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3,788,929 METHOD FOR PLASTICIZING WOOD Jouko Huttunen, Porvoo, Finland, assignor to Neste Oy, Helsinki, Finland No Drawing. Filed June 8, 1970, Ser. No. 44,636 Claims priority, application Finland, June 10, 1969, 1,713/69 Int. Cl. B27h 1/00; C09j 5/02 2 Claims

U.S. Cl. 156-307

### ABSTRACT OF THE DISCLOSURE

Wood is treated by being initially impregnated with hydrazine. Thereupon wood is shaped in the usual manner and then excess hydrazine is removed. Hydrazine is preferably used in an aqueous solution. Wood may be heated after the impregnation. The process is suitable for treating wood veneers and various wood boards.

The present invention concerns a method for plasticiz- 20 ing wood for the purpose of its shaping. Wood that has been treated according to the method can be, for instance, bent and compressed and twisted without damage to the wood, and thereby it acquires a new, stable shape.

It is known that the bendability of wood can be in- 25creased in some degree by treating it with hot water or steam. After the wood has become saturated with moisture, it can be bent and subsequently dried under pressure. In addition to the fact that in this method prolonged treatment periods are required, treatment with hot water 30 or with steam causes in the wood physical and chemical changes which impair its characteristics. It is also a fact that the bending accomplished with the aid of such treatment is not particularly permanent.

According to another method, the wood is impregnated 35 with anhydrous, liquid ammonia or aqueous solution of ammonia, whereupon the wood can be bent into desired shape, in which shape it remains after the ammonia has been removed by evaporation.

The following explanation has been given for the mechanism of this process. Ammonia is able to penetrate not only into the amorphous regions of the cell walls but also into the crystal structure of the cellulose and into the phenolic lignin bonding substance. Ammonia breaks up some of the hydrogen bonds which are responsible for the normal rigidity of wood. Softening of the cellular structure permits the sliding of structural components past each other under effect of an external force. After evaporation of the introduced ammonia, hydrogen link- 50 ages are once more formed between the polymer chains and the wood structure becomes re-cross-linked to its former rigidity.

A great drawback of the ammonia method is the low use of refrigerating and liquefying equipment indispensable. Moreover, ammonia is poisonous and special precautions are necessary for this reason.

It has now been found that ammonia, which is such an inconvenient wood-treating agent, can be replaced with 60 hydrazine, by the aid of which wood can be plasticized, or softened, so that it can be bent into desired shape. In addition to bending, also considerable compression of the wood is rendered possible by hydrazine.

Aqueous solution of hydrazine has been found to dissolve Bjöorkman's lignin and to soften the so-called lignin-carbohydrate complex. When wood is impregnated with hydrazine, it thus appears obvious that in the wood occurs partial softening and solution of lignin and its 70 dispersal in the wood substance. Furthermore, hydrazine apparently reacts with lignin, with the result of cross link2

ages between lignin molecules, whereby the molecule size of lignin is multiplied. In likeness with ammonia, hydrazine also has an effect on cellulose.

The effect that has been discovered is employed in a method according to the present invention for plasticizing wood for purposes of shaping, and the invention is mainly characterized in that the wood is impregnated with hydrazine and that subsequent to shaping the excess hydrazine is removed.

The hydrazine may be pure or in the form of a solution, e.g. an aqueous solution. Of course, other solvents or vehicles appropriate for the purpose are equally usable. The essential thing is that the impregnating fluid contains hydrazine in sufficient amount. It is thus possible to use hydrazine solutions which have concentrations between 1 and 100 percent, most appropriately 3-15%. It should be noted that the wood to be treated may also be green timber, provided that more concentrated hydrazine solution is then used.

For the purpose of impregnating the wood with hydrazine any suitable impregnating method may be used. As a rule, in such methods various combinations of vacuum and pressure treatments are employed. Impregnation is suitably effected e.g. in a pressure vessl, in which at first a vacuum is created in order to draw the air from the pores. Hydrazine solution is then admitted into the vessel and allowed to be absorbed, possibly at first under vacuum, and then at normal pressure and/or over-pressure. Impregnation may also be effected by means of brush application.

In the impregnation treatment the wood becomes pliable and soft. The plasticity may be further increased by means of a fairly short, e.g. 10-30 minutes, heat treatment (at 80-100° C.). Subsequent to impregnation the wood can be bent and/or compressed. The bendability of the wood after impregnation is very remarkable indeed. For instance, pieces of birch wood 20 mm. in thickness (with longitudinal direction of grain) can be easily bent by hand into an arc of 180 degrees with about 3 cm. bending radius, without inflicting any damage to the wood. After the hydrazine has been evaporated, the wood retains the bent shape and subsequently resists bending considerably more strongly than untreated wood.

It is also possible after impregnation to compress the wood, whereby from it escapes hydrazine solution in which lignin has been solved. With a common manually operated screw press, the volume of the wood can be easily reduced by more than 50% in the direction perpendicular to the grain. After or during compression the volatile substances may be removed by means of heat treatment, whereby the wood acquires a new, stable shape and considerably altered characteristics.

Compression is most appropriately accomplished during heat treatment. The pressing temperature may be, for boiling point of the impregnating fluid, which makes the 55 instance, 100-200° C., suitably 150° C. The pressing and drying time may vary, depending on the desired final result, wood species, dimensions, pressing temperature, etc. As a rule about 4 hours pressing and drying time is sufficient. During the pressing and drying treatment mainly hydrazine evaporates from the wood and can be recovered by condensation. Measurements of the nitrogen content of the wood after treatment have revealed that hydrazine remains in the wood in the amount of about 2%, referred to the dry weight of the wood.

Then density (volume weight) of the wood increases considerably as a result of compression. As a rule, the density of hardwoods increases more strongly than that of softwoods.

Another consequence of the compression and heat treatment is considerable increase in hardness of the wood. For instance, the hardness of birch wood in untreated con-

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dition, and measured perpendicular to the grain, is about 200 Brinell degrees, but wood treated according to the present invention may present with a corresponding Brinell hardness higher than 2000. The hardness measured in the direction of the grain is even higher, and it may be up to about 3000 Brinell degrees.

It has further been noted that treatment according to the invention improves the dimensional stability of the wood. The dimensions change more slowly and in lesser amount in humid air than those of untreated wood. Submerged in 10water, wood according to the invention swells equally as untreated wood, though at a slower rate, but on drying it largely regains its improved hardness and strength. Also, it does not check in the course of drying upon water im-

Hydrazine impregnation and subsequent pressing and heat treatment has, as a rule, a darkening effect on the color of the wood. The change in color depends on the heating temperature and heating period employed. The color of birch becomes brown, that of alder becomes reddish brown, while pine only begins to darken after prolonged heating. It is thus possible, by varying the heating periods and temperatures, to achieve color shades simulating valuable wood species in common wood.

The wood species has not been found to impose any 2 restrictions on application of a method according to the invention. Good results are thus achieved with hardwoods

as well as softwoods.

Wood treated by a method according to the invention may be used to advantage e.g. in the furniture, ski, packing 31 case and boat industries and on the whole in any applications where bending of wood comes into question. It may also be employed in any such instances of use in which the natural hardness or strength of wood does not suffice. An important fiield of application is that of the 3. board industry, in which wood boards or veneers treated according to the method in hand may be glued onto various board materials, such as plywoods, stave boards, fibre boards, chip boards, etc. It goes without saying that the pressing and drying procedure of the veneers and 40 gluing of the laminated product may be combined in one work phase. It is also possible to apply on the treated wood material an appropriate coating, e.g. a paint or lacquer coat.

The fluid which runs off when hydrazine-impregnated wood is in compression can be recovered and, if desired, reused for the impregnation of wood. From this fluid the lignin may possibly be separated by precipitation, for instance, and the dilute hydrazine thus obtained may be made up with fresh, concentrated hydrazine. It is likewise possible to recover the hydrazine evaporated in the heat treatment and to reuse it for impregnation.

The method according to the present invention is now illustrated by the following, non-restrictive examples, which disclose some of its embodiments.

## EXAMPLE 1

A piece of room-dry birch wood (moisture content 6-7%) having the dimensions 20.4 mm, x 62.3 mm, x 580 mm. was impregnated with 15% aqueous solution of 60 hydrazine as follows. The piece of wood was first placed in a pressure cylinder, which was evacuated so that the pressure became less than 5 mm. Hg. The hydrazine solution was then admitted into the cylinder and allowed to be absorbed under normal pressure for 15 minutes. The pressure was then raised to be 5 kg./cm.2 for a period of 30 minutes.

The dimensions of the specimen after impregnation were 21.5 mm. x 68.5 mm. x 580.5 mm. It was subsequently kept in a heating chamber at 80° C. for 15 minutes. The piece of wood was then bent by hand through 180 degrees with about 3 cm. bending radius. Drying was accomplished under bending pressure at 115° C. The specimen displayed no signs of fracture. The weight of the specimen 75 of wood.

was 497.1 g. prior to impregnation, 1004.5 g. after impregation and 467.1 g. after drying.

#### EXAMPLE 2

In this example the effect of hydrazine on the compresibility and hardness of specimens prepared from birch, alder and pine wood was investigated. Impregnation was carried out as in the preceding example, using 15% aqueous solution of hydrazine. Pressing was accomplished in a conventional sodium press with 6 mm. pitch of the screw and 60 cm. length of the handle bars. The press was operated by hand. The compression time was 4 hours and compression temperature, 150° C. Tables 1 to 3 show the characteristics of birch, alder and pine wood before and after treatment. The hardnesses were measured perpendicular to the grain.

TABLE 1.—BIRCH

0 Cha	racteristics	Untreated wood	Impregnated wood	Compressed and dried wood
Len Volu Weig 5 Den	ckness, mm th, mm gth, mm ime, cm. th, g, /cm. dness, ° B	97. 0 98. 8	20. 8 52. 9 97. 0 122. 5	9. 7 44. 0 97. 0 41. 2 58. 5 1. 42 2, 000

#### TABLE 2.—ALDER

30	Charact eristi cs	Untreated wood	Impregnated wood	Compressed and dried wood
35	Thickness, mm Width, mm Length, mm Volume, cm.3 Weight, g Density, g/cm.3 Hardness, B	18. 5 48. 4 99. 4 88. 7 35. 5 0. 40 120	19. 6 52. 0 99. 8 107. 7	5. 9 45. 0 99. 5 26. 4 32. 4 1. 23 1, 520

## TABLE 3.—PINE

0	Characteristics	Untreated wood	Impregnated wood	Compressed and dried wood
	Thickness, mm	19. 8 47. 9 97. 6	20. 7 50. 9 97. 7	9. <b>3</b> 47. 1 97. 5
5	Volume, cm. <sup>3</sup>	92. 6 44. 1 0. 47	73.8	42.7 41.0 0.98
	Hardness, o B	145		790

It is seen from the tables that the greatest decrease in thickness and decrease of volume occurred in alder wood. The volumes were reduced, in the case of birch, alder and pine, by the respective amounts of 58.3, 70.2 and 59.3 percent. The highest hardness was achieved with birch, and the lowest with pine wood.

It has been found that the original density of the wood to be treated has an influence on its compressibility and increase in hardness. With the increasing original density, the compressibility and the hardness achieved by treatment according to the invention both decrease.

## **EXAMPLE 3**

In order to study the effect of concentration of the hydrazine solution, pieces of birch wood of about 20 x 127 x 117.7 mm. size were impregnated with hydrazine solutions of various concentrations. Impregnation, compression in the direction of thickness and drying were carried out as in Example 2. For purposes of comparison the same experiment was also carried out, using for impregnating fluid mere water, and 25% ammonia. Table 4 shows the thickness, density and hardness of the specimens before and after treatment. The hardnesses were measured perpendicular to the grain. The results reveal that hydrazine solution of about 5% concentration still exerts a notable effect on the compressibility and hardness

TABLE 4

	Un	Untreated wood		Impregnated and pressure- dried wood		
Impregnating substance	Thick- ness, mm.	Den- sity, g./cm.3	Hard- ness, B.	Thick- ness, mm.	Den- sity, g./cm.3	Hard. ness- ° B,
Hydrazine, 15.4%	20. 1 20. 1 20. 2 20. 1 20. 2	0, 52 0, 54 0, 60 0, 60 0, 68	200 200 200 200 200 200	7. 0 8. 2 9. 9 15. 6 12. 3	1. 42 1. 20 1. 11 0. 71 1. 04	2,000 1,800 1,300 300 700

#### **EXAMPLE 4**

The wood specimens prepared in Example 2 were transferred from laboratory conditions (20° C., 25% R.H.) into 65% relative humidity (at 20° C.). The dimensions of the specimens remained unchanged for 4 days. The weight of the specimens of alder wood increased by about 1% and that of the birch specimens by about 0.4%. The volume of the pine specimens increased about 1.5% and their weight, about 3.7%. In an experiment with identical, untreated pieces of wood the increase in volume was about 3% for pine and 2.64% for birch. The increase in weight was 5.5% for pine and 4% for birch.

#### **EXAMPLE 5**

Birch veneers of 2.7 mm. thickness were impregnated with hydrazine solution and pressure-dried as in Example 2. The thickness of the veneers after drying was 1.3 mm. Tested by hand, the veneers had far greater strength than conventional veneers of equivalent thickness. The hardness had also considerably increased (1500° B.).

### EXAMPLE 6

Bars (with 5.10 x 18.55 mm. cross section) made of birch wood treated in accordance with Example 2 were tested for ultimate bending strength according to B.S.S. 373:1957. The hydrazine concentrations employed at impregnation were 15.4% and 7.7%. For comparison, the ultimate bending strength was also determined for bars made of untreated birch wood (8.4 x 25.1 mm. cross section). The results are presented in Table 5, which also states the moduli of elasticity calculated for the test bars.

TABLE	5
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5	Impregnating substance	Ultimate bending strength, kg./cm. <sup>2</sup>	Young's modulus of elasticity, kg./cm.²	
	Hydrazine, 15.4%	5, 800 5, 620 1, 680	452, 000 400, 000 181, 000	

What is claimed is:

1. A method of laminating and shaping wood veneers comprising impregnating at least one wood veneer with hydrazine, adhesively bonding said veneer to at least one surface of an untreated wooden board, whereupon the laminated product is formed into a desired shape using heat and pressure.

2. A method of laminating and shaping wooden boards comprising impregnating at least two wooden boards with hydrazine, adhesively bonding said boards to each other, whereupon the laminated product is formed into a desired shape using heat and pressure or pressure alone.

#### References Cited

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# U.S. Cl. X.R.

117—59, 108, 116, 147; 144—254, 309 B, 327; 156—196, 221; 161—411; 264—343