106-81 CROSS REFERENCE EXAMINER

Patented Aug. 31, 1954 2,687,556

UNITED STATES PATENT OFFICE

2,687,556

METHOD OF PREPARING PRODUCTS COMPRISING COMPRESSED LIGNOCELLULOSIC MATERIALS AND CHEMICALLY COMBINED SOLUBLE SILICATES

Donald F. Othmer, Coudersport, Pa., and Louis G. Ricciardi, Brooklyn, and Warren R. Smith, Crown Point, N. Y.

No Drawing. Application January 5, 1951, Serial No. 204,678

6 Claims. (Cl. 18—55)

1

This invention relates to a method of preparing bonded mixtures of lignocellulose and soluble silicates by pressing said mixtures under the application of heat, and it also relates to the bonded and pressed products produced by said method in the form of wallboard and similar materials.

Herein the term "lignocellulose" is used to describe either the liberated fibers as obtained after any grinding or defibrating means, and also the naturally occurring unliberated fibers as they exist in wood in any form including sawdust, slabs, edgings; or bagasse and other annual plant residues, and the like.

Fibers or particles of wood and annual plant material have been formed and compressed into wallboards by wet processes or methods for many years. The wet process used varies from company to company and compares closely with paper making. The raw materials (such as fiber, waterproofing agents and sizes) and the methods of production are similar. Chips from cordwood (not waste wood such as sawdust, shavings, slabs) are made into fibers of a particular mesh size by one of several methods, usually involving expensive equipment and processing steps. The fibers are washed, refined, screened and fed to a Fourdrinier machine similar to the standard paper making machine, where the board is formed as a thick continuous mat, known as "wet lap." After this "wet lap" is trimmed and cut to size, the individual pieces are placed on screens and inserted between the steam heated platens of a press and pressed at pressures of 1,000 pounds per square inch or higher, for 30 minutes or longer. The large amount of water is removed in a heating cycle of sufficient duration to dry and cure or bond the material into a firm mass. To reduce the long pressing cycle the mat may be pre-pressed first to remove much of the water or sent through a large drier. Equipment such as chippers, high pressure boilers, and washers, besides all the special handling and loading machines used in such prior art processes, are very expensive. This high cost of breaking whole wood down into its individual fibers and the attendant complications and skill required in the subsequent operations indicates that the making of wallboard by the wet process must be a very large business enterprise by itself and not something to be appended to an existing wood working industry.

2

In addition to the wet process of making wallboard, substantially as described above, there have been introduced within the past ten years or so, three different types of processes utilizing resins. These are:

(1) Dry powdered synthetic resin is mixed with substantially dry wood fiber, the moisture content of wood varying from 1–15%; and a dry forming and pressing process is used;

(2) Synthetic resin in a liquid solution or dispersion is added to relatively dry wood fiber, the moisture content of which is less than 5%; and a dry forming and pressing process is used;

(3) Synthetic resin in a liquid solution or dispersion is added to a wet slurry of wood fiber; and a wet forming process is used.

The synthetic resins used in the above processes are not only expensive, ranging up to 25 to 40 cents a pound, and therefore having a value much more than any wood waste used, but the percentage of the resins based on the wood waste used is high, since it may be from 10 to 25 percent, although lower amounts have been used with poorer results. In some cases the producer of such resin bonded boards has actually found himself in the chemical business of making the required resins or purchasing them on a large scale, rather than keeping to his own business of wood working or wood processing.

It is an object of our invention to produce a bonded product of the nature of wallboard and the like by compressing and heating lignocellulosic material of whatever origin or variety with soluble silicates.

Another object of our invention is to provide a method of producing inexpensive, water resistant, and permanently bonded products by a dry process of varying thicknesses, densities, hardnesses, shapes, and having other similar desirable properties.

These and other objectives are readily apparent from this disclosure of our invention.

According to our invention we prepare a mixture of lignocellulose material; for example, sawdust or wood flour, and soluble silicates; and we thereafter subject the prepared mixture to specific temperature and pressure ranges for definite periods of time to effect a permanent bond between the lignocellulose and the admixed lignin.

The wood used may be of the deciduous type or of the coniferous type; or it may be a lignocellu-

lose material from any of the annual agricultural products such as bagasse, coconut fibers, and the like.

The preferred size of the lignocellulose is from 16 to 30 mesh, though mesh sizes above and below this range are operable. Long fibers or thin shavings up to one inch square or larger have been used. It is desirable that the least dimension of the particle be not over about $\frac{1}{4}$".

Ground or comminuted wood prepared by any mechanical process as, for example, from an attrition mill, hammer mill, stone mill, or from any combination of a steam and mechanical disintegrating process, such as the (a) Asplund, or (b) the Masonite process, and the like, can be used.

The moisture content of the lignocelluse to be used may vary considerably, and it is preferred that the lignocellulose possess a substantially dry feel and appearance. Lignocellulose containing substantially 0 to about 50 percent moisture (based on its total weight) may be used. Thus, for example, freshly sawn wood; i. e., sawdust, or bagasse, either having a moisture content up to about 50 percent, or sawdust having the normal air-dry moisture content of about 10 percent, as well as bone-dry sawdust, are all operable and give satisfactory results in our process.

According to our invention, solutions of soluble silicates, for example, alkali metal silicates, such as sodium or potassium silicates of various grades—for example those known in the trade as Grades 33, 42 and 60—or any of the ortho- or meta- or the super-silicates, may be mixed either alone or in a somewhat diluted form with substantially dry lignocellulose particles; and this mixture is heated and pressed under control conditions herein specified to yield a hard, strong, dense, molded product.

Soluble silicates is a general term applied to a group of commercial chemical materials, the molecular structure of which is composed of different proportions of (a) sodium or potassium oxide (Na_2O or K_2O) and the like, and (b) silica (SiO_2), depending upon the molecular configuration of the alkali metal silicate.

The proportion of, for example, sodium oxide to silica, may be varied from one molecule of sodium oxide and four molecules of silica to two molecules of sodium oxide and one molecule of silica. The concentration of water may also be varied. By carefully controlling these two factors, the manufacturer is able to make various grades of alkali silicate having a wide range of physical and chemical properties.

The Grades 33, 42 and 60 are viscous solutions of high silica content. The grades having a high alkali content known as sodium meta-silicate, sodium super-silicate, and sodium ortho-silicate, are readily soluble granular solids.

It has sometimes been found advantageous to add additional lignin to increase the strength of the final or "cured" board. The lignin we use consists substantially of the non-polysaccharide constituents of wood and other lignin containing plants and is generally defined as that part of the plant which does not dissolve in a specific concentration of sulfuric acid under specified physical conditions. Lignin containing more or less cellulose is commercially available as a by-product of various paper making processes using wood or other lignocellulose from either trees or annual plants, such as (1) the soda process, and (2) the sulfate or kraft process, and (3) the sulfite process. Lignin may also be obtained from lignocellulose by using various acid hydrolysis processes. Substantially any lignin from processes separating it more or less completely from celulose may be used to bond other lignocellulose material when subjected to the temperatures and pressures for the periods of time presented hereinafter.

It is realized that modern chemistry does not have an exact comprehension of the term "lignin" and that the various commercial lignins are not chemically identical and, in fact, commercial "lignin" may be an alkali or alkaline earth salt of the organic lignin structure. Moreover, even the "lignins" obtained from the various lignicellulosic plants or trees vary slightly as to their methoxy content and other properties. The method of obtaining lignin, whether by chemical or mechanical treatment, will affect the chemical and physical properties of the lignin. Furthermore, the methoxy content of the lignin may vary in the lignin obtained from different parts of the same tree or agricultural plant. Nevertheless, all these various lignins, as prepared by any of the many known methods, are operable and are intended to be embraced within the scope of this invention and its claims.

Examples of a few operable commercial lignins are: Indulin C, a sodium salt of lignin obtained from the sulfate process; and Indulin A, substantially a "free" or "pure" lignin also obtained from the sulfate process, and both these Indulins are produced by the West Virginia Pulp and Paper Company; Meadol, a lignin obtained from the soda process and produced by the Mead Corporation; Benaloid, obtained by treating wood with high pressure steam, a product produced by the Masonite Corporation; acid hydrolysis lignin from either the Scholler Process or from the Katzen-Othmer Process (Industrial and Engineering Chemistry, vol. 34, page 314), or from any other acid wood hydrolysis process such as that of the Stora Kopparbergs Bergslags Aktiebolag of Falun, Sweden; Goulac, a lignin obtained from the sulfite process and produced by the Robeson Process Company; Silvacon, a lignin produced by the Weyerhauser Timber Company from the bark of Douglas fir; Arborite, a lignin obtained from the soda process and produced by the Howard Smith Paper Mills; and many others.

Most of the commercially available lignins are contaminated with cellulose or other polysaccharides, the amount present varying from about 20 to about 40 percent; or they are lignin salts of sodium, calcium, or other metals. Indulin A, on the other hand, is substantially a 100 percent "pure" or "free" lignin compound; and it and similar materials are obtained by acidulating with strong acids the lignin salts that are recovered from pulp and similar manufacturing processes.

We prefer to add lignin to the lignocellulose as an air-dry powder, although the moisture content of the lignin may vary widely as in the case of the lignocellulose itself, and still be usable in our invention. This lignin may come directly from a prior, wet manufacturing operation, without drying, if desired. The mesh size of the lignin powder may be important due to desirability of thorough mixing.

We may also add certain other materials to the mixture of lignocellulose, lignin and soluble silicates in order to increase the water resistance of the finished board or product. In particular, the addition of mineral, vegetable, or animal oils and fats, or petrolatum and other hydrocarbon, gives a product which is more water resistant.

In preparing wallboard and molded products according to our invention there is mixed with the lignocellulose up to 10 percent of a soluble silicate, as a concentrated solution of 50 percent solids. Added lignin may or may not be present. (Thus there is 5 percent of the soluble silicates themselves.) Powdered lignin, when employed in our process, is used in an amount varying from about 2 percent to about 50 percent, depending on the source and purity of the lignin used. (In referring to percentages in all examples the total weight of the naturally moist or air-dry lignocellulose material, plus that of the added lignin, is taken as 100%, unless otherwise specified. For example: if in a 10 pound mixture of lignocellulose material, soluble silicate and added lignin, there is present 20 percent lignin (i. e., Indulin A), and 6 percent of soluble silicate (Grade 60), this means that there is present in the mixture 2 pounds of lignin, 7.4 pounds of lignocellulose material, and 0.6 pound of soluble silicate.)

We have found in making boards of many different ratios of materials that there is an optimum amount of soluble silicate, which, when added to a particular lignocellulose fiber containing a fixed amount of lignin, gives the best results. This optimum amount of silicate, to give a board of maximum strength for the given amount of lignin added, is less with a fine mesh size of lignocellulose material than with the coarser sizes. Also, when soluble silicates are mixed with lignocellulose fibers that have a low natural lignin content, weak boards are produced as compared to those boards made from lignocellulose fibers having a very high natural lignin content (see Table II). The addition of lignin then increases the strength.

The amounts of soluble silicates which are used in our process are much too small to function as the adhesive itself in effecting the bonding action between the various lignocellulose particles, as in the use of sodium silicate as a glue. For such a glueing action per se there would be required many times as much of the soluble silicate and entirely different operating conditions.

We may dilute concentrated solutions of soluble silicates, if so desired. Use of such diluted solutions may give, in some cases, a more uniform dispersion of the soluble silicates and the lignocellulose fibers.

The mixing of the lignocellulose, soluble silicates, and lignin, if such is added, is effected by any conventional mechanical dry mixer to yield a thoroughly blended product. The blended dry mixture is then poured, or dumped, for example, into a pan having a faceplate in the bottom and sidewalls constituting a deep frame. The mixture is then covered with a wire mesh screen having about 20 wires to the inch and of a size to fit loosely in the pan. The use of this deckel arrangement for making the board may not be necessary, as boards can be made on a continuous basis or simply hot-pressed between two plates or screens. The purpose of the screen is to facilitate the escape of vapors formed during the hot pressing cycle. The pan is then placed between a pair of heated platens of a hydraulic or equivalent press and subjected to varying pressures for a specified period of time. Other methods of molding used in the art are also applicable; and in some cases it is preferred to use the screen for the bottom of the pan and the faceplate or another screen for the top.

The temperature we employ in the upper and

lower platens during the pressing operation in order to accomplish the chemical reaction which bonds the lignocellulose, the soluble silicates, and the added lignin to give a strong wallboard, may vary anywhere from 180° C. to 300° C., a range between 205° C. and 275° C. being preferred. The pressure we apply to the lignocellulose and lignin mixture also may vary within wide limits. Pressures within the range from 100 pounds per square inch to 1000 pounds per square inch are operable, though pressures within the range of 300 to 750 pounds per square inch are preferred. The time needed for hot-pressing the board may vary from 5 to 60 minutes, although 9 to 15 minutes is usually preferred. While we have found some variation between pressures, temperatures, and time to be possible; i. e., greater time may reduce the pressure used, or vice versa, we have found it necessary to have a minimum temperature of about 205° C. to obtain our desired effect. One of the unexpected advantages of our invention is the fact that the use of soluble silicates expedites the "curing" action and reduces the time cycle which would otherwise be necessary.

The following relationship between platen temperature and board temperature was found by use of thermocouples inserted in the mid-point of the loose lignocellulose-lignin-soluble silicate mixture, prior to subjecting this mixture to high temperatures and pressure, to form a board $\frac{3}{8}$ inch in thickness. Thermocouple wires were then molded into the finished board in these experiments.

Time, minutes.....	0	1	2	3	4	5	6	7	8	9
Material Temp., ° C..	25	85	135	170	170	170	194	220	245	245
Platen Temp., ° C....	240	235	238	238	238	240	240	241	245	246
Pressure in pounds per square inch....	0	100	100	200	200	300	300	400	400	600

Obviously, the final board temperature is closely related and substantially the same as the final platen temperature.

In order to demonstrate the nature of the changes taking place during the hot-pressing process, we have conducted a series of experiments whereby we analyze for the soluble silicate after mixing with the lignocellulose, and then also after making boards from the mixture. Table I reports these results and shows that an actual chemical change takes place during the making of boards according to our process from lignocellulose and soluble silicates, with or without the addition of lignin.

The high temperature, in the range of 205° C. to 250° C., effects some chemical reaction between the soluble silicates present and the components of the wood, and since very little of the soluble silicate appears in the board after being heated to this temperature, it has reacted with the other materials and thus then is formed a new chemical product, firmly bonded together. While the exact chemical changes are unknown, the obvious chemical change of the soluble silicate into an insoluble condition and the tremendous physical changes of the board itself demonstrates that an important chemical change has taken place during this heating process. The cellulose which, in any case, shows little or no bonding action and is relatively inert, has probably not been influenced chemically; and we refer to this chemical change as an "activation" of the lignin to make this serve again as the binder for bonding the cellulose fibers together, even as in the original wood.

Table I

Composition	Conditions for Pressing			Modulus of Rupture or Flexural Tests, p. s. i.	Percent Soluble Silicates as Sodium Silicate
	Temp., ° C.	Pressure, p. s. i.	Time, Min.		
1. 30 mesh white pine wood flour		Unpressed			No soluble silicates found.
2. 30 mesh white pine wood flour, 2% sodium silicate, 6% lignin.		do			1.8.
3. 30 mesh white pine wood flour, 2% sodium silicate, 6% lignin.	25	580	11		1.88.
4. 30 mesh white pine wood flour, 2% sodium silicate.	25	580	11		1.85.
5. 30 mesh white pine wood flour, 2% sodium silicate.	180	580	11	1,000 to 1,200	1.8.
6. 30 mesh white pine wood flour, 2% sodium silicate.	220	580	11	1,500 to 2,000	1.8.
7. 30 mesh white pine wood flour, 2% sodium silicate.	245	580	11	3,000 to 4,000	0.15.
8. 30 mesh white pine wood flour, 2% sodium silicate, 6% lignin.	245	580	11	4,500 to 5,500	0.15.

The percent sodium silicate in all cases is based on the solid sodium silicate present in a 50% aqueous solution of this material.

The method of analysis for determining soluble silicates was sensitive within one part of soluble silicate in 10,000 parts of solution.

The soluble alkali silicates are believed to enhance the flow of lignin associated with the lignocellulose so that, under the influence of heat and pressure, the lignin is activated by this chemical interaction of the silicates; and this "activated" lignin bonds the various lignocellulose fibers together to produce a strong, dense, board.

Numerous examples may be given showing the properties of wallboard made under variation of the conditions of our process. Table II presents the properties of only a few of the resulting wall boards produced from mixtures containing lignocellulose, soluble silicates, and added lignin. The independent variables that were considered in the wallboards reported in Table II are as follows:

(1) Wood species
(2) Mesh size of material
(3) Moisture content
(4) Temperature
(5) Pressure
(6) Time
(7) Lignin used
(8) Percentage of added lignin
(9) Percent and type of soluble silicates
(10) Quantity of initial charge

The dependent variables that were considered in Table II are as follows:

(1) Flexural strength
(2) Hardness
(3) Density
(4) Water absorption
(5) Swelling after immersion in water

In Table II we have listed the properties only of those boards made from substantially dry lignocellulose particles or fibers and those which, while containing up to 50% moisture, appear to be dry. This is because we have preferred to operate our process using a dry forming and pressing technique because of the relative cheapness and simplicity of the equipment required for such a dry process; and because of the economy in pressing and the shorter pressing cycle which is possible because the large amount of water of the wet process does not have to be handled. However, we have found that the addition of soluble silicates alone or with added lignin to lignocellulose material for making wallboard and the like molded products may also be applied to the wet process. Also, where for particular reasons, it is desired to use a wet process, we have found that the addition of soluble silicates, alone or with added lignin, to the stock or slurry, results in improvements in the board as shown by the data of Table II, which data are, however, taken from the dry process. For example: if a slurry of white pine, 30 mesh, fibrous material is pressed into a mat and then inserted between two hot platens, the temperature of the platens being 250° C. and pressed at a pressure of 600 pounds per square inch for 12 minutes, the final board will have a flexural strength of approximately 4000 pounds per square inch. On the other hand, if 6 percent lignin, for example, Indulin A, and 6 percent of a soluble silicate is added to a slurry of white pine, 30 mesh, fibrous material prior to its being formed into a mat, and this mixture of added lignin and lignocellulose material is then compressed into a cured board at a temperature of 250° C., a pressure of 600 pounds per square inch, and for a time of 12 minutes, the flexural strength of the cured board will be approximately 5500 pounds per square inch or higher. The soluble silicates dissolved in the water forming the slurry may be recovered for reuse of that water with the next batch—or otherwise—so that loss does not occur.

As can be seen from Table II, the addition of lignin in any form gives an improved modulus of rupture as well as an improved surface hardness, although very satisfactory boards may be made without its use. This invention obviously embraces the activation of the native lignin of the lignocellulose, through the chemical action taking place with the soluble silicates.

In the hydrolysis of wood (or other lignocellulose material), the cellulose is made soluble and removed. Thus by partially hydrolyzing wood, to any desired extent, the ratio of cellulose to lignin may be controlled. We have found that hydrolyzed lignocellulose (e. g., from the Katzen-Othmer process) may be used directly with soluble silicates, and without the addition of other lignin, since the lignin-cellulose ratio may then be correct for our process.

Table II

Board No.	Lignocellulose		Kind of Lignin and Percent of Total Charge	Moisture of Lignocellulose (Percent)	Sodium Silicate and Water Added (Percent)	Molding Temperature, ° C.
	Species	Mesh Size				
1	White Pine	30	Indulin A 6%	8–9	Grade 60 6%, Water 6%	250
2	do	30	do	8–9	Grade 60 2%, Water 2%	225
3	do	30	Indulin A 2%	8–9	Grade 60 10%, Water 10%	250
4	do	30	Indulin A 20%	8–9	Grade 60 6%, Water 6%	205
5	do	30	Indulin A 6%	8–9	do	250
6	do	30	Indulin A 15%	8–9	do	300
7	Bagasse	16	Indulin A 6%	8–15	do	250
8	Eucalyptus	Fibrous	do	8–15	do	250
9	Maple	16	do	8–15	do	250
10	Yellow Pine	Fibrous	do	8–15	do	250
11	Gum Poplar	do	do	8–15	do	250
12	Douglas Fir	16	do	8–15	do	250
13	Redwood	Fibrous	do	8–15	do	250
14	S. Afr. Wattle	16	do	8–15	do	250
15	Mangle	16	do	8–15	do	250
16	White Pine	30	do	8–9	Grade 33 6%, Water 6%	250
17	do	30	do	8–9	Grade 42 6%, Water 6%	250
18	do	30	do	8–9	Grade 52 6%, Water 6%	250
19	do	30	do	8–9	Sodium Metasilicate 6%, Water 6%	250
20	do	30	do	8–9	Sodium Metasilicate 2%, Water 2%	250
21	do	30	do	8–9	Sodium Supersilicate 10%, Water 10%	250
22	do	30	do	8–9	Sodium Orthosilicate 6%, Water 6%	250
23	do	30	Scholler 14%	8–9	Grade 60 6%, Water 6%	250
24	do	30	Hydro. Wood 50% (Katzen-Othmer).	8–9	do	250
25	do	30	Goulac 12	8–9	do	250
26	do	30	Nat. Sou. Lignin 8%	8–9	do	250
27	do	30	Meadol MWS 10%	8–9	do	250
28	do	30	Indulin C 10%	8–9	do	250
29	do	30	Arborite 9%	8–9	do	250
30	do	30	Benaloid 16%	8–9	do	250
31	do	30	Silvacon 12%	8–9	do	250
32	do	30	Indulin A 6%	0	do	250
33	do	30	do	5	do	250
34	do	30	do	30	do	250
35	do	30	do	50	do	250
36	do	30	None	8–9	Grade 33 6%, Water 6%	250
37	do	30	do	8–9	Grade 42 6%, Water 6%	250
38	do	30	do	8–9	Grade 52 6%, Water 6%	250
39	do	30	do	8–9	Supersilicate 10%, Water 10%	250
40	do	30	do	8–9	Orthosilicate 6%, Water 6%	250
41	do	30	do	8–9	Metasilicate 2%, Water 2%	250
42	Maple	30	Hydrolyzed Wood 84%	3–5	Grade 60 10%, Water 6%	250

Board No.	Final Pressure, p. s. i.	Pressing Time, Min.	Thickness of Molded Wallboard, Inches	Specific Gravity	Modulus of Rupture, p. s. i.	Surface Hardness, Barcol Scale	Immersion 24 Hours in Water	
							Inc. in Wt. Percent	Inc. in Thickness (Percent)
1	580	12	.185	1.10	5,960	70	24.1	9.8
2	1,000	35	.346	1.14	5,960	72	21.5	3.6
3	800	8	.235	.94	2,340	45	40.1	15.6
4	100	60	.856	1.05	4,000	60	27.1	11.8
5	750	5	.130	.93	2,610	45	34.9	14.1
6	300	30	.561	1.09	5,080	68	28.1	11.0
7	580	11	.185	1.00	4,100	45	35.1	11.2
8	580	13	.236	1.15	5,390	40	33.1	14.1
9	580	13	.218	1.16	3,800	50	33.5	13.9
10	580	12	.150	1.05	5,700	60	29.1	11.8
11	580	11	.135	1.12	5,690	65	28.1	10.7
12	580	12	.165	.99	4,500	52	30.8	10.1
13	580	8	.150	1.01	6,000	78	15.1	4.2
14	580	20	.340	1.12	3,950	55	32.1	9.5
15	580	15	.220	1.18	4,210	55	30.1	9.2
16	580	11	.193	1.15	5,250	65	29.1	12.2
17	580	11	.185	1.13	5,380	65	28.5	12.0
18	580	11	.202	1.13	5,360	65	28.5	12.2
19	580	11	.210	1.13	5,760	70	27.1	8.8
20	580	11	.195	1.13	5,050	65	29.1	9.8
21	580	11	.215	1.15	5,960	75	28.1	9.1
22	580	11	.202	1.16	5,800	72	29.2	11.1
23	580	11	.202	1.10	4,650	55	40.1	20.2
24	580	15	.253	1.16	5,000	65	20.1	10.5
25	580	12	.195	1.15	4,460	60	38.5	18.9
26	580	12	.183	1.15	5,210	65	33.3	12.8
27	580	12	.200	1.15	4,410	50	39.9	16.1
28	580	12	.190	1.13	4,652	55	37.1	14.2
29	580	12	.190	1.15	5,790	65	29.1	9
30	580	12	.215	1.16	5,310	60	35.5	15.5
31	580	12	.198	1.16	5,810	68	28.1	8.2
32	750	11	.175	1.18	5,920	75	24.1	5.7
33	750	11	.183	1.15	5,300	70	29.1	7.8
34	750	11	.181	1.18	6,200	75	23.5	4.8
35	750	11	.181	1.19	6,350	75	22.5	3.9
36	580	11	.190	1.12	3,450	45	39.1	16.2
37	580	11	.193	1.10	3,250	45	39.1	17.2
38	580	11	.195	1.10	3,450	45	38.1	15.2
39	580	11	.190	1.13	4,310	55	32.5	11.1
40	580	11	.198	1.14	4,710	60	37.1	13.5
41	580	11	.210	1.05	2,560	38	50.1	20.5
42	580	15	.195	1.18	6,250	72	20.1	4.3

We may also add fireproofing agents or insecticides to the lignocellulose fibers to make a superior product possessing fireproof or insecticidal qualities

In our discussion above we have shown the advantages which we have found in the use of combinations of our preferred materials with lignocellulose in the production of wallboard made directly from sawdust or from other lignocellulosic granules or fibers in either a dry or a wet process. We have also found that we can use the same combinations of materials and under the above specified conditions of pressure, time, and temperature which are required for the making of wallboard in the production of other shapes and articles wherein similar physical properties are desired as in wallboard. It is especially desirable to make such objects when the cost of the raw material is important, because the materials we use are very cheap. Particularly useful are our methods and the products resulting in those cases where the cross-sectional dimension of the final object is not too great, and wherein it is possible to mold the material with some provision for the escape of gases, and wherein the shape of the final object is not too complicated and does not require too great an ability of the raw material to flow in filling the mold. In this case our preferred combination of materials acts substantially as a molding powder of peculiar properties and of comparative cheapness to other similar molding powders which have been suggested and are used in the art.

We have also found that our combination of materials may be added directly as an upper or lower layer or both in a final application on the surface of an assembly of veneers for the making of plywood. Thus, a smooth, fine surface, similar to that which is produced in making wallboard by our preferred process, is obtained on the surface of the plywood, regardless of any irregularities or imperfections in the plywood itself. Besides eliminating the objectionable grain in some of the rotary cut veneers, irregularities such as knots, etc., may be removed from the surface finish by the application of a mixture of lignocellulosic granules or fibers, together with the other materials, as specified above. In some cases we have found that the same application of a fine lignocellulosic material, together with added materials, as specified above, may be applied to rough lumber or even planed lumber which has imperfections therein; thus a fine surface is achieved immediately without imperfections, and without surfracing operations which remove and make thinner the wood. The imperfections in the lumber or in the plywood are then finally out of sight beneath a hard dense surface with a very fine and smooth pattern, depending upon the particular type and size of the wood fibers or granules used. In this use we often prefer a fine granular or fibrous material such as from 30 to 80 or even finer mesh, in order to give a thin coating of smooth surface qualities.

This invention is a continuation-in-part of Serial Number 747,130, filed April 9, 1947.

While many embodiments of our invention have been presented above, it is to be clearly understood that the invention is not to be limited to these embodiments.

We claim:

1. The dry method of preparing dense, water-resistant, molded products comprising mixing substantially dry lignin powder with substantially dry lignocellulose particles and adding an aqueous solution of alkali metal silicate in an amount sufficient to activate the lignin at an elevated temperature and pressure to the resulting lignocellulose-lignin mixture and thereafter subjecting the resulting composition to said elevated temperature and pressure sufficient to activate the lignin and thereby effect a strong bond among the lignocellulose particles.

2. The dry method of claim 1, wherein an effective amount of lignin up to 50 percent of the weight of the lignocellulose is added to the lignocellulose, said lignin varying in moisture content between substantially zero percent to about 50 percent moisture.

3. The dry method of preparing dense, water-resistant wallboards consisting of mixing pulverulent lignin with lignocellulose particles of the group consisting of wood sawdust, wood shavings, and bagasse particles, adding thereto a quantity of aqueous alkali metal silicate of the group consisting of ortho-silicate and meta-silicate in an amount sufficient to activate the lignin of the resulting composition at an elevated temperature and pressure, and thereafter subjecting the said composition to said elevated temperatures and pressure to activate said lignin by said silicate whereby a strong bond is effected among the lignocellulose particles.

4. The dry method of preparing dense, water-resistant wallboard consisting of adding from one to 20 percent lignin to lignocellulose and adding thereto aqueous alkali metal silicate in an amount sufficient to activate the lignin of the resulting composition at an elevated temperature and pressure and thereafter subjecting the resulting composition to said elevated temperatures and pressure whereby the lignin is activated and effects a bond among the lignocellulose particles.

5. The process of claim 4, wherein a temperature in the range of 205° C. to 300° C. is employed to activate the lignin and a range of pressures of 100 to 1000 p. s. i. is used to mold the resulting composition for a time period up to 60 minutes.

6. The process of claim 5, wherein the temperature range is 205° C. to 275° C., the pressure range is 300 to 750 p. s. i. and the time of pressing is from 8 to 15 minutes.

References Cited in the file of this patent

UNITED STATES PATENTS

Number	Name	Date
293,785	Ravenscroft	Feb. 19, 1884
629,600	Platz	July 25, 1899
1,168,831	Skalla	Jan. 18, 1916
1,532,908	Lowe	Apr. 7, 1925
1,564,706	Oelhafen	Dec. 8, 1925
1,793,667	Baldwin	Feb. 24, 1931