. However, the bagasse fibers might denature, and those hydroxyl groups decrease when the fibers are heated. Thus, the weight of the bagasse fibers that was heated at 200 °C, 1 MPa of pressure for 5 min with a hot-press machine was also measured. As a result, the unheated bagasse fibers absorbed up to 8.2% moisture, while the heated bagasse fibers absorbed up to 7.1% moisture. How this heating affects the bagasse mass is not clear at this point; however, the decrease of the heated bagasse mass during the hot press forming would be less 1% according to TG profiles around 200 °C as shown in Fig. 8 below. Thus, if we assume all the 40 wt% bagasse fibers in the composites reach the equilibrium moisture content of 7.1%, then the weight of the composites increases from 2.8 wt% to 3.2 wt%. These estimated values agreed well with the saturated moisture absorption of the bagasse composites from 2.8 wt% to 3.1 wt% shown in Fig. 4a. It is probable that the all bagasse fibers in the composites absorbed moisture and reached equilibrium moisture content by 50 d after the formation.

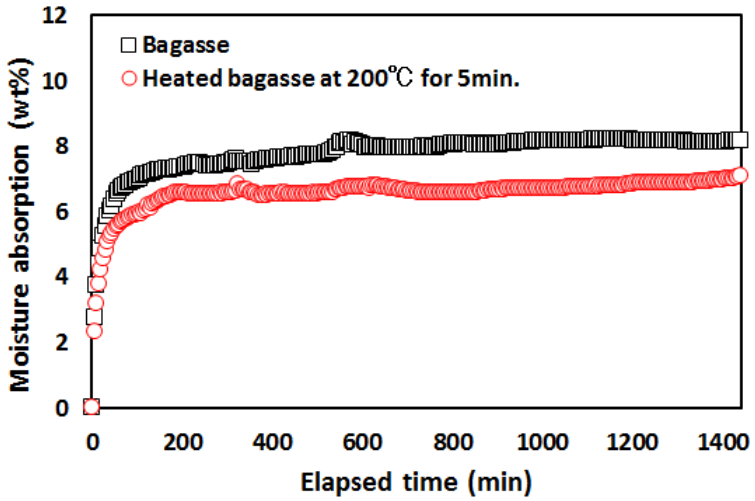

Fig. 5. Relationship between elapsed time and moisture absorption of bagasse fibers

There was little difference in moisture absorption between the short fiber and long fiber bagasse composites; however, there was a large difference in the surface roughness (Fig. 4a). In short bagasse fiber composites, the time-dependent change in the surface roughness was apparently smaller than that in the long fiber composites. However, the thickness of the short bagasse composites increased 8% at a moisture absorption of 1.5 wt%.

Figure 6 shows the surface appearance of bagasse fibers by SEM. In bagasse composites, the fibers swelled and were exposed on the surface after 50 days. This was especially obvious with the large fibers. The long fibers tend to have inhomogeneous distribution because of the large size, while the short fiber composites exhibited homogeneous dispersion. This difference in fiber distribution can be seen in a comparison of Figs. 2a and 2b. In the case of the long fiber, the fiber in proximity with the composite surface absorbs moisture and swells, while the polypropylene does not absorb moisture; this difference is assumed to be the cause of the surface roughness increase.

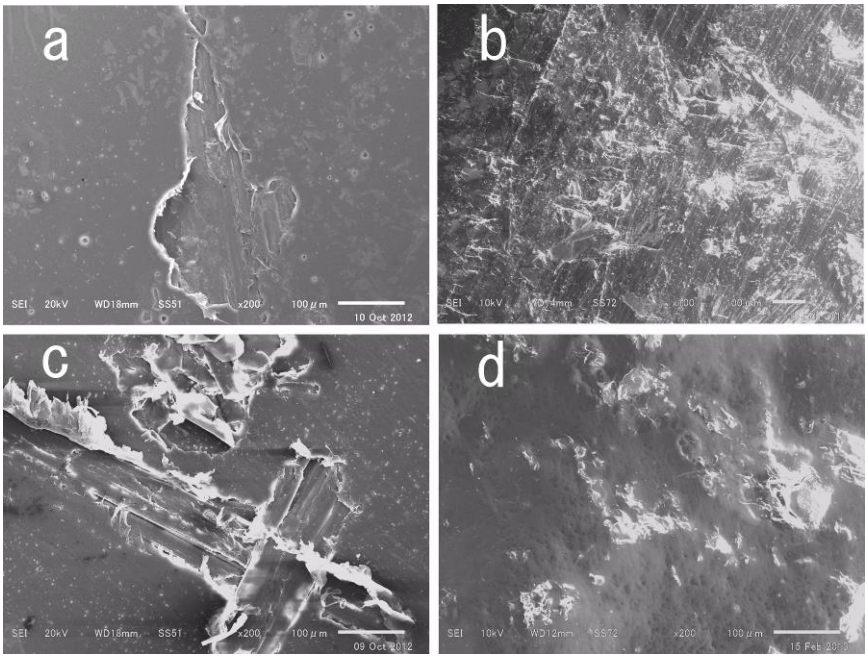

Fig. 6. Surface appearance of (a) bagasse long fiber composite at 1 d, (b) bagasse long fiber composite at 50 d, (c) bagasse short fiber composite at 1 d, and (d) bagasse short fiber composite at 50 d

In the case of the short fibers, the fibers absorb moisture as well as long fibers; however, the entire surface homogeneously swells because of the fibers homogeneous distribution. Thus, all the moisture absorption did not contribute to the surface roughness increase but rather led to an increase of thickness in the specimen. From these experimental results, the long fibers were shown to be larger than the short fibers in length and width, but with fewer numbers. The difference in the fiber size causes inhomogeneous distribution in the composites. This is the reason why the surface roughness in the long fiber composites increased more than in short fiber composites

when the fibers recoiled by water absorption. Short fiber composites absorb moisture as well as long fiber composites; however, the small fibers swell homogeneously and the increase in surface roughness is smaller.

Alternately, as shown in Fig. 4b for the palm fibers, the moisture absorption in both the long and short fiber composites was found to be lower than that in bagasse composites. In the long fiber, the surface roughness increased from 3.8 μm at day 1 to 6 μm at day 50. The final surface roughness of the long palm fibers was 55% that of the bagasse long fiber composites.

The moisture absorption of palm long and short fibers was 1.8% and 1.4%, respectively. These values also were approximately 55% that of bagasse long and short fiber composites.

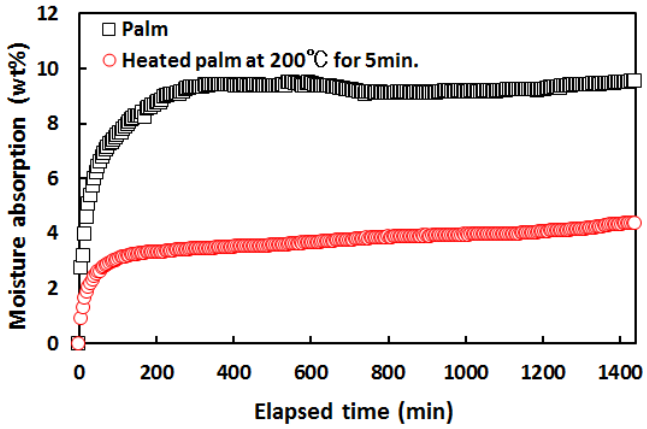

Fig. 7. Relationship between elapsed time and moisture absorption of palm fibers

Figure 7 shows the relationship between elapsed time and moisture absorption of palm fibers. The measurement condition was the same as that for bagasse fibers. The moisture absorption in the palm fibers washed with hot water reached 9.5 wt%, while that for the palm heated to 200 °C for 5 min was only 4.4 wt%. Also, the heated fibers were carbonized to a dark color, as shown in Fig. 1c and 1d. This change in appearance indicates chemical denaturation.

The palm fibers were found to have a lower thermal decomposition temperature than bagasse fibers. Generally, palm fibers contain palmitic acid, oleic acid, and linoleic acid. These saturated and unsaturated fatty acids may react with the functional groups of fibers. Through these chemical reactions, the total hydroxyl group of the fiber component such as cellulose and hemicellulose decreases dramatically and results in lower saturated moisture absorption.

The saturated moisture absorption of palm fibers heated to 200 °C for 5 min was found to be 3.5 wt%. Using this value, the moisture percentage of fiber content 40 wt% of composites corresponds to 1.4 wt% of the composite weight. This calculation estimation agreed well with the experimental results shown in Fig. 4b.

For the palm fiber composites, the saturated moisture absorption decreased because the hydroxy groups of the fibers were decreased dramatically by thermal

decomposition during heating. The swelling of fibers decreased and the surface roughness change was smaller than that in bagasse fiber composites.

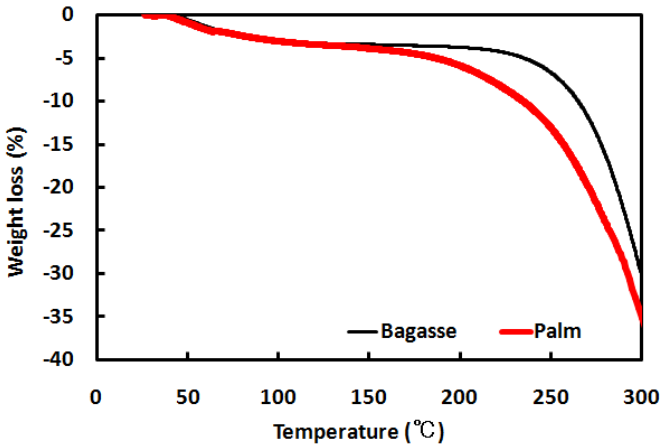

Fig. 8. TG profiles for bagasse and palm fibers

Effect of Carbodiimide Treatment on Moisture Absorption and Time-dependent Surface Roughness in the Bagasse Fiber Composites

Figure 9 shows the moisture absorption profiles of bagasse long fibers with a change in carbodiimide concentration. It was found that the moisture absorption in the bagasse fibers decreased with increasing concentration. This is because the hydroxyl groups of the cellulose were transformed to hydrophobic groups by carbodiimide treatment. Figure 10 shows the relationship between time-dependent moisture absorption of bagasse composites that used fibers treated by wt% carbodiimide 20 and elapsed time. In both bagasse composites, the initial surface roughness was 4 μm. However, 700 h later (30 days), the surface roughness of untreated fibers reached 11 μm, while that of the carbodiimide-treated bagasse fiber was 6.1 μm.

The number of hydroxyl groups in the fibers was assumed to decrease because of the substitution reaction of hydroxyl groups with carbodiimide treatment. Thus, lower moisture absorption caused a smaller change in the time-dependent surface roughness.

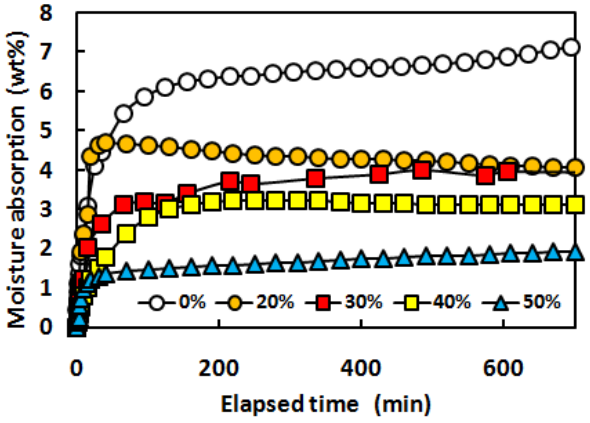

Fig. 9. Quantity of carbodiimide and moisture absorption profiles in bagasse fibers

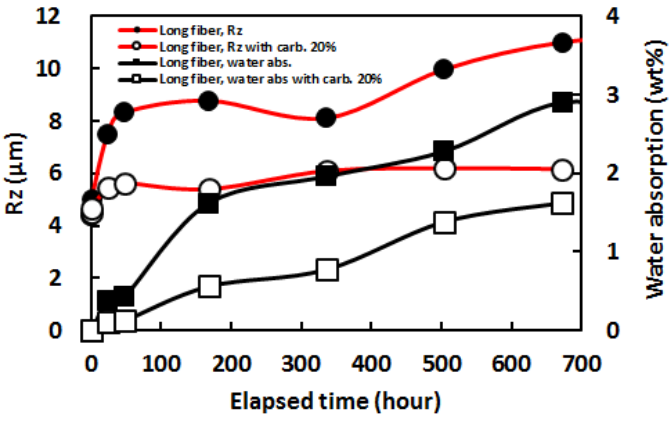

Fig. 10. Relationship between elapsed time and surface roughness in the bagasse composites with and without carbodiimide treatment

CONCLUSIONS

1. A clear relationship was found between the increase of surface roughness with time-dependent moisture absorption in the bagasse, palm, and polypropylene composites by hot press molding. This is because the hydroxyl groups of natural fibers absorb moisture in the air by diffusion, and the compressed fibers recoil. The natural fibers in the composites absorb moisture up to saturation.

2. In the case of the bagasse short fiber composites, the surface roughness increased less because of the smaller size and homogeneous distribution, although the thickness in the short fiber composites increased.

3. Comparing bagasse fibers and palm fibers, the moisture absorption in palm fibers was found to decrease to 55% that of the bagasse fibers. The hydroxyl groups in palm fibers decreased dramatically by heating because of the saturated and unsaturated fatty acid in the palm fibers. The lower moisture absorption in palm fibers resulted in lower moisture absorption in the composites and a smaller surface roughness increase.

4. To decrease the moisture absorption in bagasse fibers, carbodiimide treatment was performed. The moisture absorption and the surface roughness in treated bagasse fibers decreased to 50% that of untreated bagasse fibers.

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