

# EFFECTS OF HOT OIL TREATMENT ON COLOUR AND CHEMICAL CHANGES IN 15-YEAR-OLD ACACIA HYBRID

W Razak<sup>1</sup>\*, K Izyan<sup>1</sup>, A Roziela Hanim<sup>2</sup>, S Othman<sup>3</sup>, M Aminuddin<sup>2</sup> & H Affendy<sup>2</sup>

<sup>1</sup>Faculty of Agro Industry and Natural Resources, Universiti Malaysia Kelantan, 16100 Pengkalan Chepa, Kelantan, Malaysia

<sup>2</sup>School of International Tropical Forestry, Universiti Malaysia Sabah, 88999 Kota Kinabalu, Sabah, Malaysia

<sup>3</sup>School of Industrial Technology, Universiti Sains Malaysia, 11800 Penang, Malaysia

Received January 2010

**RAZAK W, IZYAN K, ROZIELA HANIM A, OTHMAN S, AMINUDDIN M & AFFENDY H. 2011. Effects of hot oil treatment on colour and chemical changes in 15-year-old *Acacia* hybrid.** *Acacia* hybrid trees aged 15 years were harvested and cut into samples consisting of sapwood and heartwood taken from the bottom, middle and top portions of the tree. The wood samples were thermally modified using temperatures of 180, 200 and 220 °C, and treatment times of 30, 60 and 90 min. The colour changes in the sapwood and heartwood were measured using a chroma meter and the results presented according to the CIE L\*a\*b\* colour coordinate system. The results showed that temperature at a certain treatment time enhanced and darkened the treated wood. The colour of treated sapwood could be enhanced to match the colour of natural *Acacia* hybrid heartwood. For chemical analysis, wood samples were dried and ground into sawdust. Untreated samples were used as controls. The results showed that the holocellulose contents decreased between 1.0 and 11.7% for sapwood, and 2.3 and 12.8% for heartwood. The cellulose contents decreased as much as 1.9% to 20.0% for sapwood and 2.3 to 22.1% for heartwood. The hemicellulose contents increased to 4.1% for sapwood and 5.7% for heartwood. Lignin contents increased to 15.4% for sapwood and 11.6% for heartwood.

Keywords: Sapwood, heartwood, wood colour improvement, wood chemical constituents

**RAZAK W, IZYAN K, ROZIELA HANIM A, OTHMAN S, AMINUDDIN M & AFFENDY H. 2011. Kesan rawatan minyak panas terhadap perubahan warna dan perubahan kimia dalam hibrid *Acacia* berusia 15 tahun.** Pokok hibrid *Acacia* berusia 15 tahun ditebang dan dipotong kepada sampel kayu gubal dan kayu teras yang diambil daripada bahagian bawah, tengah dan atas pokok. Sampel kayu kemudiannya menjalani modifikasi haba menggunakan suhu 180 °C, 200 °C dan 220 °C serta masa rawatan 30 min, 60 min dan 90 min. Perubahan warna pada kayu gubal dan kayu teras diukur menggunakan meter warna. Keputusannya ditunjukkan berdasarkan sistem kordinat warna CIE L\*a\*b\*. Keputusan menunjukkan bahawa suhu pada tempoh rawatan tertentu menggelapkan warna kayu yang dirawat. Warna kayu gubal yang dirawat boleh dipertingkatkan supaya setanding dengan warna semula jadi kayu teras hibrid *Acacia*. Bagi analisis kimia, sampel kayu dikeringkan dan dikisar menjadi serbuk gergaji. Sampel yang tidak dirawat digunakan sebagai kawalan. Keputusan menunjukkan kandungan holocelulosa berkurangan antara 1.0% hingga 11.7% untuk kayu gubal dan antara 2.3% hingga 12.8% untuk kayu teras. Kandungan selulosa pula berkurangan sebanyak 1.9% hingga 20.0% untuk kayu gubal dan sebanyak 2.3% hingga 22.1% untuk kayu teras. Kandungan hemiselulosa meningkat ke 4.1% untuk kayu gubal dan 5.7% untuk kayu teras. Kandungan lignin meningkat kepada 15.4% untuk kayu gubal dan 11.6% untuk kayu teras.

## INTRODUCTION

*Acacia* species have become major species in forest plantations in Malaysia. The project started in 1982 and was aimed at providing a sustainable supply of timbers for the local wood-based industry. The wood-based industry is expected to face a shortage of supply from natural forests (Shakri 1995). *Acacia* was chosen as a plantation species based on its fast growth, adaptability to local soil conditions and quality of

wood. Furniture made of *Acacia* is valued for its strength, durability, aesthetic values such as grain orientation and colour, and cost.

Young *Acacia* has distinct colour differentiation between the sapwood and heartwood. The sapwood is lighter in colour than the heartwood. The colour difference with irregular colour margin reduces its value considerably. Problems will arise when matching individual pieces of

\* E-mail: drrazakw@yahoo.com

different colours into the final product. In order to remain competitive in the market, the colour appearance of the wood needs to be modified and evaluated to ensure the quality of the products (Tolvaj & Molnar 2006). The sapwood of all timber species can be enhanced using the hot oil thermal modification process (Charrier et al. 2002).

The hot oil thermal modification process seems to be suitable because it is an environmentally-friendly process. Most wood treatments use preservatives which mostly have heavy metals and discharge toxins into the environment. Many developed countries have totally banned the use of chrome-copper arsenic (CCA) (Berard et al. 2006). In recent years, increases in environmental awareness and policies which support renewable resources and environmentally-friendly chemicals have resulted in great interest in non-biocides. As such, a lot of effort has been put into developing new wood preservatives. Another environmentally-friendly technique for wood modification is the use of biodegradable substances in wood protection (Hyvonen et al. 2006).

The objective of this study was to investigate how hot oil thermal modification process enhanced the colour appearance and chemical constituents of *Acacia* hybrid wood at different temperatures and treatment times. The result of this study will improve the value of *Acacia* wood for the local wood industry.

## MATERIALS AND METHODS

### Wood material

Three 15-year-old *Acacia* hybrid logs were obtained from a forest plantation at the Sabah Forest Development Authority (SAFODA), Kinarut, Kota Kinabalu, Sabah. The logs were harvested from trees selected based on their good physical appearance, diameter at breast height ranging from 25 to 30 cm and long straight bole with minimum branches. The logs were cut and segregated into bottom, middle and top portions. Boles of 600 mm long were cut from the middle of each portion and later planed into dimensions of 320 × 100 × 25 mm (length × width × thickness). The wood samples were then placed in a conditioning chamber set at 20 ± 2 °C and 65 ± 5% relative humidity to stabilise the moisture content (MC) to 12%. The samples were conditioned for two weeks.

### Hot oil thermal modification process

The hot oil thermal modification on *Acacia* hybrid wood was performed in a stainless steel tank, complete with thermocouples and heat generator. The tank size was of dimensions 750 × 600 × 600 mm (length × width × height) and 3 mm thick. The heat of the tank was generated using electricity. Eighty samples were prepared prior to the treatment. Palm oil was used as heating medium. It has a high boiling point of 320 °C (Rafidah 2007). Treatment temperatures at 180, 200 and 220 °C were applied for 30, 60 and 90 min. After each treatment period, the samples were removed from the tank. The samples were cooled down and then conditioned again in a conditioning chamber at 20 ± 2 °C and 65 ± 5% relative humidity.

### Colour measurement of heat-treated samples

Colour measurements were taken when the wood samples had completed the hot oil thermal modification process. The surfaces on the woods to be measured were marked before the heat treatment process. This was done so that the parts of the surfaces measured were consistent throughout the process. Using pencil, the area was marked 2 × 2 cm on the cross-sectional surface of each sapwood and heartwood. This section was regarded as the most representative area for revealing the colour difference of sample (Unsal et al. 2003). Samples for colour measurement were taken at the middle of the sapwood and heartwood in the cross-sections of the *Acacia* hybrid at each portion height. The samples were sanded down slightly for about 3 mm with sandpaper and brushed clean to minimise the risk of colour variation caused by differences in surface structure. The measurements of colour were determined according to CIE L\*a\*b\* (CIE 1986) system. A colour solid is defined by three rectangular coordinates. This system is sensitive enough to detect small differences in colour parameters to determine between tree and within species variations (Thulasidas et al. 2006). The principal axis is the lightness level L\* on a 0–100 scale, 0 being total blackness and 100 pure white, while intermediate values give grey shades. The hue is specified by the other two chromic characters a\* and b\*. The character a\* is the red–green axis

with negative values reflecting the dominance of greenness and positive ones, redness. The character  $b^*$  is the yellow–blue axis, with negative values reflecting the dominance of blueness and positive ones, yellowness.

Changes in colour of the wood surfaces due to treatment were measured using a chroma meter. These measurements were only done on clear wood surfaces. The sensor head was 8 mm in diameter. The colour reader measures colour difference on the surface of wood specimens between two colours, i.e. before and after treatments. The results were presented according to the CIE  $L^*a^*b^*$  colour coordinate system based on the D65 light source with the reflection spectra being measured in the 400–700 nm region. These values were used to calculate the colour change as a function of thermal treatment according to equations 1, 2 and 3.

$$\Delta L_t^* = L_t^* - L_s^* \quad (1)$$

$$\Delta a_t^* = a_t^* - a_s^* \quad (2)$$

$$\Delta b_t^* = b_t^* - b_s^* \quad (3)$$

where t = treated sample, s = control sample,  $\Delta$  = colour change,  $L^*$  = lightness,  $a^*$  = reddish colour,  $b^*$  = yellowish colour.

### Chemical analysis

The chemical changes studied were holocellulose, hemicellulose, cellulose and Klason lignin. All chemical analysis tests were conducted separately for heartwood and sapwood. The total amounts of chemical constituents were calculated based on the sapwood or heartwood ratios. Evaluation of chemical analysis was conducted using TAPPI T203 cm-99 (TAPPI 1999) and TAPPI T222 cm-02 (TAPPI 2002) standards.

### Sample preparation

Wood samples were divided into heartwood and sapwood to study the differences in chemical composition. The woods were chipped before grinding. Wood was ground into powder with a Wiley mill in order to pass the BS 40-mesh sieve and retained on BS 60-mesh sieve. The ground samples were then dried for seven days until the MC is in equilibrium with the surrounding before they were chemically analysed. A small

weighing bottle previously cleaned and dried in an oven was weighed on an analytical balance. An amount of 2 g of air-dried sawdust was placed in the weighing bottle and reweighed. The sample was then dried in an oven at  $103 \pm 2$  °C for three hours without the cover. Then the bottle was removed and placed in a desiccator for 15 min to let it cool down before weighing. The MC of the sawdust was determined.

## RESULTS AND DISCUSSION

### Colour changes

#### *Lightness ( $L^*$ ) of wood colour*

In this study, the main component in colour change is lightness ( $L^*$ ). Variation in  $L^*$  is a more responsive sign of colour change to the human eye than that of the chromic characters  $a^*$  and  $b^*$  (Keey 2004). Variability in darkness or lightness has been reported as the main cause of wood colour variability (Thulasidas et al. 2006).

Both sapwood and heartwood showed decrease in lightness in the first 30 min of treatment (Table 1).  $L^*$  values progressively decreased with increase in temperature and duration. However, the  $L^*$  values of heartwood did not change considerably compared with the sapwood. This may be due to the brightness of the wood colour itself as the sapwood of acacia has brighter colour than the heartwood. From the results obtained from the bottom, middle and top portions of sapwood, wood treated at 220 °C for 30 min showed  $L^*$  values having almost similar values to  $L^*$  values of untreated heartwood.

The decrease in  $L^*$  values from 200–220 °C was larger than 180–200 °C in sapwood and when compared with heartwood. The  $L^*$  values in heartwood changed slightly when the temperature exceeded 200 °C. The greatest change of treatment duration of both wood types was found between 60 and 90 min. This indicated that the change in  $L^*$  values gradually decreased with increase in treatment temperature and time.

Table 2 shows correlations between  $L^*$  and chemical components of *Acacia* wood. Positive correlations were observed between  $L^*$  and holocellulose and cellulose. Hemicellulose and lignin were negatively correlated with colour. The differences in chemical compositions such as extractives and lignin in sapwood and heartwood could probably be the main reason for dissimilarity in colour (Sundqvist 2004).

**Table 1** Average values of variation in L\*, a\* and b\* of oil heat-treated *Acacia* hybrid wood

Wood	Temperature (°C)	Treatment duration (min)	L*			a*			b*			
			B	M	T	B	M	T	B	M	T	
Sapwood	Control	Control	73.6	73.0	71.4	6.9	6.6	7.3	21.2	22.3	21.4	
		180	30	66.7	70.0	65.5	7.4	8.7	8.5	21.9	23.3	23.4
		60	65.4	62.7	63.0	8.8	9.9	9.8	25.7	26.3	24.1	
	180	90	65.6	61.9	62.9	9.3	10.9	10.2	27.3	27.5	26.3	
		200	30	63.5	63.5	58.2	9.5	10.0	9.9	26.4	26.1	26.2
		60	56.9	57.2	56.9	11.4	11.9	10.7	27.5	27.1	26.5	
	200	90	50.7	54.6	51.6	12.2	12.4	12.2	27.9	28.9	28.0	
		220	30	56.5	53.0	53.4	11.7	10.6	10.3	27.6	26.9	26.7
		60	48.7	47.9	43.2	12.3	12.7	11.2	27.8	27.6	27.8	
Heartwood	Control	Control	54.1	54.8	53.5	14.2	15.1	13.9	26.8	26.3	25.2	
		180	30	53.6	52.7	52.9	13.4	12.9	12.0	25.0	24.8	24.1
		60	51.7	51.6	50.7	11.9	11.1	11.5	23.6	23.2	23.5	
	180	90	50.4	50.2	48.2	11.2	10.8	10.7	23.0	21.5	22.3	
		200	30	51.4	52.1	48.6	11.5	10.3	11.3	23.7	24.1	23.9
		60	49.1	50.5	46.9	11.0	9.7	10.7	23.4	21.5	21.9	
	200	90	45.9	46.7	43.5	9.9	8.9	10.2	23.1	21.2	20.9	
		220	30	50.7	50.8	46.3	10.0	9.0	10.2	22.6	22.9	21.0
		60	46.6	47.4	38.8	8.3	8.3	8.4	21.1	19.8	17.2	
220	90	40.0	40.9	33.1	6.7	7.1	7.3	20.5	19.0	15.1		

L\* = lightness, a\* = red, b\* = yellow; B = bottom portion, M = middle portion, T = top portion

This is in accordance with the finding of Burtin et al. (1998) whereby during steaming at elevated temperatures, polyphenols found in hybrid walnut heartwood which gave the dark colour migrated to the sapwood region and changed the sapwood colour from light to dark.

Decrease in lightness due to heat treatment decreases certain chemical components in wood such as hemicellulose and lignin (Bourgiou et al. 1991, Mitsui et al. 2001). Changes in wood colour can be an indication of chemical modification or changes in wood (Burtin et al. 1998, Bekhta & Niemz 2003, Sundqvist et al. 2004).

#### *Chroma colour a\* (reddish colour)*

The a\* values of sapwood increased while those of heartwood decreased with hot oil treatment (Table 1). This may be because of the original colour of the wood samples. The original sapwood colour of *Acacia* hybrid is light yellowish

red while heartwood, brownish red. The a\* values reached maximum values at 220 °C. Based on the a\* values, the colour of sapwood became more red than before while the red colour in heartwood lessened. According to Hon and Minemura (1991), the reddish colour and increased saturation substantiated as a decrease in hue and increase in chroma could be due to the formation of secondary condensation or degradation products of the quinone and quinonemethide types.

Positive correlations were observed between a\* and b\*, holocellulose, cellulose and lignin (Table 2). Hemicellulose was negatively correlated with a\*. However a\* did not significantly correlate with chemical components except for lignin. According to Charrier et al. (2002) and Grelier et al. (1997), wood colours changed from yellow to brown because of the photo-oxidation of lignin and wood extractives, with the production of coloured quinone component.

**Table 2** Correlations between colour and chemical properties of 15-year-old cultivated *Acacia* hybrid

Property	L*	a*	b*	Holocellulose	Hemicellulose	Cellulose	Lignin
L*	1	-0.37**	0.11 ns	0.51**	-0.33**	0.53**	-0.75**
a*		1	0.47**	0.11 ns	-0.02 ns	0.10 ns	0.15*
b*			1	-0.07 ns	-0.07 ns	-0.04 ns	-0.01 ns
Holocellulose				1	-0.34**	0.96**	-0.69**
Hemicellulose					1	-0.59**	0.61**
Cellulose						1	-0.77**
Lignin							1

N = 216; \*\* = significant at  $p \leq 0.01$ , \* = significant at  $p \leq 0.05$ , ns = not significant; L\* = lightness, a\* = reddish, b\* = yellowish

### Chroma colour $b^*$ (yellowish colour)

Table 1 shows that  $b^*$  values for sapwood increase with increase in temperature and duration. The  $b^*$  values of sapwood showed the greatest increase when treated at 200–220 °C and 60–90 min. The yellowish colour of sapwood changed to brownish yellow.

The  $b^*$  values of heartwood decreased with increase in temperature and treatment time (Table 1). This meant that the yellowish colour in heartwood started to decrease from 180 °C and it decreased drastically when exposed to 220 °C and longer treatment times.

### Chemical changes

Table 3 shows that untreated sapwood and heartwood of *Acacia* hybrid have the highest percentages of holocellulose and cellulose compared with the other thermally modified wood in various temperatures and treatment times. These changes were due to the loss in polysaccharide material especially at temperature above 180 °C (Hill 2006). For treated sapwood, holocellulose varied between 63.1 and 70.8% while hemicellulose, between 23.0 and 26.1%. Cellulose varied between 37.7 and 46.2% while lignin, between 20.7 and 24.7% at 180–220 °C with 30–90 min treatments.

On the other hand, holocellulose for treated heartwood varied between 64.0 and 71.7%, hemicellulose between 23.2 and 26.2%, cellulose between 38.1 and 47.8% and lignin between 21.7 and 25.0% at 180–220 °C with 30–90 min treatments. The chemical compositions in the heartwood of *Acacia* hybrid were higher than in sapwood.

### Holocellulose

The holocellulose determined for sapwood and heartwood after heat treatment were between 63.1 and 70.8%, and 64.0 and 71.7% respectively (Table 3). The decrease in holocellulose content has also been observed by other researchers in the heat treatment process. Inari et al. (2007) found that holocellulose content of beech and pine woods after heat treatment was between 50 and 60%. Boonstra and Tjeerdsma (2005) also found that the holocellulose content of Scots pine heated at 180 °C decreased between 79.7 and 63.3%. The decrease is associated with loss of cellulose and hemicellulose during heating.

### Hemicellulose

The hemicellulose contents for both sapwood and heartwood showed no specific trend. This may be due to the structural heterogeneity of hemicellulose (Kotilainen 2000, Manninen et al. 2002). The hemicellulose content for sapwood degraded 5.7% when treated at 180 °C in 90 min while that of heartwood degraded from 0.8–5.3% when treated at temperature 180 °C from 30 to 90 min. Arabinose, manose, galactose and xylose which are responsible for hemicelluloses formation are significantly influenced by increasing temperatures (Rowell et al. 2005). The increment in lignin content of *Grevillea robusta* wood with treatment time confirmed higher susceptibility of hemicelluloses to thermal treatment (Mburu et al. 2008). According to Hill (2006), there are variations in the literature regarding the exact temperature for the onset of hemicellulose degradation

**Table 3** Average values of chemical changes of 15-year-old oil heat-treated *Acacia* hybrid wood

Wood	Treatment temperature (°C)	Treatment duration (min)	Chemical composition (%)				
			Holocellulose	Cellulose	Hemicellulose	Lignin	
Sapwood	Control	Control	71.5	47.1	24.4	20.8	
	180	30	70.8 (-1.0)	46.2 (-1.9)	24.6 (0.8)	20.7 (-0.5)	
		60	69.7 (-2.5)	45.1 (-4.2)	24.6 (0.8)	22.0 (5.8)	
		90	66.6 (-6.9)	43.6 (-7.4)	23.0 (-5.7)	22.1 (6.3)	
	200	30	68.6 (-4.1)	42.5 (-9.8)	26.1 (7.0)	23.9 (14.9)	
		60	65.3 (-8.7)	39.2 (-16.8)	26.0 (6.6)	23.8 (14.4)	
		90	64.5 (-9.8)	38.5 (-18.3)	26.0 (6.6)	24.7 (18.8)	
	220	30	66.4 (-7.1)	41.6 (-11.7)	24.8 (1.6)	23.7 (13.9)	
		60	64.9 (-9.2)	38.9 (-17.4)	26.0 (6.6)	24.3 (16.8)	
		90	63.1 (-11.7)	37.7 (-20.0)	25.4 (4.1)	24.0 (15.4)	
	Heartwood	Control	Control	73.4	48.9	24.5	22.4
		180	30	71.7 (-2.3)	47.8 (-2.3)	23.9 (-2.5)	21.7 (-3.1)
60			70.6 (-3.8)	46.3 (-5.3)	24.3 (-0.8)	22.9 (2.2)	
90			67.9 (-7.5)	44.7 (-8.6)	23.2 (-5.3)	23.0 (2.7)	
200		30	68.8 (-6.3)	42.6 (-12.9)	25.9 (5.7)	24.5 (9.4)	
		60	68.3 (-6.9)	42.1 (-14.0)	26.2 (6.9)	24.9 (11.2)	
		90	66.5 (-9.4)	40.9 (-16.4)	25.6 (4.5)	25.0 (11.6)	
220		30	67.1 (-8.6)	42.0 (-14.1)	25.7 (4.9)	24.7 (10.3)	
		60	65.1 (-11.3)	39.7 (-18.8)	25.4 (3.7)	24.8 (10.7)	
		90	64.0 (-12.8)	38.1 (-22.1)	25.9 (5.7)	24.9 (11.2)	

( ) = % change from control

Holocellulose is the total polysaccharide fraction of wood that is composed of cellulose and all of the hemicelluloses and what is obtained when the extractives and lignin are removed from the natural material.

although it is well known that hemicelluloses are less thermally stable than cellulose. Moreover, in the pyrolysis of hemicelluloses, more gaseous products and less charred residue are formed compared with cellulose. When wood is heated, the most thermally labile of the hemicelluloses begin to degrade, resulting in the production of methanol, acetic acid and various volatile heterocyclic compounds (Hill 2006). The acetic acid is generated when the acetylated hydroxyl groups of the hemicellulose chains are split off (Johansson 2008). It is suggested that volatile organic acids formed during heating of wood are trapped in the process and promote the degradation rate (Viitaniemi 2001).

### Cellulose

The cellulose contents for both sapwood and heartwood after hot oil thermal modification

were between 37.7 and 46.2% and 38.1 and 47.8% respectively (Table 3). Minor degradation of celluloses occurred when wood was treated at 180 °C in 30 min and it continually decreased with increase in temperature and duration of treatment. Similar observation was also made by Izyan et al. (2010) on chemical constituents of heat-treated *Acacia* hybrid. Cellulose is more resistant to hydrolysis than hemicelluloses, pectins and starch, and it generally has a more regular and crystalline structure with considerably higher molecular weight (Sundqvist 2004). Intrinsic viscosity measurement shows that heat treatment results in a considerable reduction in molecule size of cellulose (Sundqvist 2004). The crystalline structure of cellulose can change once the temperature reaches 200 °C (Yilidiz et al. 2006). Different process conditions and treatment times applied during heat treatment could influence the degradation rate of cellulose (Boonstra &

Tjeerdsma 2005). Fengel and Wegener (1989) found that the degree of polymerisation of cellulose was decreased in thermally treated spruce at temperatures above 120 °C. This is due to cleavage of the glucosidic bonding that is accelerated by the acids that are catalysing the reaction. With extended heating, chain scission of the cellulose occurs, producing alkaline soluble oligosaccharides, with a concomitant decrease in the degree of polymerisation of cellulose and degree of crystallinity (Hill 2006). The amorphous regions of cellulose are more susceptible to thermal degradation and probably exhibit similar thermal properties to the hexose components of hemicelluloses (Hill 2006).

### Lignin

Lignin in both the sapwood and heartwood increased with treatment temperature and time. Lignin composition of the *Acacia* hybrid sapwood increased from 20.8% in the untreated wood to 24.7% in the heat-treated wood. Lignin composition of the heartwood increased from 22.4% in the untreated heartwood to 25% in the heat-treated wood. The changes showed that there was a change in the lignin structure of the woods. Similar observations were also made by other researchers (Sarni et al. 1990, Yildiz et al. 2006, Inari et al. 2007, Brito et al. 2008, Mburu et al. 2008). Increase in lignin content of oak was observed as treatment temperature increased (Sarni et al. 1990). Mburu et al. (2008) also found increase in lignin content of *G. robusta* wood with treatment time. The relative mass proportion of lignin increased with both elevated temperature and treatment time, with a simultaneous decrease in the mass proportion of carbohydrates (Kotilainen 2000). Since lignin presents a higher resistance to heat, there is a significant and proportional increase in its content (Brito et al. 2008). Sundqvist (2004) reported that noticeable changes in the lignin structure occurred at temperatures about 120 °C. With rising temperatures, the changes increased. At temperature around 180 °C, the degradation of lignin in the thermal-treated wood was considerable.

### Analysis of variance on colour and chemical changes

Lightness ( $L^*$ ) through oil heat treatment was significantly affected by treatment temperature, duration and wood type (Table 4). This meant that  $L^*$  played a significant role in indicating the change of wood colour. However, for chroma colour  $a^*$  and  $b^*$ , only wood type showed significant difference. This showed that chroma colour did not have much influence on the change of wood colour. There were significant differences between treatment temperature, duration and wood type in relation to colour and chemical components such as holocellulose, cellulose and lignin. ANOVA showed that there was no significant difference between wood type and hemicellulose. It can be concluded that wood type did not affect the hemicellulose content in oil heat treatment process. Temperature and treatment duration increased the hemicellulose composition. Most of the chemical compositions in *Acacia* hybrid wood were highly affected by treatment temperature, duration and wood type in the hot oil thermal modification process.

### CONCLUSIONS

The colour and chemical properties of hot oil thermally modified cultivated *Acacia* hybrid changed during the treatment process. Changes in the colour and chemical contents of *Acacia* hybrid were significantly affected by temperature and treatment time.

The colour of the sapwood and heartwood of *Acacia* hybrid became darker once they were exposed to high temperature and longer treatment time in the hot oil thermal modification process. The degree of changes varied between both wood types. The sapwood tended to darken more than the heartwood. The increment in colour of both woods increased with temperature and treatment time. The changes in the wood could be monitored in order to get the desired colour.

The hot oil thermal modification process caused some changes in the chemical compositions of *Acacia* hybrid wood. Changes in chemical components occurred with decreases in holocellulose and cellulose contents, and

**Table 4** ANOVA of colour and chemical changes of treated 15-year-old *Acacia* hybrid

Source of variance	Dependent variable	Sum of squares	Df	Mean square	F-ratio
Temperature	L*	9538.60	3	3179.53	110.70**
	a*	9.33	3	3.11	0.42ns
	b*	35.41	3	11.80	1.10ns
Duration	L*	1257.57	2	628.79	21.89**
	a*	1.43	2	0.72	0.10ns
	b*	2.67	2	1.33	0.12ns
Wood type	L*	6537.30	1	6537.30	227.60**
	a*	94.80	1	94.80	12.82**
	b*	294.70	1	294.70	27.55**
Temperature	Holocellulose	1618.96	3	539.65	564.15**
	Hemicellulose	139.12	3	46.37	85.82**
	Cellulose	2460.23	3	820.08	1018.75**
	Lignin	368.90	3	122.97	461.45**
Duration	Holocellulose	250.82	2	125.41	131.10**
	Hemicellulose	7.43	2	3.72	6.88**
	Cellulose	210.26	2	105.13	130.60**
	Lignin	12.69	2	6.35	23.81**
Wood type	Holocellulose	96.80	1	96.80	101.20**
	Hemicellulose	0.01	1	0.01	0.01ns
	Cellulose	95.20	1	95.20	118.27**
	Lignin	52.22	1	52.22	195.94**

\*\* = significant difference at  $p \leq 0.01$ , \* = significant difference at  $p \leq 0.05$ , ns = not significant

increases in hemicellulose and lignin contents when the *Acacia* woods were exposed to oil-heat treatment process at temperatures higher than 180 °C.

## REFERENCES

- BEKHTA P & NIEMZ P. 2003. Effect of high temperature on the change in color, dimensional stability and mechanical properties of Spruce wood. *Journal of Holzforschung* 57: 539–546.
- BERARD P, LAURENT T & DUMONCEAUD O. 2006. Use of round wood of chestnut tree coppices: crack risk and effects of a hot oil bath treatment. *Journal of Holz als Roh- und Werkstoff* 64: 287–293.
- BOONSTRA MJ & TJEERDSMA B. 2005. Chemical analysis of heat treated softwoods. *Journal of Holz als Roh- und Werkstoff* 64: 204–211.
- BOURGIOS PJ, JANIN G & GUYONNET R. 1991. La mesure de couleur. une methode d'etude et d' optimisation des transformations chimiques du bois thermolyse. *Journal of Holzforchung* 45: 377–382.
- BRITO JO, SILVA FG, LEO MM & ALMEIDA G. 2008. Chemical composition changes in *Eucalyptus* and *Pinus* woods submitted to heat treatment. *Journal of Bioresource Technology* 99: 8545–8548.
- BURTIN P, JAY-ALLEMAND C, CHARPENNIER JP & JANIN G. 1998. Natural wood colouring process in *Juglans* spp. (*J. nigra*, *J. regia*, and hybrid *J. nigra* × *J. regia*) depends on native phenolic compounds accumulated in the transition zone between sapwood and heartwood. *Journal of Trees* 12: 258–264.
- CHARRIER B, CHARRIER F, JANIN G, KAMDEM DP, IRMOULI M & GONCALEZ J. 2002. Study of industrial boiling process on walnut colour: experimental study under industrial conditions. *Journal of Holz als Roh- und Werkstoff* 60: 259–264.
- CIE (COMMISSION INTERNATIONALE DE ECLAIRAGE). 1986. *Colorimetry*. Second edition. Central Bureau of the CIE, Vienna.
- FENDEL D & WEGENER G. 1989. *Wood Chemistry, Ultrastructure, Reactions*. Walter de Gruyter and Co., Berlin.
- GRELIER S, CASTELLAN A, DESROUSSEAU S, NOUR A, ODE A & PODGORSKI L. 1997. Attempt to protect wood colour against UV/visible light by using antioxidants bearing isocyanate groups and grafted to the material with microwave. *Journal of Holzforschung* 51: 511–518.
- HILL C. 2006. *Wood Modification: Chemical, Thermal and Other Processes*. John Wiley & Sons Ltd., England.
- HYVONEN A, PILTONEN P & NIINIMAKI J. 2006. Tall oil/water-emulsions as water repellents for Scots pine sapwood. *Journal of Holz als Roh- und Werkstoff* 64: 68–73.
- HON DNS & MINEMURA N. 1991. Color and discoloration. Pp. 395–454 in Hon DNS & Shirashi N (Eds.) *Wood*



- and *Cellulosic Chemistry*. Marcel Dekker Inc., New York.
- INARI GN, PETRISSANS M & GERARDIN P. 2007. Chemical reactivity of heat-treated wood. *Journal of Wood Science and Technology* 41: 157–168.
- IZYAN K, RAZAK W, MAHMUD S, OTHMAN S, AFFENDY H, HANIM RA & ANDY RM 2010. Chemical changes in 15-year-old cultivated *Acacia* hybrid oil-heat treated at 180, 200 and 220 °C. *International Journal of Chemistry* 2: 97–107.
- JOHANSSON D. 2008. Heat treatment of solid wood: effects on absorption, strength and colour. PhD thesis, Lulea University of Technology, Lulea.
- KEY RB. 2004. Colour development on drying. Pp. 33–47 in *Proceedings of the 14th International Drying Symposium (IDS 2004)*. 22–25 August 2004, Sao Paulo.
- KOTILAINEN R. 2000. Chemical changes in wood during heating at 150 °C–260 °C. PhD thesis, Jyväskylä University, Jyväskylä.
- MANNINEN AM, PASANEN P & HOLOPAINEN JK. 2002. Comparing the VOC emissions between air-dried and heat-treated Scots pine wood. *Journal of Atmospheric Environment* 36: 1763–1768.
- MBURU F, DUMARCAY S, BOCQUET JF, PETRISSANS M & GERARDIN P. 2008. Effect of chemical modifications caused by heat treatment on mechanical properties of *Grevillea robusta* wood. *Journal of Polymer Degradation and Stability* 93: 401–405.
- MITSUI K, TAKADA H, SUGIYAMA M & HASEGAWA R. 2001. Changes in the properties of light-irradiated wood with heat treatment. Part I. Effect of treatment conditions on the change in color. *Journal of Holzforschung* 55: 601–605.
- RAFIDAH MD. 2007. Effect of high temperature boiling on properties of *Gigantochloa scortechnii* Gamble (buluh semantan). MSc thesis, Universiti Putra Malaysia, Serdang.
- ROWELL RM, PETTERSEN R, HAN JS & TSHABALALA MA. 2005. Cell wall chemistry. Pp. 37–72 in Rowell RM (Ed.) *Handbook of Wood Chemistry and Wood Composites*. CRL Press, Madison.
- SARNI F, MOUTOUNET M, PUECH JL & RSBIER P. 1990. Effect of heat treatment on oak wood extractable compounds. *Journal of Holzforschung* 44: 461–466.
- SHAKRI MS. 1995. Finishing properties of *Acacia mangium*, *Paraserianthes falcataria* and *Gmelina arborea* timbers: some important parameters. *Journal of Tropical Forest Products* 1: 83–89.
- SUNDQVIST B. 2004. Colour changes and acid formation in wood during heating. PhD thesis, Lulea University of Technology, Lulea.
- TAPPI (TECHNICAL ASSOCIATION OF THE PULP AND PAPER INDUSTRY). 1999. *TAPPI Standard T203 cm-99*. TAPPI, Atlanta.
- TAPPI (TECHNICAL ASSOCIATION OF THE PULP AND PAPER INDUSTRY). 2002. *TAPPI Standard T222 cm-02*. TAPPI, Atlanta.
- THULASIDAS PK, BHAT KM & OKUYAMA T. 2006. Heartwood colour variation in home garden teak (*Tectona grandis*) from wet and dry localities of Kerala, India. *Journal of Tropical Forest Science* 18: 51–54.
- TOLVAJ L & MOLNAR S. 2006. Colour homogenization of hardwood species by steaming. *Journal of Acta Silvaticam Lignaria Hungarica* 2: 105–112.
- UNSAI O, KORRUT S & ATIK C. 2003. The effect of heat treatment on some properties and colour in *Eucalyptus* (*Eucalyptus camaldulensis* Dehn.) wood. *Journal of Maderas (Ciencia Tecnologia)* 5: 145–152.
- VIITANIEMI P. 2001. The thermal modification of wood with heat treatment. Pp. 1–21 in *VTT Building and Transport*. Espoo, Finland.
- YILDIZ S, GEZER ED & YILDIZ UC. 2006. Mechanical and chemical behaviour of Spruce wood modified by heat. *Journal of Building and Environment* 41: 1762–1766.