

ECO-CEMENT, COMPOSITIONS THEREOF, AND METHODS OF PRODUCTION THEREOF

TECHNICAL FIELD

[0001] The present disclosure relates to a cement composition and methods of producing the cement composition by using sodium carbonate as an activator/reactant. More specifically, the disclosure relates to a cement composition that is biocompatible, calcium-rich, and environmentally friendly.

BACKGROUND

[0002] Cement is a crucial binding material widely used in construction and civil engineering. It acts as a key component in concrete and mortar, providing structural integrity and durability to various constructions, including buildings, bridges, and roads. Traditionally, cement is produced by calcining limestone and other materials at high temperatures to form clinker, which is then ground into a fine powder. The most common type, Portland cement, is composed primarily of calcium silicates. Upon mixing with water, cement undergoes a hydration reaction, resulting in the formation of a hardened mass that binds aggregate materials. Despite its widespread use, conventional cement production is associated with significant environmental challenges, including high carbon dioxide emissions and energy consumption.

[0003] As a possible alternative, alkali-activated binders (AAB) are cementitious materials produced through a series of reactions between an aluminosilicate precursor and an alkali reagent at high pH, involving dissolution, polycondensation, and the formation of hardened new mineral phases at relatively low temperatures, typically below 100 °C [1-3]. The precursors generally serve as the source of silicon and aluminium and must exhibit high solubility in alkali media. Consequently, a diverse range of materials can be utilised as precursors, including but not limited to modified clay minerals, natural pozzolans, fly ash, bottom ash, and metallurgical slags [4,5]. The reagent is usually a water-soluble substance containing alkali metals, such as sodium or potassium hydroxides, silicates, carbonates, sulphates, or aluminates [6].

[0004] Studies have demonstrated that materials based on alkali-activated slag as a binder exhibit similar or superior performance compared to those made with Ordinary Portland Cement (OPC) as a binder [7-9]. However, using conventional reagents such as sodium silicate and sodium hydroxide could negatively impact AABs, including short setting times and higher shrinkage, which could hinder their large-scale application [9-11].

[0005] Accordingly, sustainable and environmentally friendly cement compositions with superior physical properties remain highly desirable.

SUMMARY

[0006] In accordance with one aspect of the present disclosure, a cement composition is disclosed, comprising: 80% to 90% by weight of a cement precursor comprising one or more alkali-activatable aluminosilicate materials; and 10% to 20% by weight of a reagent comprising Na_2CO_3 .

[0007] In some aspects, the cement composition is hydrated or further comprises water. In some further aspects, the cement composition is hydrated or further comprises water in an amount of between 37% to 42% or 40% by weight of the total cement composition.

[0008] In some aspects, the cement composition absorbs carbon dioxide to form a carbonate-based cement. In some further aspects, the cement composition is configured to, after hydration, absorb carbon dioxide to form a carbonate-based cement.

[0009] In some aspects, the reagent further comprises an accelerator comprising: anhydrous sodium metasilicate ($\text{Na}_2\text{O} \cdot \text{SiO}_2$), reactive magnesium oxide (MgO), calcium hydroxide ($\text{Ca}(\text{OH})_2$) or slaked lime, synthetic hydrotalcite, or combinations thereof.

[0010] In some aspects, a total amount of Na_2O is between 3.5% to 5% by weight of the reagent.

[0011] In some aspects, a total amount of MgO does not exceed 7% by weight of the reagent.

[0012] In some aspects, a total amount of Ca(OH)_2 does not exceed 5.2% by weight of the reagent.

[0013] In some aspects, a total amount of the synthetic hydrotalcite does not exceed 0.08% by weight of the reagent.

5 **[0014]** In some aspects, a purity of the anhydrous sodium metasilicate is at least 96%, comprising an amount of Na_2O between 49.5% to 51.5% by weight of the anhydrous sodium metasilicate, and an amount of SiO_2 between 45% to 47% by weight of the anhydrous sodium metasilicate.

10 **[0015]** In some aspects, an amount of the MgO is at least 97% by weight of the reactive magnesium oxide or at least 99% by weight of the reactive magnesium oxide, a specific surface area of the reactive magnesium oxide is at least $32 \text{ m}^2/\text{g}$, and a particle size of the reactive magnesium oxide does not exceed $125 \text{ }\mu\text{m}$.

15 **[0016]** In some aspects, an amount of the Ca(OH)_2 is at least 95% by weight of the calcium hydroxide or slaked lime or at least 97% by weight of the calcium hydroxide or slaked lime, and a particle size of the calcium hydroxide or slaked lime does not exceed $45 \text{ }\mu\text{m}$.

20 **[0017]** In some aspects, the synthetic hydrotalcite is magnesium-aluminium hydroxycarbonate comprising 73% Al_2O_3 by weight and 26% MgO by weight, or 74% Al_2O_3 by weight and 27% MgO by weight, and a particle size of the synthetic hydrotalcite does not exceed $40 \text{ }\mu\text{m}$.

[0018] In some aspects, the Na_2CO_3 comprises at least 97% by weight or at least 99% by weight of a source material.

[0019] In some aspects, a particle size of the source material does not exceed $100 \text{ }\mu\text{m}$.

25 **[0020]** In some aspects, the cement precursor comprises CaO in an amount between 35% to 52% by weight of the cement precursor.

[0021] In some aspects, a grain size of the cement precursor does not exceed $45 \text{ }\mu\text{m}$ or does not exceed $30 \text{ }\mu\text{m}$.

[0022] In some aspects, an amount of amorphous phase of the cement precursor is at least 85% by weight of the cement precursor.

[0023] In some aspects, the cement precursor comprises CaO, MgO, Al₂O₃, SiO₂, or a combination thereof.

5 **[0024]** In some aspects, a ratio of an amount of the CaO and the MgO in the cement precursor to an amount of the Al₂O₃ and the SiO₂ in the cement precursor is at least 7:10, by weight.

[0025] In some aspects, the cement precursor comprises 85% to 88% by weight of the cement composition; and the reagent comprises 12% to 15% by weight
10 of the cement composition.

[0026] In some aspects, the cement precursor comprises slag comprising blast-furnace slag, ferrous slag, non-ferrous slag, municipal solid waste slags, slag from energy facility, incineration products, or combinations thereof.

[0027] In some aspects, the cement precursor comprises glass comprising
15 recycled glass, waste glass, or both.

[0028] In some aspects, the cement composition further comprises a retarder comprising borax and/or tetrasodium 3-hydroxy-2,2'-iminodisuccinic acid (HIDS) configured to reduce a rate of cement setting.

[0029] In some aspects, the borax has a molecular weight of between 370g/mol
20 to 390g/mol, a specific gravity of between 1.6 g/cc to 1.8g/cc, and an onset of water loss at between 60°C to 64°C.

[0030] In some aspects, the HIDS has a molecular weight of between 340g/mol to 360g/mol, a density of between 1.4g/cm³ and 1.6g/ cm³ at 20°C, and a concentration of between 49% to 51% by weight in aqueous solution.

25 **[0031]** In some aspects, the cement composition is rich in calcium.

[0032] In accordance with another aspect of the present disclosure, a reinforced cement composition is disclosed, comprising: the cement composition of

any one of the above aspects; and a filler material comprising wollastonite fibres, engineered aggregates comprising silicon dioxide, or both.

[0033] In some aspects, an amount of the wollastonite fibres is between 2% to 3% by volume or 2.5% by volume.

5 **[0034]** In some aspects, a ratio between an amount of the cement composition to an amount of the engineered aggregates is between 1:0.8 and 1:3.5 by weight, between 1:0.9 to 1:2 by weight, or 1:1 by weight.

[0035] In some aspects, an amount of SiO_2 in the wollastonite fibres is between 49% to 53% by weight of the wollastonite fibres or 51% by weight of the wollastonite
10 fibres.

[0036] In some aspects, an amount of Al_2O_3 in the wollastonite fibres does not exceed 1% by weight of the wollastonite fibres.

[0037] In some aspects, an amount of CaO in the wollastonite fibres is between 43% to 47% by weight of the wollastonite fibres or 45% by weight of the wollastonite
15 fibres.

[0038] In some aspects, an amount of MgO in the wollastonite fibres does not exceed 2% by weight of the wollastonite fibres.

[0039] In some aspects, an amount of Fe_2O_3 in the wollastonite fibres does not exceed 0.5% by weight of the wollastonite fibres.

20 **[0040]** In some aspects, the engineered aggregates comprise quartz, quartz sands, or both.

[0041] In some aspects, a loss on ignition of the wollastonite fibres does not exceed 5% by weight at 1000°C , an aspect ratio of wollastonite fibres is between 15:1 to 21:1 or 19:1, and a grain size of the wollastonite fibres does not exceed $75\text{ }\mu\text{m}$.

25 **[0042]** In accordance with another aspect of the present disclosure, a method or producing cement from the cement composition of any one of the above aspects is disclosed, comprising: homogenizing the reagent by dry shear mixing; homogenizing the cement precursor by dry shear mixing; homogenizing the reagent and the cement

precursor to form cement powder; and homogenizing water with the cement powder to produce the cement or reinforced cement. In some further aspects, the reagent is mixed for 18 minutes to 22 minutes at 180 RPM to 220 RPM, and/or the cement precursor is mixed for 18 minutes to 22 minutes at 180 RPM to 220 RPM and/or the
5 water is homogenized in an amount of between 37% to 42% or 40% by weight of the total composition.

[0043] In accordance with another aspect of the present disclosure, a method for producing cement from the reinforced cement composition of any one of the above aspects is disclosed, comprising: homogenizing the reagent by dry shear mixing for
10 18 minutes to 22 minutes at 180 RPM to 220 RPM; homogenizing the cement precursor by dry shear mixing for 9 minutes to 11 minutes at 200 RPM or less; homogenizing the wollastonite fibers with the reagent and the cement precursor to form cement powder; and homogenizing water in an amount between 37% to 42% or 40% by weight of the total composition with the cement powder.

15 In accordance with another aspect of the present disclosure, a method for producing cement from the reinforced cement composition of any one of the above aspects is disclosed, comprising: homogenizing the reagent by dry shear mixing for 18 minutes to 22 minutes at 180 RPM to 220 RPM; homogenizing the cement precursor by dry shear mixing for 9 minutes to 11 minutes at 200 RPM or less; homogenizing the
20 reagent and the cement precursor to form cement powder; homogenizing the engineered aggregates with the water in an amount of 2.5% to 3.5% by weight of the engineered aggregates to form a mixture; and homogenizing the mixture with the cement powder

[0044] In some aspects, the method further comprises dissolving a
25 superplasticiser comprising polycarboxylate in water at an amount of 0.9% to 1.1% by weight of the engineered aggregates, the water in an amount of 2.5% to 3.5% by weight of the engineered aggregates; homogenizing the engineered aggregates with the superplasticiser and the water to form a mixture; and homogenizing the mixture with the cement powder.

30 **[0045]** In some aspects, a density of the cement is in a range of 2.1 to 2.6 g/cm³.

[0046] In some aspects, a compressive strength of the cement 24 hours after formation or mixing is at least 10 MPa.

[0047] In some aspects, a splitting tensile of the cement 24 hours after formation or mixing is at least 1 MPa.

5 [0048] In some aspects, the cement is a calcium carbonate cement.

BRIEF DESCRIPTION OF THE DRAWINGS

[0049] Further features and advantages of the present disclosure will become apparent from the following detailed description, taken in combination with the appended drawings, in which:

10 FIG. 1 depicts isothermal calorimetry curves of an alkali-activated, sodium carbonate-based cement composition showing heat flow and total heat released.

FIG. 2 depicts isothermal calorimetry curves of a biocompatible and an environmentally friendly cement composition, according to an example embodiment.

15 FIG. 3 depicts effect of borax addition on the hydration of a biocompatible and an environmentally friendly cement composition, according to an example embodiment.

FIG. 4 depicts effect of tetrasodium 3-hydroxy-2,2'-iminodisuccinic acid addition on the hydration of a biocompatible and an environmentally friendly cement composition, according to an example embodiment.

20 FIG. 5 depicts cumulative particle size distribution of engineered aggregates, according to an example embodiment.

FIG. 6 depicts compressive strength over ageing time of a biocompatible and an environmentally friendly cement composition, according to an example embodiment.

FIG. 7 depicts splitting tensile strength over ageing time of a biocompatible and an environmentally friendly cement composition, according to an example embodiment.

25 FIG. 8 depicts compressive strength over ageing time of a reinforced a biocompatible and an environmentally friendly cement composition, according to an example embodiment.

FIG. 9 depicts splitting tensile strength over ageing time of a reinforced biocompatible and an environmentally friendly cement composition, according to an example embodiment.

5 FIG. 10 depicts compressive strength over ageing time of a carbonated mortar produced from a reinforced biocompatible and environmentally friendly cement composition, according to an example embodiment.

FIG. 11 depicts splitting tensile strength over ageing time of a carbonated mortar produced from a reinforced biocompatible and environmentally friendly cement composition, according to an example embodiment.

10 **DETAILED DESCRIPTION**

[0050] The following description is of preferred embodiments by way of example only and without limitation to the combination of features necessary for carrying the disclosure into effect.

15 [0051] All terms are intended to be understood as they would be understood by a person skilled in the art. Unless defined otherwise, all technical and scientific terms used herein have the same meaning as commonly understood by one of ordinary skill in the art to which the disclosure pertains. The section headings used herein are for organizational purposes only and are not to be construed as limiting the subject matter described.

20 [0052] Although various features of the present disclosure can be described in the context of a single embodiment, the features can also be provided separately or in any suitable combination. Conversely, although the present disclosure can be described herein in the context of separate embodiments for clarity, the present disclosure can also be implemented in a single embodiment.

25 [0053] The following definitions supplement those in the art and are directed to the current application. Accordingly, the terminology used herein is for the purpose of describing particular embodiments only and is not intended to be limiting.

[0054] In this application, the use of the singular includes the plural unless specifically stated otherwise. It must be noted that, as used in the specification, the

singular forms "a," "an" and "the" include plural referents unless the context clearly dictates otherwise.

[0055] In this application, the use of "or" means "and/or" unless stated otherwise. The terms "and/or" and "any combination thereof and their grammatical
5 equivalents as used herein, can be used interchangeably. These terms can convey that any and all combinations are specifically contemplated. The term "or" can be used conjunctively or disjunctively, unless the context specifically refers to a disjunctive use.

[0056] Furthermore, use of the term "including" as well as other forms, such as
10 "include", "includes," and "included," is not limiting.

[0057] Reference in the specification to "some embodiments," "an embodiment," "one embodiment" "alternate embodiment", or "other embodiments" means that a particular feature, structure, or characteristic described in connection with the embodiments is included in at least some embodiments, but not necessarily
15 all embodiments, of the present disclosures.

[0058] As used in this specification and claim(s), the words "comprising" (and any form of comprising, such as "comprise" and "comprises"), "having" (and any form of having, such as "have" and "has"), "including" (and any form of including, such as "includes" and "include") or "containing" (and any form of containing, such as
20 "contains" and "contain") are inclusive or open-ended and do not exclude additional, unrecited elements or method steps. It is contemplated that any embodiment discussed in this specification can be implemented with respect to any method or composition of the present disclosure, and vice versa. Furthermore, compositions of the present disclosure can be used to achieve methods of the present disclosure.

[0059] The term "about" in relation to a reference numerical value and its
25 grammatical equivalents as used herein can include the numerical value itself and a range of values plus or minus 10% from that numerical value. The term "about" or "approximately" means within an acceptable error range for the particular value as determined by one of ordinary skill in the art, which will depend in part on how the
30 value is measured or determined, i.e., the limitations of the measurement system. For

example, "about" can mean within 1 or more than 1 standard deviation, per the practice in the art. Alternatively, "about" can mean a range of up to 20%, up to 10%, up to 5%, or up to 1% of a given value. In another example, the amount "about 10" includes 10 and any amounts from 9 to 11.

5 **[0060]** The description which follows, and the embodiments described herein are provided by way of illustration of an example of particular embodiments of principles and aspects of the present disclosure. The examples are provided for the purposes of explanation and not of limitation, of those principles of the disclosure. In the description that follows, like parts and/or steps are marked throughout the
10 specification and the drawing with the same respective reference numerals.

[0061] In the search for a cost-effective and sustainable activator for alkali-activated binders in use for cement compositions, sodium carbonate emerges as one of the most promising alternatives. Sodium carbonate is cheaper, less corrosive, and more environmentally friendly than conventional reagents. Therefore, the combined
15 use of sodium carbonate and slag to produce a binder is considered more economical and environmentally friendly, as it eliminates the need for OPC and does not require heat treatment for curing.

[0062] The reactions in slag-sodium carbonate cement compositions release heat upon adding water, a phenomenon also observed in Portland cement-based
20 materials and in reactions involving liquid sodium silicate and slag compositions. These reactions are typically preceded by a pre-induction period, related to the initial dissolution of slag particles, followed by an induction period whose duration is strongly influenced by the characteristics of the slag and the reagent. After the induction period, intensive acceleration and deceleration phases are typical, and the main
25 exothermic peak observed can be associated with the precipitation of phases responsible for strength development [12,13]. Additionally, for sodium carbonate-slag-based AAB, an extra exothermic peak is observed approximately 60 minutes after mixing, which can be attributed to the precipitation of gaylussite [14].

[0063] Despite its potential, a constraint for the widespread use of sodium
30 carbonate as an activator for slags is the prolonged time required for sodium carbonate-slag compositions to react and commence hardening, which can be 3 to 5

days [15,16]. In some cases, the strength development of sodium carbonate-based activators is not as rapid as that of liquid alkali silicate-based compositions. However, after 28 days of curing, their mechanical properties can be comparable to Portland cement-based products. Moreover, the mechanical properties of sodium carbonate-based cement can meet the requirements of most construction market applications.

[0064] The present disclosure relates to a sodium carbonate-activated cement composition settable by hydration to produce CaCO_3 as well as methods for producing cement products therefrom. In particular, the cement composition can reduce the carbon footprint of the cement material produced therefrom by incorporating environmentally friendly materials in the composition, minimizing or removing clinker content, and reducing harmful emissions during the reaction phase. As such, the cement compositions of the present disclosure may be referred to as an eco-cement composition, which can form an eco-cement (material). According to a broad aspect, the composition comprises 85% to 88% by weight of a cement precursor configured to be activated by an alkali reagent and comprising silicates, oxides, and/or aluminosilicates containing CaO ; and 11% to 15% by weight of a reagent comprising Na_2CO_3 . In particular, the disclosed cement composition can comprise a single dry reactive component comprising sodium carbonate at 10% to 20% by weight of the composition, anhydrous sodium metasilicate ($\text{Na}_2\text{O} \cdot \text{SiO}_2$) at 2% to 5% by weight of the reagent, reactive magnesium oxide at 3% to 6% by weight of the reagent, and synthetic hydrotalcite at 0.04% to 0.06% by weight of the reagent, which can be combined with dry ferrous calcium-rich slag, dry non-ferrous calcium-rich slag, or a combination thereof at 80% to 90% by weight of the composition.

[0065] Accordingly, the present disclosure can provide a cost-effective, one-part cement formulation that utilises calcium-rich ferrous slags, calcium-rich non-ferrous slags, or a mixture thereof, activated by engineered sodium carbonate-based agents. Through this process, a sustainable, environmentally friendly, CO_2 -absorbing cement matrix with high mechanical performance, equivalent to OPC cement type CEM I, can be produced, which can achieve compressive strengths exceeding 10 MPa after 1 day, 12 MPa after 7 days and 32.5 MPa after 28 days. That is, compared to standard sodium carbonate-based cements, the cement compositions disclosed herein can promote rapid high-early compressive strength, exceeding 10 MPa after

just 1 day, whereas conventional materials typically require at least 3 days to achieve comparable strength levels.

[0066] There are many possible industrial applications for the cement compositions disclosed herein. For example, traditional approaches to coastal protection infrastructure and erosion control often rely on OPC concrete and non-biodegradable materials such as polymers, contributing to environmental harm and climate change. The cement compositions disclosed herein can provide a sustainable alternative that protects coastlines while minimizing environmental impact. Further, the disclosed cement composition may exhibit better intrinsic properties for durability in coastal environments than traditional OPC, particularly in terms of resistance to chloride ingress and sulphate attack. In particular, its denser microstructure and lower permeability contribute to its enhanced durability.

[0067] The disclosed cement composition may exhibit enhanced biocompatibility and can be useful in versatile applications, particularly in marine and coastal protection initiatives, and can provide vital support to diverse plant and animal species within aquatic ecosystems. For example, the disclosed cement composition may be used in fabricating artificial reefs that emulate natural reef structures, fostering biodiversity and enhancing shoreline defences to effectively safeguard coastlines and mitigate erosion. Further, the disclosed cement composition can exhibit exceptional resilience to climate and environmental changes, ensuring long-term protection for coastal communities and significant cost savings over time.

[0068] Furthermore, the cement composition disclosed herein can contain a high calcium content and can undergo carbonation through CO₂ absorption when exposed to the environment [17], a process typically associated with the corrosion and degradation of steel armour in OPC-based concrete. To mitigate or minimise the potentially harmful effects of carbonation, it is possible to incorporate reinforcing materials such as wollastonite fibres (e.g. at 2% to 3% by volume, preferably 2.5% by volume) and engineered aggregates (at 1:0.9 to 1:1.1 wt./wt. ratio, preferably 1:1 wt./wt.), as disclosed further herein.

[0069] As such, the cement composition disclosed herein can offer numerous advantages for coastal protection applications, including enhanced durability,

resistance to aggressive marine environments, and a reduced environmental footprint, aligning with sustainable development and environmental conservation goals. In particular, the chemical stability, dense microstructure, low permeability, controlled pH levels, and environmental benefits of the cement composition disclosed
5 herein can result in a highly biocompatible cement material for marine and coastal applications. These properties can also help minimise the impact on marine ecosystems while providing durable and sustainable solutions for marine infrastructure. Specifically, the cement composition disclosed herein can be a cost-effective, innovative, adaptive, sustainable, long-lasting and smart solution for
10 addressing matters related to costal protection and flood management, particularly with regard to breakwaters and seawalls, revetments, groynes, artificial reefs, flood defences, shoreline stabilisation, underwater and submerged structures (e.g. underwater cable-protection), and marine habitat restoration and wave energy conversion structures.

15 **[0070]** Furthermore, although various references are made herein with regard to a cement composition, it should be noted that the various features, advantages, and benefits are described herein are also generally applicable to products and materials resulting therefrom.

Embodiments

20 **[0071]** Embodiments are described below, by way of example only, for the calcium comprising environmentally friendly cement composition.

[0072] In one aspect, the materials used for the synthesis of the cement disclosed in this disclosure is a combination of inorganic, solid materials. Specifically, the cement composition for synthesizing the cement can comprise materials classified
25 as precursors, reagents, and accelerators. The components of the cement composition can promote the reactions responsible for the formation of a solid and dense matrix made of carbonate-like materials in up to 24 hours at room temperature to reach at least 10 MPa of compressive strength, in particular due to the accelerator, which comprises specific mineral compositions.

[0073] In accordance with the present disclosure, the precursor may be at least one cement precursor. In one exemplary embodiment, the precursor may comprise silicates and oxides. In one exemplary embodiment, the precursor may be a single material or a combination/mixture of materials. The precursor may be configured to
5 react with a reagent (as described further herein), particularly an alkali reagent, to form a mineral phase(s) (e.g., binder or cement material/product). That is, the cement composition may be an alkali-activatable cement composition or (cement) binder. The precursor may comprise one or more materials configured to provide calcium, silicon, and aluminium in the reaction for the formation of cement products/materials (e.g., act
10 as a source of calcium, silicon, and aluminium). The precursor can exhibit solubility, preferably high solubility, in the reagent (e.g., alkali reagent). As an example, the precursor may be one or more aluminosilicate compounds, for instance alkali-activatable aluminosilicate materials. As another example, the precursor may be modified/unmodified clay minerals, natural pozzolans, fly ash, bottom ash,
15 metallurgical slags, or combinations thereof. As another example, the precursor may comprise slag, glass, or both. As an example, the slag may comprise blast-furnace slag, ferrous slag, non-ferrous slag, municipal solid waste slags, slag from an energy/research facility (e.g., slag from Waste-to-Energy Research Facility, Singapore), incineration products, alternative/other slag(s) known in the art, or
20 combinations thereof. As another example, the glass may comprise recycled glass, waste glass, alternative/other glass(es) known in the art, or combinations thereof.

[0074] In one exemplary embodiment, the precursor may comprise 80% to 90% by weight, preferably 85% to 88% by weight of the total cement composition. In another example, the material(s) may comprise calcium oxide (e.g., CaO) content in
25 a range of 30% to 60% by weight of the precursor, preferably 35% to 52% by weight of the precursor. In another example, the ratio of the sum of CaO content and MgO content in the precursor to the sum of the Al_2O_3 content and SiO_2 content in the precursor is at least 6:10 by weight, preferably at least 7:10 by weight, or greater. That is, the components of (e.g. content comprised by) the precursor, expressed as (CaO
30 + MgO) / (Al_2O_3 + SiO_2)) should be at least 0.65, preferably at least 0.7, or greater. As an example, if the precursor comprises a mixture of materials, particularly slag and/or glass, the CaO content in the precursor may be the sum of the CaO content from each of the materials comprised in the precursor.

[0075] In one exemplary embodiment, the precursor may comprise material(s) (e.g. the content(s) of the precursor) with a grain size not exceeding 50 µm, preferably not exceeding 45 µm, most preferably not exceeding 30 µm. In some embodiments, 100% of the precursor material(s) have a grain size not exceeding 50 µm, preferably not exceeding 45 µm, most preferably not exceeding 30 µm.

[0076] In one exemplary embodiment, the precursor (e.g. the content(s) of the precursor) may comprise amorphous phase or phases of at least 80% by weight of the precursor, preferably at least 85% by weight, or greater. The weight percent (e.g. amount) of the amorphous phase(s) may be determined by X-ray diffraction and/or Rietveld analysis.

[0077] In accordance with the present disclosure, the reagent can refer to one or more materials that react with the precursor to form a binder and/or cement product/material. In one exemplary embodiment, the reagent may comprise 10% to 20% by weight, preferably 12% to 15% by weight of the total cement composition. In particular, the reagent may be an activator or serves as an activator for the reaction. As an example, the reagent may initiate or facilitate the reaction with the precursor. In one exemplary embodiment, the reagent may be an alkali reagent, preferably an alkali reagent of pH 7 or higher, more preferably pH 9 or higher, even more preferably pH 11 or higher. The precursor may be dissolved in the reagent to begin the reaction to from the binder and/or cement product/material. As another example, the reagent may be water-soluble.

[0078] In one exemplary embodiment, the reagent can contain/comprise alkali metals. As an example, the reagent may comprise sodium or potassium hydroxides, silicates, carbonates, sulphates, aluminates, or combinations thereof. In a preferred embodiment, the reagent is or comprises sodium carbonate (Na_2CO_3). That is, the cement composition may be alkali-activatable, sodium carbonate-based cement composition. In one exemplary embodiment, the material(s) serving as the source of sodium carbonate comprises at least 95% sodium carbonate by weight of the source material(s), preferably at least 97% by weight, more preferably at least 99% by weight. In another example, the (average) particle size of the reagent, in particular the sodium carbonate source material(s) (e.g., white crystalline powder thereof), does not exceed

150 μm , preferably does not exceed 120 μm , more preferably does not exceed 100 μm , even more preferably does not exceed 95 μm .

[0079] In accordance with the present disclosure, the reagent can further comprise (e.g. be combined with) one or more accelerators. The accelerator may be one or more materials configured to enhance a reaction rate for the formation of the binder and/or cement product/material.

[0080] In one exemplary embodiment, the accelerator may comprise sodium metasilicate (Na_2SiO_3) or ($\text{Na}_2\text{O}\cdot\text{SiO}_2$), magnesium oxide (MgO), calcium hydroxide/slaked lime ($\text{Ca}(\text{OH})_2$), hydrotalcite, materials/compounds thereof, or combinations thereof. Particularly, the sodium metasilicate may be anhydrous sodium metasilicate, the magnesium oxide may be reactive magnesium oxide; the hydrotalcite may be synthetic hydrotalcite, preferably magnesium-aluminum hydroxycarbonate or other layered double hydroxides (LDH); materials/compounds thereof; or combinations thereof.

[0081] In one exemplary embodiment, the total amount or total equivalent content (e.g. resulting from the addition of sodium carbonate and accelerator material such as anhydrous sodium metasilicate) of Na_2O in the reagent is between 2% to 6% by weight of the reagent, preferably 3% to 5.5% by weight, most preferably 3.5% to 5% by weight. In another example, the total amount of MgO content comprised in (e.g. added to) the reagent does not exceed 10% by weight of the reagent, preferably does not exceed 7% by weight. In another example, the total amount of $\text{Ca}(\text{OH})_2$ content comprised in (e.g. added to) the reagent does not exceed 6% by weight of the reagent, preferably does not exceed 5.2% by weight. In another example, the total amount of hydrotalcite content comprised in (e.g. added to) the reagent does not exceed 1% by weight of the reagent, preferably does not exceed 0.8% by weight.

[0082] In one exemplary embodiment, the degree of purity for the sodium metasilicate is at least 90%, preferably at least 93%, most preferably at least 96%. As another example, the total amount of Na_2O content in the sodium metasilicate is in a range 45% to 55% by weight of the sodium metasilicate, preferably in a range of 49.5% to 51.5% by weight. As another example, the total amount of SiO_2 content in

the sodium metasilicate is in a range 42% to 50% by weight of the sodium metasilicate, preferably in a range of 45% to 57% by weight.

[0083] In one exemplary embodiment, the total amount of MgO content in the magnesium oxide (e.g. reactive magnesium oxide material) is at least 95% by weight of the magnesium oxide, preferably at least 97% by weight, most preferably at least 99% by weight. As another example, the specific surface area of the magnesium oxide is at least 30 m²/g, preferably at least 32 m²/g. As another example, the (average) particle size of the magnesium oxide does not exceed 150 µm, preferably does not exceed 125 µm. In particular, 100% of the particles of magnesium oxide does not exceed 125 µm, preferably does not exceed 150 µm.

[0084] In one exemplary embodiment, the total amount of Ca(OH)₂ content in the calcium hydroxide/slaked lime (e.g. material(s)/compound(s) thereof) is at least 93% by weight of the hydroxide/slaked lime, preferably at least 95% by weight, most preferably at least 97% by weight. As another example, the (average) particle size of the calcium hydroxide/slaked lime does not exceed 50 µm, preferably does not exceed 45 µm. In particular, 100% of the particles of the hydroxide/slaked lime do not exceed 50 µm, preferably does not exceed 45 µm.

[0085] In one exemplary embodiment, the total amount of Al₂O₃ content in the synthetic hydrotalcite is at least 70% by weight of the synthetic hydrotalcite, preferably at least 73% by weight, most preferably at least 74% by weight. As another example, the total amount of MgO content in the synthetic hydrotalcite is at least 25% by weight of the synthetic hydrotalcite, preferably at least 26% by weight, most preferably at least 27% by weight. As another example, the specific surface area of the synthetic hydrotalcite is at least 80 m²/g, preferably at least 84 m²/g. As another example, the (average) particle size of the synthetic hydrotalcite does not exceed 45 µm, preferably does not exceed 40 µm. In particular, 100% of the particles of the synthetic hydrotalcite does not exceed 45 µm, preferably does not exceed 40 µm.

[0086] In accordance with the present disclosure, the cement composition may further comprise at least one retarder. The retarder may be added to the cement composition (e.g., the cementitious mixture) to slow down (e.g., reduce the rate of) the chemical reactions that cause the setting and hardening of the cement. In

particular, the delay in the setting process from the reduced reaction rate can allow for extended working time, which can be beneficial in various construction scenarios.

In one exemplary embodiment, the retarder may be borax, 3-hydroxy-2,2'-iminodisuccinic acid/ tetrasodium 3-hydroxy-2,2'-iminodisuccinate (HIDS), or both. In

5 one exemplary embodiment, borax may be added to the cement composition for producing mortars and concrete. As an example, an amount of the borax comprised in the cement composition may be 7% by weight of the precursor or less, preferably 5% by weight or less. As another example, the borax may have a minimum molecular weight in a range of 350 g/mol to 410 g/mol, preferably in a range of 370 g/mol to 390
10 g/mol. As another example, the borax may have a specific gravity in a range of 1.5 g/cc to 1.9 g/cc, preferably in a range of 1.6 g/cc to 1.8 g/cc. As another example, the borax may have an onset of water loss at a range of 60 °C to 64°C, preferably at about 62 °C. In another example, an amount of the HIDS comprised in the cement composition may be 1% by weight of the precursor or less, preferably 0.5% by weight
15 or less. As another example, the HIDS is liquid HIDS and may have a molecular weight in a range of 320 g/mol to 380 g/mol, preferably in a range of 340 g/mol to 360 g/mol. As another example, the HIDS may have a density at 20°C in a range of 1.2 g/cm³ to 1.8 g/cm³, preferably in a range of 1.4 g/cm³ to 1.6 g/cm³. As another example, the concentration of the HIDS in an aqueous solution may be in a range of
20 47% to 53% by weight, preferably in a range of 49% to 51% by weight.

[0087] In accordance with the present disclosure, admixtures comprising mineral material(s) may be added to the cement composition. The mineral materials may exhibit various shapes and sizes and can be added as filler and/or aggregate for providing mechanical reinforcement or to mitigate the loss of mechanical properties
25 due to carbonation reactions. For example, mineral components from the surface of the mineral materials can react with the components in the cement composition during the cement reactions, which can promote a strong bond between the resulting cement and the filler/aggregate.

[0088] In one exemplary embodiment, the admixture can be wollastonite, preferably wollastonite fibres. In particular, wollastonite can be a needle-like mineral of natural origin. As an example, the wollastonite can be added to the cement composition in an amount of 1% to 4% by volume of the cement composition,
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preferably 2% to 3% by volume, most preferably 2.5% by volume. As another example, the wollastonite can comprise SiO_2 content in a range of 45% to 57% by weight of the wollastonite, preferably 49% to 53% by weight, most preferably 51% by weight. As another example, the wollastonite can comprise Al_2O_3 content in an amount that does not exceed 3% by weight of the wollastonite, preferably does not exceed 1% by weight. As an example, the wollastonite can comprise CaO content in a range of 40% to 50% by weight of the wollastonite, preferably 43% to 47% by weight, most preferably 45% by weight. As another example, the wollastonite can comprise MgO content in an amount that does not exceed 4% by weight of the wollastonite, preferably does not exceed 2% by weight. As another example, the wollastonite can comprise Fe_2O_3 content in an amount that does not exceed 1% by weight of the wollastonite, preferably does not exceed 0.5% by weight. As another example, the wollastonite can have a loss on ignition at 1000 °C in an amount that does not exceed 8% by weight of the wollastonite, preferably does not exceed 5% by weight. As another example, the wollastonite can have an aspect ratio in a range of 13:1 to 24:1, preferably in a range of 16:1 to 21:1, most preferably of 19:1. As another example, the (average) grain size of the wollastonite does not exceed 80 μm , preferably not exceeding 75 μm . In particular, 100% of the particles of wollastonite does not exceed a grain size of 80 μm , preferably not exceeding 75 μm .

[0089] In one exemplary embodiment, the admixture can be engineered aggregates, preferably sand-based engineering aggregates or quartz-based engineering aggregates. As an example, the engineered aggregate can comprise mixtures of silicon dioxide such as quartz and/or quartz sands that are natural and/or artificial. As another example, the engineered aggregates can be added to the cement composition in a range of 1:0.6 to 1:5 by weight ratio to the cement composition, preferably in a range of 1:0.8 to 1:3.5 by weight ratio, more preferably in a range of 1:0.9 to 1:1.1 by weight ratio, even more preferably in a range of 1:0.9 to 1:1.2 by weight ratio, most preferably 1:1 by weight ratio. Cumulative particle size distribution of an example embodiment of engineered aggregates is shown in FIG. 5, where the dashed lines represent the upper and lower limits of size distribution. As another example, the engineered aggregates can have a grain size distribution within 20% of the particle size distribution shown in FIG. 5, preferably within 5%, most preferably yielding the grain size distribution shown in FIG. 5.

[0090] In accordance with the present disclosure, the cement composition may be an alkali-activatable, sodium carbonate-based cement composition. The hydration of such a cement composition may comprise 5 stages/periods. An example isothermal calorimetry graph showing the heat flow (solid line) and total heat released (dashed line) over time for an example alkali-activatable, sodium carbonate-based cement composition is shown in FIG. 1. Referring to FIG. 1, the sequence of stages comprising dissolution, induction, acceleration, deceleration, and diffusion are also shown in the graph in reference to the flow and total heat realized.

[0091] As depicted in FIG. 1, the initial period of dissolution can correspond to the initial wetting and dissolution of precursors (e.g. glass and/or slag precursors) by the reagent (e.g. alkali reagent), which can occur seconds after they come into contact. Naturally exothermic, this reaction is represented by the first intense peak typically observed in the heat flow curves of alkali-activated materials. Subsequently, the induction period follows the dissolution and may be defined as the time between the exothermic peaks of the dissolution and acceleration periods on the heat flow curve; it is characterised by very low heat flow.

[0092] Following FIG. 1, the acceleration period, which follows the induction or dormant period, can represent the stage of nucleation and growth of gel phases in the cement composition, resulting in a more interconnected microstructure. The deceleration period follows the acceleration period and can mark the transition from rapid initial hydration to a slower and more controlled reaction phase, where the rate of heat evolution and reaction may slow significantly. During this phase, a gel-like layer of hydration products, for example primarily calcium-aluminium-silicate-hydrate (CASH), can form around the unreacted precursor particles (e.g. slag particles), which can impede the further diffusion of ions thereto and therefrom, leading to a slower overall reaction rate.

[0093] As depicted in FIG.1, in the diffusion period, the reaction rate can decrease further and may be dominated by the slow diffusion of ions through the already-formed hydration product layers. In this stage, the movement of water, ions, and other reactive species may occur primarily through the dense hydration product network. Hydration can continue at a much slower rate compared to the earlier stages,

and the formation of additional hydration products can gradually fill the pore spaces, promoting the densification of the microstructure.

[0094] In accordance with the present disclosure, a method for synthesizing cement and/or cement product/material using the cement composition is provided. As used herein, a material may be considered dry when it comprises less than 2 % by weight of moisture.

[0095] In one exemplary embodiment, the method can comprise a step of pre-processing of the cement components. As an example, the reagent component(s) (as described above) may be mixed to homogenize the reagent components. The reagent components may be mixed using dry shear mixing, for example with a planetary mill/mixer, although other/similar alternative mixing methods/tools are possible as well. In particular, the reagent components may be mixed for 18 to 22 minutes, preferably for about 20 min, at 180 to 220 RPM, preferably at about 200 RPM. As another example, when the precursor comprises more than one component, the precursor components may be mixed to homogenize the precursor components. The precursor components may be mixed using dry mixing, for example with a planetary mill/mixer, although other/similar alternative mixing methods/tools are possible as well. In particular, the precursor components may be mixed for 8 to 12 minutes, preferably for about 10 min, at a rate not exceeding 220 RPM, preferably not exceeding 200 RPM.

[0096] In one exemplary embodiment, the mixed reagent components and precursor components can be stored without being promptly consumed. As an example, the reagent, which may be in a form of a dry and/or reactive powder, can be stored in a dry and/or closed container, which can prevent moisture absorption. When stored accordingly at 25 °C, the shelf life of the mixed reagent may be 6 months. As another example, the precursor can be stored in a dry and/or closed container. When stored accordingly at 25 °C, the shelf life of the mixed precursor may be indefinite.

[0097] In one exemplary embodiment, the method further comprises a step of synthesizing cement powder. The cement powder can be synthesized by mixing the mixed/homogenized precursor and the mixed/homogenized reagent to homogenize the resulting mixture. The mixing/homogenizing method is not restrictive and should

result in a dry cement powder composition. As another example, the cement powder can be stored without being promptly consumed. In particular, the cement powder can be stored in a dry and/or closed container to avoid moisture absorption and triggering reactions. When stored accordingly at 25 °C, the shelf life of the cement powder may be 6 months.

[0098] In one exemplary embodiment, the method further comprises a step of synthesizing cement paste for producing cement and/or cement material/product. As an example, water may be added to the cement powder to initiate hydration and setting. As another example, an amount of water added to the cement powder may be in a range of 32% to 47% by weight of the cement powder, preferably in a range of 37% to 42% by weight, most preferably about 40% by weight. The resulting mixture may be subject to mechanical mixing for homogenization, the method of which is not restrictive.

[0099] In accordance with the present disclosure, a method for synthesizing reinforced cement and/or reinforced cement product/material using the cement composition is provided.

[00100] In one exemplary embodiment, wollastonite may be added to produce reinforced cement and/or reinforced cement product/material. The method may comprise adding wollastonite to the cement powder (which can be obtained as described above). The resulting wollastonite-cement composition may be subject to mechanical dry mixing for homogenization, the method of which is not restrictive. As another example, the wollastonite-cement composition can be stored without being promptly consumed. In particular, the wollastonite-cement composition can be stored in a dry and/or closed container as to avoid moisture absorption and triggering reactions. When stored accordingly at 25 °C, the shelf life of the wollastonite-cement composition may be 6 months. The method may further comprise synthesizing reinforced cement paste for producing reinforced cement and/or reinforced cement material/product. As an example, water may be added to the wollastonite-cement composition to initiate hydration and setting. As another example, an amount of water added to the wollastonite-cement composition may be in a range of 32% to 47% by weight of the wollastonite-cement composition, preferably in a range of 37% to 42%

by weight, most preferably about 40% by weight. The resulting mixture may be subject to mechanical mixing for homogenization, the method of which is not restrictive.

[00101] In one exemplary embodiment, engineered aggregates may be added to produce reinforced cement and/or reinforced cement product/material, in particular mortar. The method may comprise preparing a solution comprising water and at least one plasticiser, particularly at least one super plasticiser. The solution can comprise an amount of water in a range of 2% to 4% by weight of the engineered aggregates (as described above), preferably in a range of 2.5% to 3.5% by weight, most preferably about 3% by weight. The plasticiser may be a polycarboxylate or similar compound, which can be dissolved in the solution. An amount of the plasticiser added may be in a range of 0.7% to 1.3% by weight of the engineered aggregates, preferably in a range of 0.9% to 1.1% by weight, most preferably about 1% by weight. The solution may be mixed to homogenize the components therein, the method of which is not restrictive. The engineered aggregates may be added to the (homogenized) solution and may be mixed until a homogeneous dispersion is obtained. The resulting mixture containing the engineered aggregates can be added to the cement paste (as described above) by performing mechanical mixing as to homogenize the mixture into the cement paste, the method of which is not restrictive.

[00102] As described above, the features and embodiments of the disclosed cement composition may exhibit various technical advantages. Various examples thereof are described below.

[00103] In some aspects, the disclosed cement composition may be (e.g. largely comprised of) composed of waste derivative materials. As an example, recycled materials (e.g. glasses) and industrial by-products (e.g. slags for reusing) can be used as precursor materials.

[00104] In some aspects, the disclosed cement composition may comprise or utilize sustainable and highly available reactant in the form of baking soda (sodium carbonate, Na_2CO_3).

[00105] In some aspects, the disclosed cement composition may exhibit low carbon footprint and act as a CO_2 absorber. As an example, carbon footprint may be

reduced by using recycled materials and industrial by-products. As another example, the disclosed cement composition, as well as material and molded parts therefrom can absorb CO₂ from the environment, which can occur from the start of the alkaline reactions until the saturation over the years.

5 **[00106]** In some aspects, the disclosed cement composition is a hydraulic cement that only requires the addition of water to trigger the reactions to form a cement paste.

10 **[00107]** In some aspects, the disclosed cement composition can be biocompatible, calcium-rich, and environmentally friendly. As an example, the final cementitious material produced therefrom may be calcium carbonate (CaCO₃), which is the most common raw material found in nature. CaCO₃ is found in rocks (marble), minerals (calcite and aragonite), chalk, limestone, eggshells, gastropod shells, shellfish skeletons, pearls and coral reefs, which can make this material suitable for use in the repair and revitalisation of reefs and coastal areas. Further, the material
15 can have a dense and stable matrix, which can promote very low permeability and solubility in seawater and coastal environments, drastically reducing the leaching of harmful substances into the marine environment. As another example, the alkalinity of the disclosed cement composition may be controlled, which can be beneficial for marine organisms, as extreme pH levels can be harmful, which is the case of OPC.

20 **[00108]** In some aspects, the disclosed cement composition can result in stable and long-lasting structures. As an example, the disclosed cement composition can form dense microstructures that limit the ingress of aggressive ions and reduce the rate of deterioration, and can ensure a stable and long-lasting structure, which may be crucial for marine and coastal applications. As another example, the disclosed
25 cement composition may produce high durability materials in aggressive marine environments to ensure the structures remain stable and do not release harmful degradation products over time.

30 **[00109]** In some aspects, the disclosed cement composition and materials resulting therefrom can be 100% recyclable and renewable, which can promote circular economy and can be recycled and reused indefinitely.

[00110] In some aspects, the disclosed cement composition can promote tunable biological fouling. As an example, the disclosed cement composition can enable the control of surface characteristics, such as roughness, open porosity, and shape, to promote or discourage biological fouling on the cement material. For instance, a dense and smooth surface can be less conducive to the growth of marine biofilms and organisms, which can be beneficial in reducing maintenance costs and extending the lifespan of marine structures. Conversely, a highly open porous structure with a proper shape design and coarse surface can promote the growth of marine biofilms and organisms to produce long-lasting artificial reefs.

[00111] In some aspects, the disclosed cement composition can produce materials with high early mechanical strength at room temperature. As an example, the disclosed cement composition can, after 24 hours, develop a minimum of 10 MPa of compressive strength and a minimum of 1 MPa of splitting tensile strength. As another example, the hardening time can be controlled by the use of accelerators or retardants, as described above.

[00112] In some aspects, the disclosed cement composition can be light weight. As an example, the density of OPC typically ranges between 3.1 to 3.2 g/cm³, while the disclosed cement composition may have densities in the range of 2.1 to 2.6 g/cm³.

Examples and Experimental Results

[00113] Non-limiting examples of the disclosure will be further described in greater detail by reference to specific Examples, which should not be construed as in any way limiting the scope of the disclosure.

Example 1

[00114] A cement material was produced using a cement comprising 85% by total weight of precursor and 15% by total weight of a reagent. The reagent comprised a total of 32% CaO by weight and a total of 4% Na₂O by weight.

[00115] The normalised heat flow and the normalised heat released at room temperature, measured by isothermal calorimetry is shown in FIG. 2. This cement composition hardens in about 20 hours at 25°C after mixing and develops a

compressive strength of approximately 20 MPa 24 hours after casting and approximately 42 MPa after 28 days. According to FIG. 2, the diffusion period starts after approximately 27 ± 1 hours and continues beyond 63 days. At this point, the compressive strength reaches 49 MPa.

5 **[00116]** In certain cases, particularly when the temperature of the working environment exceeds 28 °C and the hardening time is shortened, the workability or pot life of mortars and concretes produced from the cement composition may be extended. Borax (e.g. solid, sodium tetraborate decahydrate, $\text{Na}_2\text{B}_4\text{O}_7 \cdot 10\text{H}_2\text{O}$) and
10 HIDS (e.g. suspension, biodegradable tetrasodium 3-Hydroxy-2,2'-iminodisuccinate, $\text{C}_8\text{H}_7\text{NO}_9 \cdot 4\text{Na}$) are materials that may be used as effective retarders for the hydration of the cement composition, slowing the progress of reactions and the hardening of the paste.

[00117] For example, the time required for the cement of Example 1 (as shown in FIG. 2) to reach a stiffness high enough to compromise workability is about 5 hours.
15 When up to 5% by weight of Borax (over the precursor content) and up to 0.5% by weight of HIDS (over the precursor content), are added to the cement composition, it is possible to delay the hardening of the eco-cement up to 25 hours and 5 hours, respectively, as shown in FIG. 3 and FIG.4, respectively. As depicted in FIGS. 3 and 4, the last exothermic peaks of the graphs can represent the beginning of the
20 deceleration/diffusion periods, a point from which the stiffness of the hydrated cement prevents the paste from flowing.

Example 2

[00118] An example cement composition is formed using blast furnace slag. The cement composition also comprises the below components, where the criteria for
25 each component are also listed below in respect to the associate component.

Sodium carbonate comprising:

- (a) $\geq 99\%$ by weight of Na_2CO_3 content
- (b) 100% of particles with an average grain size of $< 60 \mu\text{m}$
- (c) $D_{90} < 45 \mu\text{m}$

- (d) $D_{50} < 10 \mu\text{m}$ (preferably with $D_{25} < 5 \mu\text{m}$)

Anhydrous sodium metasilicate comprising:

- (a) 49% to 51% by weight of Na_2O content

Reactive magnesium oxide

- 5 (a) $\geq 95\%$ by weight of MgO content
- (b) specific surface area $\geq 32 \text{ m}^2/\text{g}$
- (c) 100% of particles with an average grain size of $< 60 \mu\text{m}$
- (d) $D_{90} < 45 \mu\text{m}$
- (e) $D_{50} < 10 \mu\text{m}$ (preferably with $D_{25} < 5 \mu\text{m}$)

10 Artificial hydrotalcite ($\text{Mg}_6\text{Al}_2\text{CO}_3(\text{OH})_{16} \cdot 4\text{H}_2\text{O}$)

- (a) 73% to 75% by weight of Al_2O_3 content
- (b) 25% to 27% by weight of MgO content
- (c) mass loss on ignition at $1,000^\circ\text{C} < 2.5\%$ by weight
- (d) specific surface area $\geq 85 \text{ m}^2/\text{g}$

- 15 (e) $D_{50} \leq 25 \mu\text{m}$ (preferably with $D_{25} < 5 \mu\text{m}$)

Casting Moulding and Performance Evaluation

[00119] According to an example embodiment of the cement composition, the present disclosure provides a cost-effective, one-part cement formulation that can use ferrous and non-ferrous calcium-rich slags activated by engineered sodium carbonate-based agents. Through this process, a sustainable, environmentally friendly, CO_2 -absorbing cement having high mechanical performance matrix (equivalent to OPC cement type CEM I, with strengths exceeding 12 MPa after 7 days and 32.5 MPa after 28 days) can be achieved. In addition, the cement composition can also incorporate the use of materials such as natural wollastonite fibre and sand-

based engineered aggregates to mitigate the deleterious effects of carbonation on the material's properties and also the use of specific reaction accelerators and retarders to control the hardening reaction speed.

[00120] Samples of plain and wollastonite-reinforced cement, as well as engineered aggregate-based mortar, were moulded by die casting to produce 40x40x80 mm rectangular samples and cylinders of 40 mm in diameter and 100 mm long for mechanical properties evaluation. In particular, evaluation of compressive strength is as per EN 12390 Part 3; evaluation of tensile strength is as per EN 12390 Part 6; and evaluation of setting time is as per ASTM C807.

[00121] Superior mechanical properties can be shown. As depicted in FIG. 6, plain non-carbonated cement produced by the example embodiment exhibits compressive strength exceeding 10 MPa after 1 day, 12 MPa after 7 days and 32.5 MPa after 28 days. As depicted in FIG. 7, plain non-carbonated cement produced by the example embodiment exhibits splitting tensile strength exceeding 1 MPa, 3 MPa and 2.5 MPa after 1, 7 and 28 days.

[00122] When 2.5% by volume of the cement composition of wollastonite is added to the example embodiment, the produced wollastonite-reinforced carbonated cement exhibits compressive strength exceeding 7 MPa, 50 MPa and 60 MPa after 1, 14 and 28 days, respectively, as shown in FIG. 8. The same wollastonite-reinforced carbonated cement also exhibits splitting tensile strength exceeding 1 MPa, 3 MPa and 3.5 MPa after 1, 14 and 28 days, respectively, as shown in FIG. 9.

[00123] For a carbonated mortar produced using the example embodiment and engineered aggregate at a ratio of 1:1 by weight of the engineered aggregate to the weight of the cement composition, compressive strength exceeding 7 MPa, 30 MPa and 35 MPa after 3, 14 and 28 days, respectively, are exhibited, as shown in FIG. 10. The same carbonated mortar also exhibits splitting tensile strength exceeding 0.5 MPa, 4 MPa and 5 MPa after 3, 14 and 28 days, respectively, as shown in FIG. 11

[00124] Further, the physical properties of the example embodiment are as follows:

Plain non-carbonated cement

- (a) Water absorption: $17.5 \pm 1.5\%$
- (b) Apparent density: $2.1 \pm 0.05 \text{ g/cm}^3$
- (c) Apparent porosity: $27 \pm 2\%$

Wollastonite-reinforced carbonated cement

- 5 (a) Water absorption: $21.5 \pm 0.5\%$
- (b) Apparent density: $2.3 \pm 0.05 \text{ g/cm}^3$
- (c) Apparent porosity: $33 \pm 1\%$

Carbonated mortar

- (a) Water absorption: $11.5 \pm 0.5\%$
- 10 (b) Apparent density: $2.6 \pm 0.1 \text{ g/cm}^3$
- (c) Apparent porosity: $23 \pm 1\%$

[00125] As used herein, the term “about” in the context of concentration of a substance, size of a substance, length of time, or other stated values means +/- 5% of the stated value, or +/- 4% of the stated value, or +/- 3% of the stated value, or +/-
15 2% of the stated value, or +/- 1% of the stated value, or +/- 0.5% of the stated value.

[00126] Throughout this disclosure, certain embodiments may be disclosed in a range format. It should be understood that the description in range format is merely for convenience and brevity and should not be construed as an inflexible limitation on the scope of the disclosed ranges. Accordingly, the description of a range should be
20 considered to have specifically disclosed all the possible sub-ranges as well as individual numerical values within that range. For example, description of a range such as from 1 to 6 should be considered to have specifically disclosed sub-ranges such as from 1 to 3, from 1 to 4, from 1 to 5, from 2 to 4, from 2 to 6, from 3 to 6 etc., as well as individual numbers within that range, for example, 1, 2, 3, 4, 5, and 6. This
25 applies regardless of the breadth of the range.

[00127] The disclosure illustratively described herein may suitably be practiced in the absence of any element or elements, limitation or limitations, not specifically disclosed herein. Thus, for example, the terms "comprising", "including", "containing", etc. shall be read expansively and without limitation. Additionally, the terms and expressions employed herein have been used as terms of description and not of limitation, and there is no intention in the use of such terms and expressions of excluding any equivalents of the features shown and described or portions thereof, but it is recognized that various modifications are possible within the scope of the disclosure claimed. Thus, it should be understood that although the present disclosure has been specifically disclosed by preferred embodiments and optional features, modification and variation of the disclosures embodied herein disclosed may be resorted to by those skilled in the art, and that such modifications and variations are considered to be within the scope of this disclosure.

[00128] The disclosure has been described broadly and generically herein. Each of the narrower species and subgeneric groupings falling within the generic disclosure also form part of the disclosure. This includes the generic description of the disclosure with a proviso or negative limitation removing any subject matter from the genus, regardless of whether or not the excised material is specifically recited herein.

[00129] Unless otherwise indicated, all technical and scientific terms used herein have the same meaning as commonly understood to one of ordinary skill in the art to which the disclosure belongs.

[00130] It is contemplated that any part of any aspect or embodiment discussed in this specification can be implemented or combined with any part of any other aspect or embodiment discussed in this specification, so long as such those parts are not mutually exclusive with each other.

[00131] It should be recognized that features and aspects of the various examples provided above can be combined into further examples that also fall within the scope of the present disclosure.

[00132] When used in this specification and statements, the terms "comprises" and "comprising" and variations thereof mean that the specified features, steps or

integers are included. As used herein, the term “comprising” means “including.” Variations of the word "comprising", such as “comprise” and “comprises,” have correspondingly varied meanings. Thus, for example, a composition “comprising” X may consist exclusively of X or may include one or more additional unrecited components.

[00133] The terms are not to be interpreted to exclude the presence of other features, steps or components. Further, the singular forms "a", "an", and "the" are intended to include the plural forms as well, unless the context clearly indicates otherwise.

[00134] Use of language such as "at least one of X, Y, and Z," "at least one of X, Y, or Z," "at least one or more of X, Y, and Z," "at least one or more of X, Y, and/or Z," or "at least one of X, Y, and/or Z," is intended to be inclusive of both a single item (e.g., just X, or just Y, or just Z) and multiple items (e.g., {X and Y}, {X and Z}, {Y and Z}, or {X, Y, and Z}). The phrase "at least one of" and similar phrases are not intended to convey a requirement that each possible item must be present, although each possible item may be present.

[00135] The disclosure may also broadly consist in the parts, elements, steps, examples and/or features referred to or indicated in the specification individually or collectively in any and all combinations of two or more said parts, elements, steps, examples and/or features. In particular, one or more features in any of the embodiments described herein may be combined with one or more features from any other embodiment(s) described herein.

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CLAIMS:

1. A cement composition comprising:

80% to 90% by weight of a cement precursor comprising one or more alkali-activatable aluminosilicate materials; and

5 10% to 20% by weight of a reagent comprising Na_2CO_3 ;

wherein the reagent comprises an accelerator.
2. The cement composition of claim 1, wherein the cement composition is hydrated or further comprises water in an amount of between 37% to 42% or 40% by weight of the total cement composition.
- 10 3. The cement composition of claim 1 or 2, wherein the cement composition is configured to, after hydration, absorb carbon dioxide to form a carbonate-based matrix.
4. The cement composition of any one of claims 1 to 3, wherein the accelerator comprises: anhydrous sodium metasilicate ($\text{Na}_2\text{O} \cdot \text{SiO}_2$), reactive magnesium oxide
15 (MgO), calcium hydroxide ($\text{Ca}(\text{OH})_2$), slaked lime, synthetic hydrotalcite, or combinations thereof.
5. The cement composition of claim 4,

wherein a total amount of Na_2O is between 3.5% to 5% by weight of the reagent;

wherein a total amount of MgO does not exceed 7% by weight of the reagent;

20 wherein a total amount of $\text{Ca}(\text{OH})_2$ does not exceed 5.2% by weight of the reagent;
and/or

wherein a total amount of the synthetic hydrotalcite does not exceed 0.08% by weight of the reagent.
6. The cement composition of claim 5,

wherein a purity of the anhydrous sodium metasilicate is at least 96%, comprising an amount of Na_2O between 49.5% to 51.5% by weight of the anhydrous sodium metasilicate, and an amount of SiO_2 between 45% to 47% by weight of the anhydrous sodium metasilicate;

- 5 wherein an amount of the MgO is at least 97% by weight of the reactive magnesium oxide or at least 99% by weight of the reactive magnesium oxide, a specific surface area of the reactive magnesium oxide is at least $32 \text{ m}^2/\text{g}$, and a particle size of the reactive magnesium oxide does not exceed $125 \mu\text{m}$;

- 10 wherein an amount of the $\text{Ca}(\text{OH})_2$ is at least 95% by weight of the calcium hydroxide or slaked lime or at least 97% by weight of the calcium hydroxide or slaked lime, and the particle size of the calcium hydroxide or slaked lime does not exceed $45 \mu\text{m}$; and/or

- 15 wherein the synthetic hydrotalcite is magnesium-aluminium hydroxycarbonate comprises 73% Al_2O_3 by weight and 26% MgO by weight, or 74% Al_2O_3 by weight and 27% MgO by weight, and the particle size of the synthetic hydrotalcite does not exceed $40 \mu\text{m}$.

7. The cement composition of any one of claims 1 to 6,

wherein the Na_2CO_3 comprises at least 97% by weight or at least 99% by weight of a source material; and/or

- 20 wherein the particle size of the source material does not exceed $100 \mu\text{m}$.

8. The cement composition of any one of claims 1 to 7, wherein the cement precursor comprises CaO in an amount between 35% to 52% by weight of the cement precursor.

- 25 9. The cement composition of any one of claims 1 to 8, wherein the grain size of the cement precursor does not exceed $45 \mu\text{m}$ or does not exceed $30 \mu\text{m}$.

10. The cement composition of any one of claims 1 to 9, wherein an amount of amorphous phase of the cement precursor is at least 85% by weight of the cement precursor.

11. The cement composition of any one of claims 1 to 10,

wherein the cement precursor comprises CaO, MgO, Al₂O₃, SiO₂, or a combination thereof, and

5 wherein a ratio of an amount of the CaO and the MgO in the cement precursor to an amount of the Al₂O₃ and the SiO₂ in the cement precursor is at least 7:10, by weight.

12. The cement composition of any one of claims 1 to 11,

wherein the cement precursor comprises 85% to 88% by weight of the cement composition; and

wherein the reagent comprises 12% to 15% by weight of the cement composition.

10 13. The cement composition of any one of claims 1 to 12,

wherein the cement precursor comprises slag comprising blast-furnace slag, ferrous slag, non-ferrous slag, municipal solid waste slags, slag from energy facility, incineration products, or combinations thereof; and/or

15 wherein the cement precursor comprises glass comprising recycled glass, waste glass, or both.

14. The cement composition of any one of claims 1 to 13, further comprising a retarder comprising borax and/or tetrasodium 3-hydroxy-2,2'-iminodisuccinic acid (HIDS) configured to reduce a rate of cement setting.

15. The cement composition of claim 14,

20 wherein the borax has a molecular weight of between 370 g/mol to 390 g/mol, a specific gravity of between 1.6 g/cc to 1.8 g/cc, and an onset of water loss at between 60 °C to 64 °C; and/or

wherein the HIDS has a molecular weight of between 340 g/mol to 360 g/mol, a density of between 1.4 g/cm³ and 1.6 g/cm³ at 20 °C, and a concentration of between
25 49% to 51% by weight in aqueous solution.

16. The cement composition of any one of claims 1-15, wherein the cement composition is rich in calcium.

17. A reinforced cement composition comprising:

the cement composition of any one of claims 1 to 16; and

5 a filler material comprising wollastonite fibres, engineered aggregates comprising silicon dioxide, or both.

18. The reinforced cement composition of claim 17,

wherein an amount of the wollastonite fibres is between 2% to 3% by volume or 2.5% by volume; and/or

10 wherein a ratio between an amount of the cement composition to an amount of the engineered aggregates is between 1:0.8 and 1:3.5 by weight, between 1:0.9 to 1:2 by weight, or 1:1 by weight.

19. The reinforced cement composition of claim 17 or claim 18,

15 wherein an amount of SiO_2 in the wollastonite fibres is between 49% to 53% by weight of the wollastonite fibres or 51% by weight of the wollastonite fibres;

wherein an amount of Al_2O_3 in the wollastonite fibres does not exceed 1% by weight of the wollastonite fibres;

wherein an amount of CaO in the wollastonite fibres is between 43% to 47% by weight of the wollastonite fibres or 45% by weight of the wollastonite fibres;

20 wherein an amount of MgO in the wollastonite fibres does not exceed 2% by weight of the wollastonite fibres;

wherein an amount of Fe_2O_3 in the wollastonite fibres does not exceed 0.5% by weight of the wollastonite fibres; and/or

wherein the engineered aggregates comprise quartz, quartz sands, or both.

20. The reinforced cement composition of any one of claims 17 to 19, wherein a loss on ignition of the wollastonite fibres does not exceed 5% by weight at 1000°C, an aspect ratio of wollastonite fibres is between 15:1 to 21:1 or 19:1, and the grain size of the wollastonite fibres does not exceed 75 µm.

5 21. A method for producing cement from the cement composition of any one of claims 1 to 16, comprising:

homogenizing the reagent by dry shear mixing ;

homogenizing the cement precursor by dry shear mixing;

homogenizing the reagent and the cement precursor to form cement powder;

10 homogenizing water with the cement powder to form the cement.

22. A method of producing reinforced cement from the reinforced cement composition of any one of claims 17 to 20, comprising:

homogenizing the reagent by dry shear mixing;

homogenizing the cement precursor by dry shear mixing;

15 homogenizing the wollastonite fibers with the reagent and the cement precursor to form cement powder; and

homogenizing water with the cement powder to form the reinforced cement.

20 23. The method of claim 21 or 22, wherein the reagent is mixed for 18 minutes to 22 minutes at 180 RPM to 220 RPM, and/or wherein the cement precursor is mixed for 9 minutes to 11 minutes at 200 RPM or less, and/or the water is homogenized in an amount between 37% to 42% or 40% by weight of the total composition.

24. A method for producing cement from the reinforced cement composition of any one of claims 17 to 20, comprising:

25 homogenizing the reagent by dry shear mixing for 18 minutes to 22 minutes at 180 RPM to 220 RPM;

homogenizing the cement precursor by dry shear mixing for 9 minutes to 11 minutes at 200 RPM or less;

homogenizing the reagent and the cement precursor to form cement powder;

homogenizing the engineered aggregates with the water in an amount of 2.5% to 3.5% by weight of the engineered aggregates to form a mixture; and

homogenizing the mixture with the cement powder.

25. The method of claim 24, further comprising: dissolving a superplasticiser comprising polycarboxylate in water at an amount of 0.9% to 1.1% by weight of the engineered aggregates, the water in an amount of 2.5% to 3.5% by weight of the engineered aggregates and/or homogenizing the engineered aggregates with the superplasticiser and the water to form a mixture.

26. The cement produced by the method of any one of claims 21 to 25, wherein a density of the cement is in a range of 2.1 to 2.6 g/cm³.

27. The cement produced by the method of any one of claims 21 to 26, wherein a compressive strength of the cement 24 hours after mixing is at least 10 MPa; and/or

wherein the splitting tensile strength of the cement 24 hours after mixing is at least 1 MPa.

28. The cement produced by the method of any one of claims 21 to 27, wherein the cement is a calcium carbonate cement.