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CHALLENGES IN WOOD DENSIFICATION: PROCESSING AND PROPERTIES

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ABSTRACT

Wood can be densified and moulded through the combined effects of temperature, humidity and mechanical forces. These processes so called THM treatments. Depending to the processing parameters the physical and mechanical properties of wood can improve considerably. Although, the principles of densification of small wood elements are well known, challenges are faced in scaling-up stage and on the shape memory. The aim of this paper is to explain the origin of the shape memory and the effect of combined Thermo-Hydro-Mechanical on the fixation of compression. The process of densification can be implemented in an open system, a semi-open system or a closed system. In an open system part of the boundary conditions is open to climate and control of moisture content as well temperature of the elements during densification is not possible. But in a closed system all these parameters are under control. Although, densification of wood in an open system improves the mechanical and durability of wood, but the compression-set is not stable. Post-processing THM treatments have shown to constitute an effective means of eliminating the set-recovery. We have shown that hydrolysis of hemicelluloses during THM post treatment plays an important role in the elimination of shape memory through the dissipation of the stresses (relaxation) stored in the microfibrils and the matrix during densification. We have shown that for a total elimination of shape memory it is necessary for the lignin to be in its rubbery state during the THM treatment.

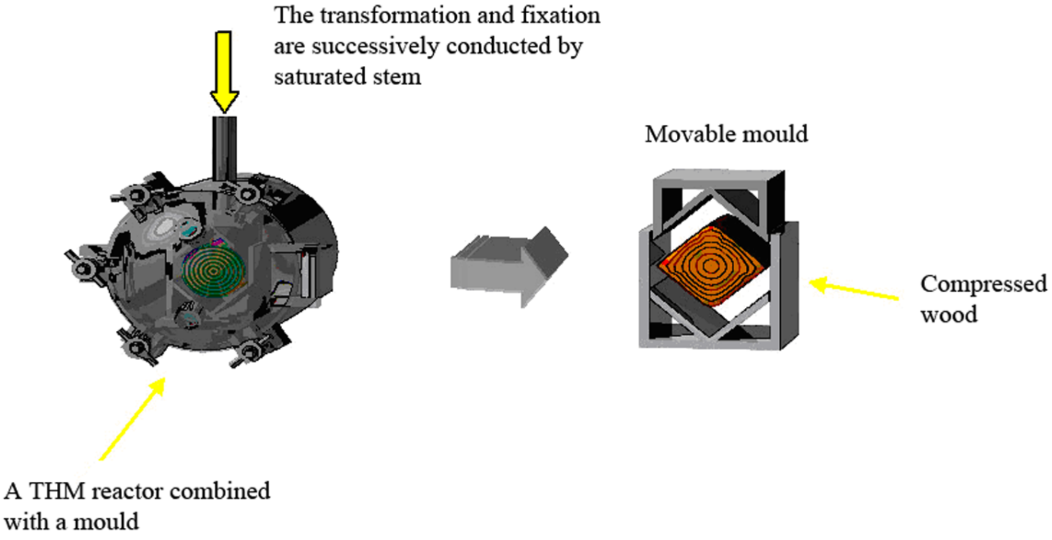
Key words: wood densification, thermo-hydro-mechanical treatments, post-treatments

INTRODUCTION AND BACKGROUND

Initially, the main aim of wood densification was to improve the mechanical behavior of wood by eliminating its porosity. Unfortunately densified wood in practice showed an undesirable behavior, tendency to return to its initial dimension, when it was subjected to heat/humidity. This phenomenon is known as „shape memory“ where „compression-set“ recovers. To eliminate this undesired phenomenon, Stamm and Seborg (1941) developed a process using wood impregnation with phenol formaldehyde resin during densification. This type of compressed-impregnated wood was industrialized under the name of „Compreg wood”. Today, in Germany, the fabrication of Compreg wood still exists in industrial scale. Stamm et al. (1946) also succeeded partially to eliminate the shape memory of densified wood by use of thermo-hydrous treatments. They showed that at 180 °C and processing time 30 minutes, the compression-set recovery of a thin plate decreased to few per cent. From these findings, a product called „Staypak“, Kollmann et al. (1975), was developed.

More recently, Norimoto et al. (1993) and Ito et al. (1998a) adapted the same process of heating to eliminate the shape memory. Tanahashi (1990), Ito et al. (1998b) have reported that the THM post-treatment of densified thin wood at 200 °C for only 4 minutes is sufficient to eliminate totally the shape memory. The primary goal of recent research on wood molding, wood densification and new techniques in THM is to extend it to wooden elements of larger dimensions for use in building construction. In Japan various investigations on THM molding of wood are developed. Two procedures have emerged from these studies. On the one hand Ito et al. (1998a) developed a system to transform small trunks of wood with round section into trunks with square section, (Figure 1), and on the other hand a second process was designed to obtain structural timber of high quality from wood with naturally weak density . This later process is known as “Compressed Lumber Processing System”. Apparently the problems related to the dimension of the wood elements (upscaling stage) such as the development of cracks and exfoliation during shaping, cooling and drying have not yet been solved in practice. Kyomori et al (2000), estimate that the acquisition of more fundamental knowledge on wood THM behavior will be required to overcome the issues that currently stand in the way of developing a new manufacturing system for “compressive molding of wood under high-pressure steam techniques”.

Since 1990s, various investigations and research works have been initiated in Europe, in the United States and most recently in Canada. Fang et al. (2011) shows the results on the post-treatment of densified wood veneer with heated oil. In Denmark, emanating from the work of Morsing (2000), a machine was developed for pre-compression of wood in the longitudinal direction. This type of longitudinal densified wood elements can be bent without any application of steam or jigs. In Switzerland, Heger (2004), Navi and Heger (2005) have investigated the origin of the set recovery and wood densification in a closed system. In Germany, Rapp et al. (2006), have developed a system for wood heat treatment based on oil at a high temperature (OHT). In this open system, rape-seed oil is used as a medium which can be heated above 200°C. The results have shown that oil-heating at a temperature above 200°C can completely eliminate the compression-set recovery of large densified spruce panels. This product has shown improved resistance against biodegradation but the strength decreases considerably.



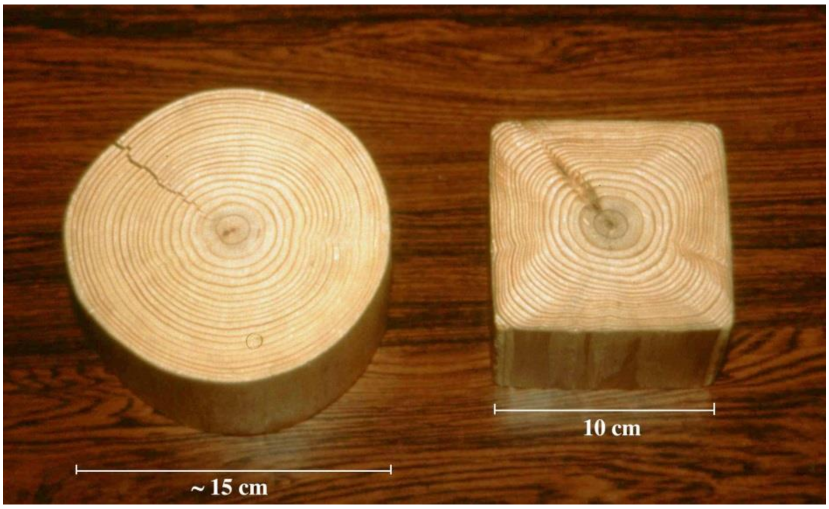


Figure 1: Transformation of a circular trunk to a square-section block by two-dimensional densification (Ito et al., 1998a).

In USA, due to the harvesting of rapidly grown wood with low density, the wood researchers have shown an interest in the possible opportunities for using densified wood in the production of composite layered materials (Kutnar et al., 2008). To densify small low-density hybrid poplar specimens, Kamke and Sizemore (2005) have developed a semi-closed THM reactor. This system might have had some advantages for the closed system developed in Japan or in Switzerland, but unfortunately the dimensions of specimens used for densification are still small and it would be difficult to evaluate the interest of this system when it is used for large-sized wood elements for building constructions purposes.

1. DENSIFICATION IN A CLOSED SYSTEM

As a consequence of wood polymeric nature and cellular structure, it is possible to densify wood transversally and longitudinally. Densification is produced by eliminating porosity through application of heat, humidity and compressive loads. There are different types of densification (Navi and Sandberg, 2012). It is possible to densify wood under open system at a high temperature (about 130 °C). This type of densification is simple, improves the mechanical and moisture-resistant properties of wood but the compression-set is unstable. One of the ways of eliminating the recovery-set is THM post-treatment, which has shown to be an effective means. This procedure usually is performed under a closed system. A photo of a small THM reactor is given in Figure 2.

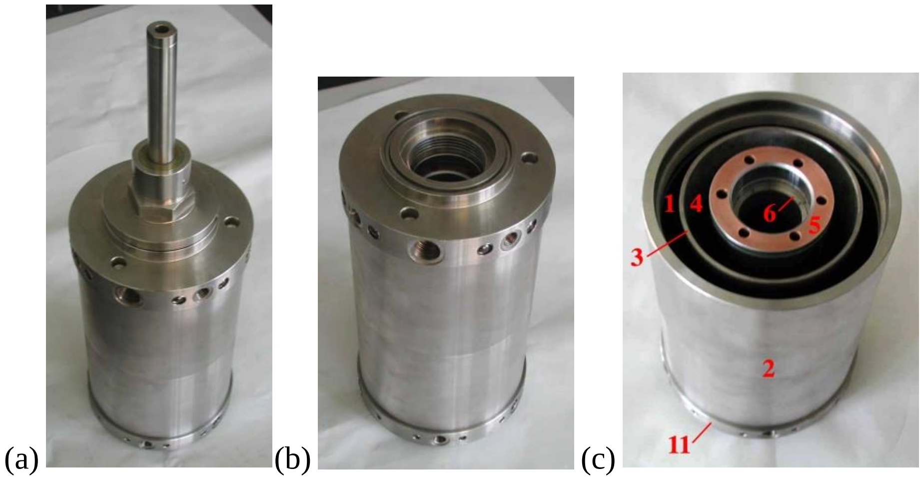


Figure 2: Photographs showing the THM reactor; (a) – THM reactor with piston, (b) – THM reactor without piston (c) – THM reactor after removing the cover. Chamber 1 and cylinder 5 (cylindrical camber) are connected to a heating circuit and are completely watertight. Chamber 4 is connected to chamber 6 (treatment-chamber and mould) by the cover, (Navi & Heger, 2005).

During THM processing chamber 1 and 5 are heated by saturated steam to reach the required temperature and the specimen placed in chamber 6 (treatment space) is heated by conduction. A diagram of a THM processing is shown in Figure 3. During densification and post treatment the temperature can be different. Beside the RH of the treatment space can vary too. A press is used to compress the specimen under a controlled displacement mode.

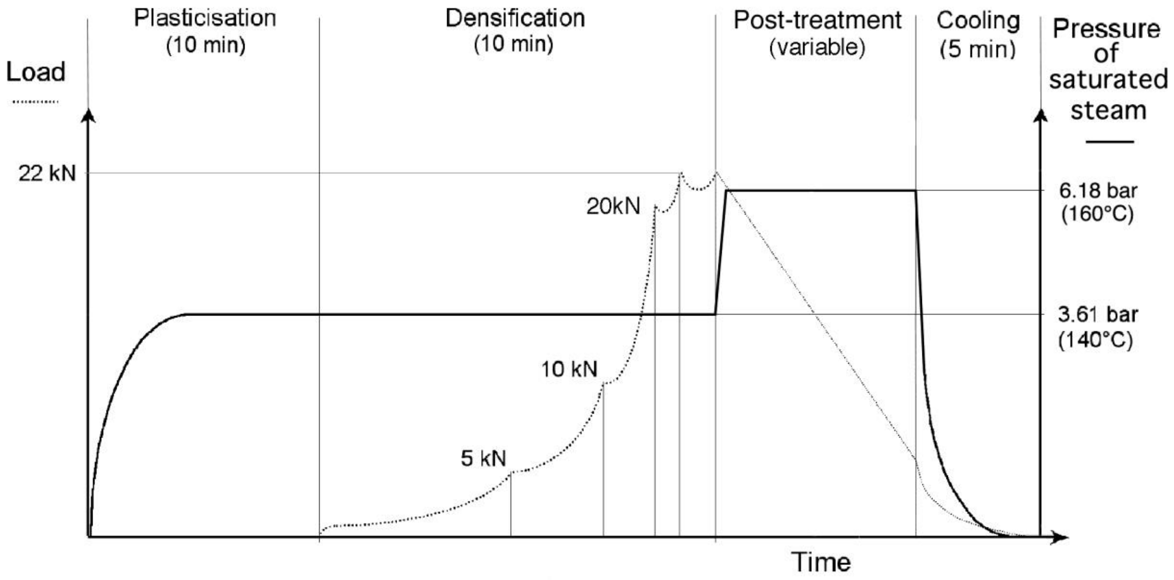


Figure 3: Load as a function of time during THM densification and post-treatment. External loading and steam pressure variations, the pressure is given for saturated steam, (Navi & Sandberg 2012)

The degree of compression-set recovery in densified wood may be investigated using a cyclic swelling/contraction test. This test consists of cycles of soaking in water at 60 °C and drying at 30 °C. Figure 4 illustrates the effect of temperature on the compression-set recovery of ten specimens of spruce densified in a closed system under saturated steam. Only the compression-set in specimen 10, which was post-treated at 200 °C for 5 minutes has been totally fixed.

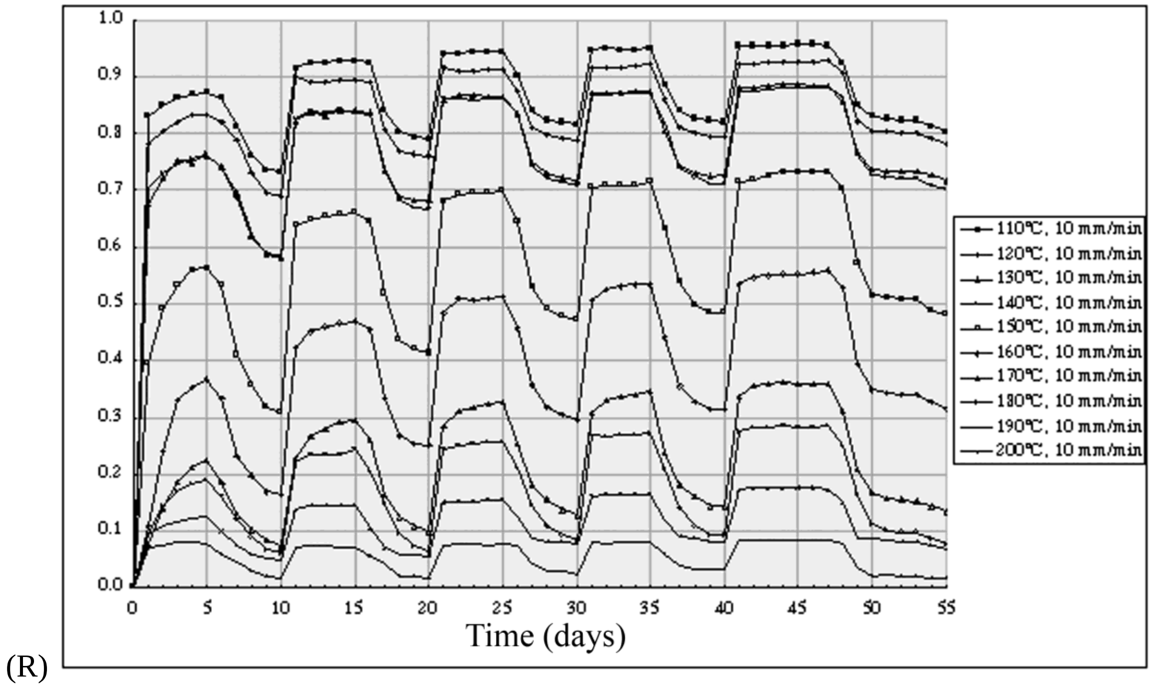


Figure 4: Recovery of compressed samples under saturation- drying cycles. Samples were subjected to compression at different temperatures between 110 °C and 200 °C (Heger, 2004).

2. ORIGINE OF THE RECOVERY OF COMPRESSION-SET

In the molecular model of composite cellulose-hemicellulose-lignin, the microfibrils are composed of crystalline and paracrystalline cellulose. The amorphous hemicellulose is connected on one side to the microfibrils by hydrogen bonding and on the other side to the lignin by covalent bonding. During the densification process, under the combined actions of water vapour, high temperature (more than110°) and compressive forces, all the polymeric components, with the exception of the crystalline cellulose, are plasticised (Black and Salmén. 1982), and undergo large deformations. When wood is transversally compressed, the macromolecules in the microfibrils are deformed elastically and the lignin undergoes elastic-viscoplastic deformation (the glass transition temperature of saturated lignin is much less than 110 °C). The elastic energy stored in the helical semicrystalline microfibrils and the lignin is considered to be the main cause of the shape memory effect. Two phenomena are responsible for the temporary fixation of the compression-set. In the first step, when the hot densified wood is cooled down to room temperature (below the glass-transition temperature of lignin) the lignin will pass from the rubbery to the glassy state and the deformed cellulose is consequently confined by a rigid matrix. On the continuation, when the sample is dried, the formation of the hydrogen bonds between hemicellulose and cellulose fixes the cellulose microfibrils in the deformed state. Then, re-humidification / heating will tend to disrupt these bonds and makes lignin rubbery, which lead to total or partial recovery of the densification.

3. FIXATION OF COMPRESSION-SET BY THM TREATMENT

Many wood products, such as bentwood, densified wood, moulded solid wood, laminated compressed wood, suffer from compression-set recovery. It is shown that high temperature saturated steaming reduces the shape memory effect significantly (Navi and Heger 2005). To investigate the mechanisms responsible for eliminating the shape memory and fixation of compression-set, a thermo-hydro-mechanical reactor was constructed. Figure 2 shows the part of THM reactor. With this system, small cylindrical samples of spruce (50x30mm diameter) were densified under saturated water vapour at 140 °C and subjected to THM post-treatments. The compression-set value (*C)* was calculated from

$$C = \frac{l_o - l_c}{l_o} \qquad \textbf{(1)}$$

where l_o and l_c are the lengths of the samples before and after densification respectively. After densification, many samples were post-treated at four different temperatures (140 °C, 160°C, 180 °C and 200 °C) and under five different relative humidity (RH) (dried, 40 %, 60 % 80 % and saturated) for different time of post treatment. All the samples then underwent the soaking-drying test to examine their recovery. The set-recovery value (*R*), is defined as

$$R = \frac{l_r - l_c}{l_o - l_c} \qquad \textbf{(2)}$$

where l_r is the length of the specimen after the set-recovery test. *R* varies between zero and one, zero means that no recovery set or total fixation of compression-set.

The *time necessary* for samples with R about zero was noted. Table 1 gives the post treatment time necessary for complete fixation of the compression-set (*R* = 0), at different temperatures under saturated and unsaturated conditions.

Table 1: Post-treatment processing time (in minutes) for complete fixation of the compression-set (*R* = 0) at different temperatures and relative humidity (RT)

Relative Humidity 0 40% 60% 80% 100%
140 °C not achieved 6000 mn. not achieved 2020 mn 210 mn*
160 °C 24250 mn not achieved not achieved 135 mn 70 mn
180 °C 5680 mn 341 mn 167 mn 50 mn 20 mn
200 °C not achieved not achieved 43 mn 16 mn 4 mn

** For various reasons we could not achieve complete fixation of the compression-set (R = 0) at some temperatures and RH. These were: 1 - when the specimens with low moisture contents were kept for long times (more than 50 minutes) at 200 °C in the reactor, they started to burn; 2 - when the specimens were treated at low temperatures with low RH, complete fixation was very difficult to achieve, probably because the lignin remained glassy.*

3.1. The role of hemicelluloses hydrolysis on fixation of compression-set

The experimental results show that the post-treatment time necessary to eliminate the compression-set at any temperature increases non-linearly with decreasing the time. To understand the mechanisms responsible for the elimination of the shape memory effect, the densified wood was subjected to chemical and physical analysis before and after post-treatment (Heger 2004). We have plotted the post treatment times given in table 1 against temperature (time in log scale) for each value of RH. The logarithm of these times varies linearly with temperature for a given RH with a slope that is approximately independent of RH. This suggested a straightforward relationship between the temperature, the relative humidity and post-treatment time. Figure 5 shows the data given in Table 1 plotted against 1/(R*T*), where R = 8,317 and *T* is the temperature on K.

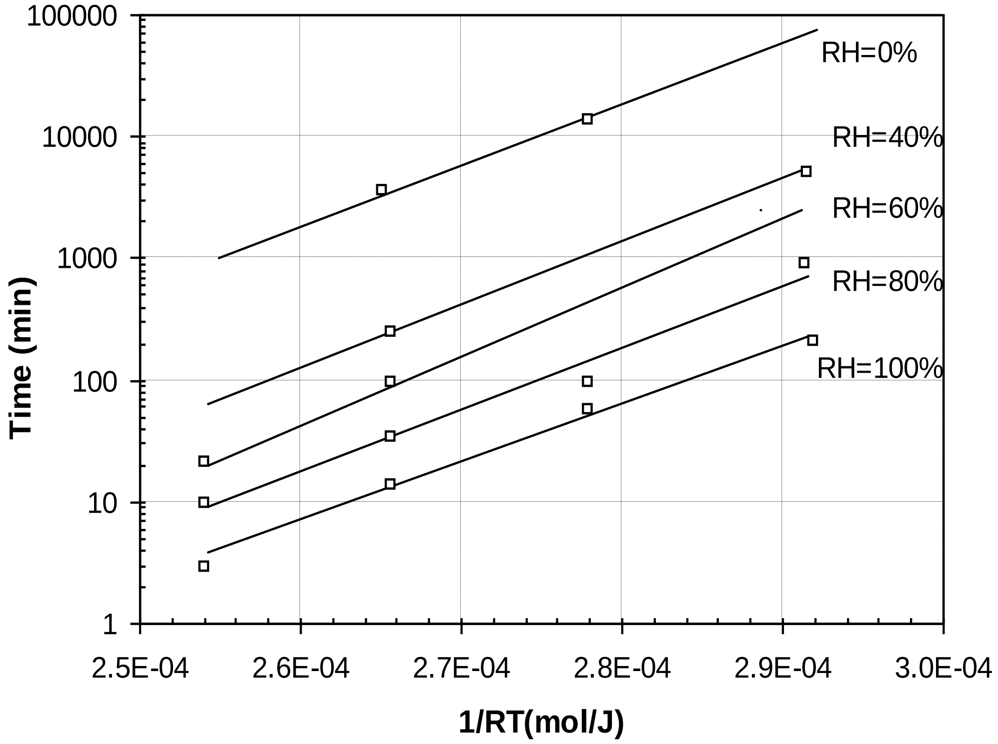


Figure 5: Value of post-treatment time to achieve complete fixation of the compression-set vs. 1/(RT).

This plot allows us to calculate the activation energy (E_A) of the process responsible for elimination of the shape memory effect for each RH through the following equation:

$$t(T,h) = \alpha\, e^{E_A/RT} \quad (3)$$

where α is a parameter dependent on RH (and also on the pH of the water during the post-treatment). The value of α is given by the intercept of the straight-line fit to the data with the time axis and E_A is the corresponding slope. The inferred values of E_A are given in Table 2.

Table 2: Values of the activation energy derived from Figure 5.

RH	**100 %**	**80 %**	**60 %**	**40 %**	**0 %**	**Average**
E_A (kJ/mol)	98,5	116,6	118,8	111,5	118,.4	112,8

E_A was roughly constant at around 98,5 – 112,8 kJ/mol, independently of RH. These measured activation energy values are very close to the value given by (Springer. 1966) for the xylan hydrolysis (E_A = 118 kJ/mol.). The relatively small value measured for 100 % RH might be because the initiation of hydrolysis is facilitated in the presence of saturated steam.

CONCLUSION

We have investigated the composite action of wood constituents on the elimination of the shape memory effect in densified wood by THM post-treatments at the molecular level. We have focussed on understanding the role of hemicellulose in the response of the cellulose-hemicellulose-lignin composite cell wall components. During THM post-treatment, splitting of the hemicelluloses chains that link the deformed crystalline microfibrils to the lignin allows internal stresses to be relaxed. We have introduced a model that links the degree of compression-set recovery to the rate of hemicellulose hydrolysis (Groux 2004, Heger 2004).

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