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Compressive-Molding of Wood by High-Pressure Steam-Treatment:

Part 1. Development of Compressively Molded Squares from Thinnings

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Summary

We attempted to compressive-mold logs of sugi (*Cryptomeria japonica* D.Don) to squares, and to permanently fix the transformed shape. The transformation and fixation were successively conducted under high-temperature saturated steam atmosphere by a newly designed apparatus which was a combined pressure vessel and a press machine. The results are summarized as follows:
(1) The logs were successively softened by steaming at 150 °C, compressively molded to square, and fixed the transformed shape by steaming at 200 °C in this apparatus.
(2) The shape of square fixed by high-pressure steam-treatment did not recover to original log shape by water-soaking and boiling. The result shows that the high-pressure steaming is effective for permanent fixation of the transformed shape.
(3) The condition for sufficient fixation of the compressive transformation was steaming at 200 °C for 3 min when a 15 cm of diameter and 10 cm of length sugi-thinning was used as a specimen.
(4) Inner stress within a compressed wood was gradually reduced during steaming and, finally, the stress was removed completely.

Introduction

Nowadays, it has become indispensable and urgent to carry out thinning operations in plantation forests. Only a few thinnings are utilized and the rest is left behind in the forest. There is therefore the need to develop a method for their utilization as a new constructive material.

There are some recent reports on efforts being made to solve the problem, for example, the studies on surface compression in the radial direction with microwave irradiation (Inoue *et al.* 1990). These methods have a problem of how to fix the compressive transformation. As one of the methods to fix the transformation, heating treatment at a dry condition was investigated. However, this method requires 20 h at 180 °C or 5 h at 200 °C of treatment for the complete fixation of the transformation. In another example, the formation of covalent crosslinking between the wood components in the deformed state was conducted (Inoue *et al.* 1991 a, b). However this method has a disadvantage of reducing the quality of wood material.

According to studies on steam explosion (Tanahashi *et al.* 1989 a; Tanahashi 1990) of wood chips, it was clarified that the crystalline form of wood cellulose was completely transformed from cellulose I α to cellulose I β, and that non-crystalline region of cellulose and hemicelluloses were rapidly hydrolyzed by steaming. Further, the crystallinity of cellulose is increased by steaming at high temperatures with or without explosion. We assumed that inner stress within compressively transformed wood remained at non-crystalline region of cellulose. In order to obtain permanent fixation of the transformed wood, it is necessary to rearrange the crystalline form of cellulose microfibril in association with the activation of the molecular movements by steaming, or to release the inner stress kept in transformed wood by partial degradation of glycoside bond with high-tensioned cellulose. It was earlier recognized that steam explosion is effective for this, so the high-pressure steam-treatment is effective means for permanent fixation of transformed wood.

We reported that almost complete fixation of compressive transformation in wood can be achieved by steaming (Tanahashi 1991 b; Inoue *et al.* 1993). However, this steaming process demands many steps and much time. The steps follow as saturation of water into the specimen, heating by microwave irradiation, compressing the wood, drying-set by heating between plates of hot press, restraining the conversion of transformation by holding with clamps and finally heating with high-pressure steam in an autoclave to fix the compressive transformation.

Six years ago, we took a patent (Tanahashi *et al.* 1989 b) and presented the research data of compressive-molded wood at the annual meeting of the Japan Wood Research Society (Tanahashi *et al.* 1991 a) and the Society of Fiber Science and Technology, Japan (Ito *et al.* 1991). This paper reports a new technique to produce completely fixed compressively molded squares from thinnings by high-pressure steam-treatment without any sawing.

Experimental

Development of compressive-molding apparatus

We developed a new high-pressure steam compressive-molding apparatus (Hisaka HTP-40/58) as shown in Figure 1. It has an inner diameter of 40cm, a depth of 58cm, and the maximum pressure used was 2.0MPa. The apparatus has a press cylinder through a vessel sealed with a metal sealer; the press is installed for the purpose of molding under high-pressure steam atmosphere. The maximum allowance on the press load is 14×10^3kg.

Permanent fixation of compressed wood by steam treatment

Sugi (*Cryptomeria japonica* D.Don) was used as the specimen after being dried at room temperature. The size of a specimen was 20mm long, 20mm in radius and 30mm thick. The specimens were treated in the compressive-molding apparatus. In this method, the specimens were softened by saturated steam at 150°C for 3min. After this, they were compressed in the direction of R (radial direction) to 10mm, and the temperature was raised to 190 or 200°C, and then treatment for shape fixation was given for 2min.

The amount of compression set (C_1) was defined by the following expression :

$$C_1 = \frac{To - Tc}{To} \times 100\,(\%)$$

where To and Tc are sizes in the direction of R (radial direction) in oven-dried condition before and after the compression, respectively.

In order to determine the effects of steam-treatment before and after compression, a recovery test was conducted. Set-recovery was determined by a test where drying, water absorption, and boiling were repeated. The size in dry condition (oven-dried condition) was measured at a completely dried state after drying the specimens at room temperature for 24h, and drying them in a vessel of 105°C for 24h. The size in water absorption condition (water saturated condition) was measured after soaking the specimens in distilled water at room temperature until saturation, reducing the pressure by an aspirator for 30min, and leaving them for 24 hours. This procedure was repeated for nine times. After the ninth water absorption stage, the specimens were treated with boiling water. The sizes after boiling were measured immediately after boiling them in hot distilled water for 30min and then, finally, specimens were measured for the size in oven-dried condition.

The set-recovery (R_1) was defined as follows:

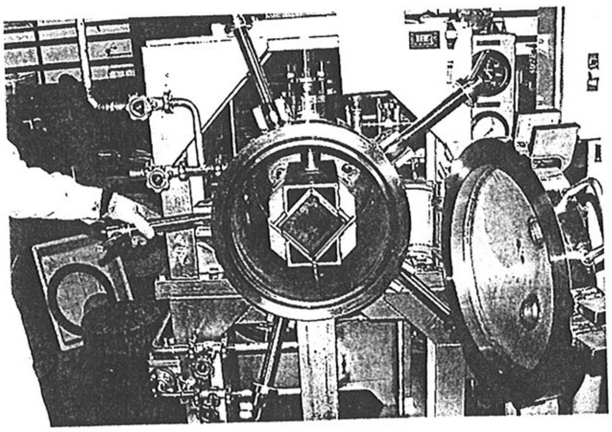

Fig. 1. The apparatus to compressive-mold by the high-pressure steam.

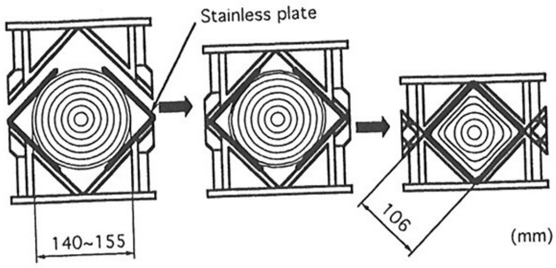

Fig. 2. Compressive-molding from a log to square.

$$R_1 = \frac{Tr - Tc}{To - Tc} \times 100\,(\%)$$

where Tr is the thickness in the direction of R (radial direction) under oven-dried conditions after the recovery treatment.

Compressive-molding process from log to square shapes

The compressive-molding apparatus contains the special jig. It is possible that the apparatus compressive-mold a log to compressively molded square as shown in Figure 2. The maximum allowance on the diameter and the length of the logs is 20cm and 34cm, respectively.

The raw logs from sugi thinning were used as samples. Air-dry specific gravity and average annual ring width were 0.38 and 1.3mm respectively. The diameter and the length of logs were about 15cm and 10cm, respectively.

A compressive-molding process was conducted as follows. First, a log was softened in the compressive-molding apparatus by saturated steam at 135 or 150°C for 1 or 3min. Secondly, it was compressive-molded to a square (10.6×10.6cm). Thirdly, the steam temperature was raised to 180°C or 200°C, and then treatment for shape fixation was given for 1 or 3min. Finally, the steam was exhausted to atmospheric pressure. The press cylinder was raised when there was no more steam from treated wood, and the compressive wood was taken out from the processing container. The time required for processing was 10–15min.

The compression set (C_2) was defined as follows:

$$C_2 = \frac{So - Sc}{So} \times 100\,(\%)$$

where So and Sc are the cross-sectional areas before and after pressing, respectively.

Recovery of shape by soaking in distilled water

In order to determine the effects of steaming before and after compression set, the recovery test was conducted. Specimens for the recovery test were cut to 10mm long. Specimens were prepared as shown in Figure 3. Water saturated specimens were prepared by impregnation under vacuum. After all specimens were soaked in water at 20°C for 24h, they were boiled in hot distilled water for 30min. Finally, the cross-sectional area of specimens were measured after drying at room temperature.

The set-recovery (R_2) was defined as follows:

$$R_2 = \frac{Sr - Sc}{So - Sc} \times 100\,(\%)$$

where Sr is the sectional area after the recovery treatment.

However, it was impossible to measure So of the type 2 specimens in the same manner, so the set-recovery (R_3) was calculated by

$$R_3 = \frac{Sr}{Sc} \times 100\,(\%)$$

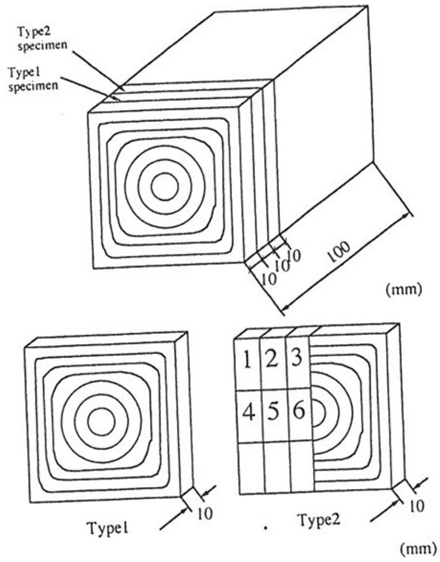

Fig. 3. Shapes of wood specimens for recovery test.

R_3 shows that the compressive transformation is fixed, when R_3 is equal to 1. R_3 increases if the compressive transformation is recovered.

Measurement of inner stress

The relation between inner temperature and inner stress in specimen was investigated by measuring the thermocouple and newly designed small stress-sensor attached in wood specimen, respectively, as a function of steaming time. The stress-sensor could be used under high-temperature and high-pressure steam conditions. Stress-sensor and thermocouple were inserted at a depth of 100 mm in wood specimen of sugi (300 mm long, 50 mm in radius and 100 mm thick) as shown in Figure 6. The specimen was softened by saturated steam at 150 °C for 5 min, and then compressed in radial direction to 16.7 mm. The compression set (C_1) was 66.6 %. After this, the temperature was raised to 200 °C, and then treatment for shape fixation was given for 70 min. During these processes, inner temperature and inner stress were measured.

Results and Discussion

Permanent fixation of compressed woods by steam treatment

Figure 4 shows the effects of the steaming temperature on the recovery of compressed specimens for the drying, wetting, and boiling cycles. In this figure, 0 % on the vertical axis means that the shape of the compressed wood is completely fixed, whereas 100 % means that it returns to its original shape. In the case of steaming for 0 min during the fixation process (referred to as "drying set" in this report), the specimen was almost recovered to the original size by twice boiling time. In the case of steaming at 190 °C for 2 min, the shape of the specimen with water at room temperature was almost fixed. However, it was found that it recovered to some extent by hot-water boiling indicating that the fixation processing was not yet complete. That is, stress was not completely relieved. However, the fixation of compressive transformation was approximately achieved by steaming at 200 °C for 2 min because the set-recovery was near to zero after any drying stage. Incidentally, the reason why the set-recovery was under 0 % after the final drying stage is that hemicellulose was extracted from the specimen and the wood shrank. The cause for it being minus is speculated that part of the hemicellulose became water-fusible and was lost. This point will be discussed in detail in the forthcoming report. The complete fixation was thus achieved in one step technique. The merit of this continuous method is that the process of softening → compressing → fixation could all be handled in one step by high-pressure steam within the same apparatus. Heat and water treatments are given to wood suddenly, so that its viscoelastical transformation is easily made and also the transformation will be fixed in a short time.

In Figure 4, fluctuation area degree of the set-recovery (R_1) in fixed specimen between dry state and water absorption condition was bigger than in drying-set ones. Table 1 shows the amount of fluctuation "t" in swelling-shrinking calculated every time drying-water-adsorption is repeated. "t" for unprocessed wood and drying-set wood was comparatively small, but "t" for steamed wood at

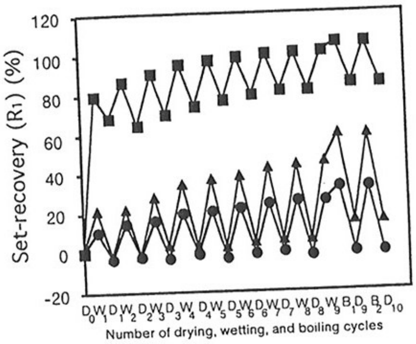

Fig. 4. Set-recovery (R_1) for steamed compressed specimens after drying, wetting, and boiling cyclic test.
Legends: ■: Steamed for 0 min in fixation process (Drying set). ▲: Steamed at 190 °C for 2 min in fixation process. ●: Steamed at 200 °C for 2 min in fixation process.
Notes: Specimens were steamed at 150 °C for 3 min in softening process. The compression set (C_1) was 50% "D", "W", "B" show oven-drying, wetting and boiling.

Table 1. The amount of fluctuation "t"(mm) in swelling-shrinking calculated every time drying-water-adsorption is repeated

water-drying -adsorption	unprocessed wood	Drying set	Fixation Process Temp.-Time (°C) (min) 190–2	200–2	200–8
W2-D1	0.4	1.8	2.6	2.0	1.5
W3-D2	0.7	2.6	3.1	2.0	2.1
W4-D3	0.7	2.5	3.3	2.5	2.0
W5-D4	0.8	2.3	3.7	2.3	1.6
W6-D5	0.8	2.1	3.8	2.8	1.2
W7-D6	0.9	2.0	4.0	2.8	1.4
W8-D7	0.9	1.9	4.0	2.8	1.4
W9-D8	0.9	2.0	4.2	3.0	1.5
Average	0.8	2.1	3.6	2.5	1.6

190 °C for 2min in the fixation process was larger. It seems to suggest that cells in the shape-fixed specimen tend to be transformed more in the direction of compression of cell shapes, by the force generated from the swelling of cell walls. In other words, the amount of fluctuation in shape caused by the swelling in the direction of thickness becomes larger, but that in the direction of width becomes relatively small. Also, "t" at 200 °C for 8 min showed even a smaller value. This seems to indicate that the amount of hemicellulose in cell walls that goes away increases with the processing time increment, and thus the amount of enlargement at the time of swelling by water adsorption gradually decreased due to the decrease of the amount of hydrophile radicals. Alternatively, it is possible to think that the non-crystalline area of cellulose within the cell wall gradually changed into a crystal area as the processing time increased, which caused a decrease in the amount of shape change that occurred in the non-crystalline area of the cellulose.

Compressive-molding process from log to square

Next, we tried to apply this technique to larger specimens, such as compressive-molding process from log to square. The method has already been described in the experimental section with a condition of 150 °C for 3 min during the softening process and 200 °C for 3 min during fixation. Figure 5 shows the photograph of thinnings before and after compressive-molding. This indicates that compressive-molding of cedar lumber from thinning into rectangular lumber is possible. Even logs with gnarls or distorted shapes can be molded into rectangular lumber with this method.

The relationship between the steam temperature and compression set is shown in Table 2. If a log of 15 cm diameter is compressive-molded to 10.6 × 10.6 cm square, the value of C_2 is about 36 %. This result shows that logs steamed at 135 °C during the softening process were not completely compressive-molded to squares. The reason may be that the temperature and time of the steaming were not enough to soften the log. When logs were steamed at 150 °C for 3 min during the softening process, they could be completely compressive-molded to squares, because the value of C_2 was close to 36 %.

Table 2. Compression set (C_2) and set-recovery (R_2) of compressively molded squares

Softening process		Compression	Fixation process		Set -recovery
Temp. (°C)	Time (min)	set (C_2) (%)	Temp (°C)	Time (min)	(R_2) (%)
135	1	19.9	170	3	86.0
135	1	17.7	180	3	64.2
135	1	21.5	190	3	56.1
135	3	25.2	170	3	75.0
150	3	33.4	170	3	79.5
150	3	34.2	180	3	72.8
150	3	33.4	190	3	62.2
150	3	34.7	200	3	4.5

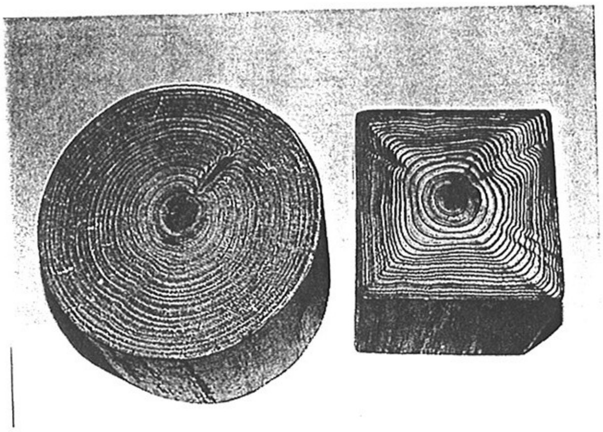

Fig. 5. Thinnings before and after compressive-molding.

Recovery of shape by soaking to water

The effect of steaming after compression set researched on the recovery tests with type 1 specimens is shown in columns 5–8 of Table 2. It is obvious that the value of R_2 decreases with an increase in fixation temperature. It is also evident that steaming at 200 °C for 3 min is required for the complete fixation of 15 cm diameter and 10 cm length logs.

Table 3 shows the results of the recovery tests with type 2 specimens to determine the effect of steaming condition in each location on fixation process. Specimens No. 4 have the maximum value for R_3. The reason is that the No. 4 specimen is the most compressed part in molded squares. Both No. 1 and No. 2 have about the same amount of compressed parts and parts extending into rectangular shape, and thus, the average compression ratio is about 1. R_3 under weak fixation conditions is also about 1. Moreover, since No. 5 and No. 6, the parts that include much heartwood, have been hardly compressed, their R_3 is about 1. From the above, it can be deduced that if the recovery of No. 4 specimen can be stopped, the recovery of the whole specimen can be suppressed. Therefore, it is evident that steaming at 200 °C for 3 min is required for the complete fixation. These facts clarify the processing conditions necessary for manufacturing compressively molded rectangular lumber from small-diameter logs and also the possibility of manufacturing cedar lumbers from thinnings by compressive molding.

Inner stress of compressive transformed wood

Figure 6 shows the relaxation process of inner stress in wood specimen by using special designed stress-sensor which can be used in high-pressure and high-temperature steam. The stress sensor developed in this project is with a distortion gauge attached inside an air-tight stainless container. It is able to measure under high-pressure and high-temperature conditions, and temperature correction of

Table 3. The effects of steaming on set-recovery (R_3) in type 2 specimens

Softening process		Compression	Fixation process		Set-recovery (R_3) Location No.					
Temp. (°C)	Time (min.)	set (C_2) (%)	Temp. (°C)	Time (min)	1	2	3	4	5	6
150	3	33.4	170	3	1.05	1.01	1.06	1.45	0,93	0.96
150	3	34.2	180	3	1.03	1.01	1.08	1.42	1.08	1.00
150	3	33.4	190	3	1.01	1.01	1.09	1.40	1.10	1.00
150	3	34.7	200	3	0.96	0.95	0.92	0.96	0.93	0.91

the sensor body is taken into account. Among the stress measuring methods reported so far is a method developed by Morooka *et al.* (1993). In this method, measurement is done outside of the pressure container, and a stress sensor is attached to a press cylinder. It was therefore necessary to correct the inside pressure of the pressure container, effects of swelling caused by the change in temperature of cylinders that penetrate pressure containers, and the friction force between side walls of pressure containers and cylinders. Also, in this method, only the average value of inner stress generated in the whole lumber could be calculated. In contrast to this, our method employs a small sensor which can be attached directly inside the processed lumber. This enables us to measure stress generated in each place separately. Also, a temperature sensor for steam pressure measurement is additionally employed, which makes it possible to accurately calculate the inner stress generated at a specific spot.

When the compression set of specimen was 66.6%, the stress per cross section caused by the press was 1.2 MPa. Inner stress during fixation process gradually decreased with time. Inner stress was completely removed at about 40 min after the temperature of compressed specimen reached 180 °C. When the inner stress was completely removed, the compressive transformation was permanently fixed. The sample treated with 180 °C steam at 3 min kept the transformed shape under high-temperature saturated steam atmosphere in the apparatus, even if the press was

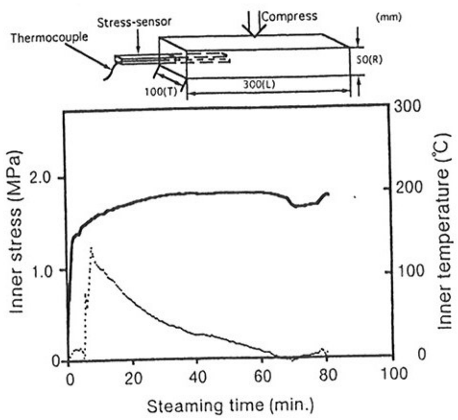

Fig. 6. Inner temperature and inner stress in wood specimen, as a function of the steaming time.
Legends: —: Inner temperature, ...: Inner stress.

liberated. This stress sensor enables us to measure the rheological behavior under high-temperature steam atmosphere to certainly establish the treating condition during the fixation process.

Thus, the transformation and fixation were successively conducted under high-temperature saturated steam atmosphere in the newly designed apparatus. The apparatus facilitates the successive treatments of softening, compressive-molding and fixation by high-temperature steam. The physical properties of the treated wood improved without destroying its excellent qualities, because no chemical was used in fixing the transformed state.

Also, regarding the recognition of the importance of the global environment which has been raised recently, one can safely argue that the steam processing, which does not do any harm to the environment of the earth and people's health, is an excellent processing method.

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