J Wood Sci (2005) 51:26–32
DOI 10.1007/s10086-003-0608-9

ORIGINAL ARTICLE

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Chemical changes in steam-pressed kenaf core binderless particleboard

Received: September 26, 2003 / Accepted: November 18, 2003

Abstract The effects of chemical changes in kenaf core binderless particleboards on the bonding performance and thickness swelling of boards were investigated by chemical and spectroscopic analyses. Mild steam-injection treatments (0.6–1.0 MPa) caused significant degradation of hemicelluloses, lignin, and cellulose. Conventional hot pressing caused a lower degree of degradation of the chemical components. The hot-pressed kenaf core board without any binders showed poor bonding performance. Thus, it was found that partial degradation of the three major chemical components of the kenaf core by mild steam-injection treatment increased the bonding performance and dimensional stability of the binderless boards, and gave better quality binderless boards than those made by hot-pressing treatments.

Key words Chemical composition · Binderless board · Kenaf core · Steam-injection pressing · Bonding performance

Introduction

A number of factors affect the final bonding properties of particleboard, such as the type of raw material, board density, type of treatment, binder type, and others. Almost all of these factors will interact with the others in one way or another. Because no resin is used in binderless boards, the self-bonding strength is improved only by activating the chemical components of the board constituents during steam/heat treatment. To date, the mechanism of self-bonding during steam/heat treatment has not been completely elucidated. However, degradation of hemicellulose during steam/heat treatment to produce furan products is believed to play an important role in self-bonding. Therefore, binderless boards are usually prepared from nonwoody raw materials, which are rich in hemicellulose.[1–4] According to Suzuki et al.,[3] the main bonding strength of binderless boards is due to the lignin–furfural linkages that are generated during the hot-press process of producing steam-exploded oil palm frond fiber.

In the mid-1980s, Shen[5] developed and patented a steam-explosion process for treatment of lignocellulosic materials to make binderless boards. A number of studies have been published regarding the manufacture of binderless boards, and they can usually be divided into three groups: the hot-pressing system,[1,2,6] the steam-explosion process before hot pressing,[3,4] and steam-injection pressing.[6,7]

Steam treatment has been known to be an effective method for improving the dimensional stability of wood-based composites. In resin-bonded boards, steam-injection pressing not only treats the wood but also affects the curing of adhesive resin. Hsu et al.[8] produced dimensionally stable wood-based composite made from steam-treated wood at 1.55 MPa for 3–4 min. The dimensional stability of binderless medium-density fiberboard (MDF) made from mixed softwood and hardwood fibers can also be improved by using steam-injection pressing, although the internal bonding (IB) is very low.[6]

Xu et al.[7] developed binderless particleboards from kenaf core using steam-injection pressing. The IB strength of these boards was excellent, even when low steam pressure was used (0.6–1.0 MPa). The thickness swelling (TS) of binderless boards also appears to be affected by steam-injection pressing. The differences in mechanical properties between hot-pressed board and steam-treated boards have been clearly shown. Considering that no binder is used, the

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Part of this report was presented at the 4th International Wood Science Symposium, Serpong, Indonesia, September 2002; and at the 53rd Annual Meeting of The Japan Wood Research Society, Fukuoka, March 2003

bonding performance and dimensional stability of the kenaf core binderless particleboards is likely to be greatly affected by chemical changes of components during steam/heat treatment. However, a certain degree of degradation of the chemical components should not cause a reduction of the bonding strength and may increase the compressibility of the boards. Unfortunately, no studies of the relationship of chemical changes of boards and their bonding performance have been published to date.

This study was designed to investigate the chemical changes of kenaf core binderless board manufactured by steam-injection pressing or hot pressing, and discuss their effects on the self-bonding performance and dimensional stability of particleboards.

Materials and methods

Materials

Kenaf core variety Chinpi-3 (*Hibiscus cannabinus L.*) binderless particleboards were used as the raw materials for chemical analyses.[7] Boards were prepared by steam-injection pressing under the following conditions: 1 MPa for 7 min, 10 min, 15 min, and 20 min; 0.8 MPa and 0.6 MPa for 20 min. Boards prepared by hot pressing were subjected to 190°C for 20 min. The target density of all binderless boards was 0.55 g/cm^3. All boards and kenaf core (control) were cut and ground to pass through 30 mesh screen, were retained on 60 mesh screen, and were then air-dried.

Chemical analysis of kenaf core and binderless boards

The kenaf core samples were extracted successively with a mixture of ethanol and benzene (1:2, v/v) for 24 h by refluxing, and then with distilled water at 60°C for 3 h. The analyses of extractives were carried out in duplicate. The water-soluble fractions were then subjected to analyses for neutral sugar composition and NMR spectroscopy. Klason and acid-soluble lignins were determined by a Klason method. Holocellulose content was determined by a Wise method. The α-cellulose content was determined using the holocellulose by extraction with 17.5% NaOH. All the chemical analyses were carried out in triplicate.

The water-soluble fraction was analyzed by ^{13}C-nuclear magnetic resonance (NMR) spectroscopy. The spectra were measured on a Jeol λ-400 NMR spectrometer (Tokyo, Japan) at 100 MHz in D_2O at 27°C using 1,4-dioxane as an internal standard (67.40 ppm).

The neutral sugar composition of the water-soluble fraction was determined as an alditol acetate by gas chromatography (Shimadzu GC-17A, Shimadzu, Kyoto, Japan) on an Ulbon HR-SS10 column (0.25 mm × 25 m, Ulbon, Kyoto, Japan) after acid hydrolysis. The acid hydrolysis was carried out with trifluoroacetic acid (TFA) at 100°C for 3 h. The column oven temperature was programmed to increase from 30°C to 210°C at a rate of 4°C/min. All analyses were run in duplicate.

Analytical pyrolysis-gas chromatography-mass spectrometry (Py-GC-MS)

Each sample (1 mg ± 0.1 mg) was analyzed by double shot pyrolysis-gas chromatography-mass spectrometry (Py-GC-MS) on a Shimadzu GCMS-QP5050A mass spectrometer equipped with a Frontier Lab double-shot pyrolyser PY-2020D (Fukushima, Japan). Separation of compounds was achieved on a fused silica capillary column, CP-Sil 8 CB (50 m × 0.25 mm i.d., Chrompack, Netherlands) using helium carrier gas. The temperatures of the injection port and separator were 280°C. Double-shot pyrograms were obtained using a two-step temperature program. For the first shot, the temperature of the pyrolyzer was maintained at 50°C for 1 min, and then raised to 250°C at a rate of 8°C/min and maintained for 1 min. The programming of the column oven temperature for GC-MS was synchronized with the temperature program of the pyrolyzer. The column oven temperature for GC-MS was first maintained at 50°C for 1 min, and then raised to 280°C at a rate of 5°C/min and maintained for 10 min. After the first shot, the column oven was cooled to 50°C. Pyrolysis was carried out by dropping the sample holder into a pyrolysis port that had been heated to 500°C. Pyrolysis was conducted for 1 min. Pyrograms were obtained by GC-MS with the same temperature program described for the analysis of volatile compounds. All analyses were run in duplicate.

Results and discussion

Chemical composition of binderless boards

Effects of steam pressure on the chemical composition of kenaf core binderless boards are shown in Fig. 1. Amount of the alcohol–benzene and hot-water extractives increased with increasing the steam pressure. Hot pressing treatment showed no significant effects on the amounts of extractives. It has been reported that steam plays an important role in chemical reactions such as the formation of acetic acid and hydrolytic cleavage of glycosidic bonds in polysaccharide chains. When boards were treated by steaming at 1 MPa or by hot pressing for 20 min, it was found that the amount of hot-water extractives from the steam-pressed boards increased to 8.2%, while those of the hot-pressed boards remained at around 2.5%. In the steam treatments, the amounts of alchohol–benzene and hot-water extractives increased with increasing pressing time, as shown in Fig. 2. Hsu et al.[8] reported that the amount of water-soluble materials extracted from aspen and lodgepole pine after steam pretreatments at 1.55 MPa for 4 min increased from 2.05% to 16.8% and 14.8%, respectively, based on the dry weight of the original material.

Figures 1 and 2 showed that hemicelluloses, Klason lignin, acid-soluble lignin, and α-cellulose also decreased with increasing steam pressure and pressing time. Lignin was more resistant to the steam treatments than hemicelluloses but some components of lignin were gradually degraded or

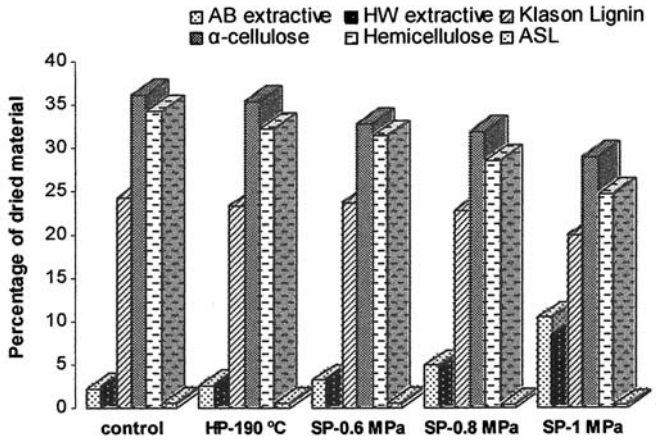

Fig. 1. The effect of steam pressure on chemical composition of binderless boards. Treatment time was 20 min, target density of binderless boards was 0.55 g/cm^3. *AB*, alcohol–benzene; *HW*, hot water; *ASL*, acid soluble lignin; *HP*, hot-pressed board; *SP*, steam-pressed board

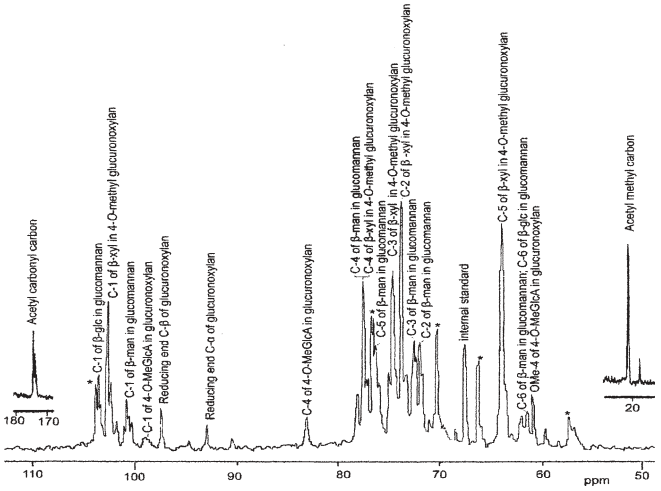

Fig. 3. ^{13}C-NMR spectrum of water-soluble polysaccharides from steam-pressed kenaf core binderless board treated at 1 MPa for 20 min. The spectrum was obtained at 100 MHz at 27°C in D_2O solutions. *Asterisk*, unidentified components

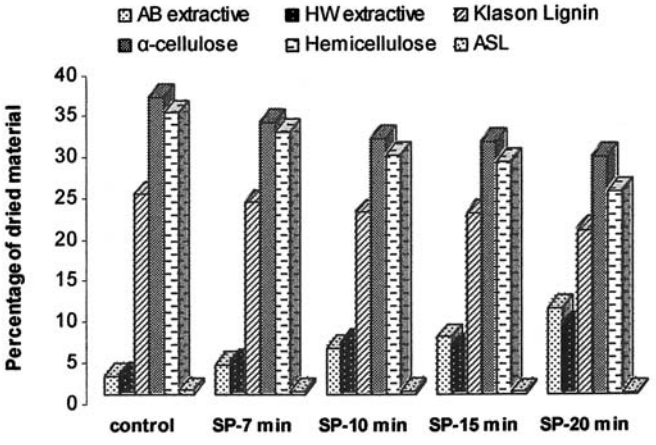

Fig. 2. The effect of pressing time on chemical composition of binderless boards. Steam pressure was 1 MPa, target density of binderless boards was 0.55 g/cm^3

modified by steaming. All of the major chemical components of kenaf core were degraded by steaming and contributed to self-bonding. The effect of steam treatment was more marked than that of hot pressing on the changes of chemical composition of the boards.

Hsu et al.[8] found that under steam pretreatment at 1.55 MPa for 1–4 min, only the hemicelluloses of hardwood and softwood decreased, while lignin and cellulose content did not decrease significantly with increasing treatment time. Okamoto et al.[6] used steam pressing in a pressure range of 0.6–1.1 MPa for 5 min to produce dimensionally stable MDF. Under these conditions, hemicelluloses and cellulose decreased with increasing steam treatment, while the lignin component did not change significantly.

The color of the steam-treated binderless boards was dark brown and became darker with increasing steam pressure and pressing time. This indicates that a high degree of hydrolysis or modification of the chemical components occurred during steam treatment. Hemicelluloses are hydrolyzed and increase their solubility in water during steam/heat treatment. Water-soluble components are mainly derived from the hemicellulose degradation products.[9]

Low-pressure steam treatment was found to have a large effect on the chemical composition of kenaf core binderless boards. Kenaf core contains hemicelluloses such as glucuronoxylan and glucomannan. Lignin is deposited in the cell walls in association with the hemicelluloses via different types of covalent linkages. Morrison et al.[10] reported low content of ester-linked and ether-linked hydroxycinnamic acid (*p*-coumaric acid and ferulic acid) in the bottom core of kenaf, being considered to be mostly ester-linked to some portion of cell walls.[11] Ester linkages, which are alkaline-labile linkages, are also susceptible to hydrolysis during steam/heat treatment. Arabinofuranosyl residues in glucuronoxylan also participate in the lignin–carbohydrate bonds through cinnamic acid residues, *p*-ferulic, and *p*-coumaric acid. Further study on the cross links in lignin–polysaccharide complexes of kenaf are to be carried out.

Analysis of water-soluble polysaccharide

Analysis of water-soluble polysaccharide by ^{13}C-NMR (see Fig. 3) showed that hemicelluloses of kenaf core mainly consist of 4-*O*-methyl-glucuronoxylan and a small amount of glucomannan, as reported by Neto et al.[12] and Ohtani et al.[13] The presence of glucuronoxylan in kenaf has also been confirmed by Das et al.[14] A part of the glucuronic acids attached to the xylan backbone are linked with lignin by ester bonds.[14]

The neutral sugar composition of water-soluble polysaccharide from kenaf core and their binderless boards is shown in Table 1. Hemicelluloses are hydrolyzed and increase their solubility in water, while cellulose was resistant to hydrolysis during steam treatment. The results demon-

Table 1. Neutral sugar composition of water-soluble polysaccharide

	Rhamnose	Arabinose	Xylose	Mannose	Galactose	Glucose
Control	0.02	0.01	0.03	0.04	0.02	0.09
HP (190°C)	0.03	0.04	0.05	0.05	0.02	0.08
SP-0.6 MPa	0.04	0.02	0.14	0.03	0.03	0.24
SP-0.8 MPa	0.09	0.04	0.99	0.10	0.10	0.29
SP-1.0 MPa	0.05	0.03	2.98	0.12	0.12	0.30

Sugar composition as percentage based on dry weight of material
HP, hot pressed; SP, steam-injection pressed

Table 2. Syringyl/guaiacyl (S/G) ratio of the boards by pyrolysis gas chromatography-mass spectrometry

	Control	HP 190°C 20 min	SP 0.6 MPa 20 min	SP 0.8 MPa 20 min	SP 1 MPa 7 min	SP 1 MPa 10 min	SP 1 MPa 15 min	SP 1 MPa 20 min
G1: Guaiacol	5.6	5.1	6.7	6.9	6.6	6.9	7.1	7.8
G2: 4-Methylguaiacol	4.1	4.2	3.9	3.9	4.6	4.5	4.9	5.2
G3: 4-Ethylguaiacol	0.7	1.6	2.2	4.3	1.4	6.7	6.8	4.9
G4: 4-Vinylguaiacol	9.6	8.7	8.6	8.2	9.6	8.4	8.8	8.9
G5: Eugenol	1.9	1.8	1.7	1.9	1.9	1.2	1.4	1.2
G6: Vanillin	1.3	2.0	2.2	1.7	1.8	1.9	1.7	1.3
G7: Isoeugenol	5.3	4.9	4.7	4.2	5.0	3.5	4.0	4.2
G8: 4-Propylguaiacol	1.2	1.4	1.1	1.0	1.1	1.0	1.0	0.9
G9: Acetoguaiacone	1.5	1.5	1.2	1.1	1.1	1.3	1.0	1.1
G10: Guaiacylacetone	1.3	1.2	1.7	1.5	1.8	1.4	1.3	1.1
Guaiacyl total	32.5	32.4	33.9	34.7	34.7	36.6	37.9	36.6
S1: Syringol	10.8	10.6	10.8	11.5	10.6	11.3	11.9	13.2
S2: 4-Methylsyringol	8.8	9.3	9.2	9.2	8.8	9.5	9.3	10.0
S3: 4-Ethylsyringol	1.6	1.8	1.7	1.6	1.5	1.7	1.6	1.7
S4: 4-Vinylsyringol	19.8	17.8	17.5	17.1	17.7	16.5	14.5	13.4
S5: 4-Allylsyringol	3.4	3.2	3.4	3.5	3.3	2.2	2.9	4.2
S6: 4-Propenyl-syringol (*cis*)	2.4	2.4	2.1	1.8	2.1	2.0	2.4	2.2
S7: Syringaldehyde	3.7	3.6	3.4	2.8	3.4	3.2	3.1	2.9
S8: 4-Propenyl-syringol (*trans*)	10.6	11.8	12.0	12.1	11.9	11.6	11.4	10.6
S9: Acetosyringone	2.3	2.5	2.2	2.1	2.1	2.0	1.7	1.9
S10: Syringylacetone	2.4	2.6	2.5	2.7	2.3	2.5	2.3	2.8
S11: Sinapaldehyde	1.6	2.1	1.3	1.1	1.6	0.7	1.0	0.4
Syringyl total	67.5	67.6	66.1	65.3	65.3	63.4	62.1	63.4
Ratio S/G	2.1	2.1	2.0	1.9	1.9	1.7	1.6	1.7

Results are given as relative percentage of total peak areas of pyrolysis products from lignin

strate that xylose content increased with increasing steam pressure. The amount of xylose in the hot-water extract of the kenaf boards after steam treatment at 1 MPa for 20 min was 3.0%, based on the dry weight of the original kenaf boards, while that obtained after hot pressing at 190°C for 20 min was 0.05%. Thus, marked differences were found in the amount of soluble xylan between the steam and hot-press treatments.

Hsu et al.[8] reported that xylose content in the water-soluble extractives from aspen treated with steam at 1.55 MPa for 4 min was 0.35%. Compared with this result, the glucuronoxylan in kenaf appears more susceptible to hydroysis by steam than aspen wood. The differences in the effects of solubilization of xylose are explained by the network of lignin–glucuronoxylan complexes involving *p*-cinnamic acid bridges in kenaf.

Pyrolysis GC-MS analysis

The pyrogram of kenaf core is shown in Fig. 4, and the syringyl–guaiacyl (S/G) ratios are shown in Table 2. Kenaf core is composed of lignin with guaiacyl, syringyl, and *p*-hydroxyl nuclei. The S/G ratio from this experiment was calculated to be 2.1 for the kenaf core. This is consistent with the values of previous reports of 1.2–2.2 for the S/G ratio for kenaf core.[10,13,15,16]

The S/G ratio decreased with increasing pressure of steam treatment. The S/G ratios of binderless boards treated by steaming at 0.6, 0.8, and 1 MPa for 20 min were 2.0, 1.9, and 1.7, respectively. This indicates that substructures containing S units are preferentially decomposed by steam treatment over those containing G units due to the more highly condensed structures of the G units than those of the S units.

In contrast to the effects of steam treatment, a S/G ratio in the hot-pressed kenaf boards was found to be close to that of original kenaf core, demonstrating that chemical modification of lignin by hot pressing is not as marked as that observed in steam treatment, although a slight decrease in Klason lignin by hot pressing was found. In the manufacture of binderless boards from oil palm fronds, decreasing S/G ratio was observed in both steam explosion and hot pressing.[3]

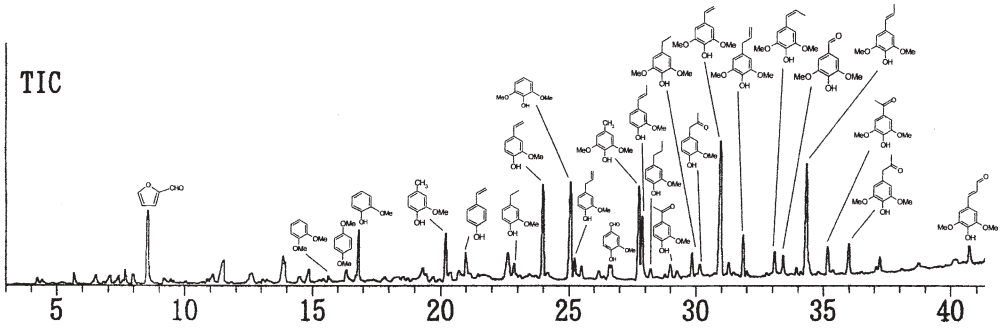

Fig. 4. Products of kenaf core pyrolyzed at 500°C for 1 min

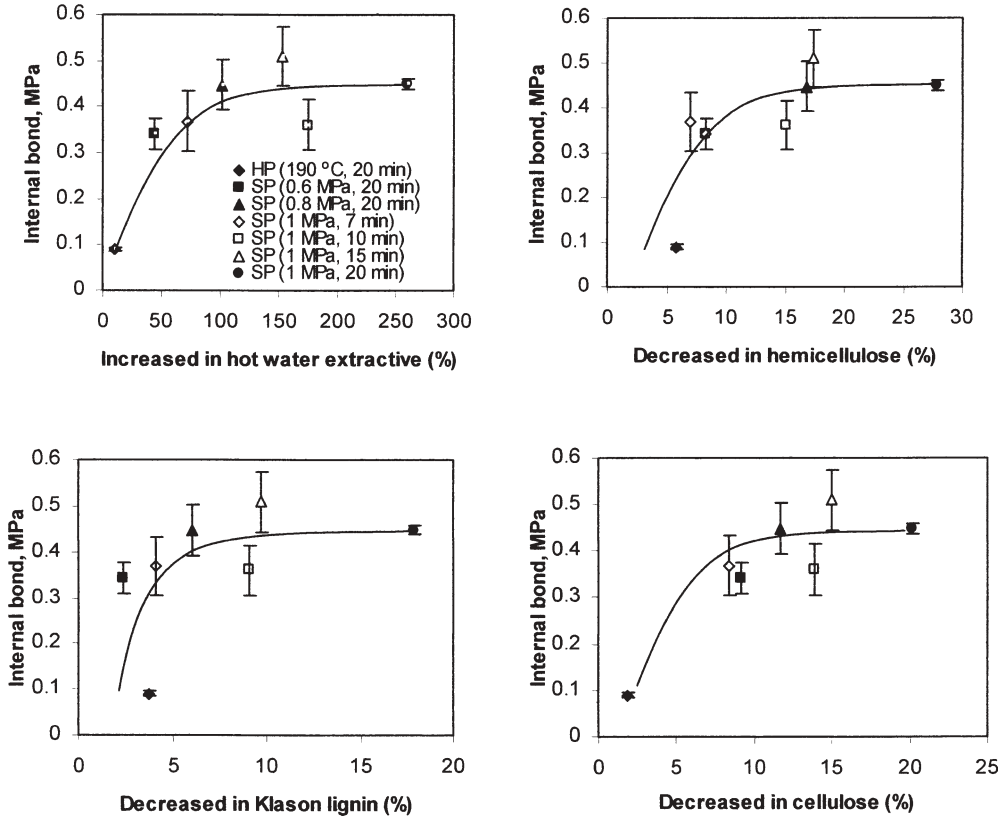

Fig. 5. The effect of chemical changes by steam/heat treatment on internal bond strength of binderless boards. The target density of binderless boards was 0.55 g/cm^3

Bonding strength and thickness swelling of binderless boards

Because resin was not used in the binderless boards, self-bonding strength was significantly affected by the changes in chemical composition of the boards caused by the steam/heat treatments. Correlation between chemical composition and IB strength of the binderless boards is shown in Fig. 5. IB strength increased with increased amount of hot-water extract and reached a plateau when the amount of hot water extract reached twice that of the untreated boards. Similar correlation was found between the IB strength and the extent of hemicelluloses, lignin, and cellulose degradation. Degradation of the three chemical components by steam pressing increased thermoplastisity, leading to deformation to a stable matrix.

In this study, low steam pressure (0.6–1.0 MPa) was applied. Compared with other binderless boards, the kenaf core binderless particleboards showed higher bondability. Cellulose gives mechanical strength to plant tissues while lignin provides rigidity and stiffness. Therefore, it is known that intensive degradation of hemicelluloses, lignin, and cellulose decreases the physical quality of the boards. Suzuki et al.[3] reported that steam explosion of oil palm frond fiber at 3 MPa produced boards of poor quality.

It should be noted that hot pressing with the lowest degradation of cell wall components gave the lowest IB strength of the board (about 0.09 MPa). Differences in the manufacturing process show a significant effect on the bonding properties. In hot pressing, steam is generated from moisture in kenaf core particles during processing. By injecting high-pressure steam, the temperature of binderless boards will rise immediately and the chemical components degrade, polymerize, and arrange a stable network in a short time during the treatment. This effect was not obtained by hot pressing, which degraded the chemical

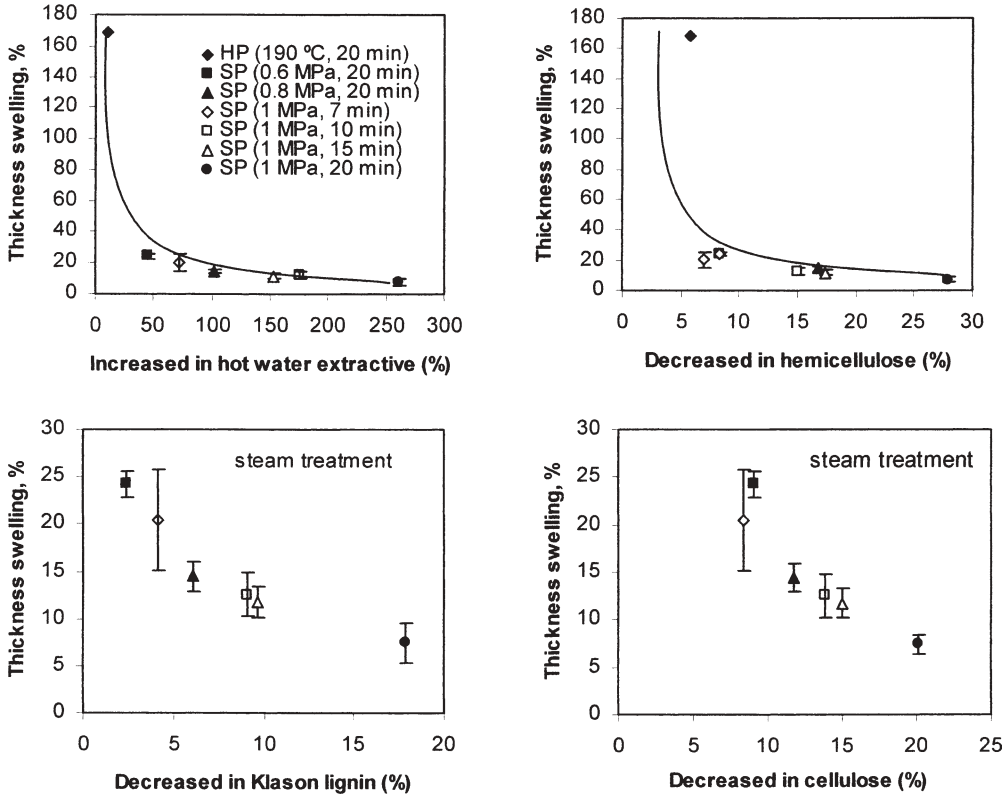

Fig. 6. The effect of chemical changes by steam/heat treatment on thickness swelling of binderless boards. The target density of binderless boards was 0.55 g/cm^3

components of kenaf much less effectively. The extent of degradation was not only dependent on the moisture content, but also on the temperature and time of treatment. Based on some research,[3,4,7] to acquire high quality binderless board, high temperature and high compactness are also needed.

In resin-bonded boards, the TS value depends on the potential thickness recovery of densified particles and breakage of the adhesive bond network.[17] Because no binder is used in the boards, the TS value will also depend on the chemical behaviors of particles during treatment.

Sekino et al.[17] reported that reduction in hygroscopicity, which because of the changes in hemicelluloses, is one main cause of improved dimensional stability. The same results are shown in Fig. 6. However, the reduction of the TS values of binderless boards is also strongly related to the decrease of lignin and cellulose contents of boards during steam treatment. These facts show that the degradation of chemical components by the steam/heat treatment to a certain degree increases the compressibility of boards and reduces the internal stress induced in each particle.

Steam treatment can improve the dimensional stability of boards. In the present study, the TS decreased significantly by using steam-injection pressing. With a steam-pressing pressure of 1 MPa, the mean TS value was 7.47% for a 20-min treatment, whereas the mean TS value of hot-pressed boards at 190°C was 169%.

An optimum chemical change has not yet been achieved. The chemical structure of the raw material also influenced the optimum conditions for the degradation of the chemical components. Further study on the bonding characterization of kenaf core is still being carried out.

Conclusions

In the manufacturing of kenaf core binderless boards by steam-injection pressing, not only hemicelluloses, but lignin and α-cellulose also contributed to self-bonding. Based on dry weight of original material, the decrease of hemicellulose (up to about 28%), α-cellulose (up to about 20%), lignin (up to about 18%), and increase of water-soluble fractions (up to about 260%) by steam-injection pressing (0.6–1 MPa) can produce kenaf core binderless boards of relatively good quality. Steam-injection pressing was an effective method to improve the bonding performance and dimensional stability of binderless boards. Hot pressing also caused degradation of chemical components, but not to a significant degree to improve their binding ability.

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