J Wood Sci (2004) 50:337–342
DOI 10.1007/s10086-003-0570-6

ORIGINAL ARTICLE

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Compressive deformation of wood impregnated with low molecular weight phenol formaldehyde (PF) resin I: effects of pressing pressure and pressure holding

Received: April 25, 2003 / Accepted: August 1, 2003

Abstract The deformation behavior of low molecular weight phenol formaldehyde (PF) resin-impregnated wood under compression in the radial direction was investigated for obtaining high-strength wood at low pressing pressures. Flat-sawn grain Japanese cedar (*Cryptomeria japonica*) blocks with a density of 0.34 g/cm^3 were treated with aqueous solution of 20% low molecular weight PF resin resulting in weight gain of 60.8%. Oven-dried specimens were compressed using hot plates fixed to a testing machine. The temperature was 150°C and the pressing speed was 5 mm/min. The impregnation of PF resin caused significant softening of the cell walls resulting in collapse at low pressures. The cell wall collapse was strain-dependent and occurred at a strain of 0.05–0.06 mm/mm regardless of whether the wood was treated with PF resin. Thus, pressure holding causing creep deformation of the cell walls was also effective in initiating cell wall collapse at low pressure. Utilizing a combination of low molecular weight PF resin impregnation and pressure holding at 2 MPa resulted in a density increase of PF resin-treated wood from 0.45 to 1.1 g/cm^3. At the same time, the Young's modulus and bending strength increased from 10 GPa to 22 GPa and 80 MPa to 250 MPa, respectively. It can be concluded that effective utilization of the collapse region of the cell wall is a desirable method for obtaining high-strength PF resin-impregnated wood at low pressing pressures.

Key words Low molecular weight PF resin · Pressure holding · Densification · Cell wall collapse

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Introduction

It is desirable in the twenty-first century to build a sustainable society based on renewable and sustainable resources. In this sense, wood may be considered as a future-oriented material even though it is one of the oldest and most common materials. However, dimensional instability due to moisture, low durability caused by biodeterioration, and relatively low mechanical properties of wood and reconstituted wood compared with other engineering materials may prevent wood from replacing materials that are based on unrenewable resources.

To overcome these drawbacks, phenol formaldehyde (PF) resin impregnation and compression seems to be a promising technique. When wood is treated with PF resin and compressed at pressing pressures of 7 MPa or above, its density reaches 1.2 to 1.35 g/cm^3. This product has been given the generic name Compreg and shows high mechanical properties, dimensional stability, and decay resistance.[1–4] Because of such properties, much research following on from the work of Stamm's has been performed, and, in recent years, the potential strength of PF resin-impregnated compressed wood has been extensively studied.[5–8] For example, when wood that was selected based on sound velocity was impregnated with low molecular weight PF resin and hot pressed at a pressing pressure of 50–80 MPa, the bending strength reached 520–530 MPa. Furthermore, when lignin and hemicellulose were removed from the wood and the specimens were impregnated with low molecular weight PF resin and hot pressed at high pressure, the Young's modulus and bending strength attained 62 GPa and 670 MPa, respectively. On the other hand, when PF resin-impregnated wood elements such as particles and powder were hot pressed under high pressure, a decrease in element size did not result in a decrease in bending strength, because high interactive forces developed between elements.[9]

The aforementioned research indicates that PF resin-impregnated compressed wood has the potential to be a substitute for other engineering materials. However, considering the production of large scale composites, the high pressing pressure required during manufacturing becomes a problem. A pressing technique to obtain highly densified wood at lower pressing pressure has to be established. Although many studies on the compressive deformation of wood as a cellular material have been done,[10–13] none has clarified the deforming behavior of low molecular weight PF resin-treated wood where PF resin acts as a plasticizer at first and then loses these effects by polymerizing or curing during hot-pressing. Therefore, in this study, compressive deformation of PF resin-impregnated wood was analyzed. Then, a method for deforming wood at lower pressing pressure was studied.

Materials and methods

Raw material

The raw material was Japanese cedar (*Cryptomeria japonica)* with an air-dried density of 0.34 g/cm^3. Flat-sawn grain specimens with dimensions of 60 mm (longitudinal, L) by 40 mm (tangential, T), and 6 mm (radial, R) were cut in a series from the sapwood portion of a block. Average annual rings width was 1.7 mm.

Treatment with phenol formaldehyde resin

Oven-dried specimens were soaked in an aqueous solution of 20% low molecular weight PF resin with an average molecular weight of about 300 (PL 2771, Gun-ei Chemical, pH 5.5, gelation time at 150°C is 10 min). The specimens were kept under vacuum for 12 h, after which release pressure and the specimens were kept at room temperature for 12 h. This process was repeated seven times in order to obtain complete penetration of the solution into the wood. After air-drying for 3 days, the treated specimens were vacuum-dried at 50°C for 12 h to remove any residual moisture (oven-dried). The weight gain determined using the oven-dried weight before and after treatment was 60.8%.

Measurement of deformation of wood

Two oven-dried specimens were compressed in the radial direction at various pressure levels using hot plates fixed to the Instron Universal Testing Machine (UTM, model 5500) (Fig. 1). This machine allows precise movement of the crosshead thus providing data on how wood can be compressed. The crosshead movement, load, and accumulated time were logged using a computer attached to the UTM. After the removal of the deformation of hot plates and their attachments, the deformation of wood in the thickness direction was evaluated.

The pressing speed was 5 mm/min and the pressing temperature was 150°C. After reaching the desired pressing pressure, the crosshead movement was stopped and the condition was kept for 30 min or the pressing pressure held for 30 min (referred as pressure holding). The relationship between pressing pressure and density was determined based on stress–strain curves.

Fig. 1. Hot plates attached to a Universal Testing Machine to evaluate the deforming behavior of wood. A cooling system was used to protect the load cell

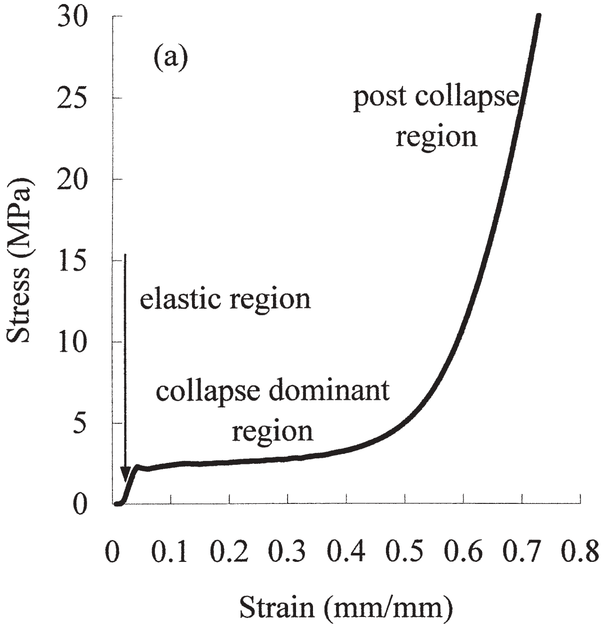
Evaluation of bending properties

Two specimens with dimensions of 50 mm (L) × 8 mm (T) were cut from each compressed wood and oven-dried for 6 h at 105°C before measurement of the oven-dried weight and final dimensions. After rounding the cut edges along the length of the specimens with sandpaper (grit #300) to eliminate the micro cracks brought about during cutting, the Young's modulus and bending strength in the oven-dried condition were evaluated by a three-point load bending test with a span of 40 mm using an Instron 4411 universal testing machine at a crosshead speed of 5 mm/min.

Results and discussion

Effects of pressing pressure

Figure 2a shows the typical stress–strain curve of oven-dried wood when compressed in the radial direction. During compression of wood, three regions are distinguished: elastic region, collapse-dominant region, and post collapse region. Wood deforms in an elastic manner at the beginning of compression. A yield point occurs at the onset of the cell wall collapse. After that wood continues to deform at a nearly constant stress level, which is the so-called collapse dominant region. The characteristic feature of this region is high compression at the lower applied stress. Because the aim of this study was to densify wood at lower pressing



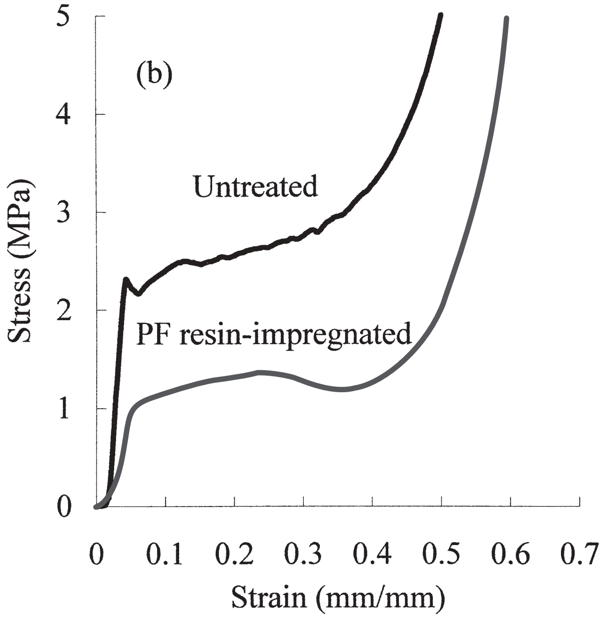


Fig. 2. Typical stress–strain curve of oven-dried wood compressed in the radial direction at a pressing temperature of 150°C and a pressing speed of 5 mm/min (**a**) and comparison with phenol formaldehyde (*PF*) resin-impregnated wood (**b**)

pressures, this region is of importance. When most of the cells have been collapsed, the post collapse region begins. The characteristic feature of this region is the densification of wood, with a rapid increase in stress as the compression continues.

As shown in Fig. 2b, PF resin-impregnated wood with a weight gain of 60.8% shows a behavior similar to that of untreated wood. However, PF resin-impregnated wood requires lower stress to initiate cell wall collapse than untreated wood. For example, to initiate collapse, PF resin-impregnated wood required a pressing pressure of 1 MPa while the untreated wood required a pressure greater than 2 MPa. This indicates that the low molecular weight PF resin in the cell wall acts as a plasticizer and causes significant softening of the cell wall, inducing a reduction of Young's modulus of the cell wall in the cross fiber direction. In addition, Fig. 2b shows that a certain strain is required to initiate collapse, irrespective of PF resin-treated or untreated condition. This result implies that the cell wall collapse in the radial direction is strain-dependent rather than stress dependent.

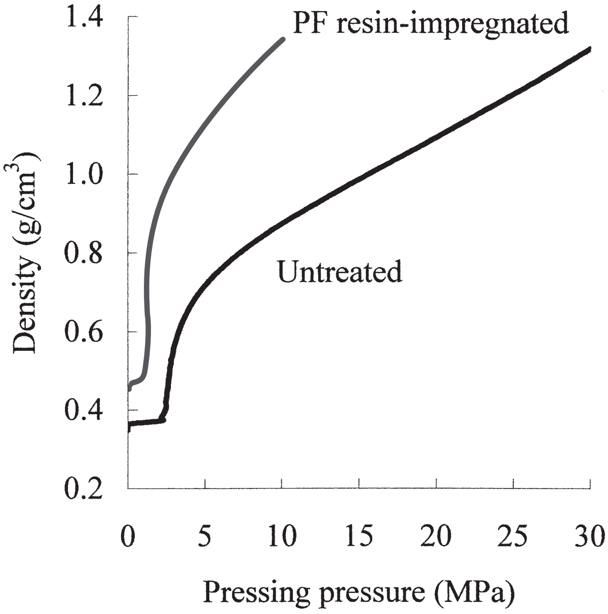


Fig. 3. Effects of low molecular weight PF resin impregnation and compression on the densification of wood

To understand the deforming behavior of wood more clearly, the relationship between pressing pressure and density was evaluated as shown in Fig. 3. Impregnation of PF resin resulted in higher densification, giving a density of 1.35 g/cm^3 at a pressing pressure of 10 MPa. However, untreated wood requires about 30 MPa to attain the same density. Figure 3 also implies that most of the densification occurred in the collapse region. As a consequence, the density of PF resin-impregnated wood increases from 0.47 g/cm^3 to 0.92 g/cm^3 with increasing pressing pressure from 1 to 2 MPa, which shows that the density increases by around two times by the application of slightly increased pressure. On the other hand, untreated wood could not attain an identical density at a pressing pressure of 2 MPa, because the cell wall collapse does not start until a pressure of above 2 MPa is applied.

In general, the mechanical properties of wood increase with increasing density, due to the increment of volume ratio of the cell wall. Figure 4 shows the Young's modulus and bending strength of PF resin-treated and untreated wood compressed at different pressing pressures. After reaching the desired pressing pressure level, the crosshead

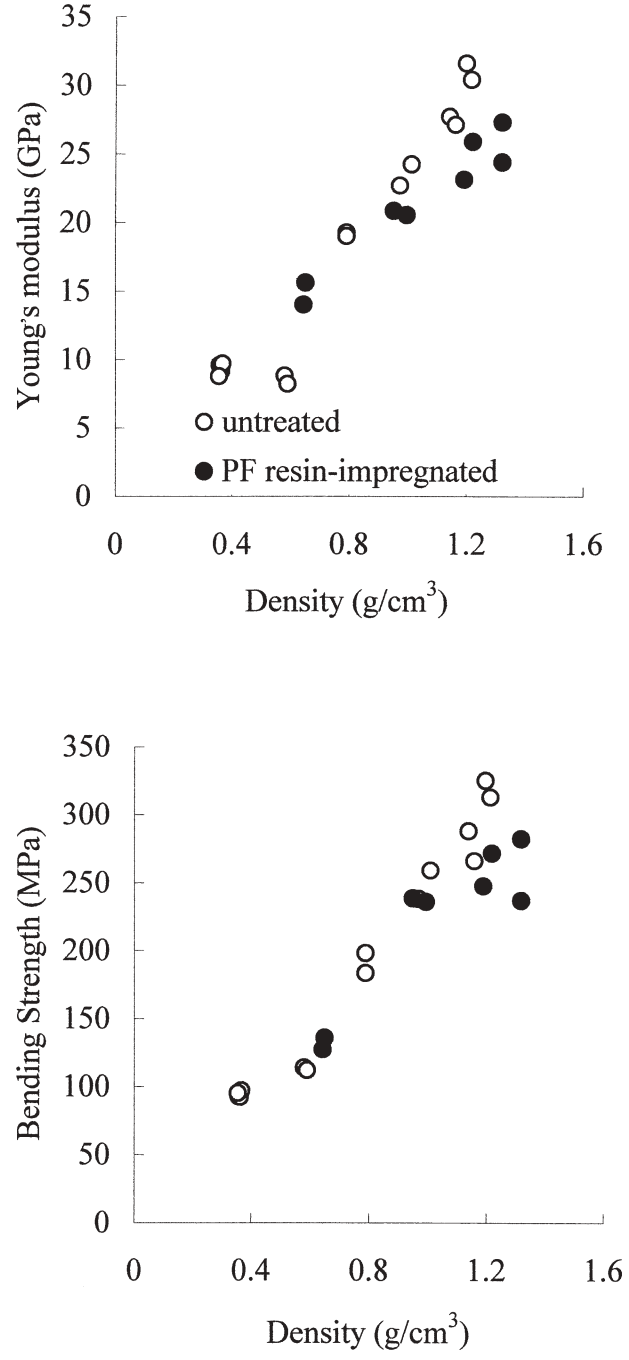


Fig. 4. Effects of low molecular weight PF resin impregnation and compression on the Young's modulus and bending strength of wood. Specimens were compressed at different pressing pressures. After reaching the desired pressure, crosshead movement was stopped and the condition was held for 30 min

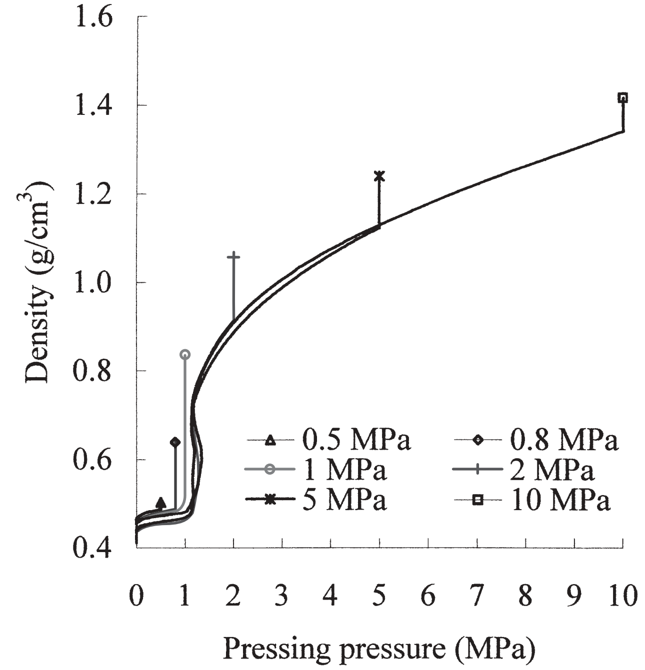


Fig. 5. Effects of pressure and pressure-holding time on the densification of PF resin-impregnated wood. The *bars* indicate a holding time of 30 min

movement was stopped and the condition was held for 30 min. The Young's modulus and bending strength increase linearly with increasing density, regardless of PF resin treatment. Furthermore, it was observed that despite the large increment of pressing pressure, that is 2 to 10 MPa (five times more pressing pressure), the increase in density in the post collapse region of PF resin-treated wood was around 1.0 to 1.35 g/cm^3, whilst the improvement in mechanical properties was small. This means that making use of the collapse region is an effective way, not only for increasing density but also, for increasing mechanical properties of wood.

Effects of pressure holding

Because the occurrence of cell wall collapse was found to be strain dependent, creep deformation of cell walls to initiate collapse at lower pressing pressures was accordingly studied at various pressing pressures ranging from 0.5 to 10 MPa. After reaching the desired pressing pressure, the pressure was held for 30 min as shown in Fig. 5. When PF resin-impregnated wood is compressed at 0.5 MPa, the effect of pressure holding on density can be negligible. However, when wood is compressed at 1 MPa, a significant increment in density occurs during the pressure-holding period. The density increases from 0.52 to 0.84 g/cm^3, which is around 70% higher than the density before pressure holding. At high pressing pressures such as 5 MPa and 10 MPa, the effect of pressure holding on density is not as significant as that at 1 MPa. This result implies that a proper combination of pressing pressure and holding period is needed to obtain marked deformation of wood at lower pressing pressures.

Hence, to understand deformation behavior during pressure holding from the viewpoint of collapse, the stress–strain curve of PF resin-impregnated wood was analyzed as shown in Fig. 6. At a pressing pressure of 0.5 MPa, although wood had already passed the collapse initiating strain of 0.05–0.06 mm/mm, the deformation stopped after a slight increase in strain due to collapse. Contrary to this, a

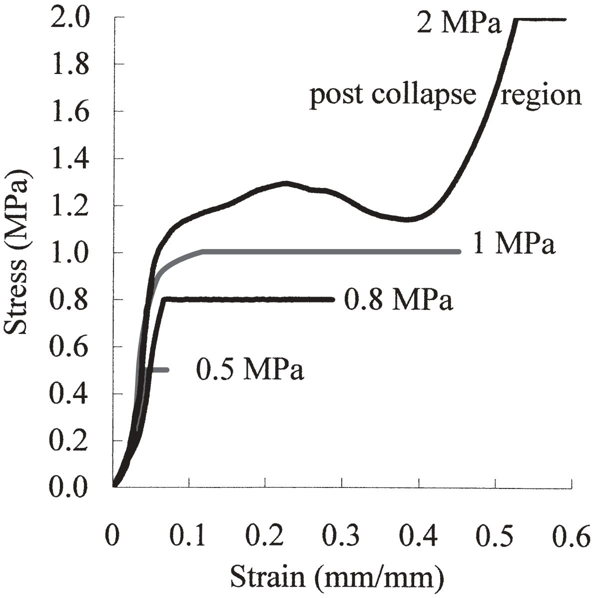


Fig. 6. Changes in strain during 30 min of holding time under different pressures

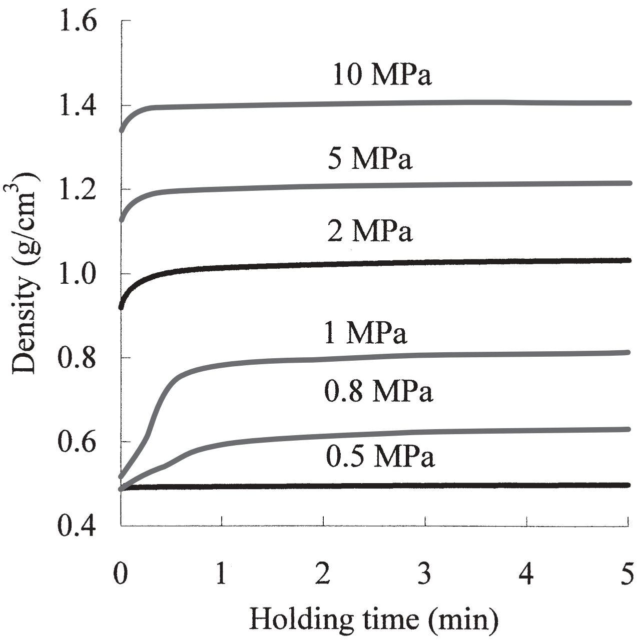


Fig. 7. Effect of duration of pressure holding on the densification of PF resin-impregnated wood

significant strain increment occurs during pressure holding at 1 MPa. The strain of PF resin-impregnated wood is 0.11 mm/mm at 1 MPa before pressure holding, and increases drastically up to 0.45 mm/mm during holding. Comparing the deforming behavior between pressure holding at 1 MPa and at 2 MPa, it can be seen that the deformation of wood compressed at 1 MPa terminates just before the post collapse region. According to the stress–strain curves of the post collapse region, as shown in Fig. 6, the slope of the curve increases with an increase of strain. It shows that the resistance of the cell wall against deformation increases with increasing strain at the post collapse region. This suggests that a certain balance between cell wall resistance and applied pressure exists during creep deformation in a collapse-dominant region as well. However, the curing of PF resin may possibly play a role in this case.

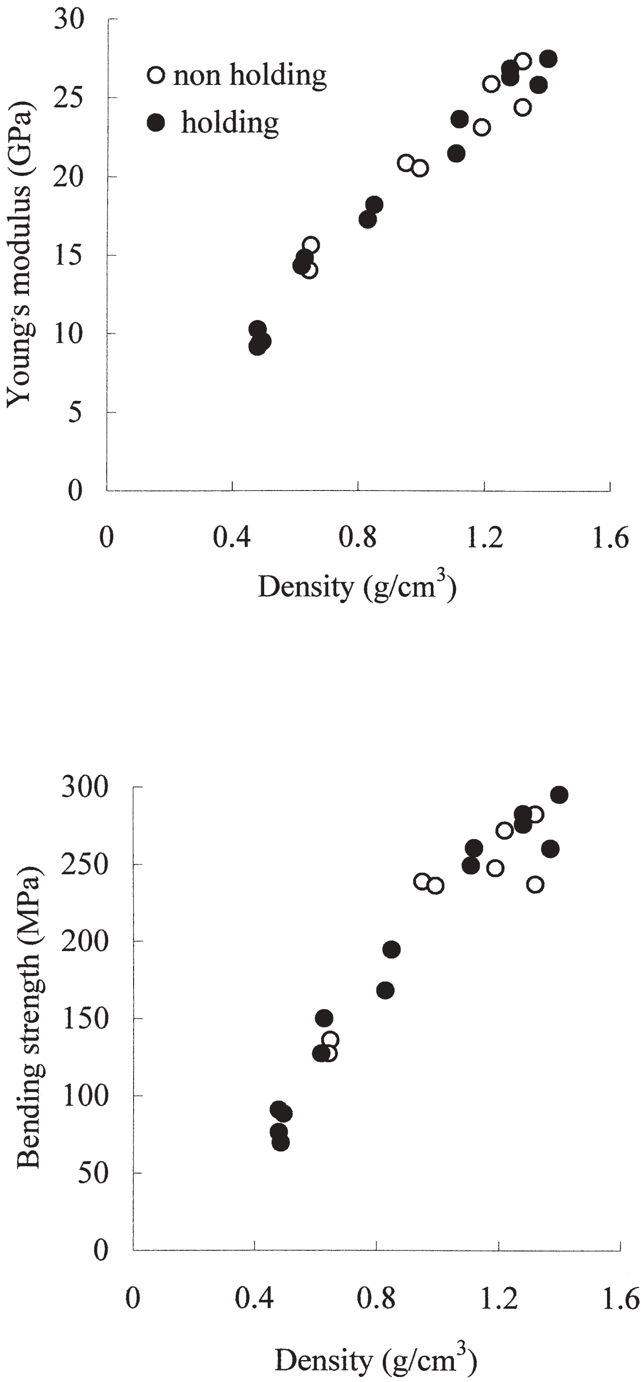


Fig. 8. Relationship between density and Young's modulus and bending strength of PF resin-impregnated wood with pressure holding and nonholding. For pressure holding, after reaching the desired pressure, the pressure was held for 30 min. For nonholding, after reaching the desired pressure, crosshead movement was stopped and the condition was held for 30 min

To clarify the effects of curing on the deforming behavior of PF resin-impregnated wood, changes in density during pressure holding were studied. Figure 7 shows the changes in density within 5 min. It can be seen that within 1 min, the density significantly increases regardless of higher or lower pressing pressures. After a holding time of 1 min, the density was almost consistent. Considering that the gelation time of PF resin used in this study is 10 min at 150°C, the stabilization of density during holding can be attributed to the balance between cell wall resistance and the applied pressing pressure. This suggests that pressure holding of 1 min is enough for the densification of wood at lower pressing pressures as long as the deformed condition is fixed.

The relationships between density and mechanical properties of PF resin-impregnated wood with pressure holding (with creep deformation) and nonholding (without creep deformation) are depicted in Fig. 8. There is no significant difference in mechanical properties between pressure holding and nonholding at the same density. As a consequence, at a pressing pressure of 2 MPa utilizing a combination of pressure holding, the density of Japanese cedar impregnated with low molecular weight PF resin increases from 0.45 to 1.1 g/cm^3. At the same time, the Young's modulus and bending strength increase from 10 GPa and 80 MPa to 22 GPa and 250 MPa, respectively. It can be concluded that effective utilization of collapse is promising for the production of high-strength wood at low pressing pressures.

Conclusions

The effect of low molecular weight PF resin impregnation on the compressive deformation behavior of wood was investigated. The impregnation of PF resin causes significant softening of the cell walls, inducing a reduction of Young's modulus and resulting in collapse at lower pressures. The cell wall collapse is strain dependent rather than stress dependent, which results in the densification of wood at lower pressing pressures. Furthermore, the proper combination of pressure and its holding results in marked deformation of wood at lower pressing pressures. By addition of pressure holding, even at a pressing pressure of 2 MPa, the density of PF resin-impregnated wood increases from 0.45 to 1.1 g/cm^3 and Young's modulus and bending strength increase from 10 GPa and 80 MPa to 22 GPa and 250 MPa, respectively. Thus, the effective utilization of collapse is a promising technique for the production of high-strength PF resin-impregnated wood at low pressing pressures.

Acknowledgments The authors thank Gun-ei Chemical Industry Ltd for supplying low molecular weight phenol formaldehyde resin. Financial assistance from Ministry of Education, Culture, Sports, Science and Technology, Japan, is greatly appreciated.

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