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Surface modification of Scots pine: the effect of process parameters on the through thickness density profile

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Abstract This study evaluated the significance of different process parameters (press temperature, closing time, holding time, moisture content and compression ratio) on solid wood surface densification and its effect on the density profile generated in Scots pine sapwood. Changes in the microstructure of the wood were also evaluated microscopically. The results showed that with a shorter closing time, densification occurred closer to the sample surface than with an extended closing time. At a compression temperature of 150 °C, the vertical density profile exhibited a sharp peak in density that was close to the wood surface. A higher temperature of 200 °C resulted in a slightly broader density peak that was less intense and further from the surface. A holding time of 10 min resulted in the wood compressing to a slightly greater extent than when using a holding time of 1 min. Higher moisture content led to more extensive deformation. The results indicate that surface modification by densification is a viable method of enhancing wood properties.

Introduction

It is well known that the density of wood correlates with its mechanical properties and, moreover, that as a porous material it can be compressed until its density reaches that of the cell wall material, which has been determined to be around 1.50 g/cm^3 [1, 2]. When heated, wood softens, with the lignin, hemicellulose and cellulose displaying different softening behaviours depending upon the temperature and moisture content [3–5]. Thus, when above its glass transition temperature, wood can be compressed without rupturing the cell walls.

Compressing wood to increase density is a well-studied method of improving its properties. Most of these studies have, however, focused on the densification of bulk wood, and several methods have been investigated. Once such approach is to compress the material in a pressure vessel under saturated steam [6–9]. Alternatively, wood may be compressed when it is water saturated [5, 10]. Bulk modification by these means is quite a long and expensive process due to the need for sophisticated pressure vessels and the fact that the wood is modified throughout. In addition to the extended process times, another negative effect of compressing bulk wood is the loss in wood volume. This may be a drawback in situations where, for example, the main aim is to improve surface hardness for say flooring applications, but where it is beneficial to retain thickness to resist bending. In such situations, an alternative approach is to densify the surface only. This targeted modification can be achieved in a hot press by carefully controlling the pressing conditions and the moisture content of the wood.

In surface densification, the objective is to compress, and thus modify, only the first few millimetres beneath the wood surface; effectively just the first few cell layers. The process starts with relatively dry wood which is heated on one surface only and is then compressed. As a result, the densification process is relatively fast, taking only a few minutes or even seconds [11–15]. It has been found that surface densification can be effected in a continuous press [16]. In this approach, a continuous press has a water

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cooling system to allow quick solidification of the wood on the compressed surface. It has been demonstrated that surface densification can alter the mechanical properties of wood as well as the surface wetting behaviour [11, 14].

It is well known that various process parameters influence the vertical density profile (VDP) created in wood-based panels such as medium density fibre board (MDF) and oriented strand board (OSB) during manufacture. In wood-based panel production, the VDP contributes greatly to the final product properties and as such has been widely studied [17–20]. As a result, VDP becomes controllable though understanding the relationship between process parameters and density profile. Similarly, in order to fully optimize the mechanical properties of surface compressed solid wood, it is important to understand the impact of the process parameters on the density through the thickness of the modified wood. At present, the relationship between process parameters and the development of the density profile in solid wood, surface densified by compression, is not clear and has not been widely studied.

The density profile developed in compressed solid wood largely depends on factors such as the initial moisture content of the wood, wood grain orientation, wood species, and also on the press temperature and closing time [21, 22]. For example, the higher the densification temperature, the higher the degree of compression that is achieved [23, 24].

The objective of this study was to investigate the surface densification of Scots pine sapwood by compression, using equipment that allows a continuous pressing process to be simulated, and to evaluate the significance of different process parameters on the density profile generated in the compressed wood. The study investigated the effects of pressing temperature, moisture content, compressing time, and compression rate on the density profile created. The density profile reveals the density throughout the thickness of the sample. This knowledge is extremely important to obtain the optimal properties for surface densified wood. It is well known that compressed wood partially recovers its initial dimensions under wet or humid conditions and that this so-called set-recovery can be eliminated by, for example, heat-treatment. This study shows only the densification process and its effect on the density profile created. To eliminate set-recovery, some form of post-treatment would be required.

Materials and methods

The wood material used in this study was the clear sapwood of kiln dried Scots pine (*Pinus sylvestris* L.) obtained from Southeast Finland, having an average density (RH 65%, 20 °C) of 0.53 g/cm^3. Specimens were machined to dimensions of 140 mm (longitudinal), 50 mm (tangential) and either 20 or 16 mm radially (so as to obtain the desired final compression). Before densification, the specimens (480 in total) were separated into three batches ($n = 160$) and each batch conditioned at relative humidities of either 35, 65 or 75% and a temperature of 20 °C, for at least 2 months, to attain different moisture contents (see Table 1).

Table 1 Summary of the process parameters used in the study

Moisture content (%)	Compression ratio (%)	Temperature (°C)	Closing time (min)	Holding time (min)
9.6	25	150	0.5	1
12.4	6	200	5	10
15.6				

Surface densification was carried out using a specially designed heated press tool fitted to a Zwick 1475 testing machine combined with an MTS Premium Elite controller (see Fig. 1). The surface of the wood was softened by an electrically heated plate incorporated into the press tool. The tool was also equipped with a water cooling system to enable the heated surface of the tool (and therefore wood) to be cooled from 200 °C to below 100 °C within approximately 30 s. A PT-100 sensor below the stainless steel surface of the heated plate monitored the plate temperature. Mechanical stops ensured that the specimens would be compressed to the same final thickness (15 mm) from the original thickness (either 16 or 20 mm). These resulted in compression ratios of 6 or 25%.

Whilst awaiting pressing, batches of 10 specimens were stored in plastic bags after removal from the conditioning rooms so as to minimise any moisture loss before pressing. Specimens were removed one-at-a-time from the bags, placed immediately on the heated platen, the upper (unheated) platen lowered to just above the wood surface and the test commenced. In all, less than a minute elapsed from placing the specimen on the heated surface to be

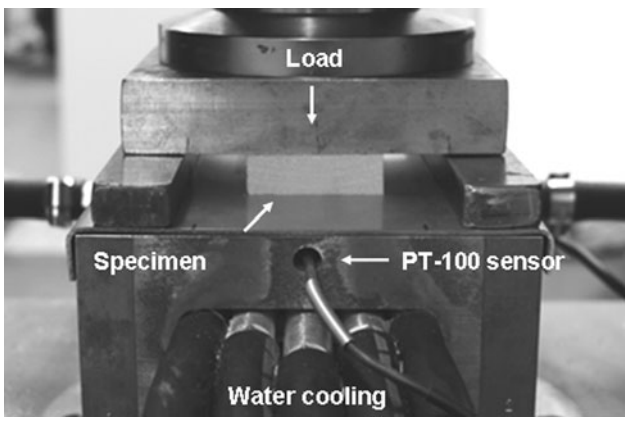

Fig. 1 Compression set-up

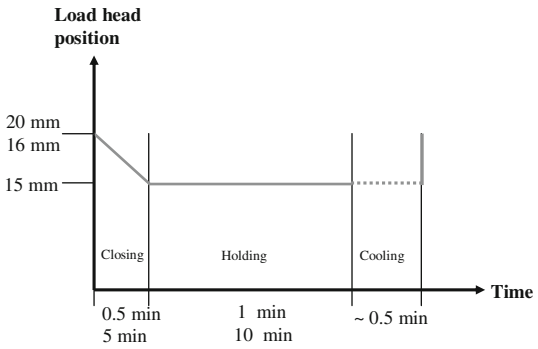

Fig. 2 The schematic illustration of the densification process

commencement of compression. Undoubtedly, some drying of the wood surface would have occurred during this time. The press was set to close in a specified time (closing time). Once the unheated loading plate had contacted with the stops, the press remained closed with the heat being applied for a specified holding time. After the desired holding period, the cooling system was turned on and the load maintained until the recorded temperature was below 100 °C (taking around 30 s) after which the tool was opened and the compressed specimen removed. A schematic illustration of the densification process is presented in Fig. 2. The closing time and holding time were controlled by the MTS controller. The process parameters used in the study are summarised in Table 1.

Density profile

After compression, the samples were conditioned (RH 65%, 20 °C) for at least 1 month. Samples for VDP measurements were machined from the surface densified materials into specimens 50 mm (longitudinal) by 50 mm (tangential) (thickness remained constant). Density was measured at intervals of 0.1 mm through the thickness of the specimens, using an ATR Density Profilometer DPM201 (1995) which operates using a gamma ray source. The density profile of ten specimens, from each of the process parameters investigated, was measured. The samples were placed in the profilometer in a manner that started the scanning from the un-densified surface and proceeded towards the densified surface.

Microscopy

The transverse surfaces of five different densified wood samples were examined by optical microscopy. Dry conditioned (RH 65%, 20 °C) specimens, approximately 10*10*10 mm^3, were first prepared using a table saw and the surfaces to be investigated abraded with fine sandpaper (grit size 1200). Microscopic analysis was carried out using both a Leica WILD MZ8 (equipped with Leica CLS150 light source) reflectance microscope and a Nikon OPTIPHOT-2 reflectance microscope. Both were equipped with a JVC 3-CCD camera.

Results and discussion

The purpose of this study was to investigate surface densification of Scots pine sapwood by compression and, more specifically, to examine the effect of different process parameters on the development of the density profile. The different process parameters investigated were closing time, holding time and the temperature at which the samples were compressed. The effect of the initial moisture content of the samples and the compression ratio used on the development of the density profile were also studied.

Wood softening behaviour is dependent upon temperature and moisture content; wood softening occurs at lower temperatures when moisture content increases because the glass transition temperature is highly dependent on moisture content [3–5]. Moreover, the role that the compression time plays is also important because the longer the surface is in contact with the heated plate the greater the heat transfer (and so potential for surface softening), but at the same time the wood dries out. The net result is that the reasons for the density profiles generated are not always obvious, since several parameters affect the process simultaneously. Some examples of the VDPs generated by different parameters are presented in Figs. 3, 4, 5, 6 and 7.

Compression ratio

In this study, Scots pine sapwood samples of two different thicknesses, 20 and 16 mm, were compressed to 15 mm thickness. The theoretical compression ratios of the two

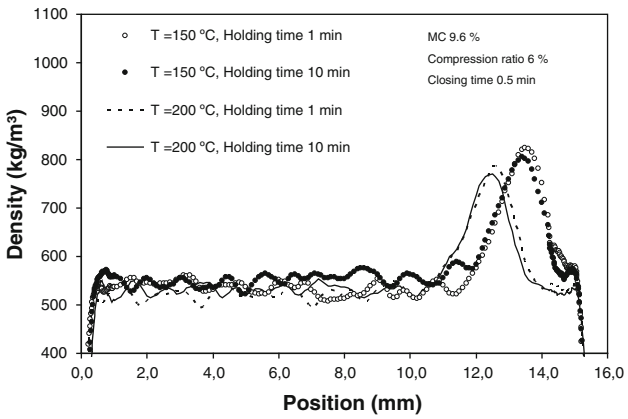

Fig. 3 Surface densified solid Scots pine board. The effect of platen temperature on the development of density profile, average of ten specimens

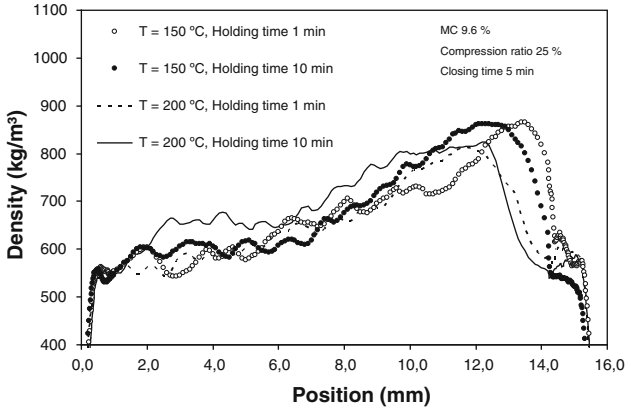

Fig. 4 Surface densified solid Scots pine board. In particularly: broad density peak developed, average of ten specimens

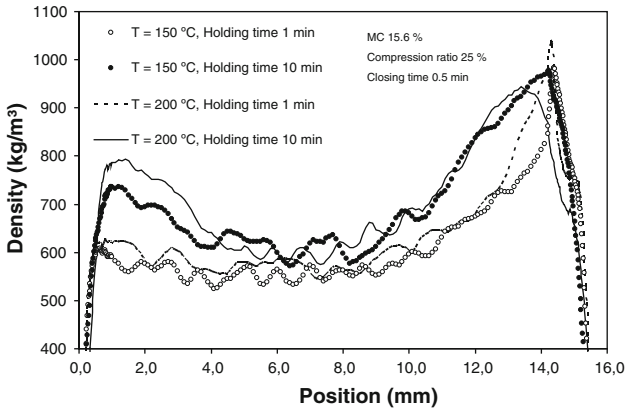

Fig. 5 Surface densified solid Scots pine board. In particularly: the effect of long holding time, average of ten specimens

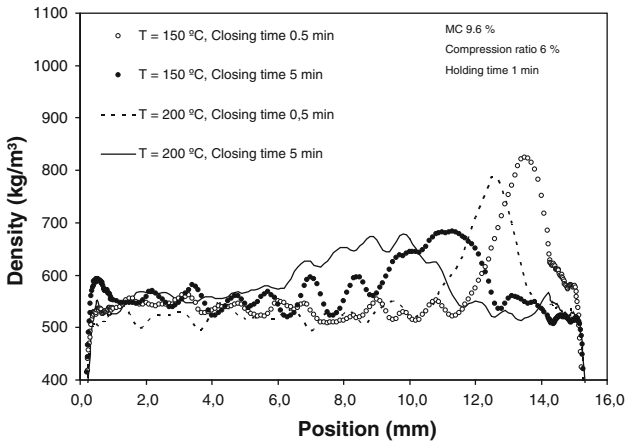

Fig. 6 Surface densified solid Scots pine board. In particularly: the effect of long closing time, average of ten specimens

different thicknesses were thus 25 and 6%, respectively. In practice, however, difficulties in controlling the initial thickness of the samples meant that the compression ratios

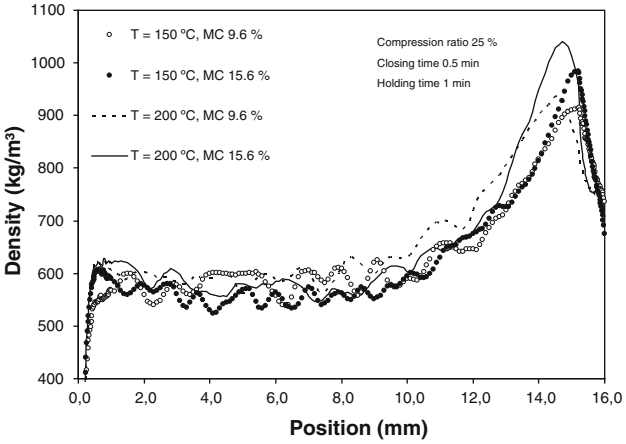

Fig. 7 Surface densified solid Scots pine board. In particularly: the effect of different moisture contents, average of ten specimens

varied slightly. This was particularly so with the 16 mm specimens, where small differences in the initial thickness resulted in a considerable change in the compression ratio.

As is clearly shown in Figs 4, 6 and 7, which show the through thickness density profiles of material conditioned to the same MC (9.6%), but compressed to different degrees (6 and 25%), not only is there a distinct increase in the overall density but, more significantly, the form of the density profiles is altered. In the case of the lower compression ratio, a sharp peak in density is seen close to one surface of the wood, being around 45% higher than the average wood density. Although the compression ratios were different, the closure times were constant (either 0.5 or 5 min), resulting in different strain rates during compression. This would most probably have led to a different response in the wood and consequently have affected the density profile generated. Moreover, the results shown in Figs. 3, 4, 5, 6 and 7 indicate that the other process parameters also have a significant influence on where the greatest density occurs.

Moisture content

The samples were conditioned to equilibrium moisture contents of 9.6, 12.4 and 15.6%. As is known, wood softens to a greater extent at a given temperature as the moisture content increases [5, 24, 25]. In general, it was found that the lowest density near the surface was obtained with the lowest moisture content, whereas the highest density resulted from the highest moisture content (Fig. 7). This phenomenon might be related to the heat transfer properties and surface drying during compression. It has been found that in MDF production higher moisture content leads to greater heat transfer towards the core of the board during hot pressing; this phenomenon most probably also occurs in solid wood compression. Furthermore, the higher density

of the MDF board caused slower heat transfer to the core [20].

Temperature

The platen temperatures used were 150 and 200 °C. As may be seen in Figs. 3, 4, 5, 6 and 7, at the lower temperature the peak of the density curve is closer to the surface of the sample; with the higher temperature, the peak density shifted further away from the surface and deeper into the specimen. Drying out of the wood surface whilst awaiting the commencement of compression might well account for the lower density right at the surface and the shift of the peak in density towards the central portion of the specimen at the higher platen temperature. This is seen particularly in Figs 3, 6 and 7. Wood softens to a greater extent at higher temperatures and thus using the lower temperature it seems probable that the wood below the surface neither heats up as quickly, nor reaches as high a temperature as the wood heated at the higher temperature and therefore this may create more resistance to compression deformation, confining the density increase to near the wood surface. Higher temperature increases heat transfer which presumably is the reason for this phenomenon. In addition, the higher temperature might dry the surface more, resulting in the very surface not deforming as much (Figs. 3, 4 and 6).

Closing time

Two different closing times were used, 0.5 and 5 min. In this study, closing time was found to exhibit the greatest influence on the formation of the density profiles. The results (Figs. 3, 4, 5 and 6) show that a closing time of 5 min resulted in a broader peak in the density profile than with a closing time of 0.5 min and that the peak shifted towards the centre of the specimen (see particularly Fig. 6). With the short closing time, the peak in density was sharper and near to the surface. These findings concur with those of previous studies on wood compression [21]. The shorter closing time most probably results in limited heat transfer, and thus softening of the wood is also limited and therefore only the surface is compressed to any great extent. Moreover, the strain rate is far greater, thus reducing the tendency for stress relaxation to occur. With the longer closure time, not only would there be greater heat transfer, but there would be a greater opportunity for the wood below the surface to deform viscoelastically, thereby broadening the density peak. In addition, with the longer closing time the surface of the wood will probably dry more whilst the inner parts of the wood have higher moisture content and thus soften more readily.

Holding time

Holding time during the compression process was either 1 or 10 min. The longer holding time resulted in the samples exhibiting a density peak slightly further from the surface than those with shorter holding time (Figs. 3, 4, 5, 6 and 7). That is to say, the samples with longer holding time exhibited a density increase over a wider portion of the sample. This was expected because the longer holding time would allow more heat transfer and thus deeper softening of the wood. Furthermore, the results show with the higher densification ratio, higher density values on the opposite surface of the wood. The most likely explanation is for this is that since the wood was compressed between two metal plates, and between them were metal stoppers, with the longer holding time the heat would transfer from the heated surface to the other surface and thus the sample may tend to soften from both surfaces.

Microscopic analysis

Microscopic examination of the cross sections of a selection of the samples was carried out to examine the extent and character of deformation in the wood as well as to see how well the density profiles correlated with the observed deformations.

Figure 8 shows the structure of same specimen at different depths, (a) from core and (c) from surface. It can be clearly seen that the earlywood deforms whilst the latewood seems to hold its form. Figure 8a shows a section of earlywood which has not compressed at all, whereas in Fig. 8b it is slightly compressed and in Fig. 8b it is highly compressed. The micrographs were also merged with the density profiles for selected samples (Figs. 9 and 10). As may be seen there is a clear correlation between the density profile and the sample microstructure. Figure 9 shows what is considered to be a successful densification, in which only the surface region has been densified.

Figure 9 further reveals that the very surface of the sample seems to have a lower density than some millimetres deeper. This phenomenon might be due to several factors. First, the spring-back effect might be present at the very surface. Second, the heated metal plate might dry the first few layers of the specimen whilst closing and thus diminish the level of deformation. Third, the very surface might have some other micro structural bias resulting from sawing, conditioning or other factors. A higher magnification micrograph (Fig. 11) of the same specimen shown in Fig. 9 exemplifies this phenomenon. Figure 10 presents an example of the density profile , which is considered to be an unsuccessful compression, because the densification has occurred in the inner part of the specimen. This type of profile was typical of longer closing times and lower compression ratios.

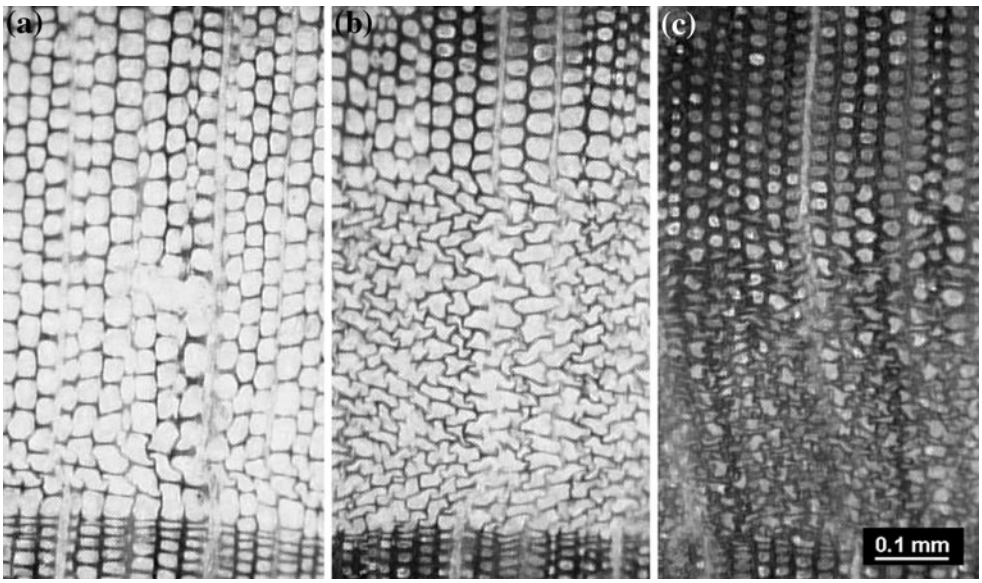

Fig. 8 Magnified images from different depths of the same compressed pine sapwood specimen. **a**: uncompressed, **b**: slightly compressed and **c**: highly compressed

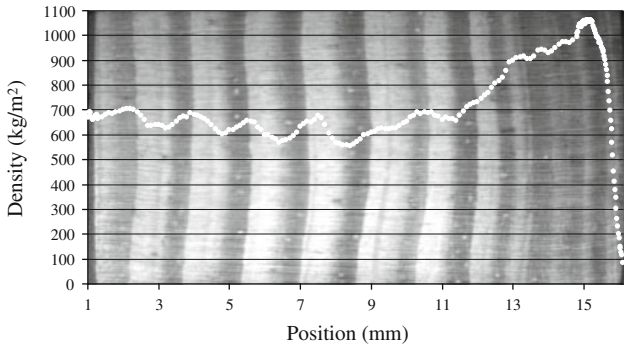

Fig. 9 Density profile curve and magnification of compressed pine sapwood (moisture content 12.4%, 150 °C, compressing time 0.5 + 10 min holding time, compressing ratio 25%)

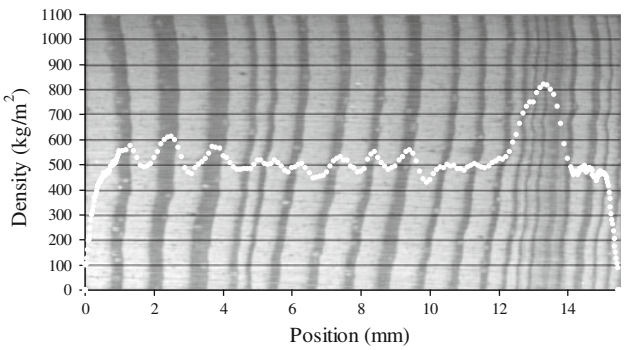

Fig. 10 Density profile curve superimposed on a micrograph of compressed pine sapwood (moisture content 12.4%, 200 °C, compressing time 0.5 + 1 min, compressing ratio 6%)

In Fig. 11, the very surface of the sample is left uncompressed. The first few uncompressed cell layers are latewood, and after that a few layers of earlywood, before

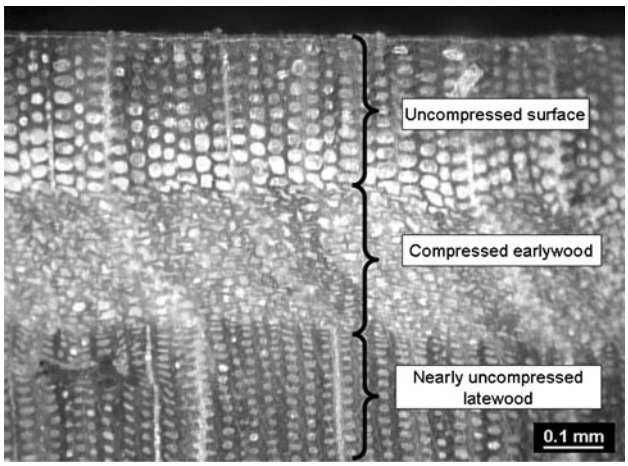

Fig. 11 Magnification of the surface of a compressed sample (moisture content 12.4%, 150 °C, compressing time 0.5 + 10 min, compressing ratio 25%) presenting the uncompressed section on the surface

the deformation begins in the earlywood section. This phenomenon was not further examined. However, according to the density profiles of several samples, it seems that the very surface has a slightly lower density. Another factor to be taken into consideration is the cupping effect of the samples after compression that is related to the moisture content difference. This most likely affects the density profiles in the same manner. The profilometer measures the density of the sample layer by layer in the tangential direction and if the sample surface is not even or the sample is cupped, the average density of the surface will seem lower than in reality.

Conclusion

The purpose of this study was to investigate surface densification of solid pine sapwood by compression and the effect of the process parameters on the VDP. Different process parameters were obtained by changing the closing time, holding time and compressing temperature. Different moisture contents of the samples were used as well as different compression ratios. Closing time exhibited an influence on the form of the density profile. A shorter closing time of 0.5 min resulted in more successful profiles where the deformation was close to the sample surface. The longer closing time of 5 min resulted unsuccessful density profiles in which the deformation mainly took place in the inner layers of the samples, but other parameters interact also. The longer holding time of 10 min resulted in the wood compressing slightly deeper than when using the shorter holding time of 1 min. The density profiles of samples compressed with the shorter closing time also exhibited a clearer peak slightly closer to the surface. Temperature also had a significant effect on the density profiles. When the compressing temperature was 150 °C, the density profile exhibited a higher peak closer to the surface. Whereas the higher temperature of 200 °C presented a slightly broader density peak that was lower in magnitude and further from the surface. The moisture content also had a significant effect on the density profiles, with the higher moisture content leading to deeper deformation. Overall, this study has shown that process parameters play a significant role in the wood surface densification process, but it has to be remembered that all these parameters are interacting each other.

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