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STEREOTYPE MAT

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This invention relates to stereotype mats, and particularly to improvements in such mats and the method of making them whereby their molding and casting properties are improved.

It is the object of the invention to provide a stereotype mat which is adapted to receive a deep, sharp, and clear-cut impression from the mold with a minimum of fracture and which will give a large number of casts which are excellent reproductions of the mold.

It is a further object of the invention to provide a stereotype mat of improved flexibility, whereby when the molten metal is poured against it, it is adapted to flatten itself readily against the supporting surface under the weight of the cast metal and correct any irregularities of its shape due to warping which may have occurred in the operations preceding the casting.

The stereotype mats which are in common use are composed of cellulose fibers, such as rag stock, cotton linters and liberated, or purified, wood pulp fibers, usually sulphite pulp fibers, and usually contain a filler such as clay. Examples of such stereotype mats are those which are disclosed in my United States Patent 1,524,155, granted January 27, 1925, and Patent No. 1,720,243, granted July 9, 1929.

A stereotype mat is molded while moistened or humidified with water, and plasticity is of utmost importance in order that the mat may take readily, a full and exact impression of the form without the application of excessive pressure. Cellulose fibers possess only limited plasticity. The impression taken by the mat is due largely to the compression and stretch undergone by the fibers. The characteristics of the fibers in these respects, therefore, place limitations upon the quality of the results which are obtained in the use of the mats.

In addition, it is important that the mat be flexible when heated for the reason that if the mat warps or buckles during the drying operation following molding and is not sufficiently flexible to conform with the supporting surface during the subsequent casting operation, the cast plate will not have the necessary uniform or level printing surface. My improved mat is flexible under the heat of the molten stereotype metal, and is adequately stiff when cold.

Attempts have been made in the past to improve the plasticity of stereotype mats by incorporating in them such materials as artificial and natural resins, but these efforts have not met with a substantial degree of success, chiefly because such mats tend to adhere to the cast metal.

In accordance with the present invention, the discovery has been made that if lignin obtained from a natural lignocellulosic material is intimately mixed with cellulose fibers of the character described heretofore, and the mixture formed into a stereotype mat, the plasticity of the resulting mat is increased, both in the cold and at elevated temperatures, and the flexibility at elevated temperatures is increased. The lignin may be in the form of finely divided solid particles thereof, or in the form of a thermoplastic lignocellulose such as is obtained by the method which is described in my co-pending joint application Serial No. 134,272, now Patent 2,196,277, in which a natural lignocellulosic material, such as wood, corncobs, straw, bagasse, cornstalks, etc., is subjected to a cooking operation with water under temperature and pressure conditions such that an appreciable portion of the lignocellulose, particularly the hemicelluloses, is rendered water-soluble. The lignocellulosic material, containing the lignin thereof in situ, is rendered thermoplastic by such cooking operation.

The product of the cooking operation may be washed with water to free it of the water-solubles, ground to the proper size, and then mixed with cellulose fibers and formed into a sheet suitable for use as a stereotype mat from a water suspension in the well-known manner. Alternatively, the cooked mass may be added directly to a water suspension of cellulose fibers and beaten to the desired size, the water-solubles passing into solution during this operation. The resulting intimate mixture of cellulose fibers and particles of thermoplastic lignocellulose is then made into stereotype mat sheets.

Prior to the cooking operation, the natural lignocellulosic material is comminuted. The time, temperature and pressure used during the cooking operation may vary within limits, temperatures between 125° C. and 225° C., and pressures up to 200 pounds per square inch being suitable. Cooking periods of from 15 minutes to one hour are usually adequate.

During the cooking operation, small amounts of acetic and formic acids are formed from the lignocellulose, and if desired, these acids may be neutralized by the inclusion prior to the cooking operation of sufficient alkali, such as sodium carbonate or hydroxide, to continuously neutralize the acids as they are produced. An excess of alkali is avoided.

When the procedure outlined above is followed, the lignin of the lignocellulose retains its nat-

ural location on the cellulose fibers and coats the fibers uniformly.

A hydrolyzed lignocellulose may be used, such as is obtained by a mild acid hydrolysis of natural lignocellulose. Such hydrolyzed lignocellulose is thermoplastic.

Where lignin is to be used instead of thermoplastic lignocellulose, the lignin may be obtained in any suitable manner, such as by a strong acid hydrolysis of lignocellulose, or by precipitation from a solution thereof obtained by extracting lignocellulose with an alkaline solution (black liquor). It may also be obtained by evaporation of the solvent from a solution thereof in an organic solvent for lignin. The lignin is comminuted to the desired particle size and then mixed intimately with the cellulose fibers and the mixture formed into sheets suitable for stereotype mats. The sheets range in thickness from approximately 0.018 to 0.035 of an inch.

Particle sizes for both lignin and thermoplastic lignocellulose which pass through a 100-mesh per inch screen have been found to be suitable, but it is preferred to use particle sizes which pass through a 200-mesh per inch screen.

The lignin, or the thermoplastic lignocellulose, is added to the cellulose fibers which, preferably, have previously been beaten to the desired degree of fineness. The proportion of lignin in the lignocellulose varies with the different kinds of lignocellulose, usually being about 40% to 45% of the weight of the lignocellulose. Varying proportions of the thermoplastic lignocellulosic material may be mixed with the fibers. Good results have been obtained with 5% to 60%, based upon the weight of the dry sheet. This represents a sheet containing up to approximately 27% of lignin. Optimum results have been obtained with approximately 25% to 45% of lignocellulose. For the lignin as such, the range may be from approximately 5% to 50%, and the proportions for optimum results are approximately 10% to 40%.

The resulting mat is essentially fibrous in character and is flexible. It has a smooth impression-receiving surface without fibers projecting therefrom. It absorbs moisture readily and uniformly and in the moistened condition is highly plastic at ordinary temperature, and is adapted to receive a deep and exact impression of the mold. Cold molding is the usual procedure, and the mat of this invention possesses markedly improved cold molding properties. Hydrolyzed lignocellulose in the moist condition is highly plastic. The mat also possesses improved hot molding properties.

There are two widely used methods for molding stereotype mats. In the first, the mat, containing 10% to 50% of moisture is placed on the form and molded by direct hydraulic pressure, or by rolling the mat under pressure. The form, without disturbing the molded mat, is transferred to a drying apparatus in which the mat is dried by the simultaneous application of heat and pressure, by means of heated platens. Temperatures of from approximately 160° C. to 170° C. are used during this drying operation. The mat is then separated from the form and is ready for the casting operation.

The second method is the one which is commonly used in newspaper plants. In this method, the mat, having 10% to 20% of moisture, is molded by direct pressure or by a roller. The mat is then separated from the form and is re-

moved to a dryer, commonly called a "scorcher," where it is heated until dry.

In both of the above described methods of molding, warping or buckling of the mat frequently occurs either during the molding operation or the drying operation.

The improved mat of this invention possesses sufficient flexibility that during the subsequent casting operation all such irregularities are straightened out under the weight of the cast metal. The lignin, or the thermoplastic lignocellulosic material, as the case may be, is rendered plastic by the heat imparted by the molten cast metal and, as a result, the mat becomes flexible and conforms readily to its supporting surface. The mat maintains its shape during the cooling of the metal, so that when the cast plate is removed, all irregularities are absent and the plate has a uniform, level printing surface. The cast plate separates readily from the mat.

In addition, the thermoplasticity of the lignin, or of the thermoplastic lignocellulosic material, causes the latter to assume a condition of fusion or plastic flow during the casting operation, and causes an intimate cementing or welding together of the thermoplastic material and the cellulose fibers. This renders it possible to make a large number of casts from the stereotype mat without impairing its surface characteristics.

The following are specific examples of the preparation of the improved stereotype mat of this invention. The first two and the fifth relate to compositions in which lignin as such is employed as the thermoplastic material, and the third and fourth are compositions in which thermoplastic lignocellulosic material is used for that purpose.

Example No. 1

Black liquor, obtained from the digestion of a natural lignocellulosic material, such as wood, by the soda or sulphate pulping process is acidified sufficiently with sulphuric acid to cause the precipitation of the lignin contained in the black liquor. Heating of the solution may be resorted to if desired to produce coagulation of the lignin. After the lignin has been precipitated, it is separated, as by filtering, from the black liquor and washed until it is substantially free from acid. The lignin may then be dried and comminuted to 200 mesh particle size and added to the cellulose fibers to make the mixture suitable for stereotype mats. Alternatively, the lignin may be allowed to remain in the moist condition and added to the cellulose fibers in the beating engine. The proportion of lignin should be approximately 15% based on the weight of the sheet. The lignin and fibers are mixed thoroughly in water and a stereotype mat is formed from the suspension in the usual manner. The mat may be dried or adjusted to the proper moisture content for use.

Example No. 2

A natural lignocellulosic material, such as wood, is treated with a concentrated sulphuric acid solution of approximately 72% sulphuric acid strength at room temperature until the carbohydrates are substantially all dissolved. The lignin remains undissolved. The solution is then diluted with water until the acid content is reduced to approximately 10%. It is then heated sufficiently to coagulate the lignin. After the lignin has been coagulated, it is separated from the solution by filtration and washed with water until substantially free of acid. This lignin is

then comminuted and mixed with cellulose fibers, and the mixture is formed into a stereotype mat in the manner described above in connection with Example 1.

Example No. 3

Comminuted wood is placed in a digester, preferably of the rotating type, and subjected to the action of steam at a pressure of 200 pounds per square inch (gauge). After the interior of the digester has attained the temperature of the steam, approximately 198° C., the steaming operation is continued for approximately 30 minutes to complete the dissolving action upon the water-solubles, and to effect a mild hydrolysis of the wood. At the end of the cooking operation about 30% of the wood has been converted into water-soluble products. These are chiefly carbohydrates. The proportion of lignin in the remainder has been correspondingly increased.

The cooked lignocellulosic product is separated from the cooking liquor, washed with water to remove the water-solubles, ground to 200 mesh size, and is then added to a water suspension of cellulose fibers which have previously been beaten to the desired extent. Approximately 25% by weight of the lignocellulose, based upon the weight of the final sheet, is added. The suspension is mixed thoroughly in a beater and is then felted into sheets of stereotype mat. The remainder of the method is carried out as described in connection with the previous examples.

Example No. 4

A natural lignocellulosic material, such as wood, is cooked in an excess of an aqueous solution containing 3% of sulphuric acid at a steam pressure of approximately 125 pounds per square inch. The cooking is continued for approximately 30 minutes after the digester has attained the temperature of the steam. In this method the hemicelluloses of the natural lignocellulosic material are rendered soluble, while the cellulose fibers and the lignin remain substantially undissolved.

After the cooking operation has been completed, the lignocellulose is separated from the cooking liquor, washed with water to remove the water-solubles, comminuted, mixed with cellulose fibers, and the remainder of the method is carried out in the same manner as described in connection with Example No. 3.

Example No. 5

Lignin may be used which has been derived from a solution thereof in an organic solvent. Such lignin appears to have a lower softening point than lignin obtained from an acid or an alkaline solution thereof, and is therefore, well suited to produce the desired plasticity in stereotype mats. To obtain such lignin, the thermoplastic lignocellulose product obtained from the cooking operations of Examples 3 and 4 may be extracted with an organic solvent. Some of the organic solvents which are suitable for the purpose are ethyl and butyl alcohols, acetone, dioxane, and chloroform.

This extraction may be carried out at room temperature, or in a closed container at temperatures up to 180° C. After the extraction has been completed, the solvent may be separated from the lignin by distillation. The lignin may then be reduced to the desired particle size and mixed with the cellulose fibers and the stereo-

type mat may be formed in the manner outlined in connection with Examples Nos. 1 and 2.

It is understood, that fillers for the stereotype mats, such as clay, may be added to the mixture in accordance with common practice. Such fillers may be omitted if desired.

Phenols or aromatic amines may be incorporated with the lignin, or thermoplastic lignocellulosic material. These substances have the property of increasing the heat plasticity of the thermoplastic material and their use is advantageous for that reason. These are preferably incorporated with the thermoplastic material before the latter is mixed with the cellulose fibers, but they may be added to the mixture. A mixture of a phenol and an aromatic amine, or amide, may be used, such mixture imparting greater benefit than the use of either a phenol or amine alone. Phenols which may be used are phenol, nitrophenol, o-, m-, and p-cresol, alpha- and beta naphthol, catechol, pyrogallol, and p-aminophenol. The amines and amides which may be used are aniline, o-, m-, and p-toluidine, alpha- and beta-naphthylamine, p-toluosulphonamide, phenyl ureas, phenyl guanidine. The amount of these phenols, amides and amines which may be used varies over a wide range, usually about 1% to 10% of the weight of the lignin or thermoplastic lignocellulose being desirable.

Since the mats prior to molding contain a certain amount of moisture, an efficient method of introducing a lignin plasticizer is to moisten the dry mat to the desired extent with an aqueous saturated solution of those plasticizers having an appreciable solubility in water, e. g., aniline and phenol. The plasticizers having very slight solubility in water may be incorporated in the mat stock in the beater.

The heating of the lignin or the lignocellulose, dry or admixed with water, with an aromatic amine or a phenol to form a condensation product, falls within the spirit of this invention. This operation is preferably carried out before the lignin or lignocellulose is mixed with the fibers.

I claim:

1. A felted plastic stereotype mat consisting essentially of an intimate mixture of cellulose fibers and thermoplastic lignin, and containing approximately 10% to 50% of water, based on the weight of said stereotype mat.

2. A felted stereotype mat consisting essentially of an intimate mixture of cellulose fibers and from approximately 5% to 60%, based on the weight of said stereotype mat, of a thermoplastic lignocellulose.

3. A felted fibrous stereotype mat having thermoplastic lignin distributed among the fibers thereof.

4. A felted fibrous stereotype mat having incorporated among the fibers thereof and in intimate mixture therewith particles of lignin.

5. A felted fibrous stereotype mat having distributed among the fibers thereof particles of thermoplastic lignocellulose.

6. A sheet-form fibrous stereotype mat, said mat being composed of felted cellulose fibers and having thermoplastic lignin distributed among and in intimate mixture with the fibers thereof.

7. A felted fibrous stereotype mat composed of cellulose fibers and having thermoplastic lignin and an inorganic filler distributed among and in intimate mixture with the fibers thereof.

8. A felted fibrous stereotype mat composed of cellulose fibers and having thermoplastic lignin,

in an amount approximately 5% to 50% of the weight of said mat, distributed among and in intimate mixture with the fibers thereof.

9. A felted fibrous stereotype mat composed of cellulose fibers and having thermoplastic lignin, in an amount up to approximately 30% of the weight of said mat, distributed among and in intimate mixture with the fibers thereof.

10. A felted fibrous stereotype mat composed of cellulose fibers and having particles of thermoplastic lignin distributed among and in intimate mixture with the fibers thereof, said particles being of a size which will pass through a 100 mesh per inch screen.

11. A felted fibrous stereotype mat composed of cellulose fibers and having distributed among and in intimate mixture with the fibers thereof particles of a natural lignocellulose which has been water cooked at a temperature of approximately 125° C. to 225° C. to render said lignocellulose thermoplastic.

12. A felted fibrous stereotype mat composed of cellulose fibers and having distributed among and in intimate mixture with the fibers thereof

particles of a natural lignocellulose which has been water cooked at a temperature of approximately 125° C. to 225° C. to render said lignocellulose thermoplastic and thereafter washed with water to remove the water solubles therefrom.

13. A felted fibrous stereotype mat composed of cellulose fibers and having distributed among and in intimate mixture with the fibers thereof particles of a thermoplastic lignocellulose, said particles being of a size which will pass through a 100 mesh per inch screen.

14. A felted fibrous stereotype mat composed of cellulose fibers and having distributed among and in intimate mixture with the fibers thereof particles of an acid-hydrolyzed natural lignocellulose.

15. The method of improving the molding and casting properties of a stereotype mat composed of felted cellulose fibers which comprises incorporating particles of thermoplastic lignocellulose among the fibers of said mat.

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