

FIXATION OF COMPRESSED WOOD USING MELAMINE-FORMALDEHYDE RESIN

Masafumi Inoue

Student
Wood Research Institute

Shigeyuki Ogata

Research Chemist
Koshii Mokuzai Co., Ltd.
Hirabayashikita, Suminoe
Osaka 559, Japan

Shuichi Kawai

Associate Professor
Wood Research Institute

Roger M. Rowell

Team Leader
USDA Forest Service
Forest Products Laboratory¹
One Gifford Pinchot Drive
Madison, WI 53705-2398

and

Misato Norimoto

Professor
Wood Research Institute
Kyoto University
Uji 611 Kyoto, Japan

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ABSTRACT

Methods to maximize wood hardness and dimensional stability include various combinations of compression, heating, and chemical treatment. In this study, wood was treated with increasing concentrations of a low molecular weight, water-soluble melamine-formaldehyde resin solution (mol wt 380) and compressed while heated. This method achieved a maximum bulking efficiency of 5% and an antishrink efficiency of 45%, showing that the chemical had not completely penetrated the cell wall. Once the wood was treated, its ability to retain the compressed state was tested by immersing wood specimens in water at different temperatures. Specimens treated with an 8% resin solution retained almost complete fixation when soaked in room-temperature water, while those treated with a 25% solution retained fixation in boiling water. Moreover, a 25% solution of resin and a compression of 54% increased hardness from 0.48 to 0.72 MPa.

Keywords: Melamine-formaldehyde, compressed wood, permanent fixation, recovery of set, surface hardness.

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INTRODUCTION

In previous work, we showed that wood could be permanently fixed in a compressed state by steaming it while under compression (Inoue et al. in press). This method increased hardness while increasing specific gravity. We have also investigated the use of water-soluble monomers that can be polymerized during the compression step (Inoue et al. 1991a, b). This method, first introduced by Stamm and Seborg (1951), involved impregnating wood with water-soluble phenol-formaldehyde to cause chemical weight gains of 25% to 30%. These authors dried the impregnated wood, compressed it to a density of 1.3 to 1.4, and finally cured it at 140 C to 150 C. The product, known as compreg, showed increases in strength and hardness properties in proportion to the increase in specific gravity.

Building on later work of Stamm (1964), we found that almost complete fixation of compressed state occurred using phenol-formaldehyde solutions at concentrations above 10% and curing at 130 C for 1 h. Scanning electron micrographs showed that no phenol resin remained in the cell lumen. The resin in the cell wall resulted in an antishrink efficiency of about 60%. The resulting dimensional stability contributed to the permanent fixation of the compressed state. A resin concentration of 15% and a compression set of 60% increased hardness by a factor of 3, compared to that of non-compressed wood, and a 40% resin concentration, by a factor of 4 (Inoue et al. 1991a).

The study reported here investigated the ability of a low molecular weight, water-soluble melamine-formaldehyde (MF) resin system to permanently fix compressed specimens and increase hardness.

MATERIALS AND METHODS

Wood specimens

All specimens [20 by 20 by 30 mm (longitudinal by radial by tangential)] of Sugi (*Cryptomeria japonica* D. Don) were cut from one large piece of the same board and extracted with benzene/ethanol (1/1, v/v). The specific

gravity of the wood was 0.36, and the annual ring width was 1.7 mm.

Treatment with melamine formaldehyde

Oven-dried specimens were soaked in solutions of melamine methylolated with formaldehyde (mol wt 380) of increasing concentrations (2%, 4%, 8%, 15%, and 25%). After air drying for 24 h, the treated specimens were slowly warmed over a 12-h period to 105 C and then compressed in the radial direction for 1 h at different temperatures (120 C, 140 C, 160 C, and 180 C). Three specimens of each concentration and temperature were prepared with a compression of 54%.

Weight percent gain (WPG) was determined using oven-dried specimen weights before and after treatment. Antishrink efficiency (ASE) was determined by soaking the specimens in water at room temperature (vacuum for 30 min and atmospheric pressure for 3 h) (Stamm 1964).

Bulking efficiency was calculated as

$$BE = \frac{V_A - V_B}{V_B} \times 100 (\%) \quad (1)$$

where V_A is oven-dry volume after treatment and V_B is oven-dry volume before treatment.

The percentage of size reduction from the original to the compressed state (C) was calculated as

$$C = \frac{T_0 - T_C}{T_0} \times 100 (\%) \quad (2)$$

where T_0 is oven-dried thickness before compression and T_C is oven-dried thickness after compression.

Recovery of compression set

Three tests helped determine to what degree the wood would recover from having been compressed. The first, a simple boiling test, involved soaking the specimens in water until saturated (30 min under reduced pressure and then 210 min at atmospheric pressure), placing them in a boiling water bath (98 C) for 30 min, and measuring their thicknesses after oven-drying. The second, a cyclic swelling test, in-

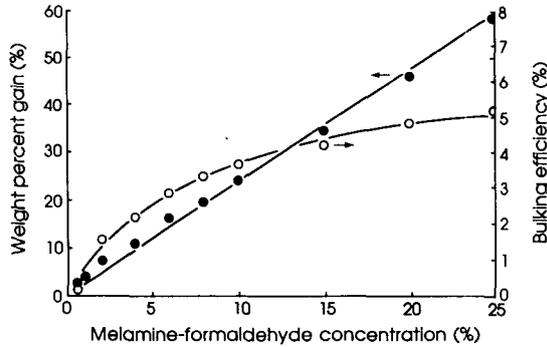


FIG. 1. Weight percent gain and bulking efficiency resulting from increasing concentrations of melamine-formaldehyde treatment.

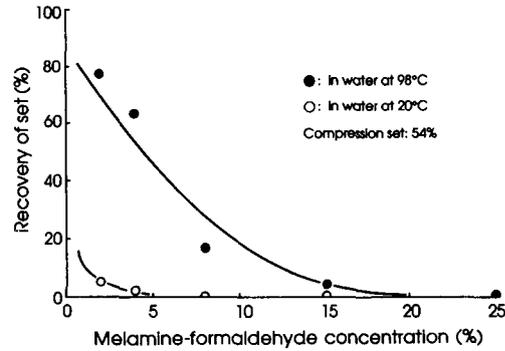


FIG. 3. Effect of melamine-formaldehyde concentration on recovery of set in water at 20 C and 98 C. Compression set, 54%.

involved soaking the specimens in water until saturated (30 min in a vacuum and 210 min at atmospheric pressure) and measuring their thickness. After drying the specimens at 40 C for 20 h and then again at 105 C for 4 h, thicknesses were measured. This procedure was repeated eight times. After the eighth wetting cycle, the specimens were placed in a boiling water bath for 2 h (98 C), measured, and then oven-dried. Three more boiling and drying cycles completed the test.

The third and final recovery test involved increasing the water temperature (starting at 20 C) in increments of 10 C seven times until it reached 98 C, measuring recovery at each interval.

Recovery (R) was calculated as

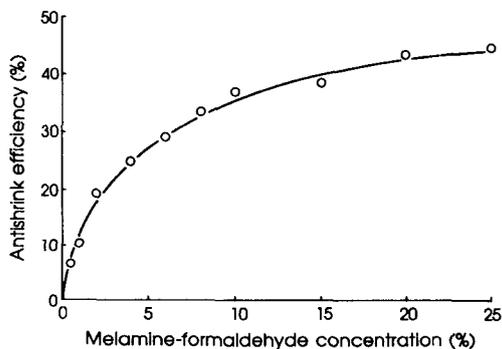


FIG. 2. Antishrink efficiency resulting from increased concentration of melamine-formaldehyde treatment.

$$R = \frac{T_R - T_C}{T_0 - T_C} \times 100 (\%) \quad (3)$$

where T_R is oven-dried thickness after recovery.

Hardness test.—The Brinell hardness test was conducted on the compressed specimens (according to the Japanese standard JIS Z2117), using a 10-mm-diameter steel ball with a head speed of 0.5 mm/min to a depth of 0.32 mm. Sixteen different points were measured for each treating condition, and the results were averaged.

RESULTS AND DISCUSSION

Figure 1 shows the WPG and bulking efficiency (BE) of wood treated with increasing concentrations of MF. The WPG increased proportionately with increasing MF concentration, but the BE did not. A maximum BE of 5% shows that the chemical occupied only about one-half of the theoretical void volume of the cell wall. Since the WPG continued to increase with increasing MF concentration while the BE did not, excess MF was in the lumen.

As with the BE, the ASE was not linearly related to MF concentrations (Fig. 2). A resin concentration of about 10% achieved a maximum ASE of about 40%; higher concentrations had little effect. This also shows that only

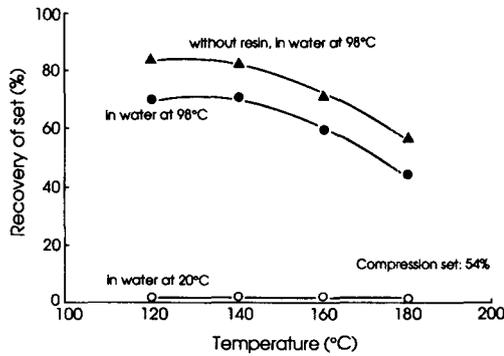


FIG. 4. Effect of press temperature for 1 h pressing on recovery of set in water at 20 C and 98 C. Compression set, 54%.

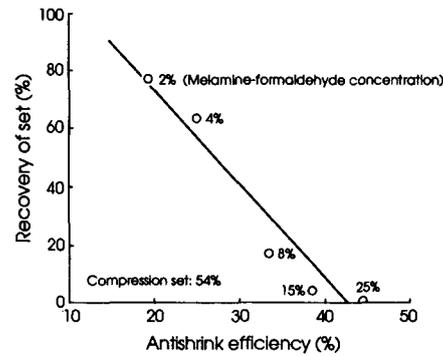


FIG. 6. Relationship between recovery of set of compressed wood and antishrink efficiency of noncompressed wood with melamine-formaldehyde treatment. Press time, 1 h; press temperature, 140 C; compression set, 54%.

a limited amount of MF resin was in the cell wall acting as a bulking agent.

Figure 3 shows the effects of increased MF concentrations on the recovery of specimens from their compressed state. Almost complete fixation was achieved with an 8% solution of MF in a water-soaking test at room temperature. Under boiling water conditions, <5% recovery occurred at a concentration of 15%, with <2% at a concentration of 25%.

The effects of pressing temperature and time were determined using a 4% MF solution. Figure 4 shows that increasing the pressing temperature from 120 C to 180 C for 1 h did not improve the resistance to recovery under room-temperature conditions. Increasing the press temperature to 180 C stabilized the set in boiling water to about 40% compared to 70% at

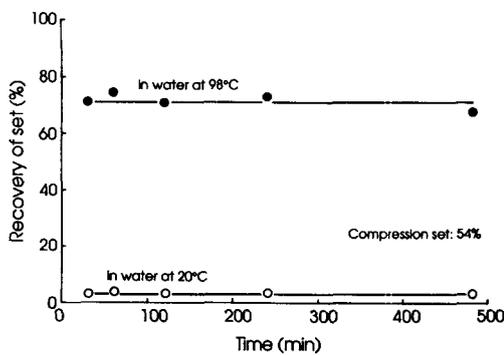


FIG. 5. Effect of press time at 140 C on recovery of set in water at 20 C and 98 C. Compression set, 54%.

120 C. Using heat in combination with MF resin improved the set stability by about 15% over using heat alone as a result of the resin's effect on cell-wall bulking.

Figure 5 shows that increasing pressing time at 140 C did not improve fixation. Once the resin is cured, which takes about 30 min at 140 C, there is no advantage in pressing for a longer time.

Figure 6 gives further evidence that cell-wall bulking, which results in dimensional stability, is primarily responsible for keeping wood in a compressed state. As the ASE increased, the wood's dimensional instability decreased. In fact, at 45% ASE, the wood retained its compressed state very effectively. From these results, we speculate that other types of chemical modifications of wood that impart even higher ASE values could cause a high degree of dimensional stability.

Figure 7 shows the results of the wetting, drying, and boiling cyclic test. Specimens soaked in 2% and 4% MF solutions showed a large increase in dimensional instability during the first eight wetting and drying cycles, while specimens soaked in 8%, 15%, and 25% MF solutions effectively resisted swelling. After the four boiling cycles, the 2% and 4% specimens recovered in almost the same way as the untreated specimens. The 8% specimens recovered to about 40%. Only the specimens treated

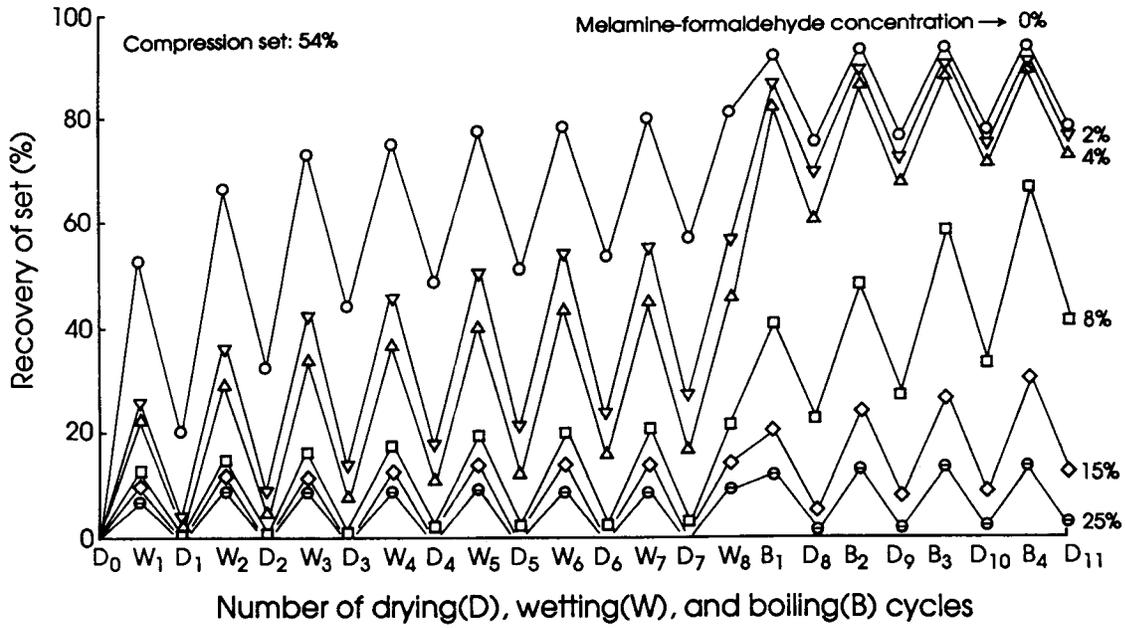


FIG. 7. Recovery of set for melamine-formaldehyde-treated compressed specimens after wetting, overdrying, and boiling cyclic test. Press time, 1 h; press temperature, 140 C; compression set, 54%.

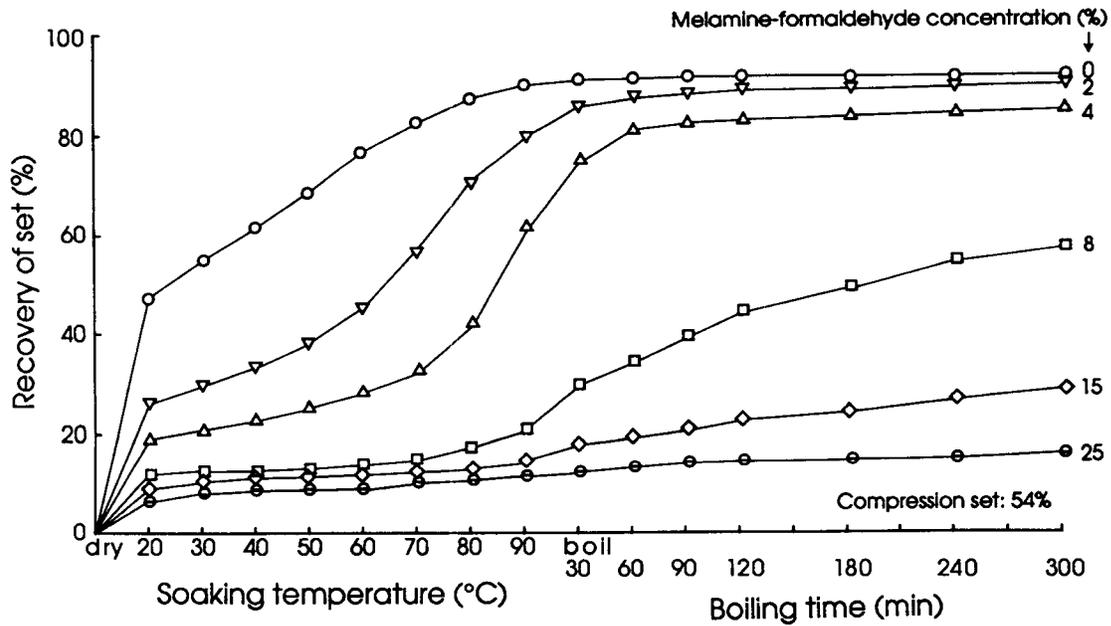


FIG. 8. Recovery of set for melamine-formaldehyde-treated compressed specimens with increasing temperature and boiling time. Press time, 1 h; press temperature, 140 C; compression set, 54%.

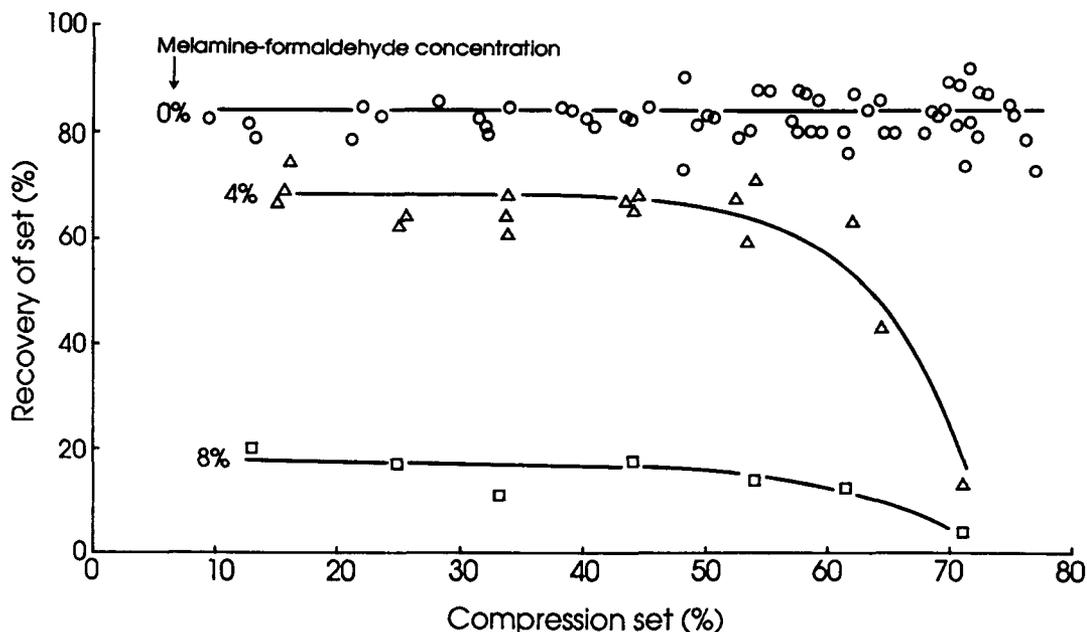


FIG. 9. Relationship between recovery of set of compressed wood and compression set for melamine-formaldehyde treatment. Press time, 1 h; press temperature, 140 C; compression set, 54%.

with 25% MF retained high dimensional stability.

The failure of the lower concentrations of MF resin to retain dimensional stability could be due to incomplete polymerization caused by the wide distribution of monomer in the cell wall. With resin concentrations exceeding 15%, a higher molecular weight polymer may form in the cell wall.

Figure 8 supports this explanation. This ex-

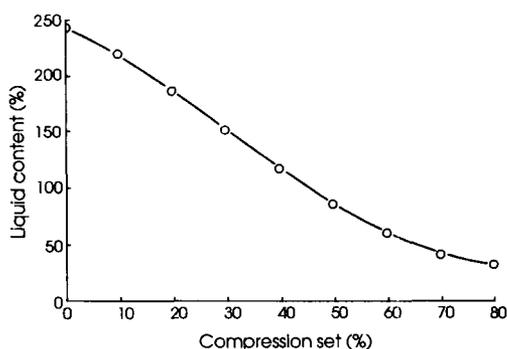


FIG. 10. Liquid volume contained in wood with increasing compression.

periment measured recovery from the compressed state as the temperature of the soaking water was increased. The specimens soaked in 2% and 4% MF solutions failed to retain compression as the temperature increased to the boiling point. Those soaked in an 8% MF solution were stable at temperatures up to the boiling point, but then they swelled as boiling time increased. Specimens treated with 15% and 25% MF solutions resisted swelling even after boiling for a period of time.

Figure 9 shows the relationship between the recovery from the compressed state and the percentage of compression. When no MF resin was used, recovery did not depend on the degree of compression. This same relationship held for the 4% MF solution up to 60% compression. Beyond 60% compression, recovery was greatly decreased; at about 60% compression, the collapsed cell walls started to touch each other. When this occurred, a concentration of chemical may have allowed a more stable polymer to form with a higher molecular weight.

Hardness increased in proportion to MF

concentration. Starting with a hardness of 0.24 MPa at 0% MF, hardness increased to about 0.48 MPa at 15% MF and to about 0.72 MPa at 25% MF when compressed 54%.

Increasing the hardness beyond 0.72 MPa would require a harder polymer at a higher level. Noncompressed wood can hold about twice its weight in water (Fig. 10). Similar weight gains of other liquids should be possible if the liquids completely penetrate the cell wall and completely fill the lumen. As the wood is compressed, the lumen volume decreases, resulting in less chemical storage capacity. At a compression of 50%, the cell wall and lumen can hold about 85% liquid, while a compression of 70% reduces liquid storage to 20%. It may be necessary to have a 60% final polymer weight gain to achieve a very high hardness value, so the wood cannot be compressed >60%. There will be a balance between hardness gained by compression and that gained by polymer weight and hardness.

CONCLUSIONS

Wood specimens with melamine-formaldehyde resin weight gains in the range of 8% to 10% retained their compression state almost completely when soaked in room-temperature water. Specimens with weight gains in the range

of 25% to 35% retained their compressed state even in boiling water. Stabilization of the compressed set was due, in part, to the bulking effect of the MF resin in the cell wall, which resulted in a limited degree of dimensional stabilization.

Research is continuing to investigate cell-wall bulking chemicals that impart a high degree of dimensional stability, with high levels of very hard polymers in the cell voids.

REFERENCES

- INOUE, M., M. NORIMOTO, Y. OTSUKA, AND T. YAMADA. 1991a. Surface compression of coniferous wood lumber. I. Permanent set of surface compressed layer by a resin. *Mokuzai Gakkaishi* 35(3):234-240.
- , ———, ———, AND ———. 1991b. Surface compression of coniferous wood lumber. II. Permanent set of compression wood by low molecular weight phenolic resin and some physical properties of the products. *Mokuzai Gakkaishi* 35(3):227-233.
- , ———, M. TANAHASHI, AND R. M. ROWELL. 1993. Steam or heat fixation of compressed wood. *Wood Fiber Sci.* 25(3):224-235.
- JAPAN INDUSTRIAL STANDARD. 1977. Surface hardness of wood. Standard JIS Z2117.
- STAMM, A. J. 1964. *Wood and cellulose science*. Ronald Press, New York, NY.
- , AND R. M. SEBORG. 1951. Resin-treated laminated, compressed wood—Compreg. Rep. No. 1381. USDA, Forest Service, Forest Products Laboratory, Madison, WI.