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Compressive deformation of wood impregnated with low molecular weight phenol formaldehyde (PF) resin IV: species dependency

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Abstract Flat-sawn specimens of eight wood species, albizia (*Paraserianthes falkata,* 0.23 g/cm^3), Japanese cedar (*Cryptomeria japonica*, 0.31 g/cm^3), red lauan (*Shorea* sp., 0.36 g/cm^3), European spruce (*Picea abies*, 0.44 g/cm^3), Douglas fir (*Pseudotsuga douglasii,* 0.50 g/cm^3), elm (*Ulmus* sp., 0.51 g/cm^3), Japanese beech (*Fagus crenata*, 0.64 g/cm^3), and Japanese birch (*Betula maximowicziana,* 0.71 g/cm^3), were impregnated with low molecular weight phenol–formaldehyde (PF) resin and their compressive deformations were compared. The volume gain (VG) and weight gain due to 20% resin solution impregnation were different among species. Furthermore, the specific volume gain (VG/specific gravity), indicating the degree of swelling of the cell wall, also varied from 17.7% for European spruce to 26.4% for elm. Oven-dried specimens of each species were compressed using hot plates fixed to an Instron testing machine. The deformation behavior of resin-impregnated wood up to 10 MPa was significantly different among the species. Stress development during cell wall collapse for low density wood was minimal. As a consequence, a significant increment of density occurred up to 2 MPa for low density wood such as albizia and Japanese cedar. When PF resin-impregnated wood was compressed up to 2 MPa and the pressure was kept constant for 30 min, the density of Japanese cedar reached 1.18 g/cm^3, about 30% higher than the density of compressed Japanese birch, which possesses an original density that is 2.5 times higher than that of Japanese cedar. The mechanical properties of resin-impregnated wood, especially low density wood, increased with density. Hence, it is manifested that low density wood species have an advantage as raw materials for obtaining high-strength wood at low pressing pressure.

Key words Species dependency · Compressive deformation · PF resin-impregnated wood · Cell wall collapse · Mechanical properties

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Introduction

Deformation of wood in the transverse direction is an important property for the production of wood composites. Many studies on the compressive deformation of wood as a cellular material have been performed;[1–5] however, no one has yet clarified the compressive deformation behavior of wood impregnated with low molecular weight phenol–formaldehyde (PF) resin where PF resin first acts as a plasticizer and then loses this effect by polymerizing or curing during hot pressing. In previous articles,[6–8] we reported that when Japanese cedar is impregnated with low molecular weight PF resin, the cell walls soften significantly, which results in collapse at lower pressing pressure. We also found that with increased PF resin content, the Young's modulus of the cell wall perpendicular to the fiber direction decreases and the collapse-initiating pressure decreases linearly with the Young's modulus. This result implied that the occurrence of cell wall collapse is strain dependent. Hence, we demonstrated that pressure holding causing creep deformation is more effective at allowing the initiation of collapse at lower pressure. Therefore, it was concluded that the combination of low molecular weight PF resin impregnation and pressure holding is a promising way to obtain highly compressed wood at low pressing pressure.

However, considering the application of this technique, the effects of the raw material or species need to be clarified, because collapse or buckling of the cell wall is a function of cell wall thickness or density. In this study, we investigated how the compressive deformation of resin-impregnated wood differs among species with different densities and different anatomical features.

Materials and methods

Preparation of raw material

Flat-sawn specimens were cut from eight wood species of different densities, including both softwoods: Japanese

Table 1. Original density, weight gain (WG), volume gain (VG), and specific volume gain of different species due to 20% phenol formaldehyde (PF) resin solution impregnation

Species	Density (g/cm^3)[a]	WG (%)	VG (%)	Specific VG (%)
Albizia (*Paraserianthes falkata*)	0.23	48.9	5.2	22.6
Japanese cedar (*Cryptomeria japonica*)	0.31	72.4	7.8	25.2
Red lauan (*Shorea* sp.)	0.36	43.0	8.0	22.2
European spruce (*Picea abies*)	0.44	47.0	7.8	17.7
Douglas fir (*Pseudotsuga douglasii*)	0.50	34.7	8.9	17.8
Elm (*Ulmus* sp.)	0.51	42.3	13.5	26.4
Japanese beech (*Fagus crenata*)	0.64	33.7	12.4	19.4
Japanese birch (*Betula maximowicziana*)	0.71	28.1	16.3	22.9

[a] Density was evaluated based on oven-dried condition for untreated uncompressed specimens

cedar (*Cryptomeria japonica*, 0.31 g/cm^3), European spruce (*Picea abies*, 0.44 g/cm^3), and Douglas fir (*Pseudotsuga douglasii,* 0.50 g/cm^3); and hardwoods: albizia (*Paraserianthes falkata,* 0.23 g/cm^3), red lauan (*Shorea* sp., 0.36 g/cm^3), elm (*Ulmus* sp., 0.51 g/cm^3), Japanese beech (*Fagus crenata*, 0.64 g/cm^3), and Japanese birch (*Betula maximowicziana,* 0.71 g/cm^3). The density given in parentheses, which will hereafter be referred to as original density, was determined based on the oven-dried weight and volume of each species. The length (longitudinal direction) of the specimens varied depending on the density of the species to allow for precise evaluation of the compressed wood mechanical properties: Japanese cedar, albizia, European spruce: 60 mm; red lauan, Douglas fir: 80 mm; and elm, Japanese beech, and Japanese birch: 100 mm. The width and thickness of each specimen were 40 mm and 6 mm, respectively.

Treatment with phenol–formaldehyde (PF) 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 was 10 min). The specimens were maintained under reduced pressure for 12 h, and then kept at ambient pressure and room temperature for 12 h. This process was repeated seven times to ensure complete penetration of the solution into the wood. After air-drying for three days, the treated specimens were vacuum-dried at 50°C for 12 h to remove any residual moisture (oven-dried). The weight gain (WG) and volume gain (VG) were determined using the oven-dried weight and volume before and after the treatment.

Measurement of deformation of wood

To gain an overview of the compressive deformation of the eight species, two oven-dried resin-impregnated specimens of each species were compressed at 150°C and 5 mm/min in the radial direction up to 10 MPa using hot plates fixed to the Instron universal testing machine 5500. Details of the compression procedure were described in a previous article.[6] After reaching a pressing pressure of 10 MPa, the crosshead movement was stopped and the distance between hot plates was held for 30 min to ensure resin curing.

Based on the results up to 10 MPa, five species were selected for compression up to 2 MPa and the pressing pressure was held constant for 30 min (this maintenance of pressure will hereafter be referred to as pressure holding). Two oven-dried untreated specimens of each species were also compressed for comparison of mechanical properties. The relationships between the pressing pressure and density were determined based on stress–strain curves.

Evaluation of bending properties

The Young's modulus and the bending strength of wood compressed at 2 MPa with pressure holding of 30 min were evaluated under oven-dried condition by a three-point bending test using an Instron 4411 universal testing machine at a crosshead speed of 5 mm/min. The span was adjusted based on the thickness of the specimens (15 times thickness).

Results and discussion

Weight gain and volume gain due to PF resin impregnation are shown in Table 1. Weight gain varied from 28.1% for Japanese birch to 72.4% for Japanese cedar despite the uniformity of the resin solution concentration. Volume gain also varied from 5.2% for albizia to 16.3% for Japanese birch. Because the volume gain is attributable to the swelling of cell walls due to resin impregnation, the specific volume gain (VG/specific gravity), a measure of the degree of swelling of the cell wall, was compared. As shown in Table 1, the specific volume gain varied from 17.7% for European spruce to 26.4% for elm. This shows that the swelling property of cell walls due to PF resin impregnation differs among species.

Figure 1 shows the deformation behavior of eight PF resin-impregnated species compressed up to 10 MPa. At the start of compression, all species deformed linearly against stress. Followed by the deformation, a yield point was exhibited. Some species showed gradual deviation from a linear response and it was very difficult to pinpoint the exact location of the collapse initiation point for Japanese beech (density of 0.64 g/cm^3) and Japanese birch (density of 0.71 g/cm^3). Hence, collapse-initiating stress was evaluated approximately as the interception point of the straight line of the linear region and a fitted line of the collapse-dominant region. The collapse-initiating stresses of elm (0.51 g/cm^3) and Douglas fir (0.50 g/cm^3) were 2.8 MPa and 2 MPa, respectively, around three times higher than that of Japanese cedar (0.31 g/cm^3) and albizia (0.23 g/cm^3).

Beyond the yield point, called the collapse-dominant region, wood tended to deform with little or no change in stress. As can be seen in Fig. 1, the stress–strain curves of the collapse-dominant regions were different among the species. Low density species such as Japanese cedar and albizia exhibited a larger collapse-dominant region while Douglas fir and red lauan displayed a shorter collapse-dominant region, and elm increased steeply even after the collapse initiation point. In general, it can be said that the higher the density, the higher the collapse-initiating stress and the shorter the collapse-dominant region.

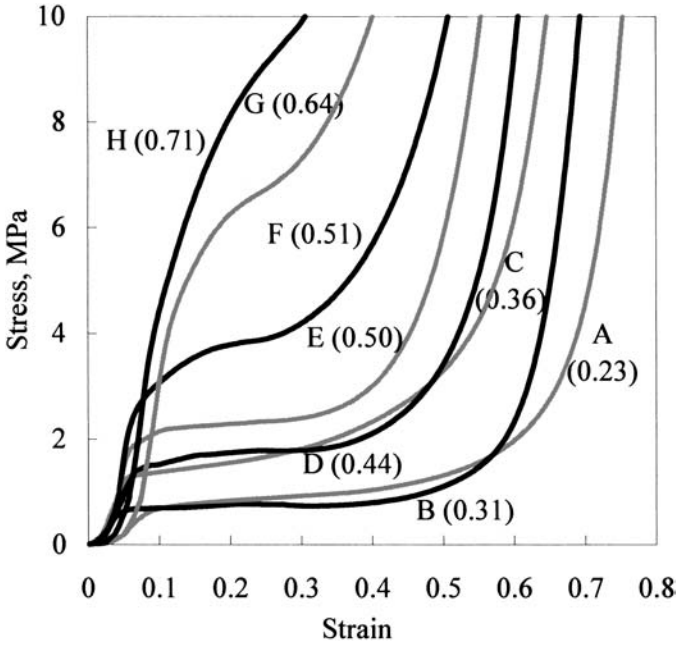

Fig. 1. Typical stress–strain curves of eight wood species impregnated with phenol–formaldehyde (PF) resin solution at a pressing temperature of 150°C and pressing speed of 5 mm/min. *A*, *B*, *C*, *D*, *E*, *F*, *G*, and *H* represent albizia, Japanese cedar, red lauan, European spruce, Douglas fir, elm, Japanese beech, and Japanese birch, respectively. The *values in parentheses* show the original density (g/cm^3) of each species

We further recognized that although the original densities of Douglas fir and elm are similar, their deformation behaviors are different. It is well known that the mechanical properties of wood in compression perpendicular to grain are affected by the anatomical characteristics of species, and softwoods, diffuse porous hardwoods, and ring porous hardwoods each behave differently.[1–5] Hence, we observed the anatomical structures of above two species.

As shown in Fig. 2, it is apparent that Douglas fir possesses a uniformly thin cell wall in the earlywood portion and low levels of ray tissue, whereas elm contains much fiber and ray tissue. According to the image of elm, the ray tissue seems to play an important role in resisting compressive deformation. It seems that not only density but also the anatomical features are critical for obtaining highly compressed wood at low pressing pressure.

As reported previously,[6] the mechanical properties of Japanese cedar increased linearly with the increase of density due to compression; hence, the relationship between the pressing pressure and density was investigated, as shown in Fig. 3. The density of Japanese cedar (original density 0.31 g/cm^3) compressed at 2 MPa was considerably higher than that of Japanese birch (original density 0.71 g/cm^3); in fact, it surpassed the density of Japanese birch at 1 MPa. The difference can be explained by the difference of collapse-initiating pressure as shown in Fig. 1. For Japanese cedar, collapse was initiated at around 0.7 MPa, and stress development during cell wall collapse was minimal; thus, a significant increment of density occurred up to 2 MPa. On the other hand, Japanese birch did not show any collapse until a pressing pressure of at least 3 MPa.

Furthermore, it was found that softwood such as Japanese cedar, European spruce, and Douglas fir were compressed more quickly after collapse was initiated than were hardwoods such as albizia and red lauan. It is worth noting that the density of European spruce (original density

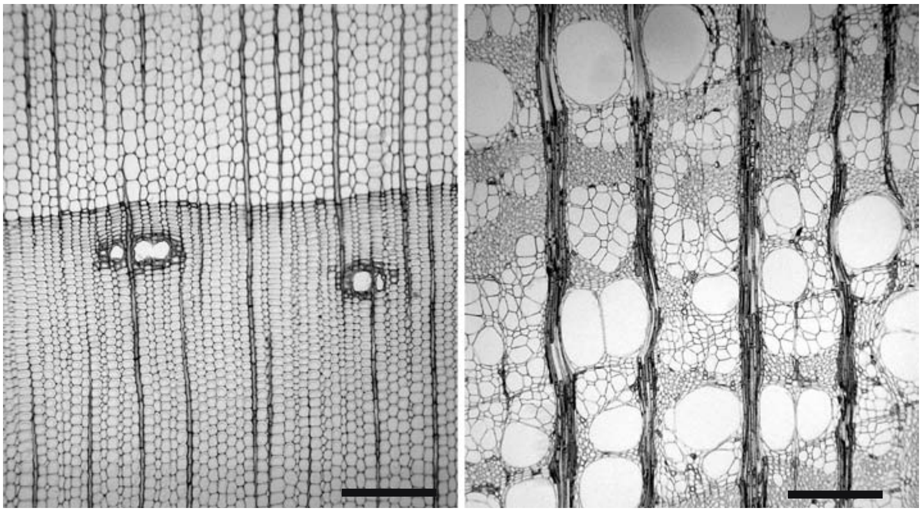

Fig. 2. Transverse sections of Douglas fir (*left*) and elm (*right*). *Bars*, 200 μm

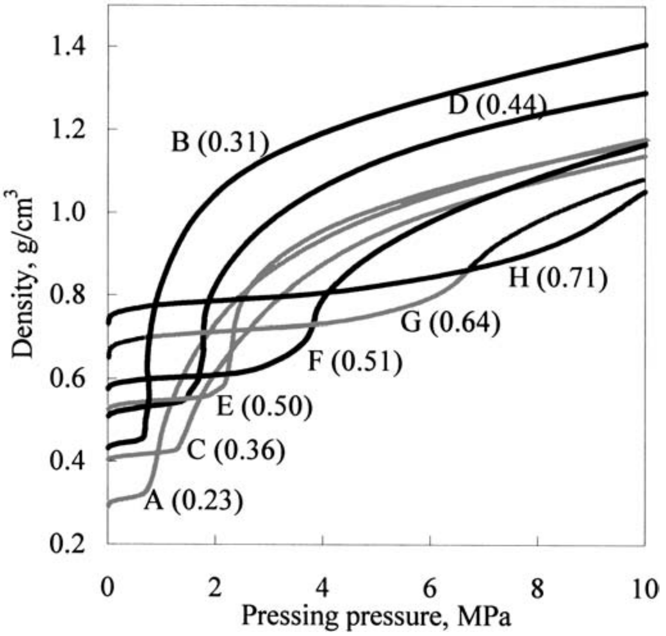

Fig. 3. Effects of low molecular weight PF resin impregnation and compression on the densification of eight species

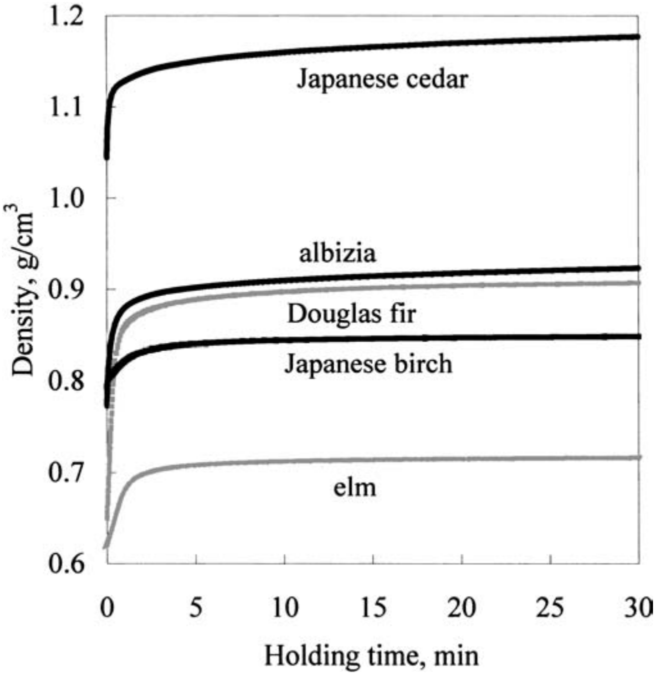

Fig. 4. Effects of holding time on the densification of wood species at 150°C and 2 MPa

0.44 g/cm^3) exceeded the density of albizia (original density 0.23 g/cm^3) at 2 MPa. This rapid deformation would be due to the relatively simple structure of the softwood.

After reviewing the species dependency in compressive deformation up to 10 MPa, we further hot pressed five species at 150°C and 2 MPa with pressure holding of 30 min, aiming at the application of this technique for the production of wood composites. In previous studies, we clarified that pressure holding causes significant creep deformation in Japanese cedar plasticized by low molecular weight PF resin.[6–8] This allowed the initiation of collapse at low pressing pressure and resulted in a considerable increment of density at low pressing pressure. Figure 4 shows that the density of PF resin-impregnated wood increased during pressure holding; however, the effects of pressure holding on the density differed among the species. Furthermore, we could see that within 1 min of holding, the density increased significantly regardless of high or low density species, and the density subsequently approached stabilization. Such stabilization is due to the resistance of the cell wall, not the curing of PF resin, which takes 10 min at 150°C.[6]

A significant increment of density occurred in Douglas fir during pressure holding. The stress–strain curve in Fig. 1 indicates that the collapse-initiating stress of PF resin-impregnated Douglas fir is around 2 MPa. Thus, during pressure holding, considerable collapse deformation occurred, resulting in a density increment from 0.63 g/cm^3 to 0.90 g/cm^3. In contrast, as can be clarified from Fig. 1, the deformation of Japanese cedar and albizia during pressure holding terminated due to the post-collapse region, where a rapid increase in stress occurred as the compression continued. The deformation of Japanese birch and elm also stopped with a slight increase in strain. This is due to the fact that until 2 MPa is reached, collapse deformation of these two species was not observed, as shown in Fig. 1. These deforming behaviors during pressure holding suggest that the optimum pressing pressure for marked deformation differs among species; the collapse-initiating pressure is a crucial indicator of these variations.

Eventually, by combining the pressing pressure with holding at 2 MPa, the density of Japanese cedar reached 1.18 g/cm^3 while Japanese birch showed a density of 0.83 g/cm^3, which did not differ from that of its uncompressed resin-treated condition. This result emphasizes the advantages of low density wood species as raw materials in the production of highly compressed wood at low pressing pressure.

As the mechanical properties of PF resin-impregnated wood improving linearly with density for Japanese cedar,[6] Young's modulus and bending strength in the longitudinal direction were compared between untreated compressed wood and PF resin-treated compressed wood. In general, it is difficut to compare the mechanical properties of compressed wood among species because the microfibril angle of the cell wall is variable; however, the changes in Young's modulus and bending strength due to compression can be compared.

Figure 5 shows the mechanical properties of PF resin-treated and untreated wood compressed at 2 MPa with pressure holding of 30 min. It was shown that PF resin-impregnated compressed wood attained higher Young's modulus, and bending strength in comparison with untreated compressed wood, especially for low density wood species. For example, the density, Young's modulus, and bending strength of PF resin-impregnated compressed Japanese cedar reached 1.18 g/cm^3, 21 GPa, and 240 MPa,

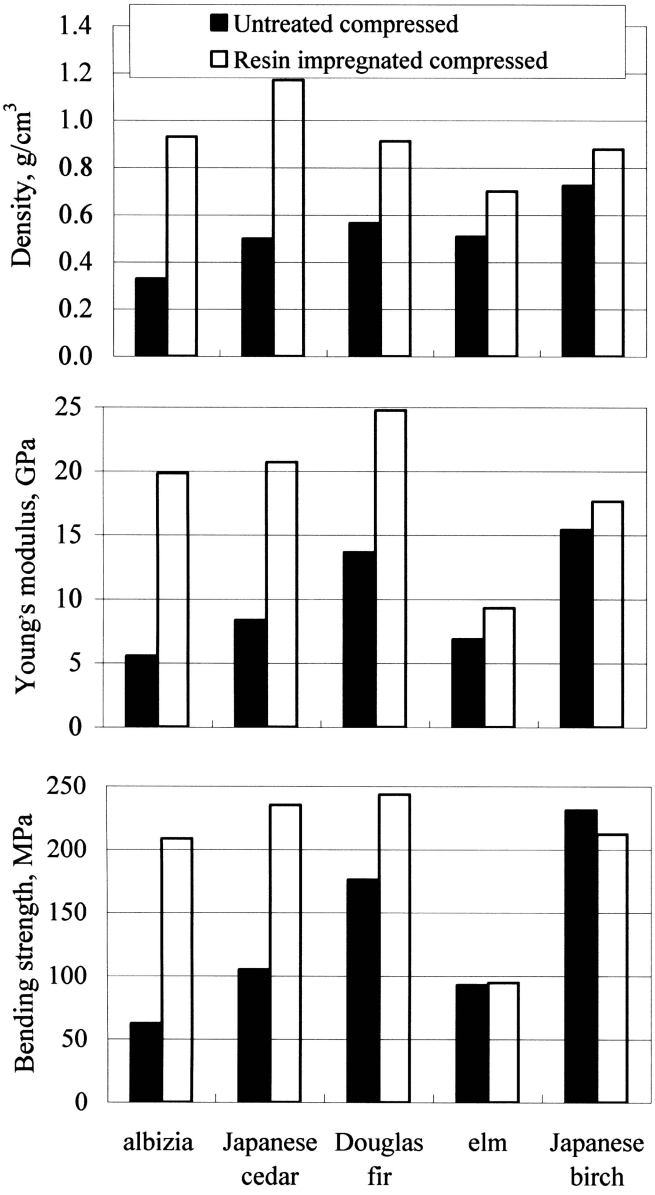

Fig. 5. Comparison of mechanical properties between untreated compressed wood and PF resin-treated compressed wood. All samples were compressed at 150°C and 2 MPa with a pressure holding of 30 min

respectively, due to high compression, while those of untreated compressed Japanese cedar were 0.50 g/cm^3, 8 GPa, and 100 MPa, respectively. Conversely, the mechanical properties of compressed Japanese birch and elm did not differ due to resin impregnation. It is concluded that as long as we use this technique involving PF resin impregnation and compression, low density wood is suitable as raw material for the production of high-strength wood at low pressing pressure.

Conclusions

To obtain highly compressed resin-impregnated wood at low pressing pressure, we investigated species dependency on the compressive deformation of PF resin-impregnated wood. It was demonstrated that differences among the species in deformation behavior were mainly attributable to the density as well as to the anatomical features of species. PF resin-impregnated wood could be further compressed during pressure holding, and the effects of pressure holding differed among species. Eventually, after pressure holding at 2 MPa, the density of Japanese cedar reached 1.18 g/cm^3, about 30% higher than that of Japanese birch, which possesses an original density that is 2.5 times higher than that of Japanese cedar. The mechanical properties of resin-impregnated wood, especially low density wood, improved with density. Thus, it is concluded that low density wood species have an advantage as raw materials for obtaining high-strength wood at low pressing pressure.

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