Title: SHAPED CONCRETE BODY

Abstract: The invention relates to a method for preparing a concrete shaped body from a moist concrete. The method comprises providing a mixture comprising cement, water, sand, gravel and starch thereby forming the moist concrete; and shaping the concrete into the desired shape. The invention further relates to a concrete mixture and a concrete shaped body, comprising starch.
Title: Shaped concrete body

The invention relates to a method for preparing a concrete shaped body from a moist concrete, to moist concrete and to a concrete body prepared from moist concrete.

Formed concrete bodies are made by filling moulds with concrete. Examples of such concrete bodies are blocks, slabs (paving stones), curbstones, etc. After removal of the moulds the shape of the concrete bodies has to be retained. The removal of the mould is at such a short time after filling that setting of cement cannot play a significant role in retaining the shape. Therefore, retaining the shape of the bodies is a critical step in the production process.

Concrete products are in direct competition with other forms of construction materials, such as metals, plastics, wood, asbestos-cement etc.

In order to obtain sufficient retention of the shape, normally concrete with a very low amount of water is used, so-called earth moist concrete. This concrete has a much higher so-called green strength than concrete with a high amount of water, such as self-compacting concrete. The green strength is the strength of fresh concrete before setting of cement plays a role. A low amount of water also ensures that the concrete does not stick to the moulds or the press.

However, the necessity to use a low amount of water has several disadvantages. In particular, filling the moulds becomes difficult. Known methodologies require a combination of extensive pressing and vibration is necessary in order to achieve sufficient filling. A bad filling by insufficient pressing or vibration leads to a larger porosity of the concrete, a lower strength, a lower wear resistance of the concrete, less resistance against frost/thaw (especially relevant to concrete blocks) and defects of the shape or surface of the concrete bodies. Another disadvantage is incomplete hydration of the cement present in the concrete which lowers the strength and can
make an increase of the amount of cement necessary.

Filling the moulds requires a lot of energy and wear and tear of the production machinery. Furthermore, this process results in a lot of noise. Another process to fill the moulds is extrusion. For both production techniques (pressing and compaction, extrusion) a good flowing of the concrete and reduction of friction is important. By increasing the amount of water or addition of a plasticizer friction can be reduced and flow can be improved. However, the use of these measures is limited, because of the demand of retaining the shape.

EP-A-0 955 277 describes a self-compacting concrete comprising a polysurfactant-like additive, which provides the concrete with desirable properties making it very easy to pour or place.

DE-A-102 09 812 is directed to the use of water soluble polysaccharide derivatives as dispersion agent in particular for mortar and concrete. The polysaccharide derivatives consist of partial hydrolysed celluloses and/or starches, wherein the starches preferably have a polymerisation degree of 40 to 500.

US-A-5 575 840 describes an additive for aiding the water retention of cementitious and adhesive compositions. The additive comprises a mixture of cold-water-soluble, unmodified starch and a polysaccharide. The additive is free of modified starches.

US-A-3 847 630 describes a sprayable water-permeable porous concrete with a water-cement ratio of between 0.32 and 0.48, which concrete comprises a macromolecular compound.

US-A-4 487 864 describes the use of a modified carbohydrate polymer comprising a polyacrylamide as a water retention aid in various cementitious compositions.

US-B-6 387 171 is directed to a lightweight concrete composition comprising granular starch.

It is an object of the invention to provide a novel method for
preparing a concrete shaped body from a moist concrete, that can be used as an alternative to known methods.

It is a further object to provide a method for preparing a concrete shaped body from a moist concrete, wherein the shape of the concrete body is retained better, also in the absence of a supporting means such as a mould, than in a comparable known method.

An additional object is to provide a method for preparing a concrete shaped body from a moist concrete with good filling properties for filling a mould with the concrete, whilst maintaining sufficient retaining properties of the shape, also if the mould is removed before the concrete has substantially set.

Another object is to provide a method which allows the use of a higher amount of water than in conventional methodology making use of moist concrete, whilst maintaining good shape retaining properties of the concrete, before substantial setting of the concrete.

Further objects will be apparent from the description and/or claims.

It has now been found that it is possible to realise one or more of these objects by using a moist concrete comprising a specific polymer.

Accordingly, the present invention relates to a method for preparing a concrete shaped body from a moist concrete, comprising
- providing a mixture comprising cement, water and at least partially modified starch thereby forming the moist concrete; and
- shaping the concrete into the desired shape, preferably by moulding,

wherein the moist concrete is a concrete according to compaction class C0, C1, or C2 as defined in EN 206-1.

In addition to cement, water and at least partially modified starch, an aggregate (sand and/or gravel) is usually present in the moist concrete.

The term “at least partially modified starch” herein means that at least part of the starch which is used in the mixture for forming the moist
concrete is modified. The modified starch can be any modified starch, such as chemically, biologically and/or physically modified starch.

The starch according to the invention is preferably soluble in cold water.

Concrete can be classified in terms of the compaction class. In the context of this invention, the term “moist concrete” is in particular used for a concrete having a compaction class C0, C1 or C2 as defined in EN 206-1. Accordingly, the degree of compactability (C) of a moist concrete is in particular at least 1.11. Preferably, C is more than 1.20, in particular more than 1.25 (i.e. class C0 or C1). According to EN 206-1, the consistence of earth moist concrete, i.e. concrete with a low water content designed to be compacted in special processes, is not classified. However, NEN 5950 and NEN 8005 classify earth moist concrete in compaction class C1.

Good results have been achieved with a concrete, wherein C is less than 1.46. Particularly good results have been achieved with an earth moist concrete (i.e. class C1 or C0), in particular an earth moist concrete of class C1 (i.e. having a C value in the range of 1.26-1.45).

C is defined herein as \( h_0/(h_0-s) \), wherein \( h_0 \) is the initial height of a column of concrete, \( s \) the difference between \( h_0 \) and \( h \) wherein \( h \) is the height after sagging (see figure 1). C may be determined according to NEN 5958.

The exact water to cement ratio of the concrete generally depends on for instance the application (the product being made), the raw materials including the use of a (super)plasticizer, and the production equipment.

In a method according to the invention, sufficient dimension stability of the shaped body is usually reached shortly after shaping (e.g. in a mould), typically within a few seconds to a couple of minutes.

Starch and starch derivatives and combinations of these with other polymers have been found to improve the production process and/or the concrete properties in a number of ways.

The internal friction of the concrete and the friction between the
concrete and production equipment may be decreased so that filling the moulds may improve. As a consequence pressing and vibration may be reduced, which leads to less wear and tear of the production machinery. The reduced internal friction can also lead to increased homogeneity of the concrete and/or an increased density of the concrete. An increased density may be desired for providing a concrete with a relatively low pore volume and/or a small average pore size. An increased density may contribute to improved thaw/frost resistance, a low efflorescence of salts (in particular free lime), a better wear resistance of the concrete and/or a higher colour intensity of an article made from the concrete.

In an embodiment, the use of starch or a combination of starch with one or more other polymers increases the green strength of the concrete which contributes to retaining the shape of the concrete body. This leads to a reduction in B-grade production (products with a relatively low quality, yet still useful for less demanding applications), and thus to an increase in A-grade production (high quality products; usually not showing a substantial amount of visible deformations).

Further, a method according to the invention allows the use of a higher amount of water and/or plasticizer, whilst maintaining good stability and/or strength. Increase of the amount of water is thought to lead to a more complete cement hydration. This makes it possible to increase the strength of the concrete and/or offers the opportunity to decrease the amount of cement in the formulation. A better cement hydration may also change the pore size distribution and lead to a decreased pore volume. As a consequence the thaw/frost resistance of the concrete may be improved because the tendency of water from the environment to penetrate into the concrete is reduced.

Moreover, the surface characteristics of the shaped bodies may be improved because of a more favourable rheology. Smooth surfaces with a low number of surface defects are obtainable in accordance with the invention.

As cement, any cement suitable for preparing concrete may be
used, in particular any cement as defined in NEN-EN 206-1 and more
specifically in EN197-1.

As aggregate (meaning sand and/or gravel) any aggregate suitable
for preparing concrete may be used as, in particular any aggregate as defined
in NEN-EN 206-1 and more specifically in prEN 12620:2000 for normal and
heavy-weight aggregate and prEN 13055-1:1997 for light-weight aggregates.

The starch may in principle be any starch, including modified
starches, such as chemically, biologically and/or physically modified starches.

In principle a starch from any starch may be used. Suitably, a
starch may be selected from potato, tapioca, maize and wheat, including
starch from varieties having a high amylopectin content, such as amylopectin
potato starch and waxy maize starch. In particular, good results have been
achieved with a potato starch.

Preferably, at least part of the starch is a chemically modified
starch. Such starches are commercially available or may be made by a
method known in the art. For an overview of known derivatisation reactions,
reference is made to O.B. Wurzburg (Ed.), "Modified Starches: Properties and
optimizes the increase of the green strength and minimizes the effect on the
hydration of cement by which a delay of setting is prevented.

Suitable chemically modified starches include starch ethers, starch
esters and crosslinked starches. Preferred examples include starch ethers
and crosslinked starch, including cross-linked starch ethers.

Very suitable is a starch that is hydroxyalkylated and/or
carboxyalkylated, in particular a starch that is hydroxypropylated and/or
carboxymethylated. In a preferred embodiment such hydroxyalkylated and/or
carboxyalkylated starch is cross-linked.

The degree of modification may be chosen within wide limits.

In an embodiment, the degree of hydroxyalkylation (DS) of the
starch is between 0.01 and 2.0 hydroxyalkylations per monosaccharide unit.
In a particular embodiment the degree of hydroxyalkylation is in the range of 0.05-0.8.

In an embodiment, the degree of carboxyalkylation of the starch (DS) is between 0.01 and 0.5 carboxyalkylations per monosaccharide unit.

A high degree of polymerisation (DP) of the polysaccharide is preferred with respect to the degradation of the starch. Therefore, the DP is preferably 100 or more, more preferably 500 or more, and most preferably 1000 or more. It is believed that a high degree of polymerization has a positive effect on the increase of the green strength.

The starch may be cross-linked with any cross-linking agent known in the art, including epichlorohydrin, linear dicarboxylic acid anhydrides, citric acid acrolein, phosphorus oxychloride, adipic/acetic mixed acid anhydrides, dichloro acetic acid, trimetaphosphate salts, acrolein, polyphosphates (e.g. hexametaphosphate), biphenyl compounds, N,N-dimethylol-imidzolidon-2 (DMEU), formaldehyde, cyanuric chloride, diisocyanates, dimethylolethylene urea and divinyl sulfones.

The cross-linking reaction may be carried out under any conditions which are known to be suitable for this type of reaction.

Crosslinking may very suitably be done to a cross-linking degree (DS) of about 0.0001 to about 0.05, in particular with epichlorohydrine as the crosslinking agent.

The starch ester can e.g. suitably be prepared as described in EP A-1 225 268 (in particular page 5, lines 25-28). Thus, it may be prepared by reacting an aqueous suspension of the starch with an alkali (such as NaOH) at a pH in the range of 6-9.5, usually at a temperature of about 20-40 °C.

In an embodiment, the degree of esterification of the starch (DS) is between 0.02 and 2 ester moieties per monosaccharide unit.

The amount of starch in the concrete may be chosen in wide limits.

In practice, an amount of starch in the range of 0.01 to 1.0 wt.%, based upon
the weight of cement, usually suffices. An amount in the range of about 0.05
to 0.3 % has been found advantageous in terms of good performance to cost
ratio.

In addition to starch, cement, water and usually aggregate, one or
more components may be added to the concrete.

In particular, good results have been achieved with a method
wherein besides starch, at least one second (organic) polymer is present, in
particular a polysaccharide, preferably at least one polysaccharide selected
from the group consisting of cellulose, xanthane gum, tamarind, welan gum
and guar.

The further polysaccharide is preferably chemically modified, more
preferably a polysaccharide ether. Optionally the further polysaccharide is
cross-linked.

With respect to the nature of the modifications the further
polysaccharide may generally be modified in a manner as indicated for the
starch. A preferred further polysaccharide is an alkylpolysaccharide - in
particular a alkylcellulose, more in particular a methylcellulose - which has a
DS of about 1-2. More preferably the alkylpolysaccharide (alkylcellulose) is
additionally (hydroxyl)alkylated, in particular with a DS of 0.05 to 0.5.

It has been found that compared to the use of starch alone, the use
of a combination of starch and the further polymer, preferably a further
polysaccharide, in particular a combination of starch ether and cellulose
ether, allows the use of a considerable extra amount of water in the concrete,
whilst maintaining a good dimension stability and a high flexural strength,
which strength may even be further improved. Surprisingly, the combination
of starch ether and cellulose ether results in a further increase of the green
strength.

When used, the total amount of the further polymer(s), in
particular the polysaccharide other than starch, more in particular a cellulose
(such as cellulose ether), is preferably 3-50 wt.% based on the combined
weight of the polymer(s) including the starch, more preferably up to 20 wt. %.

Particularly good results have *inter alia* been achieved with a further polymer, in particular a polysaccharide other than starch, more in particular a cellulose (such as cellulose ether), in an amount of at least about 5 wt. % based on the combined weight of the polymer(s) including the starch.

Further, one or more other additives may be present in the concrete. In particular an additional plasticizer and/or a water adsorbing agent may be present. Particular suitable plasticizers may be selected from the group consisting of lignosulfonates, melamine-based or naphtalene-based superplasticizers, polycarboxylates and the like; a particularly suitable water adsorbing agent is limestone powder.

The method of the invention involves shaping of the moist concrete into the desired shape. This may be done based upon a method known in the art.

Preferably, the shaping is accomplished by filling a mould with the moist concrete. The moist concrete can be distributed in a highly homogenous manner in the mould.

In an embodiment the moist concrete in the mould is subjected to vibratory ramming until it is dense and ready for removal from the mould. This facilitates densification of the concrete, which improves the dimension stabilisation of the concrete. In accordance with the invention, the required vibration may be reduced, compared to a moulding method wherein use is made of a concrete not comprising the starch, or substantial vibration may even be omitted.

In an embodiment, the concrete in the mould is compressed by an upper press. The method may be carried out with reduced pressurisation (compared to a method wherein use is made of a concrete not comprising the starch) or even without subjecting the concrete to substantial pressing on an upper surface of the concrete in the mould.

According to the invention the demoulding may be done
immediately after filling, and - if desired - vibration and/or pressing. It is preferred that the concrete has substantially not set, before removing the mould. Demoulding the concrete before setting of the cement can result in sufficient retainment of the shape, plays a significant role in the strength development of the concrete. "Sufficient retainment" of the shape means in accordance with the invention that the shape and size of the concrete product is, and remains, within the allowed tolerance ranges as defined in the appropriate standards.

For rectangular concrete paving flags, this means that the thickness, width and length of the flag does not deviate more than +/- 2 mm. Also, the difference between the length of two diagonals of a paving flag may not be more than 2 mm for flags having a diagonal of at most 850 mm, and not more than 4 mm for flags having a diagonal of more than 850 mm.

For concrete kerb units, the deviation of flatness and straightness may not be more than +/- 1.5 mm for kerbs having a length of gauge of 300 mm, not more than +/- 2.0 mm for kerbs having a length of gauge of 400 mm, and +/- 2.5 mm for kerbs having a length of gauge of 500 mm. The length of a kerb may not deviate more than +/- 1% to the nearest millimetre with a minimum of 4 mm, and not exceeding 10 mm. The other dimensions, except radius, for faces may not deviate more than +/- 3% to the nearest millimetre with a minimum of 3 mm, and not exceeding 5 mm, and for other parts not more than +/- 5% to the nearest millimetre with a minimum of 3 mm and not exceeding 10 mm.

For concrete paving blocks having a thickness below 100 mm, the length and width may not deviate more than +/- 2 mm, while the thickness may not deviate more than +/- 3 mm. For concrete paving blocks having a thickness of 100 mm or more, the length and width may not deviate more than +/- 3 mm, while the thickness may not deviate more than +/- 4 mm. Also, the difference between any two measurements of the thickness of a single block shall be at most 3 mm.
The improved properties of the concrete comprising the starch allow a reduced residence time in the mould. Thus, per mould a higher through-put of shaped bodies is realised, which is a considerable advantage. A suitable residence time can be determined empirically, without undue burden. Usually a residence time of about 1 min or more suffices. If desired, shorter residence times are feasible though, such as a residence time of less than about 30 sec, or even less than 10 sec; it is an option within the present invention to separate the shaped body from the mould, immediately after filling the mould.

From a practical point of view, the upper limit is about 45 minutes. Preferably, the residence time is not longer than 30 minutes, more preferably not longer than 20 minutes, even more preferably not longer than 10 minutes. These residence times are in sharp contrast to the residence times necessary when demoulding depends on the strength development resulting from the setting of cement. These residence times are in the order of magnitude of several hours. Thus, the present invention allows for a remarkable increase in production speed.

The invention further relates to a shaped concrete body, in particular a moulded body, obtainable by a method according to the invention. In particular, the invention relates to a shaped concrete article wherein the concrete comprises a starch as defined above, and optionally one or more ingredients as mentioned above. The article is in particular made from moist concrete, which concrete has set.

A shaped article according to the invention usually has a good appearance, such as a smooth surface and/or good strength properties.

In an embodiment, the concrete article is selected from the group consisting of blocks, slabs, paving stones, curbstones, garden ornaments, street furniture and tiles.

The invention in particular relates to a shaped concrete body, meeting the requirements put in the respective NEN-EN standards for
concrete articles, such as NEN-EN 1338 for concrete paving blocks, NEN-EN 1339 for concrete paving flags or EN-EN 1340 for concrete kerb units. These norms also describe in detail the allowed tolerances for size and shape, quoted above, as well as the methods of measuring them. For that reason, the contents of these norms is to be considered incorporated herein by reference.

Another aspect of the invention is a moist concrete which may be used in a method according to the invention. The composition of the moist concrete generally is as described above when describing the method of the invention in detail. Advantages of the use of a moist concrete according to the invention, compared to conventional (earth) moist concrete include: reduction of energy costs during the production of shaped articles of the concrete, reduction of wear and tear and/or maintenance of machinery, increased strength, shorter cycle times and/or saving on cement use by using less cement to realise the same strength. The concrete may further lead to one or more other advantages (in a product made from the moist concrete), which are thought to be at least partially caused by the increase in density, in the presence of the starch and optionally another polymer (such as cellulose or a further polysaccharide). Such advantages include: increased freeze/thaw resistance, less efflorescence, high(er) wear resistance, intensified colour, improved surface properties.

In an embodiment, the moist concrete has a degree of compactability of at least 1.11, as determined by NEN-EN 5958, preferably of at least 1.20, more preferably of at least 1.26. In practice, the moist concrete preferably has a degree of compactability of up to 1.5, in particular of up to 1.45.

An additional aspect of the invention is the use of a starch, in particular a starch as defined above, such as a starch ether, in a concrete - in particular a moist concrete such as earth moist concrete - as a reducer of the internal friction of the concrete, as a concrete-homogenising aid or as a flowing-aid.
Moreover, the invention relates to the use of a starch, as defined above, such as a starch ether, in a concrete - in particular a moist concrete such as earth moist concrete - to increase the strength of the concrete. In particular the starch may be used to increase any of the following strengths, alone or in combination: compressive strength, flexural tensile strength, tensile splitting strength and green strength.

In yet another aspect, the invention relates to the use of a starch, as defined above, such as a starch ether, in a concrete - in particular a moist concrete such as earth moist concrete - to improve smoothness of the surface of a shaped body formed of the concrete.

In yet another aspect, the invention relates to the use of a starch, as defined above, to increase the density of a concrete.

The invention will now be illustrated by the following example.

**Example: Production of concrete blocks with starch ether**

In a production environment using pressing and vibration, concrete blocks were made with two starch ether products: Solvitose FC50 (SE1) and the same starch ether containing 10% of the cellulose ether Methocel 254 (SE2). The mix design of the concrete was:

- CEM III B 80 kg/m³
- CEM II BF 250 kg/m³
- Sand 0-4 1400 kg/m³
- Gravel 2-8 550 kg/m³
- Total Water 109 kg/m³

The results are presented in Table 1 and clearly show that a large amount of water can be added to the concrete without causing stickiness or less retaining of the shape. With the mixture of starch ether and cellulose
ether (SE2) even a larger amount of water can be added to the concrete than with the pure starch ether (SE1).

Moreover, the strength increases which gives the opportunity to decrease the amount of cement in the concrete. Addition of both the pure starch ether (SE1) and the mixture with cellulose ether (SE2) lead to an increase of the density of the stones. The increased density points at a smaller pore volume. This has several advantages as mentioned above. One of those advantages is improved thaw/frost and salt stability. As pores can generally be filled with water, a smaller pore volume will also mean a smaller amount of water in the stones leading to a better thaw/frost stability.
<table>
<thead>
<tr>
<th>Test</th>
<th>Starch type</th>
<th>Dosage starch on cement (%)</th>
<th>extra amount of water</th>
<th>Stickiness</th>
<th>Dimension stability</th>
<th>7 days flexural tensile strength (N/mm²)</th>
<th>28 days flexural tensile strength (N/mm²)</th>
<th>Increase of strength from 7 to 28 days (%)</th>
<th>Density (kg/m³)</th>
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Claims

1. Method for preparing a concrete shaped body from a moist concrete, comprising
   - providing a mixture comprising cement, water, aggregate (i.e. sand and/or gravel) and at least partially modified starch thereby forming the moist concrete; and
   - shaping the concrete into the desired shape, preferably by moulding,

wherein the moist concrete is a concrete according to compaction class C0, C1, or C2.

2. Method according to claim 1, further comprising
   - demoulding the concrete after a residence time, before setting of the cement can result in sufficient retainment of the shape

3. Method according to claim 2, wherein said residence time is not longer than 45 minutes, preferably not longer than 30 minutes.

4. Method according to any one of the preceding claims, wherein at least one starch is used selected from the group consisting of chemically modified starches, biologically modified starches and physically modified starches, including cross-linked starches, starch esters and starch ethers.

5. Method according to claim 4, wherein at least one starch ether is used selected from hydroxyalkylated starches, carboxyalkylated starches, including starches that are both hydroxyalkylated and carboxyalkylated.

6. Method according to claim 5, wherein the hydroxyalkylated starch is a hydroxypropylated starch and/or the carboxyalkylated starch is a carboxymethylated starch.

7. Method according to claim 5 or 6, wherein the degree of hydroxyalkylation (DS) of the starch is between 0.01 and 2.0 hydroxyalkylations per monosaccharide unit.

8. Method according to any one of the claims 5-7, wherein the degree of
carboxyalkylation of the starch (DS) is between 0.01 and 0.5 carboxyalkylations per monosaccharide unit.

9. Method according to any one of the claims 4-8 wherein the degree of cross-linking of the starch with epichlorohydrin (DS) is between 0.0001 and 0.05.

10. Method according to any one of the preceding claims, wherein the amount of starch in the moist concrete is in the range of 0.01 to 1 wt.% based upon the weight of the cement, preferably in the range of 0.05 to 0.3 wt.% based upon the weight of the cement.

11. Method according to any one of the preceding claims, wherein in addition to the starch at least one further polymer, preferably a further polysaccharide is present.

12. Method according to claim 11, wherein the further polysaccharide is selected from the group consisting of cellulosics, xanthane gums, tamarind, welan gums and guar, in particular a cellulose, more in particular a cellulose ether.

13. Method according to claim 11 or 12, wherein the amount of further polymer is in the range of 3-50 wt.%, preferably in the range of 5-20 wt.%, based upon the total weight of starch plus further polymer.

14. Method according to any one of the preceding claims, wherein the moist concrete has a degree of compactability (C) of at least 1.11, preferably at least 1.20, more preferably 1.26-1.45.

15. Method according to any one of the preceding claims, wherein the concrete is shaped in a mould and the concrete in the mould is subjected to at least one treatment selected from: vibrating and pressing.

16. Method according to any one of the preceding claims, wherein the shaped body is selected from the group consisting of blocks, slabs, paving stones, curbstones, garden ornaments, street furniture and tiles.

17. Moist concrete as defined in any of the claims 1-14.

18. Shaped concrete body, comprising a starch as defined in any one of the
claims 4-10 and optionally a second polymer as defined in any one of the claims 11-13, said body preferably being selected from the group consisting of blocks, slabs, paving stones, curbstones and tiles.

19. Concrete body according to claim 18, wherein the body is a moulded body.

20. Use of a starch, in particular a starch as defined in any one of the claims 4-10, in a concrete to increase the density of the concrete.

21. Use of a starch, in particular a starch as defined in any one of the claims 4-10, in a concrete for increasing strength, in particular green strength, of the concrete.

22. Use of a starch, in particular a starch as defined in any one of the claims 4-10, in a concrete to improve smoothness of the surface of a shaped body formed of the concrete.

23. Use according to any one of the claims 20-22, wherein the concrete is a moist concrete, having a C value of at least 1.11, preferably of at least 1.20, more preferably of about 1.26-1.45.
Figure 1
**INTERNATIONAL SEARCH REPORT**

**INTERNATIONAL APPLICATION NO**

PCT/NL2005/000896

**A. CLASSIFICATION OF SUBJECT MATTER**

INV. C04B28/02 C04B24/38

According to International Patent Classification (IPC) or to both national classification and IPC

**B. FIELDS SEARCHED**

Minimum documentation searched (classification system followed by classification symbols)

C04B

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic database consulted during the international search (name of database and, where practical, search terms used)

EPO-Internal

**C. DOCUMENTS CONSIDERED TO BE RELEVANT**

<table>
<thead>
<tr>
<th>Category</th>
<th>Citation of document, with indication, where appropriate, of the relevant passages</th>
<th>Relevant to claim No.</th>
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<tbody>
<tr>
<td>X</td>
<td>EP 0 955 277 A (COOPERATIVE VERKoop- EN) 10 November 1999 (1999-11-10) the whole document</td>
<td>1-7, 10-20, 22, 23</td>
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<tr>
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<td>GB 922 427 A (JOHNS-MANVILLE CORPORATION) 3 April 1963 (1963-04-03) the whole document</td>
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**Date of the actual completion of the international search**

19 April 2006

**Date of mailing of the international search report**

26/04/2006

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