

ARTICLES OF MANUFACTURE AND METHODS OF PRODUCTION**5 TECHNOLOGICAL FIELD**

The invention is in the field of environmentally friendly articles of manufacture and methods of manufacturing the same.

BACKGROUND ART

References considered to be relevant as background to the presently disclosed
10 subject matter are listed below:

- International patent application publication No. WO2015173819

Acknowledgement of the above references herein is not to be inferred as meaning that these are in any way relevant to the patentability of the presently disclosed subject matter.

15 BACKGROUND

The building materials industry products is controlled by conventional cement, quick lime, gypsum and fired clay. The methods of producing those materials are one of the most environmentally polluting processes in the industrial world.

Use of earth-based mixtures for producing new products has been described by
20 WO2015173819. Specifically described are earth-based mixtures, methods of preparing mixtures and a process of forming articles of manufacture, as well as a process of manufacturing articles in molds involving compression molding of the earth-based mixtures containing one or more of sand, silt, clay, minerals and combinations of same.

GENERAL DESCRIPTION

25 The presently disclosed subject matter is based on the development of compressed articles made of earth-derived materials and/or recycled substances, also referred as CES
- Circular Engineered Stone (natural binder-based version of engineered stone or

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biogeo-agglomerated stone). The compressed articles can have a variety of shapes and uses.

Thus, in accordance with a first aspect of the presently disclosed subject matter there is provided an article of manufacture comprising a compressed mixture of

5 (a) particulates comprising a blend of clay material comprising at least one clay mineral having plasticity index above about 20% and liquid limit above about 40%, as determined according to ASTM D4318 and (ii) water insoluble inorganic material; and

 (b) fibrous material;

10 wherein a sample of said article of manufacture is characterized by at least one of

- a shape parameter of said water-insoluble inorganic material within a range of 0.24 and 0.33;
- flexural strength of at least 7MPa when measured according to
15 ASTM C99;
- compressive strength of at least 30 MPa when measured according to ASTM C170 on a cubic sample of the article of manufacture;
- compressive strength of at least 3 MPa when measured according to
20 EN 772-1 for air-dried specimens, on said sample having a shape of a hollowed masonry block;
- elastic modulus of between 2.5GPa and 15GPa when measured according to ASTM C 99,
- a ratio between Ultimate Flexural Strength (UFS) strain to yield point
25 strain of at least 1.2, in combination with a ratio between UFS toughness to yield point toughness of at least 2 when measured according to ASTM C 99;
- inorganic cation content of at least 0.00034wt%.

In accordance with a second aspect of the presently disclosed subject matter, there is provided a method for producing an article of manufacture, the method comprising:

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- placing within a mold a wet mixture comprising
 - (a) particulates comprising a blend of (i) clay material comprising at least one clay mineral having a plasticity index above about 20% and liquid limit above about 40%, as determined according to ASTM4318 and (ii) water insoluble inorganic material;
 - (b) fibrous material, and
 - (c) a liquid comprising inorganic cations, in water; the amount of said liquid being between about 5wt% and 50wt% out of the total weight of particulates and fibrous material;
- said wet mixture having a plasticity of between 8 to 1.15 when measuring according to pfefferkorn test;
- subjecting the wet mixture to compression molding under conditions that allow controlled liquid removal out of the wet mixture during application of compression forces onto said mixture to obtain a compressed mixture, said compression molding is conducted until reaching at least one of
 - removal of at least 7% of liquid out of a total weight of the wet mixture prior to said compression, and
 - removal of liquid out of a total weight of the wet mixture prior to said compression, until reaching a liquid content in said compressed mixture of less than 15%; and
- removing the compressed mixture from the mold to obtain an article of manufacture.

BRIEF DESCRIPTION OF THE DRAWINGS

In order to better understand the subject matter that is disclosed herein and to exemplify how it may be carried out in practice, embodiments will now be described, by way of non-limiting example only, with reference to the accompanying drawings, in which:

Figure 1 provides a scatter plot representing cumulative particle size distribution with an optimal shape parameter, with a target shape factor range of 0.24-0.33 being

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presented by the solid lines, and the optimal distribution being illustrated by the dashed line.

Figure 2 provides a scatter plot representing cumulative particle size distribution in an insoluble particulate material mixture with an optimal shape parameter based on fitting to the optimal distribution curve, with a target shape factor range of 0.24-0.33 being represented by the solid lines, and the optimal distribution being illustrated a dashed line, and particle size distribution of the experimental mixture being represented by a doubled line.

Figure 3 provides a scatter plot representing cumulative particle size distribution in an insoluble particulate material mixture with out-of-range shape parameter based on fitting to the optimal distribution curve, with a target shape factor range of 0.24-0.33 being represented by the solid lines, the optimal distribution being illustrated by the dashed line and particle size distribution of the experimental mixture being represented by a doubled line.

Figure 4 is a graph showing the cumulative particle size distribution of the example mixture of Table 3 with its corresponding upper and lower limits.

Figure 5 is a graph showing the flexural strength of articles prepared with different pressing pressures and different pressing duration, both influence the water extraction efficiency and its amount ("squeeze").

Figure 6 is a graph showing toughness up to yield point (elastic region area under the stress-strain curve, also called modulus of resilience), with toughness up to UFS being defined by the sum of the elastic region and plastic region areas under the stress-strain curve.

Figure 7A-7B is a graph showing the compressive strength of a full block of 75*75*75mm (Figure 7A) and of a hollow block (Figure 7B).

DETAILED DESCRIPTION

The presently disclosed subject matter concerns articles and methods of producing the same. Specifically, the articles contain or are made of earth-based materials and particularly clay material and water insoluble inorganic material that are being processed in a manner to provide resilient articles.

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In the following description all examples and definitions referring to the presently disclosed articles, are to be considered equally and equivalently to refer to the presently disclosed methods, *mutatis mutandis*. Thus, for example, when referring to clay material in the context of the presently disclosed articles, its meaning should be equally understood
5 in the context of to the presently disclosed methods, unless otherwise indicated.

In accordance with a first aspect of the presently disclosed subject matter there is provided an article of manufacture (or in short "article") comprising a compressed mixture of (a) particulates comprising a blend of (i) clay material, comprising at least one clay mineral having a plasticity index above 20% and liquid limit above 40% as
10 determined according to ASTM D4318 and (ii) water insoluble inorganic material; and (b) fibrous material.

The article is characterized by at least one of the following characteristics, as further described below:

- a shape parameter of said water-insoluble inorganic material within a
15 range of 0.24 and 0.33;
- flexural strength of at least 7MPa when measured on a sample of the article of manufacture with/without coating, said measurement being according to ASTM C 99;
- compressive strength of at least 30 MPa when measured on a cubic sample
20 according to ASTM C 170;
- compressive strength of at least 3 MPa when measured on a full hollowed masonry block according to EN 772-1;
- elastic modulus 2.5GPa up to 15GPa as measured according to ASTM C99;
- 25 - a ratio between Ultimate Flexural Strength (UFS) strain to yield point (YP) strain of at least 1.2, in combination with a ratio between UFS toughness to yield point (YP) toughness of at least 2 when measured according to ASTM C 99; and
- inorganic cation content of at least 0.00034wt%.

In the context of the present disclosure, when referring to an article that comprises
30 a compressed mixture it is to be understood to mean any one or combination of the

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following: the article is formed at least following a compression process of the mixture from which it is formed; and/or the particulates forming the article are pressed against one another such that no gaps are visible to the naked eye within the particulate mass.

The term "*mixture*" as used herein relates to a combination of at least the
5 components (a) to (b) as described hereinabove and below. The mixture may be formed in any way suitable for any scale of production of the mixture.

In some examples, the term "*clay*" per se should be understood to have its commonly acceptable meaning, namely, fine grained natural rock or soil material that combines one or more clay minerals, at times, with variable amounts of iron, magnesium,
10 alkali metals, alkaline earths, and other cations, and at times trace amounts of metal oxides.

The "*clay material*" in the context of the presently disclosed subject matter can be defined by its composition and/or by its grain size and/or by plasticity index of one or more of the clay minerals forming it, as well as by other parameters.

15 In some examples of the presently disclosed subject matter, the clay minerals forming part of the clay material comprise at least one oxide selected from the group consisting of silica, alumina, magnesia, and combination of same.

In some examples of the presently disclosed subject matter, the clay minerals comprise at least aluminum phyllosilicate.

20 In the context of the presently disclosed subject matter, the clay material can be characterized by plasticity of one or more of the clay minerals therein. The plasticity can be determined by standard test methods for liquid limit, plastic limit, and plasticity index of soils. In some examples, the clay material has a plasticity that is defined by the combination of its plasticity index and its liquid limit.

25 In the context of the present disclosure, the term "*plasticity index*" is used to define the range of the water content within which the clay material (and more specifically, the clay mineral) achieves its plastic state.

The PI is determined by the liquid limit and the plastic limit of the clay mineral according to the following acceptable equation:

30
$$I_p = W_l - W_p$$

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Where:

W_l = liquid limit

W_p = plastic limit

I_p = plasticity index

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The term "*liquid limit*" is to be understood as the water/moisture content (in weight percentage, wt%) at which the behavior of a material changes from a plastic state to a liquid state.

10 The term "*plastic limit*" is to be understood as the water/moisture content at which a thread of material with 3.2mm diameter begins to crumble.

In some examples, the PI and liquid limit are determined according to ASTM D 4318 and/or according to Israeli Standard 253, paragraphs 103.3.2, 103.4, 103.8-103.10, 206.1, 206.1.2, 206.2.2 and Table 6 therein.

15 In some examples of the presently disclosed subject matter, the clay material comprises a combination of clay minerals, each being defined by its PI and liquid limit, the liquid limit being greater than the PI.

In some examples of the presently disclosed subject matter, the clay material comprises at least one clay mineral having a PI above about 20% and liquid limit above about 40%.

20 In some examples of the presently disclosed subject matter, the clay material comprises at least one clay mineral having a PI above amount 35%.

In some examples of the presently disclosed subject matter, the clay material comprises at least one clay mineral having a liquid limit above about 55%.

25 In some examples of the presently disclosed subject matter, the clay material comprises at least one clay mineral having a PI above about 35% and a liquid limit above about 55%.

In some examples of the presently disclosed subject matter, the clay material comprises at least one clay mineral having a liquid limit above about 400%.

30 In some examples of the presently disclosed subject matter, the clay material comprises at least one clay mineral having a liquid limit above about 450%.

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In some examples of the presently disclosed subject matter, the clay material comprises at least one clay mineral having a liquid limit above about 500%.

In some examples of the presently disclosed subject matter, the clay material comprises at least the mineral ball clay.

5 In some examples of the presently disclosed subject matter, the clay material comprises at least ball clay having a PI above about 20%, at times above about 35%, and liquid limit above about 40%, at times above 55%.

In some examples of the presently disclosed subject matter, the clay material comprises at least bentonite.

10 In some examples of the presently disclosed subject matter, the clay material comprises at least bentonite having a liquid limit above about 400%, at times above 450%, at times, above 500%.

In some examples of the presently disclosed subject matter, the clay material comprises at least a combination of said bentonite and said ball clay.

15 In some examples, the clay material comprises a combination of said bentonite and said ball clay at a weight ratio of between about 1:1.1 and about 1.2:1.

In some examples, the clay material comprises a combination of said bentonite and said ball clay at a weight ratio of between about 1:2 and about 1:5.

20 In some examples, the clay material comprises a combination of said bentonite and said ball clay at a weight ratio of between about 1:2.1 and about 1:3; at times between about 1:2.1 and about 1:2.6; at times, between about 1:2.1 and 1:2.5; at times between about 1:2.1 and about 1:2.4.

In some examples of the presently disclosed subject matter, the clay material is recognized as an industrial clay.

25 The presently disclosed article also comprises said water insoluble inorganic material.

In the context of the presently disclosed subject matter, the term "*water insoluble inorganic material*" should be understood to encompass material that comprises any one of insoluble rock derived material.

In some examples of the presently disclosed subject matter, the water insoluble inorganic material comprises sedimentary rock material.

For the purposes of the present discussion, when referring to "*sedimentary rock material*" it is to be understood to include any one or combination of the three major categories of sedimentary rocks, being (1) terrigenous clastic sedimentary rocks, (2) carbonates (e.g. limestone and dolomite) and (3) metamorphic, e.g. marble and quartzite.

When referring to "*terrigenous clastic sedimentary rocks*" it is to be understood to refer to sedimentary rocks composed of the detrital fragments of preexisting rocks and minerals and are conventionally considered to be equivalent to clastic sedimentary rocks in general. Because most of the clasts are rich in silica, they are also referred to as siliciclastic sedimentary rocks. Thus, in some examples of the presently disclosed subject matter, the water insoluble inorganic material comprises at least siliciclastics. These siliciclastics can be identified or further classified on the basis of clast diameter as conglomerate and breccia, sandstone, siltstone, and finer-than-silt-sized mudrock (shale, claystone, and mudstone), all of which constitute part of the "*water insoluble inorganic material*" of the presently disclosed subject matter.

When referring to "*carbonates*" in the context of sedimentary rocks, it is to be understood to encompass one or both of limestones and dolomites, which, in turn, known to consist essentially of the minerals aragonite, calcite, and dolomite.

In some examples, the water insoluble inorganic material comprises at least terrigenous clastic sedimentary rock.

In some examples, the water insoluble inorganic material comprises at least limestone and/or dolomite.

In some examples, the water insoluble inorganic material comprises a combination of terrigenous clastic sedimentary rock and limestone and/or dolomite.

In some examples of the presently disclosed subject matter, the sedimentary rock comprises any one or combination of limestone, chalk, sandstone, shale.

In some examples of the presently disclosed subject matter, the water insoluble inorganic material comprises igneous rock. Igneous rocks are known as a type of rock

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that is formed when molten rock (rock liquefied by intense heat and pressure) cools to a solid state. A non-limiting list of igneous rock include basalt, quartz, pumice, obsidian, rhyolite, scoria, dacite, granite, gabbro, diabase, diorite, pegmatite, and peridotite.

5 In some examples of the presently disclosed subject matter, the water insoluble inorganic comprises at least one igneous rock.

In some examples of the presently disclosed subject matter, the water insoluble inorganic comprises at least basalt.

In some examples of the presently disclosed subject matter, the water insoluble inorganic comprises at least quartz.

10 In some examples, the water insoluble inorganic comprises at least metamorphic rock, e.g. marble, quartzite etc.

non-limiting examples of water insoluble inorganic material includes quartz, limestone, dolomite, chalk, sandstone, shale, pumice, obsidian, rhyolite, scoria, dacite, granite, gabbro, diabase, diorite, pegmatite, peridotite, basalt, marble, quartzite and any
15 combination of same.

In the context of the present disclosure, it is to be understood that the term "*water insoluble inorganic material*" refers to material that is different/other than the herein referred to clay material. This being said notwithstanding the fact that the clay material and the water insoluble inorganic material may have overlap in one or more of the
20 minerals they contain (e.g., in the presence of clay minerals).

In some examples of the presently disclosed subject matter, the water insoluble inorganic material comprises inorganic material consists essentially of earth derived inorganic matter.

25 In some other examples of the presently disclosed subject matter, the water insoluble inorganic material comprises recycled matter comprising water insoluble inorganic material.

The term "*recycled matter comprising water insoluble inorganic material*" is to be understood to encompass material that comprises processed earth derived material and as such, may include also synthetic material, such as inorganic binders.

In some examples of the presently disclosed subject matter, the recycled matter comprises recycled construction and demolition waste, such as concrete.

In some examples of the presently disclosed subject matter, the recycled matter comprising water insoluble inorganic material comprises at least 50wt%, at times at least 60wt%, at times, at least 70wt%; at times at least 80wt%; at times, at least 90wt%, or even, at times 100wt% of said water insoluble inorganic material.

In some examples of the presently disclosed subject matter, the recycled matter comprising water insoluble inorganic material comprises or is recycled concrete.

The combination of the clay material and the water insoluble inorganic material constitute together the particulate portion of the herein disclosed article. This being different from the (solid) fibrous material, that while being in fiber form, is distinguishable from the clay material and the insoluble inorganic material.

In accordance with the presently disclosed subject matter, the particulates have a maximum dimension of not more than 20mm, when measured along the longest axis of the particulates. In some examples of the presently disclosed subject matter, the particulates have a maximum dimension of not more than 19mm; at times, not more than 18mm; at times, not more than 17mm; at times, not more than 16mm; at times, not more than 15mm; at times, not more than 14mm; at times, not more than 13mm; at times, not more than 12mm; at times, not more than 11mm; at times, not more than 10mm; at times, not more than 9mm; at times, not more than 8mm; at times, not more than 7mm; at times, not more than 6mm; at times, not more than 5mm; at times, not more than 4mm when measured along the longest axis of the particulates.

The article of manufacture disclosed herein is characterized by one or more physical and chemical properties.

In some examples of the presently disclosed subject matter, the water insoluble inorganic material is characterized by a shape parameter of the water insoluble inorganic material.

In the context of presently disclosed subject matter, when referring to "*shape parameter*" (also known by the terms "*volumetric shape parameter*", "*volume shape parameter*" or "*shape factor*") refer to a value (q) in the formula:

$$P(D) = \left(\frac{D}{D_{\max}} \right)^q$$

wherein P(D) is the cumulative (or total integral) particle size distribution, D is the varied particle size D_{max} is the maximum size of particles in the specific mixture and q is the shape parameter. [P. Domone, C.H. Wen, Testing of binders for High Performance Concrete, Cement and Concrete Research 27 (1997) 1141– 1147.;
 5 W.B. Fuller, S.E. Thompson, The laws of proportioning concrete, Transactions of the American Society of Civil Engineers 33 (1907) 222– 298].

In some examples of the presently disclosed subject matter, the shape parameter of the water insoluble inorganic material is between about 0.24 and about 0.33; at times, between about 0.25 and about 0.30.

10 **Table 5** provides a non-limiting example of the effect of using mixture having a shape parameter within the presently disclosed range, vs. a mixture having a shape parameter outside the scope, the latter being inferior at least with respect to product's flexural strength. Further reference can be made to Figures 2 – 4 supporting the presently disclosed shape parameter range.

15 In some examples of the presently disclosed subject matter, the article is characterized by its flexural strength.

The flexural strength defines the ability of the material to withstand bending forces applied perpendicular to its longitudinal axis. The higher the value, means the article is more resilient to the bending forces applied.

20 The flexural strength of the presently disclosed article can be determined according to ASTM C 99, using three points bending or four points bending (as described in <https://www.compositesworld.com/articles/notched-testing-of-sandwich-composites-the-sandwich-open-hole-flexure-test>) with ½ load span, and the following parameters:

$$\text{Span}=0.7*L,$$

25 Speed control=1MPa/sec.

In some examples of the presently disclosed subject matter, the article has a flexural strength of at least 7MPa; at times, of at least 7.1MPa; at times, of at least 7.2MPa; at times, of at least 7.3MPa; at times, of at least 7.4MPa; at times, of at least 7.5MPa; at times, of at least 7.6MPa; at times, of at least 7.7MPa; at times, of at least

7.8MPa; at times, of at least 7.9MPa; at least 8MPa; at times, at least 9MPa; at times, at least 10MPa; at times, at least 11MPa; at times, at least 12MPa; at times, at least 13MPa.

In some examples of the presently disclosed subject matter, the article has a flexural strength that is preferably above 7MPa, at times preferably above 7.1MPa.

5 In some examples of the presently disclosed subject matter, the article has a flexural strength of at most 30MPa; at times, at most 25MPa; at times, at most 20MPa; at times, at most 15MPa.

In some examples of the presently disclosed subject matter, the article has a flexural strength within a range of above 7.1MPa and below 30MPa, where any range of
10 value within this range constitutes an independent example of the present disclosure.

Tables 4 and 5 herein provide non-limiting examples for flexural strength of a specimen indicative of the flexural strength that can be obtained for articles according to the presently disclosed subject matter. According to these non-limiting examples, the specimens have flexural strengths of 9.1 MPa (composition 1), 8.3 MPa (composition 2)
15 and 10.85 MPa (Composition 3) and 8.61 MPa (repetition of composition 1 with some of the shape parameter within the defined range), all values being well above 7MPa.

In some examples of the presently disclosed subject matter, the article is characterized by its compressive strength.

The compressive strength defines the ability of the material to withstand
20 compression forces applied perpendicular to its height axis. The higher the value, means the article is more resilient to the compression forces applied.

The compressive strength of the presently disclosed article can be determined according to either by ASTM C 170 for full blocks of 50mm or more, or by EN 772-1 for hollowed blocks, according to the parameters detailed in each standard for low
25 temperature drying conditions.

In some examples of the presently disclosed subject matter, the article has a full block compressive strength of at least at least 30MPa; at times, at least 35MPa; at times, at least 40MPa; at times, at least 45MPa; at times, at least 50MPa; at times, at least 55MPa; at times, at least 60MPa; at times, at least 65MPa.

In some examples of the presently disclosed subject matter, the article has a full block compressive strength of at most 130MPa; at times, at most 120MPa; at times, at most 110MPa; at times, at most 100MPa; at times, at most 90MPa; at times, at most 80MPa; at times, at most 70MPa.

5 In some examples of the presently disclosed subject matter, the article has a full block compressive strength within a range of above 30MPa and below 130MPa, where any range or value within this range constitutes an independent example of the present disclosure.

Figure 7A provides a non-limiting example of the compressive strength of a full
10 block according to the presently disclosed subject matter, having a value of 97MPa. In some examples of the presently disclosed subject matter, the article has a hollow block compressive strength of at least 3MPa; at times, at least 4MPa; at times, at least 5MPa; at times, at least 6MPa; at times, at least 7MPa; at times, at least 8MPa; at times, at least 9MPa; at times, at least 10MPa; at times, at least 11MPa; at times, at least 12MPa; at
15 times, at least 13MPa.

In some examples of the presently disclosed subject matter, the article has a hollow block compressive strength of at most 30MPa; at times, at most 25MPa; at times, at most 20MPa; at times, at most 15MPa.

 In some examples of the presently disclosed subject matter, the article has a
20 hollow block compressive strength within a range of above 3MPa and below 30MPa, where any range or value within this range constitutes an independent example of the present disclosure.

Figure 7B provides a non-limiting example of the compressive strength of a hollow block according to the presently disclosed subject matter, having a value of
25 14MPa with coating, and 6.1MPa without coating.

In some examples of the presently disclosed subject matter, the article is characterized by its elastic/plastic properties at flexural testing.

The flexural elastic behavior ("*elastic modulus*") defines the ability of the material to withstand deflections to its longitudinal axis up to its elastic limit.

The plastic/elastic correlation is defined by the combination of the ratio between the strain at the Ultimate Flexural Strength (UFS) point and its yield point (YP) strain (“UFS/YP strain ratio”), and the ratio between the toughness at the Ultimate Flexural Strength (UFS) point and its yield point toughness (“UFS/YP toughness ratio”). This combination defines the material total elastic-plastic behavior.

When referring to UFS/YP strain ratio it is to be understood as a measure of how much a material can be deformed before it breaks in comparison to how much it can be deformed before it undergoes permanent, non-recoverable deformation. This ratio may provide insight into the ductility of the material, and the reinforcing fibres been used.

When referring to UFS/YP toughness ratio it is to be understood as a measure of how much work (energy) is needed to invest into the material in order to break it in comparison to how much work is needed for the elastic (resilient) region before it undergoes permanent, non-recoverable deformation. This ratio may provide insight into the toughness of a material, as well as its ability to absorb energy before failure, which also relates to the reinforcing efficiency of the fibres – depending on the optimization of particle size distribution (packing density), optimization of fibres length and type, optimization of binder content and properties.

The flexural elastic behavior (referred to herein by the term "flexural elastic modulus") and the plastic/elastic correlation (referred to herein by the terms “UFS/YP strain ratio”, “UFS/YP toughness ratio”) of the presently disclosed article can be determined according to ASTM C 99, using three points bending or preferably by four points bending (as described in <https://www.compositesworld.com/articles/notched-testing-of-sandwich-composites-the-sandwich-open-hole-flexure-test>) with ½ load span, and the following parameters:

Span=0.7*L,

Speed control=1MPa/sec.

In some examples of the presently disclosed subject matter, the article has a flexural elastic modulus of between 2.5GPa and 15GPa.

Notably, the flexural elastic behavior can be deduced from the slop of the stress-strain curve at the elastic region while the flexural strength is the UFS in such curve. In this connection, reference can be made to Figure 6 which provides a non-limiting example

of the flexural elastic modulus of a specimen according to some examples of the presently disclosed subject matter, the specimen having an elastic modulus of 6.13GPa (which is the slope of the elastic region in this Figure 6).

In some examples of the presently disclosed subject matter, the article has
5 UFS/YP strain of at least 1.2, combined with UFS/YP toughness of at least 2.0.

These ratios can be deduced from an article's stress-strain curve. Specifically, the UFS/YP strain is calculated by dividing the UFS maximal point strain value (e.g. 0.0023 in Table 7) by the end of the elastic region point (e.g. 0.0015 in Table 7), while the UFS/YP toughness is deduced from dividing the total area of the plastic region (e.g.
10 0.6903 in Table 7) by the total area of the elastic region (e.g. 0.0068 in Table 7).

These properties can be increased by adding to the mixture additives such as, and without being limited thereto, fibers, or increasing length of existing fibers, or any other additives commonly known in this industry.

Without being bound by theory, the presently disclosed article, and its individual
15 properties, including the flexural strength the compressive strength and/or the plastic/elastic properties (the combination of the elastic modulus and aforesaid ratio) are independently a result of the unique method by which the articles are formed in accordance with the present disclosure. As will be further detailed below, the herein disclosed method of producing the articles involves active and controlled liquid removal.
20 During the liquid removal particles are compressed and densified one against the other, and inner voids, if exist, become smaller and fewer, resulting in an increase in strength. Moreover, without being bound by theory, densification increases reinforcing efficiency of the fibers because the particulates increase their interlock on the fibers, and thereby providing increased tensile/flexural strength.

25 In some examples of the presently disclosed subject matter, the article is characterized by its inorganic cation content.

When referring to "*inorganic cation*" it is to be understood to encompass any one or combination of potassium, ammonium, sodium, calcium, magnesium, lithium, aluminum.

30 In some examples, the inorganic cation is a monovalent cation.

In some examples, the inorganic cation is potassium.

In some other examples, the inorganic cation is ammonium.

The inorganic cation is present in an amount that is greater than its combined amount in clay material and rock derived water insoluble inorganic material from which the article is made, when they are in their natural state (i.e., in nature). The amounts being in clay material and rock derived water insoluble inorganic material as known in the art for each. The greater amount being as a result of externally added cations during the method of manufacturing, as will be further described herein below.

In some examples of the presently disclosed subject, the inorganic cation is in an amount that is equal or above 0.00034wt%.

In some examples of the presently disclosed subject, the article disclosed herein has an inorganic cation content of at least 0.00034wt%; at times, of at least 0.0007wt%; at times, of at least 0.001wt%; at times, of at least 0.003wt%; at times, of at least 0.004wt%; at times, of at least 0.005wt%; at times, of at least 0.006wt%; at times, of at least 0.007wt%; at times, of at least 0.008wt%; at times, of at least 0.009wt%; at times, of at least 0.01wt%; at times, of at least 0.011wt%; at times, of at least 0.0115wt%; at times, of at least 0.012wt%; at times, of at least 0.0125wt%; at times, of at least 0.013wt%.

Without being limited thereto, Table 6 provides one example according to the presently disclosed subject matter, showing that with potassium content (inorganic content) of at least 0.0098wt% (externally added), the flexural strength of the specimen was increased as compared to the same composition albeit without externally added potassium.

In some examples of the presently disclosed subject, the article disclosed herein has an inorganic cation content of at most 0.5wt%.

Generally, the amount of inorganic cation can be determined by any one of methods known to those versed in the art of analytical chemistry. Analysis can be made by any one of atomic emission spectroscopy (AES) and in particularly Inductively Coupled Plasma AES (ICP-AES), Inductively Coupled Plasma Mass Spectrometry (ICP-MS) or Ion Chromatography (IC).

In some examples, the article of manufacture is characterized by two of the properties selected from the shape parameter of the insoluble inorganic material, flexural strength, compressive strength, elastic modulus (and ratio), and inorganic cation content, each being as defined herein; at times by three of these properties; at times, by four of these properties; at times by all of these aforementioned properties.

In some examples of the presently disclosed subject matter, the article disclosed herein is characterized by a shape parameter of said water insoluble inorganic material within a range of 0.24 and 0.33 and a flexural strength of at least 7MPa.

In some examples of the presently disclosed subject matter, the article disclosed herein is characterized by a shape parameter of said water insoluble inorganic material within a range of 0.24 and 0.33 and compressive strength of at least 30MPa for full block or at least 3MPa for hollowed block.

In some examples of the presently disclosed subject matter, the article disclosed herein is characterized by a shape parameter of said water insoluble inorganic material within a range of 0.24 and 0.33 and flexural elastic modulus of at least 2.5GPa and UFS/YP strain of at least 1.2 combined with UFS/YP toughness of at least 2.0.

In some examples of the presently disclosed subject matter, the article disclosed herein is characterized by a shape parameter of said water insoluble inorganic material within a range of 0.24 and 0.33 and inorganic cation content of at least 0.00034wt%.

In some examples of the presently disclosed subject matter, the article disclosed herein is characterized by a flexural strength of at least 7MPa and inorganic cation content of at least 0.00034wt%.

In some examples of the presently disclosed subject matter, the article disclosed herein is characterized by a flexural strength of at least 7MPa and compressive strength of at least 30MPa for full block or at least 3MPa for hollowed block.

In some examples of the presently disclosed subject matter, the article disclosed herein is characterized by a compressive strength of at least 30MPa for full block or at least 3MPa for hollowed block and inorganic cation content of at least 0.00034wt%.

In some examples of the presently disclosed subject matter, the article disclosed herein is characterized by a shape parameter of said water insoluble inorganic material

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within a range of 0.24 and 0.33, a flexural strength of at least 7MPa and compressive strength of at least 30MPa for full block or at least 3MPa for hollowed block.

In some examples of the presently disclosed subject matter, the article disclosed herein is characterized by a shape parameter of said water insoluble inorganic material
5 within a range of 0.24 and 0.33, a flexural strength of at least 7MPa and inorganic cation content of at least 0.00034wt%.

In some examples of the presently disclosed subject matter, the article disclosed herein is characterized by a flexural strength of at least 7MPa, compressive strength of at least 30MPa for full block or at least 3MPa for hollowed block and inorganic cation
10 content of at least 0.00034wt%.

In some examples of the presently disclosed subject matter, the article disclosed herein is characterized by a shape parameter of said water insoluble inorganic material within a range of 0.24 and 0.33, a flexural strength of at least 7MPa, compressive strength of at least 30MPa for full block or at least 3MPa for hollowed block and inorganic cation
15 content of at least 0.00034wt%.

In some examples of the presently disclosed subject matter, the article disclosed herein is characterized by a shape parameter of said water insoluble inorganic material within a range of 0.24 and 0.33, UFS/YP strain of at least 1.2 and UFS/YP toughness of at least 2.0.

20 In some examples of the presently disclosed subject matter, the article disclosed herein is characterized by a shape parameter of said water insoluble inorganic material within a range of 0.24 and 0.33, UFS/YP strain of at least 1.2 and UFS/YP toughness of at least 2.0 and inorganic cation content of at least 0.00034wt%.

In some examples, the presently disclosed subject matter is characterized by the
25 combination of a shape parameter of said water insoluble inorganic material within a range of 0.24 and 0.33; and flexural strength of at least 7MPa when measured according to ASTM C99; and compressive strength of at least 30MPa when measured according to ASTM C170 on a cubic sample of said article of manufacture; and compressive strength of at least 3 MPa when measured according to EN 772-1 for air-dried specimens, on said
30 sample having a shape of a hollowed masonry block; and elastic modulus of between 2.5GPa and 15GPa when measured according to ASTM C99; and a ratio between Ultimate

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Tensile Strength (UFS) strain to yield point strain of at least 1.2 and a ratio between UFS toughness to yield point toughness of at least 2; and inorganic cation content of at least 0.00034wt%.

In some examples of the presently disclosed subject matter, the article of
5 manufacture can also be characterized by the clay material content and/or the water insoluble inorganic material content.

The amount of the clay material and/or the amount of the water insoluble
inorganic material within the article can be determined using any technique known to
those versed in the art of petrography. For example, and without being limited thereto,
10 the amount of the clay material and/or the amount of the water insoluble inorganic
material can be determined using any one or combination of Scanning Electron
Microscopy (SEM), Polarizing Light Microscopy (PLM), density gradient separation,
Thermal Gravimetry Analysis (TGA) and combinations thereof.

In some examples, the amount of the constituents is determined by Scanning
15 Electron Microscopy (SEM) equipped with an Energy Dispersive Spectroscopy (EDS)
detector.

In some examples of the presently disclosed subject matter, the article comprises
clay material in an amount of between about 10% and about 50% out of the total volume
of the article. At times, the amount of clay material in the presently disclosed article is
20 between about 10% and about 40%; at times, between about 10% and about 30%; at times,
between about 10% and about 28%; at times, between about 15% and about 30%; at times,
between about 20% and about 35%.

At times, the amount of the clay material is determined in line (based on) the
amount of aluminum phyllosilicate in the article of manufacture, as detected by e.g. SEM.

25 In some examples of the presently disclosed subject matter, the article comprises
water-insoluble inorganic material in an amount that constitutes at least 50% out of a total
volume of the article of manufacture; at times, at least 60% out of the total volume of the
article of manufacture; at times, between about 50% and about 90%; at times, between
about 60% and about 75%; at times, between about 65% and about 75%; at times, between
30 about 70% and about 85%; at times, between about 80% and about 90%.

In addition to particulate material, the article of manufacture comprises fibrous material.

In some examples, when referring to fibrous material it is to be understood to encompass at least natural fibrous material, i.e. derived from natural substances.

5 In some examples of the presently disclosed subject matter, the fibrous material is derived from plant material.

In some examples of the presently disclosed subject matter, the fibrous material comprises at least one of cellulose, hemicellulose, and lignocellulose fibers.

10 In some examples of the presently disclosed subject matter, the fibrous material comprises at least one of cellulose.

In some examples of the presently disclosed subject matter, the fibrous material comprises fibers selected the group consisting of flax, sisal, hemp, jute, cotton, abaca, bamboo, ramie, banana, and kenaf and combinations of same.

15 In some examples of the presently disclosed subject matter, the fibrous material comprises at least flax fibers.

In some examples of the presently disclosed subject matter, the fibrous material comprises basalt fibers.

In some examples of the presently disclosed subject matter, the fibrous material comprises or is synthetic fibers.

20 In some examples, the synthetic fibers comprise any one of glass fibers, polyester fibers, basalt fibers, carbon fibers.

25 In some examples of the presently disclosed subject matter, the fibrous material comprises fibers have a filamentous or thread shape, with a length (longitudinal dimension) of at least 2mm; at times, of at least 3mm; at times, of at least 4mm; at times, of at least 5mm.

In some examples of the presently disclosed subject matter, the fibrous material has a length within a range of between about 2mm and about 30mm; at times, between about 2mm and about 20mm;; at times, between about 2mm and about 15mm; at times, between about 4mm and about 18mm; at times, between about 7mm and about 12mm; at

times, between about 5mm and about 16mm; at times between about 15mm and about 20mm.

It is to be appreciated that such longitudinal dimension of the fibrous material can be determined by observation with the naked eye or by light microscopy.

5 In some examples of the presently disclosed subject matter, the content of the fibrous material within the article can be determined. In some examples, the amount of fibrous material is determined by one of polarizing microscopy, density gradient separation or Thermal Gravimetry Analysis (TGA).

10 In some examples of the presently disclosed subject matter, the fibrous material is in an amount of at least 1% out of a total volume of the disclosed article.

In some examples, the fibrous material is in an amount of between about 1% and about 20% out of a total volume of the presently disclosed article; at times, between about 1% and about 12%; at times, between about 1% and about 6%; at times, between about 5% and about 10%; at times, between about 8% and about 16%.

15 In some examples of the presently disclosed subject matter, the fibrous material is characterized by tensile strength of at least 400MPa. In this context, it is to be understood that when referring to tensile strength of the fibrous material it means the tensile strength of the elementary fibrous material, i.e. the smallest diameter fibre that can be separated by physical separation. In some examples of the presently disclosed subject
20 matter, the fibrous material is characterized by tensile strength of at least 450MPa; at times, of at least 500MPa.

 The article of manufacture disclosed herein can be characterized by any combination of the herein defined features, including one, two, three, four or more of the above features, each combination constituting a separate and independent example of the
25 present disclosure.

 In some examples of the presently disclosed subject matter, the article is essentially free of synthetic material. In this context, it is to be understood that when referring to essentially free, it means that if synthetic material is present, the amount of the synthetic material does not exceed a total of 20wt% out of the total weight of the
30 article, when in dry (or at least essentially dry) form.

The presently disclosed article can have any moldable shape and can be suitable for any purpose.

In some examples of the presently disclosed subject matter, the article of manufacture is shaped for use in construction, as a construction element.

5 Without being limited thereto, the article disclosed herein can be molded (preferably by compression molding) or injected or extruded into any one of brick, tile, block, lining, partitioning element, furniture or furniture element, roofing tile, cladding, facade, panels, boards, insulative products, interior design elements, small containers, under floor element, paving brick and element, or wall bricks.

10 The presently disclosed subject matter also provides a method of producing the herein disclosed articles of manufacture.

 The method involves, *inter alia*, compression molding. It has unexpectedly been found that when subjecting the raw/intake material for forming the articles, as will be further described hereinbelow, to compression, under controlled liquid removal
15 conditions, it is possible to significantly and unexpectedly improve the physical/mechanical properties of the resulting article (e.g. the flexural strength, compressive strength, plastic/elastic properties as defined herein).

 Independently from the molding conditions, it has been unexpectedly found that when the raw/intake material comprises water insoluble inorganic material having a shape
20 parameter within a narrow range of 0.24 and 0.33, it is also possible to significantly and unexpectedly improve the physical/mechanical properties of the resulting article (e.g. the flexural strength, compressive strength, plastic/elastic properties as defined herein).

 Further, independently from the molding conditions and/or shape parameter, it has been found that when externally adding an inorganic cation to the raw/intake material it
25 is possible to significantly and unexpectedly improve the physical/mechanical properties of the resulting article (e.g. the flexural strength, compressive strength, plastic/elastic properties as defined herein).

The presently disclosed subject matter thus also provides a method for producing the article of manufacture in accordance with the first aspect disclosed herein.

It is appreciated that the various features described above in connection with the presently disclosed article of manufacture also apply *mutatis mutandis* to the method for obtaining said article of manufacture. This applies, *inter alia*, to the clay material, the water insoluble inorganic material, the fibrous material, the inorganic cation etc.

5 Specifically, the method comprises placing within a mold a wet mixture comprising particulates comprising a blend of (a) clay material and (b) water insoluble inorganic material, and in addition to the blend, also fibrous material is included in the wet mixture. Further included in the wet mixture is liquid, e.g. water, comprising an inorganic salt comprising an inorganic cation, e.g. potassium.

10 After being placed in the mold, the wet mixture is subjected to compression molding under conditions that allow controlled liquid removal out of the mixture during application of compression forces onto the wet mixture. The controlled removal of liquid is conducted until reaching one of two criteria: 1) a removal of at least 7% of liquid out of a total weight of the wet mixture, more preferably reaching a removal of at least 10%
15 of liquid out of a total weight of the wet mixture, or 2) removal of liquid until reaching a liquid content leftover after the pressing of less than 15%, more preferably reaching a liquid content leftover after the pressing of less than 10%. to thereby obtain a compressed mixture.

 In some examples, controlled removal of liquid is conducted until at least 8wt%;
20 at times, at least 9wt%; at times, at least 10wt%; at times, at least 11wt%; at times, at least 12wt%; at times, at least 13wt%; at times, at least 14wt% or even at least 15wt% liquid out of the total weight of the wet mixture prior to placing within the mold and compression, is removed.

 Once a desired amount of liquid is removed (i.e. at least 7wt%), the resulting
25 compressed mixture can be extracted from the mold for further drying, to provide an article of manufacture.

 In some examples, controlled removal of liquid is conducted until reaching a liquid content leftover after the pressing of less than 15%; at times, less than 14wt%; at times, less than 13wt%; at times, less than 12wt%; at times, less than 11wt%; at times,
30 less than 10wt%; at times, less than 9wt% or even less than 8wt% liquid out of the total weight of the wet mixture prior to placing within the mold and compression.

Once a desired amount of liquid left within the mixture after pressing (i.e. less than 15wt%), the resulting compressed mixture can be extracted from the mold for further drying, to provide an article of manufacture.

The raw material introduced into the mold is prepared by mixing clay material, water insoluble inorganic material and fibrous material and wetting the same with the liquid containing the salt of the inorganic cation.

The amount of clay material, water insoluble inorganic material and fibrous material in the mixture can vary.

In line with the above disclosure relating to the article of manufacture, the amount of clay material in the mixture based on dry matter, can be, in accordance with some examples of the presently disclosed method, between about 10% and about 50% out of the total volume of the article. At times, the amount of clay material in the dry mixture of presently disclosed method is between about 10% and about 20%; at times, between about 15% and about 30%; at times, between about 25% and about 40%.

Further in line with the above disclosure relating to the article of manufacture, the amount of water-insoluble inorganic material in the mixture based on dry matter, can constitutes at least 50% out of a total volume of the dry mixture (before wetting with the liquid); at times, between about 60% and about 90%; at times, between about 60% and about 75%; at times, between about 65% and about 75%; at times, between about 70% and about 85%; at times, between about 80% and about 90%.

Further in line with the above disclosure relating to the article of manufacture, the amount of fibrous material in the dry mixture, prior to wetting, can be at least 1% out of a total volume of the dry mixture, at times, between about 1% and about 20% out of a total volume of the presently disclosed article; at times, between about 1% and about 6%; at times, between about 5% and about 9%; at times, between about 8% and about 12%.

The amounts of the clay material and the water-insoluble inorganic material can be determined, *inter alia*, to provide a desired particle size distribution of the water insoluble inorganic material.

In accordance with some examples of the presently disclosed subject matter, the water insoluble inorganic material is selected to provide one cumulative distribution that behaves within an upper and lower limit, derived from the mathematical equation of

$$P(D) = \left(\frac{D}{D_{\max}} \right)^q$$

(wherein $P(D)$ is the cumulative particle distribution, D is the varied particle size $D_{\max}$ is the maximum size of particles in the specific mixture and q is the shape parameter), in order to reach a desired shape parameter.

To achieve a desired shape parameter, insoluble inorganic materials with various
5 particle sizes are mixed together to achieve a particle size distribution fitting a curve corresponding to desired shape parameter, i.e. that fits the curve of **Figure 1**. The dry mixture is then wetted with the liquid.

The liquid includes water (which may be de-ionized, but not necessarily) and an inorganic salt, the latter as defined hereinabove with respect to the presently disclosed
10 article of manufacture.

The amount liquid would constitute at most 50% out of the total weight of the wet mixture.

In some examples of the presently disclosed subject matter, the amount of liquid added is such to constitutes between about 5% and about 50% out of a total weight of the
15 resulting wet mixture; at times between about 8% and about 40%; at times between about 11% and about 30%; at times between about 14% and about 20%.

In some examples of the presently disclosed subject matter, the amount of liquid added is to provide a wet mixture where the liquid constitutes between about 12% and about 20% out of a total weight of the wet mixture.

20 The liquid is prepared by dissolving the salt (namely a salt comprising the inorganic cation, as described hereinabove), in the de-ionized water.

In some examples, the amount of salt dissolved in the de-ionized water is up to about 0.1wt%; at times, up to about 0.09wt%; at times, up to about 0.08wt%; at times, up to about 0.07wt%; at times, up to about 0.06wt%; at times, up to about 0.05wt%; at times,
25 about up to 0.04wt%; at times, about up to 0.03wt%; at times, up to about 0.02wt%; at times, up to about 0.01wt%; at times, up to about 0.0099wt%; at times, up to about 0.0098wt%; at times, up to about 0.0095wt%; at times, about up to 0.009wt% out of the total weight of the wet mixture.

The wet mixture is then placed within a mold.

In accordance with the presently disclosed method, the molding process requires controlled liquid removal. When referring to controlled liquid removal, it is intended to refer, *inter alia*, to the rate of liquid removal during compression; mainly influenced by the duration of compression, the pressure applied onto the wet mixture during
5 compression, the applied heat to the mold and any combination thereof.

In some examples of the presently disclosed subject matter, the controlled compression is achieved using a mold that is equipped with (i) a pressing layer having a pressing layer front surface configured to press the mixture and extract the liquid therefrom, and an opposite pressing layer back surface; (ii) a backing plate having a
10 backing plate front surface facing the pressing layer back surface and an opposite backing plate back surface; and (iii) a liquid accumulating space extending at least partially in a plane parallel to the pressing layer and positioned between the backing plate back surface and the pressing layer front surface, the liquid accumulating space being configured to accumulate the liquid therein, as described in co-pending US patent application No.
15 63/375,360 filed on September 12, 2022.

In some examples of the presently disclosed subject matter, the controlled compression, and thus controlled liquid removal, is achieved using a mold that is equipped with (i) a pressing mechanism configured to receive the mixture therewithin and press the mixture to extract the liquid therefrom; (ii) a press member operatively
20 coupled to the pressing mechanism and configured to operate the pressing mechanism for pressing the mixture; and (iii) a biasing mechanism configured to continuously bias the pressing mechanism in a direction to pressurize the mixture and compensate for a pressure drop in the mixture due to said extraction of the liquid; as described in co-pending US patent application No. 63/375,360 filed on September 12, 2022. In some examples of the
25 presently disclosed subject matter, the pressing mechanism can comprise a first pressing layer and a second pressing layer, and the press member can be configured to move at least one of the first and second pressing layers towards other one of the first and second pressing layers during the operation of the pressing mechanism. In some examples of the presently disclosed subject matter, the biasing mechanism can be configured to
30 continuously bias at least one of the first and second pressing layers towards other one of the first and second pressing layers at least during operation of the mold. In some examples of the presently disclosed subject matter, the biasing mechanism can comprise

a spring operatively coupled to the at least one of the first and second pressing layers. In some examples of the presently disclosed subject matter, the press member can be configured to load the biasing mechanism in a direction opposite the direction in which the biasing mechanism biases the pressing mechanism.

5 In some examples of the presently disclosed subject matter, the biasing mechanism can be configured to continuously bias the pressing mechanism in a direction to pressurize the mixture to compensate for a pressure drop in the mixture due to extraction of the liquid from the mixture during its processing. It is to be understood herein that the term continuously when used in the context of biasing by the biasing
10 mechanism intends to mean that the biasing mechanism is configured to bias the pressing mechanism even in between the pressing cycles of the press member. For instance, the press member operates the pressing mechanism in discrete forces for pressing the pressing mechanism. The biasing mechanism continues the bias on the pressing mechanism even between these discrete forces. The press member operates the pressing mechanism with
15 a first pulse of force to press the mixture to a first extent. The mixture loses liquid content as a result of the pressing to the first extent. This loss of liquid causes a pressure drop in the mixture and thus the force experienced by the mixture due to the press member (the press member force) gradually reduces despite of the fact that the press member is at its position of applying the first pulse of force. The biasing mechanism continuously biases
20 the pressing mechanism in the direction of pressing the mixture. The biasing force applied by the biasing mechanism compensates for the reduction of the press-member force experienced by the mixture and consequently compensates for the drop in pressure within the mixture due to liquid loss.

 In some examples of the presently disclosed subject matter, the biasing
25 mechanism biases the first pressing layer in a first direction, for example, upwards. The direction of bias of the biasing mechanism has to be such that the bias effects the pressing of the mixture within the pressing mechanism. The press member when moves the second pressing layer to press on to the mixture to apply the first discrete pulse of force, the second pressing layer moves downwards to a first extent and a first distance is achieved
30 between the first pressing layer and the second pressing layer. The press member subsequently stops moving the second pressing layer downwards and this completes the first pulse of force, and the force applied by the press member remains constant thereafter

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until a next pulse of force applied by the press member after a time interval T1. Simultaneously, the above-mentioned action of the press member causes the first pressing layer to move downwards such that the biasing mechanism is loaded in a second direction opposite the first direction. This loading of the biasing mechanism causes the biasing
5 mechanism to bias the first pressing layer upwards, i.e., in the first direction.

During the time interval T1, the mixture loses liquid (at least as a result of the pressing) and thereby experiences a loss in volume and consequently a loss in pressure within the mixture. This loss in pressure is compensated by the biasing mechanism continuously (i.e., in between the discrete pulses of force applied by the press member)
10 biasing the first pressing layer upwards to reduce the distance between the pressing layers. This action of the biasing mechanism also results in a final product/article with uniform physical properties, especially density, throughout.

After time interval T1, the press member again moves the second pressing layer downwards repeating the whole sequence, i.e., applying a second pulse of force on the
15 mixture and loading the biasing mechanism again in the downwards direction. During a time interval T2 until a third pulse of force, the whole sequence repeats, i.e., loss in pressure in the mixture and the pressure being compensated by the biasing mechanism by continuously biasing the first pressing layer upwards. These sequences are repeated until the final product/article is obtained.

20 In some examples, the biasing mechanism can be coupled to the second pressing layer, or the mold can be used in the inverted manner, and in such examples, the first direction can be downwards.

It is to be understood herein that a biasing mechanism in addition to the press member results in continuous bias of the pressing mechanism in a direction towards
25 pressing the mixture, and thus at no time the mixture is allowed to experience a drop in pressure that could result in non-uniformity in the physical/mechanical properties of the resulting product/article (e.g. the flexural strength, compressive strength, plastic/elastic properties as defined herein). During the above-described controlled liquid removal, particles are compressed and densified one against the other, and inner voids, if exist,
30 become smaller and fewer, resulting not only in an increase in strength but also a uniform strength throughout the resulting article. Moreover, without being bound by theory,

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uniform densification increases reinforcing efficiency of the fibers uniformly throughout the resulting article because the particulates increase their interlock on the fibers, and thereby providing increased tensile/flexural strength. Accordingly, it has unexpectedly been found that when subjecting the material for forming the articles to compression,
5 under controlled liquid removal conditions, it is possible to significantly and unexpectedly improve the physical/mechanical properties of the resulting product/article (e.g. the flexural strength, compressive strength, plastic/elastic properties as defined herein).

In some examples of the presently disclosed subject matter there is provided also
10 an article of manufacture comprising a compressed mixture, the article of manufacture obtained or obtainable by molding a mixture comprising particulates comprising a blend of (i) clay material comprising at least one clay mineral having a plasticity index above about 20% and liquid limit above about 40%, as determined according to ASTM4318 and (ii) water insoluble inorganic material and fibrous material, said molding being
15 characterized by using a mold that is equipped with (i) a pressing mechanism configured to receive the mixture therewithin and press the mixture to extract the liquid therefrom; (ii) a press member operatively coupled to the pressing mechanism and configured to operate the pressing mechanism for pressing the mixture; and (iii) a biasing mechanism configured to continuously bias the pressing mechanism in a direction to pressurize the
20 mixture and compensate for a pressure drop in the mixture due to said extraction of the liquid.

Other methods for controlled water removal can include, without being limited thereto, the following molding tools,

- A molding tool comprising two punches including an upper punch and a
25 lower punch, each having a respective molding surface shaped to substantially define an external surface of the molded product. Each of the punches includes at least two layers, a first layer made of metal, such as aluminum, aluminum alloys, tool steel, or the like, making it possible to heat the mold and conduct the heat to all parts of the mold and to be a backing plate that transfer the loads from the press
30 to the exact geometry of the article, and a second layer formed of one or more porous materials, either porous metal plates that accumulate passively the liquid and can act as intermediate part from which the liquid is vacuum sucked out, or

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fibrous materials that can be natural and/or synthetic and can include felt and / or other non-woven fibrous materials that actively draw out liquid and conduct it out of the mold. The tool includes an actuator assembly including a piston cylinder arrangement to displace the upper punch with respect to the lower punch. The tool
5 operates on the wet mixture to reduce its liquid content that increases the strength of bonds between the particulate matter. Fluid extraction paths extend through both upper and lower punches through which fluid from within the wet mixture is transported away from the respective molding surface. The tool further has a fluid extraction system including a suction subsystem and a heating subsystem
10 configured to extract the liquid content from the wet mixture.

In operation, the punches are spaced apart from one another and the wet mixture is positioned between the mold surfaces. The actuator assembly is operated to displace the upper punch to compress the wet mixture between the molding surfaces. Concurrently, the suction subsystem is operated to induce a
15 negative pressure in the fluid extraction paths. The processing steps involve reducing the liquid fraction in the wet mixture. Such a tool and its operation are described also in WO2021097525, the content of which is incorporated herein by reference.

- A molding tool that includes two pressure plates connected to and operable
20 by a hydraulic pressure device via respective sliding piston rods. Two shaped bodies are arranged between the two pressure plates, each of the shaped bodies associated with a respective one of the pressure plates. The shaped bodies enclose between them a cavity in which a wet mixture according to the present disclosure is to be placed for processing in the casting mold. The shaped bodies include
25 respective bores for supplying said mixture into the cavity and/or for discharging water from the cavity. Each of the shaped body can have a water absorption layer on its side facing the cavity, and a load bearing layer on the side facing away from the cavity. The water absorption layers extract the water from the wet mixture located in the cavity and discharge it through a drainage network formed in either
30 or both water absorption and load bearing layers. Such a tool and its operation are described also in EP0234360, the content of which is incorporated herein by reference.

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- A molding tool comprising a fluid sealable cavity configured for receiving and holding the wet mixture and positioned between a bottom mold portion and a top mold portion moveable with respect to each other by a press member. The molding tool further comprises at least one active fluid duct extending through a circumferentially extending wall of the mold and comprising an inlet port and an outlet port. The fluid duct extends through the mold such that the inlet port is in fluid communication with the fluid sealable cavity and is configured for liquid extraction from the fluid sealable cavity when the mold is in operation. The compression pressure applied by the mold portions can be increased and/or decreased during the molding process depending on the change in volume of the total wet mixture due to liquid extraction. Such a tool and its operation are described also in US11,192,278, the content of which is incorporated herein by reference. Preferred molding tools in accordance with the presently disclosed subject matter comprise the molds as described in co-pending US patent application No. 63/375,360 filed on September 12, 2022, the content of which is incorporated herein, in its entirety, by reference.

The process of the present disclosure is not limited, however, to the aforementioned types of molds and molding setups and any other mold can be used, without departing from the scope of sought protection as defined by the claims, as long as the molding tool provides a controlled liquid removal from the wet mixture.

When referring to "*controlled liquid removal*" it is to be understood that at least one of the following operational features are applied:

In some examples of the presently disclosed subject matter, the controlled liquid removal is obtained by applying negative air pressure (i.e. vacuum) within the molding cavity, during at least portions of molding duration. When referring to negative pressure it means a negative gauge pressure within the mold.

In some examples of the presently disclosed subject matter, the controlled liquid removal is obtained by using a mold including a porous plate such that the liquid can be removed is extracted via the pores of the porous plate. Preferably, the extraction of the liquid via the pores is aided by applying the negative air pressure.

In some examples of the presently disclosed subject matter, the controlled liquid removal is obtained using a moisture wicking fabric. As appreciated by those versed in textile, moisture-wicking fabrics are fabrics with the ability to pull moisture away (in this case, from the wet particulate mass) using tiny, built-in capillaries. Moisture is drawn into the fabric, which makes it easier to be translocated from the mold. A non-limiting list of possible moisture wicking fabrics include wool, polyamide, polyester, polypropylene, Gore-Tex, Nylon, bamboo, Modal, acrylic, Rayon and Spandex. In some examples of the present disclosed method, the moisture wicking fabric is placed within the mold, and the wet mixture is introduced at least onto the moisture wicking fabric. Moisture wicking fabrics are known in the art and typically are fabrics with the ability to pull moisture away via capillary action.

Processing the wet mixture in accordance with some examples of the disclosed subject matter comprises heating the mold to a temperature of at least about 30°C. In some examples, the heating of the mold is prior to placing the wet mixture.

In some examples of the presently disclosed method, the mold is heated to a temperature of between about 30°C and about 140°C; at times, between about 60°C and about 90°C; at times, between about 85°C and about 105°C; at times, between about 100°C and about 120°C; at times, between about 115°C and about 135°C; at times, between about 130°C and about 140°C.

Processing the wet mixture in accordance with some examples of the disclosed subject matter further comprises applying a compression pressure to the mold. In some examples, the compression is controlled so as to maintain a constant pressure applied onto the wet mixture, for example as described herein above.

In some examples of the presently disclosed method, the pressure is controlled to maintain a constant pressure of at least about 10Bar.

In some examples of the presently disclosed method, the compression is controlled to be at range of between about 10Bar and about 300Bar; at times, between about 40Bar and about 80Bar; at times, between about 70Bar and about 120Bar; at times, between about 100Bar and about 140Bar; at times, between about 130Bar and about 170Bar; at times, between about 160Bar and about 200Bar.

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In accordance with some examples of the presently disclosed subject matter, following the compression molding process the compressed mixture article of the manufacture is removed from the mold and further dried. In some examples, the drying is to a moisture content that is between about 0.1wt% and about 1wt% out of the total
5 weight of the article of manufacture; at times, between about 0.1% and about 0.4%; at times, between about 0.3% and about 0.6%; at times, between about 0.5% and about 0.8%; at times, between about 0.7% and about 1%.

In accordance with some examples of the presently disclosed method, the drying of the extracted article comprises any one or combination of air drying, oven drying,
10 controlled humidity oven and dry air blowing.

In some examples, the drying of the extracted article in a drying chamber with low humidity of, for example, 20% relative humidity (RH) and temperature of 40°C-70°C until the moisture content is of less than 1%wt (determined, for example, based on the weight difference of the article before pressing and after being placed in the drying
15 chamber).

Without being bound thereto, it is believed that the efficiency of water removal determines the compression duration and positively affects the final physical properties of the product. In principle, it has been found herein that it is desired to remove a high amount of liquid (one of two criteria: 1) a removal of at least 7% of liquid out of a total
20 weight of the wet mixture, more preferable reaching a removal of at least 10% of liquid out of a total weight of the wet mixture, or 2) removal of liquid until reaching a liquid content leftover after the pressing of less than 15%, more preferable reaching a liquid content leftover after the pressing of less than 10%)., in a short compression duration (i.e. less than 12 minutes, preferably less than 4 minutes), so as to obtain better mechanical
25 properties. In other words, it has been found herein that faster water removal allows a shorter compression period and a better product in terms of mechanical properties of the resulting article of manufacture.

In some examples, the water content can be determined by the Pfefferkorn plasticity test. Without being limited thereto, it has been found for tiles formation an
30 advantageous physical property, as disclosed herein, are obtained if the wet mixture

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before compression had Pfefferkorn value of 8 to 1.15. In this connection, it is to be understood that a Pfefferkorn value of 1 means that the mixture has zero plasticity.

In some examples of the presently disclosed method, the controlled compression is obtained using a uniaxial compression machine (nominal force >60ton, slider stroke
5 >300mm, bed dimension min 40/40cm) under the following conditions:

- **Compression force:** min 60ton, compression pressure: of at least 10bar or of between 10bar and 300 bar.
- **Holding time:** 2-15 minutes
- **Water removal:** assisted by a combination of active water removal methods
10 such as described in Example 5c below or any other active water removal, including for example, apply any one or combination of porous plate with pressure and suction, porous plate without suction, suction with capillary medium and the like, as opposed to passive water removal using capillary medium.
- **Controlled compression:** achieved by maintaining a constant compression
15 force during water removal (notwithstanding the pressure loss due to the water removal) as described in Example 5c below.

In accordance with some examples of the presently disclosed method, the article of manufacture extracted from the mold and optionally, or preferably further dried to a
20 moisture content of less than about 1wt%, is subjected to further processing steps.

In some examples, the extracted article is subjected to a step of coating.

Coating can be achieved by any one or combination of spraying, spreading, brushing, dipping of the article in a coating agent.

Coating agents natural coating agents or synthetic coating agents.

25 In some examples of the presently disclosed subject matter, the coating agent is a natural agent.

In some examples of the presently disclosed method, the coating agent is a plant extract, this including, for example, plant extract oil or plant extract adhesive.

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In some examples of the presently disclosed method, the coating agent is a plant extracted drying oil (such as those known in the art of painting). A non-limiting list of possible drying oils include any one of walnut, tung, linseed, poppyseed, perilla oils and mixtures thereof.

- 5 In some examples of the presently disclosed method, the coating agent is a natural extract resin or plant derived resin mixture.

The article can be coated using combination of coating agents as described herein.

- 10 The presently disclosed invention will now be exemplified in the following description of experiments that were carried out in accordance with the invention. It is to be understood that these examples are intended to be in the nature of illustration rather than of limitation. Obviously, many modifications and variations of these examples are possible in light of the above teaching. It is therefore, to be understood that within the scope of the appended claims, the invention may be practiced otherwise, in a myriad of possible ways, then as specifically described hereinbelow.

15 **NON-LIMITING EXAMPLES**

Example 1: Preparing particulate mixture

Clay material – Clay material was prepared from mixing bentonite and ball clay at a ratio of 1:2.4. The bentonite and ball clay were obtained from a commercial supplier.

- 20 The bentonite and ball clay were each dried and downsized by milling until reaching D50<7 microns. Both clay materials (bentonite and ball clay) are considered non-organic high plasticity clays (according to Israeli Standard 253, paragraphs 103.3.2, 103.4, 103.8-103.10, 206.1, 206.1.2, 206.2.2 and Table 6 therein, and plasticity test according to ASTM D 4318). The ball clay was selected to have a minimum index plasticity above 30% and liquid limit above 50%, The Bentonite was selected to have liquid limit above 400%.

Water insoluble inorganic material – water insoluble inorganic material included any one of

Inorganic material 1: limestone powder

Inorganic material 2: limestone and basalt mixture at a weight ratio of about 2.2:1.

Inorganic material 3: limestone and quarts mixture at a weight ratio of about 0.75:1.

5 The insoluble inorganic material was downsized by milling and separated by sieving to different fractions distinguished by their D50 according to Table 1:

Table 1: Inorganic material composition

Fraction No.	D50 (micron)	Amount (%)
Fraction 1	9	15.70
Fraction 2	76	19.89
Fraction 3	280	16.75
Fraction 4	640	25.134
Total inorganic fractions		77.47

10 The clay material, each of the water insoluble inorganic material and Flax fibers (*fibrous material*, fiber length of 14mm) were then combined into different dry compositions (1 to 3), with the ratio of the different components as provided in **Table 2**. The amounts of the clay material and water insoluble inorganic material were selected to provide one cumulative distribution that behaves within an upper and lower limit (derived

15 from the mathematical equation of $P(D) = \left(\frac{D}{D_{max}}\right)^q$, in order to reach a desired shape parameter (q) within the range of 0.24 and 0.33.

Figure 1 provides a scatter plot representing cumulative particle size distribution with an optimal shape parameter, with a target shape factor range of 0.24-0.33 being presented by the solid lines, and the optimal distribution being illustrated by the dashed line.

20 **Figure 2** provides a scatter plot representing cumulative particle size distribution in an insoluble particulate material mixture (based on Table 2) with an optimal shape

parameter based on fitting to the optimal distribution curve, with a target shape factor range of 0.24-0.33 being represented by the solid lines, and the optimal distribution being illustrated a dashed line, and particle size distribution of the experimental mixture being represented by a doubled line.

5 **Figure 3** provides a scatter plot representing cumulative particle size distribution in an insoluble particulate material mixture (Table 5) with out-of-range shape parameter based on fitting to the optimal distribution curve, with a target shape factor range of 0.24-0.33 being represented by the solid lines, the optimal distribution being illustrated by the dashed line and particle size distribution of the experimental mixture being represented by a doubled line.

The immediate conclusion from Figures 1-3 is that it is possible to design mixtures that would fall within the desired particle size distribution and use the equation in order to determine which mixture is suitable and which is not.

Table 2: Dry Compositions (weight %)

Composition No.	1	2	3
Clay material	20.17%	20.51%	26.12%
Inorganic Material No. 1	77.47%		
Inorganic Material No. 2		77.29% (51.28% basalt + 26.01% limestone)	
Inorganic Material No. 3			71.37% (43.23% quartz + 28.14% limestone)
Flax fibers	2.36%	2.20%	2.51%
Shape parameter	0.28	0.33	0.33

15 **Example 2: Preparing wet mixture suitable for molding**

Prior to mixing the dry components of Table 2 with KCl solution, the insoluble inorganic materials (either 1, 2, or 3) were air dried for at least 48 hours, until the weight

change within an interval of 24 hours was less than 1%wt. Each of the dried insoluble inorganic material was mixed with the flax fibers under shear forces to cause opening of the fibers during mixing (the conditions included 15 minutes in a rotation mixer (15rpm) or 2 minutes in high-speed shear mixer (1500rpm)).

5 The dried components, including the clay material, were then poured into a different planetary mixer in which it was mixed with KCl solution in water, at room temperature for about 20-30 minutes, as detailed in **Table 3**.

 The amount of KCl solution was selected to provide a final concentration of KCl of 0.0115wt% out of the total weight of the dry components or to provide an amount of
10 KCl of 0.0098wt% out of the total weight of the wet composition.

 In practice, the amount of KCl was determined to be sufficient to at least saturate the clay material and in this particular example provided herein, the amount of KCl solution was 17.75%wt out of the total dry material which is equivalent to 15.07wt% out of the total weight of the wet composition.

15 **Table 3: Dry/Wet mixture composition**

Component	%Weight in dry composition	%Weight in wet mixture
Insoluble inorganic	77.44%	65.82%
Clay material	20.2%	17.1%
Flax fibers with length of 14mm	2.36%	2%
KCl		0.0098%wt.
Water		15.07%
Total	100%	100%

 The wet composition was then placed within a mold having dedicated openings for liquid release during controlled compression and being pre-heated to a temperature of 97°C. The controlled compression was achieved by controlled compensation of pressure
20 drop as a result of liquid removal, as described below. The water content was determined by the Pfefferkorn plasticity test.

 The Pfefferkorn test determines the amount of water required to achieve a 30% contraction in relation to the initial height of a test body under the action of a standard

mass (Pfefferkorn, Ein Beitrag zur Bestimmung der Plastizität in Tonen und Kaolinen Sprechsaal, 57 (25) (1924), pp. 297-299). The results express height reduction as a function of moisture content.

Measuring of plasticity according to Pfefferkorn test is based on the principle of impact deformation. A sample with a diameter of 33 mm and an initial height (h_0) of 40 mm, is deformed by a free-falling plate with a mass of 1.192 kg. The initial height is related to the impact deformation height, the result of which is the ratio of deformation. The ratios of deformation or the impact deformation heights (h_0 , initial height; h_f , final height) are plotted against the moisture content. The steeper the curve, the “shorter” the body, i.e. the more intensely the body will react to variations of the moisture content.

In the current experiment, the best compressed article was achieved if the wet mixture before compression had Pfefferkorn value of 1.54 up to 1.91. This defines the preferred range of preferred water content needed in order to obtain the desired mixture plasticity for the production of our articles.

15 **Example 3: Product preparation**

The wet mixture within a pre-heated mold was subjected to controlled compression molding using uniaxial compression machine (nominal force 60ton, slider stroke >300mm, bed dimension min 40/40cm) under the following conditions:

- **Compression pressure:** 120bar
- 20 - **Holding time:** 2-8 minutes
- **Water removal:** assisted by a combination of vacuum and porous plates as described in Example 6 below.
- **Controlled compression:** was achieved by maintaining a constant compression force during water removal (notwithstanding the pressure loss
- 25 due to the water removal) as described in Example 6 below.

During compression, fluid was extracted from the mixture via the pores of the porous plate and assisted by applying vacuum on the mold. The water removed allowed the wet composition to transform from a "plastic" fluid into a stiff solid material.

The efficiency of water removal determined the compression duration and the final properties of the product. In principle, it has been found herein that it is desired to remove a high amount of liquid so as to obtain better mechanical properties. It is preferred to perform short compression durations with a low initial water content, resulting in faster water removal. It has been found that the mold structural features allowed the improved water extraction. The minimum water extraction during the pressing was 7% from the wet mix (as determined by weighting the extracted article immediately at the end of the compression, by no more than 20 sec until weighing).

While still hot and steamy, the compressed product was extracted from the mold and further dried to a target moisture content of 0.5-1%. The further drying was done by placing the mold into a drying chamber with low humidity of 20%RH and temperature of 40°C for at least 30minutes until a moisture content of less than 1%wt was measured (based on weight differences before pressing and after drying).

The dried product was then subjected to further post curing processes including spray coating with a coating agent.

The coating is preferably to obtain a continuous coating layer, and the coating agent penetrates to a minimum depth of 0.5 mm below the surface of the dried product. This was determined based on color change examination of a cross sectional cut of the coated dried product.

Example 4: Characterization

The resulting coated products of Example 3 were characterized for their physical properties as follows:

Flexural strength as determined by a three points bending according to ASTM C 99, or by a four points bending as described in <https://www.compositesworld.com/articles/notched-testing-of-sandwich-composites-the-sandwich-open-hole-flexure-test>, with ½ load span, using the following parameters:

$$\text{Span}=0.7*L,$$

$$\text{Speed control}=1\text{MPa/sec.}$$

Density as determined by weight per volume of the final product.

The resulting products prepared based on the dry compositions of **Table 2** exhibited the flexural strength and densities, as summarized in **Table 4**.

Table 4: Physical Properties of the resulting products

Composition No.	Flexural Strength (MPa)	Density
1	9.1	1.96
2	8.3	2.00
3	10.85	2.15

5 All the samples tested shows a flexural strength of more than 7MPa. The different products also all contained an amount of potassium that is essentially the same as originally introduced into the wet composition. The presence of potassium ions in the wet composition contributed to the flexural strength of the dried product, as also exemplified hereinbelow. Without being bound by theory, it is assumed that the majority (if not
10 essentially all) the potassium externally added to the wet composition is entrapped by clay material and is not removed during liquid extraction. In principle, this can be determined by washing the final product with ion exchange buffer and measuring potassium content in the eluent.

Example 5: Comparative Example

15 Example 5A – Effect of shape parameter

In order to create the wet mixture, the different materials from **Table 3** were combined. Their amounts were chosen based on the outcome graph distribution achieved for different combinations and the identifying the compositions that provide the optimal accumulative particle size distribution.

20 **Figure 4** provides the cumulative particle size distribution of the example mixture of Table 3 with its corresponding upper and lower limits.

For comparison, a first dry mixture of the insoluble inorganic materials was constructed to fit a curve with a total particle size distribution corresponding to a 0.29 shape parameter (**Figure 2**), a second dry mixture of the insoluble inorganic materials

was constructed to fit a curve with a total particle size distribution corresponding to a 0.41 shape parameter, which is outside the range of the current disclosure.

Table 5 provides the flexural strength (according to ASTM C99) of products produced from in range distribution compared to out range distribution.

5 **Table 5: Example of two mixtures that were tested during the search for optimal shape parameter (n)**

	Out of range distribution (n=0.41)	On median distribution (n=0.29)
Flexural strength [Mpa]	6.3	8.61 (36% improvement)

Table 5 shows that when using a mixture defined by a shape parameter within the range of the presently disclosed subject matter (in Table 5, on the median distribution), the resulting product exhibits a significantly higher flexural strength.

10 Example 5B – Effect of externally added potassium

In addition, the effect of externally added potassium content vs. no externally added potassium, was determined.

Table 6 shows the flexural strength and density of a tile prepared from Composition No. 1, in comparison with a composition including the same components,
15 however without externally added potassium chloride. The results show that the addition of potassium ions improved the flexural strength of the tile:

Table 6: Potassium Content Effect on the resulted flexural strength

No.	Composition (content)	Percentage of Externally added KCl	Flexural strength (Mpa)	Density (gr/cm ³)
1	Composition 1 with slightly different shape parameter	0.0098	7.05	2.1
2	Composition 1 with slightly different shape parameter	NONE	6.16	2.1

Table 6 clearly shows that the presence of potassium in the composition contributes to the flexural strength of the product, while having no effect on the density of the product.

Example 5C – Effect of active water removal

5 In a further experiment, the effect of amount of liquid removed during the compression process was determined, the amount of liquid being removed either using 'passive liquid removal' with only permeable fabrics through which the liquid can diffuse out (**control product**) or using active liquid removal where, in addition to the permeable fabric, vacuum and a porous plate were used to extract the liquid (**test product**).

10 In both cases, compression was for 3 minutes (to allow the desired amount of liquid to be removed).

 It has been found that when no active liquid removal is applied, the resulting product (the control product) was too weak to be extracted from the mold and tended to break during attempts to remove from the mold. While for the active liquid removal the
15 3 minutes gave an article equal in properties to the one stated in US 11,192,278. Therefore, a further comparison was done on a control product that was pressed for 8 minutes (instead of 3 minutes) to obtain adequate liquid extraction. in addition, a further comparison was done on a product that been through active liquid removal for 5 minutes, to achieve the high strength that this innovation can render.

20 **Table 6** provides the flexural strength of the control and test products, and shows that when applying active liquid removal, there is 67% improvement in strength obtained as compared to the control product.

Table 6: Effect of Passive vs. Active Liquid Removal

Property	Passive liquid removal (8 min)	Active liquid removal (5 min) (Figure 5)
Amount (%) of liquid removed	7.8	10.0
Flexural strength [Mpa] of a 22.3/22.3/1 cm tile	4.43	7.4

Table 6 shows that already after 5 minutes of active water removal the flexural strength of the sample increased significantly as compared to the sample obtained with passive water removal.

The comparative results show there is a significant improvement in the physical properties of the products of the presently disclosed subject matter when at least one of the following parameters need to be applied:

- Shape parameter of the water insoluble inorganic material between 0.24 and 0.33
- External addition of potassium into the wet mixture of clay material and inorganic natural material;
- Active liquid removal (in this specific example, the combination of pressure and using a porous plate – as shown in Figure 5, further discussed below).

In addition, the effect of active removal on the flexural strength (according to ASTM C99), compression strength (according to ASTM C170 or EN772-1) elastic modulus (according to ASTM C99) and on the ratio between Ultimate Flexural Strength (UFS) strain to yield point strain together with the ratio between UFS toughness to yield point toughness were determined.

Figure 5 shows the flexural strength as a function of different pressing pressures and different pressing duration, both influence the water extraction efficiency and its amount. Specifically, Figure 5 shows that it is possible, when using a porous plate and different pressures, to reach flexural strengths above 7MPa. The increase in flexural strength can also be further improved or alternatively be achieved when removing more than 7% water during the process.

Example 6: Elastic/Plastic Properties

To further assess the beneficial physical properties of the presently disclosed articles of manufacture, the elastic and plastic properties were compared to articles obtained by passive water removal as described in US 11,192,278.

Specifically, an article prepared from the composition of Table 3, and based on the procedure of Example 3 (referred to in Table 7 by the short term "Article") was

compared with an article prepared as described in US 11,192,278 (referred to in Table 7 as the "Reference").

Table 7 provides the comparison of the different properties of the Article and the Reference.

Property	Article	Reference
Stress YP [MPa]	9.11	4.00
Stiffness [Gpa]	6.13	2.93
Strain YP [m/m]	0.0015	0.0014
Modulus of resilience (elastic region toughness) [J/m ³]	0.0068	0.0028
Toughness at UFS [J/m ³]	0.6903	1.26
Stress at UFS [MPa]	9.61	5.35
Strain at UFS [m/m]	0.0023	0.0053

5

Table 7 shows that the article according to the presently disclosed subject matter has higher strength compared to an article as disclosed in US 11,192,278, (e.g. 9.61 vs. 5.35 MPa), together with higher stiffness (elastic modulus) (e.g. 6.13 vs. 2.93 GPa). Further, the article according to the presently disclosed subject matter has a decrease of UFS/YP strain (e.g. 1.53 vs. 3.79) and a decrease of UFS/YP toughness (e.g. 101 vs. 450).

10

Notably, the toughness at UFS and strain at UFS are in normal range values.

To conclude, the better compaction and reinforcement efficiency of the article according to the presently disclosed subject matter result with increase of the article strength and stiffness, making it less ductile.

15

Further, **Figure 6** shows toughness up to yield point (elastic region (light grey) under the stress-strain curve, also called modulus of resilience), with toughness up to UFS being defined by the sum of the elastic region and plastic region areas under the stress-strain curve. Specifically, Figure 6 shows that the sample had an elastic region slop of

6.8GPa (= Stress YP [9.11] / Strain YP [0.0015]), UFS strain to yield point strain of 2.02 (= Strain at UFS [0.0023] / Strain YP [0.00153]), UFS toughness to yield point toughness of 157.95 (= Toughness at UFS [0.6903] / Modulus of resilience [0.0068]). This elastic behaviour correlates with the presently disclosed subject matter defined properties.

5 **Example 7: Compressive Strength**

Compressive Strength was determined on a full block (75/75/75mm, ASTM C170) with the composition of Table 3, prepared according to Example 4.

10 **Figure 7A** shows that the sample had a compressive strength with an ultimate strength at 97MPa which is the point at which the sample withstands the highest stress before failing.

A similar test was conducted on a hollowed block (**Figure 7B**) which are generally subjected to more eccentric load and tend to fail more by shear or buckling rather than by pure compression. Thus, the compressive strength is reduced significantly to about 6MPa without a coating but can be further improved by a coating to achieve 14MPa, or more.

CLAIMS:

1. An article of manufacture comprising a compressed mixture of

(a) particulates comprising a blend of (i) clay material comprising at least one clay mineral having a plasticity index above about 20% and liquid limit above about 40%, as determined according to ASTM4318 and (ii) water insoluble inorganic material and

(b) fibrous material;

wherein a sample of said article of manufacture is characterized by at least one of

- a shape parameter of said water insoluble inorganic material within a range of 0.24 and 0.33;
- flexural strength of at least 7MPa when measured according to ASTM C99;
- compressive strength of at least 30MPa when measured according to ASTM C170 on a cubic sample of said article of manufacture;
- compressive strength of at least 3 MPa when measured according to EN 772-1 for air-dried specimens, on said sample having a shape of a hollowed masonry block;
- elastic modulus of between 2.5GPa and 15GPa when measured according to ASTM C99;
- a ratio between Ultimate Tensile Strength (UFS) strain to yield point strain of at least 1.2 and a ratio between UFS toughness to yield point toughness of at least 2; and
- inorganic cation content of at least 0.00034wt%.

2. The article of manufacture of claim 1, comprising an oxide selected from the group consisting of silica, alumina, magnesia and combination of same.

3. The article of manufacture of claim 1 or 2, comprising aluminum phyllosilicate.

4. The article of manufacture of any one of claims 1 to 3, wherein said clay material constitutes at least 10%v out of a total volume of said article of manufacture as determined

by Scanning Electron Microscopy (SEM) equipped with an Energy Dispersive Spectroscopy (EDS) detector.

5. The article of manufacture of claim 4, wherein said clay material constitutes between about 10%v and about 50%v out of a total volume of said article of manufacture.

6. The article of manufacture of any one of claims 1 to 5, wherein said water insoluble inorganic material comprises a natural inorganic material.

7. The article of manufacture of claim 6, wherein said natural inorganic material is selected from the group consisting of igneous rocks, sedimentary rocks and metamorphic rock and any combination of same.

8. The article of manufacture of claim 6 or 9, wherein said natural inorganic material is selected from the group consisting of quartz, limestone, dolomite, chalk, sandstone, shale, pumice, obsidian, rhyolite, scoria, dacite, granite, gabbro, diabase, diorite, pegmatite, peridotite, basalt, marble, quartzite and any combination of same.

9. The article of manufacture of any one of claims 1 to 8, wherein said insoluble inorganic material comprises recycled matter comprising water insoluble inorganic material.

10. The article of manufacture of any one of claims 1 to 9, comprising said water insoluble inorganic material or recycled matter comprising said water insoluble inorganic material, the water insoluble inorganic material being in an amount that constitutes at least 50%v out of a total volume of said article of manufacture as determined using SEM equipped with an Energy Dispersive Spectroscopy (EDS) detector.

11. The article of manufacture of any one of claims 1 to 10, wherein said insoluble inorganic material constitutes between about 50%v and about 90%v out of a total volume of said article of manufacture.

12. The article of manufacture of any one of claims 1 to 11, wherein said fibrous material comprises natural fibers.

13. The article of manufacture of claim 12, wherein said natural fibers comprise plant fibers.

14. The article of manufacture of any one of claim 12 or 13, wherein said fibrous material comprises at least one of cellulose, hemicellulose, lignocellulose fibers.

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15. The article of manufacture of any one of claims 1 to 14, wherein said fibrous material comprises fibers selected from the group consisting of flax, sisal, hemp, jute, cotton, abaca, bamboo, ramie, banana, and kenaf and combinations of same.
16. The article of manufacture of any one of claims 1 to 15, wherein said fibrous material comprises flax fibers.
17. The article of manufacture of any one of claims 1 to 14, wherein said fibrous material comprises basalt fibers.
18. The article of manufacture of any one of claims 1 to 14, wherein said fibrous material comprises synthetic fibers.
19. The article of manufacture of any one of claims 1 to 18, wherein said fibrous material has a longitudinal dimension of at least 2mm.
20. The article of manufacture of claim 19, wherein the fibrous material has a longitudinal dimension of between 2mm and 30mm.
21. The article of manufacture of any one of claims 1 to 20, wherein said fibrous material has a tensile strength of at least 400MPa.
22. The article of manufacture of any one of claims 1 to 21, wherein said fibrous material is in an amount of minimum 1% out of a total volume of the article of manufacture.
23. The article of manufacture of claim 26, wherein said fibrous material is in an amount of between about 1% and 20% out of a total volume of the article of manufacture.
24. The article of manufacture of any one of claims 1 to 23, wherein said water insoluble inorganic material therein has a shape parameter within a range of 0.24 and 0.33.
25. The article of manufacture of any one of claims 1 to 24, having a flexural strength of at least 7MPa.
26. The article of manufacture of any one of claims 1 to 25, having a compressive strength of at least 30MPa when measured on a cubic sample of said article of manufacture.

- 27.** The article of manufacture of any one of claims 1 to 26, having a compressive strength of at least 3 MPa when measured on a sample of said article having a shape of a hollowed masonry block.
- 28.** The article of manufacture of any one of claims 1 to 27, having an elastic modulus of between 2.5GPa and 15GPa.
- 29.** The article of manufacture of any one of claims 1 to 28, having a ratio between Ultimate Tensile Strength (UFS) strain to yield point strain of at least 1.2 and a ratio between UFS toughness to yield point toughness of at least 2.
- 30.** The article of manufacture of any one of claims 1 to 29, wherein said inorganic cation is selected from the group consisting of potassium, ammonium, sodium, calcium, magnesium, lithium, aluminum.
- 31.** The article of manufacture of claim 30, wherein said inorganic cation is potassium.
- 32.** The article of manufacture of any one of claims 1 to 31, having a potassium content of at least 0.00034wt%.
- 33.** The article of manufacture of any one of claims 1 to 32, having a shape suitable for use as brick, tile, block, lining, partitioning element, furniture or furniture element, roofing tile, cladding, facade, panels, boards, insulative products, interior design elements, small containers, under floor element, paving brick and element, or wall brick.
- 34.** A method for obtaining an article of manufacture, the method comprising:
- placing within a mold a wet mixture comprising
 - (a)** particulates comprising a blend of (i) clay material comprising at least one clay mineral having a plasticity index above 20% and liquid limit above 40% as determined according to ASTM4318 and (ii) water insoluble inorganic material;
 - (b)** fibrous material, and
 - (c)** a liquid comprising inorganic cations, in water; the amount of said liquid being between about 5wt% and 50wt% out of the total weight of particulates and fibrous material;

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the liquid being added in an amount to provide said wet mixture with a plasticity of between 8 to 1.15 according to pfefferkorn test;

- subjecting the wet mixture to compression molding under conditions that allow controlled liquid removal out of the wet mixture during application of compression forces onto said wet mixture until reaching at least one of
 - removal of at least 7% of liquid out of a total weight of the wet mixture prior to said compression; and
 - removal of liquid out of a total weight of the wet mixture prior to said compression, until reaching a liquid content in said compressed mixture of less than 15%; and
- removing the compressed mixture from the mold to obtain the article of manufacture.

35. The method of claim 34, wherein said clay material is as defined in any one of claims 2 to 5 and 24, said water insoluble inorganic material is as defined in any one of claims 6 to 11, and said fibrous material is as defined in any one of claims 12 to 23 .

36. The method of claim 34 or 35, comprising heating the mold to a temperature of at least about 30°C prior to placing said wet mixture.

37. The method of claim 36, wherein said heating of the mold is to a temperature of between about 30°C and about 140°C prior to placing said wet mixture.

38. The method of any one of claims 34 to 37, comprising placing the wet mixture in a mold comprising a porous plate.

39. The method of any one of claims 34 to 38, comprising placing the wet mixture in a mold comprising a moisture wicking fabric.

40. The method of any one of claims 34 to 39, comprising applying negative air pressure during at least portions of said compression duration.

41. The method of any one of claims 34 to 40, wherein said compression is at a pressure of at least 10 Bar.

42. The method of any one of claims 34 to 41, wherein said compression is at a pressure of between about 10Bar and about 200Bar.

43. The method of any one of claims 34 to 42, comprising drying the compressed mixture after extraction from the mold, the drying comprises removal of liquid to a moisture content that is between about 0.1% and 1% out of a total weight of the resulting article of manufacture.

44. The method of claim 43, wherein said drying comprises any one or combination of air drying, oven drying, controlled humidity oven, and dry air blowing.

45. The method of any one of claims 34 to 44, comprising coating said article of manufacture with a sealer.

46. The method of claim 45, wherein said sealer is a natural based sealer.

47. An article of manufacture comprising a compressed mixture, the article of manufacture obtained or obtainable by molding a mixture comprising particulates comprising a blend of (i) clay material comprising at least one clay mineral having a plasticity index above about 20% and liquid limit above about 40%, as determined according to ASTM4318 and (ii) water insoluble inorganic material and fibrous material, said molding being characterized by using a mold that is equipped with (i) a pressing mechanism configured to receive the mixture therewithin and press the mixture to extract the liquid therefrom; (ii) a press member operatively coupled to the pressing mechanism and configured to operate the pressing mechanism for pressing the mixture; and (iii) a biasing mechanism configured to continuously bias the pressing mechanism in a direction to pressurize the mixture and compensate for a pressure drop in the mixture due to said extraction of the liquid.