

METHOD OF MANUFACTURING A BIOCHAR AGGREGATE AND BIOCHAR AGGREGATE MATERIAL OBTAINED

Field of Invention

- 5 The present invention relates to a carbon-negative aggregate comprising biochar, and its uses, such as in the building and highways industries.

Background of the Invention

- 10 Presently around 45% of the CO₂ emitted by humans remains in the atmosphere, which is a significant factor behind global warming.

- In addition to (or as an alternative to) avoiding the emission of CO₂, it is also important to develop methods to remove some of the CO₂ already present in the atmosphere. One such
15 key method is carbon sequestration, which refers to the capturing, removal and storage of carbon dioxide (CO₂) from the earth's atmosphere.

- Biochar has been recognised as a potential candidate for a solid medium to be used in carbon sequestration methods.

- 20 Biochar is the solid material obtained from the thermochemical conversion of biomass in an oxygen-limited environment (i.e., pyrolysis). The material structure is generally amorphous, but does contain some local crystalline structures of highly conjugated aromatic compounds. The composition of the material is predominantly carbon, but also contains hydrogen,
25 oxygen and nitrogen, along with trace heavy metals. The appearance of the material is generally a black solid. Biochar can be made from almost any source of carbon originating from a bio-based source (including agricultural waste, wood, etc.).

- As a result of undergoing a pyrolysis process, the resulting biochar can store carbon in a
30 much more stable way than the starting biomass within a given timeframe of years to decades. On the other hand, most biomass is easily broken down by bacteria, fungi and natural weathering in the environment, releasing back into the atmosphere the carbon dioxide that has been stored in the biomass originally alongside other decomposition greenhouse gases such as methane and nitrogen oxides. Biochar, instead, is much more
35 difficult for these organisms to break down and therefore the carbon removed from the atmosphere is stored within the biochar for over 100 years, which is seen as permanent carbon storage.

Although biochar has the potential to be used for sequestering carbon dioxide, there are some drawbacks associated with using pure biochar in industry. For example, pure biochar tends to be brittle and weak, making it difficult for use in industrial mixing and conveying.

- 5 In addition, mixing biochar with fine and coarse natural aggregate can result in a grinding effect on the biochar, causing potentially adverse effects in the application. For instance, in the case of asphalt compositions, the biochar grinding increases the surface area which can stiffen the asphalt composition due to more bitumen being required to coat the biochar which limits the amount of biochar that can be added before significant changes in properties
10 are observed. Typically, as the biochar content increases, the penetration depth decreases, the softening point increases and ductility decrease (Gan *et al*, PLoS One, 2021, 16(2) e0247390). This results in more bitumen being used in the composition or for it to be processed at higher temperatures to allow for adequate mixing and compaction when laid. The increased bitumen use is wasteful in terms of resource efficiency especially in a global
15 environment looking to decrease oil extraction and resulting bitumen and the increase in temperature would result in a higher energy use (which could also cause unwanted bitumen oxidation), which increases costs and has a detrimental environmental impact.

- A further disadvantage of pure biochar is the significant health and safety risk associated
20 with handling and/or being exposed to the dust and fine particles typically being produced by the biochar. The fine dust is also known to be highly flammable which has to be kept moist to prevent unwanted ignition.

- In order to enable the use of biochar for sequestering carbon dioxide at larger scale, there
25 is a need to develop new methods to convert biochar into a form that is suitable, efficient, and safe to be used in industry, such as the building industry (e.g., infrastructure sector).

Summary of the invention

- 30 The present invention seeks to address the problem outlined above by providing a carbon-negative composite biochar aggregate useable in the building industry. The use of biochar in the manufacture of an aggregate offers distinct advantages over using pure biochar, such as control of aggregate properties such as strength, density, surface area, particle size, water absorption and ease of handling.

- 35 The biochar aggregate and uses thereof provide an environmental benefit which is directly related to the amount of stored carbon in the biochar. Assuming that almost all of the carbon in the biomass used to create the biochar has come from carbon dioxide from the atmosphere, it is estimated that the biochar used to manufacture the aggregate of the

invention has removed up to x3.7 by weight CO₂ based on a carbon content of 100%. The exact percentage of carbon removal depends on the carbon purity of the biochar. For example, a biochar of 79 wt% carbon has a carbon removal value of 2.9 kg CO₂/kg.

- 5 The biochar aggregate of the present invention may be incorporated into certain products to render those products carbon-neutral or carbon-negative, such as by replacing a component part (e.g., in concrete compositions, roads). For example, it is expected that the addition of the biochar aggregate of the invention to asphalt even at between 2-5% w/w reduces the carbon footprint of the asphalt at least to net-zero.

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Accordingly, a first aspect of the invention is a method of manufacturing a biochar aggregate comprising the steps of:

- (i) mixing biochar and at least one binder to form a first composition;
- (ii) optionally providing a second composition comprising water;
- 15 (iii) optionally mixing together the first and second compositions to form a pre-mixture; and
- (iv) agglomerating the first composition or, if present, the pre-mixture to form the biochar aggregate.

20 A second aspect of the invention is a biochar aggregate obtainable by a method of the first aspect of the invention.

A third aspect of the invention is a biochar aggregate comprising:

- (i) biochar; and
- (ii) at least one binder.

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A fourth aspect of the invention is a composition comprising the biochar aggregate of the second or third aspects of the invention (including all embodiments thereof).

30 A fifth aspect of the invention is a method of manufacturing a composition of the fourth aspect of the invention, comprising the step of mixing the biochar aggregate of the second or third aspects of the invention (including all embodiments thereof) with at least one other component (e.g., a binder, resin, or paint).

35 A sixth aspect of the invention is the use of a biochar aggregate of the second or third aspects of the invention (including all embodiments thereof) to:

- (i) place the biochar aggregate in a location such that the biochar aggregate is exposed to the natural atmosphere; and/or
- (ii) fill a depression in the ground or to change the elevation of the ground; or
- (iii) insulate a building component.

A seventh aspect of the invention is an engineered fill, geological fill or insulation material, comprising a biochar aggregate of the second or third aspects of the invention.

- 5 An eight aspect of the invention is the use of a biochar aggregate of the second or third aspects of the invention in reducing pollution.

Description of the Figures

- 10 Figure 1 shows a biochar aggregate of the invention.

Figure 2 shows a diagram of an active carbonation using Flue gas/DAC.

- 15 Figure 3 shows a diagram of a modified pellet mill that involves conducting the pelletising under a CO₂ atmosphere.

Figure 4 shows a schematic for a modified pan pelletising under a CO₂ atmosphere.

Detailed description of the invention

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Unless indicated otherwise, all technical and scientific terms used herein will have their common meaning as understood by one of ordinary skill in the art to which this invention pertains.

- 25 The term "comprising" or variants thereof will be understood to imply the inclusion of a stated element, integer or step, or group of elements, integers or steps, but not the exclusion of any other element, integer or step, or group of elements, integers or steps. References to "comprising" also encompass providing basis for "consisting".

- 30 The term "consisting" or variants thereof is to be understood to imply the inclusion of a stated element, integer or step, or group of elements, integers or steps, and the exclusion of any other element, integer or step or group of elements, integers or steps.

- 35 The term "about" herein, when qualifying a number or value, is used to refer to values that lie within $\pm 5\%$ of the value specified. For example, if a particle size range is specified to be about 60 μm to about 4 mm, particle sizes of 57 μm to 4.2 mm are included.

"wt%" is a common abbreviation in the art to mean the "weight %" with respect to the total weight of the article/material referred to.

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In a first aspect, the invention provides a method of manufacturing a biochar aggregate comprising the steps of:

- (i) mixing biochar and at least one binder to form a first composition;
- (ii) optionally providing a second composition comprising water;
- 5 (iii) optionally mixing together the first and second compositions to form a pre-mixture; and
- (iv) agglomerating the first composition or, if present, the pre-mixture to form the biochar aggregate.

The biochar aggregate obtainable by the method of the first aspect of the invention may be referred to herein as the "biochar aggregate of the invention". For the avoidance of doubt, 10 the biochar aggregate of the second and third aspects of the invention may also be referred to herein as the "biochar aggregate of the invention".

The skilled person will understand that the biochar aggregate of the invention is a composite aggregate (as such, the biochar aggregate of the invention may be referred to as the 15 "composite biochar aggregate" of the invention, or similar). A "composite" material refers to a material made up of at least two constituent materials.

The term "aggregate" is used herein to refer to particulate material.

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"Biochar" is a solid material which is a stable carbon derivative, produced from the thermochemical conversion of plant and/or animal biomass in an oxygen-limited environment (i.e., pyrolysis). The composition of biochar is predominantly carbon, but also contains hydrogen, oxygen and nitrogen making up the other atomic components, along 25 with trace heavy metals.

"Pyrolysis" refers to the chemical decomposition of organic materials at elevated temperatures in a predominantly inert atmosphere (little to no oxygen or steam). Pyrolysis produces condensable liquids, non-condensable gases, and biochar. Pyrolysis processes 30 may be categorised into slow pyrolysis, fast pyrolysis and flash pyrolysis. "Slow pyrolysis" refers to slowly heating feedstock to temperatures of between 300-500°C at a low heating rate (0.1-1°C/second) by using large particle sizes or piles of biomass (>1 cm) for a time period from minutes, to hours, to days. "Fast pyrolysis" refers to heating feedstock to a temperature between 400-900°C at a more rapid heating rate (between 1°C/second and 100°C/second) by using small particle sizes (<6 mm) for a time period from seconds to tens 35 of seconds. "Flash pyrolysis" refers to very rapidly heating feedstock to a temperature between 400-900°C (>1000° C/second) by using very small particle sizes (<1 mm) or using ablation reactors for a time period of less than one second.

In some embodiments, the biochar used in step (i) was obtained by slow pyrolysis, fast pyrolysis, or flash pyrolysis, such as fast pyrolysis. As such, suitably the method comprises the pre-steps of (i) providing biomass; (ii) subjecting the biomass to slow pyrolysis, fast pyrolysis, or flash pyrolysis to obtain biochar.

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Biochar can be made from almost any source of carbon derived from a bio-based source, which results in biochars having different properties. In some embodiments, the biochar is derived from agricultural wastes, wood (e.g. hardwood, softwood), sawdust, particle board, manure, leaves, hemp, grass, rice husks, hay, straw, coconut husks, algae, mycelium, fungi, seagrasses, or seaweeds, or mixtures of two or more thereof, preferably wood (in particular wood that has been sustainably sourced).

In some embodiments, the biochar is derived from waste, by-products, and/or low value materials, preferably waste (e.g., agricultural waste).

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Alternatively, the biochar raw material can be grown and produced for the purpose of producing biochar, especially if large scale volumes of biochar from a specific source of low-availability is required. Although growing trees and plants solely for this purpose might be associated with some environmental issues, using sea-based sources negates many of such issues, with algal sources able to be manufactured far inland in algal reactors. As such, in some embodiments, the biochar is derived from a sea-based source, such as algae.

As mentioned above, the environmental impact of the biochar aggregate is directly related to the amount of carbon stored in the biochar. The carbon percentage of the biochar may be referred to in terms of purity, with 100% pure biochar having a carbon dioxide removal potential of 3.7 kg CO₂/kg biochar. Typical carbon purities have a carbon content of around 65%-90%. The purity of the biochar is determined by many factors such as maximum temperature, heating rates and time, raw material source and other factors.

In some embodiments, the biochar aggregate has a carbon dioxide removal potential of at least 0 kg CO₂/kg biochar, such as at least 0.1 kg CO₂e/kg, 0.2 kg CO₂e/kg, 0.3 kg CO₂e/kg, 0.4 kg CO₂e/kg, 0.5 kg CO₂e/kg, 0.6 kg CO₂e/kg, 0.7 kg CO₂e/kg, 0.8 kg CO₂e/kg, 0.9 kg CO₂e/kg, 1.0 kg CO₂e/kg, 1.1 kg CO₂e/kg, 1.2 kg CO₂e/kg, 1.3 kg CO₂e/kg, 1.4 kg CO₂e/kg, 1.5 kg CO₂e/kg, 1.75 kg CO₂e/kg, 2.0 kg CO₂e/kg, 2.25 kg CO₂e/kg, 2.5 kg CO₂e/kg, 2.75 kg CO₂e/kg, or 3.0 kg CO₂e/kg biochar.

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In some embodiments, the biochar aggregate has a carbon footprint of -2 kg CO₂e/kg to 0.2 kg CO₂e per kg of biochar aggregate, preferably -1.5 kg CO₂e/kg to 0 kg CO₂e per kg of biochar aggregate, more preferably -1 kg CO₂e/kg to 0 kg CO₂e per kg of biochar

aggregate. The skilled person would be able to determine the carbon footprint of a given composition using standard techniques known in the art, such as by carrying out an industry standard life-cycle assessment (LCA). A skilled practitioner would take the carbon dioxide removed as a negative value during the carbon footprint calculations.

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The at least one binder may comprise cementitious binder or non-cementitious binder, preferably cementitious binder.

10 "Cementitious binder" refers to a material or substance that adheres other materials together to form, set and harden the resulting concrete composition. "Cementitious binder" encompasses cement, a slag (such as ground granulated blast furnace slag (GGBS)), pulverised fly ash (also known as pulverised fuel ash), Portland cement, micro-silica, calcined clay, pozzolanic material or geopolymers.

15 Often, the cement comprises any one or a mixture of calcium oxide, calcium hydroxide and calcium silicate. Typically, the cement is a hydraulic cement such as Portland cement, which reacts with water via Pozzolanic reactions to cure and set. Portland cement is usually made by heating limestone and clay minerals to form a clinker, which is ground and contacted with gypsum. Portland cement typically consists of at least two-thirds by mass of calcium
20 silicates, with the remainder consisting of aluminium- and iron-containing compounds. The ratio of CaO to SiO₂ within Portland cement is at least 2:1.

"Geopolymers" are amorphous, alumina-silicate binder materials. "Geopolymers" encompass metakaolin.

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Suitably, the cementitious binder used in the present invention may comprise one or a mixture of one or more of cement, a slag (such as GGBS), fly ash (also known as pulverised fly ash), micro-silica, calcined clay, pozzolans, or geopolymers. In one embodiment, the cementitious binder may comprise cement and/or a slag (such as GGBS). More preferably,
30 the cementitious binder comprises cement. In one embodiment, the cement comprises Portland cement.

Preferably, the cementitious binder is GGBS or High Strength Cement, preferably GGBS.

35 In some embodiments, the at least one binder comprises non-cementitious binder.

In some embodiments, the non-cementitious binder is a dehydrated gypsum (i.e. calcium sulphate, also known as Anhydrite) or a lignosulphonate.

Depending on the moisture level of the biochar and the specific binder being used, the addition of water may be necessary. Moisture level is known in the field to refer to the quantity of water contained in a material. In some embodiments, the method comprises step (ii) (i.e., providing a second composition comprising water) and step (iii) (i.e., mixing
5 together the first and second compositions to form a pre-mixture). The skilled person will understand that in such embodiments, the agglomerating referred to in step (iv) is agglomerating of the pre-mixture obtained in step (iii).

The skilled person will also understand that steps (ii) and (iii) are either both required or
10 neither of them is required.

In particular embodiments, when the moisture level of the biochar in step (i) is above 25% by weight (preferably above 20 wt% such as 15 wt% or above), the method does not comprise steps (ii) and (iii).
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In particular embodiments, when the moisture level of the biochar in step (i) is less than 15% by weight (such as less than 10 wt%), the method comprises steps (ii) and (iii).

In one embodiment, the method of manufacturing a biochar aggregate comprises the steps
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- (i) providing biochar with a moisture level of more than 10 wt% and mixing said biochar and at least one binder to form a first composition; and
- (ii) agglomerating the first composition to form the biochar aggregate. Preferably the biochar provided in step (i) has a moisture level of more than 12 wt%, such as more than
25 14 wt%. Preferably the biochar provided in step (i) has a moisture level of between 10 and 20 wt%, preferably between 12 and 18 wt%, more preferably between 13 and 17wt%, such as around 15 wt%.

In one embodiment, the method of manufacturing a biochar aggregate comprises the steps
30 of:

- (i) providing biochar with a moisture level of less than 15 wt% and mixing said biochar and at least one binder to form a first composition;
- (ii) optionally providing a second composition comprising water;
- (iii) optionally mixing together the first and second compositions to form a pre-mixture; and
35 (iv) agglomerating the first composition or, if present, the pre-mixture to form the biochar aggregate. Preferably the biochar provided in step (i) has a moisture level of less than 12 wt%, more preferably less than 10 wt%, yet more preferably less than 5 wt%, such as less than 3 wt%, such as less than 1 wt%. Suitably the biochar is essentially dry.

The moisture level of the biochar and/or biochar aggregate may also affect the ability of the biochar to form strong and relatively uniform pellets, without significant fines content.

5 Suitably the biochar aggregate may have a moisture level of between 3 and 30 wt%, more preferably between 5 and 25 wt%, such as between 8 and 23 wt%, for example between 10 and 20 wt%. In more particular embodiments, the biochar aggregate has a moisture level of about 15 wt%.

10 The preferred binder GGBS represents a lower carbon alternative to Ordinary Portland Cement (OPC). The use of GGBS allows to reduce the embodied carbon of the aggregate and of the final concrete, compared to the use of OPC. GGBS is characterised by an extremely slow curing rate when hydrated and is typically activated by an alkaline solution which increases the curing rate and compressional strengths.

15 Although the slow curing rate of GGBS could represent an obstacle, during the production of the composite aggregate (such as via pelletisation, which generates high pressure and heat), the pressure and heat improves the aggregates strength due to compaction of the particle.

20 Early strength and final strength of cementitious based biochar aggregate of the invention can also be improved by the use of an activator solution, preferably obtained by adding sodium hydroxide, sodium carbonate or a combination of. A secondary advantage of this process is when scaled up (>0.5 T), the aggregate can retain the heat generated by the process and the heat generated through the hydration reaction for multiple days, this is due
25 to the inherent insulating properties of the biochar this causes improved compressional strengths.

In some embodiments, the second composition further comprises at least one inorganic base.

30 The "inorganic base" acts as an activator. Inorganic bases include a class of inorganic compounds with the ability to react with, that is neutralize, acids to form salts. These compounds comprise strong and weak bases, such as metal hydroxides, alkali metal hydroxides, ammonium hydroxides, alkali metal carbonates or bicarbonates. The term is
35 intended to comprise also substances that can generate bases, i.e. hydroxides, when in contact with water, such as metal and alkali metal oxides, alkaline silicates.

Suitably, the at least one inorganic base comprises an alkali hydroxide, alkali oxide, alkali carbonate, alkaline silicates or a mixture thereof. In some embodiments, the alkali is sodium, potassium or calcium.

- 5 Preferably the at least one inorganic base comprises sodium hydroxide, sodium carbonate or a mixture thereof.

In some embodiments of the method, when particular cementitious binders are used, an inorganic base is not required to cure the binder. As such, suitably there is provided a
10 method that does not require an inorganic base in step (ii) and the second composition. Exemplary cementitious binders include Portland cement.

The first composition or, if present, the pre-mixture obtained in step (iii) is agglomerated to form the biochar aggregate. "Agglomeration" refers to the process of accumulating
15 material into larger cohesive units. Suitably the agglomerating comprises pressure or non-pressure agglomeration of the first composition or, if present, the pre-mixture to form the biochar aggregate.

In some embodiments, the agglomeration comprises compressing and heating to form the
20 biochar aggregate. As such, the first mixture or, if present, the pre-mixture may be compressed and heated to form the biochar aggregate.

Compression is achieved by applying a force to the granulated mixture to combine it. Heat might be generated from the compression process or heat may be applied after the
25 formation of the aggregate by compression.

Suitably, compression and heating may be a single process. Preferably, the agglomerating, such as the compression, and optionally heating, comprises pelletisation. Typically during certain compression methods, including pelletisation, the heating required is generated
30 naturally from friction during the compression of the mixture being pushed through the die. However, further heating can also be applied after compression to keep the pellets warm for longer to further increase the curing speed.

"Pelletisation" is the process of compressing material into the form of a pellet. Suitably, the
35 pelletisation is die mill pelletisation, pan pelletisation, briquetting or extrusion pelletisation. Die mill pelletisation refers to converting granulated material into free flowing pellets. "Pan pelletisation" refers to mixing material (e.g. finely ground material or seed pellets) with a binder and agitating the resulting mixture until pellets of desired size have formed. The centrifugal force experienced by the pellets against the edge of the pelletiser creates

compression of the particles into a pellet. In addition, continually rotating and falling would compress and increase the density the pellets each time the aggregate falls while rotating round the pan. "Briquetting" refers to compressing material into a desired form, such as a pellet. "Extrusion pelletising" (also referred to as "compounding") refers to extruding a mixture, then passing the mixture onto a pelletiser such as a granulator to convert the extrudate into pellets.

In some embodiments, the pelletisation is die mill pelletisation (also known as press pelletisation), pan pelletisation, briquetting, or extrusion pelletising. Preferably, the pelletisation is die mill pelletisation.

Alternatively, compression and heating may be separate, so that they are carried out as part of separate processes, and/or are carried out at different locations or a different time. In such cases, compression is first carried out, followed by heating. Therefore, in some embodiments, compression and heating are separate processes, wherein compression is carried out before heating. Heat is advantageous to increase the curing of the pellets.

In some embodiments, the compression is done by pelletisation.

In some embodiments, the biochar aggregate is in the form of a pellet. In alternative embodiments, the biochar aggregate is in the form of a briquette.

In some embodiments, the agglomeration comprises masonry blockwork. In particular such embodiments, the first mixture or, if present, the pre-mixture (optionally comprising other aggregates such as sand) is run through a blockwork plant, cured, and subsequently crushed and screened. This method of production is able to create large (>10 mm) irregular shapes. When the biochar aggregate is in this form (i.e., the irregular shapes), it may be referred to as a "crush". For the avoidance of doubt, as used herein, the term "biochar aggregate pellet" includes the biochar aggregate in the form of "crush".

In some embodiments, the method of the invention does not comprise sintering of the biochar (e.g. does not comprise a step carried out after steps (i) to (iv) comprising sintering of the biochar). In particular, it would not be beneficial for the method of the invention to include sintering, as sintering would convert the biochar back into CO₂, thus removing the majority of the carbon-benefit of the aggregate.

The skilled person will understand that "sintering" refers to a process comprising heating the biochar to high temperature such that biochar particles are compacted. Sintering of the biochar may occur at temperatures such as 900 to 1300 °C.

In particular embodiments, the method of the invention does not comprise sintering of the biochar, such as sintering at a temperature of 900 to 1300 °C.

- 5 In some embodiments, the method of the invention does not comprise calcination of the biochar (e.g. does not comprise a step carried out after steps (i) to (iv) comprising calcination of the biochar).

"Calcination" of the biochar may occur at high temperatures, such as 1000 to 1300 °C.

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The skilled person will understand that in such embodiments, the method of the invention does not comprise a step of calcination to form ceramsite.

- 15 In more particular embodiments, the method of the invention does not comprise sintering or calcination of the biochar.

In some embodiments, the method of the invention further comprises one or more of the following pre-steps:

- (i) receiving biochar (e.g., from a biochar producer);
20 (ii) reducing the moisture content of the biochar, such as to between 10 and 20 wt%;
(iii) granulating the biochar to obtain a mixture of biochar particles;
(iv) treating the mixture of biochar particles to form a treated mixture;
(v) sieving the mixture of biochar particles to size separate the biochar particles;
(vi) reforming the mixture by contacting the biochar particles of different sizes with one
25 another.

- In one embodiment, the method comprises the pre-steps (i), (v) and (vi). In another embodiment, the method comprises the pre-steps (i), (iii), (v) and (vi). In a further embodiment, the method comprises the pre-steps (i), (iv), (v) and (vi). In another
30 embodiment, the method comprises the pre-steps (ii) and (iii). In a further embodiment, the method comprises the pre-steps (i), (ii), (iii), (iv), (v) and (vi).

- The production of biochar typically forms dusty particles with high carbon contents at temperatures of up to 400°C. In this state, the material is highly flammable. To combat
35 this, producers quench the biochar with water which lowers the temperature, however if too much water is used, then it can make post-processing difficult (e.g., the processing according to the first aspect of the invention).

As such, in some embodiments, the method comprises pre-step (ii) i.e., the pre-step of reducing the moisture content of biochar. In some embodiments, the method comprises pre-steps (ii) and (iii), and pre-step (ii) is carried out before step (iii) (i.e. the moisture is reduced before granulation), otherwise, there is the risk that the high-moisture biochar can clog the machinery used in producing the biochar aggregate.

Preferably, the method comprises pre-step (i) and the biochar received in pre-step (i) has a low moisture (e.g. less than 20% by weight, such as less than 18wt%, or less than 15wt%). In preferred such embodiments, the method does not comprise step (ii), as it is not necessary to reduce the moisture of such biochars.

Since biochar can be made from a wide range of sources, the resulting particle size and shape would need to be homogenised using particle reduction techniques such as granulation. As such, in some embodiments, the method comprises pre-step (iii).

The term "granulating" refers to forming into particles, i.e. discrete, solid pieces, and may be achieved by shredding (tearing or cutting), milling (pressing, crushing, hammer milling and/or grinding) and chipping (breaking off pieces).

Granulating may be achieved by any method that reduces the size of the biochar (e.g., biochar chunks) of larger size and forms it into the desired smaller particles. Granulating may be carried out by any one or a combination of methods selected from shredding, milling and chipping. In some embodiments, granulating comprises shredding. In other embodiments, granulating comprises shredding and milling. Typically, granulating comprises shredding followed by milling.

In some embodiments, the biochar particle size used in step (i) of the method is less than 2 mm, such as less than 1.5 mm, 1 mm, 0.75 mm, 0.5 mm, 0.25 mm, or 0.1 mm.

In some embodiments, in pre-step (iii), the biochar is granulated to a particle size of less than 2 mm, such as less than 1.5 mm, 1 mm, 0.75 mm, 0.5 mm, 0.25 mm, or 0.1 mm.

Suitably the treating in step (iv) may be pelletising. "Pelletising" refers to the process of densifying biochar particles into pellets using a suitable binder.

Suitably the treating in step (iv) may be agglomeration.

In one embodiment, the particles of biochar (i.e. the biochar used in step (i) of the method) have a size distribution of 0 mm to about 2 mm, preferably 0.1 mm to about 2 mm. It is

- to be understood that the size range given refers to the longest dimension of the biochar particles. At least some of the particles of the aggregate are of sizes that fall within the size range. For example, some of the particles of the aggregate may have sizes of about 1 mm, and the rest of the particles of the aggregate may have sizes greater than about 2 mm.
- 5 According to particular embodiments, substantially all (more than 90% by weight, often more than 95% by weight, for example more than 98% or 99% by weight) of the biochar particles in the aggregate are of a size from of about 0 mm to about 2 mm, or 0.1 mm to about 2 mm.
- 10 In some embodiments, the pre-mixture may further comprise an admixture. In particular embodiments, the admixture is added to the first or second compositions. In alternative embodiments, the admixture is added to the pre-mixture independently of (i.e., separately from) the first and second compositions.
- 15 As used herein "admixture" encompasses materials such as air entrainers, water reducers, set retarders, set accelerators or plasticisers. Admixture includes rosin resin, alkyl sulfonate, aliphatic alcohol sulfonate, protein salt and petroleum sulfonate, soluble inorganic salts of alkali and alkali earth metals (sodium or potassium hydroxide, calcium chloride, bromide and fluoride, sodium and calcium nitrite and nitrate, potassium carbonate, sodium and calcium thiocyanate, sulphate, thiosulphate, perchlorate, silicate, aluminate), carboxylic acids (formic, acetic, propionic and butyric, oxalic) and their salts (Calcium formate, calcium oxalate), lignosulfonates, sulfonated naphthalene formaldehyde (PNS), sulfonated melamine formaldehyde (PMS), vinyl copolymers (VCPs), and polycarboxylic ethers (PCEs).
- 20 and calcium thiocyanate, sulphate, thiosulphate, perchlorate, silicate, aluminate), carboxylic acids (formic, acetic, propionic and butyric, oxalic) and their salts (Calcium formate, calcium oxalate), lignosulfonates, sulfonated naphthalene formaldehyde (PNS), sulfonated melamine formaldehyde (PMS), vinyl copolymers (VCPs), and polycarboxylic ethers (PCEs).
- 25 In some embodiments, the width of the biochar aggregate pellet may be 0.5 to 50 mm, such as 1 to 40 mm, 1 to 30 mm, 1 to 20 mm, 5 to 40 mm, 5 to 30 mm, or 5 to 20 mm, preferably 5 to 20 mm. The length of the biochar aggregate pellet may be 0.5 to 50 mm, such as 1 to 40 mm, 1 to 30 mm, 1 to 20 mm, 5 to 40 mm, 5 to 30 mm, or 5 to 20 mm, preferably 5 to 20 mm.
- 30 An "aspect ratio" is a well-known ratio and is a proportional relationship between the aggregate's length and width. The aspect ratio between the length of the pellet and the width of the pellet may be 0.05 to 20, such as 0.1 to 10, 0.2 to 8, 0.25 to 6, 0.25 to 4, preferably 0.25 to 4. Suitably the biochar aggregate may be round and therefore it can be difficult to distinguish between length and width. Thus suitably the aspect ratio is about 1:1.
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In a preferred embodiment, the biochar aggregate has a size distribution of 0.1 to 50 mm, preferably 0.1 to 20 mm; and optionally the agglomeration step of the method comprises pelletisation. In particular such embodiments, the biochar aggregate is in the form of pellets.

- 5 In an alternative preferred embodiment, the biochar aggregate has a size distribution of 5 to 50 mm, and optionally the agglomeration step comprises pan pelletisation. In particular such embodiments, the biochar aggregate is in the form of pellets.

- 10 In an alternative preferred embodiment, the biochar aggregate has a size distribution of greater than 10 mm, such as from 10 mm to 200 mm; and optionally the agglomeration step comprises briquetting. In particular such embodiments, the biochar aggregate is in the form of briquettes.

- 15 The ratio between the biochar to binder (e.g., cementitious binder) is important to achieve the desired strength and density of the resulting aggregate. In some embodiments, the weight ratio of biochar to binder (e.g., cementitious binder) in the first composition and/or pre-mixture and/or the biochar aggregate is from 1:0.1 to 1:20, such as 1:0.5 to 1:10, 1:1 to 1:10 or 1:1 to 1:5, preferably 1:2.

- 20 In some embodiments, the weight percentage of biochar in the first composition and/or pre-mixture and/or the biochar aggregate is from 91% to 4%, preferably from 70% to 20%, more preferably 50% to 20%, such as about 35%.

- 25 In some embodiments, the biochar aggregate comprises from 5 to 95% by weight biochar, such as from 5 to 90%, 10 to 90%, 20 to 90%, 25 to 80%, 30 to 75%, 35 to 70%, preferably 25 to 85%, more preferably 30 to 80%.

- 30 In embodiments wherein the at least one binder comprises cement (e.g. Portland cement or High Strength Cement), the amount of binder may be lowered by using the method of the invention, wherein the pelletisation step is die mill pelletisation. Using lower amounts of cement provides an environmental benefit.

- 35 Therefore, in embodiments wherein the at least one binder comprises cement and the pelletisation step is die mill pelletisation, the weight percentage of biochar in the first composition and/or pre-mixture and/or the biochar aggregate may be from 20% to 90%, preferably from 25% to 80%, more preferably 30% to 80%, even more preferably 35% to 75%.

In embodiments wherein the at least one binder comprises cement and the pelletisation step is die mill pelletisation, the weight ratio of biochar to binder (i.e. cement) in the first composition and/or pre-mixture and/or the biochar aggregate is from 10:1 to 1:1, such as 8:1 to 1:1, 4:1 to 1:1, or 3:1 to 1:1, preferably 2.5:1 to 1:1, more preferably 2:1 to 1.2:1.

5

The amount of water is important for achieving hydration of the binder (e.g., cementitious binder or anhydrite) and to aid in the agglomeration (e.g., pelletisation) process. If the amount of water is too high, the resulting mixture will not process efficiently and if the amount of water is too low, the mixture can get stuck in the die and not be processed properly. In some embodiments, the weight ratio of water to the at least one binder in the first composition and/or the pre-mixture and/or the biochar aggregate is from 0.1 to 1, such as 0.1 to 0.6 or 0.3 to 0.5, preferably 0.2 to 0.5.

When the use of an inorganic base is necessary, the amount of inorganic base is important for activating the hydration of the cementitious binder and to achieve a higher final strength of the resulting aggregate (e.g., pellets). In some embodiments, the weight% of inorganic base in the pre-mixture and/or the biochar aggregate is from 0 to 15% w/w of the cementitious binder, 0.1 to 15% w/w of the cementitious binder, such as 0.5 to 12% w/w, 1 to 10% w/w, or 2 to 10 % w/w, preferably 2 to 10% w/w of the cementitious binder.

20

Suitably, the biochar aggregate (e.g., pellet) may further comprise at least one additive. Additives include admixture, strength enhancers, rheology modifier, and fibre. As such, in some embodiments, the method further comprises the step of adding an additive to the first composition, and/or second composition and/or the premixture. In some embodiments, the method comprises adding an additive during or between any one of steps (i), (ii) or (iii), preferably wherein the additive is an admixture.

25

The strength of the biochar aggregate and the resulting product (e.g., concrete) may be increased using strength enhancers. In some embodiments, the method further comprises the step of adding a strength enhancer (preferably graphene) during or between any one of steps (i), (ii) or (iii). In particular embodiments, the strength enhancer (preferably graphene) is added to the first composition during or after step (i), preferably after step (ii) and before step (iii). In particular embodiments, the strength enhancer is added to the second composition.

35

Fillers may improve the properties and the microstructure of the biochar aggregate. In some embodiments, the method further comprises the step of adding a filler during or between any one of steps (i), (ii) or (iii), preferably wherein the filler is limestone or clay. In particular embodiments, the filler is added to the first composition during or after step

(i), preferably after step (i) and before step (iii). In particular embodiments, the filler is added to the second composition.

5 The biochar aggregate may be exposed to an active carbonation process to increase the amount of carbon sequestered in the aggregate.

As such, in some embodiments, the at least one binder (e.g., cementitious binder) comprises at least one metal oxide or at least one metal silicate, and the method of the first aspect of the invention further comprises the step of:

10 (v) carbonating the at least one metal oxide or metal silicate by means of an active carbonation process to obtain a carbonated biochar aggregate.

For the avoidance of doubt, the carbonated biochar aggregate may comprise any of the features described above for the biochar aggregate. As used herein, the term "biochar
15 aggregate" includes the "carbonated biochar aggregate".

As used herein, "carbonated biochar aggregate" refers to a biochar aggregate containing increased amounts of metal carbonate species (e.g. CaCO_3 or MgCO_3) compared to the equivalent biochar aggregate which had not been exposed to a carbonation step. The metal
20 carbonate species may result, for example, from reactions of oxides (e.g. calcium oxide, magnesium oxide) or silicates (calcium silicate, magnesium silicate) with carbon dioxide. For the avoidance of doubt, the oxides and silicates that result in the metal carbonate species were present in the biochar aggregate, such as in the cementitious binder, prior to the carbonation step.

25 As used herein, "active carbonation" refers to a carbonation process wherein the rate of carbonation is increased compared to the "passive carbonation rate". Passive carbonation of the biochar aggregate refers to carbonation by exposing the biochar aggregate to ambient conditions (which atmosphere comprises around 420 ppm CO_2). As such, the passive
30 carbonation rate is the rate of carbonation of the biochar aggregate exposed only to ambient conditions, i.e., without increasing the concentration of CO_2 in the surrounding atmosphere, without heating, or watering, etc.

Suitably, the active carbonation process may comprise adding water to the binder (e.g. cementitious binder) or the biochar aggregate, such as using manual watering, automatic
35 spray systems or exposing the biochar aggregate to rain. Watering the aggregate increases the carbonation reaction rate.

Suitably, the active carbonation process may comprise heating the biochar aggregate, such as by using heating elements or a heat exchange system. Heating the biochar aggregate increases the carbonation reaction rate.

- 5 Intentionally increasing the airflow throughout the aggregate, particularly while exposing the aggregate to an atmosphere comprising CO₂, increases the rate of carbonation, and is thus an "active carbonation" process. In some embodiments, the active carbonation comprises increasing the airflow throughout the biochar aggregate, such as by placing the aggregate on a raised false floor. In some particular embodiments, increasing the airflow
10 throughout the biochar aggregate may be combined with a heat exchange system whereby waste heat from industrial processes in a gas or water form could be channelled through a pipe that is in contact with the airflow. This extra heat would increase the carbonation rate. In some embodiments, the active carbonation comprises increasing the airflow throughout the aggregate and exposing the biochar aggregate to an atmosphere comprising increased
15 CO₂ concentration (i.e., higher concentrations of CO₂ versus the typical ambient atmosphere, such as to more than 450 ppm).

- All equipment used in the watering, heating or increasing the airflow processes may be run using renewable electricity sources and energy plans as to not damage the environment
20 more than the aggregate would remove from the atmosphere.

- "Active carbonation" may also refer to exposing the biochar (such as the biochar aggregate) to a CO₂ enriched source. A CO₂ enriched source refers to a source of air comprising a higher concentration of CO₂ than that found in the atmosphere (i.e. more than
25 approximately 420 ppm), such as at least 450 ppm. Such a CO₂ enriched source may be captured flue gases from places such as power plants, cement kilns and chemical manufacturing. Suitably the CO₂ enriched source has a CO₂ concentration of or higher than 0.5% higher than the CO₂ concentration of the atmosphere, preferably of or higher than 1%, more preferably 2%, 5%, 10%, 20%, 30%, 40%, 50%, 60%, 70%, 80%, 90%, 95%,
30 99% or 100% higher than the CO₂ concentration of the atmosphere.

In some embodiments, the source of CO₂ has a concentration of at least 0.05%, 0.5%, 1%, 2%, 5%, 10%, 20%, 30%, 40%, 50%, 60%, 70%, 80%, 90%, 95%, 99% or 100% of CO₂.

- 35 "Active carbonation" may also refer to direct air capture (DAC) whereby the CO₂ is extracted directly from the atmosphere (Figure 2). Once pulled from the atmosphere, permanent storage would be required to stop CO₂ leaking back into the environment. Carbonation of the biochar aggregate may be used to store this CO₂. As such, in some embodiments, the active carbonation process comprises direct air capture (DAC).

In some embodiments, active carbonation comprises the use of a reaction chamber whereby the source of CO₂ is pumped into a vessel containing the biochar aggregate. The temperature, pressure and moisture inside the reaction chamber may be controlled. In
5 some embodiments, the biochar aggregate in the vessel has already hardened after production. Alternatively, the biochar aggregate could be introduced in the vessel as soon as the biochar aggregate has been formed, such as from compression pelletising or pan pelletising.

10 Once the biochar aggregate is produced, it could be conveyed using a sealed system comprising an enriched CO₂ atmosphere. The sealed system may be an enclosed belt conveyor or a vibrating spiral conveyor.

One or more of the aforementioned "active carbonation" processes may be used in the
15 methods of the present invention, sequentially or simultaneously.

The agglomerating and carbonating steps are simultaneous or sequential in any order. In some embodiments, the compressing, heating and carbonating steps are simultaneous or sequential in any order.

20 As such, the active carbonation step may be performed before the formation of the aggregate (i.e., before the agglomeration step, such as compressing or heating steps, i.e., during step (iii) and/or between step (iii) and (iv)), during the formation of the aggregate (i.e. during the agglomeration step, such as compressing or heating steps), as soon as the
25 biochar aggregate is formed (i.e. as a sequential step after agglomerating, such as after the compressing and heating), and/or once the biochar aggregate has hardened, such as after a couple of days.

In some embodiments, the agglomerating and carbonating steps are simultaneous. As such,
30 in some embodiments, the method comprises making the biochar aggregate in step (iv) under a CO₂ atmosphere (either at ambient temperature and pressure or at elevated temperatures and pressures) to result directly into a carbonated biochar aggregate. Figure 3 shows a diagram of a modified pellet mill that involves conducting the pelletising under a CO₂ atmosphere.

35 In embodiments wherein the agglomeration comprises compressing and heating, the compression and active carbonation (and optionally the heating) steps may be carried out simultaneously.

The carbonated biochar aggregate may then be fed into a pellet mill to pelletise the carbonated biochar aggregate. An advantage of this process is that heat is generated from the compression of the biochar aggregate which would accelerate the carbonation process.

- 5 Suitably, the biochar aggregate is tumbled around a pan pelletiser which has been modified to be sealed from the atmosphere, to form an aggregate (e.g. spherical aggregate) under a CO₂ atmosphere (Figure 4). The carbonation is accelerated in this process.

10 The carbonation process is directly related to surface area. As the biochar aggregate described herein is porous, it allows for a higher surface area and thus exposing more of the metal oxide or silicate of the binder (e.g., cementitious binder), and so aiding in the carbonation of the biochar aggregate. It has also been suggested that biochar can be used as an absorbent in direct air capture (DAC) methods. Without wishing to be bound by theory, it is believed that this property of the biochar can assist in increasing the localised
15 CO₂ concentration, resulting in an increase in the carbonation rate.

Additionally, without wishing to be bound by theory, in embodiments where sodium hydroxide is used as an inorganic base, the NaOH is expected to react with the carbonic acid to create sodium bicarbonate. According to Iversen et al (ACS Applied Materials &
20 Interfaces 2015 7 (9), 5258-5264), sodium bicarbonate is able to act as a catalyst for the carbonation of magnesium silicates.

As such, in some embodiments, the method may comprise adding a catalyst to the pre-mixture or the biochar aggregate that is able to increase the carbonation rate of the
25 aggregate (such as sodium bicarbonate).

In an alternative aspect of the invention there is provided a method of carbonating the biochar aggregate comprising:

- 30 (i) providing a biochar aggregate comprising a) biochar, b) at least one binder (e.g., cementitious binder) comprising at least one metal oxide or at least one metal silicate, and c) water; and
(ii) carbonating the at least one metal oxide or at least one metal silicate by means of an active carbonation process to obtain a carbonated biochar aggregate.

- 35 For the avoidance of doubt, the method of this alternative aspect of the invention may comprise any of the features described for any of the other aspects of the invention.

According to a second aspect of the invention, there is provided a biochar aggregate obtainable, such as obtained, by a method of the first aspect of the invention.

For the avoidance of doubt, the biochar aggregate of the second aspect of the invention may comprise any of the features described herein for the first and third to eight aspects of the invention.

5

According to a third aspect of the invention, there is provided a biochar aggregate comprising:

- (i) biochar; and
- (ii) at least one binder.

10

Suitably, the biochar aggregate of the third aspect of the invention may be in the form of a pellet or a briquette, preferably a pellet.

Suitably, the biochar aggregate (e.g., pellet) may further comprise at least one additive.

15 Additives include admixture, strength enhancers, rheology modifier, and fibre.

In some embodiments, the biochar aggregate (e.g., pellet) further comprise an admixture.

20 "Strength enhancers" are materials used to increase the strength of the composition incorporated them. Strength enhancers include microsilica, graphene and alkanolamines for example tri-isopropanolamine (TIPA) and Triethanolamine (TEA). Suitably, the biochar aggregate (e.g., pellet) may further comprise a strength enhancer.

25 In some embodiments, the strength enhancer is graphene. Graphene increases the strength of the biochar aggregate and the strength of the resulting composition in which the aggregate is incorporate (e.g., concrete composition comprising the biochar aggregate). Graphene can be functionalised with surface groups (such as graphene oxide and other forms of functionalised graphene), dispersed in a liquid using a surfactant or used without any dispersion aids, just the pure carbon form.

30

Modifying the rheological properties of concrete may improve the properties of concrete in the fresh and hardened state, which is particularly important for production and placement of special construction applications such as underwater or self-consolidating concrete. A "rheology modifier" is a material that alters the rheology (i.e. deformation or flowing response to applied forces or stresses) of a fluid composition to which it is added. Rheology modifiers include viscosity modifying agents such as cellulose ethers, natural gums (xanthan, wellan) and starch. Suitably, the biochar aggregate (e.g., pellet) may further comprise a rheology enhancer.

35

Fibre-reinforced concrete has greater tensile strength when compared to non-reinforced concrete. Fibers include cellulose fibres, natural fibres, carbon fibres, polyester fibres, glass fibres, polypropylene fibres, and steel fibres. Suitably, the biochar aggregate (e.g., pellet) may further comprise a fibre.

5

In some embodiments, the biochar aggregate (e.g., pellet) comprises from 0.01 to 15 wt% of the at least one additive (e.g., admixture), such as 0.1 to 10 wt%, 0.1 to 7 wt%, 0.1 to 5 wt%, 0.1 to 3 wt%, or 1 to 3 wt%.

10 Fillers may improve the properties and the microstructure of products incorporating the biochar aggregate such as concrete compositions. Suitably, the biochar aggregate (e.g., pellet) may further comprise at least one filler. The filler may be mica, gypsum, limestone, sand, wood, wood shavings, clay, concrete dust, dolomite, basalt, or char, preferably clay, limestone, dolomite, basalt or a mixture thereof, preferably limestone.

15

In some embodiments, the clay is calcined clay. The calcined clay may be natural or synthetically produced by high temperature kilns.

20 In some embodiments, the biochar aggregate (e.g., pellet) comprises from 0.01 to 30 wt% of the at least one filler, such as from 0.1 to 25 wt%, 0.5 to 20 wt%, 1 to 20 wt%, or 1 to 10 wt%.

In particular embodiments, the biochar aggregate comprises a large aggregate, preferably limestone. In particular such embodiments, the biochar aggregate comprises other aggregates, such as sand.

25

For the avoidance of doubt, the biochar aggregate (e.g., pellet) of the third aspect of the invention may comprise any of the features described for the first or second and for the fourth to eighth aspects of the invention.

30

According to a fourth aspect of the invention there is provided a composition comprising the biochar aggregate of the second or third aspects of the invention (including all embodiments thereof).

35 In some embodiments, the biochar aggregate is bound to at least one of the other components of the composition (i.e., the biochar aggregate is cohesive with at least one other component of the composition).

Suitably, the composition may comprise 0.5 to 75 wt%, preferably 0.5 to 65 wt%, more preferably 0.5 to 55 wt% biochar aggregate, even more preferably 0.5 wt% to 50 wt%, more preferably 0.5 wt% to 40 wt% biochar aggregate. Alternatively, the composition may comprise 1 to 50 wt%, more preferably 2 to 20 wt% biochar aggregate. The wt% of biochar aggregate refers to wt% relative to the weight of the whole composition.

In some embodiments, the composition may comprise more than 0.5 wt% biochar aggregate, preferably more than 1 wt%, 2 wt%, 3 wt%, 4 wt%, 5 wt%, 6 wt%, 7 wt%, 8 wt%, 9 wt%, or 10 wt% and/or comprise less than 75 wt%, 70 wt%, 60 wt%, 50 wt%, 45 wt%, 40 wt%, 35 wt%, 30 wt%, 25 wt%, 20 wt%, 15 wt%, 11 wt% biochar aggregate. For the avoidance of doubt, any of the aforementioned lower range end-points may be combined with any of the aforementioned upper range end-points.

In some embodiments, the composition further comprises bitumen. Such compositions may be considered an "asphalt composition". As known in the art, "bitumen" (also known as "asphalt") is a viscous form of petroleum. An asphalt composition refers to a composition comprising bitumen and a mixture of aggregates, binder and/or filler, used for constructing and maintaining roads, parking areas, railway tracks, ports, airport runways, bicycle lanes, pavement and also play- and sport areas. Typically, the bitumen functions as a binder in such compositions to bind the aggregates into a cohesive mixture.

In some embodiments, the asphalt composition is in the form of a road, parking area, airport runway, bicycle lane or pavement, preferably road, parking area or airport runway.

In particular embodiments, the asphalt composition comprises 1 to 20 wt% biochar aggregate, preferably 1 to 15 wt%, more preferably 2 to 10 wt%.

In more particular embodiments, the asphalt composition comprises 3 to 15 wt%, preferably 3 to 10 wt% biochar aggregate, such as 3 to 8 wt% biochar aggregate.

Suitably the asphalt composition comprises 1 to 30 wt% bitumen, preferably 1 to 20 wt%, more preferably 1 to 10 wt%, even more preferably 2 to 8 wt%, yet more preferably 3 to 7 wt%.

In particular embodiments, the asphalt composition represents net-zero asphalt. The skilled person will understand that net-zero refers to a balance of zero between the amount of greenhouse gas (GHG) that is produced and the amount that is removed from the atmosphere by the asphalt.

Net-zero asphalt may be obtained for asphalt compositions comprising up to 8 wt% biochar aggregate, such as between 2 wt% to 8%, for example 3 wt% to 8 wt% biochar aggregate. The skilled person will be able to determine the quantity of biochar aggregate that is needed to produce net-zero asphalt.

5

In particular embodiments, the asphalt composition represents carbon-negative asphalt. The skilled person will understand that carbon-negative refers to a negative balance between the amount of greenhouse gas (GHG) that is produced and the amount that is removed from the atmosphere by the asphalt (i.e. more carbon is removed).

10

Carbon-negative asphalt may be obtained for asphalt compositions comprising greater quantities of the biochar aggregate, for example greater than 8 wt% biochar aggregate, such as greater than 9 wt% or 10 wt% biochar aggregate. The skilled person will be able to determine the quantity of biochar aggregate that is needed to produce carbon-negative asphalt.

15

In some embodiments, the composition further comprises cementitious binder. Such compositions may be considered a "concrete composition". A concrete composition refers to a composition comprising cementitious binder and a mixture of aggregate(s) e.g., the biochar aggregate), binder and/or filler, used for constructing and maintaining roads, parking areas, railway tracks, ports, airport runways, bicycle lanes, pavement and also play- and sport areas.

20

In some embodiments, the concrete composition is in the form of foundations, block masonry, mortars, pre-cast, roofing tiles, 3D printed concrete, ready-mix and screeds.

25

In some embodiments, the concrete composition is in the form of a concrete building component, for example a precast concrete component, such as a column, beam, slab or block, preferably a concrete block.

30

In particular embodiments, the concrete composition comprises 1 to 50 wt% biochar aggregate, preferably 2 to 40 wt%, more preferably 2 to 30 wt%, even more preferably 3 to 20 wt%, such as 3 to 10 wt% biochar aggregate or 3 to 8 wt% biochar aggregate.

35

In particular embodiments, the concrete composition comprises 15 to 40 wt% biochar aggregate, preferably 20 to 35 wt%, such as 25 to 35 wt%.

In some embodiments, the composition further comprises a resin. As used herein, "resin-based" composition refers to a composition that comprises at least one resin. A resin is

understood in the art to be a solid or highly viscous substance of plant or synthetic origin that is typically convertible into polymers.

5 In some embodiments, the resin-based composition is in the form of flooring underlay, landscaping, anti-slip flooring or a product for artistic use.

10 In particular embodiments, the resin-based composition comprises 1 to 50 wt% biochar aggregate, preferably 1 to 40 wt%, more preferably 2 to 30 wt%, even more preferably 3 to 20 wt%, such as 3 to 10 wt% biochar aggregate or 3 to 8 wt% biochar aggregate.

15 In some embodiments, the composition further comprises a paint. Suitably, the paint is anti-slip paint or textured paint.

20 In particular embodiments, the composition comprising paint comprises 1 to 50 wt% biochar aggregate, preferably 1 to 40 wt%, more preferably 2 to 30 wt%, even more preferably 3 to 20 wt%, such as 3 to 10 wt% biochar aggregate or 3 to 8 wt% biochar aggregate.

25 In some instances, the above mentioned compositions may comprise 1 to 40 wt% of the bitumen, cementitious binder, resin, and/or paint, preferably 1 to 20wt%, such as 1 to 10wt%, or 2 to 8wt%, or 3 to 7wt%.

The skilled person will understand that, due to the presence of the biochar aggregate, the compositions of the fourth aspect of the invention have a lower carbon footprint compared to the same compositions but that do not comprise the biochar aggregate. As such, the biochar aggregate is used as a carbon footprint lowering additive.

30 For the avoidance of doubt, the fourth aspect of the invention may comprise any of the features described for any one of the other aspects of the invention.

35 In a fifth aspect of the invention, there is provided a method of manufacturing a composition of the fourth aspect of the invention, comprising the step of mixing the biochar aggregate of the second or third aspects of the invention (including all embodiments thereof) with at least one other component.

In some embodiments the at least one other component comprises a binder, resin or paint.

In some embodiments, the binder is a cementitious binder or bitumen.

The skilled person will understand that the method step recited in the fifth aspect of the invention may also be considered an additional step of the method of the first aspect of the invention.

- 5 In an alternative fifth aspect of the invention, there is provided the use of the biochar aggregate of the second and third aspects of the invention in manufacturing a composition of the fourth aspect of the invention.

For the avoidance of doubt, the fifth aspect of the invention may comprise any of the
10 features described for any one of the other aspects of the invention.

In a sixth aspect of the invention, there is provided the use of a biochar aggregate of the second or third aspects of the invention (including all embodiments thereof) to:

- (i) place the biochar aggregate in a location such that the biochar aggregate is exposed to
15 the natural atmosphere; and/or
(ii) fill a depression in the ground or to change the elevation of the ground (e.g., by creating a mound); or
(iii) insulate a building component.

As used herein, "natural atmosphere" refers to the outdoors atmosphere, i.e. exposed to
20 the weather.

In an alternative sixth aspect of the invention, there is provided a method comprising placing the biochar aggregate of the second or third aspects of the invention (including all embodiments thereof) in a location such that the biochar aggregate is exposed to the natural
25 atmosphere. The skilled person will understand that this method step may also be considered an additional step of the method of the first aspect of the invention.

In a further alternative sixth aspect of the invention, there is provided a method comprising filling a depression or hole in the ground, or creating a mound or otherwise changing the
30 elevation of the ground using the biochar aggregate of the second or third aspects of the invention (including all embodiments thereof). The skilled person will understand that this method step may also be considered an additional step of the method of the first aspect of the invention.

35 In a further alternative sixth aspect of the invention, there is provided a method comprising insulating a building component using the biochar aggregate of the second or third aspects of the invention (including all embodiments thereof). The skilled person will understand that this method step may also be considered an additional step of the method of the first aspect of the invention.

In some embodiments, the use or the method of the sixth aspect of the invention comprises placing the biochar aggregate in a location such that the biochar aggregate is exposed to the natural atmosphere.

5

In particular embodiments, the location is on or next to a road (in which case the biochar aggregate may be referred to as a "roadside aggregate").

In particular embodiments, the location is on or next to a railway (in which case the biochar aggregate may be referred to as a "railway aggregate").

In some embodiments, the use or the method of the sixth aspect of the invention comprises filling a depression in the ground or changing the elevation of the ground. The skilled person will understand that such a use may be referred to as use of the biochar aggregate as an "engineered fill" or "geological fill".

"Engineered fill" refers to a fill which is selected, placed and compacted to an appropriate specification in order that it will exhibit the required engineering behaviour.

"Geological fill" refers to a fill that essentially acts like soil. It may be used as a soil infill, as an alternative to traditional fill materials such as soil or concrete.

In particular embodiments, changing the elevation of the ground comprises creating a mound.

25

In particular embodiments, changing the elevation of the ground comprises changing the elevation of an embankment, landscaping or a road base.

In particular embodiments, the use or the method of the sixth aspect of the invention comprises insulating a building component. The skilled person will understand that such a use may be referred to as use of the biochar aggregate as "insulation fill".

In some embodiments, the building component comprises one or more of a foundation, a floor, a wall, a beam, a column, a roof, or stairs, preferably a wall, more preferably a cavity wall.

35

The skilled person will understand that in the sixth aspect of the invention, the biochar aggregate may be used as "loose", i.e., without performing any further steps that would change the composition of the biochar aggregate or bind the aggregates together. Typically,

a plurality of the biochar aggregates would be used in such uses and methods in order to fill or cover the required space.

As described herein, in the sixth aspect of the invention, the biochar aggregate is used, *inter alia*, in order to lower the carbon-footprint of the system comprising the location (e.g. road, the railway), the ground, or the building component described above, as appropriate.

For the avoidance of doubt, the use or the method of the sixth aspect of the invention may comprise any of the features described for any one of the other aspects of the invention.

10

In a seventh aspect of the invention, there is provided an engineered fill, geological fill, insulation fill, roadside aggregate or railway aggregate comprising a biochar aggregate of the second or third aspects of the invention (including all embodiments thereof).

15 For the avoidance of doubt, the engineered fill, the geological fill, the insulation fill, the roadside aggregate or the railway aggregate of the seventh aspect of the invention may comprise any of the features described for any one of the other aspects of the invention.

20 In addition to lowering the carbon footprint of the product or system comprising the biochar (e.g., road, railway or building component comprising the insulation fill), a further advantage of using the biochar aggregate of the invention in such products is the ability of the biochar to help control local pollution due to industrial activities due to the biochar aggregate's surface chemistry and porosity.

25 The advantages of the biochar aggregate in such applications over pure biochar are the added strength, weight (which helps stop the biochar being washed away) and durability. Due to the large and reactive surface area of the biochar, it is expected that oils, volatile organic compounds (VOCs), semi-volatile organic compounds (SVOCs), polycyclic aromatic hydrocarbons (PAHs), and/or persistent organic pollutant (POPs) from cars and railways
30 could be absorbed by the biochar aggregate, thus essentially filtering out pollutants. Consequently, this would reduce the concentration of pollutants in local systems and would reduce damage to wildlife and ecosystems.

35 Thus in an eighth aspect of the invention, there is provided the use of a biochar aggregate of the second or third aspects of the invention (including all embodiments thereof) in reducing pollution.

In some embodiments, the use of the biochar aggregate is as a pollution-reducing additive.

In some embodiments, the pollution is roadside or railway pollution. As explained above, the biochar aggregate of the present invention achieves this effect by adsorbing oils, volatile organic compounds (VOCs), semi-volatile organic compounds (SVOCs), polycyclic aromatic hydrocarbons (PAHs), and/or persistent organic pollutant (POPs) from roads and railways.

5

In some embodiments, the pollution is waterways pollution. The biochar aggregate of the present invention may be used to remove pollutants or substances that may damage the environment e.g., pharmaceuticals from water systems.

10 Without wishing to be bound by theory, the biochar aggregate of the present invention has at least the following advantages over pure biochar.

Using the biochar aggregate of the present invention improves the strength of the aggregate, allowing the biochar to better withstand the industrial processes and reducing
15 the biochar's surface area, therefore requiring less bitumen to cover the surface and lower processing temperatures. This may also allow more aggregate to be included in the mixture when kept within normal asphalt parameters, allowing for further carbon sequestration.

The improved durability of the biochar aggregate of the present invention compared to pure
20 biochar also allows for decorative concrete uses such as architectural masonry to incorporate the biochar (as the aggregate), whereas pure biochar is likely to crumble and break up due to weathering and/or abrasion.

Importantly, the biochar aggregate of the present invention also improves the health and
25 safety of handling due to lower amounts of dust and fine particles produced. This allows for a wider application use as this would require less specialised equipment for handling which is not always available at construction sites. Additionally, aggregates used in asphalt are typically dry before use, however drying pure biochar results in a highly combustible material which represents a safety risk, especially when used in large volumes. When
30 present in the biochar aggregate form, the biochar is bonded to incombustible material, reducing the fire risk.

A further advantage of the biochar aggregate over pure biochar is that the increased weight
35 and durability of the aggregate may allow for the recovery of the aggregate to be reused or regenerated.

Many of the binders used to manufacture the biochar aggregate of the invention are able to undergo carbonation. Biochar is highly porous able to absorb and hold onto moisture as well as having improved physical and chemical absorption of CO₂ to the biochar surface.
40 Using biochar in the aggregate is thus expected to result in a higher rate of carbonation of

the cementitious binders used in the biochar aggregate, resulting in more CO₂ removal. The addition of the inorganic base also increases the carbonation rate of the biochar aggregate.

- 5 The biochar has nanopores that are known to absorb CO₂, this property of the biochar should allow more CO₂ to dissolve into the moisture in the aggregate. The more CO₂ dissolved in the water in the aggregate improves the rate of reaction with minerals that have carbonation potential.

10 **Clauses**

Certain embodiments of the invention are described in the following clauses:

1. A method of manufacturing a biochar aggregate comprising the steps of:
- 15 (i) mixing biochar and at least one binder to form a first composition;
- (ii) optionally providing a second composition comprising water;
- (iii) optionally mixing together the first and second compositions to form a pre-mixture; and
- (iv) agglomerating the first composition or, if present, the pre-mixture to form the biochar aggregate.
- 20 2. A method of clause 1, wherein the agglomerating comprises pressure or non-pressure agglomeration of the first composition or the pre-mixture to form the biochar aggregate.
- 25 3. A method of any one of the preceding clauses, wherein the agglomerating comprises compressing and heating the first composition or the pre-mixture.
4. A method of any one of the preceding clauses, wherein the agglomerating comprises briquetting or pelletisation of the first composition or the pre-mixture, preferably die mill
- 30 pelletisation, pan pelletisation or extrusion pelletisation of the pre-mixture.
5. A method of any one of the preceding clauses, wherein the agglomerating comprises pelletisation and the biochar aggregate is in the form of a pellet.
- 35 6. A method of any one of the preceding clauses, wherein the second composition further comprises at least one inorganic base.

7. A method of clause 6, wherein the at least one inorganic base comprises an alkali hydroxide, alkali oxide or alkali carbonate or a mixture thereof, preferably sodium hydroxide, sodium carbonate, calcium oxide or a mixture thereof.
- 5 8. A method of any one of the preceding clauses, wherein the particle size of the biochar in step (i) is less than 2 mm, preferably less than 1.5 mm, more preferably less than 1 mm.
9. A method of any one of the preceding clauses, wherein the at least one binder comprises a cementitious binder.
- 10 10. A method of clause 9, wherein the cementitious binder is selected from GGBS, cement, fly ash, Portland cement, micro-silica, or a mixture thereof, preferably GGBS.
11. A method of any one of clauses 1 to 8, wherein the at least one binder comprises a
15 non-cementitious binder.
12. A method of clause 11, wherein the non-cementitious binder is de-hydrated gypsum (Anhydrite).
- 20 13. A method of any one of clauses 5 to 12, wherein the width of the pellet is 0.1 to 50 mm, preferably 0.1 to 20 mm.
14. A method of any one of clauses 5 to 13, wherein the aspect ratio between the length of the pellet and the width of the pellet is 0.05 to 20, preferably 0.25 to 4.
- 25 15. A method according to any one of the preceding clauses, wherein the weight ratio of the biochar to the at least one binder (e.g., cementitious binder) in the first composition and/or the pre-mixture and/or the biochar aggregate is from 1:0.1 to 1:20.
- 30 16. A method according to any one of the preceding clauses, wherein the weight ratio of water to the at least one binder (e.g., cementitious binder) in the first composition and/or the pre-mixture and/or the biochar aggregate is from 0.1 to 0.6, preferably 0.2 to 0.5.
17. A method according to any one of clauses 6 to 16, wherein the weight% of the at
35 least one inorganic base in the second composition and/or the pre-mixture and/or the biochar aggregate is from 0.1 to 15% w/w of the binder (e.g., cementitious binder), preferably 2 to 10% w/w of the binder (e.g., cementitious binder).

18. A method according to any preceding clause, further comprising the step of adding an additive during or between any one of steps (i), (ii) or (iii), preferably wherein the additive is an admixture.
- 5 19. A method according to any preceding clause, further comprising the step of adding a filler during or between any one of steps (i), (ii) or (iii), preferably wherein the filler is limestone or concrete dust.
20. A method according to clause 18 or 19, wherein the additive and/or the filler is added
10 to the first composition during or after step (i), preferably after step (i) and before step (iii).
21. A method according to any one of the preceding clauses, wherein the at least one binder comprises a metal oxide or metal silicate, and the method further comprises carbonating the at least a metal oxide or metal silicate by means of an active carbonation
15 process to obtain a carbonated biochar aggregate.
22. A method of clause 21, wherein the active carbonation process comprises adding water to the at least one binder or the biochar aggregate (such as by manual watering, automatic spray systems or exposing the biochar aggregate to rain).
- 20 23. A method of clause 21 or 22, wherein the active carbonation process comprises providing an increased airflow through the biochar aggregate.
24. A method of any one of clauses 21 to 23, wherein the active carbonation process
25 comprises heating the biochar aggregate.
25. A method of any one of clauses 21 to 24, wherein the active carbonation process comprises exposing the biochar aggregate to a CO₂ enriched source of gas.
- 30 26. A method of clause 25, wherein the CO₂ enriched source is obtained by direct air capture or is an industrial flue gas.
27. A method of carbonating a biochar aggregate pellet comprising:
i. providing a biochar aggregate pellet comprising a) a biochar particle, b) at least one binder
35 (e.g. cementitious binder) comprising at least one metal oxide or at least one metal silicate, and c) water,
ii. carbonating the at least one metal oxide or at least one metal silicate by means of an active carbonation process to obtain a carbonated biochar aggregate.

28. A method of clause 27, having any of the features of clauses 1 to 26.
29. A biochar aggregate obtainable by a method of any one of clauses 1 to 28.
- 5 30. A biochar aggregate comprising:
(i) biochar; and
(ii) at least one binder.
31. A biochar aggregate according to clause 30, wherein the biochar aggregate is in the
10 form of a pellet or a briquette, preferably a pellet.
32. A biochar aggregate according to clauses 29 to 31, wherein the weight ratio of the biochar to the at least one binder is from 1:0.1 to 1:20.
- 15 33. A biochar aggregate according to any one of clauses 29 to 32, wherein the particle size of the biochar in step (i) is less than 2 mm, preferably less than 1.5 mm, more preferably less than 1 mm.
34. A biochar aggregate according to any one of clauses 29 to 33, wherein the at least
20 one binder comprises a cementitious binder.
35. A biochar aggregate according to clause 34, wherein the cementitious binder is selected from GGBS, cement, fly ash, Portland cement, or micro-silica, or a mixture thereof, preferably GGBS or cement, preferably GGBS.
25
36. A biochar aggregate according to any one of clauses 29 to 33, wherein the at least one binder comprises a non-cementitious binder.
37. A biochar aggregate according to clause 36, wherein the non-cementitious binder is
30 de-hydrated gypsum (Anhydrite) or lignosulphonate.
38. A biochar aggregate according to any one of clauses 29 to 37, further comprising at least one inorganic base.
- 35 39. A biochar aggregate according to clause 38, wherein the at least one inorganic base comprises an alkali hydroxide, alkali oxide or alkali carbonate or a mixture thereof, preferably sodium hydroxide, sodium carbonate or a mixture thereof.

40. A biochar aggregate according to clause 38 or 39, wherein the weight% of inorganic base is from 0% to 15% w/w of the binder (e.g., cementitious binder), 0.1 to 15% w/w of the binder (e.g., cementitious binder), preferably 2 to 10% w/w of the binder (e.g., cementitious binder).
- 5 41. A biochar aggregate according to any one of clauses 29 to 40, further comprising water, preferably wherein the weight ratio of water to binder (e.g. cementitious binder) is from 0.1 to 0.6, preferably 0.2 to 0.5.
- 10 42. A biochar aggregate according to any one of clauses 29 to 41, wherein the biochar aggregate further comprises at least one additive, preferably the at least one additive is an admixture, a strength enhancer, a rheology modifier, or a fiber.
43. A biochar aggregate according to clause 42, wherein the biochar aggregate
15 comprises from 0.01 to 15 wt% of the at least one additive.
44. A biochar aggregate according to any one of clauses 29 to 43, wherein the biochar aggregate further comprises at least one filler, preferably the at least one filler is limestone, sand, wood, clay, concrete dust, or char, preferably limestone and/or concrete dust.
- 20 45. A biochar aggregate according to clause 44 wherein the biochar aggregate comprises from 0.01 to 30 wt% of the at least one filler.
46. A biochar aggregate according to any one of clauses 29 to 45 wherein the biochar
25 aggregate comprises calcined clay, limestone, high strength cement.
47. A biochar aggregate according to any one of clauses 29 to 46 having a carbon footprint of -2 kg CO₂e/kg to 0.2 kg CO₂e per kg of biochar aggregate.
- 30 48. A composition comprising a biochar aggregate of any one of clauses 29 to 47.
49. A composition of clause 48, further comprising bitumen, cementitious binder, resin, and/or paint, preferably bitumen or cementitious binder, more preferably bitumen.
- 35 50. A composition of clause 48 or 49, wherein the composition comprises 1 to 50 wt% of the biochar aggregate, preferably 1 to 20 wt% of the biochar aggregate, more preferably 1 to 15 wt%, even more preferably 2 to 10 wt%, yet more preferably 3 to 10 wt%, such as 3 to 8 wt%.

51. A composition of any one of claims 48 to 50 wherein the composition comprises 1 to 40 wt% of the bitumen, cementitious binder, resin, and/or paint, preferably 1 to 20wt%, more preferably 1 to 10 wt, even more preferably 2 to 8 wt%, yet more preferably 3 to 7 wt%.
52. A method of manufacturing a composition of claims 48 to 51, comprising the step of mixing the biochar aggregate of any one of claims 29 to 47 with at least one other component (such as a binder, resin, or paint).
53. Use of a biochar aggregate of any one of claims 29 to 47 to:
- (i) place the biochar aggregate in a location such that the biochar aggregate is exposed to the natural atmosphere; and/or
 - (ii) fill a depression in the ground or to change the elevation of the ground; or
 - (iii) insulate a building component.
54. An engineered fill, geological fill or insulation material, comprising a biochar aggregate of any one of claims 29 to 47.
55. The method, aggregate, composition or use of any preceding clause wherein the biochar aggregate has a moisture level of between 3 and 30 wt%, more preferably between 5 and 25 wt%, such as between 8 and 23 wt%, for example between 10 and 20 wt%.

Examples

Example 1

Biochar was granulated to a maximum particle of 2 mm and then dried. The dried biochar was mixed with GGBS and a solution of a mixture of sodium hydroxide (SH) and water, and was formed into an aggregate using press pelletisation using an 8 mm die.

Table 1: An example of the mix proportions of a GGBS-based biochar aggregate.

	Water:GGBS ratio	SH % of GGBS	Biochar:binder	Biochar (kg)	GGBS (binder) (kg)	Water (kg)	SH (kg)
Example 1	0.40	5.0%	1: 1.2	1.0	1.2	0.48	0.06

The resulting biochar aggregate had a particle density of 1908 kg/m³, a 24H water absorption value of 44% (both measurements were taken using a pyknometer), and a 10% fines value of 7.4 kN.

- 5 The biochar is expected to have sequestered 2.9 kg CO₂e/kg (based on known calculations used in the field), which would result in an estimated carbon footprint of -1 kg CO₂e/kg.

Example 2

- 10 The same method was used as in Example 1 but in this example, high strength cement (HSC) was used as a binder instead of GGBS.

An added advantage of this example formulation is that, as biochar is supplied with 20-50% moisture content, it is possible to use this inherent moisture to react with the cement,
15 therefore the drying of the biochar may not be required.

The biochar particles have a maximum particle size of 2 mm before being mixed.

Table 2: The mix proportions for a cement-based biochar aggregate

	Water:HSC	Biochar:binder		Biochar (kg)	HSC (kg)	Water (kg)
Example 2	0.40	1:	0.5	2.0	1.0	0.40

20 The resulting biochar aggregate had a particle density of 1900 kg/m³ and a 24H water absorption value of 61%, both measurements were taken using a pyknometer.

The biochar is expected to have sequestered 2.9 kg CO₂e/kg (based on known calculations
25 used in the field), this would result in an estimated carbon footprint of -0.9 kg CO₂e/kg.

Example 3

A die pelletiser was used to create two sample aggregates with equal carbon footprints
30 based on the amount of biochar and binder in the aggregate. Two different binders were used, ground granulated blast slag (GGBS) and CEM I, a high strength cement.

Table 3: The mix proportions of sample 1 and 2 that have the same carbon footprint

	Ground granulated slag (GGBS) wt%	High strength cement (HSC) wt%	Biochar wt%	Chemical solution (12% sodium hydroxide) wt%	Water wt%	Total %
Sample 1	47.0	0.0	31.0	19.0	3.0	100.0
Sample 2	0.0	35.5	43.8	0.0	20.6	100.0

The samples were prepared by mixing of the dry components followed by the addition of the chemical solution and water. The resulting mixture can be described as being able to "snowball", whereby the mixture is able to stick together when squeezed by hand.

The die pelletiser was preheated using a "cleaning mixture" which consists of no hardening materials, such as a mixture of sawdust, sand and water. The die consisted of 8 mm diameter holes. The samples were bagged immediately after production and left covered to cure.

Table 4: The kg CO₂e contribution by weight of the samples

	Sample 1	Sample 2
GGBS / kg CO ₂ e	0.2	0
HSC / kg CO ₂ e	0	1.8
Biochar / kg CO ₂ e	-4.9	-6.5
Total / kg CO ₂ e	-4.7	-4.7

Table 5: The carbon footprints of each of the materials used in sample 1 & 2.

Material	kg CO ₂ /kg
Biochar	-2.86
GGBS	0.0796
HSC	0.906

The carbon footprint calculations are based on multiplying the weight used in the testing mix by the carbon footprint highlighted in Table 5. However, it should be noted that for the

biochar carbon footprint, the water content has to be taken into account, which has been deemed 7%.

Results

- 5 Both samples were crushed using a standard crushing technique to determine their aggregate crushing values, i.e. how strong the aggregates are. The results are shown in Table 6.

Table 6: The 1 day 10 percentage fines testing of sample 1 and 2

Sample	10% fines test / kN
1 (GGBS)	8.10
2 (Cement)	4.24

10

Table 6 shows that sample 1 (which was produced using GGBS) was stronger than sample 2 (produced using cement). This shows that using GGBS allows for an aggregate to be produced of the same carbon footprint but having higher strength, allowing it to be used in more applications.

15

To achieve the same strength as the GGBS, sample 2 would require more cement to be added or the use of admixtures, both of which would increase cost and cause the carbon footprint to increase.

20

Therefore, using biochar in a GGBS-based rather than cement-based aggregate provides a greater environmental benefit because the sequestered carbon has more impact when the other components of an aggregate are as low carbon as possible.

Example 4

25

A pan pelletiser was used to create two sample aggregates with equal carbon footprints based on the known carbon footprints of the raw ingredients, but using two different types of binders, ground granulated blast slag and high strength cement. The carbon footprints of these mixes are the same as the one used in Example 3. However, the mix proportions are different due to the increased water required for pan pelletising.

30

Table 7: The mix proportions of samples 3 & 4

	Ground granulated slag (GGBS) wt%	High strength cement (HSC) wt%	Biochar wt%	Chemical solution (12% sodium hydroxide) wt%	Water wt%	Total %
Sample 3	45.4	0.0	29.9	18.4	6.3	100.0
Sample 4	0.0	32.7	40.5	0.0	26.7	100.0

After 4 days of curing, the aggregates were tested to find their aggregate crushing value, which is a measure of how much the aggregate breaks up when a constant force rate is applied. The results are shown in Table 8.

5

Table 8: The 4 day aggregate crushing value for samples 3 and 4

Sample	Aggregate crushing value / %
3 (GGBS)	52.2
4 (Cement)	55.7

The aggregate crushing results shown in Table 8 show that sample 3 (GGBS) is a stronger aggregate than the analogous sample 4 (cement).

10

Example 5 – Asphalt comprising biochar aggregate

The biochar-based aggregate can be used in asphalt applications. In this use, the aggregate can be mixed with other standard aggregates such as limestone and basalt.

15

Two tests were performed on a mixture of about 95% limestone and 5% of a biochar aggregate. The biochar aggregate was produced in the same way as sample 1 of Example 3, however, produced using a 6 mm diameter hole die instead of an 8 mm diameter hole. The tests were Los Angeles abrasion (LA) test and magnesium sulfate soundness test, which are common in the field. The LA test tumbled the mixture of aggregate and determined how much the mixture of aggregate fragments, and the soundness test is to mimic freeze-thaw cycles.

20

Results:

25 The minimum requirement for use in an asphalt application is an LA result of less than 30%.

The aggregate mixture achieved a value of 26%, showing that the aggregate mixture passes the requirements to be used in asphalt.

- 5 The aggregate mixture achieved a magnesium sulfate soundness value of 10.2%. A typically accepted value is 25%. This shows that the mixture of aggregate is suitable to be used in an asphalt application.

Example 6: Testing of asphalt comprising a biochar aggregate

- 10 The aggregate can be used in asphalt applications to reduce the embodied carbon footprint of the material. The asphalt prepared for the testing had a mix design that is outlined in Table 9. The biochar aggregate had the same mix design proportions as outlined in Table 3, sample 1.
- 15 The mix design used is a typical industry standard AC 20 asphalt mix design, with a proportion of the natural aggregate replaced with the biochar-based aggregate in accordance to the size.

Table 9: The mix proportions of the asphalt used for the testing

Material	wt%
Sample 1 (biochar aggregate) 0-4 mm	1.60
Sample 1 (biochar aggregate) 4-6 mm	2.00
Sample 1 (biochar aggregate) 6-10 mm	2.60
Limestone 0-4 mm	34.30
Limestone 4-6 mm	9.20
Limestone 6-10 mm	9.30
Limestone 14-20 mm	33.20
Limestone filler	3.10
Bitumen	4.7
Total	100

20

All samples were mixed in accordance to BS EN 12697-35:2016 (industry standard), with the asphalt samples prepared using an impact compactor in accordance with BS EN 12697-

30:2018, apart from the tests regarding the wheel tracking which required the samples to be prepared using a roller compactor in accordance to the BS EN 12697-33:2019 standard.

The binder & grading were performed to the procedure outlined in BS EN 12697-1-2020&2-2015 A1-2019 (Pressure Filter Method). The results of the particle grading of the asphalt mix are shown below in Table 10.

Table 10: Binder and aggregate grading

Material	Percentage by mass passing
31.5 mm	100
20.0 mm	94
14.0 mm	81
10.0 mm	68
6.3 mm	57
4.0 mm	44
2.0 mm	36
1.0 mm	30
0.50 mm	24
0.25 mm	18
0.125 mm	12
0.063 mm	8.5
Binder (bitumen) content	4.6%

10 Table 10 shows that the grading of the measured asphalt is in line with the properties expected of the asphalt in Table 9. The grading shown in Table 10 is similar to typical asphalt specification.

The asphalt was tested for air voids using the procedure outlined in BS EN 12697-6 2020 & 15 -8 2018 - Procedure D. The results are presented below in Table 11.

Table 11: The results of *in situ* air void content of the asphalt mixture

Mass of specimen (g)	Height (mm)	Diameter (mm)	In situ bulk density (Mg/m ³)	Maximum density (Mg/m ³)	In situ Void content (%)
1122.9	59.9	101.5	2.317	2.417	4.1

As used herein, "Mg" refers to megagram.

The typical void content of asphalt is 1-7%. Therefore, as shown in Table 11, the void content of the asphalt containing the biochar aggregate of the invention is within the standard range accepted in the industry.

The maximum density of the asphalt was performed using the procedure outlined in BS EN 12697-5 2018. The results are presented below in Table 12.

10

Table 12: The maximum density of the asphalt

Test temperature of de-aired water / °C	Maximum density of bituminous mixture: P _{MV} / Mg/m ³
24.2	2.417

The density of the asphalt is within the standard range accepted in the industry.

15 The stiffness of the asphalt was performed using the procedure outlined in BS EN 12697-26 2004 Annex C. The results are presented in Table 13 below.

Table 13: The ITSM (stiffness) testing results.

Load rise time (ms)	124.5
Horizontal stress (kPa)	519.9
Vertical force (kN)	4.97
Peak deformation (microns)	5.0
Stiffness modulus (MPa)	10194

20 The minimum stiffness modulus for typical asphalt use is 1800 MPa. The results in Table 13 show that the biochar aggregate asphalt is within typical specification.

The wheel tracking of the asphalt was performed using the procedure outlined in BS EN 12697-22 2020 (Procedure B Small Device). The results are presented in Table 14 below.

5 Table 14: Wheel tracking depth results

Height (mm)	98.9
Diameter (mm)	202.8
Bulk density (Mg/m ³)	2.332
Maximum density (Mg/m ³)	2.417
In situ air voids	3.5
Test temperature °C	60
Wheel tracking slope (WTS _{air} , mm/1000 cycles)	0.1
Rut depth @ 10,000 cycles (mm)	3.4
Proportional rut depth @ 10,000 cycles (%)	3.4

Asphalt tested at 60°C typically has a maximum rut depth limit of 7 mm @ 10,000 cycles and a wheel tracking slope of less than 1 mm/1,000 cycles. The asphalt tested containing the biochar aggregate was within these standard parameters.

10

The water sensitivity of the asphalt was performed using the procedure outlined in BS EN 12697-12:2018 inc. BS EN 12697-23:2017. The results are presented below in Table 15.

Table 15: The sample properties and testing results of the water sensitivity.

Parameter	Wet	Dry
Mean thickness (mm)	59.6	59.8
Mean diameter (mm)	101.5	101.6
Mean bulk density (Mg/m ³)	2.355	2.345
Volume increase after vacuum	n/a	-

Test temperature °C	15.1	15.7
ITS (KPa)	2780	2910
ITSR ratio (%)	95.5	95.5
Type of failure	C	C

Asphalt typically has a minimum value of 80% ITSR ratio to be used. The addition of the aggregate keeps the asphalt within standard specification.

5 Example 7: Carbonation of a biochar aggregate

A sealed pressure chamber was used to carbonate an aggregate with the formulation of biochar aggregate, sample 1.

- 10 The procedure for this testing was to dry the aggregates at 100°C for a minimum of 24 hours to remove all moisture. The sample was then weighed to know the original dry mass, after which water was added, to obtain a wet aggregate at different percentages.

- 15 Sample 1 was placed at the bottom of a steel pressure chamber, whereby a vacuum is pulled on the chamber to evacuate all remaining air. After all air has been removed from the chamber, carbon dioxide was then let into the chamber at room temperature and pressure and left overnight for 16 hours to undergo carbonation.

- 20 Once the set amount of time has passed, the chamber was evacuated and opened to air, after which the aggregate was removed ensuring that all of the sample has been removed and collected. It was then placed in an oven at 100°C for 24 hours to fully dry.

- 25 The maximum theoretical weight gain for the mix design is 20% which is related to the CaO and MgO content of the GGBS used in the aggregate.

Sample 1 was then weighed to determine the mass increase of the sample from the reaction with carbon dioxide.

Table 16: The overnight weight increase from carbon dioxide exposure.

Sample number	Water addition %	Initial dry weigh /g	Final dry weigh / g	Difference / g	Percentage increase %	Percentage of maximum
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						carbonation %
Sample 1	20	648.0	686.0	38	5.83	29.2
Sample 1	40	150.0	158.6	8.6	5.73	29.0

This demonstrates that the biochar aggregate is able to undergo carbonation (and quickly), thus removing carbon dioxide from the environment.

5 Example 8: Bitumen affinity test

A bitumen affinity test is conducted on aggregates that are intended to be used in asphalt and is a typical test done in the industry to determine how “sticky” the aggregates are to the bitumen binder in asphalt.

10

A biochar aggregate using the formulation of sample 1 was mixed with bitumen and then subsequently placed in a glass jar with water and rolled at 60 RPM for 6 hours and 24 hours, after which the sample was inspected to determine the coverage of bitumen on the aggregate by an expert.

15

Table 17: The bitumen affinity results for an aggregate that has the formulation of sample 1

Time / H	Bitumen affinity %
6	75
24	60

A 60% bitumen affinity result would be considered acceptable by the industry. The aggregate is within acceptable parameters.

20

Claims

1. A method of manufacturing a biochar aggregate comprising the steps of:
(i) mixing biochar and at least one binder to form a first composition, wherein the at least
5 one binder comprises GGBS;
(ii) optionally providing a second composition comprising water;
(iii) optionally mixing together the first and second compositions to form a pre-mixture; and
(iv) agglomerating the first composition or, if present, the pre-mixture to form the biochar
aggregate.
10
2. A method of claim 1, wherein the agglomerating comprises pressure or non-pressure
agglomeration of the first composition or the pre-mixture to form the biochar aggregate.
3. A method of any one of the preceding claims, wherein the agglomerating comprises
15 compressing and heating the first composition or the pre-mixture.
4. A method of any one of the preceding claims, wherein the agglomerating comprises
briquetting or pelletisation of the first composition or the pre-mixture, preferably die mill
pelletisation, pan pelletisation or extrusion pelletisation of the pre-mixture.
20
5. A method of any one of the preceding claims, wherein the agglomerating comprises
pelletisation and the biochar aggregate is in the form of a pellet.
6. A method of any one of the preceding claims, wherein the second composition further
25 comprises at least one inorganic base, preferably wherein the at least one inorganic base
comprises an alkali hydroxide, alkali oxide or alkali carbonate or a mixture thereof,
preferably sodium hydroxide, sodium carbonate, calcium oxide or a mixture thereof.
7. A method of any one of the preceding claims, wherein the particle size of the biochar
30 in step (i) is less than 2 mm, preferably less than 1.5 mm, more preferably less than 1 mm.
8. A method of any one of Claims 5 to 7, wherein the width of the pellet is 0.1 to 50
mm, preferably 0.1 to 20 mm.
- 35 9. A method according to any one of the preceding claims, wherein the weight ratio of
the biochar to the at least one binder in the first composition and/or the pre-mixture and/or
the biochar aggregate is from 1:0.1 to 1:20.

10. A method according to any one of claims 6 to 9, wherein the weight% of the at least one inorganic base in the second composition and/or the pre-mixture and/or the biochar aggregate is from 0.1 to 15% w/w of the binder, preferably 2 to 10% w/w of the binder.
- 5 11. A method according to any preceding claim, further comprising the step of adding an additive and/or a filler during or between any one of steps (i), (ii) or (iii), preferably wherein the additive is an admixture and the filler is limestone or concrete dust.
- 10 12. A method according to any one of the preceding claims, wherein the at least one binder comprises a metal oxide or metal silicate, and the method further comprises carbonating the at least a metal oxide or metal silicate by means of an active carbonation process to obtain a carbonated biochar aggregate, preferably wherein the active carbonation process comprises exposing the biochar aggregate to a CO₂ enriched source of gas, more preferably the CO₂ enriched source is obtained by direct air capture or is an industrial flue gas.
- 15 13. A biochar aggregate obtainable by a method of any one of claims 1 to 12.
14. A biochar aggregate comprising:
(i) biochar; and
20 (ii) at least one binder, wherein the at least one binder comprises GGBS.
15. A biochar aggregate according to claim 14, wherein the biochar aggregate is in the form of a pellet or a briquette, preferably a pellet.
- 25 16. A biochar aggregate of claim 14 or 15 having any of the features of claims 8 to 12.
17. A composition comprising a biochar aggregate of any one of claims 13 to 16.
18. A composition of claim 17, further comprising bitumen, cementitious binder, resin,
30 and/or paint, preferably bitumen or cementitious binder, more preferably bitumen.
19. A composition of claim 17 or 18, wherein the composition comprises 1 to 40 wt% of the biochar aggregate, preferably 1 to 20 wt% of the biochar aggregate, more preferably 1 to 10wt%.
- 35 20. A composition of claim 18 or 19, wherein the composition comprises 1 to 40 wt% of the bitumen, cementitious binder, resin, and/or paint, preferably 1 to 20wt%, such as 1 to 10wt%.

21. A method of manufacturing a composition of claims 17 to 20, comprising the step of mixing the biochar aggregate of any one of claims 13 to 16 with at least one other component (such as a binder, resin, or paint).
- 5 22. Use of a biochar aggregate of any one of claims 13 to 16 to:
- (i) place the biochar aggregate in a location such that the biochar aggregate is exposed to the natural atmosphere; and/or
 - (ii) fill a depression in the ground or to change the elevation of the ground; or
 - (iii) insulate a building component.
- 10 23. An engineered fill, geological fill or insulation material, comprising a biochar aggregate of any one of claims 13 to 16.