

# Improvements in stability, durability and mechanical properties of radiata pine wood after heat-treatment in a vegetable oil

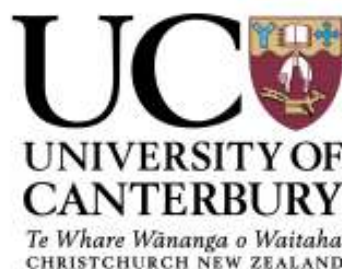
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By

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## **ABSTRACT**

Radiata pine is a major plantation grown wood in the Southern hemisphere, but has inferior dimensional stability and low durability compared to other commercial species and the improvement of these features is the focus of this thesis.

Specifically this thesis examines the dimensional stability, durability and mechanical properties of radiata pine after heat-treatment (160-210°C) in linseed oil. Changes in colour, micro-structure and chemistry with heat-treatment were studied. To optimise the treatment results, oil heat-treatment of wood was also carried out after thermo-mechanical compression of wood and the effect of the prior thermo-mechanical compression on stability, durability and mechanical properties were examined.

The oil heat-treated wood turned uniformly darker in colour. The hydrophobicity (Water Repellence Efficiency-WRE up to 30%), dimensional stability (Ant-Swelling Efficiency-ASE up to 60%) and fungal resistance (up to 36%) were improved with the extent of the changes mainly depending on treatment temperature. However, the mechanical properties of oil heat-treated wood were reduced compared to the untreated control group. Accelerated UV weathering tests have shown that the oil heat-treated wood retained its colour and dimensional stability better than the untreated wood (the control group).

The cell wall of treated wood was intact and no distinct structural changes were observed even at the most severe treatment (210°C for 6 hrs). The treatment resulted in changes to the wood chemical constituents, mainly the degradation of hemicelluloses which is believed to be principal reason for alterations in wood properties.

A study of the effect of prolonged heating on the linseed oil showed an increase in viscosity with heating time which in turn reduced the oil uptake and water repellency of treated wood. However, no significant difference in the colour and dimensional stability of the treated wood was noticed with oil of different heating ages. Oil absorbed by the wood during heat treatment was removed by organic solvent extraction and its contribution to the weight percentage change and moisture related properties were evaluated. The oil uptake percentage, determined by organic solvent extraction, was greater than the weight percentage loss that was deduced to occur during the heat treatment phase, which was attributed to mass losses or thermal degradation of wood constituents. Moisture excluding efficiency decreased after removal of the oil from treated wood, which suggested that the hydrophobicity of treated wood is affected by oil absorption. The influence of the post-treatment cooling period on properties of treated wood was studied separately. Oil uptake increased substantially with the post-treatment cooling time which in turn affected the hydrophobicity of treated wood although this effect was less important to dimensional stability.

The loss of mechanical properties due to heat-treatment was successfully countered by thermo-mechanical compression of wood prior to the oil heat-treatment. The wood was compressed to 39% of its original thickness without any visible surface checks and cracks. Spring back and compression set recovery in densified wood decreased after oil heat-treatment. This combination treatment also resulted in improved fungal resistance compared to untreated wood.

From this research, it is concluded that oil heat-treatment of radiata pine wood can improve its dimensional stability and durability obviating the need to introduce any persistent toxic chemicals. Thermo-mechanical densification of wood prior to oil heat-treatment can counter the loss of mechanical properties due to heat-treatment. The heating oil can be re-used in subsequent treatments and oil uptake can be minimised by limiting the post-treatment cooling time without any significant effect on the dimensional stability of treated wood.

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## **ABBREVIATIONS**

ASE: Anti-Swelling Efficiency

ASTM: American Standard Testing and Materials

BSI: British Standard Institute

CIE: Commission Internationale d'Eclairage (International Commission on Illumination)

DP: Degree of Polymerisation

FTIR: Fourier Transform Infrared spectrometry

LOI: Linseed Oil Impregnation

MEE: Moisture Excluding Efficiency

MOE: Modulus of Elasticity

MOR: Modulus of Rupture

MTS: Material Testing Machine

OHT: Oil Heat Treatment

RH: Relative Humidity

SEM: Scanning Electron Microscopy

TAPPI: Technical Association of Pulp and Paper Industries

TMC: Thermo-Mechanical Compression

WFD: Wet-Freeze-Dry

WL: Weight Loss percentage

WPC: Weight Percentage Change

WRE: Water Repellence Efficiency

# Chapter 1

## INTRODUCTION

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### 1.1. Background

Awareness of the society towards the impacts of human activities on the environment is increasing and environmental considerations are changing the way in which the materials are being utilized. Thus, it is important to manufacture products with minimal environmental impact. Wood is a biologically derived renewable raw material with a low processing energy demand. It is recyclable and biodegradable. Timber products with prolonged life can be used as a repository for CO<sub>2</sub> (Bruce and Lucy 2006) and can be considered as a future-oriented material to achieve the universal goal of sustainable development and a low carbon future. Recently, to promote the use of wood the Ministry of Forestry in New Zealand launched a new programme for sustainable New Zealand building which states “*all new government buildings up to four storeys must consider wood as the main structural material from 2008*” (Bodeker 2008).

In the past, people had the privilege of having abundant quantities of high quality wood from natural forests. The wood with excellent desirable inherent characteristics was generally a ‘ready to use’ naturally available raw material requiring little processing effort. But the supply of these wood raw materials has become scarce and no significant supply can be expected from the natural forests in the years to come. The dependence of wood industries on fast grown plantation wood as the main source of raw

material is predicted to significantly increase in the coming years (Carle and Holmgren 2008; FAO 2009).

Radiata pine is one of the most important fast grown plantation species in the southern hemisphere. It is considered to be the backbone of New Zealand forestry. The availability of radiata pine wood from sustainably managed forests could be 30 million m<sup>3</sup> p.a. in the next 5 years which makes it a potential wood of the future to satisfy increasing timber demands across the globe, in particular from fast growing economies such as China, India and Korea. It can be a logical alternative to tropical and native timbers as well as replacing plantation timbers coming from non-sustainable resources.

However, some of the wood properties of radiata pine, in particular dimensional stability and durability, are inferior compared to most commercial species (Bamber and Burley 1983). In the past few years radiata pine has lost markets in the construction industry to steel and concrete due to its dimensional instability (The Press 2008). Dimensional instability, low mechanical properties and low durability have put this wood at a disadvantageous position in the competitive global market (Dickson and Walker 1997; Walker 2006). The improvement of the aforesaid properties of radiata pine in an environmentally benign way is one of the biggest challenges but also an opportunity for wood technologists.

Plantation wood is generally treated with chemicals in the form of water repellents and/or preservatives to protect it against moisture fluctuation, micro-organisms and UV rays and to improve their dimensional stability and durability. Certain chemicals used in traditional water repellents and preservative solutions, such as pentachlorophenol (PCP), copper-chrome-arsenate (CCA) and creosote, are toxic to mammals and harmful to the

environment (Cooper 2001; Hale 2003; Williams and Feist 1999). Apart from human health and safety issues, disposal of such wood after the end of its service life is also a major concern (Syrjänen and Kangas 2001). Therefore, use of these chemicals is already banned or restricted for certain applications in many regions of the world such as Europe, North America and Japan (Drysdale 2002; Williams 1999). Also, there are increasing environmental concerns within furniture industries over the use of solvent-borne wood finishes (Pandey 2005). Recently, in New Zealand, the adverse effect on human health was reported from the use of wooden floors coated with moisture-cured polyurethane (3NEWS 2009). In future these concerns may restrict the use of other chemicals too, for example some environmentalists are even concerned over the use of copper which is an integral component of most of the novel wood preservatives systems (Dubey 2003; Hale 2003). These may force industry to opt for non-biocidal methods to improve the durability and stability of wood. All these developments have resulted in significant research and commercial interest mainly in Europe with wood modification technologies such as thermal modification (heat-treatment) and chemical modification of wood. In addition, the demand for high quality wood products and the decreasing availability of tropical wood species also increase the importance of these technologies (Evans 2003; Homan and Jorissen 2004). These wood modification technologies can play an important role in improving the stability and durability of wood without using any persistent toxic chemicals. The improved characteristics of the modified wood offer many potential and attractive opportunities for the wood industry and it is expected that there will be more research and commercial developments in this area in the near future.

Being a vegetable oil, linseed oil possesses favourable environmental characteristics such as renewability, low mammalian toxicity and biodegradability (Gunstone 2002; NNFC 2007). In addition, a long history of its use in wood protection, ease of drying, ready availability and a high boiling point at atmospheric pressure make this oil a preferred candidate for use in wood modification (ASTM-D234-82 1998; Chemwatch 2007; Treu *et al.* 2001). A considerable number of published research papers and reviews are available in literature on wood modification processes involving impregnations with a range of chemicals and on wood heat-treatments in air, steam or a nitrogen atmosphere (Hill 2006; Militz 2002). However, information on wood modification treatments with linseed oil is limited (Lyon *et al.* 2007; Sailer *et al.* 2000b). In the thermal treatment, certain issues need to be addressed such as loss of mechanical properties after heat-treatment (Bengtsson *et al.* 2002; Sailer *et al.* 2000a; Yildiz *et al.* 2005) and colour fading of heat-treated wood on UV exposure (Ahajji *et al.* 2009; Militz 2002; Syrjänen and Kangas 2000). In addition, in the oil thermal treatment of wood, the effect of oil ageing and the cooling period after the treatment on the properties of heat-treated wood is important and no reports on this have been found in the literature.

The aim of this study was to investigate the dimensional stability and durability of New Zealand grown radiata pine wood after (1) heat-treatment with linseed oil, and (2) thermo-mechanical compression followed by oil heat-treatment. In addition, changes in structure, chemistry, colour and mechanical properties of wood with these treatments were examined. The effect of wood densification; removal of oil from treated wood; accelerated UV weathering, oil ageing and post-treatment cooling period on the properties of the treated wood were also investigated.



## 1.2. Objectives

The specific objectives of the study are given below:

1. Investigation of dimensional stability, water repellency, durability and colour changes in wood after heat-treatment using linseed oil.
2. Examination of the micro-structure, chemistry and mechanical properties of wood after the oil heat-treatment.
3. Assessment of the effect of wood densification prior to the oil heat-treatment on dimensional stability, mechanical properties and fungal resistance of wood.
4. Measurement of oil uptake using organic solvent extraction of the oil heat-treated wood, and examination of effect of the oil uptake on water repellency.
5. Evaluation of oil heat ageing and the post-treatment cooling period on oil uptake and moisture related properties of the heat-treated wood.

## 1.3. Thesis structure

Following the introduction the overall structure of the thesis is organized as follows:

**Chapter 2** begins with a brief description of radiata pine plantation resources and their significance. Then dimensional stability and durability of the radiata pine wood is discussed after a brief review of its structure and chemical composition. Following this, the chapter introduces heat-treatment and thermo-mechanical densification of wood.

Finally, an overview of production, chemical composition, properties and application of linseed oil is provided.

**Chapter 3** contains details of materials, specimen preparation procedures and gives a brief account of wood treatment experiments. Details of different treatments are discussed later in relevant chapters. Finally, it gives the methodology used for measurement and analysis of wood properties concerned. Any additional test or changes in the experiment procedure adopted for a specific property are described in the chapter concerned.

**Chapter 4** presents experiments and results for the oil heat-treatment of wood and discuss changes in structure, chemistry and wood properties after the treatment.

**Chapter 5** examines the effect of a combination treatment involving densification of wood prior to the oil heat-treatment on the wood properties, mainly dimensional stability and mechanical properties.

**Chapter 6** investigates the colour and dimensional stability of oil heat-treated wood after an accelerated UV weathering.

**Chapter 7** examines the effect of oil ageing on the oil viscosity and properties of heat-treated wood.

**Chapter 8** is focused on the measurement of oil uptake using a solvent extraction method. Change in the hydrophobicity of heat-treated wood after removal of the absorbed oil by solvent extraction is examined. The effects of the cooling period after heat treatment on oil uptake and moisture related properties of wood are also investigated.

**Chapter 9** includes general conclusions, the significance of the findings from this work and recommendations for future work.

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# Chapter 2

## LITERATURE REVIEW

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### 2.1. Radiata pine

#### 2.1.1. Global plantation resources

The global area of plantation forestry in 2005 was estimated as more than 260 million hectares and is projected to be around 345 million hectares by 2030 (Del Lungo *et al.* 2006). Although planted forests constitute only 7% of total forest area they are very significant from social, economic and commercial points of view. Forest plantations are the main raw material resource for the forest based industries owing to decline in the commercial harvesting from the natural forests (FAO 2009). This indicates the significance of plantations to meet the socio-economic demands for forest products of current and future generations. Forest industries have to accept and accommodate themselves to the optimum utilization of plantation resources.

The annual wood production from the plantation forests is 1.2 billion m<sup>3</sup> which is two thirds of overall wood production. This is expected to increase to 1.9 billion m<sup>3</sup> in 2030 (Carle and Holmgren 2008). Among different plantation species genus *Pinus* is the largest planted group world wide with a 73.2 million hectares plantation area (Table 2.1). The plantation area of pines is also projected to increase by 30% in 2030. Pine plantations are dominant in the Oceania region, constituting 75% of total plantations.

Radiata pine is an important plantation species of the Southern Hemisphere. The botanical name of the species is *Pinus radiata* D. Don. It is also known as “Monterey Pine”, “Remarkable Pine” and “Pino Insigne”. The species is versatile, highly productive, can grow in a range of growing environments, is easy to establish, relatively free from diseases and provides a larger timber yield in a shorter time (Bootle 2005).

**Table 2.1. Species wise plantation forest area in million hectares (Carle and Holmgren 2008)**

	2005		2030 projection	
	World	Oceania	World	Oceania
Pinus	73.2	2.7	94.9	2.9
Other softwood	64.0	0.2	83.0	0.2
Acacia	9.1	-	9.9	-
Eucalyptus	13.8	0.5	21.4	0.8
Other hardwood	100.3	0.2	135.5	0.3
<b>Total</b>	<b>260.5</b>	<b>3.6</b>	<b>344.6</b>	<b>4.2</b>



Worldwide, the plantation area of the radiata pine is about 4.3 million ha (Lungo *et al.* 2006). It contributes 1.1% of global and 8.8% of the Asia-Pacific trade in forest products (FAO 2005b). The species is suitable for temperate climates and is widely planted in Argentina, Australia, Chile, Kenya, New Zealand, South Africa and Uruguay. It is also a rapidly emerging plantation species in China (Kamke 2006). Table 2.2 shows the main regions of radiata pine plantations with their plantation areas.

**Table 2.2. Major plantation regions of radiata pine in the world**

<i>Region</i>	<i>Area ( million Ha )</i>	<i>Source</i>
New Zealand	1.60	(MAF 2009)
Chile	1.30	(Toro and Stanley 1999)
Australia	0.74	(DPINSW 2007)
Spain	0.26	(DPINSW 2007)
South Africa	0.05	(Sutton 1999)

### **2.1.2. Radiata pine in New Zealand**

New Zealand is the largest plantation region of radiata pine in the world with about 1.6 million hectares of plantation area (MAF 2009). The species was introduced in 1859 as an aesthetic species from North America. Since the early 1880s, New Zealand has planted radiata pine

and today all major wood industries rely on it as the principal source of raw material (TRC 2007). Today it meets all domestic wood requirements and also contributes substantially to the export trade of logs and wood products.

Table 2.3 shows the areas of natural and plantation forests in New Zealand with the main species in each type. Radiata pine occupies more than 90% of the plantation forests and grows in a wide range of conditions. This species is fast growing with an average harvesting age of 26-28 years (Kininmonth and Whitehouse 1991).

**Table 2.3. Forest areas in New Zealand in 2007 (MAF 2009)**

<i>Type</i>	<i>Main Species</i>	<i>Area (million Ha)</i>	<i>Percentage of geographical area</i>
(1) Natural or indigenous forests	Beech, kauri, rimu, taraire and tawa.	6.5	23
(2) Plantation or exotic forests		1.79	4
(2.1) <i>Radiata forests</i>	<i>Radiata pine</i>	1.6	
(2.2) <i>Non-radiata forests</i>	<i>Douglas fir, other conifers &amp; hardwoods</i>	0.19	
Total		8.29	27

### **2.1.3. Significance of radiata pine plantations**

The use of plantation grown wood is increasing significantly owing to depletion of tropical forests and the ban or restrictions on felling in natural forests in many countries. New Zealand is a noteworthy supplier to world softwood timber markets mainly from its radiata pine resource. Forest products are the third largest export item and in 2006, New Zealand's log and lumber exports were 8 mill m<sup>3</sup> by volume and NZ\$1.33 billion by value (MAF 2009). The species has huge growth potential in the future. The annual sustainable harvest of pine forests is forecast to be more than 40 million m<sup>3</sup> and the annual export of wood products is projected to be more than NZ\$14 billion within the next 20 years (MAF 2009). Prior to export most of the surplus wood could be available for value added processing through efficient technologies such as wood modification through heat-treatment.

Asia is the biggest export market of radiata pine. The availability of significant quantities of radiata pine wood from sustainably managed forests makes it increasingly attractive to some of the fast growing economies like China, India and Korea to meet their increasing demand for wood and wood products. It could be a logical alternative to tropical and native timbers as well as a substitute for plantation timbers coming from non-sustainable resources.

Unfortunately, the quality of radiata pine wood is generally inferior in comparison to most of the commercial species of the world and a major challenge for future prospects for the species in domestic and global markets (Bamber and Burley 1983; Dickson and Walker 1997).

## **2.1.4. Radiata pine wood**

### *2.1.4.1. General structure*

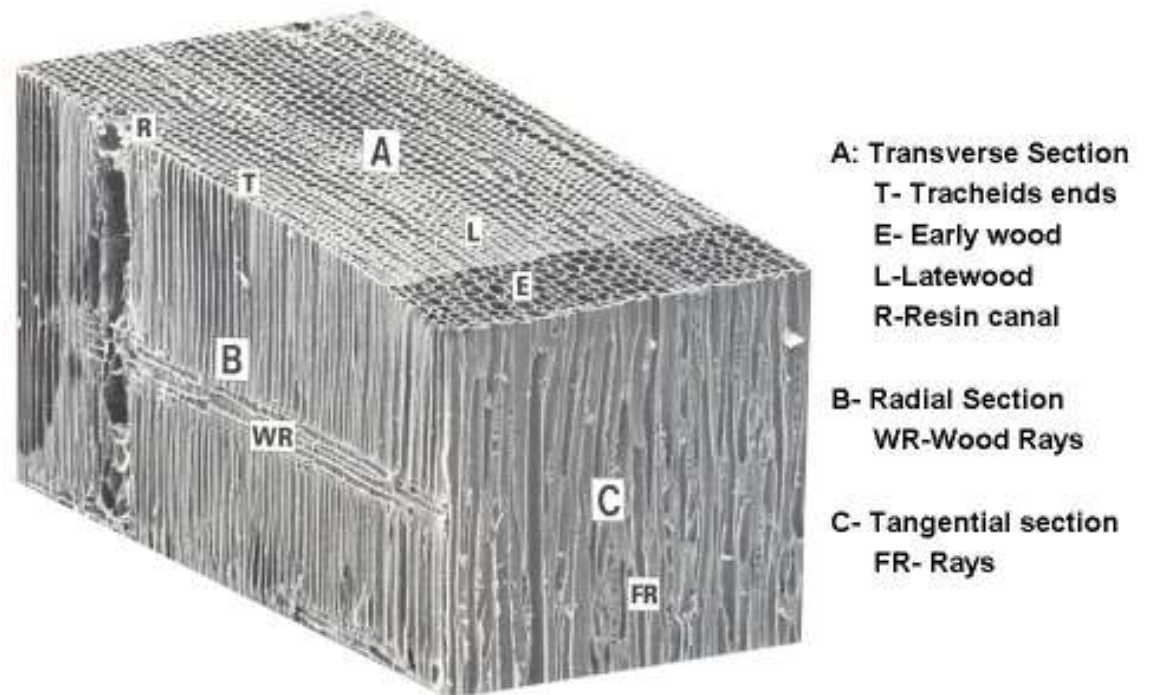
Radiata pine is a “softwood or conifer”, a botanical group of trees, usually with needle shaped leaves. The cross-section of the stem generally reveals a light or chestnut brown central portion known as “heartwood” surrounded by “sapwood” which is creamy white in colour. Sapwood is further surrounded by an outermost protective layer, “bark”. The heartwood formation starts when the tree is about 15 years old (Walker 2006) and in 20-30 year old trees, 80-90% of the volume consists of sapwood. (Cown 1992) termed radiata plantations “sapwood trees” owing to their wide sapwood zone and the moderately pale colour of their heartwood.

The cross-section of radiata pine wood is characterized by the concentric layers known as growth rings. These growth rings are composed of earlywood or springwood – cells with larger lumens and thin cell walls – and latewood or summerwood – cells with smaller lumens and thicker cell walls. The transition between earlywood and latewood is gradual which makes the wood even textured (Harris 1991b).

Figure 2.1 shows the main wood structure of radiata pine wood in three different surfaces of the block: (1) transverse or cross-sectional: the surface at the end of a board or log; (2) radial: the surface passing through the pith along rays and normal to growth rings; and (3) tangential: the surface tangential to growth rings. The three surfaces of wood are important because wood structures and properties are different in these directions.

More than 90% of the volume of radiata pine wood is composed of long and narrow tube-like cells known as “tracheids”. These cells are aligned

axially around the stem axis. They are closed, pointed at the ends and have a hollow centre known as a lumen. Tracheids are connected to each other by small openings known as pits. In radiata pine the average tracheid length, diameter and wall thickness are 3.5-4  $\mu\text{m}$ , 30-40  $\mu\text{m}$  and 4-5  $\mu\text{m}$  respectively (Cown 1992). The cell wall is composed of a thin outer layer known as primary wall and a thick inner layer, the secondary wall. The middle lamella is the intercellular layer which holds the wood cells together. The secondary wall is further divided into three sub-layers: a thin outer S1 layer, a thick middle S2 layer and a thin inner S3 layer (towards the cell lumen).



*Figure 2.1. Wood structure of radiata pine wood in three different directions (Harris 1991b)*

Among the tracheids are a few small rectangular cells. These cells are thin walled parenchyma cells. These parenchyma cells, together with ray tracheids form the “rays” which are narrow uniseriate or biseriate stripes crossing the growth rings and extending from the bark toward the centre of the tree. They assist in horizontal transport and storage of nutrients in standing trees. Resin canals are a prominent feature of radiata pine and contain and transmit resinous materials. They are the intercellular passages surrounded by thin walled parenchymal cells (Bamber and Burley 1983).

#### *2.1.4.2. Chemical composition*

Wood is mainly composed of cellulose, hemicelluloses and lignin. Extractives, pectin substances and ash are the minor constituents of wood. Table 2.4 gives the chemical composition percentage of radiata pine, along with other softwoods and hardwoods.

Cellulose is the main structural component of wood. It is a polydispersed linear homopolysaccharide of  $\beta$ -D-glucopyranose units linked together by  $\beta$  1-4 glycosidic bonds. The empirical formula of cellulose is  $(C_6H_{10}O_5)_n$ , where ‘n’ is the degree of polymerization or the number of glucose molecules per cellulose chain. The degree of polymerization of wood cellulose is of the order of 10000. A cellulose molecule is highly crystalline (60-75% degree of crystallinity) in nature and possesses three available hydroxyl units for each unit within the chain (Alen 2000; Tsoumis 1991).

Hemicelluloses are low molecular weight polysaccharides with degrees of polymerization ranges in hundreds. They are composed of more than one monosaccharide – they are copolymers – such as mannose, xylose, galactose and arabinose. Among them mannose is the most important

hemicellulose monomer in softwoods (Bowyer *et al.* 2003; Walker 2006).

Lignin is the cell wall component that differentiates wood from other cellulosic materials. It always occurs in association with cellulose (Tsoumis 1991). Lignin is a complex, amorphous, high molecular weight substance composed of phenylpropane monomer. The exact structure of lignin is still not clear because it occurs in a variety of forms and is difficult to isolate (Bowyer *et al.* 2003).

**Table 2.4. Chemical composition of wood (%)**

<i>Components</i>	<i>Radiata pine*</i>	<i>Typical Softwoods**</i>	<i>Typical Hardwoods**</i>
Cellulose	40	40-44	42-46
Hemicelluloses	31	25-29	23-33
Lignin	27	25-31	20-28
Extractives	2	1-5	1-7
<i>Ash</i>	0.2	***0.02-1.1	***0.1-5.4

(Uprichard 1991) \* (Walker 2006) \*\* (Tsoumis 1991) \*\*\*

Wood extractives are compounds that can be extracted from wood using polar and non-polar solvents. They are not part of the wood substance, but are subsequently deposited within wood cells and lumens. They have varying chemical compositions such as resins, gums sugars, oils,

starches, fats, alkaloids and tannins. In radiata pine heartwood resin acids are the main extractives and account for 70-80% of total extractives. However in radiata pine sapwood resin acids and fatty acids are found in nearly equal proportion of approximately 41% each (Walker 2006).

## 2.2. Dimensional stability and durability of wood

### 2.2.1. General wood properties of radiata pine

Radiata pine wood can be broadly described as a medium-density, even-textured softwood. Table 2.5 gives some of its physical and mechanical properties.

**Table 2.5. Typical mechanical properties of dry New Zealand radiata pine wood at 12% moisture content . \*** (Bootle 2005; Cown 1992);  
**\*\* (Dinwoodie 2000)**

<i>Property</i>	<i>*Radiata pine</i>	<i>**Softwoods</i>	<i>**Hardwoods</i>
Basic density (kg/m <sup>3</sup> )	400-500	417-769	368-977
Modulus of rupture (MPa)	62-86	72-107	54-181
Modulus of elasticity (GPa)	7.3-8.2	10.2-12.6	5.5-21
Hardness (KN)	2.2-3.6	2.1-4.9	1.9-10.4

The mature radiata pine wood is a versatile, multiple-use wood. The uniform texture makes it easy to saw, peel, pulp or machine. Furthermore, it has good nail and screw holding capacity, it is easy to



dry, treat, glue and finish. This wood is suitable for a variety of uses including sawn wood, furniture, cabinets and the manufacture of wood based panels, pulp and paper (Bootle 2005; FITEC 2007; Harris 1991a; TRC 2007).

However, the fast growing nature of wood and the reduction in the harvesting age to below 30 years affects the quality of timber significantly owing to the higher proportion of juvenile wood<sup>1</sup>. This results in poor dimensional stability, low natural durability and low stiffness of the wood (Jayawickrama 2000; Macalister 1997; Walford 1991). These poor qualities are the main reasons for the modest competitiveness of radiata pine in comparison to most of the commercial species of the world (Bamber and Burley 1983; Dickson and Walker 1997; Maclaren 2002).

### **2.2.2. Dimensional stability**

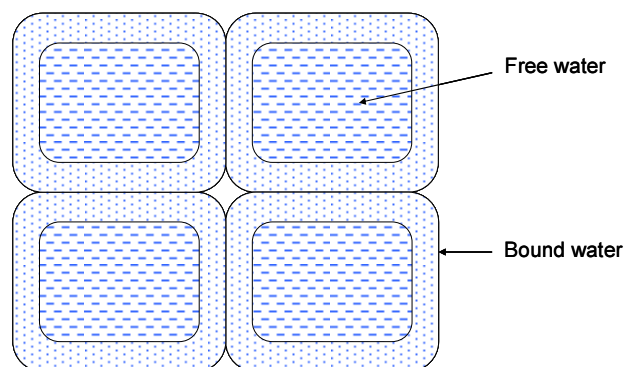
The dimensional stability of wood is an attribute describing its ability to withstand changes in its dimensions under given conditions. Wood is a natural fibrous material and hygroscopic in nature, that is, it attracts moisture from its surroundings and holds it in the form of water or water vapor. The free hydroxyl groups in the cell wall polymers form hydrogen bonds with the environmental moisture.

Hygroscopicity is an important property of wood and affects all physical, mechanical and biological wood properties (Tsoumis 1991). Water in wood exists in two forms (Figure 2.2): (1) free water: the bulk of the

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<sup>1</sup> Wood formed in the early stages of growth, is found adjacent to the pith, characterized by progressive change in cell dimension and a different microstructure from mature wood ASTM-D9 (1992) Standard terminology relating to wood. (ASTM International: West Conshohocken, USA)..

water is contained in the cell cavity and only held by capillary forces. (2)  
 Bound or hygroscopic water: water bound to the wood via hydrogen bonds.



**Figure 2.2. Free and bound water in wood**

**Table 2.6. Shrinkage values (from green to oven dry moisture content)  
 in different directions of wood**

<i>Dimension</i>	<i>Radiata pine*</i>	<i>Softwoods (North American spp)</i>	<i>Hardwoods (North American spp)</i>	<i>European** (Hardwood &amp; softwood)</i>
Longitudinal	0.2-0.25	0.1-0.2	0.1-0.2	0.1-0.6
Radial	2-3.4	2.1-5.1	3-7.9	2.3-6.8
Tangential	4-7	4.4-9.1	6.2-12.7	6-11.8
Volumetric	6.2-10.4	6.8-14	10.2-19.2	8.5-18.8

Source: (Forest Products Laboratory 1999), \*(Black 2005; Bootle 2005; Cown 1992), \*\*(Tsoumis 1991).

As wood dries the free water is drawn away first from the cell lumen and the point at which all of the free water is removed but the cell walls remain completely saturated with bound water is known as fiber saturation point (FSP). At room temperature, it is usually taken as 25 to 30% moisture content based on oven-dry weight (ASTM-D9 1992). Wood changes dimensions as it gains or loses moisture below the FSP. It swells as the moisture content increases and shrinks as the moisture content decreases in response to changes in the temperature and humidity of the surrounding air. Dry wood develops a significant amount of internal stress as it absorbs moisture which can reach a theoretical value of between 91-168 MPa (Rowell 2005). In pre-industrial societies the internal stress of wood has been used to split granite rock.

The shrinkage and swelling of wood are not uniform in different directions of the wood for the same change of moisture content (Table 2.6). Thus wood is anisotropic in nature with regard to changes in its dimensions. These dimensional changes are the least in the longitudinal direction (along the tree trunk) and much greater in the transverse directions. The lower winding angle of the microfibrils in the S2 layer of cell wall causes wood to shrink or swell more in the transverse (radial and tangential) plane than in the longitudinal direction (Bowyer *et al.* 2003; Hill 2006). The longitudinal shrinkage or swelling in mature/outer wood is generally very small. However, in the case of juvenile wood and reaction wood of radiata pine longitudinal shrinkage of up to 2.9% has been observed (Xu *et al.* 2009).

In the transverse direction, shrinkage or swelling is more in the tangential than in the radial direction by a factor of 1.5-3. This is mainly due to the anatomical features of wood such as the presence of ray tissues, frequent pitting on radial walls, microfibril arrangements and earlywood - latewood interaction (Bowyer *et al.* 2003; Pang 2002; Walker 2006).

Figure 2.3 shows the changes in board shape and the distortion on drying within a cross-section of a log below FSP. The growth rings in the board E, F and C form an angle of 45-90 degrees with the wide surface (quarter sawn). These boards show less distortion than boards A, B and D in which growth rings make an angle of less than 45 degrees with the wider surface of the board.



***Figure 2.3. Effect of shrinkage and grain direction in cross section shape (Forest Products Laboratory 1999)***

Dimensional instability is one of the major shortcomings of wood compared with synthetic materials coming from non-renewable resources. Stability is an important factor affecting wood quality and is crucial for the structural market (Walker 1998). It is of prime importance in the use of wood and wood based products, in particular, for applications of high requirements for exact and stable dimensions (Alma *et al.* 1996; Baysal *et al.* 2004). Table 2.7 gives the FRI (Forest Research Institute, Rotorua, New Zealand, now Scion) wood quality selection matrix for the stability.

This indicates the high importance of wood stability for almost all wood based industries except MDF (Medium Density Fiberboards), paper and pulp industries (Meder *et al.* 1995). The instability issue is more important for fast grown plantation species such as radiata pine in comparison to the indigenous, natural grown species owing to a higher proportion of juvenile wood in plantation species. The stability of radiata pine is considered to be inferior and one of the main reasons for its weak market in comparison with other commercial species such as ponderosa pine (Maclaren 2002) and Douglas-fir (Wang *et al.* 2010).

**Table 2.7. FRI wood quality selection matrix for Stability (Meder *et al.* 1995)**

<i>Sector</i>	<i>Priority</i>
Furniture component	3
Dry Structural	3
Finger joint	3
Preservation	2
Plywood	3
MDF	1
Pulp and paper	1

where 3-High priority; 2- Moderate priority; 1- Low priority

Dimensional instability leads to problems in exterior applications of wood products that are subjected to frequent climatic changes (Alma *et*

*al.* 1996; Baysal *et al.* 2004; Devi *et al.* 2004). It significantly decreases the value of wood and limits its application as a result of stability related defects. Anisotropic shrinkage and swelling may induce changes in the cross-sectional shape of wood products, warping and checking. Figure 2.3 shows changes in the shape of various cross-sections and warping due to differential radial and tangential shrinkage. Warping (crook and bow) and twist are identified as the two most significant factors affecting market acceptance of timber (Milliken 2008). Bow is face-to-face variation in longitudinal shrinkage while edge-to-edge variation in longitudinal shrinkage causes crook. Twist is the result of spiral grain.

Dimensional instability is also a major source of structural and visual problems for the furniture industry. Swelling may result in difficulties in opening or closing doors, windows or drawers. On the other hand, gaps in floors and weakening of furniture joints and loosening of tool handles may occur due to reduction in dimensions on drying after installation. Furthermore, it also results in splits in table tops, fractures in frame joints, poor fitting in corner blocks, problems in gluing, a shrunken glue line, poor surface finishing and inferior performance of coatings (Eckelman 2008)

### **2.2.3. Durability of wood**

Durability is a broad term and defined as resistance of wood against biotic factors for example fungi, insects and abiotic factors such as UV radiations (Tsoumis 1991). The resistance against the biotic factors is generally referred as the natural durability of wood which is defined as the inherent resistance of a timber species to decay and insect attack under favourable conditions for such attack (AS5604 2003). These biotic and abiotic external factors may cause degradation of appearance,

structure and chemical composition of wood, limiting its use and can greatly shorten the service life of the wood.

**Table 2.8. Natural durability classification of wood (AS5604 2003)**

<i>Durability Class</i>	<i>In ground life expectancy (years)</i>	<i>In ground life expectancy (years)</i>
1	> 25	> 40
2	15-25	15-40
3	5-15	7-15
4	< 5	< 7

In general sapwood is considered to be non-durable (fewer than 5-10 years service life) and particularly prone to biological attack while heartwood is considered durable mainly owing to deposition of extractives (ranging from 5-30+ service life years). Approximately half of all plantations are less than 20 years old and most of these are established by using fast grown species such as pines, acacias and eucalypts with short harvesting ages (FAO 2001). The management of most of these plantations is focused on maximizing the yield under short rotations rather than the quality. Therefore, fast grown softwood from plantations such as radiata pine is generally characterized by higher proportions of sapwood and poorly developed heartwood. Therefore the durability of plantation timber is inferior to timber from natural growth forest (Hill 2006; Rijckaert *et al.* 2000).

Radiata pine sapwood is highly prone to fungal attack. It is more susceptible to brown rot fungi than attack by white rot fungi (Bamber and Burley 1983; Butcher and Drysdale 1991). Moreover, untreated heartwood severely decayed in contact with the ground in 5 years (Butcher and Drysdale 1991). Therefore radiata pine sapwood is classified as perishable and the heartwood as non-durable. Table 2.8 gives the natural durability classes of wood as per the Australian Standard AS 5604. As per this standard, radiata pine wood is classified under Natural durability Class 4 for both in ground or above ground uses, that is wood of low durability with an expected life of not more than 7 years (AS5604 2003). Therefore, untreated radiata pine wood is at high risk of biological deterioration over a wide range of exposure conditions.

Until recently, traditional biocidal wood preservative treatments have been widely used to improve the durability of timber. Moreover, radiata pine is one of the easiest timbers to treat with preservatives such as CCA and creosote (Bamber and Burley 1983). These preservatives are very cost effective and possess excellent technical qualities required for wood treatment. However, they have come under continual scrutiny for health and environmental safety reasons in many parts of the World, particularly in North America and Europe (Drysdale 2002; Williams 1999). Apart from human health and safety issues, disposal of treated wood after the end of its service life is also a major concern (Syrjänen and Kangas 2001). Such concerns over the use of persistent toxic chemicals as wood preservatives have led to restrictions on their use, in many regions of the world. For example the European Commission Directive No. 2003/02/EC, Copper Chrome Arsenic (CCA) does not allow their use in Europe for residential construction and for all those uses which involve direct human and/or animal contact (Braunschweiler 2002). These restrictions also apply to imported wood and to waste wood containing



preservatives. Furthermore, from 2007-2008 CCA requires authorization according to the Biocidal Products Directives (Hill 2006). Similarly, in the USA, on February 2002, the Environmental Protection Agency announced a voluntary decision by the US timber industry to substitute the sale of CCA treated wood with new alternative wood preservatives by the end of 2003 (Schert 2002). These restrictions severely affected all residential uses of CCA treated wood. Similar legislation has been or will be enforced for the disposal of waste wood treated with persistent toxic chemicals. This will require significant investment to build a plant that can meet the environmental requirements (Hill 2006)

In addition, New Zealand is an important exporter of fruits and organic foods. Some importers such as the USA require the posts used in orchards to be treated with environmentally friendly chemicals rather than CCA (Millen 2009).

These concerns have led to commercial interest in wood modification technologies, including heat-treatment of wood. These technologies aim to improve wood properties without using any persistent toxic chemicals.

### **2.3. Heat-treatment of wood**

Heat-treatment of wood can be defined as the application of heat to wood to bring about desired improvement in the performance of material in an environmentally benign way (Hill 2006). It can play an important role in increasing the competitiveness of fast grown plantation wood by overcoming some of its shortcomings such as dimensional instability and low durability.

The use of heat to improve wood properties is a very old practice. In ancient Africa aborigines used to sharpen and harden their wooden stick ends by heat treating them at the bottom of heated coals followed by

constant beating under the rock. Vikings used to burn the outer edges of ships to make them hydrophobic and fire resistant. Ancient people were aware of the benefits of heat treating wood but were without any understanding of the reasons (Rowell 2006).

The history of heat-treatment or thermal modification of wood as a science is no more than a hundred years old. Since the beginning of the twentieth century, there has been academic interest in different methods of heat-treatment. Tiemann (1917) was one of the pioneers who examined the effects of high temperature on wood stability and mechanical properties. He noted a reduction in moisture sorption and a slight reduction in strength on heating air-dry wood in superheated steam at 150°C for 4 hrs.

Dr Alfred J. Stamm of the Forest Products Laboratory, Madison, USA worked on almost every area of wood modification including heat-treatment during the mid-twentieth century. He can be regarded as the father of wood modification owing to his significant pioneering work in this field. A published report of the US Forest Products Laboratory dated 1916, cited by Stamm and Hansen (1937), mentioned a significant decrease in wood hygroscopicity on heating black gum wood in dry air at 205°C for 6 hrs. Stamm (1956) studied the thermal degradation of wood and wood components. He observed greater degradation of wood by heat in a closed system than in an open system and he also found more severe degradation on heating wood in the presence of air than in the absence of air. Presence of air results in a greater loss of wood mass and mechanical properties owing to oxidation caused by atmospheric oxygen (Stamm *et al.* 1946). Therefore, an oxygen free environment is one of the key requirements for thermal modification to develop stable wood with moderate strength properties.

Owing to environmental and health concerns over uses of certain chemicals in traditional wood treatments, the last decade witnessed significant academic and commercial interest shown in certain wood modification technologies, particularly in the thermal wood modification process (Sándor *et al.* 2006). Several companies particularly in Europe are currently producing thermally modified wood (Table 2.9) for a variety of above ground uses.

**Table 2.9. Annual production of heat treated wood in Europe (Evans 2003)**

<i>Country</i>	<i>No of companies</i>	<i>Annual production (1000 m<sup>3</sup>)</i>
Finland	12	156
France	6	25
Germany	1	2
Netherlands	1	-

These include Thermowood in Finland, the Plato process in the Netherlands, Perdure and Retification in France and hot oil treatment in Germany. The total production capacity of heat treated wood in 2001 was estimated to be 165,000 m<sup>3</sup> (Evans 2003). In all these processes wood is subjected to heat-treatment at temperature ranges from 140 °C to 260 °C for several hours and the main differences are observed in the heating medium and treatment steps (Table 2.10). A temperature of less than 140 °C is insufficient to cause significant change in wood properties and a

temperature above 260<sup>0</sup>C results in severe degradation of wood material (Hill 2006; Militz 2002).

**Table 2.10. Treatment steps and heating mediums of European thermal modification processes**

<b>Process</b>	<b>Main treatment steps</b>	<b>Heating medium</b>	<b>Reference</b>
Thermo wood	High temperature kiln drying followed by intensive heat-treatments and finally cooling and moisture conditioning	Water vapour	(Syrjänen and Kangas 2000)
Platowood	Hydro-thermolysis, an intermediate drying step and curing	Steam or heated air	(Militz 2002)
Retification	Heating of dry wood (12% moisture content) in limited oxygen conditions	Nitrogen atmosphere with < 2% oxygen	(Militz 2002)
Oil heat-treatment	Heating of dry wood (10-12% moisture content) in vegetable oil in a closed vessel	Vegetable oil	(Sailer <i>et al.</i> 2000a)

There are several reports available on the heat-treatment of wood in varying air, vacuum, and steam or nitrogen environments in which solid wood is subjected to temperatures close to or above 200<sup>0</sup>C for several hours (Ahajji *et al.* 2009; Bekhta and Niemz 2003; Christmas *et al.* 2005;

Giebeler 1983; Syrjänen and Kangas 2000; Tjeerdsma *et al.* 1998; Vidrine *et al.* 2007; Viitanen *et al.* 1994). These studies reported a general increase in dimensional stability and biological durability. However, the mechanical properties were inferior, and as the colour of the wood faded the surface checks appeared in treated wood on exposure to UV radiation (Ahajji *et al.* 2009; Militz 2002; Syrjänen and Kangas 2000).

## **2.4. Thermo-mechanical compression (TMC) of wood**

Thermo-mechanical compression is an extension of thermal modification resulting in densified wood with improved mechanical properties. Wood can be compressed thermo-mechanically under plasticizing conditions without any structural breakdown: the moisture in wood and heat act as plasticizing agents (Seborg and Stamm 1941).

Thermo-mechanical compression (TMC) of wood increases the wood density by reducing the void volume, thus improving mechanical properties. This process has been studied since the mid 20<sup>th</sup> century (Seborg *et al.* 1945; Seborg and Stamm 1941; Seborg *et al.* 1962; Stamm *et al.* 1946). Densified wood has found applications in the manufacture of specialized wood products such as textile shuttles, bobbins and tool handles. Commercial manufacture of densified wood started in Germany before World War II under the trade name of *Lignostone* -compressed solid wood, *Lignofol*- compressed laminated wood and *Kunstharzschichtholz*- resin treated laminated compressed wood (Stamm 1964).

However, dimensional instability is a major drawback of compressed wood. Shrinkage and swelling of wood are generally proportional to the

amount of water absorbed and removed by the cell wall, which implies that the higher the density the greater the instability (Bowyer *et al.* 2003), therefore compressed wood is dimensionally unstable. When exposed to a high humidity environment compressed wood exhibits significant swelling and permanent spring back. This phenomenon is known as compression set recovery or shape memory (Kamke 2006). Therefore, restraining spring back is one of the main challenges to the use of densified wood.

## **2.5. Linseed oil and its use in heat-treatment**

### **2.5.1. Vegetable oils**

Vegetable oils are a mixture of triglycerides, usually liquid at room temperature, obtained from different plant parts mainly seeds or beans. The value and uses of different vegetable oils depend on the composition of their constituent fatty acids, particularly the length of the carbon chain, the degree of saturation, and the nature and position of functional groups. More than 40 species have been used for the commercial production of vegetable oils of which only nine are commercially important: palm, coconut, soyabean, palm kernel, groundnut, cottonseed, linseed, sunflower and rapeseed (FAO 2005a). These oils can be divided into three major groups (CEC 1973): (1) Edible or soft oils - oils used mainly for edible purposes, e.g. groundnut, soybean, cottonseed, sunflower, sesame, olive and rapeseed; (2) Edible-industrial or hard oils - oils used for industrial as well as for specialized food uses such as margarine, e.g. palm, palm kernel and coconut oils; (3) Industrial or drying or technical oils - oils mainly used as drying agents, lubricants or for other industrial purposes, such as castor, tung or wood oil and linseed.

Linseed oil is an important drying oil obtained from the seeds of the flax plant *Linum usitatissimum* (Figure 2.4). The oil content of flax seeds is 33-47% which is more than the average oil content of all oilseeds which is typically around 26% (Gunstone 2002).

### **2.5.2. Linseed crop**

The flax plant is an annual herb with erect stems. The crop is believed to originate from the Mediterranean and Southwest Asian regions (BIOMETNET 2007) and linseed has been grown for fibres and seeds which are rich in oil since ancient times. *Linseed*, *flax* and *dual purpose flax* are the three varieties of this species (Gunstone 2002). The *Flax* variety crop is 80-120 cm in height and mainly grown for textile fibers. The *Linseed* variety crop is 60-80 cm in height and is mainly grown for seeds. It is termed *Linseed* in India and the United Kingdom, *Flaxseed* in North America and *oil flax* or *seed flax* in many European countries. The crop variety grown for both seeds and fibre is known as *dual-purpose flax*. The *linseed crop* has more capsules and bigger sized seeds than the *flax crop*. The seeds are oval, flattened, shiny, and pale to dark brown in colour and 4-6mm in length (Figure 2.4).

The New Zealand flax plant is very distinct from the flaxseed plant (*Linum usitatissimum*). New Zealand flax is obtained from perennial plants mainly *Phormium tenax* and *Phormium cookianum*, known by the Māori names *harakeke* and *wharariki* respectively. These plants are associated deeply with the culture, history, and economy of New Zealand. They produce long leaf fibers and were traditionally used by Maori people for clothing, fishing nets and daily household items. They are also used as medicine and as paper fiber. Flax seed oil from New Zealand flax is subject to research and development and is not available commercially (DOC 2007).

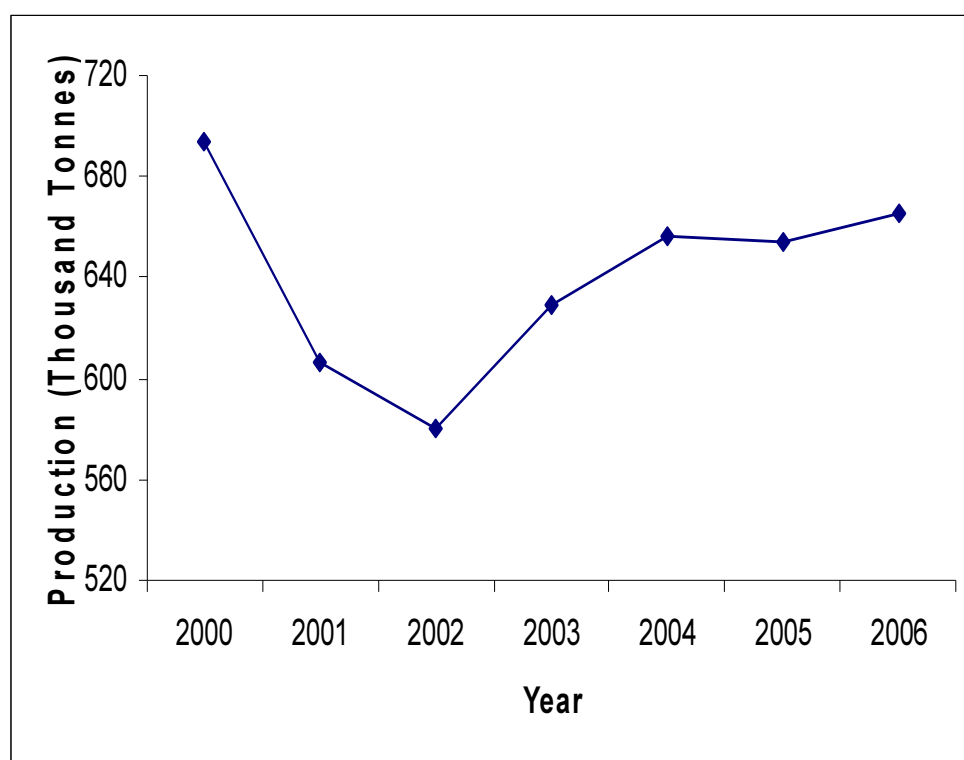


*Figure 2.4. Linseed plant (top) (BIOMETNET 2007) and seeds of flax crop(bottom)*

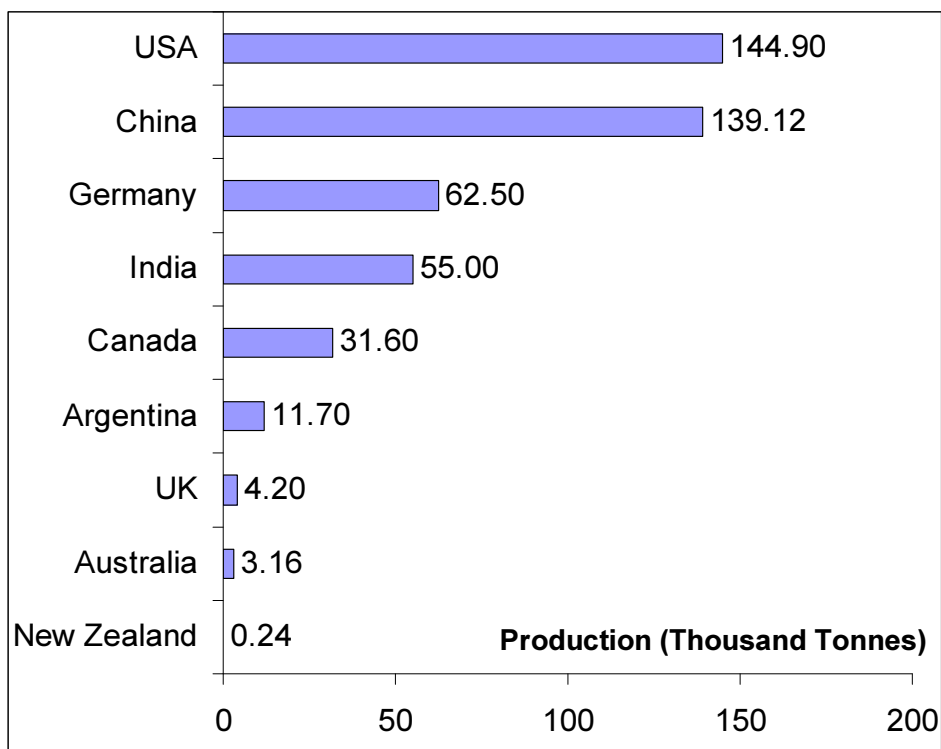


### 2.5.3. Linseed oil production

The linseed crop is grown in tropical as well as in temperate regions and the global production of linseed oil was around 665,000 tonnes per annum during 2004-2006 as shown in Figure 2.5 (FAO 2008). USA and China produce almost 43% of global production producing 21.8% and 20.9% respectively (Figure 2.6). Argentina, Australia, Canada, Germany, India, Pakistan and UK are other major linseed oil producers. Linseed oil production in New Zealand was reported to be 241 tonnes in 2006.



*Figure 2.5. Global linseed oil production during 2000-2006, Source (FAO 2008)*



*Figure 2.6. Linseed oil production in New Zealand and major producing countries in 2006 Source (FAO 2008)*

#### **2.5.4. Chemical composition**

The fatty acid composition of linseed and other vegetable drying oils is dominated by C18 fatty acids. Table 2.11 lists the fatty acid composition of the main vegetable drying oils. The most common fatty acids in drying oil are (a) saturated acid - lauric, myristic, palmitic and stearic acids (with 12, 14, 16 and 18-carbon atoms respectively) and (b) 18-carbon polyunsaturated acids - oleic, linoleic, linolenic acids (Lazzari and Chiantore 1999; Turner 1987). Linolenic, linoleic, and oleic acids are also known as omega fatty acids and the high levels of polyunsaturated fatty acids found in these oils aid in the drying of oil when applied on a surface in thin film. The drying ability of these oils depends on the chemical reactivity of triglyceride molecules arising from the double

bond of the unsaturated acids which helps in oxidation and polymerization.

**Table 2.11. Drying oil with fatty acid composition, % weight,** (Gunstone 2002; Lazzari and Chiantore 1999; Stenberg *et al.* 2005)

<i>Fatty acid</i>	<i>*C: D</i>	<i>Linseed</i>	<i>Walnut</i>	<i>Poppyseed</i>	<i>Tung</i>
Linolenic acid	18:3	48-60	2-16	5	3
Linoleic acid	18:2	14-19	57-76	72	15
Oleic acid	18:1	14-24	9-30	11	11
Palmitic acid	16:0	6-7	3-7	10	3
Stearic Acid	18:0	3-6	0.5-3	2	2
Elaeostearic acid		-	-	-	59

\* C = Number of carbon atoms; D = Number of double bonds

Linolenic acid content is 48-60% in the linseed oil which is very high in comparison to other drying oil (Table 2.11). The high linolenic acid content makes linseed oil an excellent drying oil. It can be converted to an edible oil by reducing the linolenic acid (C18:3) content from 50 to 2% and increasing the linoleic acid content to 65-75% through plant breeding (Table 2.12). These changes increase the oxidative stability of linseed oil.

**Table 2.12. Fatty acid composition (percentage values) of edible and non-edible linseed oil (Gunstone 2002)**

	<i>Non-edible linseed oil</i>	<i>Edible linseed oil</i>
Oleic acid	14-24	17-21
Linoleic acid	14-19	64-70
Linolenic acid	48-60	2

### **2.5.5. Grades of linseed oil**

Linseed oil is available in three main commercial grades: raw, refined and boiled linseed oil (Handyman 2008).

#### *(a) Raw linseed oil*

This is the raw oil obtained from the seeds without further treatment. It is pale yellow in colour with a distinctive odour. The oil dries very slowly and takes weeks to fully cure. It is generally used as a protective layer in outdoor uses such as decking and fences.

#### *(b) Refined linseed oil*

Refined linseed oil is the raw linseed oil without free fatty acids, gums and other extraneous material. It dries faster than raw oil but more slowly than boiled linseed oil.

*(c) Boiled linseed oil*

This is linseed oil with some additive materials such as manganese oxide and is light to dark brown in colour. The additive materials are incorporated to improve the drying qualities of the oil. This oil dries more quickly than raw and refined linseed oil and becomes thicker, denser and darker when it is dry. It is more expensive than raw linseed oil and is generally used to treat specialized wooden products such as cricket bats, antique furniture.

**2.5.6. Physical properties**

Raw linseed oil is pale yellow to amber in colour with a strong and distinctive odour. The oil is immiscible and floats in water. When spread on the surface it quickly forms a thin layer with good optical and mechanical properties. The main physical properties of the raw linseed oil are listed in Table 2.13.



*Figure 2.7. Raw Linseed oil colour and appearance*

**Table 2.13. Main physical characteristics of raw linseed oil**

<i>Property</i>	<i>Value</i>
Specific Gravity	0.92-0.93
Melting range	-19 °C
Boiling ranges	>340 °C
Auto ignition point	343 °C
Flash point	222 °C
Volatile component (% volume)	< 0.5
Iodine value	177
Saponification value	188-195
Unsaponifiable matter (maximum)	1.5 %

Source: (Chemwatch 2007; Gunstone 2002) (ASTM-D234-82 1998)

### **2.5.7. Uses and application in heat-treatment**

Linseed oil is one of the main industrial vegetable oils. Since the 15th century it has been used as a base in the paint industry (Maugh 2008). This oil is an excellent drying agent and used in many other products such as resins, inks, soaps, varnishes, wood treatments, soap and the pharmaceutical industry (Ulvcrona *et al.* 2005). It has a tendency to readily take up oxygen from the air and changes to a relatively hard,

tough and elastic substance when exposed as a thin film to air (ASTM-D16-03 2007). It has good auto-oxidative properties and can harden without any modification or addition of siccatives which are chemical compounds used to accelerate the drying process and improve the performance of coatings (Treu *et al.* 2001). The hardening of linseed oil is a very complex process and generally is considered to be a result of auto-oxidation followed by polymerization (Lazzari and Chiantore 1999). The high linolenic acid (C18:3) content of linseed enables the oil to dry fast and harden in sunlight.

The requirement of a low oxygen environment for heat-treatment creates scope for using vegetable oils such as linseed oil for the thermal modification of wood (Sailer *et al.* 2000b). The oil not only serves as the heat transfer medium but also excludes oxygen from the treatment specimens. Linseed oil provides an ideal medium for controlled heat-treatment of wood and gives an oxygen free environment to heat-treat the wood without any charring. The boiling range of this oil is above 334°C which is higher than that required for the heat-treatment of wood (Chemwatch 2007). Being a vegetable oil it has good environmental characteristics. It is inherently biodegradable, of low eco-toxicity and toxicity to humans, derived from renewable resources, and has no net carbon dioxide contribution to the atmosphere. The oil is also widely available in tropical as well as temperate countries. Linseed oil is a relatively inexpensive resource for drying oil (Stenberg *et al.* 2005). It can be thinned with organic solvents such as kerosene, mineral turpentine, xylene, toluene or even petrol or diesel. Before the advent of modern preservatives it was commonly used as a stand-alone preservative for wood (NNFC 2007). The boiled linseed oil is also used for the coating of cricket bats.

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## Chapter 3

# EXPERIMENT: MATERIALS, TREATMENTS AND METHODS FOR ANALYSIS

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### 3.1. Introduction

This chapter first describes the materials and specimen preparation for the experiments and then gives a brief account of wood treatments. Full details of the treatments are given in the following chapters. Finally, it describes the methodology used for measurement and analysis of the properties of treated and untreated specimens.

New Zealand grown radiata pine wood was selected as the experimental material. The main reasons for the selection of the material were: (1) the species is one of the major fast grown species of the Southern Hemisphere; (2) it is of significant importance in the New Zealand economy; (3) it is identified as a species with poor stability, low durability and variable mechanical properties; (4) no work has been reported on the oil heat-treatment of this species, and (5) it is easily available in the region.

The treated and untreated wood specimens were tested to quantify and evaluate the changes in wood properties. The 'treated specimens' mentioned in this chapter were the wood specimens subjected to either

(1) oil heat treatment; or (2) densification followed by oil heat treatment. The untreated specimens were the wood specimens that had received no treatment and were used as a 'control' to compare the effect of treatments.

The tests were performed on the end matched specimens according to the methods outlined in the relevant BSI (British Standard Institute) standards; ASTM (American Standard Testing and Materials) standards, TAPPI (Technical Association of Pulp and Paper Industries) standards, NZS (Standards New Zealand) or reported in the literature. The properties measured in treated and untreated (control) specimens were colour change, weight percentage change, dimensional stability, water repellency, durability and mechanical properties. The changes in colour and dimensional stability were also examined by an accelerated weathering test. The scanning electron microscope (SEM) was used to examine structural changes after treatments. Chemical changes in wood after treatments were examined by Fourier Transform Infrared spectrometry (FTIR) and wet chemical analysis.

## **3.2. Materials**

### **3.2.1. Linseed oil**

The raw linseed oil used in this study was obtained from a local supplier M/S Mainland Paint and Printing Ink Ltd, Christchurch, New Zealand and used as received. The unprocessed raw oil was manufactured by pressing flaxseed without any treatment or addition of chemicals. The colour of the oil was yellowish-brown with a very distinctive odour (Figure 3.1).



*Figure 3.1. Raw linseed oil used in this study for the treatments*

### **3.2.2. Wood specimens**

The specimens were prepared from 3-4 m long boards of radiata pine (*Pinus radiata* D. Don) which had dimensions of 90 mm (width or tangential) by 20-35 mm (thickness or radial). All of the boards came from a local saw mill M/s Sutherland Timber Ltd, Kaiapoi, New Zealand. They were of the same commercial grade, of dry sapwood (10-12% moisture content), flat sawn with growth rings parallel to the flat surface.

From these boards, control and treatment specimens with dimensions 200-300 mm (longitudinal) x 90 mm (tangential) x 20-35 mm (radial) were prepared as shown in Figure 3.2. From each board 5 mm sections were trimmed off from each end. The specimens were then oven dried at  $105 \pm 2^{\circ}$  C for 48 hrs, cooled over silica gel dessicator and weighed. Finally, the specimens were placed in a conditioning chamber at 65% RH

and  $20 \pm 2^\circ\text{C}$  for 2-3 weeks till the change in the specimens' weight was less than 0.1%.



*Figure 3.2. General cutting pattern of the specimens from a board where: E represents the trimmed ends; T represents the treatment specimens and C the control (untreated).*

### 3.3. Overview of wood treatments

The wood specimens which had been conditioned at 65% RH and  $20 \pm 2^\circ\text{C}$  were treated by one of following methods:

1. Wood specimens were treated in a hot oil bath of linseed oil at various temperatures and elapsed times. The details of the equipments and treatment procedures are discussed in Chapter 4.
2. Wood specimens were first compressed in a mechanical press and then heat-treated in an oil bath at the pre-set temperature. The details of this treatment are given in Chapter 5.

## 3.4. Methods of analysis

With the above mentioned treatments in Section 3.3, the end matched treated and untreated (control) specimens of a specific treatment were examined for: (1) physical properties: colour change and weight change after treatment; (2) moisture related properties: dimensional stability, water repellency; (3) mechanical properties: strength, stiffness and hardness; (4) durability: fungal resistance, UV weathering; (5) structural changes, and (6) chemical changes. Additional tests or changes in the procedure (if any) adopted for a specific property are described in the relevant chapter.

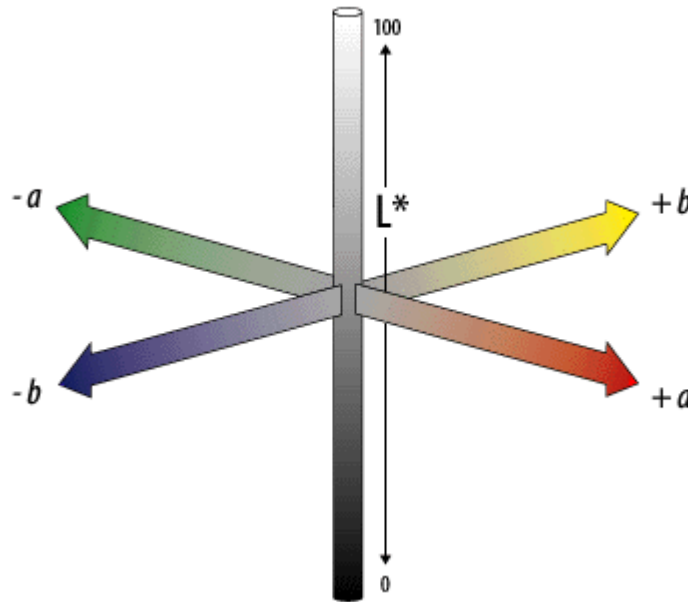
### 3.4.1. Colour

Variation in wood colour after the treatment and accelerated weathering tests (described in Section 3.4.7) was examined by visual inspection and quantified through CIE (Commission internationale de l'éclairage) 1976 (L\*a\*b) system which is a well established procedure for colour measurement (Ahajji *et al.* 2009; Esteves *et al.* 2008; St-Onge *et al.* 2005). This system measures the total colour change ( $\Delta E$ ) from the changes in three spatial coordinates using the following equation (BS3900-a 1986; BS3900-b 1986):

$$\Delta E = \sqrt{(\Delta L^*)^2 + (\Delta a^*)^2 + (\Delta b^*)^2} \quad (3.1)$$

where L\* is the brightness coordinate or luminance ranging from black (0) to white (100); a\* is the colour coordinate varying from redness (+) to

greenness (-), and  $b^*$  is colour coordinate varying for yellowness (+) to blueness (-) (Figure 3.3).



**Figure 3.3. CIELAB Coordinate System (ADOBE 2008) showing the colour changes in three coordinates ( $L^*$ ,  $a^*$  and  $b^*$ )**

Colour change in wood specimens was recorded by a Minolta spectrophotometer of model *cm2500d*. The diameter of the colour measuring circle on the specimen surface was 10 mm with a corresponding area of 0.78 cm<sup>2</sup>. The light source was the Standard Illuminant D 65 which represents the average day light including UV region and 10° standard observer. For the colour measurement, the specimens were first stabilized at 20 ± 2 °C and 65% RH before the measurements. For each specimen, six measurement points were marked and the measurements were taken both before and after the treatment or accelerated weathering. The coordinates  $L^*$ ,  $a^*$ ,  $b^*$  values of the specimens before the treatments or the accelerated weathering test were

used as the reference values to determine the colour changes with the treatment or the accelerated weathering.

### 3.4.2. Weight Percentage Change

The weight percentage change (WPC) in the specimens after the treatments were measured on the oven dry basis. Both before and after the treatment, the specimens were oven-dried at  $105 \pm 2$  °C for 48 hrs, cooled over a silica gel dessicator and weighed with an accuracy of  $\pm 0.001$  g. WPC was then determined using Equation (3.2) which gives an averaged relative change in the oven-dry weight of the tested specimens through the treatments. Six to ten replicates of treated and untreated specimens were used to determine the WPC.

$$WPC = \left[ \frac{(W_a - W_b)}{W_b} \right] \times 100\% \quad (3.2)$$

where  $W_b$  is the oven-dry weight of specimens before the treatment (g), and  $W_a$  is the oven-dry weight of specimens after the treatment (g).

### 3.4.3 Density

Densities of treated and untreated specimens were determined on the oven-dry ( $105 \pm 2$  °C) basis using Equation (3.3). The dimensions and weights of test specimens were measured to precision of  $\pm 0.03$  mm and  $\pm 0.001$  g, respectively.

$$D_{od} = \left( \frac{W_{od}}{V_{od}} \right) \quad (3.3)$$

where  $W_{od}$  is the oven dry weight in grams and  $V_{od}$  the oven dry volume in  $\text{cm}^3$ .

### **3.4.4. Dimensional stability and moisture repellency**

#### *3.4.4.1. Testing conditions*

Moisture related properties (dimensional stability and water repellency) were measured by exposing treated and untreated specimens to either of following sets of conditions.

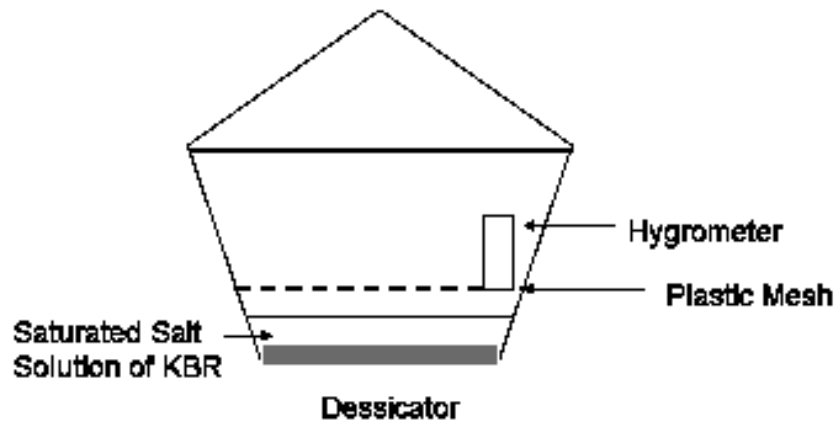
##### **3.4.4.1.1. Water immersion test**

Specimens were immersed in a water bath at  $20 \pm 2$  °C for varying periods of time up to three-four weeks. The water was circulated throughout the test and replaced every 24 hours. The samples were weighed and their dimensions were measured in longitudinal, tangential and radial directions at preset elapsed times during the test.

##### **3.4.4.1.2. High humidity test**

Wood specimens were placed in a high relative humidity condition at  $20 \pm 2$  °C for varying periods of time. The desired relative humidity environment was achieved by using a saturated salt solution at a controlled temperature (ASTM-D4993-91 1992; Martin 1962). A saturated salt solution of potassium bromide (KBR) in a sealed dessicator at  $20 \pm 2$  °C maintained the humidity of  $85 \pm 5\%$  RH (Figure 3.4). The temperature and RH were continuously monitored by using a temperature and RH data recorder and by a handheld humidity meter.





*Figure 3.4. Set up of a humidity chamber for high humidity conditions in the stability test*

#### 3.4.4.1.3. Dry-freeze-wet (DFW) cycle test

Wood specimens were exposed to three cycles of dry-freeze-wet conditions. Each cycle consisted of three days of dry conditions in an oven at  $35 \pm 2^\circ\text{C}$  followed by a day of a freezing at  $-20^\circ\text{C}$  and three days of water immersion at  $20 \pm 2^\circ\text{C}$ .

#### 3.4.4.2. Determination of dimensional stability and moisture repellency

Before the tests the oven-dry weight and dimensions of the specimens were recorded. In water immersion and high humidity tests, the specimens were weighed at different intervals. Once the weight changes of the test specimens were less than 0.1%, it was assumed that the specimens had reached equilibrium. Then the specimen dimensions in longitudinal, width (tangential) and thickness (radial) directions were measured. In DFW cycle tests the weight and dimensions of the

specimens were measured after the end of three cycles of 21 days. The weight was measured with an accuracy of  $\pm 0.001$  g and the accuracy for the dimension measurement was  $\pm 0.01$  mm. Eight to ten replicates of untreated and treated specimens were used for each test.

Water repellence efficiency (WRE) or moisture excluding efficiency (MEE) was determined using the following equation:

$$WRE \text{ or } MEE = \left( \frac{W_c - W_t}{W_c} \right) \times 100\% \quad (3.4)$$

where  $W_c$  is the percentage of water/moisture uptake by the untreated specimens (%) and  $W_t$  is the percentage of water/moisture uptake by the treated specimens (%). Both of the uptake percentages were determined from the oven-dry weight specimens and the specimen weight after wetting as given in Equation (3.5). A positive, high value of *WRE* or *MRE* means that the treated wood absorbs less water/moisture than the untreated wood thus indicating an improvement in terms of reduction in the water/moisture uptake through the oil heat treatment.

$$W_c \text{ or } W_t = \left( \frac{W_2 - W_1}{W_1} \right) \times 100\% \quad (3.5)$$

where  $W_1$  is oven dry weight of the specimens before test (g) and  $W_2$  is the weight of the specimens after testing for a specific period (g).

In a similar way, the anti-swelling efficiency (ASE) was determined from Equation (3.6) given below (Hill 2006; Rowell and Banks 1985; Stamm 1964):

$$ASE = \left( \frac{S_c - S_t}{S_c} \right) \times 100\% \quad (3.6)$$

in which  $S_c$  is the volumetric swelling of untreated specimens and  $S_t$  is the volumetric swelling of the treated specimens. They were determined by the following equation:

$$S_c \text{ or } S_t = \left( \frac{V_2 - V_1}{V_1} \right) \times 100\% \quad (3.7)$$

where  $V_1$  is the oven-dry volume of the specimens before the test ( $\text{cm}^3$ ) and  $V_2$  is the volume of the specimens after the test ( $\text{cm}^3$ ). Once again, the positive and high value for  $ASE$  means that the treatment reduced the volumetric swelling significantly. Anti-swelling efficiency was also determined in tangential and radial directions using a similar equation to Equation (3.6) as follows:

$$ASE \text{ (tangential or radial)} = \left( \frac{D_c - D_t}{D_c} \right) \times 100\% \quad (3.8)$$

where  $D_c$  is the percentage swelling in the tangential or radial direction of untreated specimens and  $D_t$  is the percentage swelling in the tangential or radial swelling of the treated specimens .

#### **3.4.5. Biological durability**

The biological efficacy of the treatments was evaluated by the Wood Mycology Laboratory at Scion, Rotorua, New Zealand. A pure cultural decay trial was performed using an agar block decay test as per the

standard “Sutter jar” method (Sutter 1978). This method is based on the same principles as those used in the European Standards EN-113 and British Standard BS 6009:1982 (Callahan and Chittenden 2009). However it differs in exposure time to the decay organism and uses the plastic Petri dish containers instead of glass Kolle flasks (Stahlhut *et al.* 2008).

Eight replicates of treated and untreated specimens with dimensions of 35 mm (tangential) x 20 mm (radial) x 7 mm (longitudinal) were used. The specimens were first leached for two weeks and air-dried for several days. They were then oven-dried at  $102 \pm 2^{\circ}\text{C}$  for 18 hrs and weighed after cooling down in silica gel desiccators. After this, the specimens were sterilized by ethylene oxide gas. Finally, the specimens were placed into prepared Sutter agar containers using standard “Sutter trial” procedures. The incubation was at  $26^{\circ}\text{C}$  and 75% RH for six weeks. The test fungus used was *Oilgoporus placenta* a common brown rot fungus found in leaky buildings in New Zealand (Stahlhut *et al.* 2008).

After incubation, specimens were cleaned, oven-dried and weighed. Weight loss percentages (WL) of the specimens were calculated from the oven-dry weight of the specimens before the incubation ( $M_b$ ) and after the incubation ( $M_a$ ) by using the following equation:

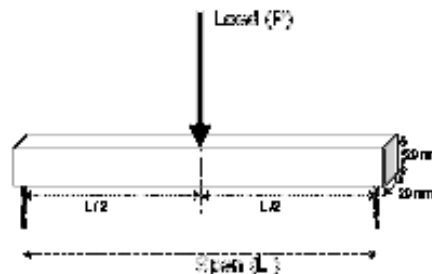
$$WL = \left( \frac{M_b - M_a}{M_b} \right) \times 100\% \quad (3.9)$$

### 3.4.6. Mechanical properties

#### 3.4.6.1. Strength and Stiffness

Strength and stiffness in bending are generally expressed as modulus of rupture (MOR) and modulus of elasticity (MOE), respectively. These properties were determined by three-point bending as per the procedure mentioned in British Standard BS 373.

Samples for the bending tests were prepared with cross dimensions of 20x20 mm (in perpendicular to longitudinal direction) from both treated and untreated specimens (Figure 3.5). The surfaces were dressed and the dimensions and moisture content of specimens recorded. The specimens were conditioned at 65% RH at 20 °C for two weeks before testing.



*Figure 3.5. A small bending sample and loading based on BS373*

Testing was performed in the ambient condition at  $20 \pm 2$  °C and 65% RH using a standard Material Testing System (810 MTS machine). The orientation of the growth ring in test specimens was parallel to the direction of loading. The loading head was moved at a speed of 6.6 mm per minute. Fifteen replicates of untreated and treated specimens were

used for the test. The MOR and MOE were calculated using the Equation 3.10 and 3.11 respectively (BS373 1957):

$$MOR = \frac{3P_{max} \cdot L}{2b \cdot h^2} \quad (3.10)$$

$$MOE = \frac{P \cdot L^3}{4\Delta \cdot b \cdot h^3} \quad (3.11)$$

In Equations (3.10) and (3.11),

$b$  is the breadth of the test specimen (mm),

$h$  is the depth of the test specimen (mm),

$L$  is the supporting span (mm),

$P_{max}$  is the maximum load when the beam is broken (N),

$P$  is the load in N within the proportional deflection (N),

$\Delta$  is the deflection at mid-length below the proportion deflection limit (mm).

#### 3.4.6.2. Hardness

The hardness of untreated and treated wood was determined by the Janka indentation test method in accordance with the British Standard BS 373.

In this test hardness is the load required to force the hemispherical end of a steel bar into the test specimen, to a depth of half of its diameter, i.e. 0.222" or 5.64 mm (BS373 1957). For the hardness tests, fifteen test specimens with the same cross dimensions as those in the bending tests (20 x 20 mm) were prepared for untreated and treated wood. The specimens were conditioned at  $20 \pm 2$  °C and 65% RH for two weeks. The testing was under ambient conditions of  $20 \pm 2$  °C and 65% RH using a standard Material Testing System (810 MTS machine). The rate of penetration of the hardness tool was 6.35 mm per minute. On each specimen, four penetrations were made, two penetrations on the radial faces and two on the tangential faces (Figure 3.6).



Figure 3.6. Janka indentation test at the MTS machine

### **3.4.7. Accelerated Weathering tests**

Accelerated weathering for both the treated and the non-treated specimens was conducted according to US standard ASTM G 154 (ASTM-G154 2006). Wood specimens were first cut to dimensions of 90 mm (tangential) x 20 mm (radial) x 200 mm (longitudinal). Then, each specimen was further cut into two halves along the mid-thickness along the specimen length thus each half had dimensions of 90 mm (tangential) x 10 mm (radial) x 200 mm (longitudinal). One half of the specimens were exposed for accelerated weathering and the other half were kept unexposed in a dark room for comparison. Test specimens were exposed to repetitive cycles of UV light and moisture under controlled environmental conditions in a weathering box (Figure 3.7). UVA 340 light was used and the moisture effect was produced by spraying the specimens with water. All the specimens were kept 100 mm from the light source. The specimens were exposed for 2100 hrs in cycles of 8 hour UV 340 at  $60 \pm 3^{\circ}\text{C}$  black panel temperature and 4 hr condensation at  $50 \pm 3^{\circ}\text{C}$  black panel temperature (ASTM-G154 2006). During the 4 hour condensation period water was sprayed for 15 seconds in every 2 minutes.

The changes in colour and stability properties were measured after the test. The measurements of colour by spectrophotometer were made at regular intervals after 250, 500, 750, 1000, 1250, 1500, 1750 and 2100 hours.





*Figure 3.7. Accelerated UV weathering system*

#### **3.4.8. Scanning electron microscopy (SEM)**

The microstructures of cross-sections of the treated and untreated specimens were examined using the Leica 440 Scanning Electron Microscope (SEM). The SEM was operated at a voltage of 20 kV EHT and a filament current of 2.8 A. Specimens were first trimmed to small blocks and boiled for 2-3 minutes. Then the specimens were cross-cut precisely using a new blade for each surface. Then the surfaces were sputter coated with gold by Emitec K550-X sputter coater for 2 minutes at 20mA and 1.2 KV. The scanning data were analyzed at 500x, 1000x and 5000x magnifications. Six to seven SEM images were taken and analyzed for each specimen.

### **3.4.9. Fourier Transform Infrared (FTIR) spectrometry**

The infra red spectra of treated and untreated specimens were examined by a Perkin Elmer System 2000 FTIR spectrometer. The wood samples were powdered and screened through a 12 mesh sieve (0.12mm). The extractives were removed as per the procedure in the TAPPI Standards 264 (TAPPI-264 1995). The powdered samples were vacuum-dried at  $70 \pm 3^\circ\text{C}$  for 48 hrs. The dried samples were mixed with spectral grade potassium bromide (KBr) and compressed in pellets by a hydraulic press. These pellets were analyzed in the absorption mode in the range of 900-1800  $\text{cm}^{-1}$  with an accumulation of 64 scans at the resolution of 4  $\text{cm}^{-1}$ . The FTIR spectra are baseline-corrected and normalized at the absorption band of maximum and minimum spectra according to the procedure given in the manual of Essential FTIR analysis software.

### **3.4.10. Wet chemical analysis**

The chemical composition of treated and untreated wood was quantitatively determined by wet chemical analysis. The wood samples were ground to a sieve size of 25-40 mesh (0.25-0.4mm). The ethanol-benzene (1:2) mixture and hot water were used to remove the polar extractives and water soluble extractives as per the procedure mentioned in the TAPPI Standard(TAPPI-264 1995).

In extractive-free wood, acid insoluble lignin or klason lignin percentage was calculated according to the TAPPI Standard (TAPPI-T222 1988). About 2.5 g of extractive free wood sample was used to determine the content of holocellulose which includes both cellulose and hemicelluloses, according to the sodium hypochlorite method (Rowell

2005). In the holocellulose fraction,  $\alpha$ -cellulose content was determined by the sodium hydroxide method (Rowell 2005). The hemicellulose content was determined by subtracting the  $\alpha$ -cellulose fraction from the holocellulose content. All the reported percentages of chemical constituents were an average of three replicates to the nearest of  $\pm 0.1\%$ .

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# Chapter 4

## OIL HEAT-TREATMENT

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### 4.1. Introduction

Wood can be and has been heat-treated in air, nitrogen atmosphere, presence of water and superheated steam. Heat-treatment of wood in a controlled environment can be an effective way of altering wood to achieve the desired properties improvements in fast grown plantation wood. The maximum temperature used for thermal modification processes of wood is generally no more than 260°C (Hill 2006). Thus, vegetable oils can be a suitable heat transfer media for thermal modification since the boiling points of many vegetable oils are higher than 260°C (Gunstone 2002). However, there is very limited published work on the use of vegetable oils for the thermal modification of wood (Sailer *et al.* 2000a; Wang and Cooper 2005b; Welzbacher *et al.* 2005).

Stamm *et al.* (1946) were the first to use a liquid medium for the thermal modification of wood. They heat-treated wood over a range of temperatures from 160-320°C in a bath of molten metal and developed dimensionally stabilised wood termed Staybwood. The degree of reduction in hygroscopicity and anti-swelling efficiency (ASE) almost doubled with every 10<sup>0</sup>C rise in the temperature range tested. Recently, the Menz Holz Process was developed in Germany in which wood is heat-treated at 180-220<sup>0</sup>C in refined rapeseed and linseed oil in a closed process vessel (Sailer *et al.* 2000a; Sailer *et al.* 2000b). Welzbacher and Rapp (2002) found superior decay resistance of wood which was heat-

treated in oil compared to that heat-treated in air under identical treatment conditions. It is also reported that the stiffness of oil heat-treated wood was better than that of the wood heat-treated in air (Hill 2006). Tjeerdsma *et al.* (2005) examined the heat-treatment of wood using modified (maleinised) linseed oil, while Wang and Cooper (2005) compared the dimensional stability and water repellency of wood which was heat-treated in different types of oils including palm oil, soy oil and slack wax. Wang and Cooper (2005) have noted that the performance of oil heat-treated wood is affected by the oil type used. The properties of heat-treated wood are also species-dependent owing to anatomical differences among species (Hill 2006). Manalo and Acda (2009) reported an improvement of wood in water absorption and thickness swelling properties for three bamboo species which were heat-treated at 160-200°C in coconut oil. In comparison to Europe, the thermal modification process is less commercialised in New Zealand and the availability of significant quantities of radiata pine wood from sustainably managed plantation opens the opportunity for future commercial ventures.

In this study radiata pine was heat-treated in a linseed oil bath. The treated wood was then investigated for changes in physical, moisture related, biological and mechanical properties resulting from the treatment. In addition, changes in the microstructure and chemistry of the treated wood were examined.

## **4.2. Experimental**

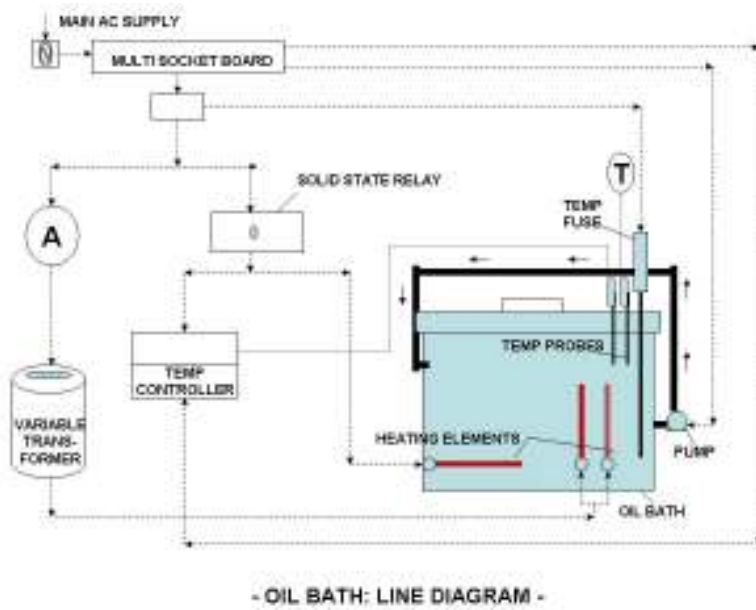
The wood specimens of 300 mm (longitudinal) x 90 mm (tangential) x 20 mm (radial), which were prepared as described in Chapter 3, were firstly conditioned at 65% RH and 20°C in a controlled chamber with

corresponding equilibrium moisture content ranging 10% to 12%. After the equalisation, the specimens were heated in an oil bath at 160, 180 and 210°C for 1, 3 and 6 hrs. The oil bath used for the treatment and its flow diagram are shown in Figures 4.1 and 4.2 respectively. Commercial grade raw linseed oil supplied by M/S Mainland Paint and Prinking Ink Ltd was used as the heating medium.

For the heat-treatment of wood, the oil was firstly heated to the target temperature (160, 180 or 210°C), and once the temperature was achieved, the wood specimens were immersed in the oil and treated for a preset period of time (1, 3 or 6 hrs). The oil bath was covered with a lid but no external pressure was applied during the treatment. The fumes from the treatments were removed by an exhaust system. Three heating coils were placed within the oil bath to achieve and maintain a uniform target temperature in the oil. The heated oil was kept circulating throughout the treatment using an electric pump. The temperature of the oil was recorded at three points in the bath. After treatment, the heating was turned off and lid of the oil bath removed. Following this, specimens were kept in the oil bath for 15 minutes then the oil was drained. Then the samples were removed from the bath, wiped and transferred immediately to an oven at 102°C temperature. After 24 hours in the oven, the samples were removed, cooled in a dessicator and weighed. After this, the treated specimens were conditioned at 65% RH and 20 °C. Changes in weight, colour, dimensional stability, biological durability and mechanical properties were examined after the treatments according to the procedure described in Chapter 3.



*Figure 4.1. Oil bath used for heat-treatment.*



*Figure 4.2. Line diagram of the oil bath used for the treatment.*



## 4.3. Results and discussion

### 4.3.1. Chemical changes

The chemical changes in treated wood were examined by wet chemical and Fourier Transform Infrared (FTIR) analysis as per the method given in Sections 3.4.9 and 3.4.10, respectively.

#### 4.3.1.2. *Wet chemical analysis of wood*

The mean values for holocellulose, alpha cellulose, hemicelluloses and Klason lignin contents for treated and untreated wood are given in Table 4.1. No significant changes in chemical constituents were found with 160°C treatment. The holocellulose content was decreased by 15 and 32% at 180 and 210°C respectively. The alpha cellulose content was not changed significantly at 180°C however it decreased by about 5% at 210°C. The most significant change was noted for hemicelluloses content which decreased by approximately 35 and 69% with 180 and 210°C treatments respectively.

Thus, the results suggest that the chemical constituents of wood are changed after oil heat-treatment and are strongly dependent on the treatment temperature. The decrease in holocellulose content was mainly due to degradation of hemicelluloses. These observations agree with (Mazella *et al.* 2004) who showed that the chemical transformation of hemicelluloses increased rapidly at temperatures above 200°C during heat treatment in air.

Hemicelluloses are the least thermally stable wood polymers mainly owing to their low molecular weight, amorphous nature and branched structure which facilitate a rapid degradation compared to other wood components (Fengel and Wegener 1984; Windeisen *et al.* 2007). The acetyl groups in hemicelluloses are thermally unstable and lead to the formation of acetic acid which is auto-catalysed by the degradation of polysaccharides in particular the hemicelluloses (Tjeerdsma *et al.* 1998).

**Table 4.1. Chemical composition of untreated and heat-treated wood.**  
*Average values are the mean of three determinations. Standard deviations are in parenthesis.*

<i>Treatment</i>	<i>Acid soluble lignin</i>	<i>Holocellulose</i>	<i>Alpha cellulose</i>	<i>Hemicelluloses</i>
	%	% (A)	% (B)	% (A – B)
Control	26.0 (0.5)	72.2 (1.9)	42.0 (1.3)	30.2
160°C	25.5 (1.6)	70.2 (1.5)	43.1 (1.0)	27.1
180°C	36.0 (1.8)	61.3 (0.9)	41.6 (0.9)	19.7
210°C	48.4 (1.8)	49.4 (0.5)	39.8 (0.9)	9.5

The alpha cellulose content remained unchanged at 160 and 180°C treatments and fell only slightly at 210°C, indicating that some cellulose degradation may have occurred at the higher temperature. This can be possibly explained by the fact that cellulose is thermally more stable than hemicelluloses owing to its crystalline structure which provides strong cohesive bonding that protects it against acid attack during hydrolysis (Fengel and Wegener 1984). Furthermore, accessibility of glucosidic bonds in cellulose, which can be degraded by released acids, is restricted compared to that of hemicelluloses (Boonstra and Tjeerdsma 2006). (Kim *et al.* 2001) noted that the degradation of the crystalline region occurs at a temperature range of 300-340°C. However, (Hill 2006) reported that cellulose degradation begins at temperatures of 210-220 °C and its degradation rate increases significantly at temperatures above 270°C when levoglucosan and other breaking-down products are formed.

Lignin is considered the most thermal stable wood constituent. In temperatures up to 600°C, mass losses of hemicelluloses amount to over 95%, and those of cellulose to more than 80%, whereas mass losses of lignins do not exceed 60% (Bartkowiak and Zakrzewski 2004). Temperatures of more than 200°C are required for lignin degradation, but significant lignin degradation only occurs at a temperature above 280°C (Akgül *et al.* 2007). The relative proportion of lignin content in wood remained unchanged at 160°C but increased for 180 and 210°C treatments. The relative increase in lignin content is due to the simultaneous loss of holocellulose. The condensation reaction of lignin at temperatures higher than 200°C may also contribute to change in lignin content (Tjeerdsma and Militz 2005). However, it should be treated with caution as some of the thermally degraded carbohydrates may be incorporated within the lignin during Klason lignin determination

that can lead to a falsely high estimation of lignin content (Yildiz *et al.* 2006; Zaman *et al.* 2000).

#### 4.3.1.2. Fourier Transform Infrared (FTIR) spectrometry

Thermal degradation of treated wood is a complex process and it is difficult to draw firm conclusions based on the change in percentages of individual wood components. In addition, chemical analytical processes may result in concomitant degradation of wood polymers (Pandey 1999a). Therefore, chemical changes of heat-treated wood have been analyzed by FTIR spectroscopy. This technique has been shown to be useful for examining chemical variations after the heat-treatment since it is able to determine the intensity of specific bonds and functional group in polymers (Labbe *et al.* 2006; Tjeerdsma and Militz 2005; Windeisen *et al.* 2007). The assignments of the main infrared bands for wood are given in Table 4.2. The bands at  $1738\text{ cm}^{-1}$  (C=O) and  $1510\text{ cm}^{-1}$  (C=C) are pure bands for carbohydrates and lignin, respectively. The band at  $1650\text{ cm}^{-1}$  and all the bands below  $1430\text{ cm}^{-1}$  are complex in nature with contributions from both carbohydrates and lignin (Pandey 1999b).

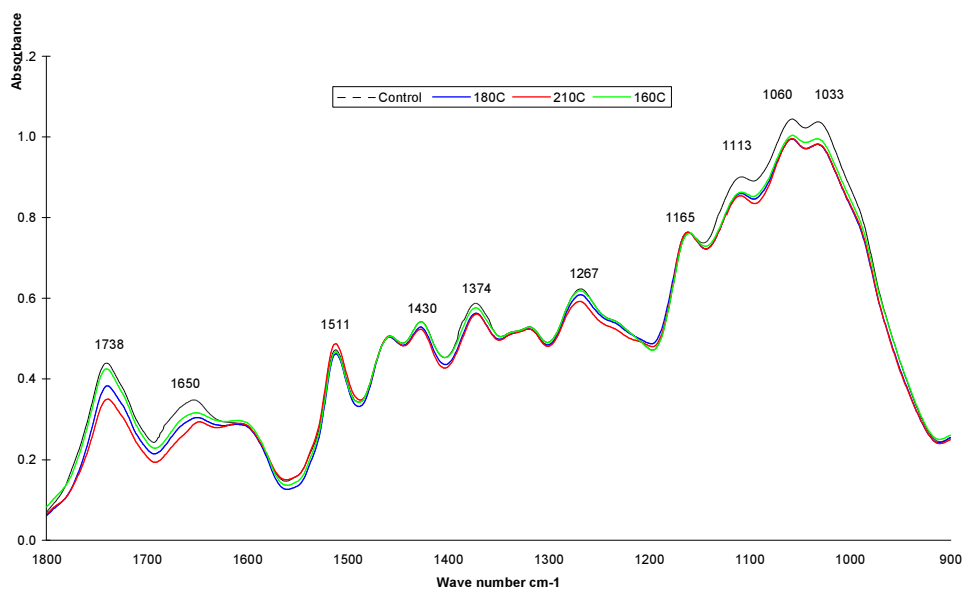
Figure 4.3 compares the FTIR spectra of untreated and treated radiata pine wood in the finger print region of  $1800\text{-}900\text{ cm}^{-1}$ . The peaks that showed important alterations were  $1738$ ,  $1650$ ,  $1511$ ,  $1430$ ,  $1374$ ,  $1267$ ,  $1165$ ,  $1113$ ,  $1060$  and  $1030\text{ cm}^{-1}$ , otherwise, the general aspect of the spectra remained unchanged (Pandey 2009).

**Table 4.2. Assignments of important IR bands of wood after (Labbe et al. 2006; Pandey 1999b).**

<i>Band position <math>cm^{-1}</math></i>	<i>Assignments and remarks</i>
1720-1740	Unconjugated C=O stretch vibration in hemicelluloses
1647-50	Absorbed OH and conjugated C=O
1511	Aromatic skeletal and C=C stretch vibration in lignin
1425-30	C-H deformation in lignin and carbohydrates
1371-75	C-H deformation in cellulose and hemicelluloses
1330-20	C-H vibration in cellulose and C=O vibration in syringyl derivatives
1268	C-O stretch vibration in lignin, acetyl and carboxylic vibration in hemicelluloses
1158-63	C-O-C vibration in cellulose and hemicelluloses
1113-1110	O-H association band in carbohydrates
1030-1060	C-O stretch in cellulose and hemicelluloses

The most significant change was observed at the  $1738\text{ cm}^{-1}$  peak which was sharply reduced after the heat-treatment. The drop was the greatest for the  $210^{\circ}\text{C}$  treated specimens which indicates the highest degradation

of the holocellulose (mainly hemicelluloses) at the higher treatment temperature. This is consistent with the greatest decreases in hemicelluloses content observed in the wet chemical analysis. The decline and shift of the  $1650\text{ cm}^{-1}$  band reflects the decrease in absorbed water with increasing treatment temperature. For the  $160^\circ\text{C}$  and  $180^\circ\text{C}$  treatments there were no changes in the shape of the peak shoulder at  $1511\text{ cm}^{-1}$ , which relates to lignin. However, for the  $210^\circ\text{C}$  treatment the small increase in the band has reflected some lignin modification. At temperatures above  $200^\circ\text{C}$  the free radical content of wood increases sharply which leads to coupling reactions between aromatic rings. This may result in more extensive cross-linking within the lignin network and a more condensed lignin structure (Tjeerdsma *et al.* 1998; Wikberg and Liisa Maunu 2004; Windeisen *et al.* 2007).



**Figure 4.3. FTIR spectra of untreated and heat-treated radiata pine wood.**

The bands at  $1430$  and  $1267\text{ cm}^{-1}$  that correspond to C-H deformation in hemicelluloses and lignin remained unaffected at  $160^\circ\text{C}$  and decreased

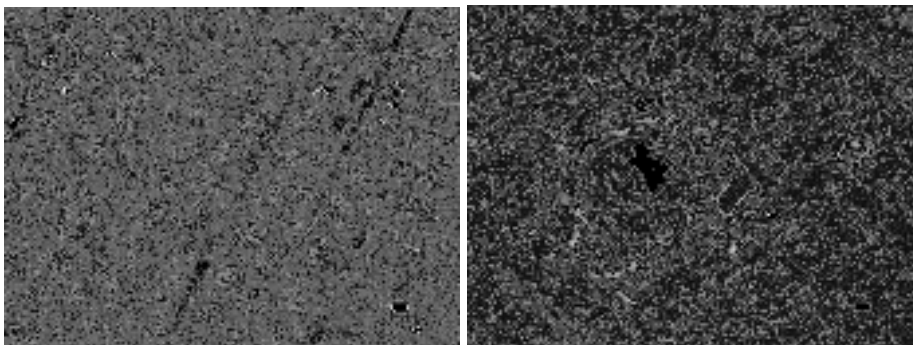
for the 180°C and 210°C treatments. There was a general decrease at the 1374 cm<sup>-1</sup> band corresponding to C-H deformation in holocellulose. Modification of H-bonding in carbohydrates due to thermal treatment was indicated by the drop in the 1113 cm<sup>-1</sup> band. The bands at 1060 and 1033 cm<sup>-1</sup> correspond to C-O stretching in cellulose and hemicelluloses. These bands decreased after the treatments which indicate depolymerisation of polysaccharides.

#### **4.3.2. Microstructure**

The microstructure of treated wood was examined by SEM with images presented in Figures 4.4 and 4.5. The cell wall structure retained its structural integrity even for the most severely heat-treated wood at 210 °C for 6 hours (Butterfield 2009). With the heat treatment, no obvious changes in the microstructure were observed and the cell walls remained intact with no visible damage or deformation. No tangential or radial cracks were noted in heat-treated specimens. Even parenchyma cells were found to be intact. These cells are considered very sensitive to damage owing to their non-lignified thin cell walls. These observations were in contrast to findings by (Boonstra *et al.* 2006) who observed radial cracks and damage to parenchymal cells in pine sapwood after a two stage heat treatment process in an aqueous environment at a temperature range of 165-185°C. This is probably due to the uniform heat transfer to wood in the oil medium in comparison to heat-treatment in other mediums (Sailer *et al.* 2000b). Bordered pits were found intact and unaffected as shown in Figure 4.4(c). The pit membrane (margo) also appeared undamaged.

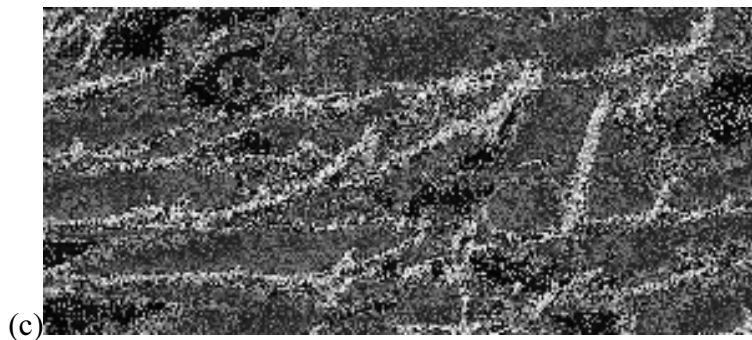
No deposition of linseed oil was observed in the lumens of specimens treated at 210 and 180°C. However, specimens treated at 160°C (Figure

4.5b) clearly showed lumens filled with depositions (Butterfield 2009) which indicates deposition of heat-polymerized linseed oil. This may be due to more oil uptake by specimens treated at 160°C than specimens treated at 180 and 210°C. This matter is discussed further in Section 4.3.3.



(a)

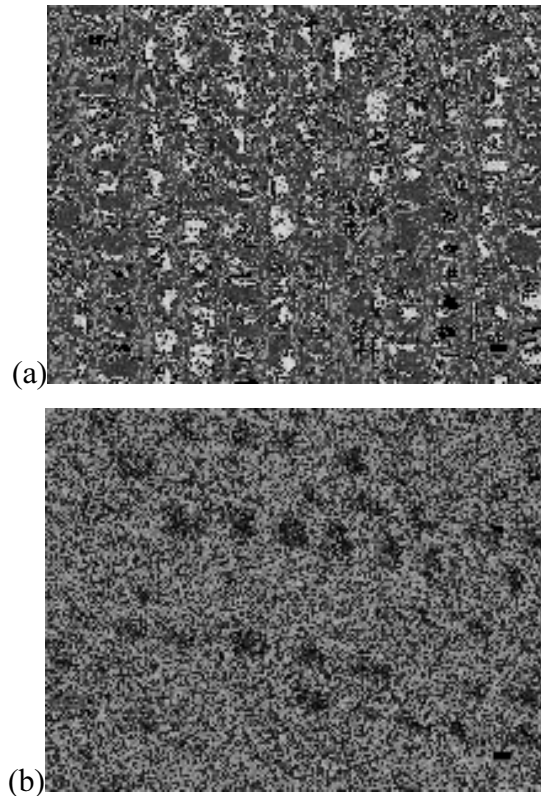
(b)



(c)

***Figure 4.4. SEM micrograph of heat-treated radiata pine wood at 210°C for 6 hours: (a) Cross section surface, (b) resin canal surrounded by parenchymal cells, and (c) radial section showing border pits.***

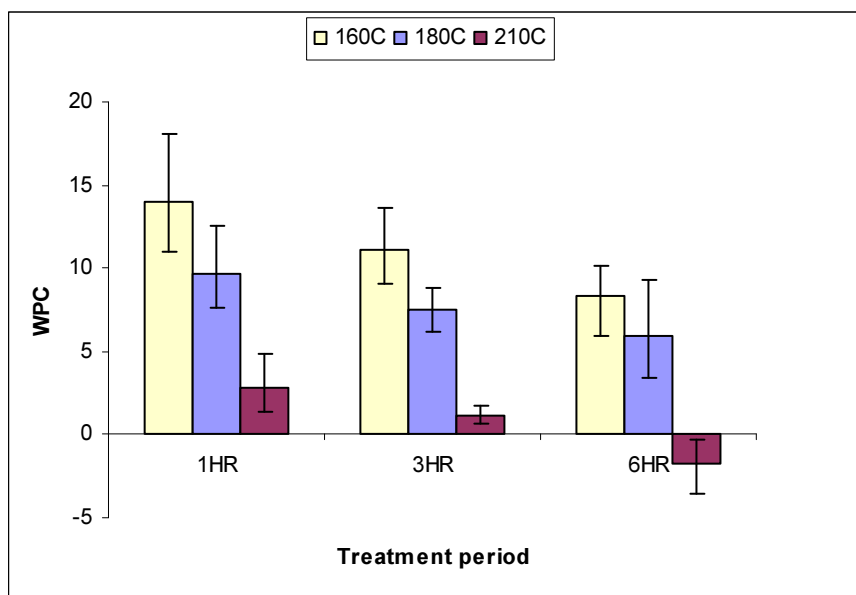




*Figure 4.5. SEM images of cross sections of radiata pine wood heat-treated at 180°C (a) and 160°C (b)*

#### **4.3.3. Weight and colour changes of the treated wood**

Figure 4.6 shows the effect of treatment temperature and time on the mean weight percentage change (WPC) of treated specimens. It is found that the weight gain decreased with an increase in the treatment temperature and duration. For the treatment duration of 3 hours, the WPC at 160, 180 and 210°C were approximately 11, 7.5 and 1%, respectively. At a given treatment temperature, the WPC values were maxima for the 1 hour treatments and minima for the 6 hour treatments. For the specimens treated at 210°C for 6 hours, a net weight loss of 1.8% was recorded.



**Figure 4.6. Effect of treatment temperature and duration on mean WPC of wood specimens (number of specimens,  $n= 8$ ).**

The heat-treatment also affected the wood colour, as expected, with the wood turning darker, however colour change was uniformly all over the surface. There were no surface cracks, warping or twisting observed in the treated wood. The surface appeared shiny near the rays which may be due to a high uptake of oil by parenchyma cells compared to other wood tissues. The colour change was identical on both flat surfaces (Table 4.3) with the difference between the two surfaces being reduced on increasing the treatment temperature. When the wood was treated at 180 and 210°C, the total colour change ( $\Delta E^*$ ) and brightness variation ( $\Delta L^*$ ) between the two surfaces were not more than 1.5 units although the wood surface became darker. The human eyes can distinguish colour change only if  $\Delta L^*$  is more than 3 units (Phelps *et al.* 1994) or the value of  $\Delta E^*$  is greater than 2 (Sundqvist 2002).

**Table 4.3 CIELAB values at the two surfaces of treated and untreated specimens**

		$L^*$	$a^*$	$b^*$	$\Delta L^*$	$\Delta a^*$	$\Delta b^*$	$\Delta E$
<b>Untreated</b>	Surface 1	68.1	11.51	33.8				
	Surface 2	64.3	12.2	31.8	3.8	-0.8	2.0	<b>4.3</b>
<b>OHT 160°C</b>	Surface 1	65.9	12.3	38.2				
	Surface 2	62.5	13.2	37.0	-3.3	0.9	-1.2	<b>3.6</b>
<b>OHT 180°C</b>	Surface 1	61.2	13.7	36.8				
	Surface 2	59.8	14.1	36.6	-1.4	0.4	-0.3	<b>1.5</b>
<b>OHT 210°C</b>	Surface 1	47.1	13.8	26.3				
	Surface 2	46.1	13.3	25.6	-0.9	-0.5	-0.7	<b>1.2</b>

Colour change was noted throughout the cross-section of the treated specimens and the intensity of the colour was greater on the surfaces than in the core (Figure 4.7). Table 4.4 gives the values of CIE  $L^*$   $a^*$   $b^*$  coordinates for the surface and core of treated wood. The average lightness values ( $L^*$ ) for the surface are 22-28% less than the corresponding values at the core. This difference in colour between the surface and the core decreased with increasing treatment temperature, i.e. for specimens treated at 210°C, the total colour difference between the surface and the core was 15 compared to the corresponding values of 15.9 and 16.7 for the specimens treated at 180°C and 160°C, respectively.



*Figure 4.7. Wood colour at surface (left) and core (right) of the specimen treated at 210°C*

*Table 4.4. CIELAB values at the surface and core of treated specimens*

		$L^*$	$a^*$	$b^*$	$\Delta L^*$	$\Delta a^*$	$\Delta b^*$	$\Delta E$
<b>OHT 160°C</b>	Surface	55	13.4	31.8				
	Core	70	7.3	27.5	15	-6.1	-4.2	<b>16.7</b>
<b>OHT 180°C</b>	Surface	61.2	12	33.5				
	Core	74.7	6.9	27	13.6	-5.1	-6.6	<b>15.9</b>
<b>OHT 210°C</b>	Surface	36.9	11.3	15.9				
	Core	47.3	11.8	26.7	10.4	0.6	10.8	<b>15</b>

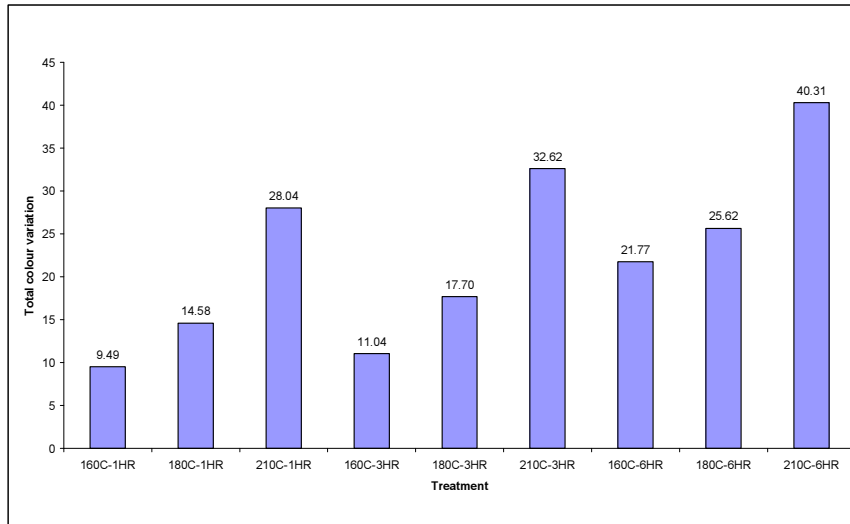
Further examination of the wood colour found that the colour of treated wood also varied with the treatment temperature as shown in Figure 4.8. The colour of wood treated at 160, 180 and 210°C for six hours was, respectively, yellowish brown, reddish-brown and dark-chocolate (Figure 4.8 a). Thus, the darkness of treated wood increased with the treatment temperature. The treatment time also affected the colour change. Specimens treated for six hours were darker in colour than those treated for one hour at the same temperature (Figure 4.8b).



***Figure 4.8. (a) Effect of treatment temperature at six hour treatment and (b) treatment period at 160°C on the colour of the wood***

Figure 4.9 shows the total colour change ( $\Delta E^*$ ) of oil heat-treated wood. The total colour change increased with treatment temperature and duration. At a given treatment time, the total colour change was greater for specimens treated at 210°C than for the specimens treated at 180°C which, in turn, was greater than for the specimens treated at 160°C. Similarly, specimens treated at a given temperature for six hours were darker than those treated at the same temperature for one hour. However, the treatment temperature has a more profound influence than the treatment time as the total colour change was greater for specimens treated at 210°C for one hour than for specimens treated at 180°C for six

hours. Similarly, specimens treated at 180°C for one hour were darker than those treated at 160°C for six hours.



**Figure 4.9. Total colour change ( $\Delta E L^*a^*b^*$ ) value with different temperatures and times**

The changes in the physical properties of wood (mean weight percentage change and wood colour changes) with heat-treatment are mainly the result of a combination of the following factors: (1) heat degradation of the chemical constituents of wood (2) the removal or migration of extractives and other compounds and (3) oil uptake in wood.

As discussed in previous sections, heat-treatment resulted in changes in wood chemical constituents mainly degradation of hemicelluloses (Table 4.1 and Figure 4.3). Some of the products formed after thermal degradation of hemicelluloses and lignin are coloured which leads to colour change in wood (Bekhta and Niemz 2003). These heat-degraded compounds along with wood extractives and other compounds such as

low molecular weight sugars and amino acids tend to migrate towards the surfaces during the heat-treatment process (Sundqvist and Morén 2002; Theander *et al.* 1993). Some of these compounds may transfer in the oil bath or be deposited at the surfaces. The transfer of heat degraded compounds and extractives in the heating oil are also evidenced by an increase in oil viscosity on heating with wood. This is further discussed in detail in Section 7.3.1. The migration of heat degraded and extractive compounds towards the surfaces also resulted in darker surfaces than at the core.

The colour of two flat surfaces of the oil heat-treated specimens was found to be identical without any resin patches. This contrasted with previous findings that reported a non-homogenous distribution of colour in thermally treated wood at 175-200°C for 1-10 hours in steam (Johansson and Morén 2006). This can be explained by the fact that during the heat-treatment in air or steam migration of extractives towards the surface leads to the formation of unsightly resin spots in wood (Mayes and Oksanen 2002), whereas in oil-heat treatment some of these extractives and other degraded products might have been transferred in the heating oil. Another possible explanation might be due to uniform heat transfer with an oil medium compared to that in the air or another heating medium (Sailer *et al.* 2000a).

The greatest mean weight increase (14%) of the specimens was found for wood treated at 160°C for 1 hour which can be attributed to the oil uptake (Figure 4.6). However, the actual oil uptake by specimens was greater as account must be taken of weight losses owing to wood degradation and removal of extractives at high temperature. The weight gain tended to be less at higher temperature treatment as the wood degradation was severe on increasing the treatment temperature, which is confirmed by the fact

that the treated wood actually lost about 2% of its weight in heat-treatment at 210°C for six hours.

The results of this study have shown that the oil heat-treated wood turned darker depending upon treatment time and temperature. The colour changes made the two flat surfaces of the specimens more identical without any colour patches observed, although the surfaces were darker than the core. Colour homogeneity between flat surfaces, and between core and surface increased with the increase in treatment temperature. The colour of wood is an important factor for aesthetic applications. While discolouration of wood during drying is undesirable, the darkening of wood during heat-treatment can be a tool to achieve a darker, richer coloured wood with improved properties. Furthermore, unlike the traditional method of painting to colour the wood, heat-treated wood does not involve any emission of health hazardous volatile organic compounds such as toluene and xylene (Mitsui 2004). Therefore, oil heat-treatment could be an effective and environmentally friendly way to change the colour of wood without any surface cracking or bending.

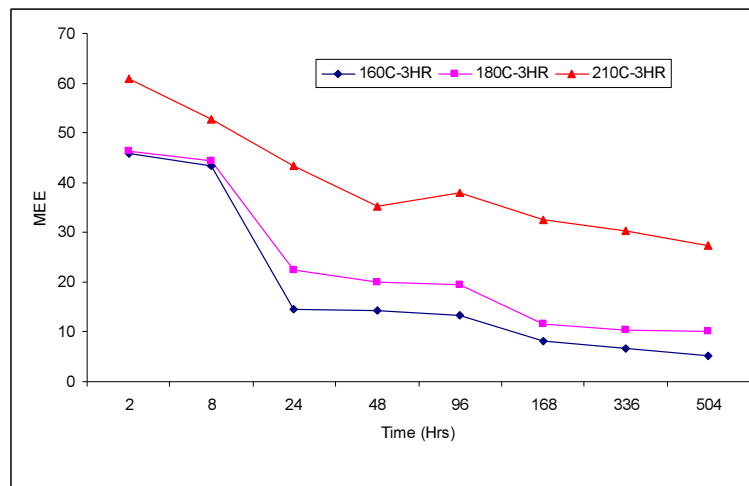
#### **4.3.4. Stability, durability and mechanical properties**

##### *4.3.4.1. Moisture repellency and dimensional stability*

The moisture repellency and dimensional stability are represented by Moisture Excluding Efficiency (MEE) and Anti Swelling Efficiency (ASE), respectively. The MEE was measured in a high humidity environment (85% RH and 20°C) while ASE was measured in a water bath, a high humidity environment and a dry-freeze-wet cycle. The measured results of MEE for the heat-treated wood are shown in Figure



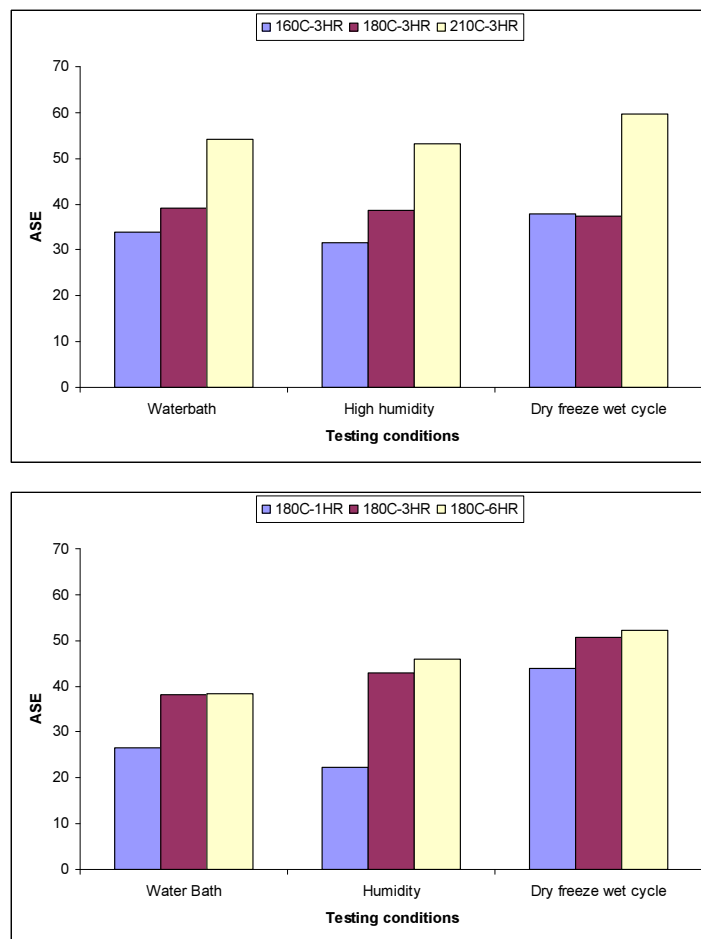
4.10 as a function of test duration. From the figure, it is seen that the treated wood became more hydrophobic and the MEE increased with the increase in treatment temperature. After 21 days in the high humidity condition, the MEE of specimens treated at 160, 180 and 210 °C were around 5, 10 and 27% respectively.



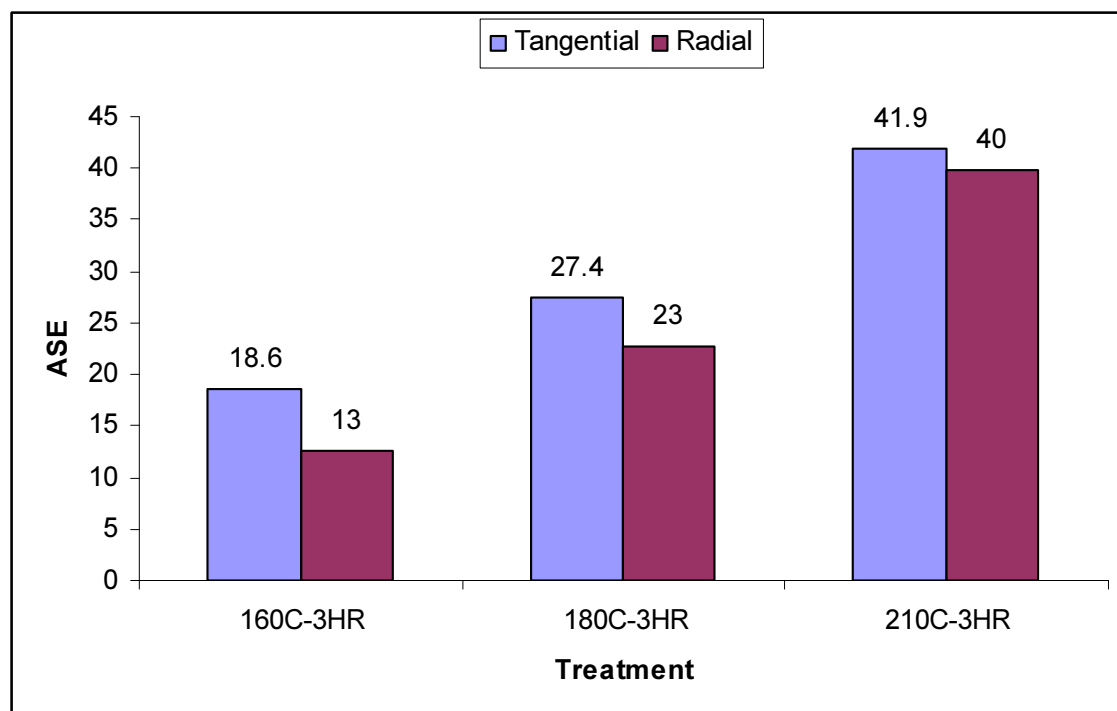
**Figure 4.10. Moisture excluding efficiency (MEE) after oil heat treatment in the high humidity test**

In Figure 4.11, the Anti Swelling Efficiency (ASE) of treated wood is shown in the three testing conditions: water immersion, high humidity environment and dry-freeze-wet cycle. In all three testing conditions, the volumetric swelling of the treated wood decreased substantially. The highest ASE of 53-60% was found in specimens treated at 210° C. There was little difference in the ASE between specimens treated at 160°C (ASE 31-38%) and specimens treated at 180°C (ASE 37-39%). At a given treatment temperature, the ASE were the lowest for of specimens treated for one hour. However, no significant difference was observed between specimens treated for three hours and specimens treated for six hours (Figure 4.11b).

The dimensional stability in the tangential and radial directions in the water immersion test is represented in Figure 4.12 in which the ASE in the tangential direction was greater than in the radial direction at all treatment temperatures. ASE at 160°C in tangential and radial directions was 19 and 13% respectively while the corresponding values at 210°C were 42 and 40%. Thus, the difference in the ASE between tangential and radial directions decreased with the increase in the treatment temperature. This suggests that at high temperatures in oil treatment wood became less anisotropic.



**Figure 4.11. ASE of treated wood in dimensional stability at (a) different temperature (b) treatment period.**

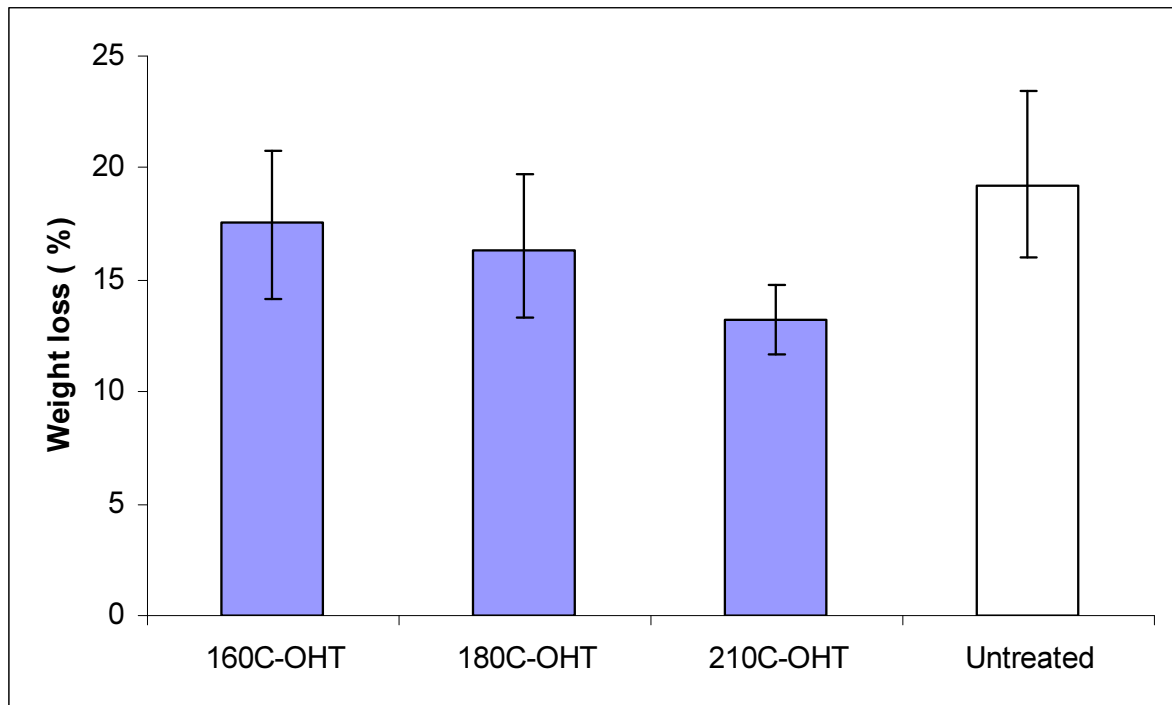


**Figure 4.12. ASE in radial and tangential direction**

#### 4.3.4.2. Biological durability

The weight loss percentages in a laboratory pure culture decay trial against brown rot fungus *Oilgoporus placenta* are given in Figure 4.13. The fungus *Oilgoporus placenta* was reported to cause a greater weight percentage loss in heat-treated wood compared to other wood decaying fungi namely *Coniophora putanea* and *Coriolus versicolor* (Welzbacher and Rapp 2002). The resistance of wood against this fungus was improved. The weight loss percentage in untreated wood was 19.2% and 13.2 -17.6% for treated wood. No significant difference ( $p < 0.5$  ANOVA single factor) in weight loss percentage was found with 160°C treatment in comparison to untreated wood. However, resistance of the treated specimens was improved with treatment temperatures of 180 and 210°C by 15% and 31% respectively. This matches the findings of others (Mazella *et al.* 2004; Paul *et al.* 2007) who reported that significant

fungus resistance of thermally modified wood can be achieved at a temperature above 200°C.



**Figure 4.13.** Mean weight loss percentages of oil heat-treated wood in a pure laboratory decay trial against *Oilgoporus placenta* (Number of specimens,  $n = 8$ )

#### 4.3.4.3. Mechanical properties

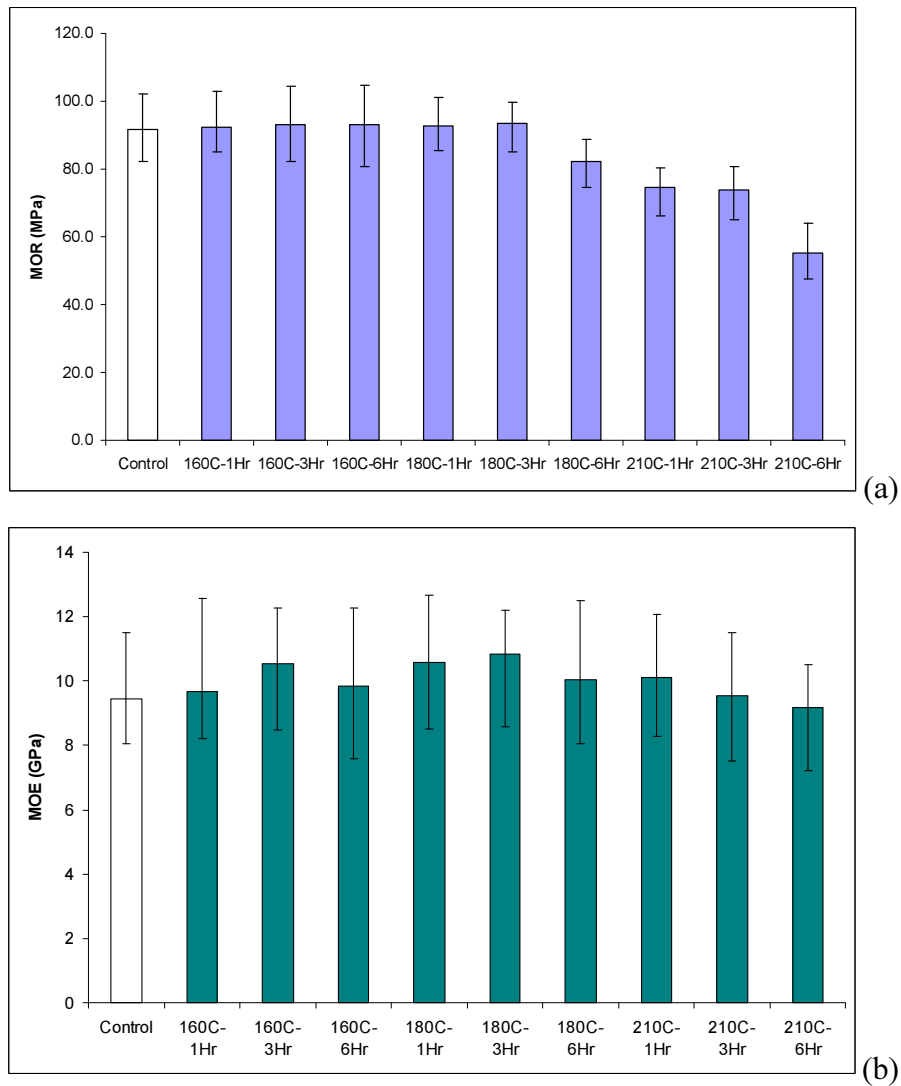
The average values of strength (modulus of rupture or MOR) and stiffness (modulus of elasticity or MOE) determined by a three point bending test are given in Table 4.5 and Figure 4.14. The MOR of the untreated wood were about 92 MPa while those of treated wood lay in the range of 55-93 MPa depending upon treatment temperature and time. As

shown in Figure 4.14 the MOR of specimens treated at 160°C for all treatment periods and 180°C for one and three hours were almost the same as those of the untreated specimens with no significant difference ( $p > 0.05$ , ANOVA single factor). However, the MOR for wood treated at 180°C for six hours and treated at 210°C (for all treatment periods) dropped significantly. The MOR of specimens treated at 180°C for six hours were 10% lower than those of the untreated specimens. In the case of specimens treated at 210°C the drop in MOR was sharper being 19, 20 and 40% less for the treatment periods of one, three and six hours respectively.

Unlike strength, the stiffness of the wood was generally unchanged or increased after treatment. The MOE values of treated wood were up to 12% higher than those of the untreated wood except for wood treated at 210°C for six hours where the elastic modulus decreased by only 3%. Stiffness was generally higher for the mild heat-treatments. Some previous studies also noted an improved MOE after heat-treatment (Bekhta and Niemz 2003; Kocaefe *et al.* 2008; Santos 2000). Conversely, some researchers observed a significant drop (up to 45%) in stiffness on heat-treatment over an identical temperature range (Esteves *et al.* 2005; Yildiz *et al.* 2002). (Shi *et al.* 2007) reported a drop of up to 28% in MOE in some species and up to a 30% increase in MOE after thermal treatment in similar conditions. Thus, the effect of temperature on MOE is very species dependent.

**Table 4.5. Mean strength (MOR) and stiffness (MOE) of wood samples in a three point bending test. The given values are averages of 15 replicates and SD denotes standard deviation.**

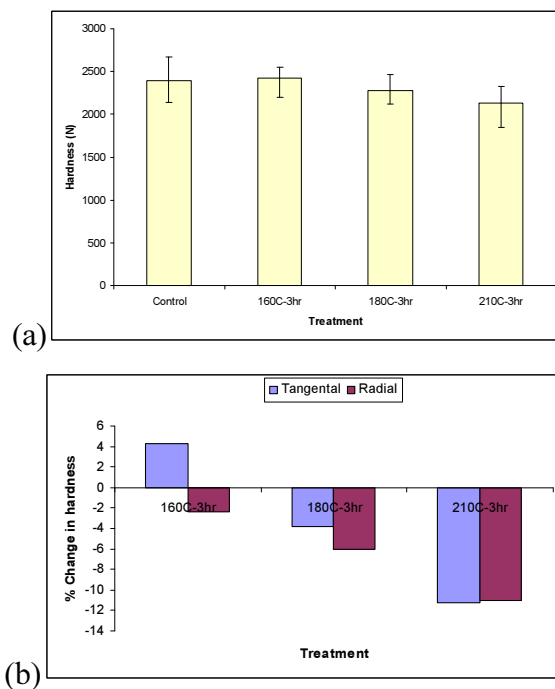
<i>Treatment Temperature (°C)</i>	<i>Treatment period (hr)</i>	<i>MOR (MPa)</i>		<i>MOE (GPa)</i>	
		<i>Mean</i>	<i>SD</i>	<i>Mean</i>	<i>SD</i>
Control		91.7	4.9	9.5	1.05
160	1	92.5	5.4	9.7	1.39
	3	93.1	6.3	10.5	1.07
	6	92.9	5.9	9.9	1.13
180	1	92.7	5.1	10.6	1.09
	3	93.4	4.1	10.8	1.01
	6	82.4	4.1	10.0	1.15
210	1	74.6	4.4	10.1	1.17
	3	73.7	5.5	9.5	0.99
	6	55.3	5.8	9.2	0.86



**Figure 4.14. Mean strength (a) and mean elastic modulus (b) of treated and untreated wood in a three point bending test. (Number of specimens,  $n = 15$ )**

Table 4.6 and Figure 4.15a give the hardness of untreated and treated (160, 180 or 210°C for 3 hours) and untreated wood determined by Janka indentation tests. Hardness is a measure of wood resistance against penetration of foreign bodies (Tsoumis 1991). The average hardness of untreated wood was 2.4 kN and this generally decreased with the heat-

treatment along with the extent of the decrease in the treatment temperature. No significant effect on average hardness was observed with 160°C treatment ( $p > 0.5$  ANOVA single factor). Tangential hardness was greater than radial hardness for both untreated and treated wood. Tangential hardness of wood treated at 160°C for three hours actually increased by 4.3% compared to the untreated samples. However at higher treatment temperatures of 180 and 210°C, the average tangential hardness decreased by 3.8% and 11.3%, respectively. In the radial direction, the hardness decreased for all of the three treatment temperatures by 2.3 to 11.3% depending on the treatment temperature (Figure 4.15b). At 210°C of heat-treatment, the hardness reduction in radial and tangential directions was similar at 11.3%.



**Figure 4.15. Average hardness of treated and untreated wood (a), and percentage change in radial and tangential hardness of treated wood compared to untreated wood (b).**



**Table 4.6. Hardness of oil heat-treated radiata pine wood in the Janaka indentation test (The given values are an average of 12 replicates and values in parentheses are standard deviation)**

<i>Treatment</i>	<i>Hardness (N)</i>		
	Tangential	Radial	Average
	[A]	[B]	[(A + B)/2]
160°C-3hr	2550.8 (160.3)	2295.9 (149)	2423.3 (123.1)
180°C-3hr	2353.4 (118.8)	2207.8 (131.9)	2280.6 (102.5)
210°C-3hr	2169.6 (192.4)	2091.8 (171.4)	2130.7 (175)
Control	2445.4 (220.6)	2350.4 (177.4)	2397.9 (188.1)

#### **4.3.5. Factors influencing dimensional stability, fungal resistance and mechanical properties.**

The oil heat-treated radiata pine wood was more moisture repellent, dimensionally stable and displayed improved fungal resistance. The mechanical properties of the treated wood were generally adversely affected (except stiffness) by the heat-treatment with the extent of the impact depending on treatment temperature and time. There are three main possible reasons for these changes as described as follows: (a) change in wood chemistry at high temperatures; (b) oil uptake and the accumulation of oil on the surface of wood and (c) the heating medium for the treatment.

##### *(a) Changes in wood chemistry*

In wood, cellulose is represented in both the crystalline and non-crystalline parts of the cell wall while the structure of hemicelluloses and lignin are amorphous (Akgül *et al.* 2007).

More than 60% of the wood cellulose is crystalline in nature which has strong resistance to moisture (Stamm 1964). Hillis (1984) reported that changes in wood moisture content lead to changes in its bond configurations of cellulose molecules with the non-crystalline regions of adjacent cellulose molecules and of hemicelluloses, resulting in swelling or shrinkage of the wood. Rowell and Banks (1985) summarized that hemicelluloses, non-crystalline cellulose, surfaces of crystalline cellulose and, to a small degree, lignin are responsible for hygroscopicity of wood. Of these, the hemicelluloses are the most hygroscopic in nature (Evans

2003; Hillis 1984; Seborg *et al.* 1953). Hemicelluloses are also nutrients for several fungi and considered to be a key component affecting wood decay (Kocaefe *et al.* 2008; Winandy and Morrell 1993).

Oil heat-treatment of wood resulted in changes in the structure and proportions of the major chemical constituents of wood. The thermal stability of softwood (radiata pine) hemicelluloses is lower than that of hardwoods (Kollmann and Fengel 1965). Significant changes in hemicelluloses have been revealed from chemical analysis of treated radiata pine wood (Table 4.1, Figure 4.3). Wet chemical analysis of treated wood revealed that the hemicelluloses percentage decreased by as much as 70% during heat treatment. The degradation of the hemicelluloses was further confirmed by FTIR analysis. Degradation of hemicelluloses results in reduction of available free polar adsorption sites mainly the hydroxyl group for water (Feist and Sell 1987; Hillis 1984). Furthermore, at very high temperatures (210°C), hemicelluloses may be changed to less hygroscopic substances like furfural polymers which are also toxic to fungal growth (Kamdem *et al.* 2002; Stamm 1964). Therefore, improved dimensional stability, water repellency and biological durability in oil heat-treated wood are mainly due to decomposition or transformation of hemicelluloses at high temperatures. As the hemicelluloses degradation increases rapidly at a very high treatment temperature (210°C), the moisture repellency, dimensional stability and fungal resistance of specimens treated at 210°C were found to be greater than those treated at 160 and 180°C. Boonstra *et al.* (2007) also reported that heat-treatment of wood can induce chemical transformation of other wood constituents, essential for fungal infestation such as minerals, vitamins and low molecular mass carbohydrates. This may also contribute to the fungal resistance of heat-treated wood.

Cellulose is considered to be mainly responsible for the mechanical strength of wood owing to its high degree of polymerisation and crystallinity (Mayes and Oksanen 2002; Pandey 1999b). The cellulose percentage was lowered to a lesser extent compared to the hemicelluloses in wood heat-treatment but this was significant for treatment at 210°C, therefore, greater losses of MOR and hardness were noted in the specimens treated at this temperature.

Stiffness and hardness are comparatively less affected in oil heat-treated wood which has been treated in a temperature range of 160-210°C. A marginal increase in MOE and hardness in oil heat-treated wood at mild heat treatments (160°C) for shorter duration may be due to the decrease in the relative proportion of low stiff hemicelluloses as compared to stiff cellulose microfibrils from the sample (Bergander and Salme' N 2002). However, at a higher temperature all the cell wall constituents start degrading and therefore both stiffness and hardness decline.

*(b) Oil uptake and the accumulation of oil on the surface of wood.*

Linseed oil is hydrophobic in nature (ASTM-D234-82 1998; Chemwatch 2007). It is considered as a non-swelling chemical as the size of linseed oil base molecule is too large to penetrate in the cell wall (Hill 2006; Olsson *et al.* 2001). Rosenqvist (2000) also confirmed that triglycerides found in linseed oil are not able to penetrate the wood cell wall. Thus, most of the absorbed oil in treated wood remains in the cell lumens. As the oil is not chemically bound with the cell wall it is expected that its contribution to dimensional stability of oil heat-treated wood is limited. This is further confirmed by the fact that specimens treated at 160°C with highest oil uptake exhibit more volumetric swelling (Figure 4.11) and fungal degradation (Figure 4.13) than specimens treated at 210°C with

the lowest oil uptake. This suggest that chemical changes at higher temperatures are more influential for dimensional stability and fungal resistance than the oil uptake, although, the latter also contributes to improve the stability and durability of wood (Wang and Cooper 2005).

Fungi require adequate moisture (1) to prevent their desiccation; (2) as a medium to diffuse enzymes and other metabolites in to the cell wall and for the nutrients and degradation products to enter the hyphae and (3) for the breakdown process catalysed by enzymes (Archer and Leebow 2006; Bowyer *et al.* 2003). Thus, oil uptake may contribute to dimensional stability and improved fungal resistance through blocking of some water-accessible sites by hydrophobic linseed oil. Furthermore, being a drying oil, the linseed oil improves the water repellency of wood by providing an additional surface barrier to the absorption of water. This protective surface barrier is also a contributing factor for the improved performance of oil heat-treated wood.

### *(c) Heating medium*

The mechanical properties of oil heat-treated wood were generally reduced compared to the untreated wood. However, strength losses were less severe than reported in previous studies on thermally modified wood at similar temperatures that reported up to 50% or more loss in strength on heating wood in different atmospheres (Bengtsson *et al.* 2002; Sailer *et al.* 2000a; Stamm *et al.* 1946; Yildiz *et al.* 2005).

The probable reason for comparatively smaller decreases in MOR in the current study is that oil was used as the heating medium. The oil as a heating medium transfers heat to wood more uniformly and provides an

oxygen-free environment as heating in oxygen results in more severe wood degradation and strength loss (Beall 1969; Kubojima *et al.* 2000; Mitchell 1988). In addition, heating wood using an open system with venting of fumes, i.e. without any external pressure also results in less thermal degradation because no acidic vapour builds up which can further catalyse the polysaccharide hydrolysis (Boonstra and Tjeerdsma 2006; Stamm 1956).

#### **4.4. Conclusions**

Radiata pine wood was heat-treated in a linseed oil bath over a range of temperatures and times. This study has shown that the microstructure of heat-treated wood was intact without distinct structural changes even at the most severe treatment (210°C for 6 hours). Wet chemical and FTIR analysis confirmed that the wood heat-treatment caused most notably the degradation of hemicelluloses (up to 70%). These analyses also indicated slight degradation of celluloses (about 5%) and further lignin condensation at 210°C. The treated wood turned darker with the total colour changes varying with the treatment temperature. Colour changes were uniform on both the surfaces and the difference between the colour of the core and the surface decreased on increasing the heating temperature. The treated wood was less hygroscopic and more dimensionally stable. The MEE and ASE of treated wood were up to 30 and 60% respectively. The ASE in the tangential direction was greater than in the radial direction; however the differences decreased with higher treatment temperatures. The resistance of treated wood against a brown rot fungus *Oilgoporus placenta* also improved by 8-30% depending on the treatment temperature with the best result at the highest treatment temperature of 210°C. The degradation of hemicelluloses

contributes largely to improvement in dimensional stability and fungal resistance of the treated wood.

The strength of the treated wood was decreased by 19-40% depending upon the process temperature. The MOR at 160 and 180°C dropped by about 20% while at 210°C it dropped sharply up to 40%. Similarly, the hardness of wood treated at 180 and 210°C was decreased by 5 and 11%, respectively. In contrast, the stiffness of the treated wood was generally unchanged or increased (up to 12%). Changes in the wood properties of treated wood were mainly due to change in chemistry as a result of high temperature. Degradation of hemicelluloses contributed largely to improvement in dimensional stability and fungal resistance of treated wood. Loss of mechanical properties particularly at 210°C was considered to be mainly due to cellulose degradation.

Based in the findings of this study, it is concluded that oil heat-treatment of radiata pine wood results in wood darkening, but becoming more dimensionally stable and durable with improved fungal resistance. However, the mechanical properties were generally reduced except for the stiffness with the heat-treatment. The wood retains its microstructure. The chemical changes mainly degradation of hemicelluloses, is considered as the main cause of changes in the wood properties.

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# Chapter 5

## OIL HEAT-TREATMENT OF THERMO-MECHANICALLY COMPRESSED WOOD

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### 5.1. Introduction

The objectives of this chapter are: (1) to improve or incur the loss of the mechanical properties of oil heat-treated wood through densification, (2) to examine the spring back and compression set recovery after densification and oil heat-treatment, and (3) to observe the colour and fungal resistance of the densified oil-treated wood.

In the previous chapter, it was found that the dimensional stability and fungal resistance of radiata pine wood could be improved after oil heat-treatment. However, the oil heat-treatment also resulted in a reduction in mechanical properties. The mechanical properties losses with heat treatment in varying environments were also reported in other studies (Bengtsson *et al.* 2002; Sailer *et al.* 2000; Stamm *et al.* 1946; Yildiz *et al.* 2005). To overcome or minimize the mechanical property loss is one of the major technical challenges in the successful heat-treatment of wood (Hill 2006).

Thermo-mechanical compression of wood increases the wood density by reducing the void volume, thus it can improve the wood's mechanical properties. Most of the mechanical properties are positively correlated

with the wood density (Bowyer *et al.* 2003). High temperature densification of wood in a mechanical press has been studied since the beginning of the twentieth century (Seborg *et al.* 1945; Seborg and Stamm 1941). However, densified wood shrinks and swells more owing to the relatively higher proportion of cell wall and less lumen volume (Spear and Walker 2006). On exposure to high humidity conditions, the compressed wood tends to regain its original compressed dimension and this phenomenon is known as compression set recovery (CSR) or shape memory (Kamke 2006). Several investigations have been attempted to minimize the compression set recovery and most of these efforts were to fix the set recovery either through steam treatment, before or during the compression (Higashihara *et al.* 2001; Inoue *et al.* 1993a; Inoue *et al.* 2008; Kawai *et al.* 1992; Navi and Girardet 2000; Navi and Heger 2004; Satoshi *et al.* 2008), or through resin impregnation (Chris and Frederick 2008; Inoue *et al.* 1993b; Shams *et al.* 2006; Shams *et al.* 2004; Stamm and Baechler 1960; Yano 2001). The existing knowledge on the fixation of compression set recovery through heat-treatment is limited or inadequate in some cases (Dwianto *et al.* 1997; Welzbacher *et al.* 2005).

This study is intended to evaluate the dimensional stability and mechanical properties of thermo-mechanically compressed radiata pine wood with oil heat-treatment. In this study, wood was first compressed in a radial direction then heat-treated in linseed oil at different temperatures. Spring back percentage, compression set recovery and anti-swelling efficiencies were determined to examine the dimensional stability. Mechanical properties were evaluated based on changes in strength, stiffness and surface hardness of the wood after treatment. In addition, change in colour and fungal resistance against a brown rot fungus were also investigated.

Throughout this chapter, the following key abbreviations are used:



- TMC: Thermo-mechanical compression;
- OHT: Oil heat-treatment;
- TMC+OHT: Thermo-mechanical compression followed by oil heat-treatment.

## **5.2. Experimental**

### **5.2.1. Specimen preparation**

The wood specimens of dimensions 200 mm (long) x 90 mm (wide) x 35 mm (thick) were prepared from 3-4 m long boards of radiata pine. All boards were of the same commercial grade, dry sapwood (10-12% moisture content), flat sawn with growth rings parallel to a flat surface. Specimens were conditioned at 65% RH and 20°C for 2-3 weeks till the change in the specimens' weight was less than 0.1%. These specimens were then marked and their thicknesses were recorded nearest to  $\pm 0.01$  mm at the three marked places.

### **5.2.2. Thermo-Mechanical Compression**

Wood specimens were densified in radial direction in the Gibitre laboratory press (as shown in Figure 5.1) to the desired thickness by applying pressure of 30-32 bars at 180 °C for 20 minutes. The metal stoppers were placed on both sides of the specimens to achieve the targeted compression. The moisture content of the specimens before pressing was 14-16%. After compression, specimens were cooled down from 180 to 35°C with the cooling rate of 15°C per minute. Cooling of the specimens was carried out in the press without releasing the compression pressure. After cooling, specimens were removed from the press and their thicknesses were immediately recorded at the marked places. Then, the specimens were kept in a conditioning chamber at 65%

RH and 20°C until they attained equilibrium and finally their thicknesses were measured at the marked places.



*Figure 5.1. Laboratory press used for TMC of wood specimens*

### **5.2.3. Oil heat-treatment**

Conditioned thermo-mechanically compressed specimens with moisture content ranging between 9-11% were heated in the oil bath at 160, 180 and 210°C for 3 hours. The oil heat-treatments were carried out as per the procedure described in Section 4.2. For the comparison studies, some end-matched wood specimens were also heat-treated in the oil bath without any thermo-mechanical compression. After the oil heat-treatment, the specimens were placed in the conditioning chamber at 65% RH and 20°C till they attained equilibrium and their thicknesses were again measured at the marked places.

## 5.3. Properties Measurement

The colour change, oven dry density, water repellence efficiency, mechanical properties and fungal resistance were determined according to the methods described in Chapter 3.

### 5.3.1. Compression set, spring back and set recovery percentages

Compression set (CS) is the amount of dimensional changes in compressed direction, expressed as percentage of changes in compressed specimens [conditioned at 65 % RH and 20°C](#) compared to the original dimension. It was determined according to Equation (5.1) defined by (Navi and Girardet 2000). In addition, compression percentages in latewood and early wood were also estimated by measuring the changes in the respective bandwidths before and after compression through image analysis using the software Image J1.0G.

Compressed material has a tendency to partially regain its original shape after removal of applied pressure owing to elastic recovery. This behaviour is known as spring back (SB) and results in change in compressed dimension (Garcia-Romeu *et al.* 2007). In this study, the spring back percentage was calculated using Equation (5.2) with the dimensions being measured two weeks after the compression force removal. During the two week period, the test specimens were kept in the conditioning chamber at 65% RH and 20°C.

On exposure to high humidity, the thickness of the compressed wood further increases which is known as compression set recovery (CSR). In this study, the compressed specimens were exposed to water and a high humidity (85% RH, 20°C) condition and the CSR after 2 and 672 hours

exposure were calculated according to Equation (5.3) defined by (Navi and Girardet 2000).

$$CS(\%) = \left( \frac{T_1 - T_3}{T_1} \right) \times 100\% \quad (5.1)$$

$$SB(\%) = \left( \frac{T_3 - T_2}{T_2} \right) \times 100\% \quad (5.2)$$

$$CSR(\%) = \left( \frac{T_4 - T_3}{T_1 - T_3} \right) \times 100\% \quad (5.3)$$

In Equations (5.1), (5.2) and (5.3),  $T_1$  is the original thickness of specimens conditioned at 65% RH and 20°C before pressing (mm);  $T_2$  is the thickness of specimens immediately after removal from the press (mm),  $T_3$  is the thickness of specimens after conditioning at 65% RH and 20°C for two weeks (mm), and  $T_4$  is the thickness of specimens after exposure in water or high humidity (mm).

### 5.3.2. Swelling percentage and ASE

Swelling percentages (SW) in radial (compressed), tangential and longitudinal directions were determined with the water immersion test (described in Section 3.4.4.1) as per Equation (5.4).

$$SW = \left[ \frac{(D_a - D_b)}{D_b} \right] \times 100\% \quad (5.4)$$

In which,  $D_a$  is the dimension of specimens after water immersion (mm), and  $D_b$  is the dimension of oven dry specimens before water submersion (mm).

Anti Swelling Efficiency (ASE) of the specimens was determined using Equation (5.5).

$$ASE = \left( \frac{S_c - S_t}{S_c} \right) \times 100\% \quad (5.5)$$

Where,  $S_c$  is the dimensional or volumetric swelling percentage of the untreated or only thermo-mechanically compressed specimens and  $S_t$  is corresponding swelling percentage of the thermo-mechanically compressed and oil heat treated specimens (mm).

## 5.4. Results and discussion

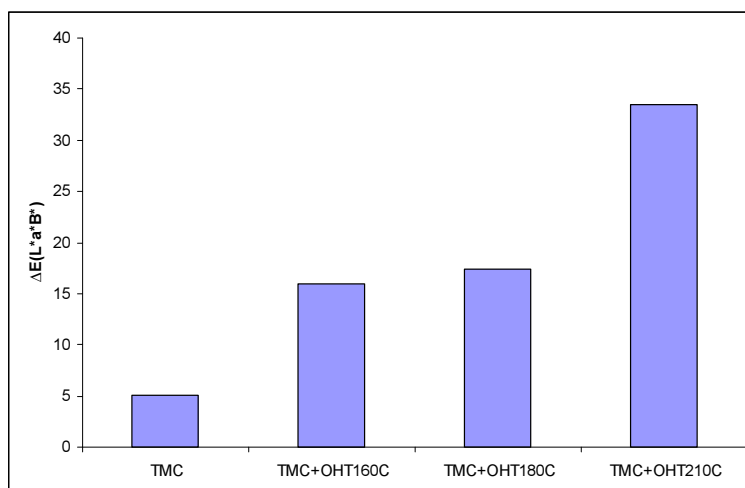
### 5.4.1. Colour

From the visual inspection of thermal-mechanically treated specimens as shown in Figure 5.2, it is observed that the wood colour appeared unchanged. The value of total colour change ( $\Delta E$ ) obtained by spectrophotometer for thermal-mechanically compressed (TMC) wood was 5.1 (Figure 5.3). This also confirms that the wood colour change after TMC densification was insignificant. However, after oil heat-treatment (OHT), the TMC densified wood turned dark and showed significant colour change with values varying from 15.9 to 33.5, increasing with the oil heat-treatment temperature (Figure 5.2 and 5.3). The wood treated at 210°C turned out to be dark chocolate brown with the highest colour change ( $\Delta E=33.5$ ). The colour change in specimens treated at 160 and 180°C was nearly same with  $\Delta E$  values of 15.9 and 17.4 respectively. Figure 5.4 compares the colour change in oil heat-

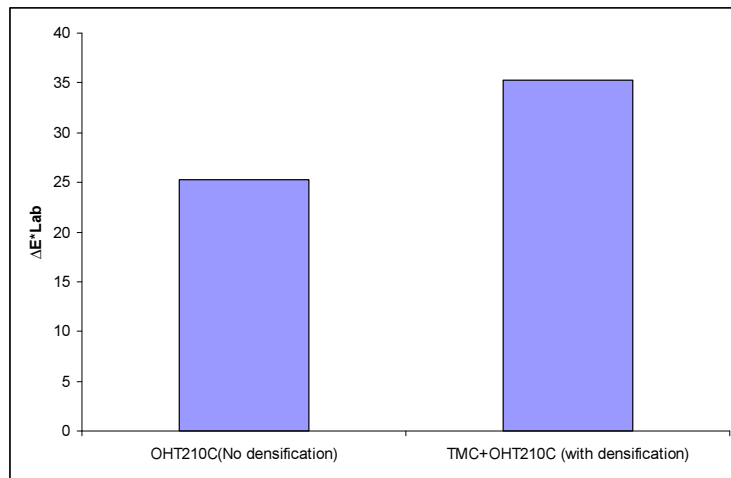
treated wood with and without prior densification. It clearly shows that densified wood became much darker than un-densified wood in the oil heat-treatment. This suggests that increase in wood density results in greater colour change with heat-treatment.



*Figure 5.2. Colour of wood in TMC wood and TMC+OHT wood treated at different temperatures*



*Figure 5.3. Total Colour change ( $\Delta E$ ) value in TMC wood and TMC+OHT at different temperatures*



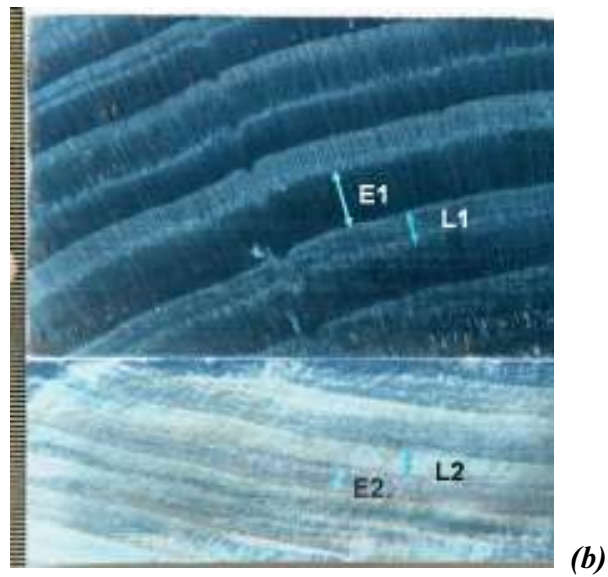
**Figure 5.4. Total Colour change ( $\Delta E$ ) value in oil heat-treated wood with and without prior densification**

#### **5.4.2. Compression set and spring back**

Figure 5.5(a) shows the dimensional changes of the wood specimen in compression direction (thickness). No visible checking or cracking was observed in the compressed specimens. The mean compression set was measured to be 39.4% with statistical analysis results given in Table 5.1. The compression in the compression direction was further analysed to investigate the differences between earlywood and latewood as seen from Figure 5.5(b). The images were used to quantify the compression set and the results are given in Table 5.2 which shows that the earlywood was compressed more than the latewood. The compression set determined in this way was 56.5% for earlywood bands and 20.6% for latewood bands. These findings are in agreement with the observations of Tabrasa (1997) who used light micrographs to examine wood densified at 100-200°C. These results were expected owing to the large lumen and thin cell walls of the earlywood tracheids compared to the latewood which has small lumen and thick cell walls (Bowyer *et al.* 2003).

**Table 5.1. Compression set percentage in wood after TMC**

<i>Mean</i>	39.4%
<i>Standard deviation</i>	0.8
<i>Y+ (Upper range from mean)</i>	1.7
<i>Y- (Lower range from mean)</i>	1.5
<i>Number of specimens (n)</i>	24



**Figure.5.5. (a) Thermo-mechanically compressed wood: before (left) and after compression (right); (b) Difference between earlywood and latewood compression: E1 and L1 - earlywood and latewood before compression, E2 and L2 the corresponding wood after compression.**



**Table 5.2. Compression sets of earlywood and latewood bands within a specimen based on percentage change in pixels measured by image analysis software Image J1.0G.**

	<i>Early wood</i>			<i>Latewood</i>		
Ring No.	Before compression	After compression	Percentage change	Before compression	After compression	Percentage change
	*pixels	Pixels	%	pixels	pixels	%
1	260	120	53.8	78	58	25.4
2	261	118	54.6	102	83	18.1
3	170	65	61.6	126	104	17.5
4	260	119	54.2	111	89	19.8
5	265	110	58.5	106	82	22.3
Mean			<b>56.6</b>			<b>20.6</b>
**SD			3.4			3.3

\* 1 Pixel = 0.02mm, \*\* SD is Standard deviation

**Table 5.3. Spring back (%) of treated wood. The given values are mean of seven specimens and SD is standard deviation. Y+ and Y- are the upper and lower range from mean respectively.**

Treatment	Mean	SD	Y+	Y-
TMC only	6.2	1.0	2.8	1.2
TMC followed by OHT at 160°C	6.0	1.4	1.7	2.0
TMC followed by OHT at 180°C	4.6	0.7	1.3	1.0
TMC followed by OHT at 210°C	1.7	0.8	1.1	1.1

The results of the measured spring back are given in Table 5.3. From the results it was found that the spring back in thermal-mechanically compressed (TMC) wood was 6.2%. Spring back with TMC followed by oil heat treatment (OHT) at 160°C was nearly the same as that of the TMC wood. However, the TMC wood followed by OHT at 180 and 210°C gained much less spring back, only 4.6 and 1.7%, respectively.

The results suggest that radiata pine wood can be successfully compressed to about 40% of its original thickness without any surface defects and cracks. The spring back percentage was less than 6.2% after TMC treatment and this can be reduced by a combination of TMC followed by OHT treatment, with the level of reduction decreasing with increasing OHT temperature.

There are several possible explanations for the wood compression and spring back with the above treatment. Wood cell walls consist of

amorphous or non-crystalline matrix of lignin and hemicelluloses in which strong and stiff cellulose microfibrils are embedded (Spear and Walker 2006). The glass transition temperature for isolated and dry cellulose, hemicelluloses and lignin are respectively 200-230°C, 165-175°C and 200°C (Laborie 2006). However, the glass transition temperature of the wood as a whole is different from the isolated polymers. This indicates that interaction among wood polymers plays an important role in the thermal softening of wood. The oven dry wood starts to soften at 180°C and reaches the maximum softening at 380°C (Chow 1971; Shiraishi 2001). The peak softening temperature decreases with an increase in the moisture content of wood and reported as 160°C at 13-14% moisture content (Chow 1971) and 60-90°C for water saturated wood (Nakajima *et al.* 2008). During TMC treatment of wood at 180°C, amorphous regions of wood become softened and the physical state of amorphous polymers of wood changes from glassy to rubbery facilitating a significant compression of wood. In contrast, microfibrils remain in a glassy state and are less affected by temperature. It has been suggested that on application of a load at the high temperature, the matrix is softened, facilitating relative displacement of microfibrils so that elastic deformation takes place in the whole framework of wood polymers (Dwianto *et al.* 1998b; Inoue *et al.* 1993a).

Another possible explanation for achieving compression without any surface defects is preheating of the mechanical press to the target temperature (180°C) before placing the specimens for compression. This caused rapid moisture movement within the specimens and moisture loss from the specimen surfaces which resulted in further decreases in compression modulus owing to the mechano-sorption effect (Kamke 2006). In addition, cooling down the specimens from 180 to 35°C under the full pressure also lowered the spring backs. This is supported by the

findings of (Kamke 2006; Seborg *et al.* 1945) who reported that spring back in densified wood could be minimised by cooling down the TMC wood under full pressure to 100°C or a lower temperature which facilitates the return of the glassy state in the amorphous region.

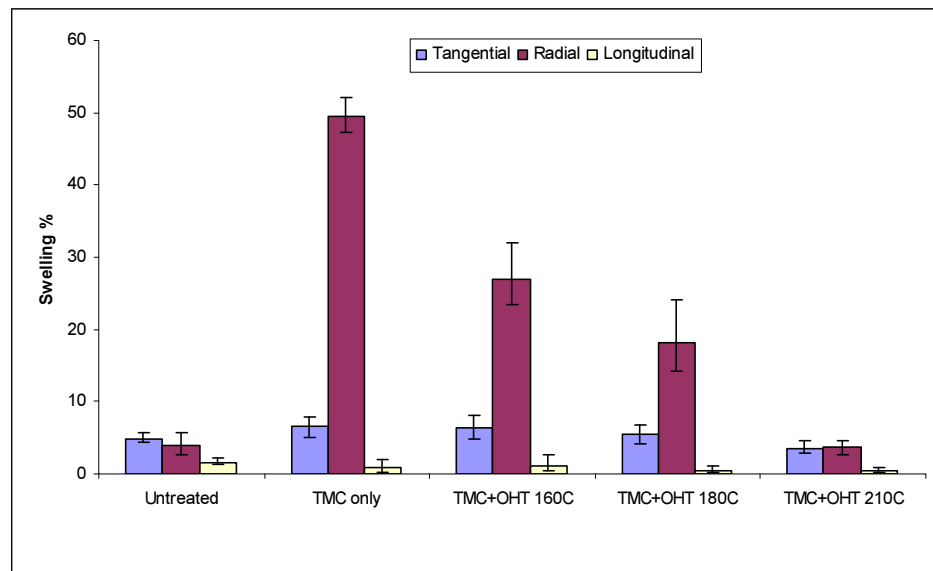
Oil heat-treatment of densified wood leads to further degradation of wood polymers, mainly hemicelluloses which can relieve the residual stresses built up during the densification (Dwianto *et al.* 1997; Hsu *et al.* 1988). This can be used to explain the reduced spring back in the TMC treatment of wood followed by OHT compared to only TMC treatment. The degradation of wood polymers increases with the heating temperature thus spring back percentages were the minimum for the specimens with oil heat-treatment at 210°C.

#### **5.4.3. Compression set recovery, anti swelling efficiency and water repellency**

During TMC treatment, both elastic and plastic deformation takes place in wood. A gradual decrease in press temperature during the cooling phase leads to the return of the glassy state in the amorphous region and thus deformation in the wood becomes elastic owing to the so called ‘frozen densification’ (Dwianto *et al.* 1998a; Inoue *et al.* 1993a). However, this frozen state is only temporary as on exposure to high humidity or water, most of this elastic deformation tends to recover. This phenomenon is known as compression set recovery. On exposing TMC wood to water or high humidity conditions, both reversible and irreversible swelling takes place. The former is due to the hygroscopic nature of wood while the later is the result of compression set recovery (Hsu *et al.* 1988).

In the experiments, swelling of both treated and untreated, matched wood specimens were measured in different wood directions after four weeks

of water immersion. The results are given in Figure 5.6 which shows that the swelling percentages of the untreated wood were 4.9, 4 and 1.6% in tangential, radial and longitudinal directions respectively. However, for the TMC treated wood, the swellings in the radial direction (49.4%) were much greater than for those of the untreated wood. In addition to the natural swelling of wood, compression set recovery is the main reason for the excessive swelling of the TMC treated wood in the radial direction. The swelling of the TMC treated wood followed by OHT decreased to a varying extent depending on the OHT temperature. For specimens with OHT at 160 and 180°C the swelling percentages in tangential and radial directions were still greater than untreated specimens but lower than only TMC densified wood. The specimens with OHT at 210°C gave the best results with 3.5, 3.7 and 0.3% swelling in tangential, radial and longitudinal directions, respectively, which were lower than those of the untreated wood.



***Figure 5.6. Swelling percentages in different directions after four weeks in water immersion. The given values are the means of ten replicates.***

Table 5.4 provides compression set recovery of the treated wood in the water immersion and high humidity tests. Even after two hours of water immersion, large compression set recovery of 46% was observed in the TMC treated wood which further increased to 66% after four weeks of water immersion. The compression set recovery was reduced significantly with OHT of the TMC densified specimens and the extent of the reduction dependent upon the OHT treatment temperature. After four weeks in water immersion, the set recovery of the TMC wood followed by OHT treatment at 160, 180 and 210°C was measured to be 37, 26 and 6%, respectively. In high humidity tests, the corresponding values for the set recovery were 19, 16 and 3%. However, the set recovery after two hours of exposure was less than 1% for all the treatments. These results suggest that oil heat-treatment reduced the set recovery of the TMC densified wood. The OHT at 210°C can effectively fix the deformation in densified wood. This result is consistent with earlier observations by (Dwianto *et al.* 1997) who showed that the post heat-treatment (in air, vacuum or molten metal) of densified wood above 200°C for 3 hours resulted in a compression set recovery of less than 5%.

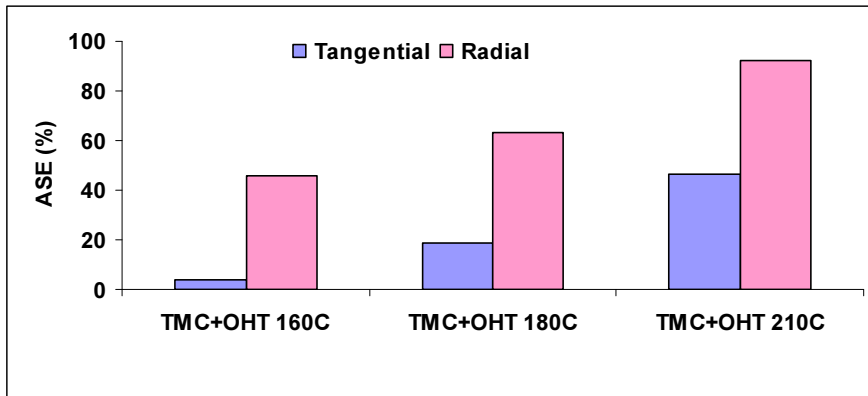
The reduction in swelling and compression set recovery of the TMC treated wood followed by OHT are mainly attributed to increased water repellence and reduced wood hygroscopicity and release of residual stresses in the microfibrils and matrix.

The oil heat-treatment of wood resulted in increased water repellency and reduced hygroscopicity. Dwianto *et al.* (1998a) studied the relationship between set recovery of densified specimens and ASE of non-densified specimens heat-treated at 160-200°C for varying periods in a molten metal bath, air and vacuum. They observed a linear decrease in set recovery of densified wood with the increasing ASE (reduction in hygroscopicity) of non-densified wood.

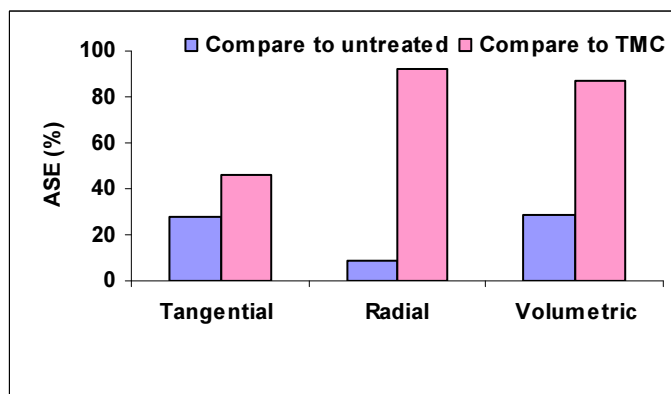
High temperature treatment of wood also results in degradation of cell wall polymers. This leads to the release of residual stresses in the matrix and microfibrils and may results in the fixation of compression set (Dwianto *et al.* 1998a; Navi and Girardet 2000).

**Table 5.4. Compression Set Recovery (CSR) in water immersion and high humidity test. The given values are the means of ten replicates and SD is standard deviation**

<i>Treatment</i>	<i>Water immersion test</i>				<i>High humidity test</i>			
	After 2 hrs		After 672 hrs		After 2 hrs		After 672 hrs	
	Mean	SD	Mean	SD	Mean	SD	Mean	SD
TMC only	45.9	10.3	66.3	4.6	0.8	0.2	22.5	3.3
TMC+OHT 160°C	12.8	5.9	37.2	4.7	0.5	0.2	18.8	2.3
TMC+OHT 180°C	9.6	3.9	25.9	4.3	0.3	0.1	16.2	1.9
TMC+OHT 210°C	2.4	2.3	5.6	0.8	0.2	0.1	3.5	1.8



**Figure 5.7. ASE in tangential and radial directions compared to TMC wood**



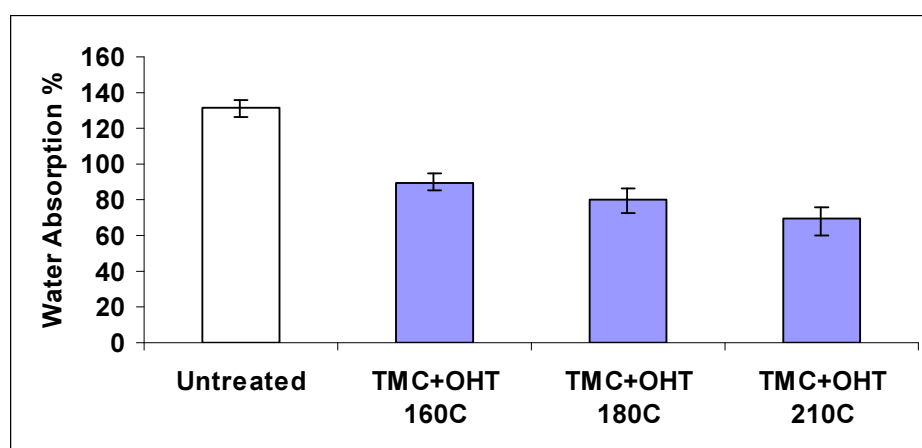
**Figure 5.8. ASE of TMC+OHT at 210°C wood compared to untreated and TMC wood**

The improvement in dimensional stability of the TMC treated wood followed by OHT is also evidenced by improved anti-swelling efficiency ASE and water repellency (WRE). Figure 5.7 shows the ASE in radial, tangential and longitudinal directions for the TMC+OHT wood in comparison to only TMC treated wood. The values of ASE in the radial directions were 46, 63 and 93%, respectively, for the oil treatment at 160, 180 and 210°C. In the tangential direction, the corresponding ASE values

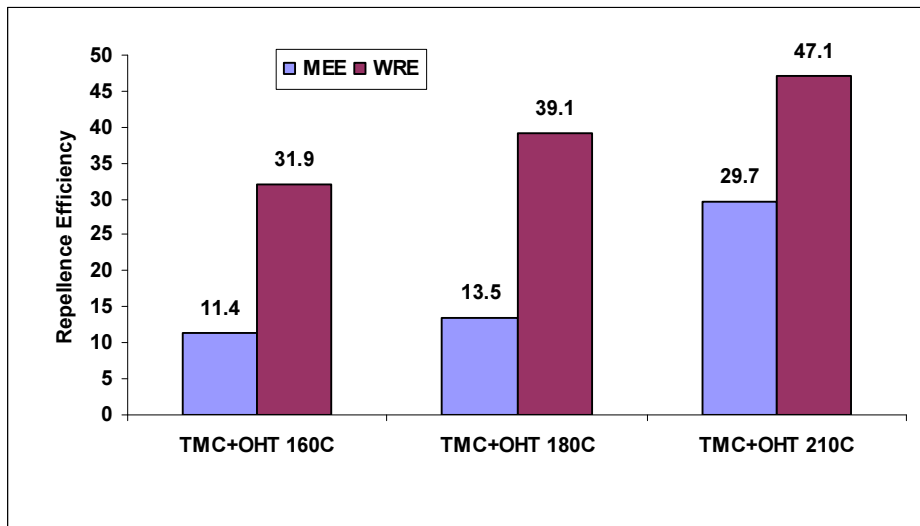


were 4, 19 and 46% which were much less than those in the radial direction. The ASE of TMC treated wood followed by OHT was much higher when assessed with respect to TMC treated wood as compared to the assessment with respect to untreated wood (Figure 5.8). For example, the volumetric ASE of wood of TMC treatment followed by OHT at 210°C was 29 and 87% evaluated with respect to untreated and TMC wood respectively.

Figure 5.9 gives water absorption percentages by wood specimens after four weeks of water immersion. This data shows TMC wood after oil heat treatment absorbs less water than untreated wood, with water absorption decreasing with increasing OHT temperature. The corresponding water repellence efficiency (WRE) of the densified wood followed by OHT at 160, 180 and 210°C were 32, 39 and 47%, respectively, as shown in Figure 5.10. Figure 5.10 also shows the moisture excluding efficiency (MEE) in high humidity tests for the treated specimens with the corresponding MEE values being 11, 13 and 30%, respectively, for the same treatment as for the WRE values.



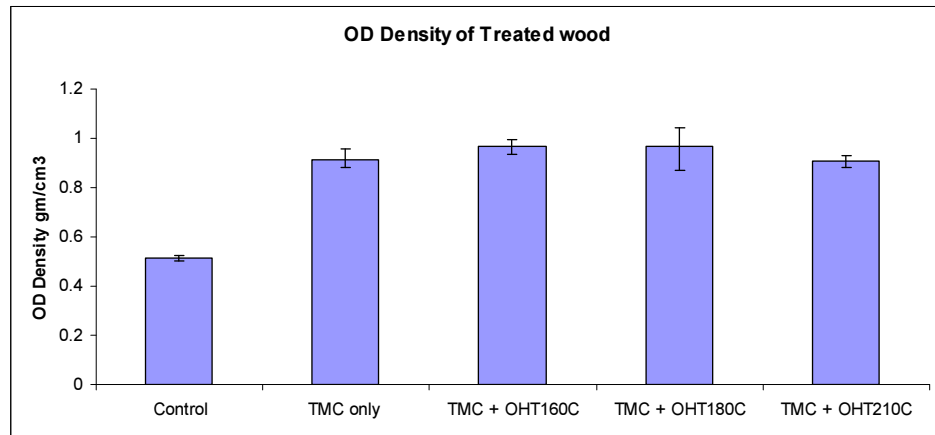
**Figure 5.9. Water absorption percentages after four weeks in the water immersion test. The given values are the means of ten replicates.**



*Figure 5.10. Moisture excluding efficiency and water repellence efficiency of treated wood in comparison to untreated wood in the high humidity test and water immersion test respectively*

#### **5.4.4. Oven-dry density and mechanical properties**

The oven-dry densities of the treated wood are given in Figure 5.11. As expected the oven-dry density of wood increased significantly after the TMC densification with its value being  $0.92 \text{ g/cm}^3$  which was about 80% higher than that of uncompressed specimens ( $0.51 \text{ g/cm}^3$ ). The oven-dry density for the specimens of TMC densification followed by OHT treatment was further increased with 160 and 180 °C temperature (up to  $0.97 \text{ g/cm}^3$ ). This increase in density may be due to net oil uptake during the OHT treatment. However, for OHT treatment at 210°C, the oven-dry density of the wood decreased slightly to  $0.91 \text{ g/cm}^3$ , indicating a net wood mass loss at the high temperature.



**Figure 5.11. Oven dry density of treated wood. The given values are an average of 10 specimens**

To assess the effect of various treatments on the mechanical properties of the treated wood, strength, modulus of elasticity and hardness of untreated, OHT treated (at 210°C) without densification, TMC densified and TMC+OHT (at 210°C) treated wood were determined and compared. Table 5.5 presents the mean strength and MOE in three point bending tests of the treated wood. The MOR and MOE of the untreated radiata pine were 76 MPa and 6.7 GPa respectively. The corresponding results for the OHT treated wood (without TMC densification) were, respectively, 67MPa and 6.5 GPa. Thus, oil heat-treatment at 210°C resulted in 12% loss in the strength and 3% in MoE. The strength (MOR) and elastic modulus (MOE) values for the TMC densified wood were 139 MPa and 9.6 GPa, thus, the TMC densification resulted in an 83% and 43% increase in the strength and the elastic modulus, respectively, compared to the untreated wood. With OHT treatment following the TMC densification, the wood MOR was 110 MPa which is 20% lower than that of TMC densified wood, but was 45% higher than the untreated wood. There was no reduction observed in the elastic modulus in the TMC+OHT treated wood compared to the TMC densified wood.

**Table 5.5. Elastic modulus and strength of treated and untreated wood in the static bending test. The given values are averages of 15 specimens and SD is standard deviation. Y+ and Y- are the upper and lower range from the mean respectively.**

<i>Treatment</i>	<i>MOR (MPa)</i>				<i>MOE (GPa)</i>			
	Mean	SD	Y+	Y-	Mean	SD	Y+	Y-
Untreated	76	4.7	6.8	9.9	6.7	0.7	1.4	1.0
OHT at 210°C	67	5.8	8.6	11.1	6.5	0.8	1.5	1.1
TMC only	139	5.6	9.9	8.6	9.6	1.0	1.8	1.5
TMC+OHT 210 °C	110	6.0	6.6	11.1	9.6	1.0	1.8	1.5

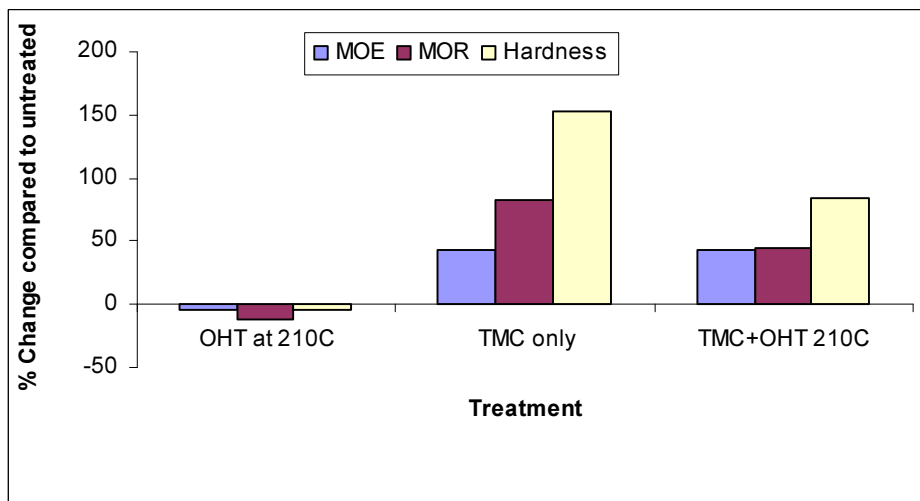
The results of wood hardness obtained from Janka indentation tests are given in Table 5.6 for various treatments. The total hardness of OHT treated wood without densification was 1.77 kN which was 5% lower than the untreated wood of 1.9 kN. The TMC densified wood resulted in hardness of 4.7 kN which was more than 150% higher than that of the untreated wood. This is in agreement with previous findings which also noted a significant increase in surface hardness after thermal-mechanical densification of wood (Clevan and Meng 2007; Navi and Girardet 2000). With OHT treatment following the TMC densification, wood hardness was 3.4 kN which was less than that of the TMC densified wood but 84%

higher than that of the untreated wood. As a general trend, the tangential hardness was greater than that in the radial direction in all cases. The differences between the tangential and the radial hardness were greater in the TMC densified wood. This difference was reduced in the specimens with OHT treatment following TMC densification.

Figure 5.12 illustrates the percentage changes in mechanical properties in treated wood compared to those of untreated wood. It clearly shows that the mechanical properties of wood decreased with OHT treatment, but strongly increased with TMC densification. In the case of TMC densification followed by OHT treatment, the wood strength and hardness were lower than those of the TMC densified wood but were still significantly higher than those of the untreated wood. The improvement in the wood mechanical properties with the TMC densification or TMC followed by OHT treatment is mainly due to the increase in wood density as a result of compression (Figure 5.11). A strong relationship between mechanical properties and wood density have been reported in the literature (Dinwoodie 2000; Tabarsa and Chui 1997). The greater increase in hardness compared to strength and elastic modulus is in agreement with observations of Bowyer *et al.* (2003). Unlike MOR and MOE, the relationship of surface hardness with density is an exponential function and the hardness increases more rapidly with the density, thus, the hardness is more affected by the TMC densification than strength and elastic modulus.

**Table 5.6. Hardness of untreated wood and treated wood. The given values are averages of 15 specimens and values in bracket are standard deviation**

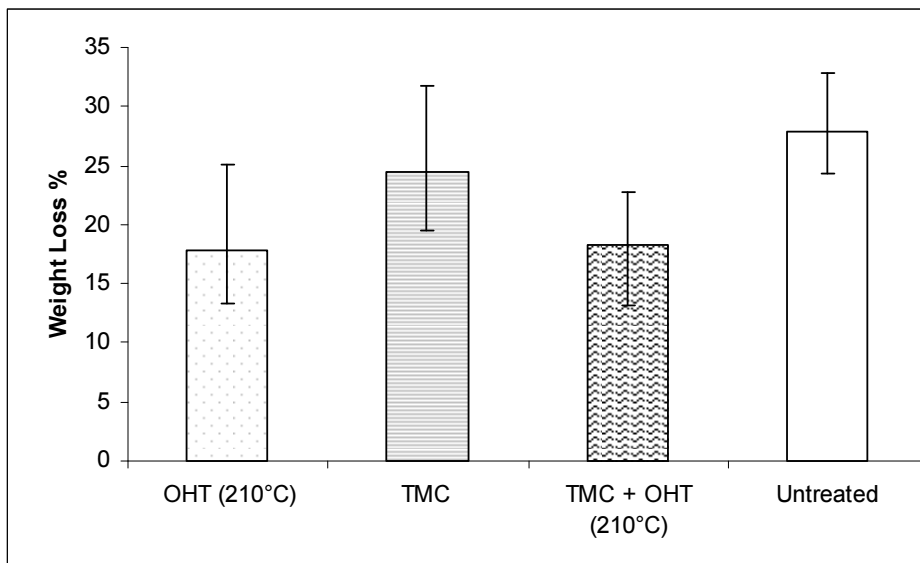
<i>Treatment</i>	<i>Hardness (N)</i>		
	<i>Radial</i>	<i>Tangential</i>	<i>Total</i>
Untreated	1678 (149)	2040 (196)	1859 (133)
OHT at 210°C	1633 (177)	1908 (154)	1771 (97)
TMC	3931 (429)	5493 (427)	4712 (291)
TMC+OHT at 210°C	3319 (278)	3531 (289)	3425 (269)



**Figure 5.12. Percentage change (compared to untreated wood) in mechanical properties after OHT, TMC wood and TMC+OHT**

#### 5.4.5. Fungal resistance

The resistance of treated wood against a brown rot fungi *Oligoporus placenta* was assessed in an accelerated laboratory fungal decay test. Figure 5.13 gives the weight loss percentages of both treated and untreated wood. The weight loss percentage of the untreated wood was 28% which was reduced to about 17.8, 24.4 and 18.3%, respectively, for wood with OHT treatment at 210°C, wood with TMC densified and wood with TMC densification followed by OHT treatment at 210°C. Among all of the treatments, oil heat-treatment gave the most fungal resistance, improving weight loss by 36% compared to the untreated wood. The TMC densified wood showed only a 12% improvement in the fungal resistance over the untreated wood, however, this can be further improved with oil heat-treatment following the TMC densification which was equivalent to that of the OHT treated wood. The improvement in fungal resistance in heat-treated densified wood was also observed by Welzbacher *et al.* (2005) who have noted the improved fungal resistance in densified spruce wood against a brown rot fungus *Poria placenta*. The possible reasons for improved fungal resistance of OHT treatment, TMC densification or OHT treatment following TMC densification are unfavourable conditions through the treatment for fungal growth such as lack of moisture and chemical and structural changes in wood substrates. Skyba and Schwarze (2005) examined fungal colonization in thermo-hydro-mechanically densified wood and noted that fungal hyphae failed to grow in wood cells with completely occluded cell lumina and were confined to partly occluded lumina. Thus, in addition to chemical and structural changes as a result of pressing temperature and oil heat-treatment, restriction of fungal growth due to occlusion of cell lumina as a result of densification is a contributing factor in the improved fungal resistance.



*Figure 5.13. Weight loss percentage of treated radiata pine Sutter blocks exposed to a brown rot fungi *Oligoporus placenta*. The given values are averages of 8 specimens and SD is standard deviation.*

## 5.5. Conclusions

In this study, radiata pine wood was thermo-mechanically compressed (TMC) followed by oil heat-treatment (OHT) at different temperatures. After the treatments colour, dimensional stability, mechanical and biological properties were measured and compared.

Radiata pine wood was compressed effectively to the higher compression set (39%) without any surface defects and cracks. Deformation took place more in the earlywood band than in latewood. The OHT treatment following TMC densification of wood resulted in improved dimensional stability. The spring back in the TMC densified wood was about 6% which further decreased to approximately 2% followed with OHT treatment, with the extent of improvement depending upon OHT treatment temperature. On exposure to water or high humidity conditions, the oil heat-treatment following the TMC densification showed much less



swelling and low compression set recovery. The compression deformation was almost fixed after the OHT treatment at 210°C with only less than 4% set recovery after 672hrs in high humidity conditions. The mechanical properties of the TMC densified wood followed by OHT treatment were less than those of the only TMC densified wood but were still significantly better than those of the untreated wood. The strength, elastic modulus and surface hardness of the TMC densified wood followed by OHT treatment at 210°C were, respectively, 45%, 43% and 84% higher than those of the untreated wood. The wood colour with this combined treatment was darker than that of only OHT densified wood. It was also shown that the treated wood with a combination of TMC followed by OHT was 36% more resistant to a brown rot fungus than the untreated wood.

Thus in general, this study has shown that the wood treated with a combination of TMC followed by OHT leads to low spring back and set recovery percentages even at about 39% compression which, in turn, shows improved mechanical properties. Therefore, controlled densification of wood can improve the mechanical, stability and durability properties of plantation wood. It is recommended that the aforesaid properties of wood at the lower compression level be determined because a loss of saleable wood volume is a major drawback to the wood densification process.

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# Chapter 6

## ACCELERATED WEATHERING OF OIL HEAT-TREATED WOOD

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### 6.1. Introduction

The aim of this part of the study was to investigate the colour and dimensional stability of oil heat-treated wood after an accelerated UV weathering test.

The colour of wood is very important for applications of aesthetic purposes. Colour preferences are cultural and depend on the end use and location and change with time. Some markets such as Scandinavia and Japan prefer light coloured wood for certain end uses (Esteves *et al.* 2008; Wahl *et al.* 2002), whereas in South Asia people like dark coloured wood as it is considered to be a symbol of strength, stability and durability (Dubey 2001). Similarly, in Portugal also darker woods are preferred as the dark colour is associated with high quality tropical species (Esteves *et al.* 2008). In the latter markets mentioned above, species of pine, poplar and eucalypts are considered to be less attractive owing to their light or pale colour. In addition, homogenous coloured wood is considered to be an advantage for use on large surfaces such as flooring. This makes certain species like black locust (*Robinia pseudoacacia*) less preferable for such applications owing to its non-homogenous colour caused by deposition of extractives (Tolvaj and Molnar 2006). Similarly, red alder wood (*Alnus rubra* Bong.) acquires a

mottled orange colour after felling which is undesirable for decorative applications (Thompson *et al.* 2005).

Heat-treatment of wood in varying environments resulted in darkening of wood and improved dimensional stability (Bekhta and Niemz 2003; Gonza' lez-Pena and Hale 2009; Stamm 1964; Stamm *et al.* 1946; Syrjänen and Kangas 2000). Unlike traditional wood finishing or painting, it does not involve any emission of health hazardous volatile organic compounds such as toluene and xylene (Mitsui 2004). Therefore, the heat-treatment could be an effective and environmentally friendly way to darken the wood colour and, at the same time, to improve wood stability (Gonza' lez-Pena and Hale 2009; Johansson and Morén 2006).

However, studies also showed that the colour of heat-treated wood is not stable and fades with the time on exposure to UV radiations (Ahajji *et al.* 2009; Militz 2002; Syrjänen and Kangas 2000). In addition to discolouration, Mayes and Oksanen (2002) reported that surface checking occurred for the heat-treated wood on exposing to UV radiations resulting in poor aesthetics. Several reports are available on the study of colour after accelerated weathering of wood heat-treated in air, vacuum, nitrogen or steam. The exposure time to UV radiations in most of the previous studies was not more than 900 hours (Ahajji *et al.* 2009; Ayadi *et al.* 2003; Deka *et al.* 2008; Letourneau *et al.* 2005; Temiz *et al.* 2006). However, investigation of the dimensional stability of heat-treated wood after accelerated weathering is very limited (Feist and Sell 1987) and there has been no published work on the accelerated weathering of oil heat-treated wood.

This study examined the colour changes and dimensional stability of wood, after accelerated UV weathering of treated and untreated wood. Radiata pine wood was heat-treated in linseed oil over a range of

temperatures. Treated and untreated specimens were exposed to an accelerating UV weathering environment for 2100hrs. Changes in colour were studied by a spectrophotometer using the CIE (L\*a\*b\*) colour coordinate system and volumetric swelling percentages were determined after a water immersion test.

## **6.2. Experimental**

### **6.2.1. Oil heat-treatment**

Wood specimens of dimensions 300 mm (long) × 90 mm (wide) × 20 mm (thick) were heat-treated in linseed oil at 160, 180 and 210°C for 3 hours as per the procedure described in Section 4.2 of the thesis.

### **6.2.2. Accelerated weathering test**

After the above oil heat-treatment, control and treated wood specimens were cut into two halves at the mid-thickness along the specimen length, thus each half was 90 x 10 x 300 mm. One of each pair of specimens was exposed for accelerated UV weathering and the other was kept unexposed in a dark room for comparison. The UV weathering tests followed the US standard ASTM G 154-2006. During the UV weathering tests, specimens were exposed to repetitive cycles of UV light and moisturisation under controlled environmental conditions in a weathering box. UVA 340 light was used and moisturisation was produced by spraying the specimens with water. Each test lasted 2100 hours consisting of 175 cycles, each of which included 8 hour UV 340 exposure at  $60 \pm 3^\circ\text{C}$  black panel temperature and 4 hour condensation at  $50 \pm 3^\circ\text{C}$  black panel temperature (ASTM-G154 2006). During the 4 hour condensation period water was sprayed for 15 seconds in every 2 minutes. All of the specimens were placed 100 mm beneath the light source.

### **6.2.3. Measurement of properties**

#### *6.2.3.2. Colour*

Colour change in treated and untreated wood after accelerated UV weathering was examined by visual inspection and quantified through CIE (Commission internationale de l'éclairage) L\*a\*b coordinates system as per the method described in Section 3.4.1.

#### *6.2.3.3 Dimensional stability*

Dimensional stability was determined by measuring the volumetric swelling percentage of the specimens which were immersed in a water bath at a controlled temperature of 20°C. During the tests, the water in the water bath was re-circulated continuously to maintain the required temperature (20°C). The specimens were weighed every 24 hours and, once the weight changes of the test specimens were less than 0.1%, it was assumed that the specimens had reached equilibrium. Then the dimensions in longitudinal, width (tangential) and thickness (radial) directions were measured to an accuracy of ±0.01 mm. The volumetric swelling percentages were determined by Equation (6.1) given below.

$$S = \left( \frac{V_2 - V_1}{V_1} \right) \times 100\% \quad (6.1)$$

Where  $V_1$  is the oven-dry volume of the specimens before water immersion ( $\text{cm}^3$ ) and  $V_2$  is the volume of the specimens after the water immersion ( $\text{cm}^3$ )

### **6.3. Results and discussion**

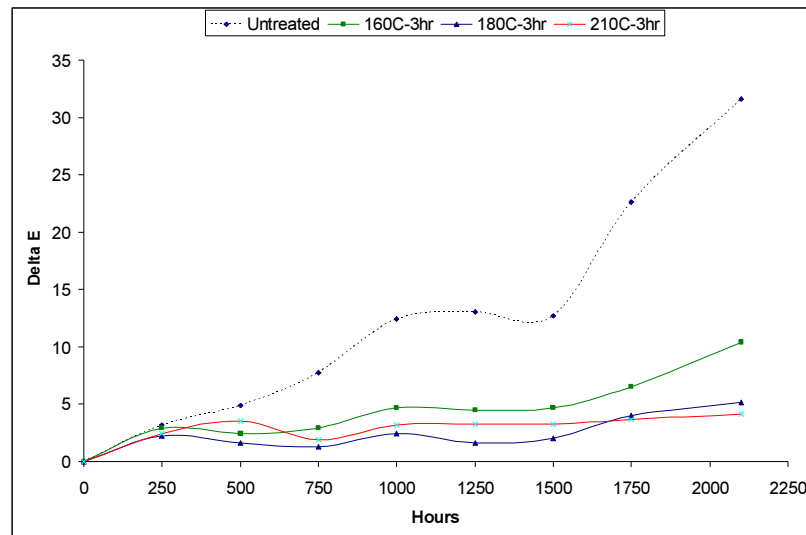
Representative colour changes after 2100hrs accelerated UV weathering test are shown in Figure 6.1 for both heat-treated and untreated samples.



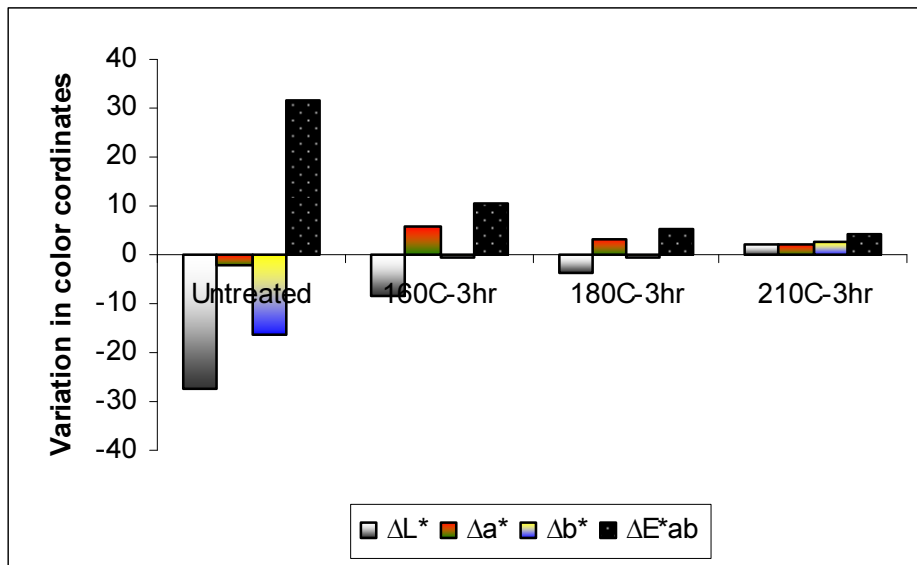
The unexposed and exposed samples belong to the same piece cut from middle of the thickness of the same specimen. Figure 6.2 shows the results of total colour change ( $\Delta E^*$ ) during the accelerated UV weathering as a function of exposure time. The untreated specimens displayed the greatest total colour change throughout the exposure period. Among the treated wood, values of  $\Delta E^*$  for specimens treated at 160°C were generally greater than those treated at 180 and 210°C. The  $\Delta E^*$  values of specimens treated at 180°C and 210°C were very close.



**Figure. 6.1. Comparison of colour in unexposed and exposed specimens after 2100 hrs of accelerated weathering..**



**Figure. 6.2. Total colour change for untreated and heat-treated wood with accelerated UV weathering tests for different exposure times**

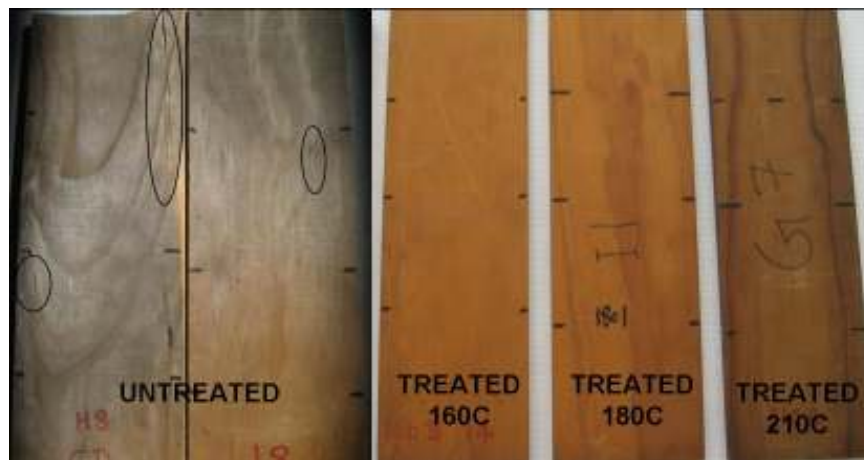


**Figure 6.3. Changes in CIELAB coordinates after 2100 hrs of accelerated UV weathering**

Figure 6.3 compares changes in chromatic coordinates ( $\Delta a^*$ ,  $\Delta b^*$ ) and the lightness coordinate ( $\Delta L^*$ ) after 2100 hrs of exposure to the accelerated UV weathering. The untreated wood clearly showed greater total colour change ( $\Delta E^* = 32$ ) than the treated wood ( $\Delta E^* = 4-10$ ). The darkness and blueness of the untreated wood increased significantly with weathering ( $\Delta L^* = -27$  and  $\Delta b^* = -16$ ), but after oil heat-treatment the colour of radiata pine wood was more stable and colour stability generally increased with treatment temperature. For example, the total colour change ( $\Delta E^*$ ) was 10 for wood treated at 160°C compared to corresponding values of 5 for wood treated at 180°C and 4 treated at 210°C. All specimens were darker without any fading ( $\Delta L^* = -4$  to  $-8$ ) except those treated at 210°C which slightly faded ( $\Delta L^* = 2$ ) during the weathering. Interestingly, specimens treated at 160°C became redder as was evident from the increase in the value of the red/yellow chromatic coordinate ( $\Delta a^* = 6$ ) in Figure 4 (b). In all of the observations of wood

colour change through CIE (L\*a\*b\*) system, the lightness coordinate (L\*) was the most affected and sensitive parameter.

Figure 6.4 shows weathered surfaces of untreated and oil heat-treated wood. The surfaces of treated wood were smooth without or with only minor checks on visual inspection. However, clearly visible cracks and checks appeared on the untreated specimens.



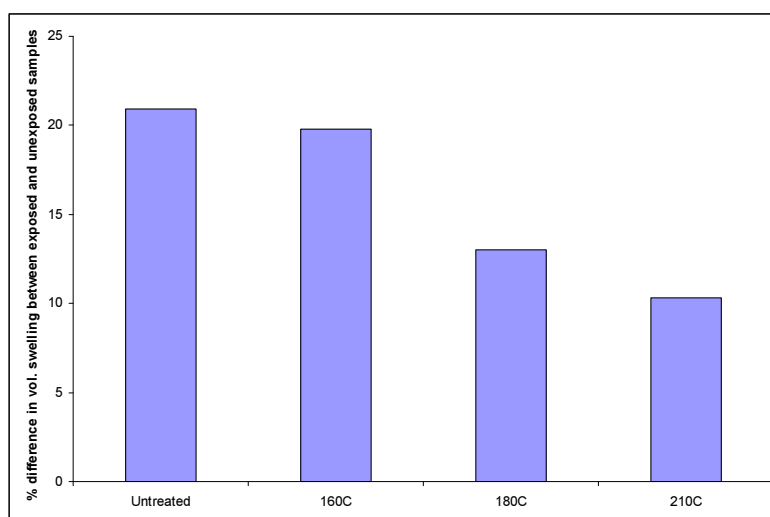
***Figure 6.4. Surfaces of treated and untreated wood after accelerated weathering. The cracks and checks which appeared on untreated wood are marked.***

Table 6.1 presents the results of volumetric swelling in water immersion tests. The results include the influences of oil heat-treatment (unexposed to weathering) and accelerated UV weathering (exposed to weathering). After weathering the volumetric swelling in untreated wood was 32-55% more than that of the treated wood. With the treated wood the volumetric swelling decreased with increasing treatment temperature, which was true both for the unexposed and exposed to the UV weathering. However, with the UV weathering, the untreated wood gave higher increases in the volumetric swelling than the treated wood. For example, the volumetric swellings of the untreated wood increased from 13.8% to 17.5% with the UV exposure while the specimens treated at 210°C increased from 7% to

7.9%. The difference in the volumetric swelling between exposed and unexposed specimens was about 21% for untreated wood, while for the treated wood this difference was about 20%, 13% and 10% for the specimens treated at 160, 180 and 210°C, respectively (Figure. 6.5).

**Table 6.1. Volumetric swelling of wood specimens in water immersion tests. The given values are an average of six replicates and SD is standard deviation.**

Treatment	Volumetric swelling (%)			
	Unexposed to weathering		Exposed to weathering	
	Mean	SD	Mean	SD
Untreated	13.9	1	17.5	0.9
OHT at 160°C	9.5	0.4	11.9	1.2
OHT at 180°C	9.5	0.7	11	0.7
OHT at 210°C	7	0.6	7.9	1.1



**Figure. 6.5. Difference in volumetric swelling percentage (after two weeks of water immersion) between exposed and unexposed specimens to UV weathering.**

In this study, no fading of treated wood was observed after weathering except for wood treated at 210°C which was slightly faded. This result was in line with the findings of Deka *et al.* (2008) who noted improved colour stability of heat-treated wood (in a vacuum at 210°C for 2 hours) during UV light irradiation for 400 hours. In contrast, Syrjänen and Kangas (2000) reported that heat-treated wood starts fading if not protected from UV light and Ahajji *et al.* (2009) observed mixed results depending upon the species in the artificial weathering of wood heat-treated at 210-250°C for one hour. Thus, oil heat-treated wood demonstrated better colour stability than untreated wood after the accelerated UV weathering test.

Similarly, after accelerated UV weathering, volumetric swelling of the treated wood increased less than that of the untreated wood which indicates treated wood is more stable. The oil heat-treated wood did not show noticeable checks with the UV weathering. This differs from Mayes and Oksanen (2002) who reported that surface checks after weathering were observed for wood heat-treated at 70-240°C for 3 hours in the steam environment.

The superior stability of colour and dimensional changes of the oil heat-treated wood compared to untreated wood during artificial weathering might be the result of an increase in lignin stability owing to it undergoing condensation reactions during heat-treatment (Ayadi *et al.* 2003; Deka *et al.* 2008). Another possible reason for these improvements with the oil heat-treatment can be related to the lower moisture absorption by the oil heat-treated wood. Apart from the UV radiation, moisture is a critical factor in photo-induced degradation of wood as it facilitates light penetration into the wood surface (Ayadi *et al.* 2003). Owing to the high linolenic acid (C18:3) content, the linseed oil has a tendency to fast dry and harden on the wood surface after the oil heat-

treatment (Gunstone 2002). This process forms a protective layer of oil on the surfaces of the treated specimens. Therefore, the oil layer acts as a barrier for the UV ray penetration and for water absorption, and thus improves the water repellency in the treated wood.

## 6.4 Conclusions

This study examined the colour change and dimensional stability of oil heat-treated radiata pine wood after accelerated UV weathering.

Oil heat-treated wood exhibited better colour stability after the UV weathering test. No colour fading was observed in the specimens treated at 160 and 180°C temperatures for 3 hours and those treated at 210°C slightly faded. In general, no surface checks were observed on the weathered surfaces of treated specimens. The percentage swelling after weathering in treated specimens was less than for untreated wood.

The results of this study indicate that oil heat-treatment can be an effective way to darken the wood with improved stability. The improved performance of oil heat-treated wood in the weathering test can be explained by the changes in wood chemical constituents and the formation of a protective oil layer on the wood surfaces.

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# Chapter 7

## AGEING EFFECT OF HEATING OIL ON THE PROPERTIES OF HEAT- TREATED WOOD

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### 7.1. Introduction

The objectives of this part of thesis were to determine the effect that the age of the heating medium (linseed oil) has on wood properties and to determine the period for which the oil can be reused.

Linseed oil is an industrial or drying oil, obtained from an annual herb, flax (*Linum usitatissimum*). The oil is used in many industries such as paints, resins, inks, soaps, varnishes, wood treatments and linoleum due to its excellent drying properties (Ulvcrona *et al.* 2005). The high linolenic acid (C18:3) content of the linseed means that the oil dries fast and hardens under sunlight (Treu *et al.* 2001). The advantages of the linseed oil, such as improved wood protection, ease of drying after treatment, wide availability and high boiling point (higher than 334°C), make this oil a preferred candidate for use in the heat treatment of wood. However, the linseed oil converts to “stand oil” when heated for a prolonged time at high temperature, which is a thickened heat-polymerised oil (Chemwatch 2007; Tollenaar and Bolthof 1946). Any change in consistency or viscosity of the oil with recycling may affect wood properties after treatment, about which no information has been found in literature.

In this study *Pinus radiata* wood specimens were heat-treated in a bath using linseed oil. The aging effect of the oil was investigated under the treatment condition of 180°C and 3 hrs. Before the wood treatment, the oil had been preheated for up to 27 hrs. After the treatment with oils of varying preheating periods, changes in oil viscosity and wood properties were examined with the oil preheated for different time. The results were compared with untreated specimens and specimens treated in fresh oil. The effect of oil viscosity on these properties was also examined.

## **7.2. Experimental**

### **7.2.1 Treatment**

The wood specimens with dimensions of 200 mm (long) x 90 mm (wide) x 35 mm (thick) were prepared as described in Section 3.2.2. The specimens were then oven-dried for 48 hrs, cooled over silica gel in a dessicator and weighed before being placed in a conditioning chamber under conditions of 65% RH and 20°C for 2-3 weeks to attain equilibrium.

After having reached the equilibrium with the moisture content of 10-12 %, the specimens were heated in an oil bath using commercial grade raw linseed oil (M/S Mainland Paint and Printing Ink Ltd.) as the heating medium. Eight specimens were treated in each run. In the first run fresh raw linseed oil was used. The oil was heated to 180°C before the specimens were immersed and then these specimens were treated for 3 hrs. During the heat treatment, the oil was re-circulated continuously and no external pressure was applied.

Once being treated, the specimens remained in the oil bath to cool down for 15 minutes then the oil was drained. Next the treated specimens were oven-dried for 48 hrs, cooled over silica gel in a dessicator and weighed.

Finally, the specimens were placed in a conditioning chamber under conditions of 65% RH and 20°C for 2-3 weeks until equilibrium was attained.

The above experiment was repeated by re-using “used” linseed oil that had been heated to a temperature of 180°C for 3, 9, 15, 21 and 27 hrs, respectively.

## **7.2.2. Property measurement and determination**

### *7.2.2.1. Wood properties*

The weight percentage change and colour change between, end-matched treated and untreated specimens were measured using the procedures described in sections 3.4.1 and 3.4.2, respectively. Dimensional stability and moisture repellency were determined by water immersion tests as described in section 3.4.4.

### *7.2.2.2. Oil Viscosity*

Viscosity of fresh linseed oil and pre-heated linseed oil were measured using a Haake viscometer with MV1 sensor with a shear rate of 1000 per second, measured at  $20.2 \pm 0.1^\circ\text{C}$  (Figure 7.1). The mean of 5 replicates was used for each condition. In addition, the oil was heated at 180°C for 3 hrs with and without wood specimens in the oil to compare the effect of oil heating in presence of wood on the oil viscosity. The mean of 10 replicates was used for this examination.



*Figure 7.1. Haake viscometer used for measurement of viscosity of linseed oil*

## **7.3. Results and Discussion**

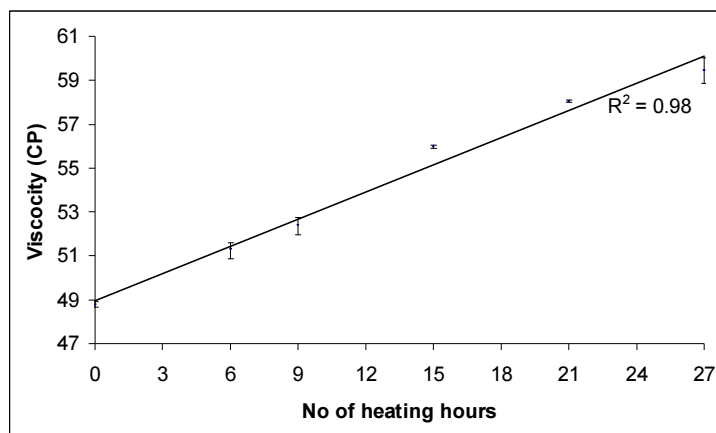
### **7.3.1. Colour and viscosity of the oil**

The colour of fresh linseed oil is pale yellow. With increased heating time (27 hrs), the oil colour changed to dark brown as shown in Figure 7.2. The mean viscosity of the oil increased linearly with the heating time, changing from around 49 cp for the control (fresh oil) to 59 cp after 27 hr heating (Figure 7.3). The increase in the oil viscosity is mainly due to evaporation of volatile components and polymerisation of the linseed oil at high temperatures (Mabery 1923; Powers 1950).

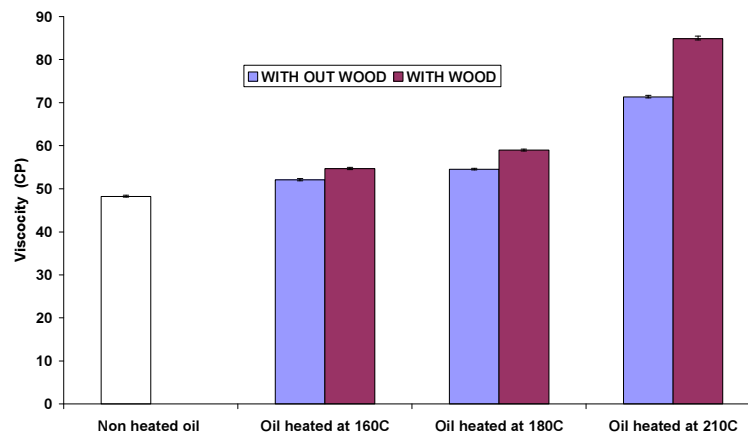
Figure 7.4 compares the oil viscosity between heating with wood and heating without wood in it. The viscosity of the oil heated with wood was higher than that without wood and this difference was increased with the treatment temperature. This confirms the view that the release of wood extractives and wood degradation products contributes, to some extent, to the increase in the oil viscosity during the wood heat treatment.



**Figure. 7.2.** Colour change in linseed oil with prolonged heating, from left to right showing fresh oil, and that heated for different hrs



**Figure. 7.3.** Effect of prolonged heating on viscosity of linseed oil (the given values are mean of 5 replicates).



**Figure. 7.4.** Mean viscosity of fresh oil and oil heated with and without wood. The values are mean of 10 replicates.

### 7.3.2. Weight percentage change

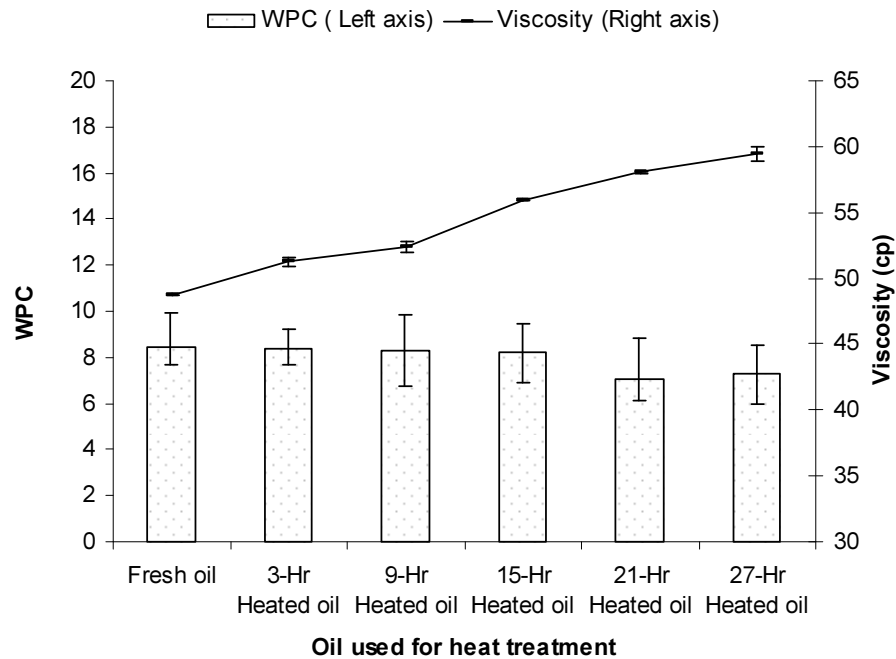
Table 7.1 presents the percentage weight change after treatments with linseed oil that had been preheated for different period of time. The mean weight of the treated specimens was increased by 7 to 8.4%, with the greatest increase occurring with fresh oil. The weight gain of the treated specimen was affected by the cumulative oil pre-heating time as shown in Figure 7.5. The wood treated in fresh oil gained around 16% more weight than those treated in the pre-heated oil. The lower weight gains using the oil pre-heated for 21 and 27 hrs may be due to poorer penetration of the oil, resulting from the increase in oil viscosity when the oil was preheated.

***Table 7.1. Weight percentage change (WPC) of wood after treatment.***

***The values are mean of 5 replicates and SD is the standard deviation.***

***Y+ and Y- are upper and lower range from mean respectively.***

Treatment medium	WPC%	SD	Y+	Y-
Fresh oil	8.4	0.7	1.5	0.8
3-hrs heated oil	8.4	0.4	0.8	0.7
9-hrs heated oil	8.3	0.9	1.6	1.6
15-hrs heated oil	8.2	0.8	1.2	1.4
21-hrs heated oil	7.1	0.9	1.8	1.0
27-hrs heated oil	7.3	0.8	1.3	1.4



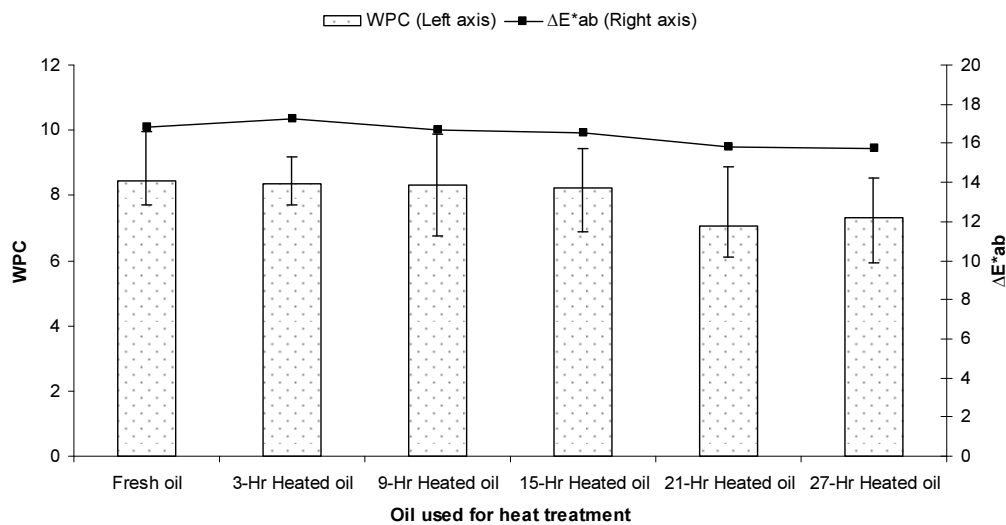
*Figure. 7.5. Effect of age of the heating oil used in wood treatments on the specimen weight percentage change and oil viscosity.*

### 7.3.3. Wood colour change

Wood colour changed significantly after the oil heat treatment with the specimens turning brownish yellow. The total colour change between the untreated specimens (controls) and the heat-treated specimens was more than 15 units (Table 7.2). Among the colour coordinates, the lightness ( $L^*$ ) of the treated wood decreased remarkably whereas the reddish ( $b^*$ ) and the yellowness ( $a^*$ ) increased, to some extent, after the oil heat-treatment. The greater value in the changes of  $\Delta b^*$  compared to  $\Delta a^*$  indicates more yellowing of the treated wood than the changes in reddish. However, colour changes were broadly similar among wood treated in oils being pre-heated for different durations and the differences between

different pre-heating duration were difficult to differentiate by the naked eye.

This is further supported by the data presented in Table 7.2 and Figure 7.6. The differences in the total colour change between wood specimens heat-treated using fresh oil and preheated oils were less than 1.2 units. Generally, the human eye is unable to differentiate colour change of less than 2-3 units (Sundqvist and Morén 2002). This implies that the effect of oil heating age on wood colour was insignificant when the oil age was equal to or less than 27 hrs.



**Figure. 7.6. Percentage weight change (WPC) and total colour change of heat treated wood using fresh oil or oil preheated for different time.**



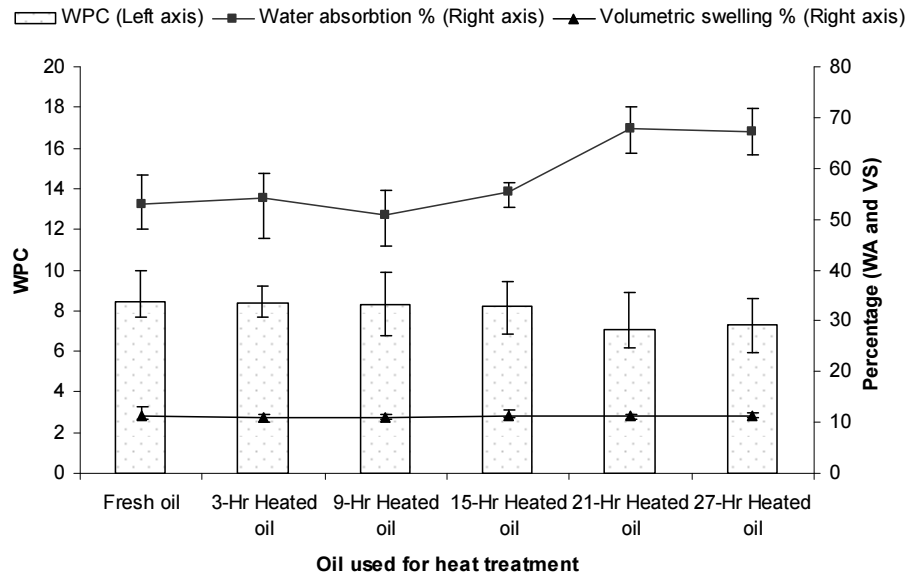
**Table 7.2. Change in colour coordinates (CIE 1976-L\*a\*b\*system)  
after treatment at 180°C.**

Treatment in fresh oil or preheated oil	Colour coordinates			Change in colour coordinates of treated specimens <u>compare to untreated specimens</u>				Change in colour coordinates of specimens treated in heated oil <u>compare to specimens treated in fresh oil</u>			
	L*	a*	b*	$\Delta L^*$	$\Delta a^*$	$\Delta b^*$	$\Delta E^*$	$\Delta L^*$	$\Delta a^*$	$\Delta b^*$	$\Delta E^*$
Specimens	L*	a*	b*	$\Delta L^*$	$\Delta a^*$	$\Delta b^*$	$\Delta E^*$	$\Delta L^*$	$\Delta a^*$	$\Delta b^*$	$\Delta E^*$
Controls	75.5	6.6	23.7	0.0	0.0	0.0	0.0				
Fresh oil	61.4	10.8	31.8	-14.1	4.1	8.2	16.8	0.0	0.0	0.0	0.0
Preheated oil for 3hr	60.8	11.3	31.3	-14.8	4.7	7.7	17.3	0.6	0.6	-0.5	0.9
Preheated oil for 9hr	61.3	11.4	31.1	-14.2	4.7	7.4	16.7	0.6	0.6	-0.8	1.2
Preheated oil for 15hr	61.6	11.4	31.2	-14.0	4.8	7.5	16.6	0.6	0.6	-0.6	1.1
Preheated oil for 21hr	62.6	10.8	31.6	-12.9	4.2	8.0	15.8	0.3	0.3	-0.2	0.4
Preheated oil for 27hr	62.4	11.0	31.3	-13.1	4.3	7.6	15.8	0.2	0.2	-0.6	0.6

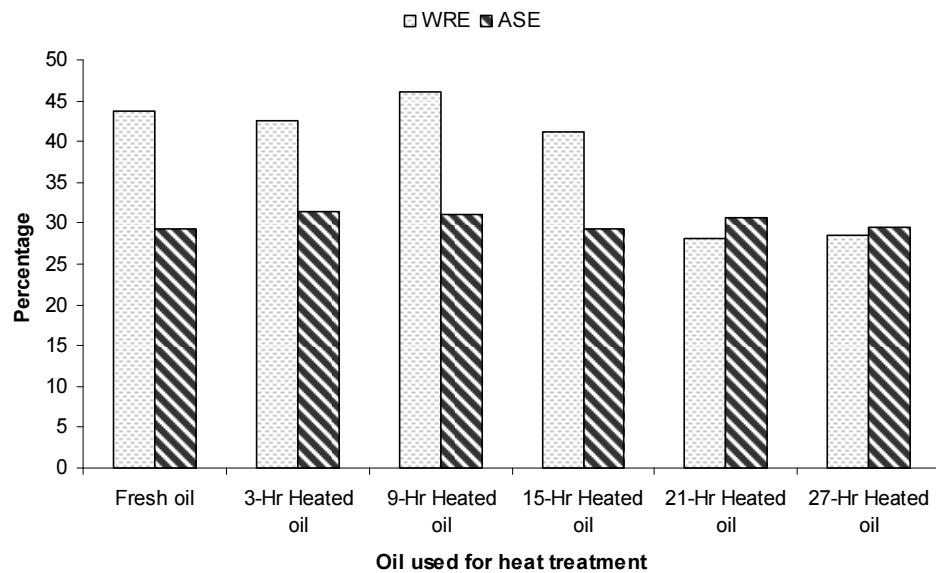
#### **7.3.4. Water repellency and dimensional stability**

As shown in Figure 7.7 and 7.8 wood became more water repellent and dimensionally stable after the heat treatment. The heat-treated wood achieved water repellence efficiencies of between 28 to 46% indicating less water uptake than untreated wood. Similarly, volumetric swelling of the treated wood decreased by 29-31%. There was almost no difference in water absorption for wood treated when using fresh oil and preheated oil for less than 21 hrs. However, wood treated using oil preheated for 21 hrs and 27 hrs showed a significant increase in the water absorption (Figure 7.7) and significant drop (around 35 %) in WRE (Figure 7.8). This was probably due to less oil being absorbed by these specimens compared to those treated in fresher oil.

However, no significant difference was found in volumetric swelling percentage among different treatments indicated by the anti swelling efficiency (ASE). This indicates that oil uptake has more influenced on water absorption than on volumetric swelling. The increase in dimensional stability of oil heat-treated wood was mainly due to chemical changes within wood at high temperature and was not much affected by oil uptake. This suggests that oil uptake by wood specimens has more influence on water absorption than the volumetric swelling and improvement in dimensional stability is mainly due to chemical changes caused by heat-treatment as discussed earlier in Section 4.3.1 of the thesis and also reported in a previous study (Wang and Cooper 2005).



**Figure. 7.7. WPC, water absorption percentage and volumetric swelling percentage of specimens heat-treated using oil of different heating age.**



**Figure. 7.8. Water repellence efficiency and anti-swelling efficiency of heat-treated wood using oil of different heating age compared to untreated control.**

## 7.4. Conclusions

Oil aging was firstly examined by preheating the oil from 3 up to 27 hrs, and the results show that the oil age increased the oil viscosity by up to 22%.

After oil heat-treatment of radiata pine wood at 180°C for 3 hrs either in fresh linseed oil or in preheated oil, it is found that the wood was darkened and became more water repellent and dimensionally stable. Due to the viscosity increase with the oil age, the oil uptake in the treated wood decreased by 16.2% for the oil preheated for 21 and 27 hrs. Water repellence efficiency of the oil heat-treated wood also decreased (around 35%) by using the preheated oil for the heat treatment. However, no significant difference was observed on total colour change and anti swelling efficiency of the treated wood for the wood heat treated either using fresh oil or using preheated oil. This implies that improvement in dimensional stability of the heat-treated wood in oil was mainly due to chemical changes in wood due to high temperature in the heat treatment. Thus, the pre-heated oil can be used for heat treatment without affecting noticeable colour change and dimensional stability of the treated wood. Further work is recommended to investigate the impact of longer heating age of the oil on wood properties in the heat treatment.

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# Chapter 8

## SOLVENT EXTRACTION OF OIL HEAT-TREATED WOOD AND THE EFFECT OF THE COOLING PERIOD ON THE OIL UPTAKE

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### 8.1 Introduction

The objectives of this part of the thesis were (1) to extract oil from oil heat-treated wood and determine the oil uptake; (2) to examine the effect of oil extraction on the hydrophobicity of the treated wood; (3) to investigate the effect of the cooling period on the oil uptake by the treated wood.

In previous chapters the oil uptake as estimated by the percentage change in the oven-dry weight of specimens after oil heat-treatment did not reflect the actual oil uptake by wood as there were weight losses of extractives and other heat degraded products for the oil heat-treated wood. The actual oil uptake was believed to be greater than that determined by weight percentage change.

The improved moisture related properties of the oil heat-treated wood are due to the combined effects of chemical changes during the heat-treatment and the oil uptake. Thus, it is also desirable to find the impact of linseed oil extraction on the moisture repellency of wood.

Linseed oil is soluble in solvents which do not cause wood swelling to a measurable extent such as benzene and carbon tetrachloride (Schneider 1980). In this study linseed oil was extracted from the treated wood using the solvents of benzene and alcohol as described in the relevant TAPPI standards (TAPPI-204 1995; TAPPI-207 1995; TAPPI-264 1995). The oil uptake percentages in the treated wood were determined after solvent extraction and compared with that determined based on net weight percentage changes. The moisture repellency in the treated wood before and after solvent extraction was measured to examine the effect of oil removal on the hydrophobicity of the treated wood. In addition, the effect of cooling specimens in the oil bath after heat treatment on oil uptake and moisture related properties were also investigated.

## 8.2 Experimental

### 8.2.1 Determination of oil uptake by wood specimens

The wood specimens with dimensions of 90 mm (wide) x 35 mm (thick) and 200 mm (long) were treated in a linseed oil bath at 160, 180 and 210°C for three hours, following the procedure given in Section 4.2. After the heat-treatment the specimens further remained in the oil bath for a period of 15 minutes with the heating turned off and the lid of the oil bath removed to aid cooling. Then the oil was drained. Oil uptake was determined by the following methods.

#### 8.2.1.1 Based on change in weight percentage

The weight percentage change (WPC) was determined from Equation (8.1):

$$WPC = \left[ \frac{(W_a - W_b)}{W_b} \right] \times 100\% \quad (8.1)$$



$W_b$  is the oven-dry weight of specimens before the treatment (g), and  $W_a$  is the oven-dry weight of specimens after the treatment (g).

#### 8.2.1.2 Based on the extraction of oil by organic solvents

Oil uptake percentage (OUP) was determined after solvent extraction through the following procedures:

1. Extraction of both treated and untreated samples with hot water to remove water soluble materials such as extractives like tannins, gums, sugars, starches and colouring matter.
2. Extraction of untreated samples by an ethanol and benzene mixture (2:1 ratio) for 12 hours and determination of the percentage of extractives soluble in organic solvent. The mass of extracted matter in this way is  $m_A$  (g/g oven dry wood).
3. Extraction of oil heat-treated samples by an ethanol and benzene mixture (2:1 ratio) for 12 hours and determination of percentage extractives and linseed oil soluble in organic solvent. The change in mass of the oil heat-treated wood after solvent extraction is attributed to the combination of extracted oil, removal of solvent soluble extractives and heat degradation products. The mass of extracted matters from the treated wood is  $m_B$  (g/g oven dry wood) which is also based on the original oven dry mass of the wood before oil heat-treatment.
4. The oil uptake percentage (OUP) of the oil heat-treated samples is determined by following equation:

$$\text{OUP} = (m_B - m_A) \times 100\% \quad (8.2)$$

The values for  $m_A$  and  $m_B$  in this study were the average of five replicates. Wood samples for solvent extraction were prepared from the same specimens used for the determination of weight percentage change after heat-treatment. Four slices (two from 20 mm from each end and two from the centre) were cut from the specimen of each treatment. The dimensions of each slice were 90 mm (wide) x 35 mm (thick) and 15 mm (long).

These slices were first broken down in a knife mill followed by grinding in an ultra centrifugal mill *Retsch ZM100*. Finally, the particles were sieved down to 25-40 mesh (0.25-0.4mm) size as recommended in the TAPPI standard (TAPPI-257 1995). The hot water and organic solvent extractions of wood specimens were carried out according to the procedures given in TAPPI standards (TAPPI-204 1995; TAPPI-207 1995; TAPPI-264 1995). The solvents used were a mixture of ethanol (approximately 95 % by volume) and reagent grade benzene in the ratio of 2:1. Owing to health and environmental risks associated with benzene, it was handled following the safety precautions described in the relevant Chemwatch Material Safety Data Sheet (Chemwatch 2008).

### **8.2.2 Moisture absorption after extraction**

The wood particles before and after organic solvent extraction were placed in high humidity conditions ( $85 \pm 5\%$  RH,  $20^\circ\text{C}$ ) and moisture excluding efficiency (MEE) of the treated wood was determined as per Section 3.4.4 of the thesis.

### **8.2.3 Effect of cooling time on oil uptake and moisture related properties**

This investigation was conducted for one treatment temperature with various cooling periods. In the experiments, specimens were heated in

linseed oil at 210<sup>0</sup>C for 3 hours. They were then removed immediately without any cooling period in the treatment oil. The treated specimens were wiped and transferred to an oven at 102<sup>0</sup>C for 24 hours. After this, the specimens were cooled in a dessicator and weighed. Following this, the treated specimens were conditioned at 65% RH and 20<sup>0</sup>C. Subsequently seven more runs were conducted using new specimens which were firstly oil-heat-treated at the same temperature and for the same duration as in the above. However, after the end of the 3 hour treatment period the heating was switched-off, the lid of the oil bath removed and oil was allowed to cool in atmospheric condition. The specimens were removed, respectively, after ½, 1, 1½, 2, 4, 8 and 12 hours of cooling in the oil bath. Six specimens were heated in each run. The temperature of the oil at the time of removal of specimens was noted.

The net oil uptake was determined by changes in the oven-dry weight percentage after the treatment using Equation (8.1). Water uptake and tangential (width) swelling percentages were determined by soaking specimens in a water bath at 20 °C for 168 hrs. The water uptake (WU) and tangential swelling (TS) percentages after water immersion were determined according to Equations (8.3) and (8.4), respectively.

$$WU = \left( \frac{W_2 - W_1}{W_1} \right) \times 100\% \quad (8.3)$$

where  $W_1$  is the oven dry weight of the specimens before water immersion (g) and  $W_2$  is the weight of the specimens after 168 hours water immersion (g).

$$TS = \left( \frac{T_2 - T_1}{T_1} \right) \times 100\% \quad (8.4)$$

where  $T_1$  is the oven-dry tangential dimension of the specimen before water immersion (mm) and  $T_2$  is the tangential dimension of the after 168 hours water immersion (mm).

### 8.3 Results and discussion

Table 8.1 gives the weight percentage change (WPC) in treated samples and the oil uptake percentages (OUP) determined based on solvent extraction. For specimens treated at 160, 180 and 210°C, nominal uptakes were 11.8, 5.2 and 1.4%, respectively.

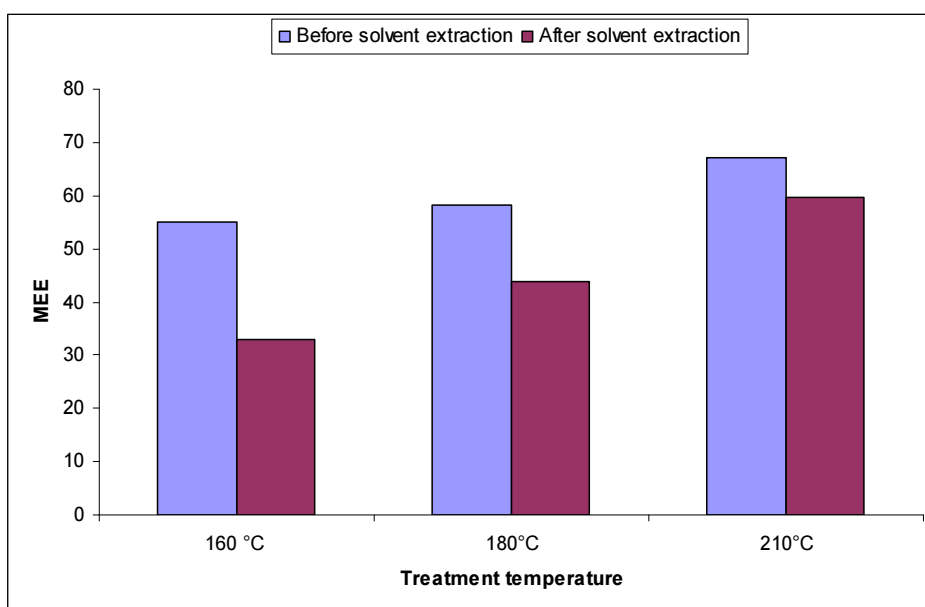
**Table 8.1. Mean oil uptake percentages (OUP) determined by solvent extraction and WPC. Values in parentheses are standard deviations.**

Treatment	WPC after oil heat-treatment (Y, %)	Mass change after solvent extraction (%)	OUP (X, %)	Difference (X-Y, %)
Control	-	0.8 (0.1)		
160°C-3hr	11.8 (1.5)	12.9 (1.3)	12.1	0.3
180°C-3hr	5.2 (0.5)	6.6 (1.2)	5.8	0.6
210°C-3hr	1.36 (0.5)	4.3 (0.5)	3.5	2.1

The change in weight after oil heat-treatment was the result of oil uptake offset by loss of extractives and losses due to thermal degradation of cell wall constituents. This implies that the actual oil uptake was greater than that determined by the nominal change in weight percentage. This was confirmed by the higher values of the oil uptake percentage (OUP) determined after solvent extraction. The OUP determined by the solvent extraction method was 12.1, 5.8 and 3.5 % for the specimens treated at 160, 180 and 210°C, respectively. The differences between the WPC and OUP determined by the two methods were small for 160 and 180°C and greater at 210°C. This implies that the mass loss after oil heat-treatment was greater at the higher treatment temperature, which was in agreement with previous studies which reported that mass losses in wood after heat-treatment in air or steam increased significantly at temperatures higher than 200°C (Esteves *et al.* 2008; Zaman *et al.* 2000).

Figure 8.1 compares the moisture excluding efficiency (MEE) of the oil heat-treated wood before and after solvent extraction. The MEE of the treated wood was 55-67 % depending upon the treatment temperature. After solvent extraction the MEE was reduced to 33-59 %. The MEE was increased with the increase in treatment temperature and this was true for wood both before and after solvent extraction. At all treatment temperatures, the MEE of the heat-treated wood was reduced with solvent extraction. This clearly indicated that oil uptake by wood increased the hydrophobicity of the heat-treated wood. This was further confirmed by the greater difference in MEEs with the 160°C treatments where oil uptake percentage was the greatest. On the other hand, specimens with the lowest oil uptake (treated at 210°C) showed small difference in MEEs. The decrease in MEE after solvent extraction may be due to greater moisture absorption as a result of the increased porosity

of wood due to removal of absorbed oil. However, it should be treated with caution as it is possible that the solvents may also have extracted some substances in wood or produced during the oil treatment that also contributed to prevent moisture uptake.



***Figure 8.1. Comparison of moisture excluding efficiency (MEE) of oil heat-treated wood before and after solvent extraction.***

Table 8.2 presents the mean change in oven-dry weight percentage (WPC) for different cooling periods in the oil bath after the oil heat-treatment. The WPC in wood specimens taken out immediately after the treatment period was less than 2%. No visible surface cracks and warping of the specimens were observed even after removing them without any cooling. The WPC increased rapidly with increasing cooling time with maximum asymptotic oil absorption being approached with a 4 hour cooling period.

These data confirmed that oil absorption mainly occurred during the cooling phase. A probable explanation is that during the heat-treatment process there was no differential pressure between the interior of the

wood and the bulk oil to facilitate oil penetration into wood (Moreira *et al.* 1999). On cooling in the oil bath the residual air in the wood cells contracts resulting in an internal negative pressure sucking the oil into the wood (Awoyemi *et al.* 2009).

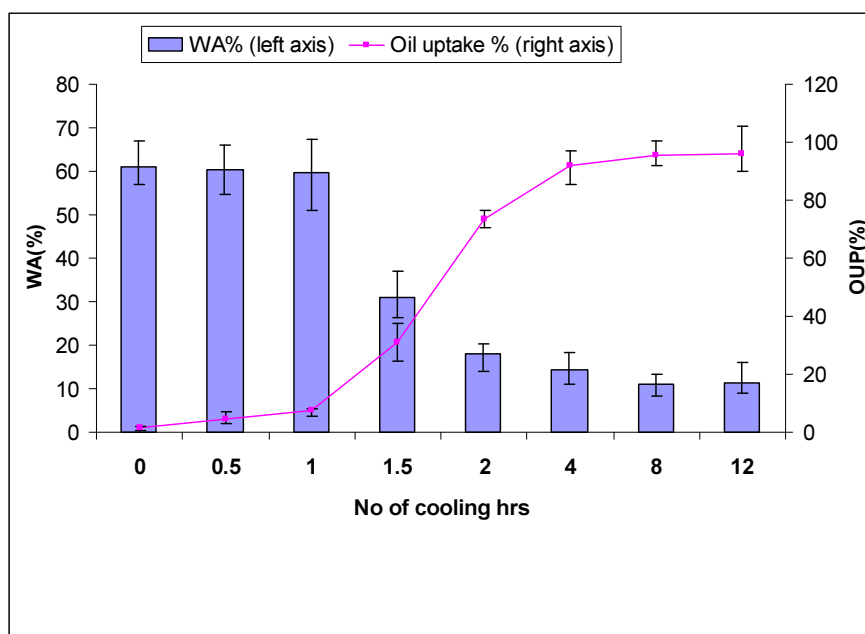
**Table 8.2. Effect of the cooling period after oil heat treatment on the WPC in wood specimens. Values for WPC are the average of six specimens.**

<i>Cooling period (Hr)</i>	<i>Weight percentage change (%)</i>	<i>Standard deviation (%)</i>	<i>Oil temperature (°C)*</i>
0	1.4	0.6	210
0.5	4.6	1.5	192
1.0	7.3	0.8	184
1.5	30.8	5.1	150
2.0	73.7	2.2	132
4.0	91.8	4.8	105
8.0	95.6	2.7	87
12.0	96.1	5.3	55

Note: \* Oil temperature at the time when the specimens were removed from the oil bath.

The oil uptake increasing with cooling time is not simply a factor of time alone, but of the falling temperature in the bath. Although, after removal from the oil bath samples were wiped-off immediately, in between this period (removal from the oil bath and wiping-off) some surface oil absorption may have also taken place in the wood owing to a further drop in temperature (Moreira *et al.* 1999). In summary, oil uptake in wood occurred during the cooling period and uptake increased with the time spent in oil bath during the cooling process.

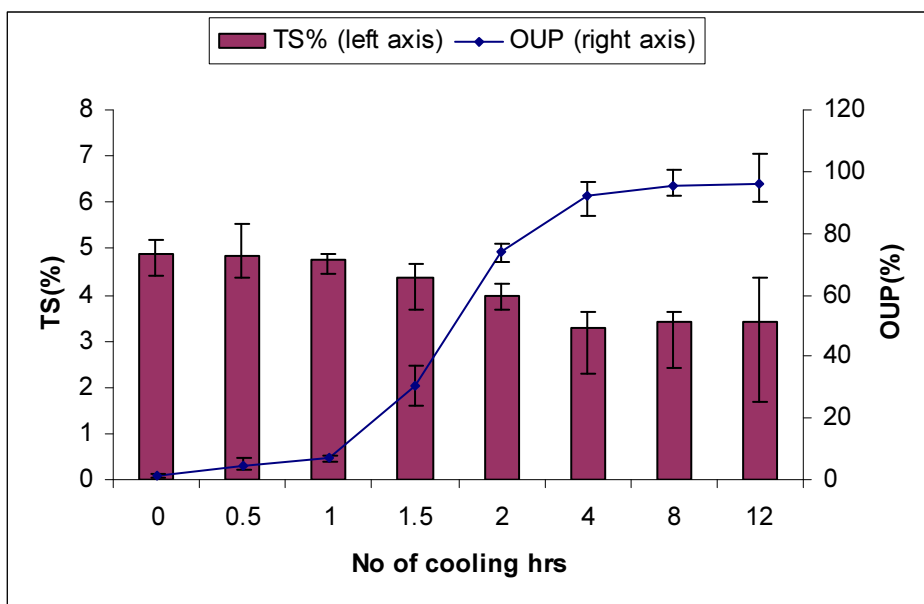
Figures 8.2 and 8.3 show the effect of the cooling period on oil uptake, water absorption and tangential swelling percentages respectively. The water absorption in untreated wood was about 73 % and 11-61% in treated wood. Similarly, tangential swelling in untreated and treated wood was respectively 7.2 % and 3.3-4.9 %. Thus, both the water repellency and dimensional stability of wood increased after the treatment compared to that for untreated wood.



**Figure. 8.2. Effect of the cooling period on the oil uptake percentage (OUP) and water absorption percentage**



Oil uptake increased with the oil bath cooling time while water absorption (WA) and tangential swelling (TS) decreased. As shown in Figure 8.2, the water absorption and the oil uptake had opposite trends. The large increase in oil uptake and the sharp decrease in water absorption percentages were observed with 1.5 hours of cooling. There was no significant further reduction in water absorption observed when the cooling time was longer than 4 hours. However, the tangential swelling percentage was less affected by oil uptake, although less tangential swelling was found for lower water absorption (Figure 8.3). Unlike the water absorption percentage, no sudden drop in the tangential swelling percentage was observed after 1.5 hours of cooling. This suggests that increased oil absorption in wood owing to prolonged cooling mainly affects the water repellency of the treated wood and is less influential on the dimensional stability. This again suggests that that dimensional stability is more affected by changes in wood chemistry (degradation of hemicelluloses) as a result of high temperature treatment (Temiz *et al.* 2006; Wang and Cooper 2005). Also it should be noted that the temperature of the oil bath was falling during the cooling period, being only 55°C after 12 hours. The final temperature of the oil bath may affect not only the oil uptake but also the water repellency and dimensional stability of wood.



*Figure. 8.3. Effect of the cooling period on the oil uptake percentage (OUP) and tangential swelling percentage*

## 8.4 Conclusions

In this part of the thesis, the oil uptake by oil heat-treated wood was determined by weight percentage change and organic solvent extraction, and the results of these two methods were compared. The effect of oil uptake and thermal modification on water repellency of the oil-heat treated wood was also investigated with organic solvent extraction. In addition, the effects of cooling time on oil uptake, water repellency and dimensional stability were also examined.

The results of this study show that oil uptake percentage determined by the solvent extraction method was greater than that determined by the weight percentage change after oil heat-treatment. This confirmed that there are some losses of wood mass during the oil heat-treatment. The difference in oil uptake determined by these two methods increased with

increasing temperature which suggests more mass loss in heat-treated wood at higher temperatures. Moisture excluding efficiencies of oil heat-treated wood after solvent extraction were reduced which suggest that oil uptake by oil heat-treated wood affects its hydrophobicity.

The oil absorption by wood increased significantly with the increase in cooling time which in turn decreased the water absorption. However, comparatively, tangential swelling in treated wood was less affected by increased oil uptake as a result of the prolonged cooling time.

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# Chapter 9

## CONCLUSIONS AND RECOMMENDATIONS

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### 9.1. Conclusions

Improvement in dimensional stability and durability of plantation grown timbers is a significant issue to meet the growing demand of wood from sustainable resources. The applications of such materials and improved processing with minimum adverse impact on human health and environment have been growing around the world. To contribute towards these important aspects this study investigated moisture-related properties, durability, physical and mechanical properties of New Zealand grown radiata pine after heat-treatment using linseed oil. The overall findings of the research are summarised in the following sub-topics:

#### 9.1.1. Heat-treatment in linseed oil

Heat-treatment of radiata pine wood was carried out in an oil bath at a range of temperatures and times using linseed oil. The treated wood turned darker and was more dimensionally stable and durable, with the extent of changes depending mainly on treatment temperature. The colour changes made the two wide faces of the boards more similar and uniform without any colour patches, although the surfaces were darker

than the core. Colour homogeneity between faces and the difference between core and surface colour decreased with the increase in treatment temperature.

The wood became more hydrophobic with a significant improvement in dimensional stability. The anisotropy of wood decreased with the oil heat-treatment and this was evidenced by the fact that differences of anti-swelling efficiency (ASE) in tangential and radial directions were reduced on increasing the treatment temperature. The fungal resistance of treated wood was also improved with the best result achieved at the highest treatment temperature of 210°C. No distinct structural changes were observed in the wood cell wall. The treatment resulted in wood chemical changes mainly in the form of degradation of hemicelluloses which was measured by wet chemical analysis and FTIR analysis. The chemical changes are believed to be the main cause of changes in wood properties. The poor mechanical properties, in particular the strength loss, were the major technical draw-back of the oil heat-treatment.

### **9.1.2. Thermo-mechanical compression followed by oil heat-treatment**

The loss of mechanical properties during the oil heat-treatment was successfully countered by thermo-mechanical compression of wood in the radial direction prior to the oil heat-treatment. In the thermo-mechanical compression, high compression set (39%) was achieved without any surface checks and cracks. The specimens undergoing thermo-mechanical compression followed by oil heat-treatment showed much less swelling and low compression set recovery in high humidity conditions. The mechanical properties of the densified wood followed by oil heat-treatment were less than those of “only densified” wood but were still significantly better than those of the untreated wood. The fungal

resistance of wood after the combination treatment of thermo-compression and oil heat-treatment was also increased significantly compared to untreated wood and wood treated using thermo-compression only.

### **9.1.3. Accelerated UV weathering of oil heat-treated wood**

After the accelerated UV weathering test, the oil heat-treated wood retained its dimensional stability and colour better than the untreated wood. After the UV weathering, no surface checks were observed in oil heat-treated wood and the volumetric swelling was also less compared to the untreated wood.

### **9.1.4. Oil heat ageing**

The effects of oil aging (up to 27 hrs) on the properties of heat treated wood were investigated. Oil viscosity was increased with the age of the heated oil, resulting in a slight decrease in the weight percentage gain of the treated wood. Water repellence efficiency of the treated wood decreased when the aged oil was used for the heat treatment. The decrease was more significant with the age of the oil. However no significant differences in colour change and dimensional stability were observed between specimens treated with oils of varying heating ages. This implies that at given treatment parameters (treatment temperature, time, cooling period), the oil can be re-used for subsequent treatments (up to 27 hrs) and the main reason for improvement in dimensional stability of the heat treated wood in oil was the chemical changes in wood under high temperature conditions.

### **9.1.5. Solvent extraction**

The percentage of net oil uptake in treated wood was determined after organic solvent extraction and compared with the weight percentage

changes with the treatment. The results of the investigation showed that the oil uptake percentage determined by the solvent extraction method was greater than that determined by the weight percentage change. The difference in oil uptake determined by these two methods increased with increasing the treatment temperature which indicated that greater mass loss of woody material in the heat-treated wood occurred at the higher temperature. It was also found that moisture excluding efficiencies of oil heat-treated wood after solvent extraction were reduced which suggested that oil uptake by heat-treated wood affected its water repellency.

#### **9.1.6. Cooling period**

The effect of cooling the specimens in the oil bath after heat treatment on oil uptake and moisture related properties were examined. The oil uptake by wood increased significantly with the cooling time which, in turn, decreased the water absorption by wood. However, the influence of the cooling time on the volumetric swelling in treated wood was less significant although the oil uptake was increased with the prolonged cooling time. This implied that increased oil absorption in wood due to a longer cooling time mainly affected the water repellency of the treated wood and was less important to the dimensional stability.

## **9.2. Significance of findings**

The present study confirms some of the previous findings and contributes additional evidence that suggests that the dimensional stability and durability of radiata pine wood can be improved by oil heat-treatment without application of any persistent toxic chemicals. The treatment methods used in this study may be applied to other plantation species either using the tested linseed oil or other vegetable oils.



Oil heat-treated radiata pine wood can be suitable for those uses in which dimensional stability and durability is of prime importance and strength is not a decisive factor such as cladding, decking, garden furniture, gates, fences, doors and window components. Where strength, stability and durability are all important, the wood can be treated using a combination of thermo-compression followed by oil heat-treatment. Favourable environmental values and easy availability of linseed oil in many countries including New Zealand may open the door for future commercial ventures.

This study also found that the colour of oil heat-treated wood was darker, homogenous and more stable during accelerated UV weathering than untreated wood. An implication of this is the possibility of using oil heat-treatment to achieve dark homogenous coloured wood with a tropical flavour for specific markets.

In addition, this study provided evidence that dimensional stability and colour of treated wood is not significantly affected by the oil heating age and post-treatment cooling-period. Thus, the cost of the treatment can be reduced by recycling used oil for subsequent treatments and by reducing the oil uptake in wood by minimising the cooling period.

### **9.3. Recommendations**

Based on findings of this study, future research is recommended as follows:

- Further study might explore the use of cheaper oils to heat-treat wood such as crude tall oils (a byproduct of the paper and pulp industry), used cooking oils from major food chains and restaurants. Additives such as insecticides, fire-retardants, waxes or dyes can be mixed in heating oil and their effect on the relevant

properties can be investigated. It would be also interesting to study the behaviour of wood and the effect on wood properties by immersing the wood in cold oil immediately after the heat-treatment.

- From this study it was found that the heating age of oil (up to 27 hours) does not affect the colour change and dimensional stability of treated wood. Further work is recommended to investigate the impact of a longer heating age of oil on these wood properties and mechanical properties to determine the maximum time that the oil can be reused for the heat-treatment.
- The current study also observed that controlled pre-densification (39% compression set) of wood can counter the loss of mechanical properties of wood due to the oil heat-treatment. However, loss of saleable wood volume is a major drawback to the densification process, thus it is recommended that the mechanical and other wood properties of densified oil heat-treated wood at the lower compression level be examined.
- A study of wood properties alone is not enough to advocate oil heat-treatment in commercial practice. Commercialization of “combined operation” involving both thermo-mechanical compression and oil heat-treatment can be explored. Study of product specific integrated operation and optimization of the treatment parameters e.g. pre-compression (compression percentage, temperature), oil heat-treatment (treatment temperature and time), cooling (time) and oil uses would be beneficial for the commercial adoptability of technology. Keeping the novel technologies cost-effective is a major challenge. Further studies should be carried out on the economic analysis of the

treatments including availability and cost of raw materials, and the cost of additional preparation and processing to the existing set up. The cost-benefit analysis should also take account the intangible benefits from such treatments such as environmental and health benefits.

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