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(54) Title: METHOD OF MAKING A FINISHED PRODUCT FROM A FEEDSTOCK, A PORTLAND CEMENT, AND A THERMOSETTING RESIN

(57) Abstract: A method of making a finished product includes the steps of (a) providing a dry feedstock selected from the group consisting of particles or fibres of a lignocellulosic material; particles of an expanded mineral; particles of a foamed set hydraulic binder; particles of an undensified silica fume or a mixture of two or more thereof; (b) mixing the feedstock with: (i) a suitable amount of a thermosetting resin in finely divided dry powder form; and (ii) a suitable amount of Portland cement in finely divided dry powder form, to give a starting material; (c) subjecting the starting material of step (b) to suitable conditions of temperature and pressure to cause the thermosetting resin to set to form a cohesive product; (d) providing to the cohesive product of step (c) water in an amount sufficient for the complete hydration of the Portland cement, while simultaneously or sequentially but before the Portland cement has set, introducing carbon dioxide into the cohesive product of step (c) to assist in the setting of the Portland cement; and (e) allowing the Portland cement to set to form the finished product. The finished product may, for example, be a board.

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**METHOD OF MAKING A FINISHED PRODUCT FROM A FEEDSTOCK,
PORTLAND CEMENT, AND A THERMOSETTING RESIN**

BACKGROUND OF THE INVENTION

THIS invention relates to a method of making a finished product such as a board, from a feedstock which is a suitable particulate or fibrous material, a binder, and a thermosetting resin.

Various types of particle board are known, manufactured from particles of a suitable feedstock, e.g lignocellulosic particles or fibres, bound together with either a hydraulic binder or a thermosetting resin or a combination thereof.

There are certain problems associated with the manufacture of particle boards where the binder comprises or includes a hydraulic binder such as Portland cement, including the time taken for the hydration of the Portland cement to proceed to completion, the possible formation of undesirable products, and the like.

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Research has been done by the Wood Research Institute of Kyoto University, Japan and the Nichiha Company Limited of Japan on accelerating the curing of Portland cement bound particle board, as well as improving the mechanical properties of the particle board, by post treating the pre-pressed and initially set particle board with carbon dioxide, either in super critical or gaseous form. One disadvantage of this process is that before such a treatment may be carried out, either the particle board must be pre-set which implies that the chemical reactions in the conventional hydration of the Portland cement have already commenced, or alternatively the particle board must in some way be pressed and clamped during treatment.

SUMMARY OF THE INVENTION

According to a first aspect of the invention there is provided a method of making a finished product including the steps of:

- (a) providing a dry feedstock selected from the group consisting of particles or fibres of a lignocellulosic material; particles of an expanded mineral; particles of a foamed set hydraulic binder; particles of an undensified silica fume; or a mixture of two or more thereof;
- (b) mixing the feedstock with:
 - (i) a suitable amount of a thermosetting resin in finely divided dry powder form; and
 - (ii) a suitable amount of Portland cement in finely divided dry powder form,to give a starting material;
- (c) subjecting the starting material of step (b) to suitable conditions of temperature and pressure to cause the thermosetting resin to set to form a cohesive product;
- (d) providing to the cohesive product of step (c) water in an amount sufficient for the complete hydration of the Portland cement, while simultaneously

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- or sequentially but before the Portland cement has set, introducing carbon dioxide into the cohesive product of step (c) to assist in the setting of the Portland cement; and
- (e) allowing the Portland cement to set to form the finished product.

According to a second aspect of the invention there is provided a finished product made by the method described above.

DESCRIPTION OF EMBODIMENTS

The crux of the invention is a method of making a finished product from a feedstock, Portland cement and a thermosetting resin.

The feedstock may be particles or fibres of a lignocellulosic material.

A lignocellulosic material refers to any plant material emanating from the photosynthetic phenomenon.

The particles or fibres of the lignocellulosic material are preferably finely divided. For example, the fibres may be provided as unifibres or bundles of a small number of unifibres. In other words, the lignocellulosic material is broken down into single fibres or bundles of a small number of fibres. Examples of suitable finely divided lignocellulosic fibres are paper mill sludge or recycled paper waste, or the fibres used in the production of medium density or high density fibre board. Alternatively, the fibres may be sourced from agriculture such as sisal, kenaf, hemp or the like, with a length of from 5 to 12 mm inclusive, preferably from 2 to 6 mm inclusive.

When the lignocellulosic material is provided in particle form, the particles preferably have a size of from 0.1 mm to 2 mm diameter inclusive.

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An important factor in the choice of the lignocellulosic material is that it should not swell on water wetting, i.e during impregnation of the cohesive product with the water to hydrate the Portland cement.

The feedstock may also be particles of an expanded mineral such as exfoliated vermiculite, expanded perlite, an expanded clay, an alumina, a pumice or diatomaceous earth.

The vermiculite particles preferably have a particle size of 2 mm diameter or smaller, down to a particle size of 100 micron diameter.

The expanded perlite particles preferably have a particle size of 2 mm diameter or smaller, down to a particle size of 100 micron diameter.

The other expanded minerals which may be used must also have a particle size of 2 mm diameter or smaller, down to a particle size of 100 micron diameter.

The feedstock may also be particles of a foamed set hydraulic binder, e.g a foamed hydraulic cement aggregate which is then milled or ground into particles having a size of 2 mm diameter or smaller, down to a particle size of 100 micron diameter.

The feedstock may also be particles of an undensified silica fume, having a particle size of from 5000 m²/kg to 20000 m²/kg.

The feedstock may also be a mixture of two or more of the various feedstocks mentioned above.

For example, where a finished product is to be classified as non-combustible, the organic percentage of the finished product should be below 7,5%.

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Assuming that the thermosetting resin, which is regarded as an organic material, is present in an amount of 5%, then the maximum amount of lignocellulosic material which can be present is 2,5%. The remainder of the feedstock may then be comprised of exfoliated vermiculite particles or expanded perlite particles or a mixture thereof.

The preferred feedstocks are lignocellulosic particles or fibres, exfoliated vermiculite particles, expanded perlite particles, particles of undensified silica fume, or a mixture of two or more thereof.

The feedstock is mixed firstly with a suitable amount of a thermosetting resin in finely divided dry powder form.

The thermosetting resin is preferably a novolac phenol formaldehyde resin, i.e a resin in which the molar ratio of phenol to formaldehyde exceeds parity, which is used with a suitable catalyst.

An example of a suitable catalyst for use with a novolac phenol formaldehyde resin is hexamethylene tetramine.

An example of a suitable novolac phenol formaldehyde resin catalyst combination is a two stage resin with a hexamethylene tetramine content of between 6 and 14%, with a hot plate gel time at 150°C of between 40 and 120 seconds, with a flow in mm at 125°C of between 30 and 75 mm, and with a particle size sieve analysis percentage retained on a 200 mesh screen of a maximum of 2%. Examples are the PRP Resins of South Africa, code Varcum 7608 which may be used as a modifier for a slow curing phenolic system such as Varcum 3337, or which may be used on its own.

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As indicated, the thermosetting resin must be provided in finely divided dry powder form. By finely divided there is meant that the thermosetting resin must have a particle size of 98% passing a 200 mesh screen.

The thermosetting resin is preferably used in an amount of from 1% to 20% inclusive by mass of the starting material, i.e the feedstock, the thermosetting resin and the Portland cement, more preferably in an amount of from 2% to 5% inclusive by mass of the starting material.

The feedstock is also mixed with a suitable amount of a binder which is Portland cement.

The Portland cement may include a pozzolan which is ground granulated blast surface slag, fly ash, or silica fume, or a mixture of two or more thereof.

The Portland cement (and the pozzolan when present) must also be provided in finely divided dry powder form. By finely divided there is meant that the Portland cement (and pozzolan) must have a particle size of from 300 to 1000 μm inclusive.

The Portland cement may be used in an amount of from 20% to 2000% inclusive of the Portland cement to 100% of the feedstock, i.e the mass ratio of the Portland cement to the feedstock is from 1 to 5 to 20 to 1. The preferred range is from 1 to 5 to 3 to 1.

As the product of step (c) is cohesive, i.e is held or stuck together, there is no need to press or clamp the cohesive product to hold it together during step (d).

There may also be added into the mixture of the feedstock, the thermosetting resin and the Portland cement additional components including inorganic filler

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materials in particle or fibre form, such as for example wollastonite, 200 mesh or finer, and synthetic fibres, or mixtures thereof.

It is to be noted that all the components of the starting material are used and mixed in dry form to ensure that the Portland cement does not set before step (e).

After step (b) in which the feedstock is mixed with the thermosetting resin and the Portland cement, the starting material so produced is subjected to suitable conditions of temperature and pressure to cause the thermosetting resin to set to form a cohesive product.

For example, the starting material may be pressed at a temperature of up to 200°C preferably in a range of from 120° to 200°C and a pressure of from 20 to 70kg/m² inclusive, preferably 20 to 30kg/m² inclusive, for a time of up to 15 seconds/mm thickness, or usually about 8 seconds/mm thickness, to form the cohesive product.

In this step, the thermosetting resin sets to form the cohesive product, while the Portland cement remains unset.

In step (d) of the method of the invention, there is provided to the cohesive product of step (c), water in an amount sufficient for the complete hydration of the Portland cement while simultaneously or sequentially introducing carbon dioxide into the cohesive product of step (c).

This may be achieved in various ways.

Firstly there may be added to the cohesive product of step (c) a carbonation reagent selected from the group consisting of carbon dioxide; and an aqueous

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solution of a compound or compounds which release carbon dioxide; under conditions that cause release of carbon dioxide in the cohesive product.

When the carbonation reagent is carbon dioxide at super critical, near super critical or high carbon dioxide density conditions, then when this carbonation reagent is added to the cohesive product of step (c), any calcium hydroxide in the Portland cement reacts with the carbon dioxide to form a carbonate plus free water. This water then can be used for the complete hydration of the Portland cement. Thus in these circumstances, it may not be necessary to add additional water.

Alternatively water may be added as such, or may form a solvent or diluent for the carbonation reagent.

In this case the carbonation reagent may be carbon dioxide, e.g gaseous carbon dioxide, liquid carbon dioxide, super critical or near super critical carbon dioxide, or a solution of carbon dioxide in a solvent.

The carbonation reagent may also be a compound which releases carbon dioxide, in solution.

The carbonation reagent may also be a combination in solution of compounds which together, under suitable conditions, release carbon dioxide

For example, the carbonation reagent may be a carbonate or bicarbonate, in solution, such as for example carbonic acid or sodium bicarbonate.

The carbonation agent may be for example a potassium carbonate of formula $K_2CO_3 \cdot 2H_2O$ or sodium carbonate or sal soda of formula $Na_2CO_3 \cdot 10H_2O$.

Step (d) is carried out under conditions that cause release of carbon dioxide from the carbonation reagent in the cohesive product. When the carbonation reagent is for example a compound which releases carbon dioxide at a particular temperature, then the cohesive product must be raised to this temperature or higher in step (d). For example, sal soda releases carbon dioxide at a temperature of about 35°C, and thus, when using sal soda as the carbonation reagent, the cohesive product should be raised to a temperature of 35°C or higher in step (d).

Other conditions which cause the release of carbon dioxide may simply be the presence of water, as with an acid/base couple, or pressure, or a combination of temperature and pressure or the like.

When using carbon dioxide itself, then it is simply required that the carbon dioxide penetrate the cohesive product.

Secondly there may be added to the starting material in step (b) a carbonation reagent which is a dry compound or dry compounds which release carbon dioxide under specified conditions. This compound or these compounds thus form part of the cohesive product of step (c).

In step (d) the cohesive product of step (c) is then subjected to the specified conditions to cause the release of carbon dioxide in the cohesive product.

For example the compound may be sodium carbonate or potassium carbonate, in which case the addition of water in step (d), optionally together with the application of heat, leads to release of carbon dioxide.

Alternatively the compounds may be an acid/base couple, in which case the addition of water in step (d) leads to release of carbon dioxide.

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The introduction of carbon dioxide assists in the setting of the Portland cement. It may accelerate the setting of the Portland cement and/or may control the microstructure of the Portland cement in the finished product.

For example the carbon dioxide propagates the rapid formation of hydration products such as calcium silicate hydrate and calcium carbonate, in a time span of from between 10 and 60 minutes, at ambient temperatures.

The use of carbon dioxide is particularly efficacious where the feedstock contains components which may interfere with the curing of the Portland cement. For example, where the feedstock contains a lignocellulosic material, the water soluble extracts of the lignocellulosic material may inhibit the setting of Portland cement. The inclusion of carbon dioxide serves to lessen or overcome this problem.

The preferred carbonation reagents are either gaseous carbon dioxide or super critical carbon dioxide. When the carbonation reagent is super critical carbon dioxide, it may be forced into the cohesive product at elevated pressures of up to 100 atmospheres. Likewise, when the carbonation reagent is gaseous carbon dioxide, it may be forced into cohesive product at elevated pressures of preferably no less than 30 atmospheres.

For example, the cohesive product from step (c) may be placed in a pressure cylinder and subjected to a vacuum for up to 15 minutes. Thereafter, the cylinder is flooded with water which is then subjected to a positive pressure of typically 6 atmospheres for a further 15 minutes, resulting in water impregnation right through the cohesive product. The cylinder is then drained of water and the cohesive product is subjected to a post vacuum in order to extract from the cohesive product all excess water which "boils" from the cohesive product, as well as to extract any interstitial air. The excess water is drained from the cylinder and compressed carbon dioxide is introduced into the

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cylinder which penetrates through the cohesive product. The compressed gas feedline may be left open for a period of 30 to 60 minutes in order to allow the various reactions to go to completion. The product is then withdrawn from the cylinder and allowed to dry to provide the finished product in which the Portland cement has substantially set.

Additional hydrophobisation of the finished product may be imposed by including in the water of hydration, a silicone micro emulsion concentrate, dispersed in water at a level of from 0,2 to 8% inclusive by mass, more generally at a level of from 1% to 3% inclusive by mass, thereby forming silicone mesophases. Examples of suitable silicone micro emulsions are Wacker BS1306, Wacker BS1000 and Wacker 1311.

Examples of the invention will now be given.

Example 1

Portland cement	35%
Dicalite 471 (Chemserve perlite)	30%
RSU.Exfoliated vermiculite (Chemserve vermiculite)	30%
Code 602 novolac resin (Schenectady)	4%
Dolanit PAN fibre	1%

(All percentages by mass).

The above composition was mixed and then pressed at 180°C to 12 mm thickness and a density of 900 kg/m³ for 2 minutes, to form a cohesive product. This product was subsequently impregnated with water to a 20% uptake by mass. After a 12 hour humidity drying time, the product was impregnated with super critical carbon dioxide at 70 atm pressure for 8 minutes at 25°C, to give the finished product

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Example 2

Portland cement	50%
Undensified silica fume	10%
Exfoliated vermiculite with a particle size spectrum of 100 to 300 micron	35%
Novolac resin	5%
(All percentages by mass)	

The above composition was mixed and then pressed at 180°C to a dry density of 1250 kg/m³ at a thickness of 12 mm to form a cohesive product. This cohesive product was subsequently impregnated in a pressure cylinder with a 50% solution by mass in water of sal soda at 30°C.

The resulting product was then humidity dried to 8% by mass of water and then warmed, in the pressure cylinder to a temperature of 60°C, to cause release of carbon dioxide. The product was retained in the pressure cylinder until the cure was complete. The carbon dioxide reacted with various cement components, particularly hydroxides of calcium which include not only calcium hydroxide but also other calcareous hydrated cement components such as calcium silicate hydrate, to give the finished product.

CLAIMS

- 1 A method of making a finished product including the steps of:
 - (a) providing a dry feedstock selected from the group consisting of particles or fibres of a lignocellulosic material; particles of an expanded mineral; particles of a foamed set hydraulic binder; particles of an undensified silica fume; or a mixture of two or more thereof;
 - (b) mixing the feedstock with:
 - (i) a suitable amount of a thermosetting resin in finely divided dry powder form; and
 - (ii) a suitable amount of Portland cement in finely divided dry powder form,to give a starting material;
 - (c) subjecting the starting material of step (b) to suitable conditions of temperature and pressure to cause the thermosetting resin to set to form a cohesive product;
 - (d) providing to the cohesive product of step (c) water in an amount sufficient for the complete hydration of the Portland cement, while simultaneously or sequentially but before the Portland cement has set, introducing carbon dioxide into the cohesive product of step (c) to assist in the setting of the Portland cement; and
 - (e) allowing the Portland cement to set to form the finished product.
- 2 A method according to claim 1 wherein the feedstock is selected from the group consisting of particles or fibres of a lignocellulosic material; exfoliated vermiculite particles; expanded perlite particles; particles of undensified silica fume; and a mixture of two or more thereof.

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- 3 A method according to claim 1 or claim 2 wherein the thermosetting resin is a novolac phenol formaldehyde resin.
- 4 A method according to any one of claims 1 to 3 wherein the thermosetting resin is used in an amount of from 1% to 20% inclusive by mass of the combined mass of the feedstock, the thermosetting resin and the Portland cement.
- 5 A method according to any one of claims 1 to 4 wherein the Portland cement is used in an amount of from 20% to 2000% inclusive of the Portland cement to 100% of the feedstock.
- 6 A method according to any one of claims 1 to 5 wherein in step (c) the starting material of step (b) is pressed at a temperature of up to 200°C and a pressure of from 20 to 70 kg/m² to form the cohesive product.
- 7 A method according to any one of claims 1 to 6 wherein in step (d) there is added to the cohesive product of step (c) a carbonation reagent selected from the group consisting of carbon dioxide; and an aqueous solution of a compound or compounds which release carbon dioxide; under conditions that cause release of carbon dioxide in the cohesive product.
- 8 A method according to claim 7 wherein the carbonation reagent is selected from the group consisting of super critical carbon dioxide, near super critical carbon dioxide, and high density carbon dioxide which is impregnated into the cohesive product of step (c) under pressure and which provides to the cohesive product of step (c) water in an amount sufficient for the complete hydration of the Portland cement.

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- 9 A method according to claim 7 wherein in step (d) there is added to the cohesive product of step (a) water in an amount sufficient for the complete hydration of the Portland cement while simultaneously or sequentially but before the Portland cement has set adding to the cohesive product of step (c) a carbonation reagent which is selected from the group consisting of gaseous carbon dioxide, liquid carbon dioxide, super critical carbon dioxide, or a solution of carbon dioxide in a solvent.
- 10 A method according to claim 7 wherein in step (d) there is added to the cohesive product of step (c) a carbonation reagent selected from the group consisting of a compound which releases carbon dioxide; and compounds which together release carbon dioxide; as a solution in water in an amount sufficient for the complete hydration of the Portland cement.
- 11 A method according to claim 10 wherein the carbonation reagent is selected from the group consisting of potassium carbonate and sodium carbonate.
- 12 A method according to any one of claims 1 to 6 wherein in step (b) there is added to the starting material a carbonation reagent which is a dry compound or dry compounds which release carbon dioxide under specified conditions, and in step (d) subjecting the cohesive product of step (c) to the specified conditions to cause the release of carbon dioxide in the cohesive product.
- 13 A method according to claim 12 wherein the carbonation reagent is selected from the group consisting of potassium carbonate and sodium carbonate.
- 14 A finished product made by the method of any one of claims 1 to 13.