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METHOD FOR TREATING CELLULOSE AND
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Upson Company, Lockport, N. Y., a corpora-
tion of New YorkNo Drawing. Application July 7, 1949,
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8 Claims. (Cl. 8—115.6)

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This invention relates to dimensionally stabilized materials of cellulose fibers, particularly cellulose structural materials, and to methods for stabilizing such materials against dimensional change caused by change in the humidity of the environment surrounding such cellulose materials. More particularly, the invention relates to the stabilization of structural cellulose fiber boards as well as wood and paper and fabrics made of cotton, linen and other cellulose materials to render them more resistant to dimensional changes resulting from variations in the ambient humidity and to improve the strength of such products.

It is well known that materials made up entirely or predominantly of cellulose fibers expand and contract with variations in humidity in the ambient atmosphere, such materials suffering an increase in their dimension upon absorption of moisture from the atmosphere and a contraction when moisture is given up to the atmosphere upon a decrease in the humidity thereof. It is also well known that in articles wherein fibers are directionally oriented, such expansion and contraction usually occurs to the greatest extent in a direction perpendicular to the predominant direction of the fibers. The present invention is, therefore, adapted particularly in preventing or minimizing the dimensional change which occurs across the fibers with change in humidity in cellulose materials, although it also reduces dimensional change in the direction of the fibers with humidity change.

Various expedients have been heretofore employed for the purpose of dimensionally stabilizing materials made up pre-dominantly of cellulose fibers as, for instance, plywood, wood boards, pulp products and combinations thereof, and solid paper boards. A degree of dimensional stabilization is obtained in the manufacture of plied or laminated articles by arranging the laminations with their fiber directions disposed angularly to one another rather than parallel. Although improvement in dimensional stabilization is obtained, the operation is laborious since it requires cutting and proper selection and assemblage of the plies.

It has also been suggested to densify the products under heavy pressure and to thereby set the cellulose fibers. Very expensive presses and extensive auxiliary equipment is required for this operation, and the product lacks low density and some of the flexibility desired for many uses of structural cellulose fiber board.

In my application Serial No. 627,966, filed

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November 10, 1945, now abandoned, of which this is a continuation-in-part, as well as in my copending application Serial No. 95,872, filed May 27, 1949, now Patent No. 2,629,701, I have disclosed that certain organic compounds having at least two hydroxyl groups in their molecules, particularly partial esters of polycarboxylic acids and polyhydric alcohols having at least one hydroxyl group in the residue derived from the polyhydric alcohol and at least one carboxyl group in the residue derived from the polycarboxylic acid, stabilize structures made up of cellulose fibers against expansion and contraction due to variation in atmospheric humidity. In my copending application Serial No. 103,527, filed July 7, 1949, now Patent No. 2,629,674, I have disclosed that polyhydric alcohols are effective stabilizing materials, and in my copending application Serial No. 103,528, filed July 7, 1949, now Patent No. 2,629,648, I have disclosed that polycarboxylic acids are also effective stabilizing materials.

In accordance with the present invention, I have found that materials made up predominantly of cellulose fibers may be wholly or partially stabilized against dimensional change by introducing into such cellulose materials certain specified chemical compounds which also appear to have a particular affinity for the cellulose fibers. Compounds which produce dimensional stabilization are organic in nature and have at least two hydroxyl groups, at least one of which is a part of a carboxyl group, and possess certain other characteristics with respect to volatility. The series of compounds possessing dimensional stabilizing characteristics are the hydroxy-carboxylic acids.

In this case, as well as in the partial esters mentioned above, at least one of the hydroxyl groups forms a part of a carboxyl group. The hydroxy-carboxylic acids are usually either soluble in water or in low molecular weight aliphatic alcohols or ketones or mixtures of these solvents. When in solvent solution, they rapidly penetrate into the fibrous structures, between the fibers and into the fiber cells, and in fact many of them rapidly penetrate such fibrous structures even in the absence of a solvent. In most cases, however, greater dimensional stability is obtained when the hydroxy carboxylic acids are employed in form of a solution for impregnating the fibrous structures. Further properties and characteristics of the stabilizing chemicals will be more fully described hereinafter.

For purposes of illustration only, the invention will be described in detail in its application to the production of dimensional stability in laminated structural cellulose fiber boards. Such products are best exemplified upon the market by the structural building panels sold under the name "Upson Board." These cellulose fiber boards are generally manufactured from so-called fiber boards, that is, a fiber sheet with a caliper greater than about 0.030 inches. These fiber boards are assembled and bonded to one another to produce a laminated or plied article having, for instance, from two to about seven plies. The resulting laminated structural cellulose fiber board occurs in standard sized panels of from $\frac{1}{8}$ inch to $\frac{3}{8}$ inch or more in thickness, and of specified length and widths. The original cellulose board is manufactured from any conventional type of cellulose pulp stock as, for instance, ground wood fiber, chemical wood fiber, rag fiber and other conventional pulp fibers and mixtures thereof. The initial cellulose board which constitutes the individual ply may be made either upon a conventional cylinder machine, as is generally the case, or may be made upon a Fourdrinier machine. It will be understood, however, that the invention is of general application to structural cellulose materials as, for instance, fiber insulation board, sound absorbing board, table top board, structural board for the interior of an airplane, and the like.

The compounds employed to effect stabilization in the structural cellulose fiber board against dimensional change induced by change in humidity may be introduced into the fibers from which the board is made, into the individual plies of the ultimate laminated structure or into the final laminated assembly itself. The choice of the place of introduction of the stabilizing compound and the manner in which it is to be introduced will be dictated by the type of fiber available and the type of structural panel to be produced.

Thus, when operating a closed board machine system wherein all water is recycled, the impregnating compound may be added to the beater or to the stock prior to paper formation, as for instance in the head chest, assuming that a stabilizing compound has been chosen which is not readily subject to hydrolytic change at the temperature and pH of the pulp suspension. Or, the impregnating compound may be added at any other point in the wet end of the machine.

Where the individual cellulose structural board is already formed, the stabilizing compounds may be introduced into the board by immersing the board in the compound or a solution thereof or by impregnating the board with a spray containing the treating compound or by applying it with padding rolls, all conventional methods of impregnation. Where a laminated board has already been formed by bonding a plurality of individual boards together, the resultant laminated article may be immersed in the stabilizing compounds or a solution thereof and the impregnated board subsequently dried. The impregnation under such circumstances will generally be desirably performed by subjecting the board to vacuum, at which time occluded gases and volatile materials are removed from the board, then permitting the impregnating solution to flow into the evacuated chamber containing the board generally placed therein in an upright position and spaced apart, whereby

the boards are enveloped in the solution which is, in effect, forced into the boards. This penetration of the solution may then be increased by relieving the vacuum and, if desired, raising the pressure above that of the atmosphere to enhance the speed and depth of penetration.

It is therefore an object of the present invention to provide a simple and inexpensive impregnating method for dimensionally stabilizing and strengthening structures made up substantially of cellulose and to produce dimensionally stabilized cellulose products.

In broad aspect, therefore, the invention comprehends the incorporation into structural cellulose fibrous materials of an hydroxy carboxylic acid or mixture of such acid whereby the usual expansion and contraction of such cellulose materials is considerably minimized by reason of change in humidity conditions in the atmosphere surrounding such materials. This stabilizing effect is dependent upon the quantity of the stabilizing compound incorporated in the fibrous cellulose structural element. Effective dimensional stabilization has been accomplished by incorporating in the fibrous material from about 2 to 50% of the stabilizing compound based upon the weight of oven dried fiber. The exact quantity to be incorporated into the cellulose material will be dictated by the type of material, the type of hydroxy carboxylic acid employed as a stabilizing compound, and the amount of the usual expansion or contraction which it is desired to remove. Thus, under certain conditions of use, the removal of so little as 12 or 15% of the normal expansion or contraction of a cellulose structural material may be suitable, while in other conditions of use, it may be desired to remove 50, 60 or 70% or more of the normal expansion or contraction encountered with a particular change in humidity conditions in the surrounding atmosphere.

The stabilizing compound may be incorporated into the cellulose fibers, whether the same be in fibrous form, unfibrated or felted fibrous form, by the employment of aqueous solutions, solutions in hydrophilic solvents, or mixtures thereof with water or in some instances may be incorporated without the employment of a solvent. However, the usual mode of incorporation will be to employ as an impregnating solution an aqueous or hydrophilic solution of the stabilizing compound.

The cellulose structural materials may be treated with the impregnated stabilizing material or solution thereof at substantially any desired temperature, although the usual impregnating temperatures will range between 20° C. and 50° C. However, temperatures as high as 100° C. may frequently be employed.

While the actual mechanism of the stabilizing action of the present invention for cellulose fibers is not fully understood, it is believed that their penetrating power and their fixation on and in the cellulose fibers is due to the particular molecular structure, that is to say, the presence of hydroxyl groups in both the cellulose and the stabilizing material.

After their incorporation in the cellulose material to be dimensionally stabilized, the stabilizing compounds show considerable resistance to removal by water and solvents, and it is believed, therefore, that probably there is some loose form of physico-chemical combination between the cellulose molecule and the stabilizing chemical. This resistance to removal of the stabilizers by

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water and solvents is quite marked, particularly if the impregnated cellulose products are heated to elevated temperatures as, for instance, between 100° C. and 200° C. It is further believed that the fixation of the stabilizing materials in and on the cellulose fiber may be due to the ability of the molecules of the stabilizing materials to react with each other, as well as with the cellulose, whereby polymerization takes place with the formation of long chain molecules of high molecular weight. The presence of free hydroxyl and carboxyl groups in the hydroxy-carboxylic acid suggests that, on heating, the hydroxy-acid reacts with the hydroxyl groups of the cellulose to modify the chemical structure thereof. It is believed that such modification of the cellulose results in increased dimensional stability of the cellulose structural material and also increases its strength and water resistance.

The fixation of the stabilizing compounds in and on the cellulose fibers can be enhanced by employing them in combination with thermosetting resins which, in their partially reacted state are soluble in the volatile, hydrophilic solvents for the stabilizers such as water, low molecular weight alcohols and ketones or mixtures thereof. The thermosetting resins, after setting, are believed to cover and protect the stabilizing materials in and on the cellulose fibers from attack by solvents. In this connection, it is further believed that the stabilizing materials penetrate farther into the cellulose fibers than the thermosetting resins, thus producing a protective coating of thermosetting resins.

The incorporation of thermosetting resins into the cellulose structure modifies to some extent the effect of the hydroxy-acid in such a way that the hardness and water resistance of the resulting cellulose fiber structures impregnated by the stabilizers are increased. The requirement of the properties determined by the ultimate use of the resulting article will guide the selection of the stabilizing material and its use either separately or jointly with a thermosetting resin.

Suitable thermosetting resins which may be employed in combination with the stabilizing material of the present invention include phenol formaldehyde, urea formaldehyde, and melamine formaldehyde, which are soluble in the volatile, hydrophilic solvents. Any other thermosetting resins which in their partially reacted state have the property of being soluble in such solvents may also be employed. The resins become insoluble and infusible upon advancement and prevent attack by water or solvents upon the stabilizing materials and reaction products thereof deposited in and on the cellulose fibers. The amount of thermosetting resin may be varied within a considerable range, for example, between 5% and 50% of thermosetting resin in the final cellulose fiber product based on the dry weight of fiber.

It will be understood that the following examples are given merely as illustrative of the invention and are not to be deemed limitative since there will be many variations of temperature, fibrous product, and composition, and concentration of impregnant suitable for use in obtaining the results of the invention. The principles of the invention are shown in the specific examples relating to dimensional stabilization of cellulose fiber board.

Example 1

Cellulose fiber board strips, measuring 0.051" x 2" x 12" and extending in their largest dimen-

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sion perpendicular to the predominating directions of the fibers were impregnated with a solution kept at 26° C. and composed of 85 parts of lactic acid and 15 parts of water. The impregnation of strips was conducted by keeping them beneath the surface of the solution for six minutes at which time a control sample showed that the solution penetrated to the center of the strip. The impregnated strips were dried at 100° C. for 20 minutes and then heated at 130° C. for 15 minutes. The strips were then measured perpendicular to the predominating direction of the fibers in the strip. Subsequently, the strips were placed into an atmosphere of 90% relative humidity and kept there until they reached their maximum expansion, whereupon their length was measured and compared to the length of an unimpregnated cellulose fiber sheet subjected in like manner to the same high humidity. The impregnated fiber board strips contained on the average 57% of lactic acid. They lost 96% of their properties of expansion and contraction with the variation of humidity in the atmosphere and absorbed only a small fraction of water within 24 hours immersion as compared to that of unimpregnated fiber board sheet treated in like manner.

A fiber board strip impregnated with a solution containing 22.5 parts of lactic acid, 3.75 parts of water and 73.75 parts of ethyl alcohol contained, after drying and heating, 14.2% of lactic acid. It lost 50% of its original ability of contracting and expanding when subjected to an atmosphere of 70% humidity. Cellulose fiber board strips impregnated with lactic acid and with a few per cent of urea-formaldehyde resin or phenol-formaldehyde resin showed, after drying and curing, improved dimensional stability, rigidity, flexural strength and water-resistance.

Example 2

A cellulose fiber board strip impregnated with a solution composed of 100 parts methyl polylactate, 50 parts isopropanol and 50 parts water contained, after drying and heating, 39% of methyl polylactate. The so impregnated board was subjected to high humidity as in Example 1. The tests indicated that 83% of contraction and expansion were removed in the treated strip and that considerable improvement in water-resistance and strength of the fiber board was attained. Other samples of boards containing 23% and 16% of methyl polylactate lost 70 and 66% of their normal contraction and expansion, respectively, induced by 90% humidity.

Example 3

A cellulose fiber board was impregnated with a solution composed of 50 parts gallic (3,4,5-trihydroxy benzoic) acid, 100 parts isopropanol, and 50 parts water. The board after drying contained 48% of gallic acid and lost 68% of contraction and expansion under varying humidity in the atmosphere. A cellulose fiber board impregnated with a solution composed of 25 parts of gallic acid, 50 parts of 50% cresol-formaldehyde aqueous solution and 100 parts of isopropanol contained 32% of active ingredients after drying at 100° C. for 20 minutes and heating for 10 minutes at 130° C. The contraction and expansion removed amounted to 47%. The tests showed that the board possessed improved water-resistance, rigidity and flexural strength.

Example 4

A fiber board strip was immersed in a 33% salicylic acid solution in isopropanol kept at 50° C. A complete impregnation was attained in 4 minutes immersion of the strip. The strip, after evaporation of solvent, contained 17% of salicylic acid. The impregnated board lost 54% of its contraction and expansion under the varying humidity in the atmosphere as described in Example 1. Strips containing 9% and 8% of salicylic acid lost 35% and 31% of their contraction and expansion, respectively. All of the impregnated boards possessed improved water-resistance.

Example 5

A cellulose fiber board was impregnated with a solution composed of 25 parts of alpha-hydroxycaproic acid, 37.5 parts of water and 37.5 parts of isopropanol. The board, after drying, contained 22% of alpha-hydroxycaproic acid. It showed a 77% loss of its contraction and expansion under varying humidity in the atmosphere as stated in Example 1. Likewise, a substantial improvement in dimensional stability, water-resistance, flexural strength and rigidity was obtained when the boards were impregnated with solutions containing various quantities of alpha-hydroxycaproic acid and of phenol-formaldehyde resin followed by drying, heating and pressing.

Example 6

A cellulose fiber board was impregnated with 20% alpha-hydroxylauric acid solution in ethanol. After drying and heating, the board contained 12% of alpha-hydroxylauric acid. The board thus treated lost 61% of its contraction and expansion under varying humidity of the atmosphere and possessed good water-resistance. A board impregnated with a solution composed of 5 parts of alpha-hydroxylauric acid, 11 parts of cresol-formaldehyde resin and 53 parts of ethanol contained, after drying and heating, 22% of active ingredients. The tests showed that nearly one-half of the original contraction and expansion had been removed (47%) and that the board possessed good water-resistance, flexural strength and rigidity.

Example 7

A cellulose fiber board was soaked for 4 minutes in a solution kept at 50° C. and composed of 16 parts of alpha-hydroxycaprylic acid, 36 parts of isopropanol and 36 parts of water. After drying, the board was found to have lost 78% of its contraction and expansion properties under varying humidity in the atmosphere. A loss of 56% of contraction and expansion was recorded in a board containing 13% of alpha-hydroxycaprylic acid and 13% of cresol-formaldehyde resin and a loss of 51% of the original contraction and expansion was found in boards containing 8% alpha-hydroxycaprylic acid and 8% of cresol-formaldehyde resin.

Example 8

Cellulose fiber boards were impregnated with solutions of various concentrations of alpha-hydroxy-myristic acid in a solvent composed of 2 parts of isopropanol and 1 part of water. After evaporation of solvents the impregnated boards containing 32%, 19%, 18% and 4% of alpha-hydroxy-myristic acid showed losses of 80%, 72%, 65% and 41% of their contraction and expansion,

respectively. The boards obtained possessed greatly improved water-resistance. Cellulose boards were impregnated with solutions containing equal quantities of alpha-hydroxy-myristic acid and cresol-formaldehyde dissolved in various quantities of a solvent composed of 2 parts isopropanol and 1 part of water. After drying and subsequent heating the boards contained 36%, 18%, 13%, 12% and 8% of active ingredients, and showed 74%, 69%, 59%, 41% and 37% loss, respectively, in their property of contracting and expanding with varying humidity in the atmosphere. A further improvement in dimensional stability was obtained when the boards were subjected to moderate pressure during their heat treatment.

Example 9

Strips of cellulose fiber boards were impregnated with a solution containing 50%, and smaller percentages, of alpha-hydroxy-butyric acid in a solvent composed of equal quantities of isopropanol and water. Impregnated strips containing after drying 34%, 20%, 12% 10% and 4% of alpha-hydroxy-butyric acid lost 76%, 66%, 48%, 43% and 18%, respectively, of their original contraction and expansion in varying humidity of the atmosphere. Tests on cellulose board impregnated with solutions of various concentrations containing equal quantities of alpha-hydroxy-butyric acid and cresol-formaldehyde resins contained 22%, 19% and 9% of active ingredients after drying and heating and showed a removal of 66%, 52% and 41% of contraction and expansion, respectively. All impregnated boards showed improvement in water-resistance, rigidity and flexural strength. A further improvement in properties of the boards containing alpha-hydroxy-butyric acid and a hydrophilic thermosetting resin, such as phenol-, urea-, and melamine-formaldehyde, was obtained when the boards were subjected to pressure during their heat treatment.

Example 10

Cellulose fiber board was impregnated with a 50% aqueous solution of d-gluconic acid. After evaporation of solvents, the board contained 37% of gluconic acid and on testing showed a loss of 61% of its original contraction and expansion in varying humidity in the atmosphere. A sample of the above board was impregnated with a solution composed of 50 parts of d-gluconic acid, 100 parts of water, 37.5 parts of water-soluble urea-formaldehyde resin and 50 parts of isopropanol. After subsequent evaporation of solvents and heating of the board, it contained 21% of active ingredients and lost more than one-half of its original property of contracting and expanding with varying humidity in the atmosphere.

Example 11

A cellulose fiber board was impregnated with a 50% aqueous solution of malic acid. After evaporation of water the board contained 61% malic acid and showed a reduction in contraction and expansion by 91% in an atmosphere of varying humidity. A cellulose fiber board impregnated with a solution containing 25 parts of malic acid, 31 parts of water-soluble cresol-formaldehyde resin, 120 parts of water, and 200 parts of isopropanol contained after evaporation of solvents and curing of the resin, 19% of active ingredients. The tests showed that 54% of original con-

traction and expansion due to varying humidity in the atmosphere was removed. The board possessed improved water-resistance, flexural strength and rigidity.

Example 12

A cellulose fiber board was impregnated with a 50% aqueous solution of tartaric acid. The solutions were subsequently diluted with water and after each dilution an additional board was impregnated with a diluted solution. The tests showed that the boards contained, after drying and heating, 60%, 32%, 10.8% and 4.2% tartaric acid and that the reductions in contraction and expansion in varying humidity of the atmosphere were 72.5%, 45%, 22.5% and 15%, respectively. A substantial dimensional stabilization of cellulose fiber board and improvement in water-resistance, flexural strength and rigidity was also obtained when cellulose fiber boards were treated in like manner, employing solutions containing tartaric acid and a water-soluble thermosetting resin.

Example 13

A cellulose fiber board was impregnated with a 50% aqueous solution of citric acid. The board after drying and heating contained 60.5% citric acid and showed a reduction in contraction and expansion in varying humidity of atmosphere by 63%. Likewise, a substantial dimensional stabilization, reduction in water-absorption and improvement in flexural strength and rigidity were obtained when more dilute solutions of citric acid alone and of citric acid in combination with water-soluble thermosetting resins were employed for impregnation.

Example 14

Cellulose fiber boards were impregnated with solutions of various concentrations containing 3-hydroxy-2-naphthoic acid dissolved in isopropanol. After subsequent drying and heating, the boards showed a substantial improvement in dimensional stabilization, flexural strength, and water-resistance. Similar improvements were also obtained when the impregnating solutions contained an alcohol-soluble cresol-formaldehyde resin in addition to the 3-hydroxy-2-naphthoic acid.

Example 15

An impregnating solution was prepared containing a mixture of hydroxycarboxylic acids and a thermoplastic resin known to the trade as Poly-Pale resin and composed largely of polymers of rosin acid, such as pimaric and abietic acids. 12-parts of alpha-hydroxylauric acid, 13.7 parts of alpha-hydroxy-caprylic acid, 24.2 parts alpha-hydroxy-myristic acid, 30.9 parts of alpha-hydroxy-stearic acid and 103.5 parts of Poly-Pale resin were dissolved in 103.5 parts of isopropanol. This solution and more dilute solutions containing the above materials in the same ratio were used for impregnation of cellulose fiber board strips. The solvents were subsequently removed by drying the strips in a ventilated oven and the strips were heated afterwards for 15 minutes at 130° C. Strips containing 29%, 20%, 13% and 10% of above active ingredients lost 46%, 39%, 36% and 26%, respectively, of their original contraction and expansion when they were subjected to varying humidity in the atmosphere. All of the strips possessed very good water-resistance, and showed improvement in flexural strength and plasticity.

Where impregnation of the fibers is attempted prior to the preparation of a fiber board, economic and operational restrictions will narrow the selection of hydroxy-carboxylic acids employed under such circumstances to those which are soluble in water. Comminuted cellulose fibers can be impregnated, however, with the stabilizing chemicals dissolved in organic solvents and structural members made therefrom show excellent dimensional stability under extremes of humidity conditions. This is shown in the following example:

Example A

An aqueous pulp suspension of a consistency of 1% was prepared containing 25% concentration of lactic acid based on solution. Sheets of fiber board were prepared from this pulp, cut to size and the expansion determined by increasing the humidity from 0% to 90%. When this expansion was compared with that of board made from another portion of the same pulp without the presence of the stabilizer, it was found that a 14.2% content of the acid in the board, based on the weight of dry fiber, eliminated 50% of the normal expansion.

The same type of results were obtained when applying a solution of the stabilizing chemicals to the wet end of the paper making machine. This operation gives somewhat greater flexibility in the choice of stabilizing compound to be employed, as compared with addition to the beater or head chest, for example, since it is entirely practicable to use organic solvent solutions of the stabilizer, for instance, a solution made of equal parts water and isopropyl alcohol and containing 25% concentration of alpha hydroxycaproic acid. When applying such a solution to the wet lap in amounts to provide 22% of acid in the board on a dry fiber basis, reductions in the normal expansion of 77% were obtained. At lower dilutions, good results were also obtained but, in many instances, operating technique will dictate the employment of relatively concentrated solutions when application is made to the wet lap.

Laminated cellulose structural fiber board may be impregnated with the dimensional stabilizer in any suitable fashion although immersion in the dimensional stabilizer or a solution thereof is recommended. In general, the temperature of the liquid in which the laminated cellulose structural fiber board is immersed will be at room temperature. Where a laminated product of an exceptional high caliper is to be impregnated, the temperature of the liquid may be elevated to facilitate penetration. The laminated board may be soaked in the impregnating solution until such time as the desired quantity of dimensional stabilizer has been absorbed by or combined in some physico-chemical manner with the cellulose.

It may be found expedient when treating laminated cellulose structural fiber boards, or other cellulose elements which are relatively rigid, to pack the same in a chamber, preferably in an upright position, having the boards spaced slightly apart to facilitate free circulation. It will also be found expedient to subject the chamber to vacuum whereby gases and other volatile materials, which interfere with free penetration of the solution into the board, are removed. Liquid containing the dimensional stabilizer is then admitted to the evacuated chamber containing the cellulose material and penetration throughout the body of the cellulose elements is facilitated. The impregnated boards are then removed from

the solution and passed through any conventional form of drier.

To summarize, the stabilizing materials in accordance with the present invention are either aromatic or aliphatic hydroxy carboxylic acids or mixtures thereof. In addition to such chemical requirements they should be soluble in all proportions in at least one volatile hydrophilic solvent as defined above and should have a boiling point at least as high as 150° C. Since solubility in volatile hydrophilic solvents depends upon several factors such as the number of hydrophilic groups, for example hydroxyl and carboxylic groups, the saturation of the compound, and arrangement of carbons as well as the number of carbons, it is impossible to more definitely specify the nature of the effective compounds by chemical characteristics. A similar situation exists as to the boiling points of the effective compounds.

Examples of effective hydroxy carboxylic acids are lactic acid; gallic acid (3,4,5-trihydroxy benzoic acid); salicylic acid; alpha-hydroxycaproic acid; alpha-hydroxylauric acid; alpha-hydroxycapric acid; alpha-hydroxycaprylic acid; alpha-hydroxymyristic acid; alpha-hydroxybutyric acid; d-gluconic acid; malic acid; tartaric acid; citric acid; 3-hydroxy-2-naphthoic acid; alpha-hydroxy-stearic acid; glycolic acid; hydracrylic acid; alpha-hydroxyisobutyric acid; alpha-hydroxyglutaric acid; beta-hydroxyglutaric acid; hydroxypivalic acid; alpha-hydroxy-dimethylacetic acid; 12-hydroxystearic acid; alpha-hydroxyvaleric acid; alpha-hydroxydecane-alpha-carboxylic acid; alpha-hydroxypalmitic acid; vinyl glycolic acid; recinoleic acid; recinstearolic acid; glyceric acid; 9,12-dihydroxystearic acid; 2,3,4-pentanetriol-carboxylic-1 acid; tartronic acid; citramalic acid; alpha-hydroxy-glutaric acid; itamalic acid; alpha-methyl-beta-ethyl malic acid; saccharic acid; dihydroxymaleic acid; desoxalic acid; mesoxalic acid; 2-hydroxyphenylacetic acid; mandelic acid; 5-hydroxy-ortho-toluic acid; melilotic acid; beta-hydroxy-beta-ethyl-alpha-phenyl acrylic acid; 6-phenylsalicylic acid; 2,3-dihydroxy-benzoic acid; resorcylic acid; 3,5-dihydroxy-ortho-toluic acid; 1,4-hydroxy-2-naphthoic acid; dihydroxytartaric acid; 3-hydroxy-1,2-phthalic acid; umbellic acid; genitissic acid; protocatechuic acid; pyrogallol carboxylic acid; phloroglucinol carboxylic acid; trihydroxy-glutaric acid; and like acids. As indicated above, the esters of low molecular weight aliphatic alcohols, for example the methyl and ethyl esters of any of the above acids, may be employed as the impregnating material, either alone or in solution in a volatile hydrophilic solvent, but in such cases the alcohol radical of the ester is driven off during the drying of the impregnated cellulose fiber so that the acid alone constitutes the actual impregnating material.

The hydroxy carboxylic acids or their polymers may also be incorporated into the fibrous articles in the form of their esters with low molecular weight alcohols. During heating and drying of the impregnated articles, however, the low molecular weight alcohols volatilize from the product, indicating that these alcohols split off either by hydrolysis or by alcoholysis, leaving an acid residue which combines with the cellulose material under treatment. In many cases, the esters of the hydroxy carboxylic acid and the low molecular weight alcohol has a boiling point below the temperature employed in heat treating the impregnated cellulose (100 to 200° C.). In such

cases, the esters may be converted, prior to impregnation, by heating in the presence of an acidic catalyst, into a polymerized ester having a boiling point above the temperature of heat treatment of the cellulose. In such cases, the low molecular weight alcohol still volatilizes during the heat treating, leaving a polymerized hydroxy carboxylic acid residue chemically combined with cellulose materials.

What is claimed is:

1. The method of stabilizing fiber wall boards consisting essentially of felted cellulose pulp fibers against expansion and contraction with changes in atmospheric humidity, which process comprises, impregnating said boards substantially throughout said boards with a solution in a volatile hydrophilic solvent of an impregnant consisting predominantly of at least one organic compound selected from the group consisting of hydroxy carboxylic acids, polymers of hydroxy carboxylic acids, and methyl and ethyl esters of hydroxy carboxylic acids and their polymers and drying the resulting impregnated structure, said compound being soluble in a volatile hydrophilic solvent and having a boiling point at least as high as 150° C.

2. The method of stabilizing fiber wall boards consisting essentially of felted cellulose pulp fibers against expansion and contraction with changes in atmospheric humidity, which process comprises, impregnating said boards substantially throughout said boards with between approximately 1% and 50% of an impregnant consisting predominantly of at least one hydroxy carboxylic acid, and drying the resulting impregnated structure said hydroxy carboxylic acid being soluble in a volatile hydrophilic solvent and having a boiling point at least as high as 150° C.

3. The method of stabilizing fiber wall boards consisting essentially of felted cellulose pulp fibers against expansion and contraction with changes in atmospheric humidity, which process comprises, impregnating said boards substantially throughout said boards with a solution in a volatile hydrophilic solvent of an impregnant consisting predominantly of at least one hydroxy carboxylic acid and drying the resulting structure, said hydroxy carboxylic acid being soluble in a volatile hydrophilic solvent and having a boiling point at least as high as 150° C., and said solution having a concentration of said acid between approximately 1 and 60%.

4. The method of stabilizing fiber wall boards consisting essentially of felted cellulose pulp fibers against expansion and contraction with changes in atmospheric humidity, which process comprises, impregnating said boards substantially throughout said boards with a mixture of a thermosetting resin and at least one organic compound selected from the group consisting of hydroxy carboxylic acids, polymers of hydroxy carboxylic acids, and ethyl and methyl esters of hydroxy carboxylic acids and their polymers, and drying the resulting impregnated structure said compound and thermosetting resin being soluble in a volatile hydrophilic solvent, said compound having a boiling point at least as high as 150° C., the amount of said thermosetting resin being between approximately 5 and 50% of said mixture.

5. As a product of manufacture, a fiber wall board consisting essentially of felted cellulose pulp fibers and containing as an impregnant substantially throughout said boards between approximately 1% and 50% of at least one organic compound selected from the group consisting of

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hydroxy carboxylic acids, polymers of hydroxy carboxylic acids, and methyl and ethyl esters of hydroxy carboxylic acids and their polymers, said compound being the predominant impregnant in said board and being soluble in a volatile hydrophilic solvent and having a boiling point at least as high as 150° C.

6. As a product of manufacture, a fiber wall board consisting essentially of felted cellulose pulp fibers and containing as an impregnant substantially throughout said boards between approximately 1% and 50% of a mixture of a thermosetting resin and at least one organic compound selected from the group consisting of hydroxy carboxylic acids, polymers of hydroxy carboxylic acids, and methyl and ethyl esters of hydroxy carboxylic acids and their polymers, said thermosetting resin and said compound being soluble in a volatile hydrophilic solvent and said compound having a boiling point at least as high as 150° C., the amount of said thermosetting resin being between approximately 5 and 50% of the total impregnant.

7. As a product of manufacture, a fiber wall board consisting essentially of felted cellulose pulp fibers and containing as an impregnant substantially throughout said boards between approximately 1% and 50% of at least one hydroxy carboxylic acid, said hydroxy carboxylic acid being the predominant impregnant in said board and being soluble in a volatile hydrophilic solvent and having a boiling point at least as high as 150° C.

8. As a product of manufacture, a fiber wall

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board consisting essentially of felted cellulose pulp fibers and containing as an impregnant substantially throughout said boards between approximately 1% and 50% of a mixture of a thermosetting resin and at least one hydroxy carboxylic acid, said thermosetting resin and said hydroxy carboxylic acid being soluble in a volatile hydrophilic solvent and said hydroxy carboxylic acid having a boiling point at least as high as 150° C., the amount of said thermosetting resin being between approximately 5 and 50% of the impregnant.

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