Thermo-mechanical Densification of *Populus tomentosa* var. *tomentosa* with Low Moisture Content

Dengyun Tu,* Xiaohua Su, Tingting Zhang, Wenjun Fan, and Qiaofang Zhou

This study used thermo-mechanical densification technology to compress low-moisture content (3~5%) rapid-growth *Populus tomentosa* var. *tomentosa* trees to produce specimens with a low-compression ratio (small volume loss) and a uniform density profile and desirable properties. Furthermore, the densified specimens were subjected to post-heat treatment at 180, 190, and 200 °C for 2, 3, and 4 h, respectively. Microscopic examination was performed to observe the changes that occurred in the wood vessels after densification. To determine the influence of post-heat treatment on the set recovery, the specimens were subjected to eight cycles of soaking and drying in 20 °C water and two cycles in boiling water. The density profile tendencies of the densified specimens were in accord with undensified specimens. Microscopic observation revealed that the deformations present in the densified wood resulted from the viscous buckling of cell walls without fracture. The volume of the void areas in the specimens decreased uniformly. Post-heat treatment can decrease compressive deformation, especially when applied at 200 °C for 4 h. After two boiling water cycles of soaking and drying, the densified wood still had a certain set recovery. Therefore, densified wood should be used sparingly in high temperature and high humidity environments.

Keywords: Post-heat treatment; Morphology; Density Profile; Set recovery

*Contact information: Department of Wood Science and Engineering, College of Forestry, South China Agricultural University, Guangzhou city, China; *Corresponding author: tudengyun@163.com*

INTRODUCTION

Wood is one of the oldest furniture materials for good reason, as it inherently has many advantageous properties, such as its high strength-to-weight ratio, excellent workability, beautiful grain, and excellent potential for modification. In future years, the use of wood will continually be important. For years, low-density wood from fast-growing trees has been utilized in great quantities. However, because of low-density wood's undesirable characteristics (*i.e*., lower surface hardness and wearing resistance), it is mainly utilized exclusively for its fibers (*i.e.*, producing manmade board and paper). Wood densification technology has been developed to overcome low-density, fast-growing wood limitations with respect to furniture applications.

Wood can be compressed, without fracturing its cell walls, when subjected to temperatures above its glass transition temperature, T_g. Wood is composed of polymeric constituents (*i.e.,* lignin, hemicellulose, and cellulose), and its detailed structure provides for porosity in the wood. Therefore, it is possible to densify wood transversally, producing a compression set by eliminating the pores either partially or totally using heat, water, and a compressive load (Navi and Heger 2004). Wood softens when it is subjected to significant amounts of heat, as its lignin, hemicellulose, and cellulose display different softening behaviors depending on the temperature and moisture content (Hillis 1984; Uhmeir *et al*. 1998). The dominant T_g is associated with lignin. There are many reports depicting the relationship between T_g and moisture content (Goring 1963; Irvine 1984; Sakata and Senju 1975; Takamura 1968). Kwei (1984) created a theoretical model to calculate T_g; the Kwei equation has been utilized by several researchers (Kelley *et al*. 1987; Wolcott *et al*. 1990) to describe the inverse relationship between T_g and moisture content in wood. Densification of wood *via* compression perpendicular to the grain requires that the cell walls are in a rubbery phase (Kutnar *et al*. 2009). The cell wall's rubbery state is essential for obtaining the compression deformation of cell wall buckling without cell wall fracture (Kutnar *et al*. 2009), which has a very important effect on the improved mechanical properties of densified wood (Blomberg and Persson 2004; Kutnar *et al*. 2008). Former studies often used high moisture content (12% or higher) to densify wood (Kutnar *et al*. 2008, 2009; Navi and Girardet 2000; Navi and Heger 2004). High moisture can soften the wood, but it also produces internal water-vapor pressure, causing the explosion of densified wood. Scheffler *et al*. (1981) reported an average moisture content (MC) of 8%, leading to an internal water-vapor pressure of approximately 1.55 MPa at a temperature of 200 °C. Navi and Girardet (2000) stated that the MC of samples for compression at 140 °C should be limited to 13% to eliminate the risk of explosions while pressing. Consequently, low moisture contents were assessed in densified wood to avoid vapor-pressure induced structural damage to the wood, as well as to guarantee the health and safety of the press-operating staff. To diminish internal water-vapor pressure, a longer closing time (*i.e.*, densification time) needs to be applied. Hong (1984) and Welzbacher (2008) reported reduced swelling in wood after it had been subjected to a prolonged densification period. Additionally, the process of lowering moisture content in densified wood can shorten the closing time significantly, increasing work efficiency. Because of experimental difficulties (*i.e.*, high-demand for machine performance and moisture control) and high wood variability (*i.e.*, wood species and density), the compression creep behavior of wood at low moisture contents has not been studied extensively.

When wood is compressed, cellulose macromolecules store elastic energy due to their semicrystalline nature. The elastic energy stored in these regions is considered to be the main cause of the shape-memory effect in compressed wood (Navi and Heger 2004). When compressed wood is heated and subsequently rewetted, its internal elastic energy will relax, and it will recover its initial shape. Heat-treatment technology has been developed to relax the internal elastic energy to avoid set-recovery. Treatment time and temperature have a great effect on set-recovery. Various studies on compressed solid wood (Dwianto *et al*. 1998, 2000; Inoue *et al*. 1993; Ito *et al*. 1998; Navi and Gigardet 2000) have reported that the set-recovery process can be almost totally eliminated if a high-temperature post-treatment is carried out, particularly in a closed press system under humid conditions.

The effects of density profile and heat treatment on low-moisture content, compressed *Populus tomentosa* were investigated in this study. The dimensional thickness swelling of different post-heating treatments at incremental temperatures and times was analyzed. This study considered both the wood products markets and the use of under-utilized, low-density wood species. This technology can enhance wood body properties that would suit selected end applications, such as furniture.

EXPERIMENTAL

Materials

The average oven-dried density of *Populus tomentosa* samples used in this study ranged from 0.39 to 0.50 g/cm^3. Plain-sawn specimens were prepared at dimensions of 300 mm long (grain direction) by 120 mm wide (tangential direction) by 24.8 mm thick (radial direction). Specimens were air-dried and dried in conventional dry kiln to the moisture content 10%, after that the specimens were dried in the oven with the temperature 50 ^{o}C to the moisture content of 3 to 5%, cooling the specimens in the dry condition.

Methods

The densification process

Densification was carried out on a typical, man-made board hot-platen machine, with the temperature of the upper and lower platens at 180 °C. The specimens were preheated under the hot-platen for 350, 400, and 450 s. The recorded pressure was in the range of 18 to 22 MPa, and the feed speed of the press was 2 mm/s. The specimens were densified from their original 24.8 mm thickness to the target thickness of 18.8 mm with a densification ratio of 24.2%. After densification, the specimens maintained compression for 480 s. Finally, after the specimen was removed from the press, it was assumed that densification had finished. Each treating process had 10 replicates. The details of the pressing parameters are given in Table 1.

Table 1. Parameters of Densification Treatments

Initial thick-ness (mm)	Target thick-ness (mm)	Densification ratio (%)	Pressure (MPa)	Feeding speed (mm/s)	Temperature (°C)	Pre-heating time (s)	Closing time (s)
24.8	18.8	24.2	18-22	2	180	350	480
						400	
						450	

Compression set

The compression set, or densification ratio, is defined as the ratio of the change in dimension to the original dimension and expressed as a percentage. The densification ratio can be calculated using Eq. 1,

$$c = [(T_0 - T_c) / T_0] \times 100\% \quad (1)$$

where c is the densification ratio (%) and T_0 and T_c are the thickness of the specimen before and after densification (mm), respectively. The thickness of the specimen was measured with a Mahr digital indicator (Germany) prior to, and immediately after, densification.

Set-recovery

Post-heat treatment was performed on densified specimens with preheating time 400 s at 180, 190, and 200 °C for 2, 3, and 4 h. For the measurement of set-recovery, slices (each 20 mm thick, 20 mm wide, and 30 mm long) were cut from specimens treated at various temperatures and times. Twenty replicates were used in different temperatures and times. Slices were first dried in a convection oven over night at 103 ± 2 °C to establish initial oven-dry dimensions. Then, the slices were soaked in 20 °C water for 3.5 h and again oven-dried. The procedure was repeated for a total of eight wet/dry cycles. After eight wet/dry cycles, the specimens were soaked in boiling water for 2 h and once again oven-dried. This procedure was repeated two times. After each cycle, the thicknesses of the slices were recorded. Set-recovery was calculated using Eq. 2, as follows,

$$D_r = [(T_r - T_c)/ (T_0 - T_c)] \times 100\% \quad (2)$$

where D_r is the set recovery (%), T_r is the oven-dried thickness of the specimen after eight 20 °C water cycles and two boiling water cycles, T_c is the oven-dried compressed thickness of the specimen, and T_0 is the oven-dried initial thickness of the specimen.

Microscopic examination

Microscopic examination was performed on both control and densified specimens. Before examination, both groups of specimens were conditioned in a controlled environment, with 65% relative humidity at 20 °C, until a constant weight was obtained. The cellular structure of the control and densified specimens, before and after densification, were examined with a Leica DFC295 stereoscopic microscope (Germany). Both control and densified specimens were evaluated with five replicates, allowing a representative image of the microstructure to be obtained.

RESULTS AND DISCUSSION

Morphology of Wood Subjected to Densification

Wood was densified to enhance its properties, essentially by reducing the cell lumen volume. In the densification process, the morphology of wood changes significantly. Typically during radial compression, the weakest earlywood deforms at a much faster rate than the latewood (Tabarsa and Chui 2001). However, *Populus tomentosa* is a diffused porous hardwood, with only minor differences in density between earlywood and latewood. Consequently, after it was densified, the earlywood and latewood buckled together. This buckling effect makes it possible for a uniform density profile, relative to undensified wood, to be derived as earlywood and latewood soften.

The anatomical characteristics of the densified *Populus tomentosa* were analyzed through the micrographs shown in Fig. 1. From the micrographs, it is clear that the cell lumen volume was reduced and the cells were deformed without fracture of the cell walls. Vessels collapse and flatten in the direction of the compression. Because the structural differences between earlywood and latewood in *Populus tomentosa* are minor, morphological differences between the specimens after the densifying process could not be distinguished. In this study, the morphology is inconsistent with previous results for poplar (Gernot *et al*. 2013; Jourez *et al*. 2001). There are two main reasons: (1) the low degree of densification; a high densification ratio will lead to a higher rate of deformation in the cell lumens, as well as a large reduction in the volume of the void spaces; and (2) the set-recovery; densified wood stores stress, and as it is subjected to heat treatment, the inner stresses relaxed and deformation was partly recovered.

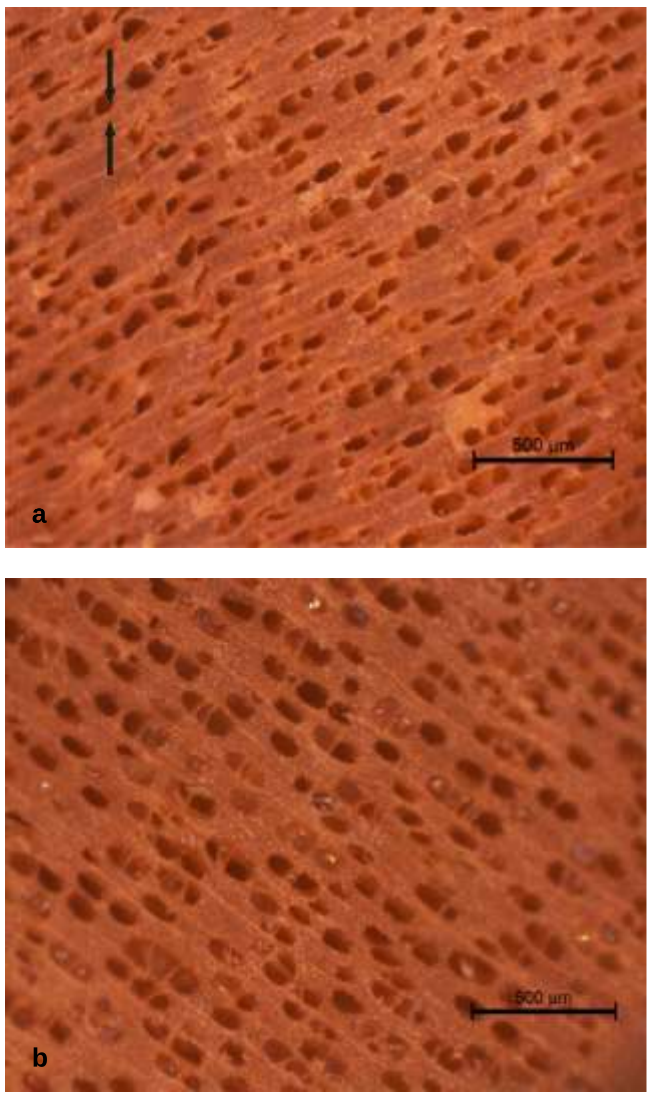


Fig. 1. Light photomicrographs of the (a) densified and (b) control wood specimens, which were heat-treated at 190 °C for 2 h. The compression was in the vertical direction, as shown by the arrows.

Density Profile

The density profile of the densified solid wood is dependent on the process parameters as well as the initial properties of the wood specimens, such as wood species, growth rate, and density. Processing time, temperature, and initial moisture content all affect the density profile (Camm and Quilter 2001; Kutnar *et al*. 2009; Wang and Cooper 2005). Figure 2 shows the density profiles of undensified and densified specimens after heat treatment (190 °C, 2 h) in the radial direction. For this study, the density profile varied with the heating time. When the heating time was 350 s, the wood surface temperature was high, meaning that the surface was softened more than the core layers of the wood. At this time, the densified wood had an M-shaped density gradient. The specimens had high-density surface regions and a low-density core. This kind of density profile is typical for wood-based composite panels. When the heating time was 400 s, the densified wood had the same shape with the undensified wood. The density in the core layers increased when the heating time was increased to 450 s. A long heating time means a high temperature and higher moisture content in the core layers, which leads to a softening effect. Kutnar *et al*. (2009) studied the density profiles of viscoelastic thermal compressed hybrid poplar (*Populus deltoides*× *P. trichocarpa*), grouping the specimens into three degrees of densification (63, 98, and 132%). They found a relationship among the three densification groups, surface density, core density, and density throughout the wood. They claimed that the effects of moisture content, temperature, press closing speed, compression force, and time in the press all can influence the density profile. Furthermore, thickness also has a great effect on the density profile. Thinner wood specimens lose moisture more rapidly and closely approach a uniform distribution, more so than thick specimens. For this study, the regulation of the density profile was in accordance with the findings by Kutnar *et al*. (2009). The densification process and ratio of densified wood for this study were different from that of Kutnar *et al.* (2009); however, both studies obtained similar end results. Consequently, the style of density profile is dependent on the distribution of moisture content and temperature. Accordingly, the study of the regulation of moisture and temperature coupling transfer in wood has theoretical significance for densified wood.

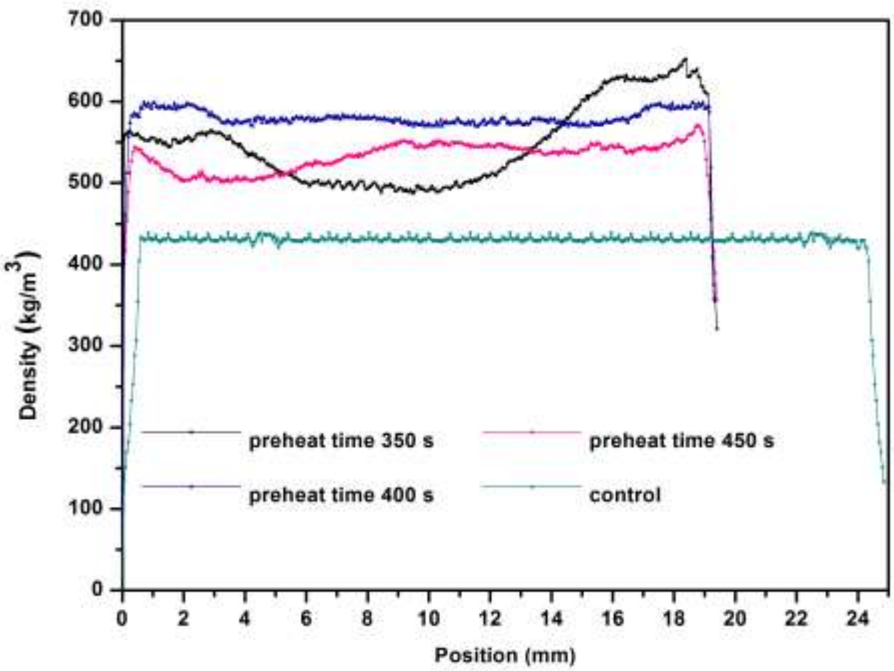
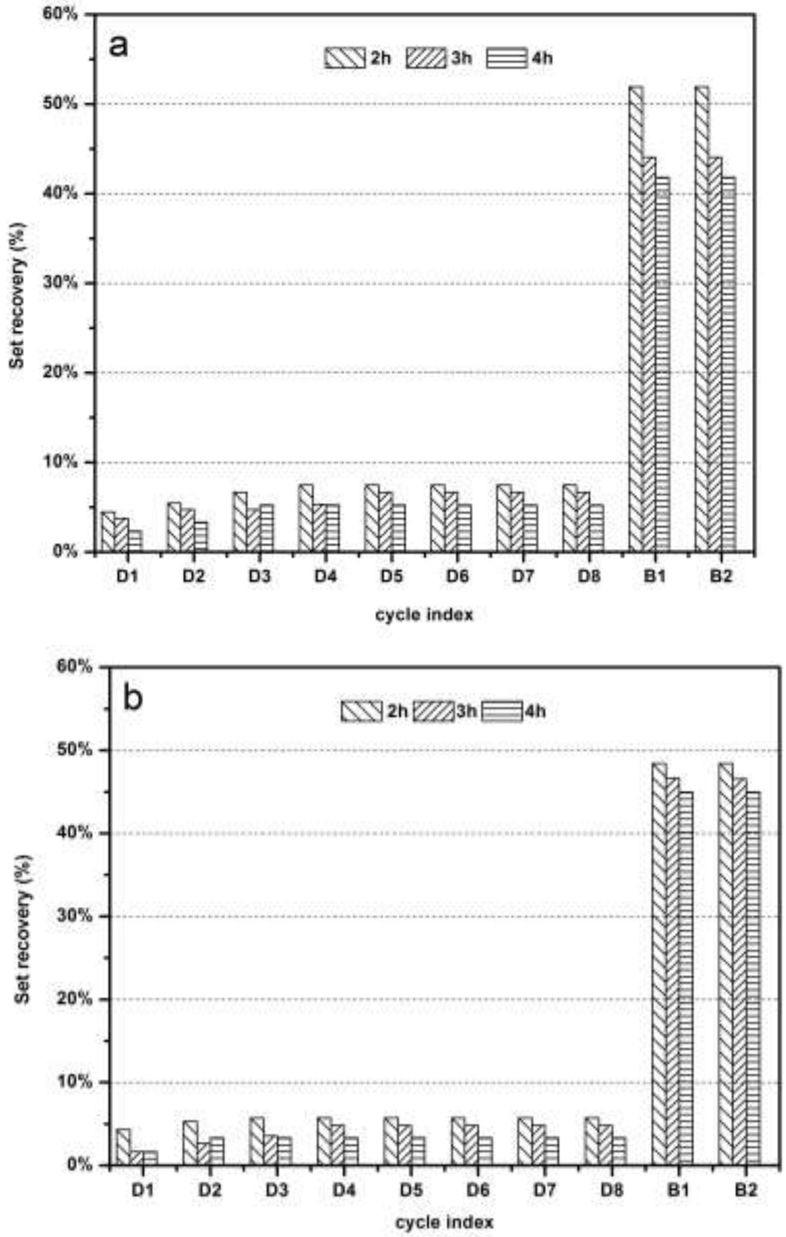


Fig. 2. Density profile of the control and densified wood at heating times of 350, 400, and 450 s, with post-heat treatment at 190 °C for 2 h

Set-recovery

Norimoto *et al*. (1993) considered the following three mechanisms to be essential in preventing deformation recovery: (1) the formation of cross-linkages between molecules of the matrix constituents, preventing the relative displacement of microfibrils; (2) the relaxation of the stresses stored in the microfibrils and the matrix; and (3) treatment to isolate the hydrophilic cell wall constituents (especially the hemicelluloses) from moisture to prevent their re-softening. These three mechanisms may be achieved through post-heat treatment. Inoue *et al*. (1993) found that near-complete fixation can be achieved by post-steaming compressed wood for 1 min at 200 °C or for 8 min at 180 °C. Reynolds (2004) investigated the swelling characteristics of densified wood and concluded that thickness swelling could be significantly reduced with exposure to saturated steam at 200 °C. Dwianto *et al*. (1999) studied the mechanism of the permanent fixation of compressive deformation of wood through high-temperature steaming of sugi (Japanese cedar). The results showed that the recovery of compressive deformation decreased with steaming time and reached almost 0 after 10 min at 200 °C. Permanent fixation from steaming below 200 °C was considered to be due to the chain scission of hemicelluloses, accompanied by a slight cleavage of lignin.

The effects of set-recovery were dependent on both the temperature and time of the post-heat treatment (Fig. 3). In this study, the densified specimens were subjected to post-heat treatment at temperatures of 180, 190, and 200 °C for 2, 3, and 4 h. The specimens were subjected to eight cycles of soaking and drying in 20 °C water and two cycles in boiling water. From Fig. 3, it is apparent that the post-heat treatment played a significant role in reducing set-recovery. Both temperature and time had an effect on set-recovery, with the effect of temperature being more significant than that of time. High-temperature heat treatment resulted in smaller set-recovery in all examined heat-treatment conditions. After eight soak/dry cycles (at the same time interval of 2 h and temperatures ranging from 180 to 200 °C), the set-recovery dropped sharply, from 10% to 6.8%. The cycle at 200 °C for 4 h seemed to be critical, in that the set-recovery was only slight (3%, average).



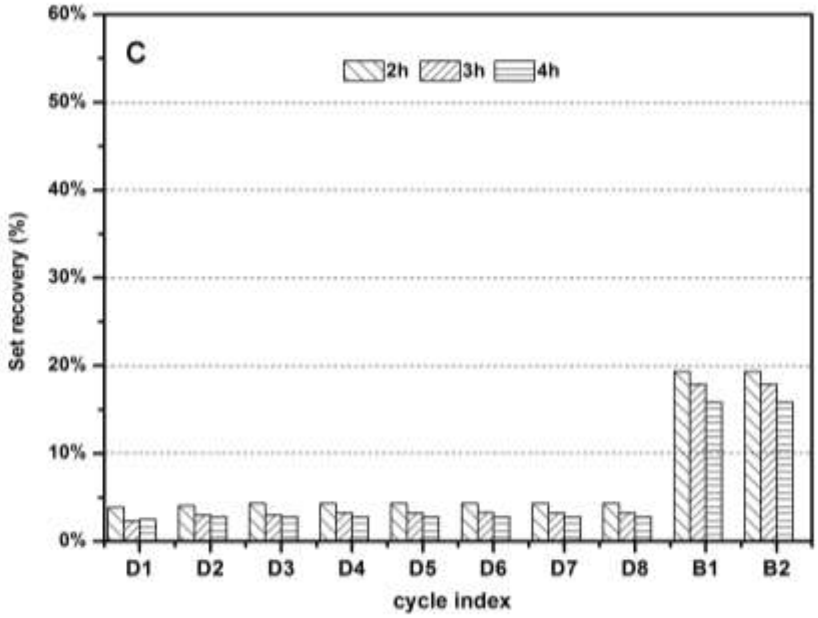


Fig. 3. The average set-recovery of specimens post-heat-treated at (a) 180 °C, (b) 190 °C, and (c) 200 °C for 2, 3, and 4 h; results are shown after eight 20 °C water (D1through D8) and two boiling water (B1 through B2) soaking and drying cycles

After two soak/dry cycles in boiling water, densified wood still had a certain set recovery (that at 200 °C for 4 h was 13.8%). Steam post-treatments were not able to improve the durability of densified specimens; such improvement is necessary for high-temperature conditions or outdoor use (Welzbacher *et al*. 2008). The set-recovery in this study could not be completely fixed after post-heat treatment, which differed from the findings based on oil heat-treatment (Welzbacher *et al*. 2008) and a thermo-hydro-mechanical process (Kutnar and Kamke 2012; Navi and Heger 2004). These differences can be attributed to the presence of steam in the heat-treatment process used in this study. The induced steam in the heat-treatment process had a negative effect on the dimensional stability of the densified specimens.

CONCLUSIONS

1. The microscopic examination of densified wood established that the densifying process adequately plasticizes wood. Deformations in the densified wood are largely the result of the viscous buckling of cell walls, without any fracture taking place.
2. A preheat time of 400 s, hot-platen temperature of 180 °C, and closing time of 480 s can produce a uniform density profile. Compressing wood with low moisture content is a valid technical approach for improving the density of soft wood.
3. Post-heat treatment can reduce set-recovery effectively; the specimens' exposure to 200 °C for 4 h after compression resulted in the smallest set recovery, 3%. However, the densified wood had some set-recovery after the boiling water test, and one must use caution when using densified wood in a high-temperature and high-humidity

environment, such as sauna room where densified wood may easily develop uneven thickness because of set recovery.

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