Maderas-Cienc Tecnol 24(2022):6, 1-23
Ahead of Print: Accepted Authors Version

DOI:10.4067/S0718-221X2022005XXXXXX

SIMULTANEOUS TREATMENT WITH OIL HEAT AND DENSIFICATION ON PHYSICAL PROPERTIES OF *Populus* × *canadensis* WOOD

Antonio Villasante[1*], Santiago Vignote[2], Alvaro Fernandez-Serrano[1], Rubén Laina[2]
[1] University of Lleida, Department of Agricultural and Forest Engineering, Lleida, Spain.
[2] Universidad Politécnica de Madrid, E.T.S.I. Montes, Forestales y del Medio Natural, Madrid, Spain.
*** Corresponding author:** antonio.villasante@udl.cat
Received: October 07, 2020
Accepted: October 06, 2021
Posted online: October 07, 2021

ABSTRACT

Samples of wood from *Populus* × *canadensis* (9,5 % moisture) were treated with olive oil at 195 °C simultaneously with 15 % or 30 % compression densification, and the results were compared with samples subjected to oil heat treatment without densification, and control samples. The density of the treated samples increased by 18 %, 43 % and 1,5 % respectively, and barely changed over the six subsequent months stored inside the laboratory room (at approximately 65 % RH, 20 °C). This was due to the fact that the slight weight increment caused by the additional moisture content was offset by the increase in volume from the springback effect. When subjected to atmospheres with different relative humidities, the treated samples stabilised at the same time as the control samples, although the treated samples had a significantly lower moisture absorption than the control samples. It was also observed that the hygroscopic shrinkage in oil heat densification treatment samples was approximately half those of the control samples. The initial densification was partially lost as a result of springback: approximately 3 % in the first springback at a relative humidity of 65 % RH, and an additional 4 % in the second springback to a relative humidity of 85 % RH. Once this latter relative humidity had been attained, no new losses in densification were observed. The ageing of the oil used in the treatment caused a slight loss of densification in the densest samples.

Keywords: Age of the oil, compression-set, olive oil, springback, wood density

INTRODUCTION

Wood is a renewable material that offers undoubted advantages but also has its drawbacks. Various researchers have developed techniques for modifying wood to avoid or reduce its unfavourable properties. The objective of modified woods is to improve certain properties, for example, by increasing its dimensional stability, resistance to bases, acids and ultraviolet radiation and decreasing biodeterioration and hygroscopicity, or improving its mechanical performance (Forest Products Laboratory 2010).

One of the modification techniques is heat treatment, which consists of maintaining the wood at high temperatures for some hours (Kamke 2006, Kutnar and Sernek 2007, Forest Products Laboratory 2010). In Finnish ThermoWood and PLATO wood the treatment occurs by heating the air around the wood, while in rectification wood the process is carried out in an atmosphere with a high nitrogen content and less than 2 % oxygen (Rapp 2001). In oil heat treatment (OHT), the oxygen is separated from the wood by immersion in hot oil (Dubey *et al.* 2012b). The OHT samples presented a significant reduction in equilibrium moisture content and water absorption compared to the untreated samples (Bak and Nemeth 2012, Lee *et al.* 2018). Bak and Nemeth (2012) reported that water absorption is not blocked in samples treated with OHT, but merely reduced as it decreases the amount of places where the water can connect. This reduction is due to the lower number of hydroxyl groups in which the water can bind and to the increased crosslinking in the lignin (Lee *et al.* 2018). Equilibrium moisture is achieved at the same time in the OHT and the untreated samples (Bak and Nemeth 2012).

The shrinkage in the wood also improve with the OHT content. Dubey *et al.* (2012a) observed that this treatment causes a reduction in the volumetric swelling percentage and increases anti-swelling efficiency (ASE). The improvement in the dimensional stability of the wood is greater when the oil temperature and treatment time increases (Bak and Nemeth 2012). Rapp (2001) also observed a higher ASE in the samples treated at higher temperatures. Bak and Nemeth (2012) reported that in OHT wood treated with three vegetable oils the increase in ASE was greater in the tangential direction then in the radial direction. Although the anisotropy of the wood decreased, it is not completely removed. Okon *et* *al.* (2018) also observed greater reductions in shrinkage in the tangential than in the radial direction in wood treated with silicone oil as a heating medium.

One factor that can affect the OHT treatment is the type of oil used. The most commonly used oils are of vegetable origin. Some studies reported differences in the results based on the type of oil. Wang and Cooper (2005) found that palm oil was more effective than soybean oil for achieving dimensional stability. Lyon *et al.* (2007) considered that the most important characteristic of the oil is its unsaturation degree. Oils with a higher proportion of polyunsaturated fatty acids are more recommended for OHT treatments. Tomak *et al.* (2011) reached the same conclusion and used iodine values in their work to establish the unsaturation degree. However, studies by other authors observed no significant differences in equilibrium moisture or moisture absorption between OHT samples treated with sunflower oil, linseed oil and rapeseed oil (Bak and Nemeth 2012). Dubey *et al.* (2011) observed less moisture absorption in OHT samples treated with fresh oil than in samples treated with pre-heated oil, although there were no significant differences in volumetric swelling. In that work, the authors justified these differences as being due to the evaporation of volatile compounds and the heat polymerization of the oil.

Another common type of modification of the wood is densification, developed since the early 20th century (Kollmann *et al.* 1975). Densification consists of increasing density by applying pressure to the previously heated wood in such a way that the lumens partially collapse (Song *et al.* 2018). Wood is usually heated by means of liquid water or steam. Some authors have used other fluids, as in the case of Song *et al.* (2018), who successfully applied densification in a solution of NaOH and Na_2SO_3 in their work. Densification is a technique that is particularly suited to low-density woods (Kamke 2006), because it enables them to be used for purposes where high density woods would normally be chosen (Kutnar and Sernek 2007). Kawai *et al.* (1992) indicated that during densification the cellulose crystals become reoriented, the hemicelluloses are partially hydrolysed and the lignin is partially degraded. The main application of densification is to increase the mechanical properties of the wood (Welzbacher *et al*. 2008; Gašparík *et al*. 2016; Laskowska 2020). Kutnar *et al.* (2008) obtained increases in MOE and

MOR of between 35 % and 100 %, depending on the degree of densification. Sotomayor (2016) reported twice the MOE in densified wood than in the control samples. Kamke (2006) obtained MOE values in densified wood that were three times greater than in the control samples. It is important to highlight that the effect of densification may be reversible when the moisture content in the wood increases (Kamke 2006; Welzbacher *et al.* 2008), a phenomenon known as springback, or recovery from compression. This occurs when the internal stresses that appear in the densification are relaxed, and part of the wood's internal structure seeks to recover its original form (Morsing 1998). Springback is reduced through several mechanisms: by making the cell wall inaccessible to water, forming crosslinks between the wood components in the deformed state, and releasing stresses in the microfibrils during compression (Morsing 1998). Welzbacher *et al.* (2008) found that springback depends more on the temperature of the treatment then on the duration of the compression, and reported a particularly significant effect at temperatures over 180 °C.

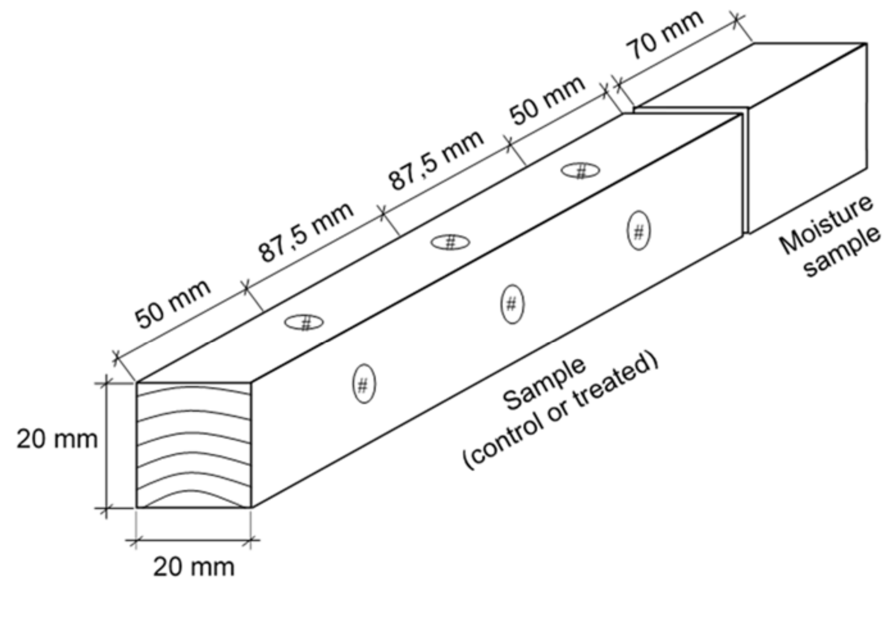
To combine favourable effects, some researchers have analysed the behaviour of wood subjected to densification and subsequently to OHT. Fang *et al.* (2011) observed that in wood treated in this way the equilibrium moisture and the springback were reduced, and this reduction was greater in the samples treated with OHT at higher temperatures. Hsu *et al.* (1988) and Laborie (2006) indicated that in this combined treatment there was a joint occurrence of a reversible swelling due to the hygroscopicity of the wood and another irreversible swelling due to the compression-set recovery. Fang *et al.* (2011) quantified the incidence of the irreversible swelling by performing saturation tests on the samples, and observed that it decreased as the time and the treatment temperature increased.

In spite of the advantages obtained by performing the densification of the wood in combination with the OHT treatment, no works were found in which these two processes are carried out simultaneously to benefit of the heating caused by the oil treatment for wood densification. The aim of the present work was to establish the behaviour in regard to moisture content in low density wood of *Populus* × *canadensis* when the densification and OHT are carried out simultaneously.

MATERIAL AND METHODS

Samples preparation

Ten logs were used from the I-214 clone of *Populus* × *canadensis* Moench, measuring 0,35 m long and with a diameter of 0,2 m to 0,3 m. They were obtained from a wooden fruit packaging company that uses trees from the Ebro Valley (Spain). Each log was identified with a letter (from A to J) to control the log factor. The largest possible quantity of strips with cross-sectional dimensions of 22 mm x 22 mm and 350 mm long was extracted from each log, with the rings parallel to the transversal edges. The strips were stored inside the laboratory until the samples attained a moisture content of near 12 %. They were subsequently processed with a thickness planer until they had a final section of 20 mm x 20 mm, when they were stored for another 15 days. Immediately before treatment, each 350 mm strip was cut transversally to obtain a sample of 275 mm in length, and a moisture sample of 70 mm which was only used to estimate the moisture content of the probe (Figure 1).



= Measurement point of the transversal measures

Figure 1: Sample preparation.

OHDT and OHT treatments

Four samples were randomly selected from each of the ten logs, one for each treatment type. The following treatments were performed:

$OHDT_h$: OHT treatment simultaneously with high densification, with an approximate 6 mm reduction in thickness.

$OHDT_l$: OHT treatment simultaneously with low densification, with an approximate 3 mm reduction in thickness.

OHT: treatment in hot oil (without densification).

Control: samples without densification and without OHT.

The main samples of 275 mm were measured lengthwise with a 1 mm precision measuring tape and crosswise with a 0,01 mm precision micrometer. Both the radial and tangential crosswise measures of the samples were taken in three measurement points: two at 50 mm from the ends and one in the centre (Figure 1). Each measurement point was marked by circling the zone of contact of the micrometer with a pencil to be able to repeat the measures at exactly the same point. The sample was weighed on a 0,01 g precision scale.

The oil used in the treatment was virgin olive oil from Aceites Toledo S.A. (Los Yébenes, Spain). Fullana *et al.* (2004) indicated a smoke point of 210 °C for this type of oil, whereas Li *et al.* (2016) indicated a range between 205 °C and 215 °C. It was decided to use a temperature of 195 °C ± 4 °C in order not to reach the smoke point, which could lead to the premature degradation of the oil. The treatment was performed in a Nevir NVR-6522F fryer, and the temperature was checked with a Digiflex TP101 digital thermometer.

The densification of each sample was carried out individually, with a press formed by two steel plates with a thickness of 8 mm and 283 mm in length, joined with eight M10 cap screws (four each side of the probe). The time required for the interior of the sample to attain a temperature of 190 °C was calculated using MacLean's formula, based on Fourier' differential equation (Kollmann 1959). For the section of the samples of 20 mm x 20 mm, the estimated time was two minutes. To maintain a minimum temperature of 190 °C at all points of the sample for 60 minutes, a preheating phase was implemented, submerging the sample in oil at 195 °C for 62 minutes. In this phase the sample was loosely attached to the outside of the steel press with a steel wire with a section of 0,3 mm to prevent it from floating, and ensure that the oil was in contact with the entire surface of the wood. When the sample was introduced in the oil, an intense bubbling was observed caused by the expulsion of air and moisture from the interior of the wood. At the end of the preheating phase, the initial bubbling ceased.

After the preheating phase, the compression was performed by placing the sample between the two plates of the press, centring it between the two rows of cap screws. The plates were closed without tightening the cap screws, simply by adjusting the discs to the sample, and it was once again submerged in the oil. The press was removed from the hot oil every 15 minutes and each cap screw was tightened a quarter turn, four times (to avoid imbalances in the plates). The quarter-turn tightening sequence is similar to the one used in cylinder heads in combustion engines. In each extraction of the hot oil, the samples were tightened approximately 1,5 mm and it took one minute to become re-immersed in the hot oil. The compression was done in a radial direction to achieve plastic yielding and the gradual collapse of the wood cells (Reiterer and Stanzl-Tschegg 2001), locating the tangential faces of the sample in contact with the plates of the press.

In the $OHDT_l$ treatment the plates were tightened twice, so a final densified thickness of approximately 17 mm was estimated (a decrease of 3 mm due to two tightenings of 1.5 mm each). The treatment time of the $OHDT_l$ samples was 92 minutes (62 minutes of preheating and two of heating before tightening periods of 15 minutes each).

In the $OHDT_h$ treatment the plates were tightened four times, so a final densified thickness of approximately 14 mm was estimated (decrease of 6 mm due to four tightenings of 1.5 mm each). The treatment time of the $OHDT_h$ samples was 122 minutes (62 minutes of preheating and four of heating before tightening periods of 15 minutes each).

Conditioning of samples

After the treatment, the samples were left to cool outside the oil and in the press for 20 hours. Once this time had elapsed, the samples were removed from the press, the radial dimensions were measured in the three measurement points, and they were weighed again. The treated samples and the control samples were stored inside the laboratory. The OHT and OHDT samples were treated simultaneously over three months. Once the treatment was finalised, all the samples were preconditioned for 15 days in a conditioning chamber at 65 % RH and 20 °C and subsequently stored for five months, grouping the ten samples of each treatment type in a plastic bag. The response to moisture content was analysed in five stable environmental conditions (SEC), alternating relative humidities 65 %, 85 %, 65 %, 85 %, and 65 % (SEC_1 to SEC_5, respectively), all at 20 °C. At the start and end of each SEC the samples were weighed and measured in each measurement point. In the first four days of each SEC the samples were weighed every day, and on subsequent days the samples were weighed every four days. The moisture content of the wood was considered to be stable when the difference in weight taken four days apart was less than 0,2 %.

As the samples were not oven-dried in any part of the process, the moisture content of the samples was estimated from the 70 mm long moisture sample (Figure 1).. The moisture content of the moisture samples was obtained from the difference in weights before and after oven drying at 103 °C, following EN 13183-1:2002 (EN 2002). Based on this moisture, the oven dry mass (m_0) of the treated and control samples was estimated using Equation 1,

$$m_0 = \frac{m_{at}}{1+\frac{\omega_{MS}}{100}} \times 100 \qquad (1)$$

where m_{at} is the mass of the sample before starting treatment and ω_{MS} (in %) is the moisture content obtained from the 70 mm long moisture sample (Figure 1).

From the mass of each sample (m_i) in a SEC and from the estimation of its m_0, the percentage of total sorption of oil and water (TS_{ow}, in %) can be obtained by means of Equation 2. In the case of control samples, TS_{ow} corresponds exclusively to water and is therefore equivalent to moisture content. In the samples treated with oil ($OHDT_h$, $OHDT_l$, and OHT), TS_{ow} corresponds to a combination of water and oil.

$$TS_{ow} = \frac{m_i - m_0}{m_0} \times 100 \qquad (2)$$

Similarly, the partial sorption of water (PS_w, in %) was calculated by means of Equation 3. This indicates the change in the mass of water that occurs between the start of a SEC_i (m_i) and the end (m_{i+1}).

$$PS_w = \frac{m_{i+1} - m_i}{m_i} \times 100 \quad (3)$$

To determine the recovery of form in the compressed samples, Fang *et al.* (2011) used the compression-set recovery variable. This was obtained by measuring the samples in oven-dry moisture content. In the present study, the compression-set recovery was not calculated because the aim was to simulate the behaviour of wood in natural conditions, and artificial oven-drying could produce irreversible alterations. This was also the reason that the anti-swelling efficiency was not used (Forest Products Laboratory 2010). The intensity of the densification of the samples was estimated by means of compression set (Welzbacher *et al.*; 2008; Wehsener *et al.* 2018) according to Equation 4,

$$CS = \frac{R_{bt} - R_{SEC_i}}{R_{bt}} \cdot 100 \quad (4)$$

where CS is compression set (%), and R_{bt} and R_{SECi} are the mean of the radial dimensions in the three measurement points before treatment and after the SEC_i stage respectively.

Statistical analysis

The differences between the groups were analysed by ANOVA, followed by Tukey's post hoc test. The densities of the samples before treatment, after treatment and before SEC_1 were compared by means of the paired samples T-Test. Linear regression was used to establish the influence of the age of the oil on the shrinkage. The statistical analysis was performed with the R program, version 3.6.1 (R Core Team 2019) with a significance level of 0,05.

RESULTS AND DISCUSSION

The treated samples darkened as a result of the treatment, and this change was maintained throughout the entire SEC. This effect coincides with the results of Dubey *et al.* (2011) in samples of *Pinus radiata* treated with linseed oil. Lee *et al.* (2018) justified the change in colour as being due to the formation of an oil layer on the wood surface and the caramelisation of soluble sugars produced from hydrolysed hemicellulose during heat treatment.

The densities of the samples according to the type of treatment are shown in Table 1. The mean density before treatment was 0,456 g/cm^3 (with 9,6 % moisture content), coinciding with the interval 0,420 to 0,480 g/cm^3, included in AITIM (1997). Other authors obtained lower densities in their studies (Istok *et al.* 2016). This difference can be explained by the intrinsic variety in the properties of the wood, and is increased by the diversity of the development techniques that are applied to this species (plantation framework, pruning, watering, fertilisation, etc.).

Table 1: Densities (in g/cm^3) before treatment, after treatment and before conditioning. In brackets Coefficient of variation (in %). The lower part in each case shows the increase in density between stages.

Density	$OHDT_h$	$OHDT_l$	OHT	control
Moisture content before treatment (%)	9,6 ^{a} (3,63)	9,6 ^{a} (4,51)	9,5 ^{a} (6,23)	9,5 ^{a} (2,35)
Density before treatment	0,456 ^{a} (9,82)	0,450 ^{a} (8,70)	0,455 ^{a} (6,90)	0,464 ^{a} (8,49)
Density after treatment	0,651 ^{c} (7,87)	0,532 ^{b} (7,65)	0,462 ^{a} (6,80)	0,463 ^{a} (8,51)
Increase (g/cm^3)	+ 0,195***	+ 0,082***	+ 0,007***	- 0,001***
Density before Stable Environmental Condition 1 (SEC_1)	0,647 ^{c} (7,54)	0,538 ^{b} (7,52)	0,472 ^{a} (6,88)	0,463 ^{a} (8,43)
Increase (g/cm^3)	- 0,004**	+ 0,006***	+ 0,010***	+ 0,0005*

$^{a\ b\ c}$ Values with a different superscript letter in the same row present significant differences between treatments. Comparison of density increases between stages. * = p-value between 0,01 and 0,05; ** = p-value between 0,001 and 0,01; *** = p-value less than 0,001; ns = non-significant.

In the control samples, the density changed less than 0,001 g/cm^3 during the six months before treatment and before SEC_1. The $OHDT_h$ treatment caused a significant increase in density of 43 %, and an increase of 18 % in $OHDT_l$ as a result of densification. A slight increase in density (1,5 %) was observed during the OHT treatment (without densification), representing less than 0,001 g/cm^3. In the six months after the treatment, the treated probes underwent slight changes in density. In these months the moisture content only increased from 9,5 % to 12 %. There were two overlapping effects in this period, which modified the density of the OHDT samples in opposite ways: on the one hand there was a slight increase in moisture content, which led to an increase in density; and on the other there was a springback effect (Kamke 2006; Kutnar and Sernek 2007) which increased the volume of the wood and therefore decreased its density. The springback was more significant in the $OHDT_h$ samples, which were more densified. A slight decrease in density was observed in these samples, as indicated by the negative increase detected in the density. In contrast, in the less densified $OHDT_l$ samples the effect of the moisture content was slightly greater than the springback, so the density increased slightly.

The changes in the amount of water and oil absorbed are shown in Figure 2. The stabilisation of the moisture content in each SEC of the treated samples and the control samples occurred at the same time, so the decrease in the moisture uptake rate in treated samples can be attributed to the lower water storage capacity (Bak and Nemeth 2012). The TS_{ow} values corresponding to the control samples showed the usual hygroscopic behaviour for wood. In the control samples it was observed that in the different SECs of 65 % RH, a moisture content of between 11 % and 13 % was found; lower values were obtained when this was achieved from lower relative humidity (SEC_1) and higher values from higher relative humidities (SEC_3 and SEC_5). This difference in values is explained by sorption hysteresis (Forest Products Laboratory 2010). The treated samples followed parallel behaviours of TS_{ow}, ordered according to the intensity of the densification. All the treated samples had the same TS_{ow} before treatment, and separation occurred during treatment. From this point on the distances remained approximately constant. It is worth noting that the values of TS_{ow} were higher in the treated samples than in the control samples. This appears to contradict the results of other authors (Jalaludin *et al.* 2010) who indicated that OHT wood exhibited a marked reduction in equilibrium moisture content. This discrepancy is explained because TS_{ow} includes both water and the oil absorbed in the treatment, so PS_w must be used when comparing only water contents. Another aspect that can be seen in Figure 2 is that the changes in TS_{ow} between the 65 % SEC and 85 % SEC were always lower in the treated samples than in the control samples, both in adsorption and in desorption. This indicates a more mitigated exchange of moisture content in the treated than in the control samples. This result coincides with Bak and Nemeth (2012), who indicated that moisture uptake is not blocked in OHT samples but merely decreases. The explanation may be due to crosslinking caused by the polycondensation reactions in lignin and to diminishing amounts of water-affinity hydroxyl groups owing to the heat during the treatment and the crystallisation of the cellulose (Lee *et al.* 2018).

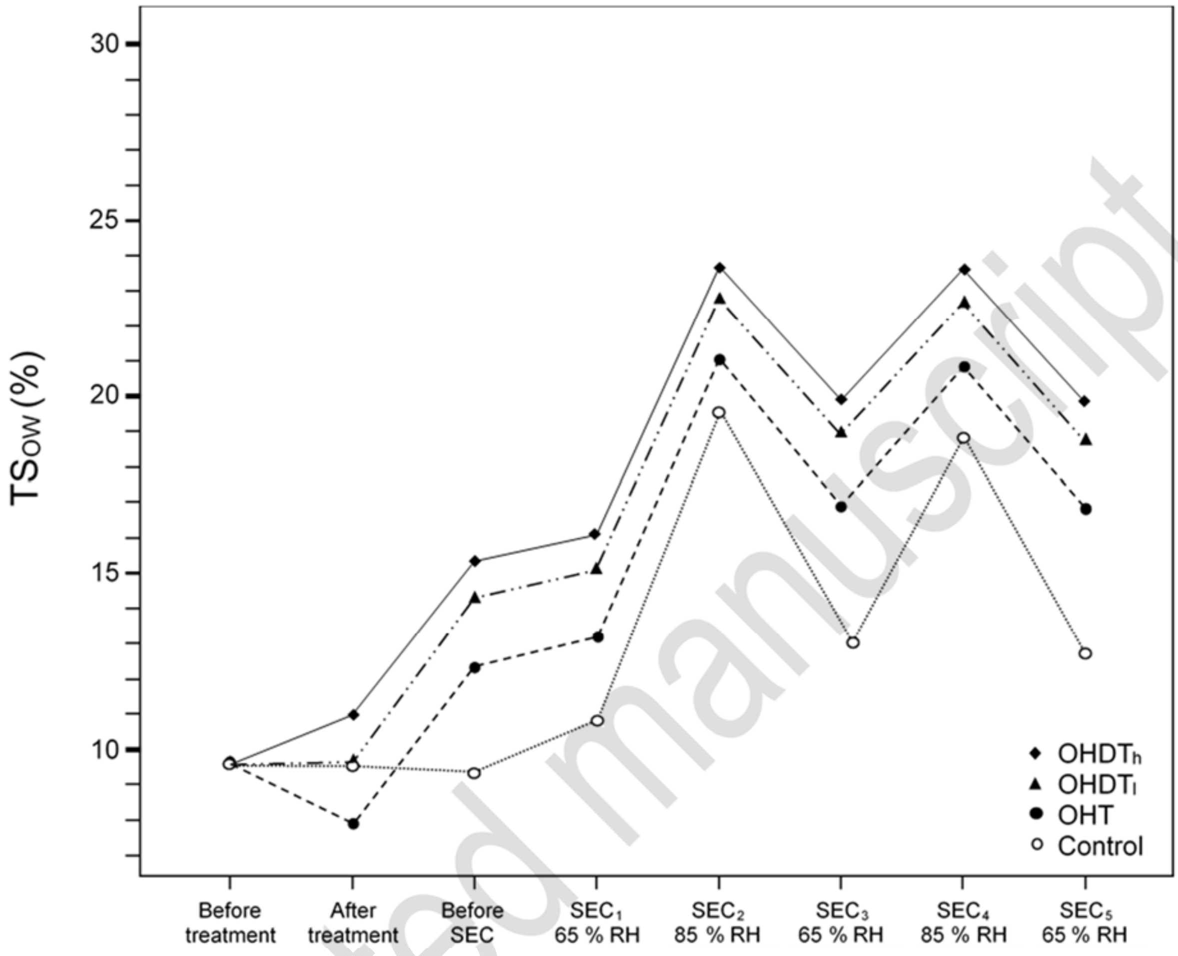


Figure 2: TSow in each stable environmental condition (SEC), grouped by treatment type.

The changes in PS_w are shown in Table 2. At the end of SECs 3 to 5, the PS_w values in the treated samples (approximately 3 %) were significantly lower than in the control samples (approximately 5 %). This confirms the mitigation of the moisture exchanges that were detected in the TS_{ow}. Other authors have also found lower equilibrium moistures in samples treated with hot oil (Jalaludin *et al.* 2010; Bak and Nemeth 2012; Lee *et al.* 2018). The exception was the stage that elapses after treatment and before SEC_1, in which the control samples practically maintained their PS_w constant whereas the treated samples had a significantly greater increase of approximately 4 %. This initial increase that occurred over six months of stable relative humidity conditions reflected a partial recovery of the hygroscopic character of the treated samples. In this initial phase no significant differences were detected in the increases in PS_w between the OHDT and OHT samples.

Table 2: Values of PS_w (in %) in stable environmental conditions (SEC_i). The coefficient of variation is shown between brackets (in %).

	$OHDT_h$	$OHDT_l$	OHT	control
Before SEC_1	3,98 ^{b} (6,60)	4,21 ^{b} (7,97)	4,10 ^{b} (10,28)	- 0,19 ^{a} (41,6)
After SEC_1 65 % RH	0,66 ^{c} (6,44)	0,69 ^{c} (7,28)	0,75 ^{b} (2,55)	1,34 ^{a} (4,05)
After SEC_2 85 % RH	6,57 ^{c} (4,76)	6,69 ^{bc} (3,79)	6,95 ^{b} (3,28)	7,84 ^{a} (4,29)
After SEC_3 65 % RH	- 3,05 ^{c} (5,72)	- 3,19 ^{bc} (6,20)	- 3,41 ^{b} (4,29)	- 5,42 ^{a} (5,31)
After SEC_4 85 % RH	3,10 ^{b} (5,13)	3,18 ^{b} (6,40)	3,37 ^{b} (3,74)	5,09 ^{a} (5,52)
After SEC_5 65 % RH	- 3,05 ^{c} (4,92)	- 3,14 ^{bc} (5,94)	- 3,31 ^{b} (2,89)	- 5,14 ^{a} (5,53)

$^{a\ b\ c}$ Values with a different superscript letter in the same row present significant differences between treatments.

A marked difference in PS_w can be observed between SEC_2 and SEC_4 (with the same 85 % RH in both cases). This is because SEC_2 comes from an environment of 65 % RH obtained by adsorption, and SEC4 comes from an environment of 65 % RH obtained by desorption. This difference is explained by sorption hysteresis (Forest Products Laboratory 2010).Within the SECs, the OHT samples presented significantly higher PS_w then the $OHDT_h$ samples, with some exceptions. The $OHDT_l$ samples with intermediate densification also had intermediate values. The moisture absorption was significantly lower in the samples that had been more densified.

The CS values are shown in Figure 3 and 4. It can be seen that the OHT treatment (hot oil without densification) produced a 1 % shrinkage in thickness (Figure 3) due to the replacement of water by oil.. The difference in thicknesses with regard to the control samples decreased to 0,4 % in the period prior to the SECs and remained more or less constant in all the SECs. The results showed some CS in the OHT samples which reached -0,7 % (equivalent to an increase in swelling of 0,7 %). The CS of the control samples were greater and reached -1,2 %, a less favourable value as it is farther from 0 %. In all SECs the control samples presented a CS of approximately double the one obtained in OHT samples.

The decreases in shrinkage in the samples treated with hot oil coincided with the results of Dubey *et al.* (2012a) who detected improvements in the volumetric swelling percentage in samples of *Pinus radiata* treated with linseed oil. Several authors have also reported improvements in anti-swelling efficiency in samples treated with OHT (Rapp 2001; Dubey *et al.* 2012a; Bak and Nemeth 2012). Fang *et al.* (2011) reported the extreme case in which the compression recovery of aspen wood veneers was eliminated with OHT treatment. This could be explained by the thinness of this type of samples.

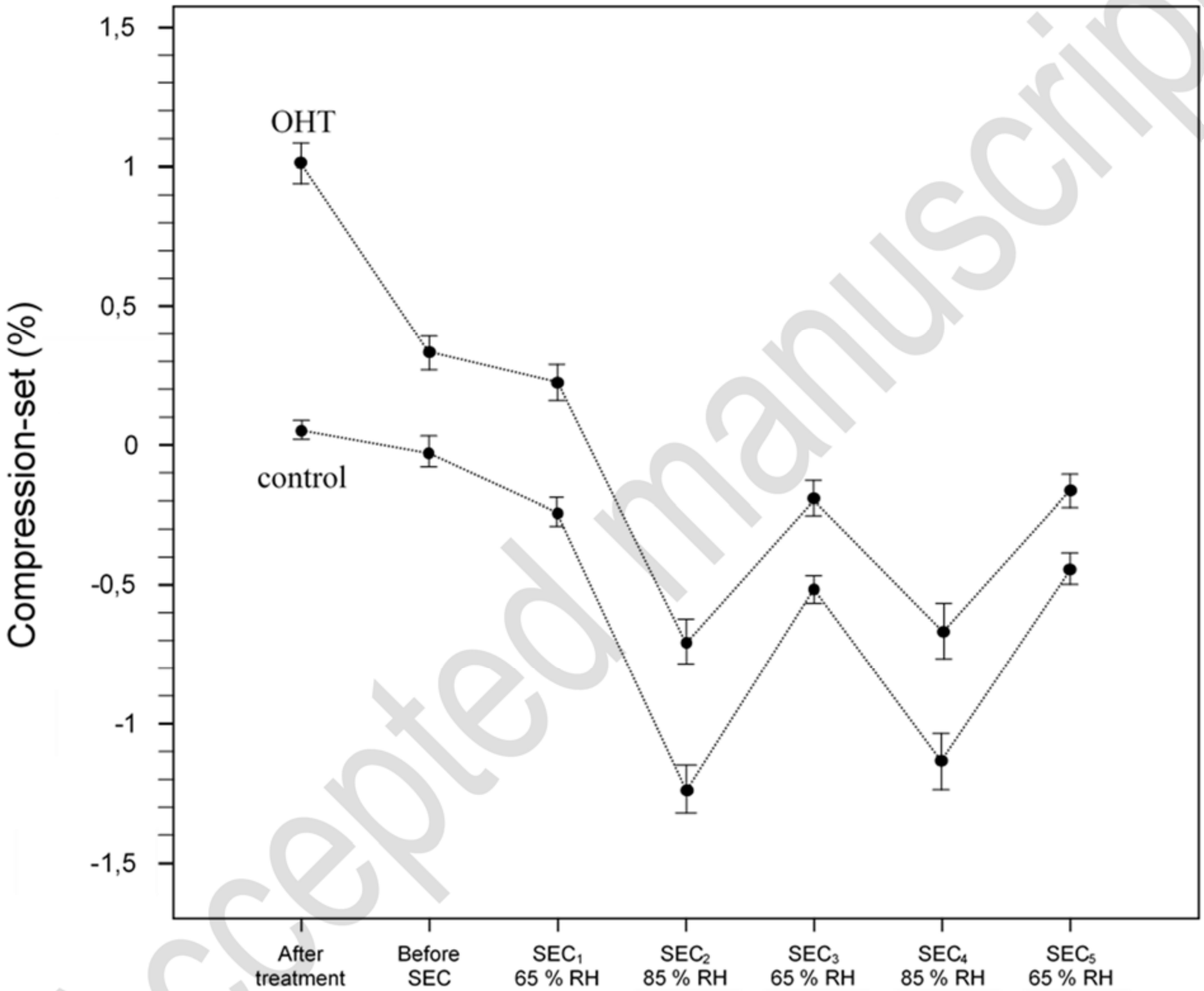


Figure 3: Compression-set of the OHT samples and control in each stage in each stable environmental condition (SEC).

The CS values in the densified samples (Figure 4) were higher than those obtained in the OHT and control samples due to densification. The initial CS obtained with densification, 30 % in the $OHDT_h$ samples and 15 % in the $OHDT_l$ samples, decreased throughout the rest of the measurements. The main cause of this reduction is springback, which occurred in the initial phase at a relative humidity of 65 % (first springback) and in the phase of high relative humidity at 85 % (second springback).

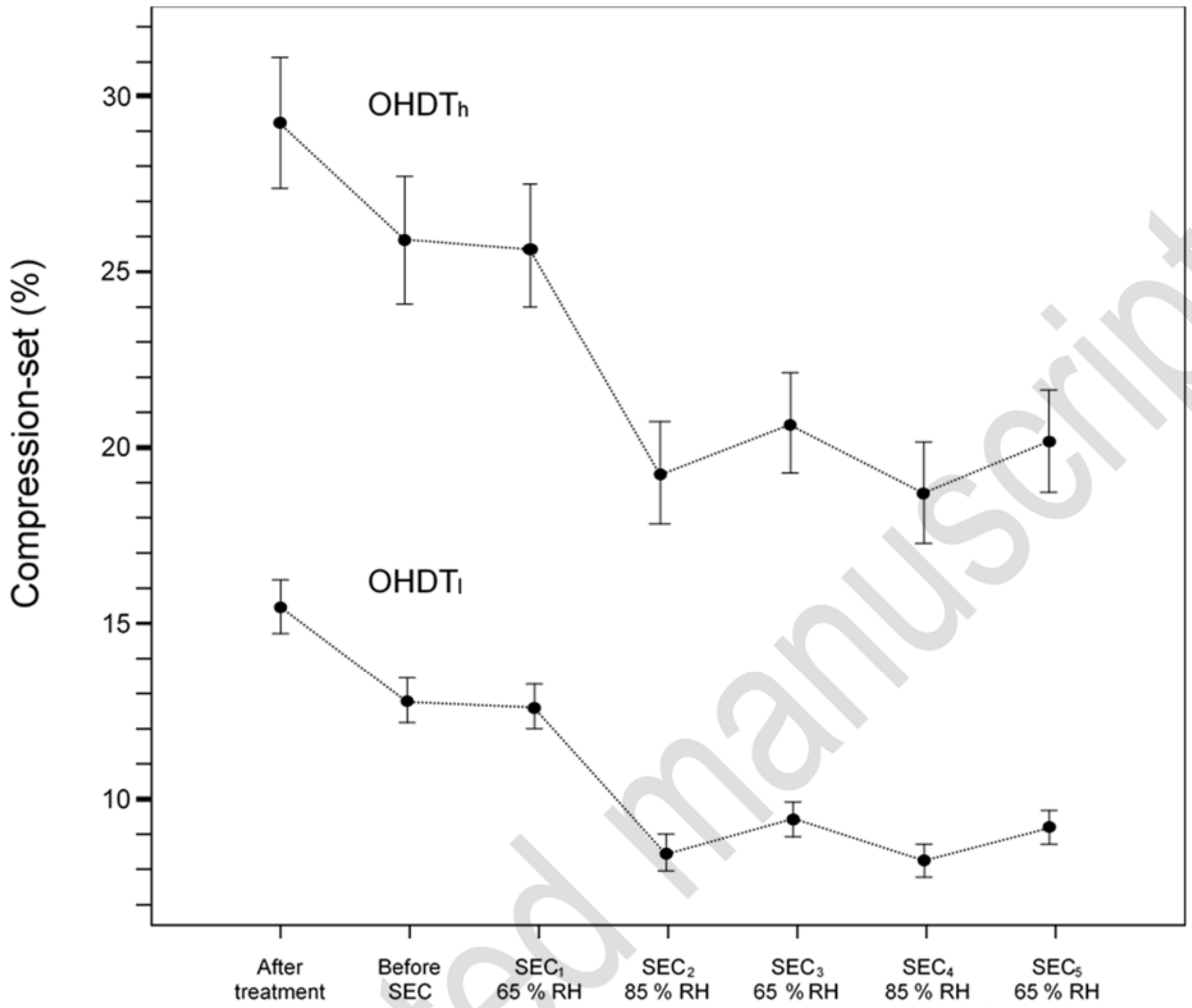


Figure 4: Compression-set of the OHDT samples in each stable environmental condition (SEC).

The first springback can be seen in the step between after treatment and SEC_1 which led to a 3,3 % loss of compression in the $OHDT_h$ samples and 2,6 % in the $OHDT_l$ samples. These values were similar to the 4,6 % and 1,7 % found by Dubey *et al.* (2016) in densified samples of *Pinus radiata* subsequently treated with oil at 180 °C and 210 °C. The second springback presented higher values of 5,0 % in $OHDT_h$ samples and 3,1 % in $OHDT_l$ samples. These correspond to the step between SEC_1 and SEC_3: the two first SECs with 65 % relative humidity, separated by a SEC with 85 %, although part of this decrease was due to the fact that SEC_1 was caused by adsortion and SEC_3 was due to desorption, which could explain only a small part of the change. Kamke (2006) also found that the swelling of the densified wood occurred with greater intensity when the samples were submitted to high moisture content (second springback).

The presence of springback in the results of the present study coincides with the works of Navi and Girardet (2005) who observed that part of the densification is lost if the wood is re-wetted. As in the control and OHT samples, from the equilibrium in SEC_2 at 85 % RH, the changes in CS between the remaining SEC remained constant, indicating that springback had already totally occurred. The rate of swelling that occurred between environments of 65 % RH and 85 % RH differed depending on the treatment. The control samples had an oscillation in CS of 0,7 %, which was reduced to 0,5 % in the OHT samples. In the case of the densified samples, the oscillations rose to 1,6 %, and 1 % for $OHDT_h$ and $OHDT_l$ respectively. Spear and Walker (2006) also observed an increase in shinkrage in the densified samples, which they attributed to the increased ratio of cell wall mass to lumen volume. To verify this hypothesis, the CS values were compared in each SEC for the four treatments. No significant differences were detected between the CS of the control samples and the OHT samples in any of the five SECs. In contrast, the CS values in the densified samples were significantly greater than the control samples in the $OHDT_l$ samples, and even greater in the $OHDT_h$ samples.

Table 3: Regression slope between the age of the oil (in minutes from previous treatments) and CS (in %).

	$OHDT_h$	$OHDT_l$	OHT
Before SEC	$- 2,34 \times 10^{-3}$ *	$- 1,2 \times 10^{-4}$ ns	$- 3,1 \times 10^{-5}$ ns
SEC_1 65 % RH	$- 2,35 \times 10^{-3}$ *	$- 1,1 \times 10^{-4}$ ns	$- 2,8 \times 10^{-5}$ ns
SEC_2 85 % RH	$- 2,02 \times 10^{-3}$ **	$- 3,0 \times 10^{-4}$ ns	$- 6,8 \times 10^{-5}$ ns
SEC_3 65 % RH	$- 2,01 \times 10^{-3}$ **	$- 3,0 \times 10^{-4}$ ns	$- 2,7 \times 10^{-5}$ ns
SEC_4 85 % RH	$- 2,01 \times 10^{-3}$ **	$- 3,0 \times 10^{-4}$ ns	$- 2,0 \times 10^{-5}$ ns
SEC_5 65 % RH	$- 2,10 \times 10^{-3}$ **	$- 2,6 \times 10^{-4}$ ns	$- 4,1 \times 10^{-5}$ ns
* = p-value between 0,01 and 0,05; ** = p-value between 0,001 and 0,01; ns = non-significant.			

The influence of the age of the oil on CS can be seen in Table 3. Significant effects were only detected in the $OHDT_h$ samples. The negative value of the slope indicates that as the oil ageds, the capacity to maintain the densification decreaseds. This decrease remaineds more or less constant in all the SECs of the $OHDT_h$ samples, with a 0,12 % decrease in CS for each hour of prior use of the oil. In the $OHDT_l$ and OHT samples, the decrease was merely a trend, as all the slopes were negative even though the values were not significant. Significance was only found in the most densified samples, where the CS values are higher. However, in woods without densification or with slight densification, the influence of other effects may have masked the significance of the age of the oil. Figure 5 shows an example in SEC_5. Dubey *et al.* (2011) detected significant differences in water absorption between woods of *Pinus radiata* treated with fresh linseed oil and pre-heated linseed oil. However, coinciding with the results of the present work, they found no significant differences in OHT samples when they compared volumetric swelling.

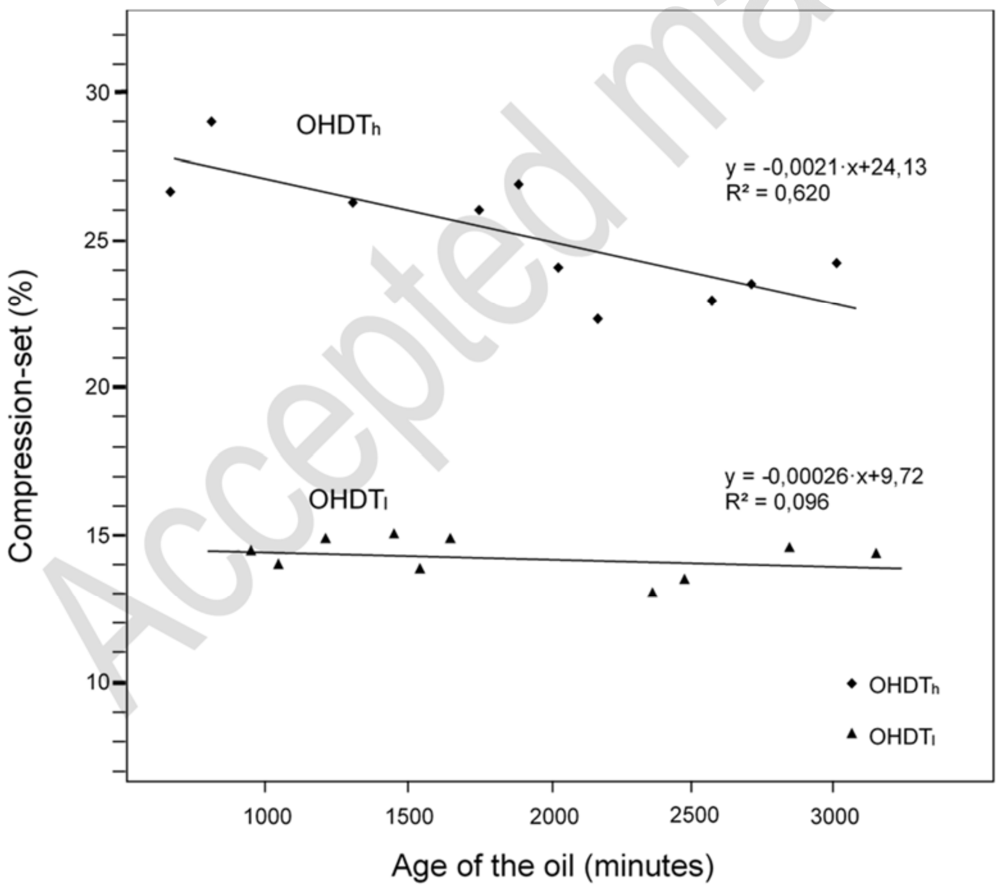


Figure 5: Relation between the age of the oil and compression-set in stable environmental condition 5 (SEC5) for the OHDTh and OHDTl samples.

CONCLUSIONS

Once the samples had stabilised at 85 % RH, the OHDT samples presented significantly less water absorption than the untreated samples. This absorption was lower in the samples that were subjected to greater densification.

The springback in OHDT samples occurred in two ways: a first springback to constant relative humidity and a second springback when they were subjected to greater relative humidities. Once the balance had been attained at a higher relative humidity, the springback remained constant for equal or lower humidities.

In the more densified OHDT samples, the age of the oil used in the treatment had a significant influence on the loss of densification due to moisture.

The OHDT treatment made it possible to obtain the advantages produced by the densification of the wood after an OHT process, but with a single treatment. This produces an energy savings as heat only needs be to be supplied in one of the phases. The treatment time is also reduced compared to when the sequential densification process is used for the OHT treatment.

REFERENCES

AITIM. 1997. *Especies de maderas para carpintería, construcción y mobiliario*. Asociación de Investigación Técnica de Industrias de la Madera y Corcho (AITIM), Madrid, Spain.

Bak, M.; Nemeth, R. 2012. Modification of wood by oil heat treatment. In Proceedings of the International Scientific Conference on Sustainable Development & Ecological Footprint. Sopron, Hungary.

Dubey, M.K.; Pang, S.; Walker, J. 2011. Effect of oil heating age on colour and dimensional stability of heat treated *Pinus radiata*. *Eur J Wood Prod* 69: 255–262. https://doi.org/10.1007/s00107-010-0431-0

Dubey, M.K.; Pang, S.; Walker, J. 2012a. Changes in chemistry, color, dimensional stability and fungal resistance of *Pinus radiata* D. Don wood with oil heat-treatment. *Holzforschung* 66(1): 49–57. https://doi.org/10.1515/HF.2011.117

Dubey, M.K., Pang, S.; Walker, J. 2012b. Oil uptake by wood during heat-treatment and post-treatment cooling , and effects on wood dimensional stability. *Eur J Wood Prod* 70: 183–190. https://doi.org/10.1007/s00107-011-0535-1

Dubey, M. K.; Pang, S.; Chauhan, S.; Walker, J. 2016. Dimensional stability, fungal resistance and mechanical properties of radiata pine after combined thermo-mechanical compression and oil heat-treatment. *Holzforschung* 70(8): 793–800. https://doi.org/10.1515/hf-2015-0174

European Committee for Standardization. 2002. EN 13183-1: Moisture content of a piece of sawn timber. Part 1: Determination by oven dry method. CEN. Brussels, Belgium. https://standards.cen.eu/dyn/www/f?p=204:110:0::::FSP_PROJECT,FSP_ORG_ID:7839,6156&cs=100F48F7330BA2CCD26D59BF5D87DFAD5

Fang, C.H.; Cloutier, A.; Blanchet, P.; Koubaa, A.; Mariotti, N. 2011. Densification of wood veneers combined with oil- heat treatment. part I: dimensional stability. *BioResources* 6(1): 373–385. https://ojs.cnr.ncsu.edu/index.php/BioRes/article/view/BioRes_06_1_0373_Fang_CBKM_Densification_Wood_Veneers_Oil_Heat

Forest Products Laboratory. 2010. *Wood handbook—Wood as an engineering material*. Forest Products Laboratory, Department of Agriculture, Madison, WI, USA. https://www.fpl.fs.fed.us/documnts/fplgtr/fpl_gtr190.pdf

Fullana, A.; Carbonell-Barrachina, A.A.; Sidhu, S. 2004. Comparison of volatile aldehydes present in the cooking fumes of extra virgin olive, olive, and canola oils. *J Agric Food Chem* 52(16): 5207–5214. https://doi.org/10.1021/jf035241f

Gašparík, M.; Gaff, M.; Šafaříková, L.; Vallejo, C.R.; Svoboda, T. 2016. Impact bending strength and Brinell hardness of densified hardwoods. *BioResources* 11(4): 8638–8652.

https://doi.org/10.15376/biores.11.4.8638-8652

Hsu, W.E.; Schwald, W.; Schwald, J. 1988. Chemical and physical changes required for producing dimensionally stable wood-based composites. *Wood Sci Technol* 22: 281–289. https://doi.org/10.1007/BF00386023

Istok, I.; Sedlar, T.; Sefc, B.; Sinkovic, T.; Perkovic, T. 2016. Physical Properties of Wood in Poplar Clones 'I-214' and 'S1-8'. *Drv Ind* 67(2): 163–170. https://doi.org/10.5552/drind.2016.1604

Jalaludin, Z.; Hill, C.A.S.; Samsi, H.W., Husain, H.; Xie, Y. 2010. Analysis of water vapour sorption of oleo-thermal modified wood of Acacia mangium and Endospermum malaccense by a parallel exponential kinetics model and according to the Hailwood-Horrobin model. *Holzforschung* 64(6): 763–770. https://doi.org/10.1515/hf.2010.100

Kamke, F.A. 2006. Densified radiata pine for structural composites. *Maderas-Cienc Tecnol* 8(2): 83–92. https://scielo.conicyt.cl/pdf/maderas/v8n2/art02.pdf

Kawai, S.; Wang, Q.; Sasaki, H.; Tanahashi, M. 1992. Production of compressed laminated veneer lumber by steam pressing. In Proceedings of the Pacific Rim Bio-Based Composites Symposium, Rotorua, New Zealand. pp. 121–128.

Kollmann, F. 1959. *Tecnología de la madera y sus aplicaciones.* Vol I. 1st edition. Instituto Forestal de Investigaciones y Experiencias y Servicio de la Madera, Madrid, Spain.

Kollmann, F.P.; Kuenzi, E.W.; Stamm, A.J. 1975. *Principles of wood science and technology. Vol. II Wood based materials.* 1st edition. Springer-Verlag, New York-Heidelberg-Berlin.

Kutnar, A.; Kamke, F.A.; Sernek, M. 2008. The mechanical properties of densified VTC wood relevant for structural composites. *Holz Roh Werkst* 66: 439–446. https://doi.org/10.1007/s00107-008-0259-z

Kutnar, A.; Sernek, M. 2007. Densification of wood. *Zbornik Gozdarstva in Lesarstva* 82: 53–62. http://www.gozdis.si/zbgl/2007/zbgl-82-6.pdf

Laborie, M.P.G. 2006. The temperature dependence of wood relaxations: A molecular probe of the woody cell wall. In: Proceedings of the Characterization of the Cellulosic Cell Wall, Blackwell Publishing, Grand Lake, Colorado, USA. pp 87–94. https://doi.org/10.1002/9780470999714.ch7

Laskowska, A. 2020. Impact of cyclic densification on bending strength and modulus of elasticity of wood from temperate and tropical zones. *BioResources* 15(2): 2869–2881. https://bioresources.cnr.ncsu.edu/wp-content/uploads/2020/03/BioRes_15_2_2869_Laskowska_Impact_Thermo_mechan_Densification_Bending_Str_MOE_Wood_Zones_16914.pdf

Lee, S.H.; Ashaari, Z.; Lum, W.C.; Halip, J.A.; Ang, A.F.; Tan, L.P.; Chin, K.L.;Tahir, P.M. 2018. Thermal treatment of wood using vegetable oils : A review. *Constr Build Mater* 181: 408–419. https://doi.org/10.1016/j.conbuildmat.2018.06.058

Li, X.; Bremer, G.C.; Connell, K.N.; Ngai, C.; Pham, Q.A.T.; Wang, S.; Flynn, M.; Ravetti, L.; Guillaume, C.; Wang, Y.; Wang, S.C. 2016. Changes in chemical compositions of olive oil under different heating temperatures similar to home cooking. *Journal of Food Chemistry and Nutrition*, 4(1): 07–15. https://esciencepress.net/journals/index.php/JFCN/article/view/1532

Lyon, F.; Thevenon, M.F.; Hwang, W. J.; Imamura, Y.; Gril, J.; Pizzi, A. 2007. Effect of an oil heat treatment on the leachability and biological resistance of boric acid impregnated wood. *Ann For Sci* 64: 673–678. https://doi.org/10.1051/forest:2007046

Morsing, N. 1998. *Densification of wood - The influence of hygrothermal treatment on compression of beech perpendicular to the grain* (Series R, N 79). Department of Structural Engineering and Materials, Technical University of Denmark, Lyngby, Denmark. https://core.ac.uk/download/pdf/13738419.pdf

Navi, P.; Girardet, F. 2005. Effects of thermo-hydro-mechanical treatment on the structure and properties of wood. *Holzforschung* 54(3): 287–293. https://doi.org/10.1515/HF.2000.048

Okon, K.E.; Lin, F.; Lin, X.; Chen, C.; Chen, Y.; Huang, B. 2018. Modification of chinese fir (*Cunninghamia lanceolata* L.) wood by silicone oil heat treatment with micro-wave pretreatment. *Eur J Wood Prod* 76: 221–228. https://doi.org/10.1007/s00107-017-1165-z

R Core Team. 2019. *R: A language and environment for statistical computing*. Version 3.6.1. R Foundation for Statistical Computing, Vienna, Austria. Retrieved from https://cran.r-project.org/

Rapp, A.O. 2001.Review on heat treatments of wood. In: Proceedings of the Special *Seminar* COST E22, Antibes, France. https://projects.bre.co.uk/ecotan/pdf/Heat_treatment_processes_Andreas_Rapp%20.pdf

Reiterer, A.; Stanzl-Tschegg, S.E. 2001. Compressive behaviour of softwood under uniaxial loading at different orientations to the grain. *Mech Mater* 33(12): 705–715. https://doi.org/10.1016/S0167-6636(01)00086-2

Song, J.; Chen, C.; Zhu, S.; Zhu, M.; Dai, J.; Ray, U.; Li, Y.; Kuang, Y.; *et al.* 2018. Processing bulk natural wood into a high-performance structural material. *Nature* 554: 224–228. https://doi.org/10.1038/nature25476

Sotomayor, J.R. 2016. Efecto del densificado de la madera de *Gyrocarpus americanus* Jacq . en su módulo dinámico determinado por ondas de esfuerzo [Effect of the densified of *Gyrocarpus americanus* Jacq. wood in its dynamic modulus established by stress waves]. *Ciencia Amazónica* 6(2): 162–171. https://doi.org/10.22386/ca.v6i2.117

Spear, M.; Walker, J.C.F. 2006. Dimensional instability in timber. In: *Primary Wood Processing, Principles and Practice*, J.C.F. Walker (Ed). Springer, Dordrecht, Netherlands. pp. 95–120. https://doi.org/10.1007/1-4020-4393-7_4

Tomak, E.D.; Hughes, M.; Yildiz, U.C.; Viitanen, H. 2011. The combined effects of boron and oil heat treatment on beech and Scots pine wood properties. Part 1: Boron leaching, thermogravimetric analysis, and chemical composition. *J Mater Sci* 46: 598–607. https://doi.org/10.1007/s10853-010-4859-8

Wang, J.Y.; Cooper, P.A. 2005. Effect of oil type, temperature and time on moisture properties of hot oil-treated wood. *Holz Roh Werkst* 63: 417–422. https://doi.org/10.1007/s00107-005-0033-4

Wehsener, J.; Brischke, C.; Meyer-Veltrup, L.; Hartig, J.; Haller, P. 2018. Physical, mechanical and biological properties of thermo-mechanically densified and thermally modified timber using the Vacu^3-process. *Eur J Wood Prod* 76: 809–821. https://doi.org/10.1007/s00107-017-1278-4

Welzbacher, C.R.; Wehsener, J.; Rapp, A.O.; Haller, P. 2008. Thermo-mechanical densification combined with thermal modification of Norway spruce (*Picea abies* Karst) in industrial scale – Dimensional stability and durability aspects. *Holz Roh Werkst* 66: 39–49. https://doi.org/10.1007/s00107-007-0198-0