THE EFFECT OF HEAT TREATMENT ON THE CHEMICAL AND COLOR CHANGE OF BLACK LOCUST (*ROBINIA PSEUDOACACIA*) WOOD FLOUR

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The aim of this study was to investigate the effects of oxygen and moisture content (MC) on the chemical and color changes of black locust (*Robinia pseudoacacia*) wood during heat treatment. The wood flour was conditioned to different initial MCs and heated for 24 h at a constant temperature of 120ºC in either oxygen or nitrogen atmosphere. The pH values and chromaticity indexes were examined. Diffuse reflectance UV-Vis (DRUV) and Fourier transform infrared (FTIR) spectra were used to characterize the changes of chromophores upon heating. The study demonstrated that the pH values decreased after heat treatment, and it was lower when the heat treated was in oxygen than in nitrogen. The L^* decreased significantly, while a* and b* increased. The total color difference ΔE^* increased with increasing initial MC until a plateau was reached after 30% MC. The color change was greater in oxygen than in nitrogen. The hydroxyl groups decreased after heat treatment. The releases of acid and formation of quinoid compounds and carboxylic groups during heat treatment were confirmed. Discoloration of wood is due mainly to the condensation and oxidation reactions, which are accelerated by oxygen. Higher MCs are required to obtain the greatest color change of wood in inert atmosphere.

Keywords: Heat treatment; Chemical changes; Discoloration; Wood; DRUV; FTIR-ATR

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INTRODUCTION

Wood's color is an important surface attribute. It is determined by the spectral composition of light reflected from surfaces and influencing people's perceptions. In some cases consumers select a species of wood based solely on its decorative properties. Enhancing the appearance of less attractive wood without chemical additives is of great interest to consumers and researchers. Wood drying is a necessary step for the manufacture of most wood product in the wood processing industry. In the wood drying process, the exposure of wood to high temperatures and humidity is a method being considered to permanently change both the appearance and chemical properties of wood (Nuopponen et al. 2004; Sundqvist 2004; Sundqvist and Morén 2002). In some cases, heat treatment can add value to wood by imitating the appearance of tropical hardwoods that are valued in many countries (Syrjänen 2001; Militz 2002; Patzelt et al. 2003; Bekhta and Niemz 2003; Christmas et al. 2005). It is important to understand the fundamentals of the kiln drying

process to achieve a controlled and desired color change in wood.

The color of a solid material is attributed to the reflection, scattering, and absorption of light within the visible range. The absorption of visible light is a characteristic of the material, and it is caused by certain molecules called chromophores (Hon and Shiraishi 1991). In wood, light is mainly absorbed by lignin below 500 nm and by phenolic extractives, such as tannins, flavanoids, stilbenes, and quinones above 500 nm (Hon and Shiraishi 1991). The discoloration reactions that occur during heat treatment are often justified by the formation of colored oxidation and degradation products involving the cell wall constituents and extractives located in cell vacuoles (Sehistedt-Persson 2003; Sundqvist 2004). In lignin, there is a cleavage of β-O-4 linkages, resulting in a higher concentration of phenolic groups (Runkel 1951; Kollmann and Fengel 1965) and a reduction of methoxyl content, leading to a more condensed structure by autocondensation of lignin (Wikberg and Maunu 2004). Changes in phenolic extractives have been identified as another potential cause for the discoloration of wood (Hiltunen et al. 2006); however, since the content of wood extractives varies with the species, so may the discoloration mechanisms. Discolorations of European oaks (*Quercus robur*) and sugar maple (*Acer saccharum*) have been linked to polymerisation or oxidation of phenolic extractives during kiln drying (Charrier et al. 1995; Miller et al. 1990). Temperature and oxygen evoke polymerisation of ellagitannins, while vacuum drying effectively prevents discoloration due to lack of oxygen (Charrier et al. 1995).

It has been reported that the presence of oxygen, moisture, and elevated temperatures combined are important factors in darkening during heat treatment. Oxidative and hydrolytic reactions are primarily considered to be the cause for the production of chromophores during thermal treatment of wood, where hydrolytic reactions are generally the dominant process when moisture is present (Fengel and Wegener 1989). Maillard reactions between reducing sugars and nitrogen compounds occur without enzymes, but they are accelerated at high temperatures and pH (McDonald et al. 2000; Rizzi 1994). It is known that during thermal treatment of wood under moist conditions, carbonic acids, mainly acetic acid, will initially be formed as a result of the cleavage of the acetyl groups of particular hemicelluloses (Kollmann and Fengel 1965; Dietrichs et al. 1978; Bourgois and Guyonnet 1988). Research into this phenomenon reveals that wood darkening caused by high temperatures varies from one species to another and is a function of moisture content (MC). Sullivan (1967) found that maples were manifested by a nearly linear relationship between darkening and MC, where increased temperature primarily increased the rate of darkening. Sawn wood of silver birch (*B. pendula*) easily darkened and reddened during vacuum drying if high temperatures were employed at the beginning of drying when the moisture content of the wood was over 25% (Lahtinen and Tolonen 2001).

Black locust is a fast-growing wood species that can be used for outdoor and indoor applications due to its hardness and durability. It can be easily discolored during drying due to its high extractive content, specifically polar extractives (Fan et al. 2010). The aim of the present work is to study the effect of oxygen and moisture content on the chemical and color changes in black locust (*Robinia pseudoacacia*) wood during heat treatment. To develop a more understandable relationship between discoloration and chemical changes in whole wood upon heating, black locust wood flour was used instead

of timber in this study. Samples with different MCs were subjected to heat treatment for 24 hours at a constant temperature of 120 ºC in an atmosphere of either oxygen or nitrogen. Chromaticity index, pH value, DRUV, and FTIR-ATR spectra were used to monitor color and chemical changes after heat treatment. The conditions for heat treatment employed in this study were selected to coordinate with the industrial wood drying process in order to provide important information on the control of wood discoloration during drying.

EXPERIMENTAL

Wood Sample Preparation

Black locust (*Robinia pseudoacacia*) logs were harvested from Heilongjiang province in northeast China. The logs were debarked and dried in ambient conditions, then ground with a Wiley mill and screened. The 40 to 60 mesh range of wood flour was selected for testing and stored in a polyethylene bag at room temperature and 13% RH until required for further experimentation.

Heat Treatment

The wood flour was conditioned with distilled water to obtain moisture contents of 0, 10, 30, 50, and 70% before being subjected to heat treatment at 120 ºC for 24 h. The initial MC was ensured by three steps: measuring the water contents, adding distilled water to the wood flour, and well mixing according to the scheduled MC. The equilibrium was accomplished in a sealed plastic bag for 24 h.

The heat treatment was carried out in a sealed stainless steel autoclave with a Teflon vessel in either oxygen or nitrogen atmospheric conditions in an electrical oven. The heat-treated samples were cooled and vacuum-dried for more than 24 hours at room temperature in the dark to ensure the MC to be less than 5% and avoid the influence of light before further analysis. Three replicates of each formulation were performed.

Extraction with Water and Determination of pH Value

After heat treatment, wood flour samples were immersed in distilled water for 48 h at room temperature, and the pH values were determined using an E-201-C acidometer (Shanghai Leiyun Corporation). The pH values were measured using three replicates of each sample, and average value was reported.

Measurement of Color Parameters

The wood flour was pressed into pellets with a die of 30 mm in diameter, and the color parameters were recorded with DF110 colorimeter (Guangzhou ABC Co. Ltd) using a D65 standard illuminant and 10° standard observer. The aperture was 18 mm in diameter. Color parameters were measured using three replicates and three times of each sample. The average value was reported.

The color parameters L^*, a^*, and b^* were determined for untreated and treated wood by the CIELAB system, in which L^* denotes lightness from 0% (black) to 100% (white), a^* denotes from green (-a) to red (+a), and b^* denotes from blue (-b) to yellow (+b). The

ΔL^*, Δa^*, and Δb^* were calculated using the unheated sample as a reference (e.g. $\Delta L^* = L^*_{treated} - L^*_{ref}$), and their corresponding total color difference (ΔE^*) was calculated using the following formula:

$$\Delta E^* = (\Delta L^{*2} + \Delta a^{*2} + \Delta b^{*2})^{1/2} \tag{1}$$

Diffuse Reflectance UV-Vis Spectra (DRUV)

The diffuse reflectance spectra from control and heat-treated wood samples were recorded at room temperature on a UV-3100 UV-Vis near-IR spectrophotometer (Shimadzu, Japan) equipped with an integrating sphere. The reflectance spectra were recorded against $BaSO_4$ as a white (R_∞) optical standard. The wood flour samples were pressed into pellets. The study was carried out over the wavelength range 240 to 800 nm. The reflectance spectra of wood samples were converted into *K/S* spectra using the Kubelka-Munk equation:

$$K/S = \frac{(1-R)^2}{2R} \tag{2}$$

where R is the measured reflectance and K and S are the absorption and scattering coefficients, respectively. Difference spectra were calculated by subtracting the reflectance spectrum of original sample from one of the heat-treated samples (Eq. 3), and plotted as a function of wavelength to identify the apparent absorption maxima. Therefore, the difference spectrum represents a net change in brightness reversion. Typically, the reflectance (R) values were in the range of 15 to 90%, and based on the Kubelka–Munk theory, we can assume that $\Delta(K/S)$ is approximately linear to *delta* chromophores in this range of reflectance values.

$$\Delta(K/S) = (K/S)_{after} - (K/S)_{before} \tag{3}$$

FTIR-ATR Spectra

FTIR-ATR spectra were recorded on a Tensor 27 spectrophotometer (Brüker Corporation) with an ATR unit using Brüker OPUS software to provide detailed information about the functional group presented at the surface of the samples. Spectra were measured at a resolution of 2 cm^{-1} and 32 scans. The peaks were normalized at the maximum absorption band at 1,050 cm^{-1} as the internal standard (Windeisen and Wegener 2008; Rosu et al. 2010), which corresponds to the C-O stretching vibrations in cellulose and changes least upon heating.

RESULTS AND DISCUSSION

Variation of Wood pH Values after Heat Treatment

Table 1 shows the pH values of wood flour obtained after being conditioned at various MCs followed by heat treatment in either oxygen or nitrogen atmosphere. The pH of the control wood flour (before heat treatment) was 6.1, and that of the wood flour after

heat treatment in oxygen and nitrogen decreased significantly to 3.9 and 4.4, respectively, when the initial MC was 0%. The decrease in pH can be explained by the production of organic acids during heat treatment, mainly by the degradation of hemicellulose, in which acetyl groups (at carbon 2 or 3 of the glucuronoxylan backbone) split off (Theander and Nelson 1988; Nuopponen et al. 2004). The phenolic carboxylic acid and 4-O-methyl-glucuronic and galacturonic acids produced as a result of the hydrolysis of wood also contribute to wood acidity (Windeisen et al. 2007). Heat treatment in oxygen led to lower pH values than in nitrogen, which suggests that the presence of oxygen enhanced acid production. It can be observed that increasing MC had little effect on pH.

Table 1. pH Values of Wood Flour Heat Treated at Different Moisture Contents in either O_2 or N_2 Atmosphere

	0% MC [a]	30% MC	70% MC
Control	6.1	—	—
Heat treated in O_2	3.9	3.7	4.3
Heat treated in N_2	4.4	4.8	4.9

[a] Vacuum dried for 7 days at room temperature over phosphorus pentoxide

Variation of Color Parameters before and after Heat Treatment

Color parameters of wood flour after heat treatment in oxygen and nitrogen can be seen in Tables 2 and 3, respectively. The black locust wood flour changed its color characteristics in terms of L^*, a^*, and b^* values when subjected to heat treatment. The initial L^* value for control was 66.9. Heat treatment induced a significant decrease in L^* values, measured at 42.8 (in oxygen) and 54.2 (in nitrogen), when the initial MC was 0%. The a^* values increased while b^* values decreased after heat treatment.

Table 2. Color Parameters L^*, a^*, b^* of Wood Flour Heat Treated at Different initial MCs in Oxygen Atmosphere

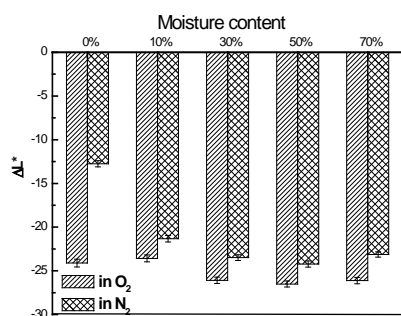|	Control	0%	10%	30%	50%	70%
L^*	66.9 ($\pm$0.4)[a]	42.8 ($\pm$0.5)	43.4 ($\pm$0.4)	40.9 ($\pm$0.4)	40.4 ($\pm$0.3)	40.8 ($\pm$0.4)
a^*	5.0 ($\pm$0.2)	8.4 ($\pm$0.2)	8.6 ($\pm$0.2)	8.7 ($\pm$0.2)	8.1 ($\pm$0.2)	7.5 ($\pm$0.2)
b^*	23.3 ($\pm$0.3)	16.9 ($\pm$0.3)	18.5 ($\pm$0.3)	17.6 ($\pm$0.3)	16.4 ($\pm$0.3)	15.0 ($\pm$0.3)

[a] Numbers in parentheses represent the standard deviation of five replicates.

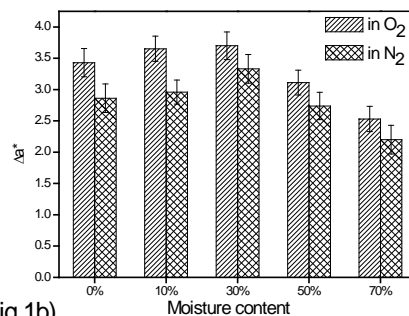**Table 3.** Color Parameters L^*, a^*, b^* of Wood Flour Heat Treated at Different initial MCs in Nitrogen Atmosphere

	Control	0%	10%	30%	50%	70%
L^*	66.9 ($\pm$0.4)[a]	54.2 ($\pm$0.4)	45.6 ($\pm$0.4)	44.0 ($\pm$0.3)	42.7 ($\pm$0.4)	43.8 ($\pm$0.3)
a^*	5.0 ($\pm$0.2)	7.9 ($\pm$0.2)	8.0 ($\pm$0.2)	8.3 ($\pm$0.2)	7.7 ($\pm$0.2)	7.2 ($\pm$0.2)
b^*	23.3 ($\pm$0.3)	18.4 ($\pm$0.2)	16.9 ($\pm$0.3)	16.6 ($\pm$0.2)	15. 5 ($\pm$0.2)	14.3 ($\pm$0.2)

[a] Numbers in parentheses represent the standard deviation of five replicates.

The change in color parameters ΔL^*, Δa^*, Δb^*, and ΔE^* of wood flour with various initial MCs heat treated in either oxygen or nitrogen atmosphere are shown in Fig. 1.
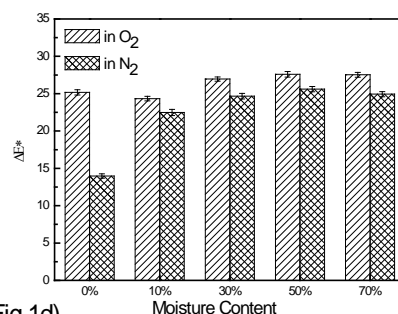
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(Fig.1a)

(Fig.1b)

(Fig.1c)

(Fig.1d)

Fig. 1. Changes in color parameters for wood flour heat treated with different initial MCs in either oxygen or nitrogen atmosphere: (a) ΔL^*, (b) Δa^*, (c) Δb^*, (d) ΔE^*

A decrease in brightness (i.e. a decrease in L^*) can be seen after heat treatment in oxygen and nitrogen, and a more rapid decrease was observed when the heat treatment took place in an oxygen atmosphere. The greatest difference between the two atmospheres was observed when the initial MC of the wood flour was 0%; the decrease in ΔL^* after heat treatment in oxygen was almost two times the decrease observed after heat treatment in nitrogen (Fig. 1a). This suggests that oxygen as an oxidation medium plays a vital role in the darkening of wood during heat treatment. The ΔL^* values of all the samples increased slightly with the increasing MC up to 50%, followed by a slight decrease at 70% MC.

Heat treatment in both atmospheres resulted in an increase in redness (i.e. increase in a^*). The Δa^* increased as initial MC increased from 0% to 30%, then decreased as initial MC increased from 30% to 70% (Fig. 1b). The variation was remarkably greater in oxygen than that in nitrogen. These observations imply the formation of condensation products and degradation and/or oxidation products. It is well known that the proanthocyanidin, lignin and other related extractives can be readily condensed under low pH on heat, and the byproducts thus formed absorb the complementary light of reddish color and contribute to the red color of wood.
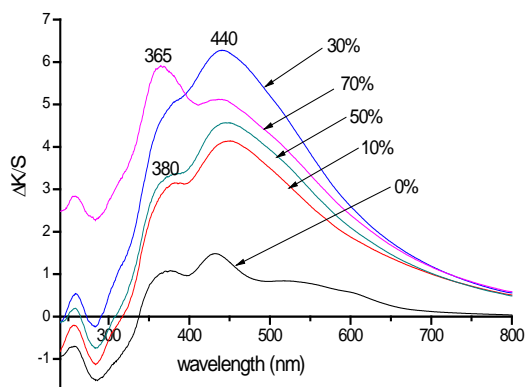
In both atmospheric conditions, all the heat treated wood flour experienced a shift from yellow toward blue (i.e. decrease in b^*). The Δb^* generally decreased as initial MC increased (Fig. 1c). There was a considerable difference in Δb^* variation between the samples treated in oxygen and nitrogen, as well as between different initial MCs. The Δb^* value was much higher for samples treated in oxygen than that in nitrogen when the

initial MC was 0%, which demonstrates that oxygen plays an important role in color shift toward blue during heat treatment. In the presence of water, combining high temperature and oxygen atmosphere, heating generally induces hydrolysis reactions in wood, which could result in the formation of lower molecular weight yellow phenolic substances, such as flavonoids. Higher MCs in the inner environment of wood result in more yellow color.
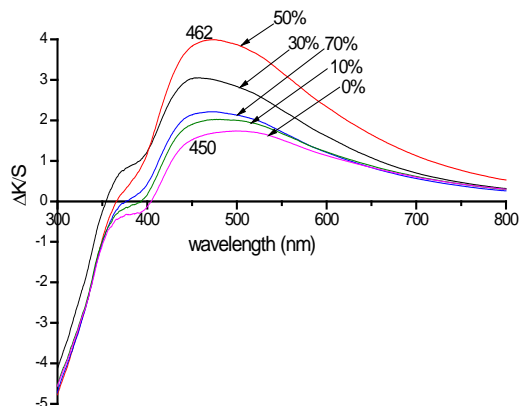
The total color difference (ΔE^*) of heat-treated wood is suggested to be an indicator of its degree of overall chemical modification. It can be seen that the total color change was larger in the case in oxygen compared to that in nitrogen (Fig. 1d). Total color change showed a slight increase with increasing initial MC from 0% to 30%, with little change in color as the initial MC increased beyond that. It can be found that oxygen is more critical than nitrogen in relation to discoloration. The color changes can be due to the formation of secondary condensation products and/or degradation products, quinoid substances, for instance, which are known to be intensely colored. The reactive compounds can include degradation products from the cleavage of α- and β-aryl ether bonds in lignin and degradation products from hemicelluloses. These may produce various colored condensation products (Lai 1991). Hemicelluloses are hydrolyzed in the presence of moisture to produce colored degradation products. Such changes also result in the increase in ΔE^* (Sehistedt-Persson 2003; Sundqvist 2004) relative to untreated wood.

UV-Vis Diffuse Reflectance Spectra Analysis

UV-Vis reflectance spectroscopy provides a tool by which to characterize the wood chromophores and changes in their concentration correlating to the discoloration of wood during heat treatment. The difference absorption spectra ($\Delta K/S$), shown in Fig. 2, can represent a net change due to heat treatment. The darkening of the wood color resulted in increasing absorption at longer wavelengths. The apparent absorption change after heat treatment suggested the generation of some types of chromophores in the degradation, condensation and oxidation progresses. However, it is very difficult to differentiate structural differences between these chromophores.

(Fig.2a)   (Fig.2b)

Fig. 2. Absorption spectra $\Delta K/S$ of wood flour heat treated at different initial MCs in: (a) oxygen and (b) nitrogen

It is apparent that there was an absorption change at longer wavelengths between 350 and 650 nm, and it was more obvious for samples heat treated in oxygen than in nitrogen. This indicates that more chromophores, which contribute to the dark color of the heat-treated wood, are generated during heat treatment in oxygen through oxidation reactions. The fact that more chromophores were generated in oxygen was also supported by the observation that there were higher ΔL^* and Δa^* values of samples treated in oxygen compared with nitrogen at the same initial MC.

Heat treatment in oxygen appeared to significantly enhance the formation of chromophores in lignin and extractives, contributing to the 350-380 nm absorption. This absorbance has been frequently attributed to the presence of aromatic ketones, conifer-aldehydes, and/or α-,β-unsaturated ketone structures (Polcin and Rapson 1971; Ternay 1979). It appears from this study that the formation of these groups is especially sensitive to oxygen. The presence of oxygen also resulted in an absorbance increase at 440 nm, which can be attributed to the formation of colored quinoid compounds originating from degradation and oxidation of the aromatic hydroxyl groups of lignin and aromatic extractives upon heat treatment (Chen et al. 2010).

Moisture content is another factor that influences color differences upon heat treatment. It can be observed in Fig. 2 that the absorption increased with the presence of moisture, and similar intensity variations resulting from MC can be seen in the samples treated in nitrogen. The strongest absorption in the range 400 to 650 nm can be found from the samples treated at an initial MC of 30% and 50% in oxygen and nitrogen, respectively. This suggests that some dark colored substances were readily produced when the wood flour with an initial MC of 30% was subjected to heat treatment in oxygen and when wood flour with an initial MC of 50% was subjected to heat treatment in an inert atmosphere.
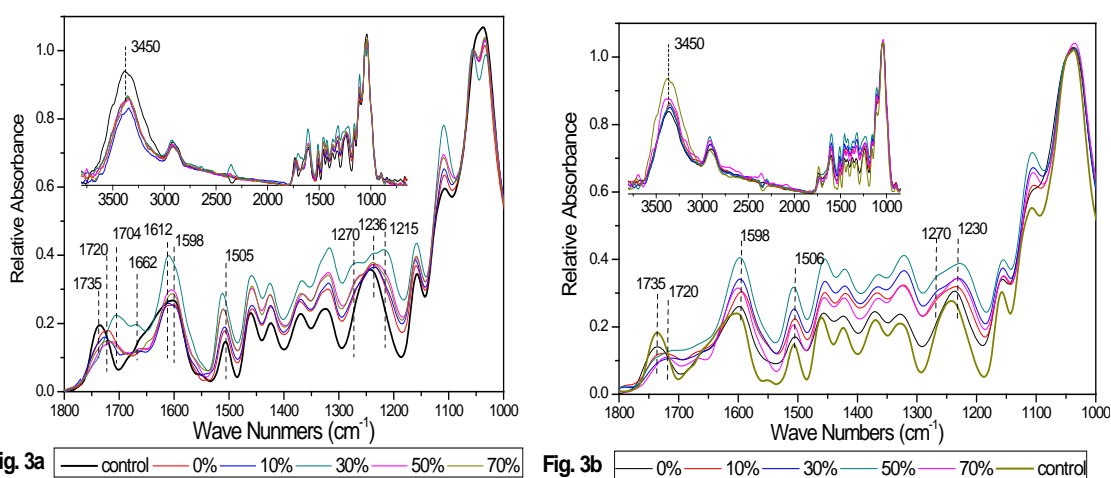
FTIR-ATR Spectra Analysis

Fig. 3. FTIR-ATR spectra (normalized at 1,050 cm^{-1}) of wood flour with different initial moisture content after heat treatment in an environment of: (a) oxygen, and, (b) nitrogen

The FTIR-ATR spectra of the samples with different initial MCs heat treated in either oxygen or nitrogen atmosphere are presented in Figs. 3a and 3b. It can be seen that the absorbance at 3450 cm^{-1} and 1735 cm^{-1}, which are assigned to the hydroxyl groups and unconjugated carboxyl groups respectively (Leschinsky et al. 2008; Pétrissans et al. 2003; Mononen et al. 2005), decreased after heat treatment in either oxygen or nitrogen atmosphere. This suggests the release of acid substances, specifically acetic acid, after degradation of acetyl groups in polyose (Tjeerdsma and Militz 2005). Such a release is consistent with the decrease in pH value after heat treatment, as stated previously. Acids can catalyze the condensation and degradation reactions in lignin structures (Lundquist 1970) and extractives after heat treatment in the presence of water and contribute to the discoloration of wood. These decreases, in combination with the new peak arising at 1720 cm^{-1}, describe the increase in esterified structures (Faix 1992).

In the case of oxygen, it can be seen that the absorption at 1505 cm^{-1} shifted to 1512 cm^{-1} gradually with the increase in MC. This is believed to be caused by the decrease in β-O-4 linkages (i.e., increasing degrees of condensation), as demonstrated by deconvolved FTIR spectra of lignin and lignin polymer models (DHPs) in this region (Faix and Beinhoff 1988). Such a process could result in the increase in phenolic hydroxyl group, and which, as an auxochrome group, could also contribute to the darkening of the heat treated wood flour.

The absorption at 1612 cm^{-1}, which can be assigned to C=O conjugated to aromatic ring (Ke and Dong 1998), rises from a plateau at 0% MC to a peak at 30% MC, just next to the signal of aromatic ring at 1598 cm^{-1} and overlies the band at 1600 cm^{-1}. This implies the formation of new conjugated structures, such as coniferaldehyde and aromatic ketones, due to oxidation and dehydration reactions. This hypothesis can be supported by the formation of formaldehyde (1704 cm^{-1}) (Lundquist et al. 1970).

As can be seen from Fig. 3a, a new peak appeared at 1662 cm^{-1} when the sample with 30% MC was heat-treated in an oxygen atmosphere. This can be attributed to the formation of quinoid structures (Keating et al. 2006) during heat treatment. This was well consistent with the results obtained from chromaticity index analysis. It can be noted that the absorption at 1662 cm^{-1} decreased with the further increase in MC. This observation implies that heat treatment at 30% MC is more effective in accelerating the oxidation and dehydration reactions. The absorption at 1270 cm^{-1} and 1720 cm^{-1} increased when samples were heat-treated at higher MC, which suggests the formation of carboxylic groups (Jokic et al. 2004) due to oxidation reactions.

Condensation should be the main class of chemical reactions during heat treatment in a nitrogen atmosphere. It can be seen from Fig. 3b that the absorptions at 1704 cm^{-1}, 1612 cm^{-1}, and 1512 cm^{-1} disappeared when samples were heat treated in nitrogen. This suggests that the appearance of these peaks rely on the presence of oxygen. Without oxygen, it seems that higher MC is needed to obtain the most discolored wood.

It has been established that there is a mixture of extractable components, such as tannins and the derivatives of robinetin and dehydrorobenetin, in black locust wood (Dünisch et al. 2010). Lignins and these extractives are subject to condensation reactions under acidic conditions (Funaoka et al. 1990; Sears and Casebier 1968) and result in a dark color of wood flour. In order to elucidate the effect of condensation on discoloration, the condensation index (CI) is calculated based on the protocol proposed by Faix (1991).

The condensation index as a function of MC after heat treatment is plotted in Fig. 4. It can be seen that, compared to the non-heated control, the CI values of heat treated wood flour were higher in either oxygen or nitrogen atmosphere. This implies the occurrence of quite serious condensation reactions in the wood flour by heating. When referring to Table 1, it also can be inferred that condensation reactions should be accelerated by low pH values.
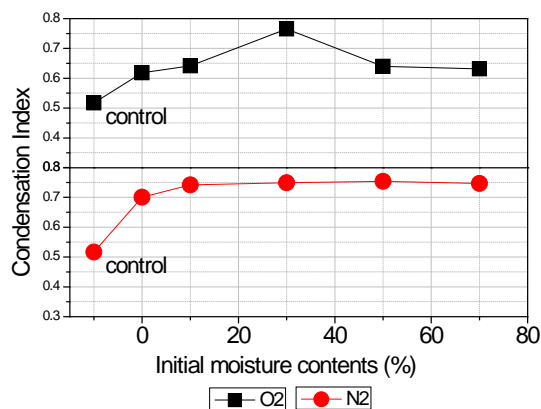

Fig. 4. The variation of condensation index as a function of moisture content of wood flour heat treated in oxygen and nitrogen atmosphere

CONCLUSIONS

1. The pH values of black locust wood flour decreased after heat treatment in either oxygen or nitrogen atmosphere due to the production of organic acids. Lower pH values are obtained upon heat treatment in an oxygen atmosphere, in comparison to an inert atmosphere.

2. Oxygen as an oxidation medium plays an important role in the darkening of wood during heat treatment. The lightness L^* decreases and the chromaticity index a^* and b^* increase during heat treatment in both atmospheres. The changes of the chromaticity are greater when heat treatment takes place in oxygen than that in nitrogen due to oxidation. The total color differences (ΔE^*) are larger in the case of oxygen compared to that in nitrogen due to the formation of condensation and oxidation products in the presence of oxygen.

3. Diffuse reflectance in the UV-visible range (DRUV) displays an intensified absorption at longer wavelengths upon heating as a result of the generation of chromophores by degradation, condensation, and oxidation progresses. Greater color change of wood flour heat-treated in oxygen atmosphere than in nitrogen indicates that more chromophores are generated in the presence of oxygen.

4. The formation of chromophores is more sensitive to oxygen. The presence of oxygen results in the formation of colored quinoid compounds by degradation and oxidation reactions of aromatic structures.

5. Moisture content is also an important contributor to the color change. The greatest chemical and color changes are obtained at an initial MC of 30% and 50% in oxygen

and nitrogen, respectively. Higher MCs are required to obtain the most color changes of wood when it is heat treated in inert gases.

6. As a result of heat treatment, hydroxyl groups decreased. Acid groups and esterified structures increased. The increase in condensation index makes a contribution to the dark color of the heat treated wood flour.

7. Quinoid structures are produced when wood is heat treated in the presence of oxygen, as a result of oxidation/dehydration reactions.

8. Discoloration of wood during heat treatment is due mainly to the condensation and oxidation reactions. Both of them lead to the formation of extensive conjugation structures.

ACKNOWLEDGMENTS

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