Original article

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In-situ penetration of ionic liquids during surface densification of Scots pine

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Abstract: The moisture-induced recovery of compressed wood is one of the major problems of wood densification technology. Achieving a cost-efficient surface densification process without the need for additional resins to eliminate the set-recovery may lead to an increase in value of low-density wood species. A previous study has shown that a pre-treatment with ionic liquids (ILs) can nearly eliminate the set-recovery. It was however observed that during the pre-treatment process the IL did not penetrate sufficiently deep into the wood to explain the achieved reduction in set-recovery. Based on these findings, the hypothesis was posed that further penetration of the IL into the wood occurs during the densification stage as a consequence of the applied heat and pressure. Thermo-gravimetric analysis (TGA) and gas-chromatography mass-selective-detection (GC-MSD) showed that the depth of penetration of the IL was greater after the densification process than before. Digital image correlation (DIC) showed that in regions with a high IL concentration, there was almost no set-recovery, and it gradually increased with a decrease in the IL concentration, as observed with TGA and GC-MSD analysis.

Keywords: compressed wood; digital image correlation; set-recovery; spectroscopy; thermo-gravimetric analysis; wood modification.

1 Introduction

Surface densification, i.e. compression in the transverse direction, of Scots pine sawn timber can lead to a more than twofold increase in surface density, resulting in a hardness level comparable to more expensive wood species, such as oak (Rautkari et al. 2013). Densifying only a relatively thin layer of the wood, as opposed to bulk-densification, reduces the needed process resources and maintains a larger share of the wood volume, without detrimental effects on the desired improvement of the material properties. This potentially facilitates the development of a large-scale and commercially viable surface densification process, which would open up new market opportunities for widely available low-density wood species, among them Scots pine.

In the past decade, many studies on surface densification of wood have explored the influence of different variables and process parameters on the material properties. Pizzi et al. (2005) and Rautkari et al. (2009) successfully densified the surface of wood with friction welding equipment, but the vast majority of surface densification studies converged on using a simple hot press (Gong et al. 2010; Lamason and Gong 2007; Rautkari et al. 2011; Rautkari et al. 2013).

Arguably the biggest obstacle preventing surface-densified wood from large-scale commercialisation is the set-recovery effect, i.e. the moisture-induced re-shaping of the compressed wood cells back to their shape before densification, which results in a loss of all improvements in the material properties. Even though, there is no universally accepted theory describing the exact mechanism of set-recovery on a molecular level, Navi and Sandberg (2012), and Navi and Pizzi (2014) suggest that during the densification process, the hemicellulose and lignin matrix surrounding the crystalline cellulose is plasticised, usually by moisture and heat, which allows plastic deformation, whereas the crystalline cellulose is not plasticised, and thereby only undergoes elastic deformation. After cooling the wood before opening the press, this elastic deformation is locked by the solidified hemicellulose-lignin matrix. However, when subsequently exposed to moisture, the matrix re-plasticises,

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which releases the inner stresses and the elastic deformation in the crystalline cellulose, resulting in the recovery of the deformed cells. Typically, the set-recovery of surface-densified wood is about 80–90%, i.e. it is almost complete (Laine et al. 2013; Neyses et al. 2017).

In the past, many studies have been carried out with the aim to reduce, or even eliminate, the set-recovery of both bulk-densified and surface-densified wood by applying methods of chemical modification, resin impregnation, or thermo-hydro-mechanical (THM) processing. THM processing (Kutnar and Kamke 2012a; Navi and Giradet 2000) and post-treatments with steam in a closed process (Inoue et al. 1993; Navi and Heger 2004) for up to 20 min can lead to an almost complete elimination of the set-recovery. Laine et al. (2012) achieved similar results with thermal post-treatment in an open system, but the process duration was several hours. Resin impregnation treatments have also been successful, as shown by Stamm (1964), Gabrielli and Kamke (2010), Pfriem et al. (2012) and Khalil et al. (2014). Unfortunately, there are several drawbacks that limit the commercial potential of these approaches: they require a closed system to achieve a short process duration, which makes integration into continuous processing chains – as common in the wood industry – very difficult, whereas thermal post treatments and resin impregnation methods consume great amounts of time of high energy input.

The mechanism of set-recovery as proposed by Navi and Sandberg (2012), and Navi and Pizzi (2014) suggests that the set-recovery will be greatly reduced if plastic deformation of the crystalline cellulose can be achieved prior to or during the densification process. This means that the development of a resource-efficient method for plasticising crystalline cellulose might result in a significant step towards a time- and cost-efficient industrial surface densification process.

Since the beginning of the 21st century, studies on ionic liquids (ILs) have become increasingly popular in the pulp and paper and in the textile industries. ILs are generally defined as salts with melting points below 100 °C, and in some cases, below room temperature (Hanabusa et al. 2018). Among others, Swatłowski et al. (2002) and Kilpeläinen et al. (2007) reported the ability of imidazolium-based ILs to plasticise and even dissolve cellulose. Rieland and Love (2020) suggested that the cellulose dissolving ability of ILs is due to

their ability to form hydrogen bonds with the hydroxyl groups of the cellulose chains. The anion starts the reaction, swelling the biomaterial and allowing the usually bulkier cations to form hydrogen bonds, thereby forming a “solvation cage” and creating repulsion between the cellulose chains.

For this reason, a fast treatment of Scots pine with different ILs prior to surface densification was tested in a previous study by Neyses et al. (2020), resulting in a substantial reduction in the set-recovery. In addition, the Brinell hardness was increased by a factor of 1.4 over untreated and surface-densified wood. The pre-treatment was carried out by applying an IL diluted with ethanol to the wood surface, followed by a waiting time of 1 min, a procedure then repeated three times, after which the treated wood was surface-densified at temperatures above 200 °C for 4 min.

In order to determine the depth of penetration of the ILs into the wood, a dye was added to the IL solution. The penetration depth of the IL before densification was approximately 1 mm, but to achieve the measured reduction in set-recovery the depth of penetration must have been at least 4 mm in the undensified state (Figure 1).

To explain this apparent contradiction, the following hypothesis was formulated (Figure 2): during the densification process at temperatures above 200 °C, (1) the IL starts to evaporate and is transported deeper into the wood, as it cannot evaporate through the press platen (2). This continues throughout the densification process, as heat is transferred from the press platen into the wood surface, activating the IL, which cleaves hydrogen bonds in the crystalline cellulose regions and increasingly plasticises the wood (3, 4). After some time, the maximum level of penetration is achieved and the ionic liquids starts to decompose, which opens up the hydrogen bond sites between the cellulose chains (5). As the wood is still under heat and pressure, the cellulose chains can fuse together permanently and form new bonds, thereby preventing the set-recovery. Thus, the core of the hypothesis is that a critical part of the penetration of the ILs into the wood surface happens during the densification process.

Several studies have explored the decomposition behaviour of different ILs (Ngo et al. 2000; Ohtani et al.

Figure 1: The contradiction between observed penetration of the IL-solution into the undensified wood surface (left) and the measured set-recovery after densification (right).

Figure 2: Cross-sectional view of a IL-treated wood specimen in a single-platen heated press, illustrating the hypothesis of *in-situ* penetration of ILs into the wood surface during the densification process.

2008), and a study by Efimova et al. (2013) is of particular relevance for the present study. The authors conducted thermo-gravimetric analysis (TGA) and evolved gas analysis (EGA) on the IL 1-butyl-3-methylimidazolium chloride (BmimCl), and showed that BmimCl starts to decompose at approximately 220 °C. This shows that TGA might be a suitable method to study the penetration and decomposition behaviour of ILs in densified wood.

The objective of this study was to test the hypothesis described above through a multi-sensor data fusion approach, derived from thermo-gravimetric (TG), chromatography mass-selective-detection (GC-MSD) and digital image correlation (DIC) data on densified wood treated with ILs.

2 Materials and methods

2.1 Specimen preparation

Specimens of Scots pine (*Pinus sylvestris* L.) sapwood were cut to dimensions of $19 \times 24 \times 140$ mm (R $\times$ T $\times$ L). The oven-dry density was between 384 and 499 kg m⁻³. The annual growth-ring orientation in the cross section of the specimens were within $\pm 20^\circ$ from being parallel with the densified surface. The specimens were dried in an oven at a temperature of 70 °C for two days, until reaching a moisture content (MC) of 0%, followed by treatment with the IL 1-butyl-3-methyl-imidazolium chloride (BmimCl) of 98% purity (Acros Organics, Geel, Belgium) and densification. The outer (bark-side) tangential surface of the specimens was used for the pre-treatment and surface densification in the radial direction:

C_W: control group of untreated wood.

D_W: densified wood. The specimens were pre-heated in a hot press at 250 °C, without applying any pressure for 30 s, after which a pressure of 2.9 MPa was applied for 90 s. The press was then cooled to a temperature of 80 °C, over a period of about 120 s, before opening.

BmimCl_W: wood treated with BmimCl. A solution of BmimCl in ethanol (20/80 wt%) was applied with a pipette to the surface of the specimens. This procedure was repeated three times in 1 min intervals, followed by evaporation of the ethanol in an oven at 70 °C.

BmimCl_D_W: BmimCl-treated and densified wood. The IL treatment and densification was carried out in the same way as for the groups BmimCl_W and D_W.

BmimCl: pure BmimCl as provided by the supplier.

Each group except for the pure BmimCl consisted of 10 specimens.

2.2 Thermo-gravimetric analysis and evolved gas analysis

For the TGA and EGA measurements, specimens of wood powder were sanded from each group as shown by Figure 3, with two replicates being taken for each group. To avoid any end-effects, about 50 mm of material was cut away from each side in the length direction, and 5 mm of material was cut away from the width of the specimens. The MC of the sanded specimens was approximately 6%.

The TGA measurements were conducted in a nitrogen atmosphere on a Perkin Elmer TGA 4000 Thermogravimetric Analyzer. The test procedure started with a 5 min holding period at a temperature of 30 °C, followed by a linear increase in temperature at a rate of 10 °C min⁻¹, until reaching 730 °C. After a holding period of 10 min, the TGA chamber was cooled to a temperature of 30 °C. The specimen weight was measured once per second. To determine the highest rate of weight loss, the first derivative dw/dt (DTG) was calculated over the entire temperature range.

To validate the results from the TGA measurements, evolved gas analysis (EGA) was done on a Perkin Elmer Frontier Fourier-transform-infrared (FT-IR) spectrometer, which was connected to the TGA device via a tube. It measures the gases evolved from the TGA and the corresponding Gram-Schmidt profile plots the absorbance of gases over the temperature at which they evolved. Ideally, the peaks in the DTG curve should overlap with the peaks in the Gram-Schmidt profile. The data points for the Gram-Schmidt profiles were collected at a rate of 1 s⁻¹.

Figure 3: Experimental design of the thermo-gravimetric analysis to study the penetration of ILs during the densification process. The black horizontal bars in the cross-sectional view of the specimens indicate the depths beneath the specimen surface from where wood powder for the TGA analysis were collected. C_W = untreated wood; D_W = densified wood; BmimCl_W = undensified wood treated with BmimCl; BmimCl_D_W = densified wood treated with BmimCl.

2.3 Gas-chromatography mass-selective-detection

GC-MSD was used to detect the existence of BmimCl within the densified wood material. Specimens of the groups C_W, BmimCl_W, and BmimCl_D_W were cut to a dimension of 1×1 mm. To determine the depth of penetration of the BmimCl solution, small wood chips were cut with a knife, parallel to the densified wood surface. The cutting depth was measured with a caliper. For each 0.5 mm, the wood flakes were collected, and this formed one sample. BmimCl was extracted by soaking each sample in 5 ml of methanol for 2 h.

GC-MSD analysis was performed on an Agilent 5890 gas chromatograph equipped with a Gerstel automatic liquid sampler coupled to an Agilent 5977B High Efficiency Source (HES) single-quadrupole mass detector. Capillary GC analysis was performed on an HP5-MS ($30 \text{ m} \times 0.25 \text{ mm id}$, $0.25 \mu\text{m}$) capillary column with helium as the carrier gas. The GC starting temperature was 50°C , held for 1 min, then gradually increased to 280°C at a rate of 5°C min^{-1} , and then held again for 1 min, with a carrier gas flow rate of 1.2 ml min^{-1} . One micro litre of sample was injected; splitless mode. The Injection temperature was 250°C . The transfer line, ion source, and quadrupole analyzer temperatures were maintained at 290°C , 230°C , and 150°C , respectively. A solvent delay of 4.0 min was selected. In the full-scan mode, electron impact ionization (EI) mass spectra in the range of $33\text{--}600 \text{ m z}^{-1}$ were recorded at 70 eV electron energy.

2.4 Digital image correlation

DIC is a non-contact full-field measurement technique typically used for spatial studies of shape, velocity, displacement, and strain of structures. DIC can be based on patterns that are applied to the object surface, such as a speckle pattern, which are then correlated in the image data to obtain measurement data related to the full field. DIC can also be based on stochastic natural patterns at the surface of the object (Sutton et al. 2009). In a study on wood, DIC was used to measure crack propagation at the scale of the cell walls within the annual growth rings (Thuvander et al. 2000).

A macro-DIC analysis was applied on the stochastic natural object-based pattern of surface-densified wood in order to study the set-recovery on a local early-/latewood annual ring level. Two specimens with dimensions of $17 \times 24 \times 30 \text{ mm}$ ($R \times T \times L$) were cut from larger specimens of the BmimCl_D_W group. Images of the cross sections from both specimen sides were taken and further investigated with DIC analysis. The reference image stage was captured in the surface-densified state before any set-recovery occurred, and a second image stage was captured after soaking in water for 24 h, followed by oven drying for 24 h, which initiated set-recovery. The MC in both cases was 0%. This method of initiating the set-recovery is widely established in the field of wood densification (Kutnar and Kamke 2012b; Laine et al. 2013).

The images were captured with a Nikon D850 camera, equipped with a FX, CMOS, $35.9 \times 23.9 \text{ mm}$ image sensor of 45.7 effective megapixels. This camera has a back-illuminated image sensor for the reduction of image noise. A Nikon AF-S 105/2.8 macro lens of a fixed focal length and a macro extension tube with a length of 20 mm and a diameter of 62 mm was used.

Four fibre-light gooseneck illuminators were placed at an angle of 45° relative to the cross-sectional surfaces of the specimens (Olympus Highlight 3000 Microscope Fiber-Light Illuminator, and Fiberoptic Heim Fiber Optic Light Source type FI L151). Three polarizing filters

(Hoya Polarisation 62 mm linear filter, Tokina Co., Ltd., Japan) were used, one on the macro lens and two on the fibre-light sources. Cross polarisation using orthogonal linear polarisers on light sources and camera improves DIC measurements, as it enables a higher spatial precision, and can eliminate saturated pixels that degrade the DIC measurements, and the cross polarisation also reduce reflections and glare which leads to an increased image contrast and saturation (LePage et al. 2016). A standard white panel was used as a reference to set the white balance. The exposure used was ISO 100, with a shutter speed of $1/3 \text{ s}$ and an aperture of $f/18$. To reduce possible vibrations and minimise blurring caused by the shutter, the shutter was raised and fixed in the mirror-up position between images.

Prior to testing, the lamps and the camera system were allowed to reach a stable operating temperature. Testing was performed in a clean and dust-free laboratory with no external fluorescent light or sunlight. The distance between the specimens and the camera image sensor was 300 mm. DIC analysis was performed using the software ARAMIS 2019 (GOM GmbH, Braunschweig, Germany).

The DIC data described in this work were obtained from TIFF images, each having a resolution of 8256×5504 pixels. As the images were captured for both cross-sectional surfaces of each of the two specimens, before and after inducing the set-recovery, this resulted in eight image sets. For each of these image sets, 10 images were taken to derive their median, which was then used for the DIC analysis. This approach improves the spatial resolution for differentiating between small details and reduces random system noise.

The output from the DIC analysis was based on a technical strain formulation in the epsilon Y-direction (ϵ_y), which is used to measure the local set-recovery of the cross-section surfaces. For the DIC surface component, bicubic subpixel interpolation was used, with a maximum residual of 15, a maximum intersection deviation of 0.1 pixels, and a minimum pattern quality of 0.1 pixels. Spatial median filter one was used, and no temporal filters and no tolerance sources were used. Subset matching was conducted against the reference stage, with automatic subset starts, a strain tensor neighbourhood of one in a hexagonal shape function mesh, and an interpolation size of 15. The full-field strain representation was three-sigma-scaled. As the basis for the DIC mesh, a subset size of 30×30 pixels at a step size of 13 pixels was used. One pixel corresponds to 0.0034 mm on the specimen surface.

3 Results and discussion

3.1 Thermo-gravimetric analysis and evolved gas analysis

The thermogravimetric (DTG) curves and Gram-Schmidt profiles of the specimens taken from a depth of $0\text{--}0.5 \text{ mm}$ beneath the densified surface are shown in Figure 4. The vertical axis ($dw \text{ dt}^{-1}$) is without scale, as the absolute values between the specimen groups did not correspond well to each other, and in this case only the peak positions and relative peak sizes were of interest. The DTG curve of untreated and densified wood (group D_W) was omitted for simplicity, as it was virtually identical to the curve of the

untreated wood (group C_W). The peaks of the DTG curves and Gram-Schmidt profiles corresponded well to each other, thereby validating the TGA measurements.

In the DTG curves, the untreated wood and the pure IL (BmimCl) each have one clear peak at 364 °C and at 308 °C, respectively. The location of the IL peak is similar to that reported by Efimova et al. (2013). The specimens taken from the ionic liquid-treated wood and the ionic liquid-treated + densified wood both have a second peak, which is at the same location as the peak of the pure IL. These findings showed that TGA can be a suitable approach to test the hypothesis formulated for this study. The size of the IL peak in relation to the wood peak was larger in the specimens of the BmimCl_W group than that of the BmimCl_D_W group. As the latter was densified at a high temperature, part of the decomposed IL probably disappeared during or after the densification process.

Figure 5 shows the intensity of the IL-peak as a function of the depth beneath the wood surface of undensified and densified wood, both treated with ILs. The IL peak disappears at a similar depth in both densified and undensified wood (Figure 5, left image). However, when correcting for densification by virtually expanding the densified wood to its original thickness, the IL penetration increases from 2 mm to approximately 4 mm in the densified and expanded state (Figure 5, right image). These data support the hypothesis of *in-situ* penetration of the IL during densification.

3.2 Gas-chromatography mass-selective-detection

The results from the GC-MSD study are summarised in Figures 6 and 7, which show the concentrations of 1-butylimidazole – a decomposition product of BmimCl –

Figure 4: Thermogravimetric curves (1st derivative: dw/dt , solid lines) and Gram-Schmidt curves from evolved gas analysis (dashed lines).

Figure 5: Intensity of the ionic-liquid peak from the TGA analysis as a function of the depth below the wood surface: ionic liquid-treated vs. ionic liquid-treated and densified. The right figure shows the effective penetration after virtually expanding the densified wood to its initial thickness before densification. The colours (red > orange > yellow > grey) indicate the intensity of the IL peak (first peak) in relation to the wood peak (second peak).

as a function of the depth beneath the wood surface in undensified and densified wood. Figure 8 compares the penetration depth of the IL between BmimCl_W and BmimCl_D_W. Graphs of the control group C_W were omitted, as they did not show any sign of the IL. In the undensified specimens, the IL concentration rapidly decreased, and from a depth of 1.5 mm onward the IL was virtually absent. In the densified specimens the IL penetrated further into the material, with IL concentration decreasing much less dramatically, and disappearing only at approximately 2.5–3.0 mm.

For both BmimCl_W and BmimCl_D_W low concentrations were visible throughout the entire measurement depth. This was most likely due to contamination between the layers during specimen preparation, as indicated by the orange control curves in Figures 6 and 7. However, this effect was negligible in relation to the general picture observed when comparing the two specimen groups.

For the densified specimens, the penetration depth of the IL matched very well between the TGA and GC-MSD

Figure 6: GC-MSD analysis showing concentrations of 1-butylimidazole in BmimCl_W as a function of depth beneath the densified wood surface.

Figure 7: GC-MSD analysis showing concentrations of 1-butylimidazole in BmimCl_D_W as a function of depth beneath the densified wood surface.

Figure 8: Concentration peak-values of 1-butylimidazole in BmimCl_W vs. BmimCl_D_W, as a function of depth beneath the densified wood surface.

analysis; however, for the undensified specimens it appeared that the TGA overestimated the depth of penetration. In fact, the results from the GC-MSD were in line with the initial observation of only marginal penetration when a dye was added to the IL solution. Currently, the reasons for this discrepancy are unclear. Nonetheless, the findings from the GC-MSD analysis again support the hypothesis of *in-situ* penetration.

3.3 Digital image correlation

The contour plot in Figure 9 shows full-field data of the strain in the radial direction (ϵ_y), i.e. the set-recovery determined with the DIC analysis for the group BmimCl_D_W. The set-recovery was virtually zero in the region just beneath the wood surface. From a depth of approximately 1–1.5 mm, the set-recovery gradually increased and reached its peak at a depth of 2–2.5 mm, and then gradually decreased again towards the undensified wood. This finding aligned almost perfectly with the GC-MSD analysis, which showed a high concentration of IL to a depth of about 1.5 mm, after which the concentration decreased more rapidly until almost disappearing at a depth of 2.5 mm (Figures 6–8).

Figure 9: Digital image correlation (DIC) data of the strain in radial direction (ϵ_y) of surface-densified wood that was pre-treated with an ionic liquid (BmimCl_D_W).

The set-recovery in the transition zone between a depth of 1.5 and 5 mm occurred almost exclusively in the earlywood. This was not surprising, as the earlywood is weaker than the latewood and therefore more easily densified, and in general the set-recovery increases with an increase in compression ratio (Navi and Sandberg 2012). The TGA and GC-MSD analysis did not provide data about differences in the level of IL concentration in earlywood versus latewood.

The correlation of the data from the DIC, TGA and GC-MSD analyses is summarised in Figure 10. There are two locations of interest with regard to the hypothesis of this study: the drop-off point where the IL concentration starts to decrease substantially, and the location of the greatest set-recovery determined by the DIC. The IL drop-off point coincides with the depth at which the set-recovery starts to increase, and the set-recovery reaches its maximum at roughly the same depth at which the IL peak disappears completely. An explanation for this behaviour is that this transition zone had a sufficient concentration of the IL to enable the densification of the weak earlywood, but the temperature in this region was likely too low to cause

Figure 10: The local strain determined by the digital image correlation (DIC) (purple dots and the purple trend curve) and the concentration of the ionic liquid measured with thermo-gravimetric analysis (TGA) (orange curve) and gas-chromatography mass-selective-detection (GC-MSD) (green curve), as a function of the distance from the IL-treated and densified surface.

the decomposition of the ionic liquid, thereby preventing the cellulose chains from forming new hydrogen bonds. Due to its thicker cell walls, the latewood has a higher absolute amount of cellulose than the earlywood, and as a result the ionic liquid concentration was perhaps too low to sufficiently plasticise those regions to allow for densification.

This study and the preceeding one reported by Neyses et al. (2020) have important implications on the commercial potential of the IL pre-treatment in the form it is presented here: firstly, a high densification temperature was required to increase the penetration depth of the IL solution into the wood surface, and secondly, the IL decomposed and was thereby consumed during the densification process. In order to improve upon these drawbacks, further studies will have to explore the possibility of diluting the IL in water to support penetration by swelling the cell walls, and of recovering the IL for reuse before the densification stage. Studies by Parviainen (2016) and Tanaka et al. (2018) on IL-treated paper have indicated the viability of both ideas.

4 Conclusions

In this study, a novel multi-sensor data fusion approach – combining chemical analyses with digital image correlation (DIC) – was applied to understand a previously unexplained penetration phenomenon, which can occur when wood is surface-treated with ionic liquids (ILs) and then densified. This unique combination of methods made it possible to track the concentration of the IL in the wood as a function of the depth beneath the surface at various processing stages, and correlate these findings with high-spatial resolution set-recovery measurements on a sub-annual ring level.

With the thermo-gravimetric analysis (TGA) it was possible to determine the penetration depth of the IL into the wood before and after the densification process. The data supported the hypothesis that a substantial portion of the penetration occurs during the densification stage, and this finding was confirmed by the gas-chromatography mass-selective-detection (GC-MSD) analysis. The full-field DIC data strengthened these findings, as the set-recovery at different depths beneath the specimen surface matched the concentrations of the IL in the wood as determined with TGA and GC-MSD. In the region just beneath the surface with a high concentration of the IL, the set-recovery was almost completely eliminated.

DIC proved to be a powerful tool to study the surface densification process and its effects on the densified wood on a scale that fills the gap between ‘macro-measurements’,

such as the set-recovery or Brinell hardness, and ‘micro-measurements’ on the cellular level. In the present study, DIC was used to compare two static stages (densified wood before and after set-recovery), but with a more sophisticated setup it may be possible to track the behaviour throughout the complete densification process.

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