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Effect of Drying Temperature on the Physical Properties of Binderless Fiberboard from Bagasse: Study of Water Absorption

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ABSTRACT

The objective is to study the utilization of bagasse as fiberboard by relying only on the role of lignocellulose to replace the use of chemical adhesive. Binderless fiberboard was produced through a wet process to enhance hydrogen bonding between celluloses. Various drying temperatures were applied during the pressing process as an independent variable which varied from 110ºC, 130ºC, 150ºC, 170ºC, and 190ºC. The effect of drying temperature on the physical properties of fiberboard, which are water absorption and dimension swelling, was then investigated. The calculations show that the absorption and thickness swelling increased from temperature of 110ºC to 130ºC by 95.23% to 101.02% and by 65.30% to 68.03% respectively and then decreased from temperature of 130ºC to 190ºC by 101.02% to 37.06% and by 68.03% to 22.75%. Meanwhile, the length swelling decreased along with the increase in temperature from 110ºC to 190ºC by 1.97% to 0.83%. As the density of fiberboards is 1.0164-1.0210 g/cm^3 included in high-density fiberboard (HDF), the results indicate that none of the fiberboards meet the classification of standard board for HDF with required water absorption below 35% (JIS A 5905 (2003)). Regarding its absorption behavior, the diffusion mechanism on each fiberboard was classified as pseudo-Fickian model with release exponent condition below 0.5. The model indicates that rate water diffusion is much less than polymeric relaxation process.

Keywords: Bagasse; High-density fiberboard; Hydrogen bonding; Drying temperature; Water absorption

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1. Introduction

Recently, binderless fiberboard has been relatively much studied related to its production through wet-forming process in replacing conventional fiberboard. Conventional fiberboard is commonly produced by adding chemical adhesives, such as urea-formaldehyde resin ($(OCNHCH_2NH)_n$), to help strengthening its structure, but vapor which occurs when the adhesives are heated is considerably harmful to living things around [1]. Meanwhile, binderless fiberboard, which relies only on the enhancement of hydrogen bonding between adjacent fibers using water medium, tends to be more environmentally friendly [2]. The presence of water helps celluloses to extend outwards from fiber surfaces and reach the required minimum distance of cellulose-cellulose interaction (0.15-0.35 nm). This mechanism is called the diffusion theory of Voiutskii [3]. The outside crystalline regions and the entire amorphous regions of cellulose chains tend to interact more with water by forming hydrogen bonds before eventually the water interference is reduced through the pressing and drying process to allow hydrogen bonding between extended cellulose chains on each adjacent fiber surfaces to occur. Besides OH-O and CH-O hydrogen bonds, it is known that electrostatic attraction, polar interactions, and van der Waals forces also make an important contribution [3-4].

There are several research papers studying the mechanical properties of this type of fiberboard successfully with various biomass and forming conditions, such as corn straw with various pressures (T. Wu, 2015), rice straw with various drying temperatures (J. Zhang, 2020), rice straw and corn stalk with various mass ratios (Y. Hua, 2018), and so on [2, 5-6]. However, the study of physical properties, such as water absorption, is relatively still few in number where in fact water absorption is an important property required to expand the range of exterior applications (e.g., siding and furniture) and compete more efficiently against synthetic fiber. It is because material resistance to moisture is closely related to other important properties, such as dimensional and mechanical stability [7, 20]. Therefore, current research focuses on the study of water absorption, specifically due to the effect of drying temperature.

A considerable research on natural fiber is conducted by considering biomass material used. Bagasse is one of the potential materials due to its chemical composition and environmental benefits. Focusing on the basic idea of binderless fiberboard making, bagasse shows a higher cellulose content than the other content. According to Soccol (2011), bagasse has percentage of cellulose of 32.0-44.0%, hemicellulose of 27.0-32.0%, lignin of 19.0-24.0%, and ash of 4.5-9.0% [8]. Meanwhile, on the environmental aspect, sugarcane is known to be cultivated on about 25.9 million hectares in more than 90 countries with a worldwide harvest of approximately 1.84 billion tons in 2017 [9]. Based on 9% of the total plant mass, sugarcane production may generate a large amount of bagasse with estimated waste of 165.74 million tons from the extraction process, and it is expected to grow along with sugarcane production by 1.1% p.a in 2018 [9, 21]. However, most industries prefer to dispose it as agricultural waste or burn it directly due to time efficiency. These practices are considered to have negative impact on the environment as a cause of air, water, and soil pollution [22]. Therefore, by utilizing bagasse as a fiberboard, it can be a safe alternative way to reduce organic waste and save 87.74% of raw material cost ($15.69/ton) compared to green wood ($128/ton) [23]. This practice is not only a form of waste management but also a form of participation in a project of BR&D Board and California ASAE International Meeting 2001 which one of them is to emerging bio-based products [10].

2. Materials and Methods

2.1 Sugarcane bagasse

Bagasse (Saccaharum spp.) was obtained from farm areas around Mie University, Japan. It was then dried under sunlight for several days to reduce moisture content and inhibit decay.

2.2 Fiberboard making methods

Fiberboard was made through 5 processes, namely cutting, soaking, refining, concentration (bulk density) determination, and forming (see Fig. 1). Dried bagasse was cut off into chips using an agricultural cutter machine, SU-16 Kowa. The chips were soaked in water for 96 hours before being poured into the inlet of beatfiner. Beatfiner, Type-A Satomi, was used to break down chips into fiber pulp by pumping them through rotating blades repeatedly for 25 minutes. Afterwards, fibers were sifted using a sieve with mesh size of 4 mm. The uniformity of fibers in fiberboard was set by determining the concentration of dried fiber in fiber pulp. Five samples of 100 ml fibers were dried in laboratory oven for 24 hours under a temperature of 110ºC. The concentration (bulk density) was obtained at 0.035 gr/ml. Forming was done by using a hot-press machine, RH-50 Tsushima. Five hundred ml fiber pulp were poured into metal mold with holes of 2 mm in diameter, inter-hole matrixes of 7 mm, and dimension of 100x100x40 mm (in length, width, and depth). After that, the rest of the molding tools were stacked on the metal mold in sequence. Five MPa loads were applied to the stack for 2 hours while being heated with various drying temperature of 110ºC, 130ºC, 150ºC, 170ºC, and 190ºC [20].

Fig. 1. Process of making binderless fiberboard [20].

2.3 Fiberboard testing methods

Water absorption and dimension swelling of fiberboard were determined with JIS A 5905 (2003) (ratio-modified) at the same time using 2 samples from each drying temperature with dimension of 5x5 cm in length, as seen in Fig. 2. The mass, thickness, and length of samples were measured both before and after immersion for 24 hours. Samples were immersed at a depth of 2 cm under the water surface and at a temperature of 20ºC. Meanwhile, removing excessive water was done by holding samples between 10 blotting papers with dimension of 6x6 cm and under load with weight of 750 g for 30 seconds. The result was then calculated using Eq. (2.1), (2.2), and (2.3) [11]:

$$WA = \frac{m_2 - m_1}{m_1}, \tag{2.1}$$

$$TS = \frac{T_2 - T_1}{T_1}, \tag{2.2}$$

$$LS = \frac{l_2 - l_1}{l_1}, \tag{2.3}$$

where WA is water absorption (%), TS is thickness swelling (%), LS is length swelling (%), m_1 is mass of sample before immersion (g), m_2 is mass of sample after

immersion (g), T_1 is thickness of sample before immersion (cm), T_2 is thickness of sample after immersion (cm), l_1 is length of sample before immersion (cm), and l_2 is length of sample after immersion (cm).

Absorption kinetic was determined in accordance with the report of Petchwattana in 2017 using 2 samples from each drying temperature with dimension of 5x5 cm in length, as seen in Fig. 2. The test method remained the same as the water absorption test with measurement of mass only. However, specifically, the mass of samples was measured every 2 hours for 24 hours. The result was then calculated using Eq. (2.1) and (2.4) [11-12]:

$$log\left(\frac{M_t}{M_\infty}\right) = log(k) + n\,log(t), \qquad (2.4)$$

where M_t is water absorption at t time (%), M_∞ is water absorption at saturation point (%), t is immersion time (hour), k is constant of network structure, and n is constant of release exponent.

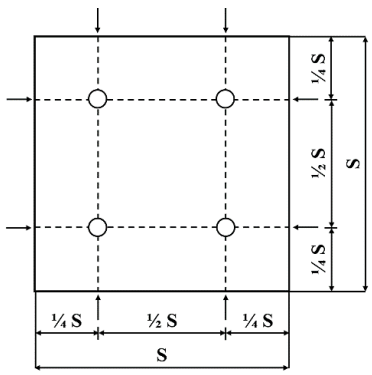

Fig. 2. Appearance of water absorption test sample and its measurement point [20].

3. Results and Discussion

3.1 Water absorption and swelling

The calculations show that the increase in drying temperature gave fluctuating results of the water absorption and dimension swelling of fiberboards (see Fig. 3). Both water absorption and thickness swelling increased from temperature of 110ºC to 130ºC by 95.23% to 101.02% and by 65.30% to 68.03%, respectively. Then, both of them decreased from temperature of 130ºC to 190ºC by 101.02% to 37.06% and by 68.03% to 22.75%. Meanwhile, the length swelling decreased along with the increase in temperature from 110ºC to 190ºC by 1.97% to 0.83%. The results indicate that drying temperature has an inversely proportional relationship with water absorption and dimension swelling, specifically above temperatures of 130ºC. As the density of fiberboards is 1.0164-1.0210 g/cm^3 which is included in high-density fiberboard (HDF), the results also indicate that none of the fiberboards meet the classification of standard board for HDF with required water absorption below 35% based on JIS A 5905 (2003) [11].

The unmet standard due to high water absorption is caused by the basic nature of woody material which is able to be rewetted in a particular condition called water sorption and plasticization [14, 16]. According to Jakes (2019), water is absorbed in amorphous portion of lignocellulosic polymers in many ways (see Fig. 4): (1) 'hydrogen bonded water' where water forms hydrogen bonds with hydrophilic moieties, such as hydroxyl groups (-OH), in the polymer network to break formed polymer-polymer hydrogen bonding and lubricate motions between neighboring polymers; (2) 'molecular solution water' where water is randomly mixed in the polymer network that results in increase in free volume; (3) 'absorption in "holes" water' where water molecules are absorbed in holes, which are large free volume elements, without perturbing polymer structure even though a hydrogen bond may still occur on hole borders in the polymer network; and (4) 'water cluster' where water agglomerates into clusters after reaching high moisture content [16]. Eventually, the gain of moisture above fiber saturation point (FSP) (>27%; free water region) leads the dimension of material into

swelling [15]. Swelling occurs due to particular absorbed waters related to plasticization, namely hydrogen bonded and molecular solution. Swelling is represented by the polymer chains being pushed apart by the water molecules (see Fig. 4) [16].

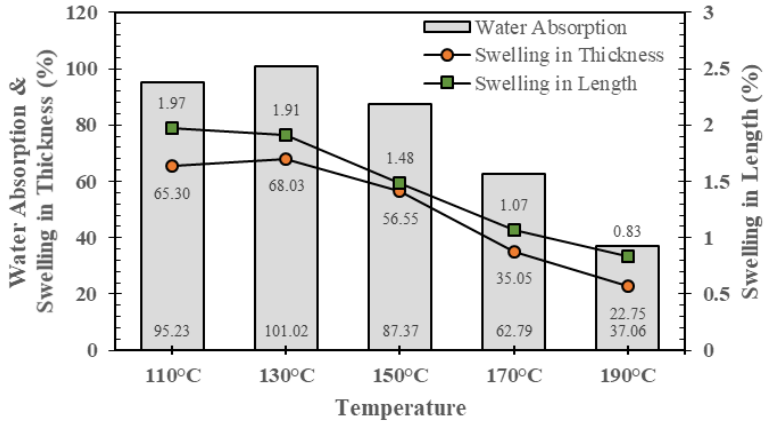

Fig. 3. Effect of various drying temperatures on water absorption, thickness swelling, and length swelling.

Furthermore, the increase in water absorption and dimension swelling from temperature of 110ºC to 130ºC may be caused by enlarged free volume. According to Jakes (2019), generally free volume increases along with the increase in temperature. A larger free volume may accommodate more water molecules being absorbed in the polymer structure even without perturbing and provide a high contact area for plasticization to give greater mobility and quicken relaxation of polymer segments which physically occur as swelling [16]. Meanwhile, the decrease in water absorption and dimension swelling from temperature of 130ºC to 190ºC or even since 110ºC may be caused by softened lignin and de-polymerization of hemicellulose. According to Boon (2019) and Luo (2014), thermal treatment at certain temperature promotes softening lignin and allows lignin to surround fiber matrixes, including polymer structures and free volume of cellulose and hemicellulose, which causes inhibition of water absorption due to its hydrophobic property [17-18]. Supported by Basu (2013), lignin starts to soften at temperatures of 80-90ºC and sets prominently at 120-150ºC. Basu also stated that at temperatures of 150-200ºC, de-polymerization of hemicellulose is initiated which leads to the decrease in water attraction [14]. As is widely known, hemicellulose has the highest hydrophilicity among lignocellulosic polymers with the order regarding the accessibility of water to their hydroxyl groups (-OH) as follows: hemicellulose $>$ cellulose $>$ lignin [13]. In addition, hemicellulose contributes about 39.12% of the theoretical lignocellulosic hydroxyl content in bagasse biomass (hemicellulose of 15.15 mmol/g, cellulose of 18.52 mmol/g, and lignin of 5.06 mmol/g) [24]. Hence, the decrease of material ability in absorbing water results in the decrease in dimension swelling.

3.2 Water absorption kinetic

The calculation results show that the constant of release exponent decreased with the increase in temperature from 110ºC to 150ºC by 0.0664 to 0.0527 and then increased from 150ºC to 190ºC by 0.0527 to 0.3473 (see Table 1). The results indicate that the diffusion mechanism of all fiberboards meets the classification of pseudo-Fickian with required constant of release exponent below 0.5 [19].

Fig. 5 shows the obtained curve characteristic of diffusion mechanism which in this case is pseudo-Fickian. According to Lars (1992), pseudo-Fickian curves resembles true Fickian curves inasmuch as they do not exhibit any inflection in slope. The curve is also known as initial two-stage behavior which indicates swelling part in diffusion mechanism. The initial portion of the curve changes linearly, but the rest of the curve changes insignificantly in plateau in which the approach to final equilibrium is very slow [19]. Furthermore, the initial portion of curve of fiberboard 190ºC appears to have longer duration in reaching plateau which may be caused by the inhibition of water absorption due to softened lignin and de-polymerization of hemicellulose marked with the significant increase in constant of release exponent of 0.3473 (see Table 1).

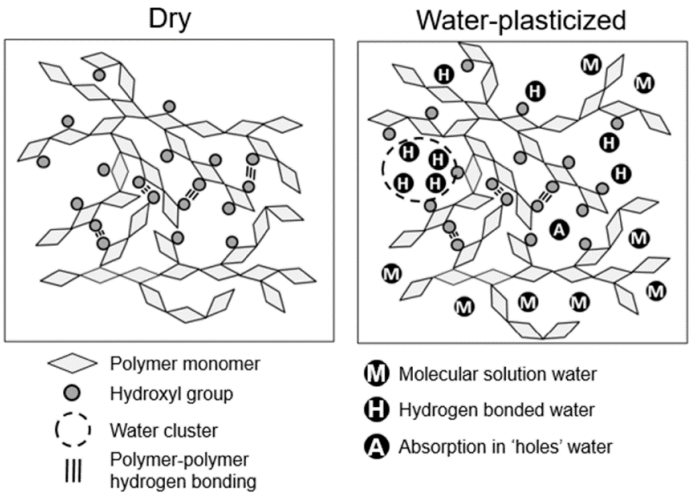

Fig. 4. Schematics showing different states of polymers in dry and water-plasticized condition or after rewetting due to water sorption and plasticization [16].

Linear fitting of logarithmic water absorption ratio and immersion time is carried out to obtain the constant of release exponent, as seen in Fig. 6 and Table 1. All fiberboards show having the constant of release exponent below 0.5 classified as pseudo-Fickian or Less Fickian model ($n<0.5$). The model indicates that molecular relaxation is much faster than the diffusion process ($R_{relax} \gg R_{diff}$). For the fluctuating results, bigger constants mean that the gap keeps decreasing ($R_{relax} > R_{diff}$). The faster the molecular relaxation towards diffusion process, the quicker water enters and leaves the polymer matrix [16]. The diffusion can also be promoted by the higher amounts of free volume, and the increase in temperature can accommodate it, as seen from temperature 110ºC to 150ºC, marked with the decrease in constants. However, the existence of softened lignin and de-polymerized hemicellulose due to higher temperature, as seen from 150ºC to 190ºC, causes lower amounts of free volume and slower molecular motions marked with the increase in constants.

Table 1. Data summary on linear fitting of logarithmic diffusion curve.

Fiberboard	Average		
	WA (%)	n	k
110ºC	94.1434	0.0664	0.8065
130ºC	96.8992	0.0532	0.8358
150ºC	86.3532	0.0527	0.8334
170ºC	62.8809	0.0633	0.8092
190ºC	41.2359	0.3473	0.3281

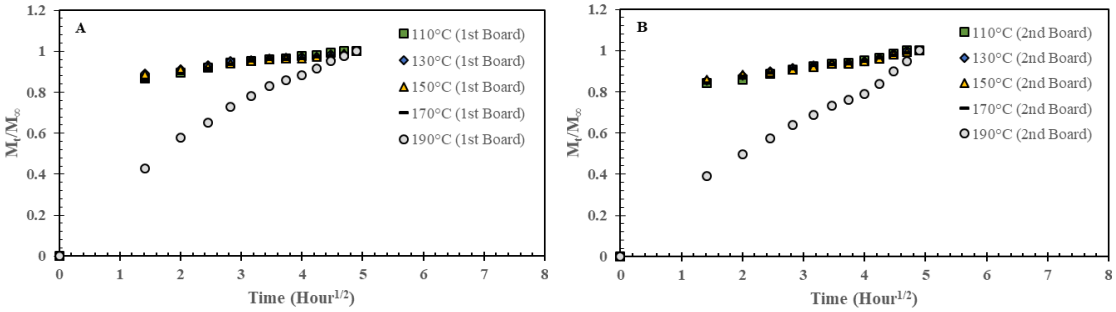

Fig. 5. Obtained pseudo-Fickian curve showing relationship of water absorption ratio to square root of immersion time, A). 1st sample; B). 2nd sample.

Meanwhile, based on the advanced analysis technique of Nuclear Magnetic Resonance (NMR), the increase in aggregation size of cellulose chains due to temperature elevation contributes to the decrease in water intake and molecular relaxation by forming cross-linking of hydrogen bonding (OH-O; strong link and CH-O; weak link) in fiber microstructure [4, 25]. The effect can be clearly indicated by cellulose crystallinity index as the nature of cellulosic material may generate an irreversible agglomeration which is resistant to water exposure [26]. However, according to Salmen (2018) using NMR analysis technique on a spruce pulp, the formed cellulose crystallinity is not significantly affected at drying temperature of 80ºC and 140ºC with results of about 56.8% (initial condition), 57.4% (at 80ºC), 58.4% (at 140ºC) [27]. As for the results in the current study, it is believed that the effect may be seen significantly when the temperature is over than 190ºC.

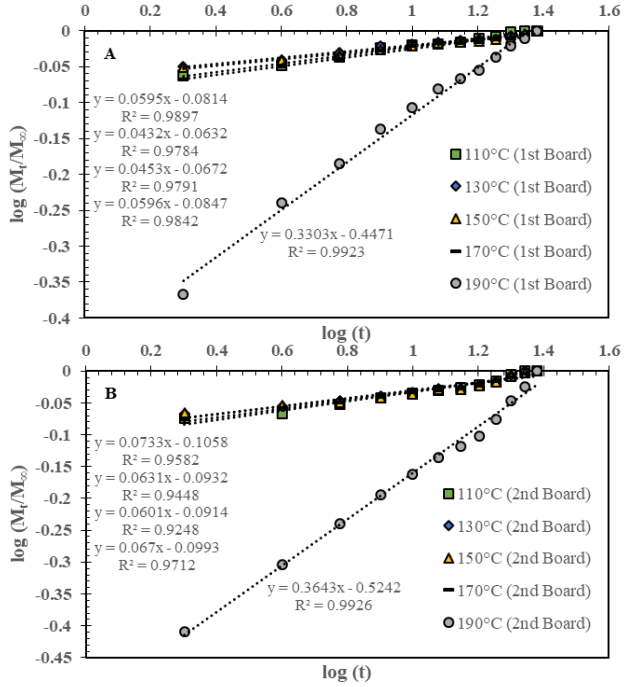

Fig. 6. Linear fitting of logarithmic diffusion curve, A). 1st sample; B). 2nd sample.

4. Conclusion

Fiberboards have been produced successfully by utilizing bagasse without using chemical adhesive. The effect of various drying temperatures on the physical properties of binderless fiberboard have been investigated. From the results obtained, the following conclusions are drawn:

- None of the binderless fiberboards meet any standard of water absorption parameter based on JIS A 5905 (2003) for high-density fiberboard.
- The most optimum drying temperature in the research is 190ºC. On average, the fiberboard has water absorption of 37.06%.
- Generally, the trends of physical properties, such as water absorption, dimension swelling, and diffusion mechanism, fluctuate related to the changes in the lignocellulosic polymer structures which mostly can be identified around temperature 150ºC.

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