

A single- or multiple component composition for producing a hydrogel

Technical field

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The invention relates to compositions used for producing (meth)acrylic hydrogels. The invention also relates to method for producing hydrogels and injection materials from the compositions and to method for sealing and/or filling of cracks, voids, flaws, and cavities in construction industry.

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Background of the invention

15 Swellable polymer materials such as acrylic based hydrogels are commonly used in construction industry as injection materials or sealants. Hydrogels are produced by polymerizing and cross-linking water-soluble monomer and/or prepolymer compounds in water based solutions or by contacting superabsorbent polymers with water. The polymerization mixture typically comprises, in addition to monomer or prepolymer compounds, an initiator for
20 the polymerization reaction and optionally a catalyst. The monomer and prepolymer compounds must be water-soluble in order to achieve the hydrogel upon polymerization. Monomer and prepolymer compounds which are not water-soluble cause separation of the water from the formed polymer material.

25 The ingredients for producing hydrogels are usually stored and delivered in a form of two-, three- or four-component compositions or kits due to the reactivity of the ingredients. In a two-component composition, for instance, one of the components contains the monomer/prepolymer compounds and optionally the catalyst, and the other component contains the initiator. In a
30 three-component composition, the monomer/prepolymer compounds, the initiator, and the catalyst are each provided as a separate component.

One of the disadvantages of the above mentioned multi-component compositions is the excess time needed for mixing several components and the risk that at the time of use the proportions of polymerizable material, initiator and catalyst are not measured correctly resulting in deterioration of the physical properties of the hydrogel.

Summary of the invention

The objective of the present invention is to provide a single- or multiple-component composition for producing (meth)acrylic hydrogels, which obviates the disadvantages and drawbacks of state of the art compositions.

According to the invention, the aforementioned objectives are achieved with the single- or multiple-component composition according to claim 1.

The main concept of the invention is that the monomer/prepolymer compounds are present in the single- or multiple component composition as supported on a solid carrier or in solid particulate form. The single- or multiple component composition further comprises a free radical initiator and optionally a catalyst for the free radical initiator.

One of the advantages of the present invention is that the composition can be delivered as a ready-to-use single component package to the construction site and only water has to be added to the composition to initiate the radical polymerization reaction. Consequently, the proportions of the reactive constituents such as the amount of the polymerizable monomer or prepolymer compounds, the amount of the initiator, and the amount of the catalyst can be determined in the manufacturing process. The end user at the construction site has to determine only the correct amount of water to be added to the composition to form a hydrogel with desired physical properties. This not only saves time at the construction site but also reduces the risk that proportions of constituents would be incorrectly measured.

Another advantage of the present invention is that the single- or multiple component composition has a long shelf-life even without the use of polymerization inhibitors since the monomer and/or prepolymer compounds are present in the composition as supported on a solid carrier or in solid particulate form.

In another aspect of the present invention, a method for producing a (meth)acrylic hydrogel, a (meth)acrylic hydrogel obtainable by the method, a (meth)acrylic injection material, and a method for sealing and/or filling of cracks, voids, flaws, and cavities in a building structure, is provided.

Detailed description of the invention

The term "(meth)acrylic" designates methacrylic or acrylic. Accordingly, (meth)acryloyl designates methacryloyl or acryloyl. A (meth)acryloyl group is also known as (meth)acryl group. A (meth)acrylic compound can have one or more (meth)acryl groups (mono- di-, tri- etc. functional (meth)acrylic compounds).

The term "(meth)acrylic" hydrogel designates a water containing gel, which contains hydrophilic (meth)acrylic polymer. In particular, the hydrophilic (meth)acrylic polymer in the hydrogel is usually crosslinked, e.g. via covalent bonds (chemical gel) or via non-covalent bonds such as ionic interaction or hydrogen bonds (physical gel). A (meth)acrylic polymer is a polymer of one or more (meth)acrylic compounds or monomers and optionally one or more co-monomers, which are co-polymerizable with the (meth)acrylic compounds or monomers.

Substance names beginning with "poly" designate substances which formally contain, per molecule, two or more of the functional groups occurring in their names. For instance, a polyol refers to a compound having at least two

hydroxyl groups. A polyether refers to a compound having at least two ether groups.

The term "water-soluble compound", e.g. a water-soluble (meth)acrylic
5 compound, designates compounds having a solubility of at least 5 g/100 g water, at a temperature of 20°C.

The term "room temperature" designates a temperature of 23 °C.

10 The term "gelling time" designates a time frame within which a hydrogel is formed once the constituents forming the hydrogel have been mixed with each other.

The term "open time" designates a time period for which the composition can
15 still be processed, such as injected into a crack, after the constituents forming hydrogel have been mixed with each other. The end of the open time is usually associated with such an increase in viscosity of the mixture that processing of the mixture is no longer possible.

20 The present invention relates in a first aspect of the invention to a single- or multiple-component composition for producing a (meth)acrylic hydrogel comprising constituents:

- a) a radical polymerizable monomer or prepolymer system,
- 25 b) a free-radical initiator,
- c) optionally a catalyst for said initiator,

wherein the radical polymerizable monomer or prepolymer system comprises:

- a1) at least one at room temperature liquid water soluble (meth)acrylic
30 compound supported on at least one solid carrier and/or
- a2) at least one at room temperature solid or liquid water-soluble (meth)acrylic compound dissolved in a liquid solution, preferably in an

aqueous solution, which liquid solution is supported on at least one solid carrier.

The term "single-component composition" refers to a composition, in which all
5 the constituents of the composition are contained in one single component.
Single-component compositions can be provided packaged in a single
compartment or package whereas in two-component or multi-component
compositions the components are provided packaged in physically separated
compartments or in separate packages.

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Single-component reactive systems are used in wide variety of sectors such as
in sealants, adhesives, coatings and floor coverings. In these systems, the
reactive constituents, e.g. catalyst, cross-linking agent, accelerator or initiator,
have to be immobilized in order to prevent initiation of the reaction before the
15 time of use. In case the single-component composition is a moisture or oxygen
curing composition, the immobilization of the reactive constituents is
accomplished by providing a diffusion barrier between the composition and
atmosphere. Premature reaction of the composition can also be prevented by
encapsulation of one or more of the reactive constituents, which are then
20 released by rupturing, dissolving or otherwise opening the capsules when the
composition is used.

The terms "a compound supported on a solid carrier" and "a compound
immobilized on a solid carrier" are used interchangeably and they refer to a
25 compound, which has been substantially completely adsorbed on or absorbed
in the solid carrier or encapsulated by the solid carrier. The solid carrier is
preferably in the form solid particles, more preferably in form of free-flowing
powder. By the term "substantially completely adsorbed on or absorbed in or
encapsulated by" is meant that more than 99.0 % by weight, preferably more
30 than 99.5 % by weight, even more preferably 99.9 % by weight, most
preferably 100.0 % by weight of the liquid compound is adsorbed on or
absorbed in the solid carrier or encapsulated by the solid carrier.

In adsorption, the molecules of the liquid component are taken up by the surface of the carrier whereas in absorption, the molecules are taken up by the volume of the carrier. The absorption can be based on a physical or chemical phenomenon (physical/chemical absorption). In encapsulation, the liquid
5 compounds are present in a core, which is surrounded by an encapsulant to form a stable encapsulated particle. A liquid compound supported on a solid carrier is available in form of solid particles, preferably in form of free flowing powder. Liquid compounds that are supported on a solid carrier and are then available as free flowing powder, are also known as dry liquids.

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The term "loading of a solid carrier" refers to the weight percentage of the amount of the liquid component supported on the carrier in relation to the total amount of the solid carrier including the liquid compound. For example, in case a liquid compound has been supported on a solid carrier "with a loading of 50
15 % by weight", the solid carrier particles contain 50 % by weight of the liquid compound based on the total amount of the solid carrier particles including the amount of the liquid compound.

The maximum amount of a liquid compound that can be supported on a solid
20 carrier depends on the properties of the liquid compound and the solid carrier and on the mechanism by which the liquid compound is supported on the solid carrier. In case the liquid compound is supported to a solid carrier by adsorption, the ability of the solid carrier to uptake the liquid compound depends mainly on the specific surface area of the solid carrier. On the other
25 hand, if the liquid compound is attached to the solid carrier by absorption, the loading degree depends mainly on the swelling properties of the solid carrier.

The term "liquid solution" refers to a homogeneous mixture of two or more substances. In such a mixture, a solute is a substance dissolved in another
30 substance, known as a solvent. The term "aqueous solution" refers to a solution, in which water is the solvent.

- The at least one at room temperature liquid or solid water-soluble (meth)acrylic compound may be a monomer, an oligomer or a polymer. The at least one at room temperature liquid or solid water-soluble (meth)acrylic compound may have e.g. a molecular weight or, if it is an oligomer or polymer with a molecular weight distribution, a weight average molecular weight of not more than 12000 g/mol, preferably not more than 8000 g/mol and more preferably not more than 4000 g/mol. The weight average molecular weight can be determined by gel permeation chromatography (GPC) with a polystyrene standard.
- 10 The at least one at room temperature liquid or solid (meth)acrylic compound is water-soluble in order to achieve the hydrogel upon polymerization. (Meth)acrylic compounds which are not water-soluble cause separation of the water from the (meth)acrylic polymer formed. Preferably, the at least one at room temperature liquid or solid (meth)acrylic compound has a solubility of at least 5 g/100 g water at a temperature of 20°C. Most preferably water and the at least one at room temperature liquid or solid (meth)acrylic compound are completely soluble in each other, i.e. they form a homogenous phase at any mixing ratio.
- 20 The at least one at room temperature liquid or solid water-soluble (meth)acrylic compound may have one, two or more than two (meth)acryloyl groups. Preferably, the at least one at room temperature liquid or solid water-soluble (meth)acrylic compound has one, two or three (meth)acryloyl groups.
- 25 The radical polymerizable monomer or prepolymer system preferably comprises at least one at room temperature liquid or solid water-soluble (meth)acrylic compound having one (meth)acryloyl group and at least one at room temperature liquid or solid water-soluble (meth)acrylic compound having two or three (meth)acryloyl groups. Water-soluble (meth)acrylic compounds having four or more (meth)acryloyl groups may be contained in addition, but this is usually not preferred.
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One of the characteristics of the present invention is that the single- or multiple-component composition forms a (meth)acrylic hydrogel after being mixed with such an amount of water that in the thus resulting mixture, the weight ratio of the total amount of the water-soluble (meth)acrylic compounds to water is in the range of 0.1 to 5, preferably in the range of 0.1 to 3. In calculating the said weight ratio, the total amount of the water-soluble (meth)acrylic compounds does not include the weight of the at least one solid carrier or the weight of solvent(s) if present in the single- or multiple-component composition.

10 Preferably, the (meth)acrylic hydrogel is formed in 30 s – 240 min, more preferably in 1 – 120 min, most preferably in 5 – 90 min after the single- or multiple-component composition has been mixed with water.

Preferably, the single- or multiple-component composition has a shelf life of at least one month, preferably at least two months, most preferably at least six months at a temperature of 50 °C and at a relative humidity of 50 %. The term “shelf-life” refers to a period of time for which the composition can be stored at the specified conditions without any significant changes in the application properties and reactivity of the composition. The changes in application properties refer to the changes in open time and/or gelling time and/or swelling properties of the hydrogel.

According to one embodiment, the single- or multiple-component composition is a single-component powder composition, preferably a free flowing powder composition, wherein the constituents a), b), and c) are present in one single component K. The term “free-flowing powder” refers to a powder, in which the particles do not stick together to form aggregates.

In order to create a single-component powder composition, all the compounds contained in the composition have to be present in the composition either in solid state, such as in solid particulate form or as liquid supported on the at least one solid carrier. It is preferable that in case the both constituents b) and c) contain one or more liquid compounds supported on the at least one solid

carrier, these liquid compounds are not supported on the same solid carrier in order to prevent a premature reaction of the free-radical initiator and the catalyst for the free-radical initiator.

5 According to another embodiment, the single- or multiple-component composition is a two component composition composed of a first component K1 and a second component K2, wherein two of the constituents a) – c) are present in a first component K1 and the third constituent is present in a second component K2. The constituents a) – c) can be arranged to the first and
10 second components K1 and K2 in any conventional way, e.g. the constituents a) and b) can be present in the first component K1 and the constituent c) in the second component K2, or constituents a) and c) can be present in the first component K1 and constituent b) in the second component K2, or the constituent a) can be present in the first component K1 and the constituents b)
15 and c) in the second component. The two component composition can be stored in two separate packages or in one package having two chambers that are separated from each other. Suitable two-chamber packaging formats include, e.g., dual cartridges such as twin or coaxial cartridges, multi-chamber poaches or bags with adapters.

20 Even though the constituents b) and c) can be provided in the same component, it is preferred that they are not provided in the same component in a two-component composition. However, in case the constituents b) and c) are present in the same component of a two-component composition, all the
25 compounds contained in these constituents are preferably either supported on the at least one solid carrier or are in solid particulate form. It is furthermore preferable that in case the constituents b) and c) contain one or more liquid compounds supported on the at least one solid carrier, these liquid compounds are not supported on the same solid carrier in order to prevent a premature
30 reaction of the free-radical initiator and the catalyst for the free-radical initiator.

According to another embodiment, the single- or multiple-component composition is a three component composition composed of a first component

K1, a second component K2 and a third component K3, wherein the constituent a) is present in the first component K1, the constituent b) is present in the second component K2, and the constituent c) is present in the third component K3.

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There are no particular restrictions for the at least one solid carrier, which can be used to immobilize the liquid compounds contained in the constituents of the single- or multiple-component composition. However, it should be possible to support the liquid compounds on the solid carrier with a reasonably high loading since a too high amount of solid support material has been found to have a negative effect on the application properties of the hydrogel. The suitability of a solid carrier depends also on, which compounds contained in the constituents a), b), and c) is to be supported on it. The (meth)acrylic compounds are preferably supported on the at least one solid carrier with a relatively high loading of at least 30 % by weight, more preferably of at least 40 % by weight, most preferably at least 50 % by weight, due to their high amount in the single- or multiple component composition. On the other hand, initiators and catalyst can be supported on the at least one solid carrier with a lower loading, such as with a loading of at least 5 % by weight, more preferably at least 10 % by weight, most preferably of at least 15 % by weight, without significantly increasing the amount of solid carrier in the single- or multiple component composition. In particular, different solid carriers can be used to support (meth)acrylic compounds, initiators, and catalysts.

25 In case the liquid compound is supported on the solid carrier mainly by adsorption, the solid carrier preferably has a high specific surface area. Preferably, the at least one solid carrier has a specific surface area measured with a BET method according to EN ISO 9277:2010 standard of 50.0 – 2000 m²/g, more preferably of 100.0 – 1500 m²/g, most preferably of 200.0 – 1000 m²/g.

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Preferably, the at least one solid carrier has a median particle size d_{50} of 0.1 – 1500.0 μm , more preferably of 0.1 – 1000.0 μm , even more preferably of 1.0 – 750.0 μm , most preferably of 5.0 – 500.0 μm .

- 5 The term median particle size d_{50} refers to a particle size below which 50 % of all particles by volume are smaller than the d_{50} value. The term “particle size” refers to the area-equivalent spherical diameter of a particle. The particle size distribution can be measured by laser diffraction according to the method as described in standard ISO 13320-1:2009.

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It can also be preferable that the at least one solid carrier has an adsorption capacity of dioctyl adipate (DOA) of 10 – 1000 ml/100 g, more preferably 50 – 750 ml/100 g, even more preferably 50 – 700 ml/100 g most preferably 75 – 500 ml/100 g.

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- Preferably, the at least one solid carrier is selected from the group consisting of particles of perlite, expanded perlite, glass, phonolite, calcium silicates, fumed silica, precipitated silica, silica gel, foamed polyurethane, polysaccharides, expanded vermiculite, clay minerals, fumed metal oxides, zeolites, kieselguhr, and mixtures thereof, more preferably from the group consisting of particles of perlite, expanded perlite, glass, phonolite, calcium silicates, fumed silica, precipitated silica, foamed polyurethane, polysaccharides, expanded vermiculite, clay minerals, kieselguhr, and mixtures thereof.

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- 25 Preferably, the single- or multiple component composition contains 5.0 – 90.0 % by weight, more preferably 10.0 – 70.0 % by weight, even more preferably 10.0 – 60.0 % by weight, most preferably 15.0 – 50.0 % by weight of the at least one solid carrier, based on the total weight of the single- or multiple component composition. In calculating the total weight of the at least one solid carrier, the weight of the at least one solid carrier does not include the weight of the liquid compounds or solutions supported on the solid carrier.

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The at least one at room temperature liquid water-soluble (meth)acrylic compound is preferably selected from the group consisting of hydroxyl-functional (meth)acrylates, carboxyl-functional (meth)acrylic compounds, low molecular weight polyether (meth)acrylates, (meth)acrylamides, or mixtures thereof.

A hydroxyl-functional (meth)acrylate is a (meth)acrylate having one or more hydroxyl groups. Examples of suitable at room temperature liquid water-soluble hydroxyl-functional (meth)acrylates are hydroxyethylmethacrylate (HEMA), hydroxyethylacrylate (HEA), hydroxypropylmethacrylate (HPMA), hydroxypropylacrylate (HPA), hydroxybutylmethacrylate (HBMA) and hydroxybutylacrylate (HBA).

A carboxyl-functional (meth)acrylic compound is a (meth)acrylic compound having one or more carboxylic groups such as e.g. (meth)acrylic acids or (meth)acrylic acids having one or more additional carboxylic groups. Examples of suitable carboxyl-functional at room temperature liquid water-soluble (meth)acrylic compounds and anhydrides thereof are methacrylic acid, methacrylic acid anhydride, acrylic acid, acrylic acid anhydride, and adduct of hydroxyethylmethacrylate.

Polyether (meth)acrylates are polyethers having one, two, three or more (meth)acrylate groups, respectively, preferably at the terminal ends thereof, wherein the polyether is preferably a polyethylene glycol (PEG), a methoxy polyethylene glycol (MPEG), a polyethylene glycol polypropylene glycol (PEG/PPG) copolymer, in particular block copolymer, an ethoxylated trimethylolpropane or an ethoxylated pentaerythritol. When the polyether is a PEG/PPG copolymer or blockcopolymer, respectively, the amount of PEG therein is preferably at least 30 % by weight, in order to achieve a suitable water solubility. The polyether (meth)acrylate is preferably a polyether having one (meth)acrylate group or a polyether di(meth)acrylate.

Polyether (meth)acrylates and polyether di(meth)acrylates also include polyethers having one or two (meth)acrylate groups, respectively, wherein the polyether includes further structural units such as urethane groups, e.g. oligomers or prepolymers obtained by reaction of polyetherpolyols, in particular
5 polyetherdiols, or polyethermonools with compounds having two functional groups which are reactive to hydroxyl groups such as polyisocyanates. For instance, polyether (meth)acrylates and polyether di(meth)acrylates may be obtained by reaction of polyetherpolyols or polyethermonools such as PEG, MPEG, PEG-PPG or MPEG-PPG or MPPG-PEG block copolymers, with
10 polyisocyanates to obtain an isocyanate-functional product which is subsequently reacted with a hydroxyl-functional (meth)acrylic compound such as hydroxyethyl methacrylate. With respect to water solubility, also in this case the PEG/PPG blocks preferably have an amount of PEG of at least 30 % by weight.

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Examples of suitable at room temperature liquid polyether (meth)acrylates and polyether di(meth)acrylates are low molecular weight PEG-di(meth)acrylates such as PEG 200 dimethacrylate, PEG 400 dimethacrylate, PEG 600 dimethacrylate, low molecular weight MPEG-(meth)acrylates such as MPEG
20 350 (meth)acrylate, and MPEG 550 (meth)acrylate. Such compounds are commercially available, e.g. from Sartomer, France, e.g. SR252 which is polyethylene glycol (600) dimethacrylate, from Geo Specialty Chemicals, USA, e.g. Bisomer MPEG-350MA, which is methoxy polyethyleneglycol methacrylate. The term "low molecular weight PEG/MPEG-di(meth)acrylate"
25 refers here to PEG-di(meth)acrylates having a molecular weight of 700 g/mol or less.

Examples of suitable at room temperature liquid ethoxylated trimethylolpropane (meth)acrylates and ethoxylated pentaerythritol (meth)acrylates are an
30 ethoxylated trimethylolpropane tri(meth)acrylate or an ethoxylated pentaerythritol tetra(meth)acrylate. Such compounds are commercially available, e.g. from Sartomer Americas, USA, e.g. SR415 which is ethoxylated (20) trimethylolpropane triacrylate (20 mole ethoxylated per mole TMP), SR454

which is ethoxylated (3) trimethylolpropane triacrylate (3 mole ethoxylated per mole TMP) or SR494 which is ethoxylated (4) pentaerythritol tetraacrylate (4 mole ethoxylated per mole PE).

5 The radical polymerizable monomer or prepolymer system may optionally comprise one or more at room temperature liquid water-soluble co-monomers, which are supported on the at least one solid carrier. These co-monomers are co-polymerizable with the acrylic and/or methacrylic compounds or monomers, respectively. In particular, the water-soluble co-monomers have a solubility of
10 at least 5 g/100 g water at 20°C. It goes without saying that the water-soluble co-monomer is different from the acrylic and/or methacrylic compounds. The water-soluble co-monomer is preferably a vinyl compound such as a vinyl ester, a divinyl ester, a vinyl ether or a divinyl ether, preferably a hydroxyl-functional vinyl ether or a hydroxyl-functional divinylether.

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The one or more water-soluble co-monomers, if used, are preferably used in relatively low amounts with respect to the acrylic and/or methacrylic compounds, e.g. in an amount of not more than 15 % by weight, preferably not more than 5 % by weight, more preferably not more than 1 % by weight, based
20 on the total amount of acrylic and/or methacrylic compounds and water-soluble co-monomers contained in the single- or multiple-component component composition.

Preferably the at least one at room temperature liquid water-soluble
25 (meth)acrylic compound is selected from the group consisting of hydroxyethylmethacrylate (HEMA), hydroxypropylmethacrylate (HPMA), low molecular weight polyethylene glycol dimethacrylate (PEG-DMA), and ethoxylated trimethylolpropane tri(meth)acrylate (TMP-TMA), or mixtures of these.

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Preferably, the single- or multiple component composition contains 10.0 – 90.0 % by weight, more preferably 12.5 – 80.0 % by weight, most preferably 15.0 – 75.0 % of the at least one at room temperature liquid water-soluble

(meth)acrylic compound supported on the at least one solid carrier, based on the total weight of the single- or multiple component composition. In calculating said weight percentages, the amount of the at least one at room temperature liquid water-soluble (meth)acrylic compound does not include the amount of
5 the at least one solid carrier, on which the at least one at room temperature liquid water-soluble (meth)acrylic compound has been supported.

Preferably, the at least one at room temperature liquid water-soluble (meth)acrylic compound is supported on the at least one solid carrier with a
10 loading of at least 10.0 % by weight, more preferably of at least 30.0 % by weight, even more preferably of at least 40.0 % by weight, most preferably of at least 50.0 % by weight.

Preferably, the single- or multiple component composition contains 10.0 – 90.0
15 % by weight of the at least one at room temperature liquid (meth)acrylic compound, based on the total weight of the single- or multiple component composition, which at least one at room temperature liquid water-soluble (meth)acrylic compound is supported on the at least one solid carrier with a loading of 30.0 – 95.0 % by weight, more preferably of 40.0 – 95.0 % by
20 weight, most preferably of 50.0 – 95.0 % by weight.

The at least one at room temperature liquid water-soluble (meth)acrylic compound may also be dissolved in a liquid solution, preferably in an aqueous solution, which liquid solution is supported on the at least one solid carrier with
25 a loading of at least 10.0 % by weight, more preferably of at least 30.0 % by weight, even more preferably of at least 40.0 % by weight, most preferably of at least 50.0 % by weight.

Preferably, the concentration of the at least one at room temperature liquid
30 (meth)acrylic compound in the said liquid solution is 10.0 – 95.0 % by weight, more preferably 50.0 – 95.0 % by weight, most preferably 70.0 – 90.0 % by weight, in relation to the total weight of the liquid solution.

According to one embodiment, the radical polymerizable monomer or prepolymer system further comprises:

a3) at least one at room temperature solid water-soluble (meth)acrylic compound in solid particulate form.

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Preferably, the at least one at room temperature solid water-soluble (meth)acrylic compound in solid particulate form has a median particle size d_{50} of less than 5000.0 μm , more preferably of less than 3000.0 μm , even more preferably of less than 1500.0 μm , most preferably of less than 1000 μm .

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Preferably, the at least one at room temperature solid water-soluble (meth)acrylic compound is selected from the group consisting of solid carboxyl functional (meth)acrylic compounds, salts of a carboxyl functional (meth)acrylic compounds, high molecular weight polyether (meth)acrylates, (meth)acrylates having a sulfonic acid group, (meth)acrylamides having a sulfonic acid group, salts or esters of (meth)acrylates having a sulfonic acid group, (meth)acrylamides having a sulfonic acid group, (meth)acrylates having a quaternary nitrogen containing group, (meth)acrylamides having a quaternary nitrogen containing group, and mixtures thereof.

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Examples of suitable at room temperature solid water-soluble carboxyl functional (meth)acrylic compounds are itaconic acid, maleic acid, maleic anhydride, adduct of maleic anhydride, and succinic acid anhydride.

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The cation for the salts mentioned above and below may be any common cation used in such compounds. Examples of suitable salts are metal salts, in particular alkali metal salts or earth alkaline metal salts, such as sodium salts, potassium salts or magnesium salts, or ammonium salts. Examples of suitable salts of carboxyl-functional (meth)acrylic compounds are salts of (meth)acrylic acids such as sodium acrylate, sodium methacrylate, potassium acrylate, potassium methacrylate, magnesium diacrylate and magnesium dimethacrylate.

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- Examples of suitable at room temperature solid water-soluble polyether (meth)acrylates and polyether di(meth)acrylates are high molecular weight PEG-di(meth)acrylates such as PEG 2000 dimethacrylate and high molecular weight MPEG-(meth)acrylates such as MPEG 1000 (meth)acrylate and MPEG 2000 (meth)acrylate. The terms "high molecular weight PEG-di(meth)acrylate" and "high molecular weight MPEG-(meth)acrylate" refer here to PEG-di(meth)acrylates and MPEG-(meth)acrylates having a molecular weight of more than 700 g/mol, respectively.
- Examples of suitable at room temperature solid water-soluble (meth)acrylates or (meth)acrylamides having a sulfonic acid group, and salts or esters thereof are 2-acrylamido-2-methylpropane sulfonic acid (AMPS®) or the sodium salt of 2-acrylamido-2-methylpropane sulfonic acid (Na-AMPS®) and sulfatoethyl methacrylate.
- Examples of suitable at room temperature solid water-soluble (meth)acrylates and (meth)acrylamides having a quaternary nitrogen containing group are 2-trimethylammoniummethyl methacrylate chloride and 3-trimethylammoniumpropyl methacrylamide chloride.
- Preferably the at least one at room temperature solid water-soluble (meth)acrylic compound is selected from the group consisting of maleic acid, itaconic acid, magnesium diacrylate, sodium acrylate, potassium acrylate, potassium salt of 3-sulfopropylacrylate, high molecular weight PEG-dimethacrylate, 2-acrylamido-2-methylpropane sulfonic acid (AMPS®), sodium salt of 2-acrylamido-2-methylpropane sulfonic acid (Na-AMPS®), and mixtures thereof.
- Preferably, the single- or multiple component composition contains 0.0 – 60.0 % by weight, more preferably 0.5 – 40.0 % by weight, most preferably 1.0 – 20.0 % of the at least one at room temperature solid water-soluble (meth)acrylic compound in solid particulate form, based on the total weight of the single- or multiple component composition.

According to one embodiment, the radical polymerizable monomer or prepolymer system comprises at least one at room temperature solid water-soluble (meth)acrylic dissolved in a liquid solution, preferably in an aqueous solution, which liquid solution is supported on the at least one solid carrier, wherein the at least one at room temperature solid water-soluble (meth)acrylic compound is selected from the group consisting of magnesium diacrylate, sodium acrylate, potassium acrylate, potassium salt of 3-sulfopropylacrylat, high molecular weight PEG-dimethacrylate, 2-acrylamido-2-methylpropane sulfonic acid (AMPS®), sodium salt of 2-acrylamido-2-methylpropane sulfonic acid (Na-AMPS®), and mixtures thereof.

Preferably, the single- or multiple component composition contains 10.0 – 90.0 % by weight, more preferably 12.5 – 80.0 % by weight, most preferably 15.0 – 75.0 % by weight of said at least one at room temperature solid water-soluble (meth)acrylic compound dissolved in a liquid solution, based on the total weight of the single- or multiple component composition. In calculating said weight percentages, the amount of the at least one at room temperature solid water-soluble (meth)acrylic compound dissolved in a liquid solution does not include the weight of the at least one solid carrier or the weight of the solvent.

Preferably, said liquid or aqueous solution of the at least one at room temperature solid water-soluble (meth)acrylic compound is supported on the at least one solid carrier with a loading of at least 10.0 % by weight, more preferably of at least 30.0 % by weight, even more preferably of at least 40.0 % by weight, most preferably of at least 50.0 % by weight.

Preferably, the concentration of the at least one at room temperature solid (meth)acrylic compound in said liquid or aqueous solution is 10.0 – 95.0 % by weight, more preferably 50.0 – 95.0 % by weight, most preferably 70.0 – 90.0 % by weight, in relation to the total weight of the liquid solution.

- The radical polymerizable monomer or prepolymer system can also comprise or consist of a mixture of the at least one at room temperature liquid water-soluble (meth)acrylic compound supported on a first solid carrier and the at least one at room temperature solid (meth)acrylic compound dissolved in a liquid solution, which liquid solution of the at least one at room temperature solid water-soluble (meth)acrylic compound is supported on the first solid carrier or on a second solid carrier. The first and second solid carriers can be of same or of different material.
- 10 The radical polymerizable monomer or prepolymer system can also comprise or consist of a mixture of the at least one at room temperature solid water-soluble (meth)acrylic compound in solid particulate form and at least one at room temperature solid (meth)acrylic compound dissolved in a liquid solution, which liquid solution of the at least one at room temperature solid water-soluble (meth)acrylic compound is supported on the at least one solid carrier.

Preferably, the sum of the:

- a1) at least one at room temperature liquid water soluble (meth)acrylic compound supported on at least one solid carrier,
- a2) at least one at room temperature solid or liquid water-soluble (meth)acrylic compound dissolved in a liquid solution, preferably in an aqueous solution, which liquid solution is supported on at least one solid carrier, and
- a3) at least one at room temperature solid water-soluble (meth)acrylic compound in solid particulate form

- amounts to 10.0 – 95.0 % by weight, more preferably 12.5 – 90.0 % by weight, even more preferably 15.0 – 85.0 %, most preferably 15.0 – 80.0 % by weight of the total weight of the single- or multiple component composition. In calculating said weight percentages, the amounts of a1), a2) and a) do not include the weight of the at least one solid carrier or the weight of solvent(s).

The single- or multiple-component composition can further comprise:

d) at least one inorganic base, preferably an alkali or earth alkali metal oxide or hydroxide, preferably at least one of potassium hydroxide (KOH), calcium hydroxide ($\text{Ca}(\text{OH})_2$), sodium hydroxide (NaOH),
5 magnesium hydroxide ($\text{Mg}(\text{OH})_2$).

According to one embodiment, the radical polymerizable monomer or prepolymer system comprises at least one at room temperature solid water-soluble (meth)acrylic compound in solid particulate form and/or at least one at
10 room temperature solid water-soluble (meth)acrylic compound dissolved in a liquid solution, preferably in an aqueous solution, which liquid solution of the at least one at room temperature solid water-soluble (meth)acrylic compound is supported on the at least one solid carrier, wherein the at least one at room temperature solid water-soluble (meth)acrylic compound is selected from the
15 group consisting of itaconic acid, maleic acid, maleic anhydride, adduct of maleic anhydride, succinic acid anhydride, and mixtures thereof, and the single- or multiple-component composition further comprises at least one inorganic base, preferably an alkali or earth alkali metal hydroxide, preferably at least one of potassium hydroxide (KOH), calcium hydroxide ($\text{Ca}(\text{OH})_2$),
20 sodium hydroxide (NaOH), magnesium hydroxide ($\text{Mg}(\text{OH})_2$).

The free-radical initiator serves to initiate polymerization of the (meth)acrylic compounds. These initiators are known to those skilled in the art. The initiator may be e.g. an organic or inorganic hydroperoxide, an organic or inorganic
25 peroxide such as a peroxydisulfate or persulfate salt, an azo compound, or any other material, which is known to the expert of being capable to generate radicals.

According to one embodiment, the free-radical initiator comprises at least one
30 at room temperature solid initiator in solid particulate form. Preferably, said at least one at room temperature solid initiator is selected from the group consisting of azobisisobutyronitrile (AIBN), sodium persulfate (NAPS), potassium persulfate or ammoniumpersulfate, and mixtures thereof.

Preferably, the at least one at room temperature solid initiator in solid particulate form has a median particle size d_{50} of less than 5000.0 μm , more preferably of less than 3000.0 μm , even more preferably of less than 1500.0 μm , most preferably of less than 1000 μm .

Preferably, the single- or multiple component composition contains 0.05 – 5.0 % by weight, more preferably 0.1 – 3.5 % by weight, most preferably 0.1 – 2.5 % by weight of the at least one at room temperature solid initiator in solid particulate form, based on the total weight of the single- or multiple component composition.

According to one embodiment, the free-radical initiator comprises at least one at room temperature liquid initiator supported on the at least one solid carrier. Preferably, said at least one at room temperature liquid initiator is selected from the group consisting of hydrogen peroxide, cumene hydroperoxide, tert-butyl hydroperoxide, diisopropylbenzene hydroperoxide, and mixture thereof.

The at least one at room temperature liquid initiator can be supported on the same solid carrier (the first solid carrier) as the at least one at room temperature liquid (meth)acrylic compound, if present in the single- or multiple-component composition, or on the same carrier (the second solid carrier) as the liquid/ aqueous solution of the at least one at room temperature solid water-soluble (meth)acrylic compound, if present in the single- or multiple-component composition. Preferably, the at least one at room temperature liquid initiator is supported on a third solid carrier. The third solid carrier can be of same material as first and second solid carriers or of different material.

Preferably, the single- or multiple component composition contains 0.05 – 5.0 % by weight, preferably 0.1 – 3.5 % by weight, most preferably 0.1 – 2.5 % by weight of the at least one at room temperature liquid initiator supported on the at least one solid carrier, based on the total weight of the single- or multiple component composition. In calculating said weight percentages, the amount of

the at least one at room temperature liquid initiator does not include the amount of the at least one solid carrier.

Preferably, the at least one at room temperature liquid initiator is supported on the at least one solid carrier with a loading of at least 5.0 % by weight, more preferably of at least 10.0 % by weight, even more preferably of at least 15.0 % by weight, most preferably of at least 20.0 % by weight.

Preferably, the single- or multiple component composition contains 0.05 – 5.0 % by weight of the at least one at room temperature liquid initiator, based on the total weight of the single- or multiple component composition, which at least one at room temperature liquid initiator is supported on the at least one solid carrier with a loading of 30.0 – 95.0 % by weight, more preferably of 40.0 – 95.0 % by weight, most preferably of 50.0 – 95.0 % by weight.

15

The free-radical initiator can also comprise a mixture of the at least one at room temperature solid initiator in solid particulate form and the at least one at room temperature liquid initiator supported on the at least one solid carrier.

According to one embodiment, the free-radical initiator comprises at least one at room temperature solid initiator dissolved in a liquid solution, preferably in an aqueous solution, which liquid solution is supported on the at least one solid carrier, wherein the at least one at room temperature solid initiator is selected from the group consisting of azobisisobutyronitrile (AIBN), sodium persulfate (NAPS), potassium persulfate or ammoniumpersulfate, and mixtures thereof.

The liquid/aqueous solution of the at least one at room temperature solid initiator can be supported on the same carrier (the first solid carrier) as the at least one at room temperature liquid (meth)acrylic compound, if present in the single- or multiple-component composition, or on the same carrier (the second solid carrier) as the liquid/aqueous solution of the at least one at room temperature solid water-soluble (meth)acrylic compound, if present in the single- or multiple-component composition, or on the same solid carrier (the

30

third solid carrier) as the at least one at room temperature liquid initiator, if present in the single- or multiple-component composition. Preferably, the liquid/aqueous solution of the at least one at room temperature solid initiator is supported on a fourth solid carrier. The fourth solid carrier can be of same material as first, second and third solid carriers or of different material.

Preferably, the single- or multiple component composition contains 0.05 – 5.0 % by weight, more preferably 0.1 – 3.5 % by weight, most preferably 0.1 – 2.5 % by weight of said at least one at room temperature solid initiator dissolved in a liquid solution, based on the total weight of the single- or multiple component composition. In calculating said weight percentages, the amount of the at least one at room temperature solid initiator dissolved in a liquid solution does not include the weight of the at least one solid carrier or the weight of the solvent.

Preferably, said liquid or aqueous solution of the at least one at room temperature solid initiator is supported on the at least one solid carrier with a loading of at least 5.0 % by weight, more preferably of at least 10.0 % by weight, even more preferably of at least 15.0 % by weight, most preferably of at least 20.0 % by weight.

Preferably, the concentration of the at least one at room temperature solid initiator in the said liquid or aqueous solution is 10.0 – 95.0 % by weight, more preferably 50.0 – 95.0 % by weight, most preferably 70.0 – 90.0 % by weight, in relation to the total weight of the liquid solution.

The free radical initiator can also comprise a mixture of the at least one at room temperature liquid initiator supported on the at least one solid carrier and the at least one at room temperature solid initiator dissolved in a liquid solution, which liquid solution is supported on the at least one solid carrier.

Preferably, the sum of the said initiators contained in the constituent b) amounts to 0.05 – 5.0 % by weight, more preferably 0.1 – 3.5 % by weight, most preferably 0.1 – 2.5 % by weight of the single- or multiple component

composition. In calculating said weight percentages, the amounts of the initiators do not include the weight of the at least one solid carrier or the weight of solvent(s).

- 5 The catalyst for the initiator serves to catalyze the polymerization reaction of the (meth)acrylic compounds. The catalyzing effect of the catalyst may be e.g. based on interaction with the initiator promoting radical generation. It is preferable that the single- or multiple component composition further comprises a catalyst for the initiator.

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According to one embodiment, the catalyst for the initiator comprises at least one at room temperature solid catalyst in solid particulate form. Preferably, said at least one at room temperature solid catalyst is selected from the group consisting of ascorbic acid, sodium formaldehyde sulfoxylate (SFS), sodium
15 thiosulfate, organic sulfinic acid derivatives and salts thereof such as Bruggolite FF6 and FF7 (commercially available from Bruggeman Chemical), toluidine derivatives, transition metal salts, transition metal complexes, and mixtures thereof.

- 20 Preferably, the at least one at room temperature solid catalyst in solid particulate form has a median particle size d_{50} of less than 5000.0 μm , more preferably of less than 3000.0 μm , even more preferably of less than 1500.0 μm , most preferably of less than 1000 μm .

- 25 Preferably, the single- or multiple component composition contains 0.05 – 7.5 % by weight, more preferably 0.1 – 5.0 % by weight, most preferably 0.1 – 3.5 % by weight of the at least one at room temperature solid catalyst in solid particulate form, based on the total weight of the single- or multiple component composition.

30

According to one embodiment, the catalyst for the initiator comprises at least one at room temperature liquid catalyst supported on the at least one solid carrier. Preferably, the at least one at room temperature liquid catalyst is

selected from the group consisting of alkanol amines, ethoxylated alkanol amines, and mixtures thereof. The at least one at room temperature liquid catalyst can be selected from the group consisting of diethanolamine (DEA), triethanolamine (TEA), N-Butyldiethanolamine, 2-Amino-2-methyl-1,3-
5 propandiol, dimethylaminopropyl methacrylamide (DMAPMA), dimethylaminoethyl methacrylate (DMAEMA), and mixtures thereof

The at least one at room temperature liquid catalyst can be supported on the same carrier (the first solid carrier) as the at least one at room temperature
10 liquid (meth)acrylic compound, if present in the single- or multiple-component composition, or on the same carrier (the second solid carrier) as the liquid/aqueous solution of the at least one at room temperature solid water-soluble (meth)acrylic compound, if present in the single- or multiple-component composition. However, the at least one at room temperature liquid catalyst is
15 preferably not supported on the same solid carrier (the third solid carrier) as the at least one at room temperature liquid initiator, if present in the single- or multiple-component composition or on the same solid carrier (the fourth solid carrier) as the liquid/aqueous solution of the at least one at room temperature solid initiator, if present in the single- or multiple-component composition.
20 Preferably, the at least one at room temperature liquid catalyst is supported on a fifth solid carrier. The fifth solid carrier can be of same material as first, second, third and fourth solid carriers or of different material.

Preferably, the single- or multiple component composition contains 0.05 – 7.5
25 % by weight, more preferably 0.1 – 5.0 % by weight, most preferably 0.1 – 3.5 % by weight of the at least one at room temperature liquid catalyst, based on the total weight of the single- or multiple component composition. In calculating said weight percentages, the amount of the at least one at room temperature liquid catalyst does not include the amount of the at least one solid carrier.

30

Preferably, the at least one at room temperature liquid catalyst is supported on the at least one solid carrier with a loading of at least 5.0 % by weight, more

preferably of at least 10.0 % by weight, even more preferably of at least 15.0 % by weight, most preferably of at least 20.0 % by weight.

Preferably, the single- or multiple component composition contains 0.01 – 7.5
5 % by weight of the at least one at room temperature liquid catalyst, based on the total weight of the single- or multiple component composition, which at least one at room temperature liquid catalyst is supported on the at least one solid carrier with a loading of 30.0 – 95.0 % by weight, more preferably of 40.0 – 95.0 % by weight, most preferably of 50.0 – 95.0 % by weight.

10

The catalyst for the initiator can also comprise a mixture of the at least one at room temperature solid catalyst in solid particulate form and at least one at room temperature liquid catalyst supported on the at least one solid carrier.

15 According to one embodiment, the catalyst for the initiator comprises at least one at room temperature solid catalyst dissolved in a liquid solution, preferably in an aqueous solution, which liquid solution is supported on the at least one solid carrier, wherein said at least one at room temperature solid catalyst selected from the group consisting of ascorbic acid, sodium formaldehyde
20 sulfoxylate (SFS), sodium thiosulfate, organic sulfinic acid derivatives and salts thereof such as Bruggolite FF6 and FF7 (commercially available from Bruggeman Chemical), transition metal salts, transition metal complexes, and mixtures thereof.

25 The aqueous liquid/solution of the at least one at room temperature solid catalyst can be supported on the same carrier (the first solid carrier) as the at least one at room temperature liquid (meth)acrylic compound, if present in the single- or multiple-component composition, or on the same carrier (the second solid carrier) as the aqueous solution of the at least one at room temperature
30 solid water-soluble (meth)acrylic compound, if present in the single- or multiple-component composition, or on the same solid carrier (the fifth solid carrier) as the at least one at room temperature liquid catalyst, if present in the single- or multiple-component composition. However, the aqueous solution of the at least

one at room temperature solid catalyst is preferably not supported on the same solid carrier (the third solid carrier) as the at least one at room temperature liquid initiator, if present in the single- or multiple-component composition or on the same solid carrier (the fourth solid carrier) as the aqueous solution of the at least one at room temperature solid initiator, if present in the single- or multiple-component composition. Preferably, the aqueous solution of the at least one at room temperature solid catalyst is supported on a sixth solid carrier. The sixth solid carrier can be of same material as first, second, third, fourth and fifth solid carriers or of different material.

10

Preferably, the single- or multiple component composition contains 0.05 – 7.5 % by weight, more preferably 0.1 – 5.0 % by weight, most preferably 0.1 – 3.5 % by weight of said at least one at room temperature solid catalyst dissolved in a liquid solution, based on the total weight of the single- or multiple component composition. In calculating said weight percentages, the amount of the at least one at room temperature solid catalyst dissolved in a liquid solution does not include the weight of the at least one solid carrier or the weight of the solvent.

Preferably, said liquid or aqueous solution of the at least one at room temperature solid catalyst is supported on the at least one solid carrier with a loading at least 5.0 % by weight, more preferably of at least 10.0 % by weight, even more preferably of at least 15.0 % by weight, most preferably of at least 20.0 % by weight.

Preferably, the concentration of the at least one at room temperature solid initiator in the said liquid or aqueous solution is 10.0 – 95.0 % by weight, more preferably 50.0 – 95.0 % by weight, most preferably 70.0 – 90.0 % by weight, in relation to the total weight of the liquid solution.

The catalyst for the initiator can also comprise a mixture of the at least one at room temperature liquid catalyst supported on the at least one solid carrier at least one at room temperature solid catalyst dissolved in a liquid solution,

30

preferably in an aqueous solution, which aqueous solution is supported on the at least one solid carrier.

Preferably, the sum of the said catalysts contained in the constituent c)
5 amounts to 0.05 – 7.5 % by weight, preferably 0.1 – 5.0 % by weight, most preferably 0.1 – 3.5 % by weight of the single- or multiple component composition. In calculating said weight percentages, the amounts of the catalysts do not include the weight of the at least one solid carrier or the weight of solvent(s).

10

According to one preferable embodiment, the radical polymerizable monomer or prepolymer system comprises at least one at room temperature liquid water-soluble (meth)acrylic compound supported on a first solid carrier and/or at least one at room temperature solid water-soluble (meth)acrylic compound in solid
15 particulate form, the free radical initiator comprises at least one at room temperature solid initiator in solid particulate form selected from the group consisting of azobisisobutyronitrile (AIBN), sodium persulfate (NAPS), potassium persulfate or ammoniumpersulfate, and the catalyst for the initiator comprises at least one at room temperature solid catalyst in solid particulate
20 form selected from the group consisting of ascorbic acid, sodium formaldehyde sulfoxylate (SFS), sodium salts of organic sulfinic acid derivatives such as Bruggolite, toluidine derivatives, transition metal salts, and transition metal complexes, wherein the first solid carrier is selected from the group consisting of particles of perlite, expanded perlite, glass, phonolite, calcium silicates,
25 fumed silica, precipitated silica, silica gel, foamed polyurethane, polysaccharides, expanded vermiculite, clay minerals, fumed metal oxides, zeolites, kieselguhr, and mixtures thereof.

According to another preferable embodiment, the radical polymerizable
30 monomer or prepolymer system comprises at least one at room temperature liquid water-soluble (meth)acrylic compound supported on a first solid carrier and/or at least one at room temperature solid water-soluble (meth)acrylic compound in solid particulate form, the free radical initiator comprises at least

one at room temperature solid initiator in solid particulate form selected from the group consisting of azobisisobutyronitrile (AIBN), sodium persulfate (NAPS), potassium persulfate or ammoniumpersulfate, and the catalyst for the initiator comprises at least one at room temperature liquid catalyst selected
5 from the group consisting of alkanol amines, ethoxylated alkanolamines, and mixtures thereof, preferably from the group consisting of diethanolamine (DEA), triethanolamine (TEA), N-Butyldiethanolamin, 2-Amino-2-methyl-1,3-propandiol, dimethylaminopropyl methacrylamide (DMAPMA), dimethylaminoethyl methacrylate (DMAEMA), and mixtures thereof, which at
10 least one at room temperature liquid catalyst is supported on the first solid carrier or on a second solid carrier, wherein the first and second solid carriers are selected from the group consisting of particles of perlite, expanded perlite, glass, phonolite, calcium silicates, fumed silica, precipitated silica, silica gel, foamed polyurethane, polysaccharides, expanded vermiculite, clay minerals,
15 fumed metal oxides, zeolites, kieselguhr, and mixtures thereof.

According to another preferable embodiment, the radical polymerizable monomer or prepolymer system comprises at least one at room temperature liquid water-soluble (meth)acrylic compound supported on a first solid carrier
20 and/or at least one at room temperature solid water-soluble (meth)acrylic compound in solid particulate form, the free radical initiator comprises at least one at room temperature liquid initiator selected from the group consisting of hydrogen peroxide, cumene hydroperoxide, tert-butyl hydroperoxide, diisopropylbenzene hydroperoxide, which at least one at room temperature
25 liquid initiator is supported on the first solid carrier or on a second solid carrier, and the catalyst for the initiator comprises at least one at room temperature liquid catalyst selected from the group consisting of alkanol amines, ethoxylated alkanolamines, and mixtures thereof, preferably from the group consisting of diethanolamine (DEA), triethanolamine (TEA), N-
30 Butyldiethanolamin, 2-Amino-2-methyl-1,3-propandiol, dimethylaminopropyl methacrylamide (DMAPMA), dimethylaminoethyl methacrylate (DMAEMA), and mixtures thereof, which at least one at room temperature liquid catalyst is supported on the second solid carrier or on a third solid carrier, wherein the

first, second and third solid carriers are selected from the group consisting of particles of perlite, expanded perlite, glass, phonolite, calcium silicates, fumed silica, precipitated silica, silica gel, foamed polyurethane, polysaccharides, expanded vermiculite, clay minerals, fumed metal oxides, zeolites, kieselguhr, and mixtures thereof.

The single- or multiple-component composition can further comprise:

e) at least one filler selected from the group consisting of inert mineral fillers, organic fillers, mineral binders, and mixtures thereof.

The term "filler" refers to solid particulate materials, which have a low adsorption capacity for the liquid components contained in the single- or multiple-component composition of the present invention and, therefore, are not preferably used as solid carriers as described above. Typically, a filler is not suitable for use as the solid carrier since any liquid component contained in the single- or multiple-component composition could be supported on the filler only with very small loading, such as less than 5.0 % by weight, preferably less than 2.5 % by weight, more preferably less than 2.0 % by weight, most preferably less than 1.0 % by weight. Although some solid carriers (described above) could be in principle used as filler and vice versa, it is preferred that different materials are used as solid carriers and fillers.

Preferably, the at least one filler also has a water-solubility of less than 0.1 g/100 g water, more preferably less than 0.05 g/100 g water, most preferably less than 0.01 g/100 g water, at a temperature of 20°C.

Preferably, the at least one filler has a particle size d_{50} in the range of 0.1 – 200.0 μm , more preferably of 0.1 – 100.0 μm , most preferably 0.1 – 50.0 μm .

30

The term "inert mineral filler" refers to mineral fillers that are not chemically reactive. They are produced from natural mineral sources by mining followed by comminution to required particle size and shape. In particular, inert mineral

fillers include quartz, ground or precipitated calcium carbonate, crystalline silicas, dolomite, clay, talc, graphite, mica, Wollastonite, barite, diatomaceous earth, and pumice.

- 5 The term "organic filler" refers to fillers comprising or consisting of organic materials. In particular, organic fillers include materials comprising or consisting of cellulose, polyethylene, polypropylene, polyamide, polyester, and dispersible polymer compositions such as Vinnapas (from Wacker Chemie AG) and Axilat 8510 (from Hexion).

10

The term "mineral binder" refers to hydraulic, non-hydraulic, latent binders, and pozzolanic binders. In particular, mineral binders include materials comprising or consisting of cement, cement clinker, hydraulic lime, non-hydraulic lime, and gypsum.

15

Preferably, the single- or multiple component composition contains 0.0-35.0 % by weight, more preferably 1.0-30.0 % by weight, most preferably 5.0-25.0 % by weight of the at least one filler, based on the total weight of the single- or multiple component composition.

20

The single- or multiple-component composition may optionally contain inhibitors. Inhibitors are often added to (meth)acrylic compounds, in particular in commercial products, in order to avoid spontaneous polymerization and/or to adjust open times and reaction times, respectively. Examples of suitable
25 inhibitors include butylhydroxytoluene (BTH), hydroquinone (HQ), monomethyl ether hydroquinone (MEHQ), PTZ (phenothiazine), and 4-hydroxy-2,2,6,6-tetramethylpiperidin-1-oxyl (4-hydroxy-TEMPO).

30

Apart from the above mentioned ingredients, the single- or multiple-component composition may optionally contain one or more further additives, which are common in this field. Examples are, color dyes and water-soluble diluents such as polyethylene glycol. Color dyes may be suitable to label the mixture.

In another aspect of the present invention, a method for producing a (meth)acrylic hydrogel is provided, the method comprising the steps of

- 5 i) Providing a single- or multiple-component composition according to the present invention,
- ii) Mixing the single- or multiple-component composition with water such that in the thus resulting mixture, the weight ratio of the total amount of the water-soluble (meth)acrylic compounds to water is in the range of 0.1 to 5, preferably in the range of 0.1 to 3,
- 10 iii) Letting the mixture form a (meth)acrylic hydrogel.

In calculating said weight ratio of step ii), the total amount of the water-soluble (meth)acrylic compounds does not include the weight of the at least one solid
15 carrier or the weight of solvent(s) if present in the single- or multiple-component composition.

The mixing step ii) is usually carried out by combining the single- or multiple-component composition with water under mixing. Suitable means for mixing
20 are a static mixers and dynamic mixers, in particular container-stirrer type mixers such as rotor-stator type mixers, dissolver mixers, colloidal mixers and other high shear mixers. The choice of suitable mixing apparatus depends on the open time of the composition. In case of long open time, the mixing can be conducted with a container equipped with a stirrer whereas in case of short
25 open time, a static mixer is preferably used in mixing.

Preferably, the time needed to complete step iii) is 30 s – 240 min, preferably 1 – 120 min, most preferably 5 – 90 min.

30 The viscosity of the mixture at the beginning is usually relatively low, since the mixture is mainly based on water, water-soluble ingredients, and solid carriers. The viscosity can be adjusted, e.g. by adjusting the ratio of water to the radical polymerizable monomer or prepolymer system and/or by adjusting the

molecular weight of the (meth)acrylic compound(s) and/or by adjusting the type and amount of rheology additives and/or by adjusting the type and amount of fillers.

- 5 The polymerization reaction preferably takes place at ambient temperatures, e.g. at temperatures in the range of -10 to 60 °C, more preferably in the range of 0 to 50 °C.

In another aspect of the present invention, a (meth)acrylic hydrogel obtainable
10 by the method as described above, is provided.

In still another aspect of the present invention, a (meth)acrylic injection material is provided, which a (meth)acrylic injection material is obtainable by mixing a single- or multiple-component composition of the present invention with water
15 such that in the thus resulting mixture, the weight ratio of the total amount of the water-soluble (meth)acrylic compounds to water is in the range of 0.1 to 5, preferably in the range of 0.1 to 3. In calculating said weight ratio, the total amount of the water-soluble (meth)acrylic compounds does not include the weight of the at least one solid carrier or the weight of solvent(s) if present in
20 the single- or multiple-component composition.

Preferably, the (meth)acrylic injection material has an open time of 30 s – 240 min, preferably 1 – 120 min, most preferably 5 – 90 min.

- 25 The viscosity of the (meth)acrylic injection material, measured with a Brookfield viscometer at temperature of 23 °C with a rotational speed of the viscometer in the range of 150 – 200 revolutions per minute, is preferably less than 500 mPa·s, more preferably less than 200 mPa·s, most preferably less than 150 mPa·s. Said viscosity of the (meth)acrylic injection material is measured
30 immediately after the complete dissolution of the constituents of the single- or multiple component composition in water.

In another aspect of the present invention, a method for sealing and/or filling of cracks, voids, flaws, and cavities in a building structure is provided, the method comprising steps of:

- 5 i) Providing a single- or multiple-component composition of the present invention,
- ii) Mixing the single- or multiple-component composition with water such that the weight ratio of the total amount of the water-soluble (meth)acrylic compounds to water in the mixture is in the range of 0.1 to 5, preferably in
10 the range of 0.1 to 3,
- iii) Applying the mixture to the site to be sealed/filled in the building structure and letting the mixture to form (meth)acrylic hydrogel.

In calculating said weight ratio, the total amount of the water-soluble
15 (meth)acrylic compounds does not include the weight of the at least one solid carrier or the weight of solvent(s) if present in the single- or multiple-component composition.

The polymerization reaction starts almost immediately upon mixing of the
20 single- or multiple-component composition and water. Therefore, the step iii) should be started soon after provision of the mixture and in any case within the open time of the mixture.

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Examples

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The followings compounds and products, respectively, were used in the examples:

Table 1

HEMA	Hydroxyethyl methacrylat (HEMA) including 400 ppm hydrochinon monomethylether (HMME) as inhibitor	Visiomer HEMA 98, Evonik Industries
ETMA	Ethyltriglycol methacrylate	Visiomer® ETMA, Evonik Industries
MPEG350MA	Methoxy polyethyleneglycol methacrylate, average molecular weight 430 g/mol	Bisomer® MPEG350MA, Geo Specialty Chemicals, USA
SR252	Polyethylene glycol (600) dimethacrylate, molecular weight 736 g/mol	Sartomer® SR252, Sartomer, France
Potassium acrylate	Potassium acrylate	CAS # 10192-85-5
Potassium methacrylate	Potassium methacrylate	CAS # 6900-35-2
AMPS	2-Acrylamido-2-methylpropane sulfonic acid, 50% solution in water	AMPS® 2405, Lubrizol
Sipernat 33	Silicon oxide	Sipernat® 33, Evonik Industries
Vinnapas 7055N	Vinyl acetate, ethylene and vinyl ester	Vinnapas® 7055 N, Wacker Chemie
Vinnapas 5045E	Vinyl acetate, ethylene and vinyl ester	Vinnapas® 5045 E, Wacker Chemie
PUR-mehl	Polyurethan particulate	CAS # 9009-54-5, Beisswenger GmbH
Cab-O-Sil M5	Untreated fumed silica	Cab-O-Sil® M-5, Cabot Corporation
Expanded perlite	Silicous rock	Perlite 0-1 mm, AG für Steinindustrie
NAPS	Sodium persulfate (as solid or diluted with	CAS # 7775-27-1

	water to yield a 20 wt.% solution)	
Trigonox TMBH-L	Tetramethylbutylhydroperoxide	Trigonox® TMBL-L, AkzoNobel
Trigonox TAHP-W85	Tert-Amyl hydroperoxide	Trigonox® TAHP-W85, AkzoNobel
TEA	Triethanolamine (technical grade 85%TEA /15% DEA)	Triethanolamin 85, Ineos Oxide
Ascorbic acid	Ascorbic acid	CAS # 50-81-7
Bruggolite FF7	2-Hydroxy-2-sulfonatoacetic acid and 2-hydroxy-2-sulfonatoacetic acid with sodium bisulfite	Bruggolite® FF7, Brügemann Chemical
Bruggolite L40	Composition based on sodium hydroxymethanesulfinate	Bruggolite® L40, Brügemann Chemical
Manganese 10HS	Composition based on metal salts octanoates with 10 % manganese	Octa-Soligen® Manganese 10 HS, OMG Borchers GmbH
ABL	Acetylbutyrolactone	CAS # 517-23-7
Axilat 8510	Vinyl acetat copolymer latex	Axilat® 8510, Hexion
Mikrodur R-U	Cement	Mikrodur® R-U, Dyckerhoff

The example compositions Ex1 to Ex38 were prepared by supporting the liquid compounds to solid carriers followed by mixing the ingredients with water to provide hydrogel forming mixtures. The mixing of the ingredients was conducted in a dissolver mixer with a mixer speed of 1000 revolutions per minute for three minutes. The ingredients and their amounts in parts by weight (pbw) are presented in Tables 2 to 4.

In the example compositions Ex1-Ex25 and Ex35-Ex38, the liquid triethanolamine (TEA) catalyst supported on Sipernat 33 particles with a loading of 50 % by weight (weight ratio TEA:Sipernat was 1:1) was used. The amount TEA in Tables 2-4 refers to the amount of TEA without the solid carrier, if used.

15

In the example compositions Ex1-Ex24, a sodium persulfate (NAPS) catalyst in solid particulate form was used and in the example compositions Ex25-Ex27 and Ex35-Ex38 an aqueous solution containing 20 % by weight of NAPS supported on Sipernat 33 particles with a loading of 50 % by weight (weight ratio aqueous solution:Sipernat was 1:1) was used. The amount of liquid

20

compounds supported on a carrier in Tables 2-4 refers to the amount of these compounds without the solid carrier.

In each of the example compositions Ex1 to Ex38, the polymerization started
5 when the ingredients were mixed with water and the mixture was finally transformed into a hydrogel. The properties of the hydrogels were tested in terms of gelling time, gel consistency and swelling. These results are also provided in Tables 2-4. The term "nd" means that the value was not determined.

10

Loading of carrier particles

The liquid compounds were supported on the solid carriers by mixing the carrier particles with the liquid compound in a dissolver mixer using a mixer speed of 2000 revolutions per minute with a mixing time of two minutes. The
15 loading percentage of the carrier particles was determined based on the measured weights of the particles before and after supporting of the liquid compounds on the particles.

Viscosity

20 The viscosity was measured before the initiator, such as NAPS, was added to the mixture and before any polymerization reaction took place. A qualitative analysis was carried out by observing the flow behavior of the mixture. In some cases the viscosity could not be determined due to inhomogeneous mixture or too high viscosity of the mixture.

25

A Brookfield DV2T viscosity measurement apparatus with ULA-DIN-85 spindle was used in viscosity measurements. The diameter of the spindle was 2.55 cm and the diameter of the vessel was 2.7 cm. All viscosities were determined at a temperature of 23 °C and with rotational speed in the range of 150 – 200
30 revolutions per minute.

Gelling time (min)

The gelling time at 23°C was determined by visual inspection (gel time is achieved at the time when gel-like structures are detected).

Gel consistency

- 5 The gel consistency of the hydrogel obtained was tested haptically.

Swelling 7 days in water

- 10 For the measurement of the swelling properties, a test specimen with dimensions of 1.0 x 1.5 x 1.5 cm was cut from the hydrogel material produced from each of the example compositions. The test specimens were stored in tap water at a temperature of 23 °C and the change in weigh of the test specimen was determined at the end of the 7 days test period. The hydrogel material was
- 15 cured for 24 hours before conducting the swelling test. The swelling values presented in Tables 2-4 were determined as the percentage change in weight of the test specimen during the swelling test.

Table 2

	Ex 1	Ex 2	Ex 3	Ex 4	Ex 5	Ex 6	Ex 7	Ex 8	Ex 9	Ex 10	Ex 11
Ingredients											
HEMA (pbw)	45.0					30.0	40.0	43.0	43.0	43.0	43.0
ETMA (pbw)		45.0	40.0								
MPEG350MA (pbw)				45.0	40.0	15.0					
SR252 (pbw)			5.0		5.0		5.0				
Potassium acrylate (pbw)								2.0			
Potassium methacrylate (pbw)									2.0		
AMPS (pbw)										2.0	2.0
Sipernat 33 (pbw)	15.0	15.0	15.0	15.0	15.0	15.0	15.0	15.0	15.0	15.0	15.0
Carrier loading (wt.-%)	75	75	75	75	75	75	75	75	75	75	75
Water (pbw)	60.0	60.0	60.0	60.0	60.0	60.0	60.0	60.0	60.0	60.0	90.0
^a NAPS (pbw)	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5
^b TEA (pbw)	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00
Total (pbw)	121.5	121.5	121.5	121.5	121.5	121.5	121.5	121.5	121.5	121.5	151.5
Results											
Viscosity (mPa·s) (without NAPS)	22	23	50	45	39	17	25	21	22	26	12
Gelling time at 23°C (min)	12	12	120	9	8	15	11	14	12	16	15
* Gel consistency	M	S	H	H	H	S	M	H	H	H	M
Swelling 7 days in water (%)	7	nd	23	184	196	40	39	88	258	46	30

^a in solid particulate form^b supported on Sipernat 33 carrier with 50 wt.-% loading

* S = soft, M = medium soft, H = hard

Table 3

	Ex 12	Ex 13	Ex 14	Ex 15	Ex 16	Ex 17	Ex 18	Ex 19	Ex 20	Ex 21	Ex 22	Ex 23	Ex 24
Ingredients													
HEMA (pbw)	45.0	42.0	42.0	42.0	42.0	48.0	48.0	48.0	40.0	21.0	21.0	16.0	45.0
SR252 (pbw)									8.0			5.0	
Sipernat 33 (pbw)	15.0												
PUR-mehl		18.0	18.0										
Expanded perlite				18.0	18.0								
Cab-O-Sil M-5						12.0	12.0	12.0	12.0				
Vinnapas 7055 N										39.0	39.0	39.0	
Vinnapas 5045 E													55.0
Carrier loading (wt.-%)	75	70	70	70	70	80	80	80	80	35	35	35	45
Water (pbw)	90.0	75.0	90.0	70.0	90.0	90.0	132.0	152.0	132.0	45.0	60.0	60.0	45.0
^a NAPS (pbw)	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5
^b TEA (pbw)	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00
Total (pbw)	151.5	136.5	151.5	131.5	151.5	151.5	193.5	213.5	193.5	106.5	121.5	121.5	146.5
Results													
Viscosity (mPa·s) (without NAPS)	9	nd	114	nd	nd	nd	34	57	26	703	183	170	nd
Gelling time at 23°C (min)	15	16	21	23	20	20	18	21	13	16	15	22	15
* Gel consistency	M	H	H	H	H	H	S	S	H	S	S	S	M
Swelling 7 days in water	7	31	26	15	14	39	8	18	6	25	nd	nd	40

^a in solid particulate form^b supported on Sipernat 33 carrier with 50 wt.-% loading

* S = soft, M = medium H = hard

Table 4

	Ex 25	Ex 26	Ex 27	Ex 28	Ex 29	Ex 30	Ex 31	Ex 32	Ex 33	Ex 34	Ex 35	Ex 36	Ex 37	Ex 38
Ingredients														
HEMA (pbw)	22.3	22.4	22.3	22.4	22.2	22.3	22.3	22.3	22.3	22.3	45.0	45.0	45.0	45.0
Sipernat 33 (pbw)	7.8	7.8	7.8	7.8	7.7	7.7	7.8	7.8	7.8	7.7	15.0	15.0	15.0	15.0
Carrier loading (wt.-%)	74	74	74	74	74	74	74	74	74	74	75	75	75	75
Water (pbw)	37.0	37.0	37.0	37.1	37.1	37.0	37.1	37.0	37.0	37.2	58.0	58.0	58.0	58.0
^a NAPS (pbw)	2.54	2.5	2.52								2.5	2.5	2.5	2.5
^b Trigonox TMBH-L				3.00	3.02	2.96	3.04							
^c Trigonox TAMP-W85								3.04	3.01	3.01				
^d TEA (pbw)	1.01										1.00	1.00	1.00	1.00
Ascorbic acid		0.24		0.26				0.53						
Bruggolite FF7			1.24		1.25				1.25					
^e Bruggolite L40							1.25			1.28				
^f Manganese 10 HS						0.65								
^g ABL						0.64								
Filler														
Axilat 8510											10.0	20.0		
Cement Microdur R-U													10.0	20.0
Total (pbw)	70.7	70.0	70.9	70.5	71.3	71.3	71.5	70.6	71.3	71.5	131.5	141.5	131.5	141.5
Results														
Gelling time at 23°C (min)	16	10	14	11	6	12	7	5	2	16	19	23	9	10
*Gel consistency	M	S	S	S	S	S	S	S	S	M	H	M	H	H

^a 20 wt.-% solution supported on Sipernat 33 carrier with 50 wt.-% loading^{b-g} supported on Sipernat 33 carrier with 50 wt.-% loading

* S = soft, M = medium, H = hard

Claims

1. A single- or multiple-component composition for producing a
5 hydrogel comprising constituents:
a) a radical polymerizable monomer or prepolymer system,
b) a free-radical initiator,
c) optionally a catalyst for said initiator,
- 10 wherein the radical polymerizable monomer or prepolymer system
comprises:
a1) at least one at room temperature liquid water-soluble
(meth)acrylic compound supported on at least one solid carrier
and/or
15 a2) at least one at room temperature solid or liquid water-soluble
(meth)acrylic compound dissolved in a liquid solution, preferably
in an aqueous solution, which liquid solution is supported on at
least one solid carrier.
- 20 2. The single- or multiple-component composition according to claim 1,
wherein the composition is a single-component powder composition,
preferably a free-flowing powder composition, wherein the
constituents a), b), and c) are present in a single component K.
- 25 3. The single- or multiple-component composition according to claim 1,
wherein the composition is a two component composition composed
of a first component K1 and a second component K2, the first
component K1 comprising at least the radical polymerizable
monomer or prepolymer system and the second component K2
30 comprising the free-radical initiator and/or the catalyst for said
initiator.

4. The single- or multiple-component composition according to any of claims 1-3, wherein the at least one solid carrier has a specific surface area measured with a BET method according to ISO EN 9277:2010 standard of 50.0 – 2000 m²/g, more preferably of 100.0 – 1500 m²/g, most preferably of 200.0 – 1000 m²/g.
5. The single- or multiple-component composition according to any of claims 1-4, wherein the at least one solid carrier is selected from the group consisting of particles of perlite, expanded perlite, glass, phonolite, calcium silicates, fumed silica, precipitated silica, silica gel, foamed polyurethane, polysaccharides, expanded vermiculite, clay minerals, fumed metal oxides, zeolites, kieselguhr, and mixtures thereof.
6. The single- or multiple-component composition according to any of claims 1-5, wherein the at least one at room temperature liquid water soluble (meth)acrylic compound is selected from the group consisting of hydroxyl-functional (meth)acrylates, carboxyl-functional (meth)acrylic compounds, low molecular weight polyether (meth)acrylates, (meth)acrylamides, and mixtures thereof, preferably from the group consisting of hydroxyethylmethacrylate (HEMA), hydroxypropylmethacrylate (HPMA), low molecular weight polyethylene glycol dimethacrylate (PEG-DMA), ethoxylated trimethylolpropane tri(meth)acrylate (TMP-TMA), and mixtures thereof.
7. The single- or multiple-component composition according to any of claims 1-6, wherein the radical polymerizable monomer or prepolymer system further comprises:
- a3) at least one at room temperature solid water-soluble (meth)acrylic compound in solid particulate form, which at least one at room temperature solid water-soluble (meth)acrylic compound is preferably selected from the group consisting of

5 magnesium diacrylate, sodium acrylate, potassium acrylate, potassium salt of 3-sulfopropylacrylate, high molecular weight PEG-dimethacrylate, 2-acrylamido-2-methylpropane sulfonic acid (AMPS), sodium salt of 2-acrylamido-2-methylpropane sulfonic acid (Na-AMPS), and mixtures thereof.

8. The single- or multiple-component composition according to any of claims 1-7, wherein sum of the water-soluble (meth)acrylic compounds contained in the constituent a) amounts to 10.0 – 95.0 %
10 by weight, preferably 12.5 – 85.0 % by weight, most preferably 15.0 – 80.0 % by weight of the total weight of the single- or multiple component composition.
9. The single- or multiple-component composition according to any of
15 claims 1-8, wherein the free-radical initiator comprises at least one at room temperature solid initiator in solid particulate form and/or at least one at room temperature solid initiator dissolved in a liquid solution, which liquid solution is supported on the at least one solid carrier, wherein said at least one at room temperature solid initiator is
20 selected from the group consisting of azobisisobutyronitrile, sodium persulfate, potassium persulfate or ammoniumpersulfate, and mixtures thereof, and/or at least one at room temperature liquid initiator supported on the at least one solid carrier, which at least one at room temperature liquid initiator is selected from the group
25 consisting of hydrogen peroxide, cumene hydroperoxide, tert-butyl hydroperoxide, diisopropylbenzene hydroperoxide, and mixtures thereof.
10. The single- or multiple-component composition according to any of
30 claims 1-9, wherein the catalyst for the initiator comprises at least one at room temperature solid catalyst in solid particulate form and/or at least one at room temperature solid catalyst dissolved in a liquid solution, which liquid solution is supported on the at least one

solid carrier, wherein said at least one at room temperature solid catalyst is selected from the group consisting of ascorbic acid, sodium formaldehyde sulfoxylate, 2-hydroxy-2-sulfinatoacetic acid and/or salt thereof, 2-hydroxy-2-sulfonatoacetic acid and/or salt thereof, toluidine derivatives, transition metal salts, transition metal complexes, and mixtures thereof, and/or at least one at room temperature liquid catalyst supported on the at least one solid carrier, which at least one at room temperature liquid catalyst is selected from the group consisting of alkanol amines and ethoxylated alkanol amines, and mixtures thereof.

11. The single- or multiple-component composition according to any of claims 1-10 further comprising:

e) at least one filler selected from the group consisting of organic fillers, inert mineral fillers, mineral binders, and mixtures thereof.

12. A method for producing a (meth)acrylic hydrogel comprising steps of

i) Providing a single- or multiple-component composition according to any of claims 1-11,

ii) Mixing the single- or multiple-component composition with water such that the weight ratio of the total amount of the water-soluble (meth)acrylic compounds to water in the mixture is in the range of 0.2 to 3,

iii) Letting the mixture form (meth)acrylic hydrogel.

13. A (meth)acrylic hydrogel obtainable by the method according to claim 12.

14. A (meth)acrylic injection material obtainable by mixing a single- or multiple-component composition according to any of claims 1-11 with water such that that the weight ratio of the total amount of the water-

soluble (meth)acrylic compounds to water in the mixture is in the range of 0.2 to 3.

15. A method for sealing and/or filling of cracks, voids, flaws and
cavities in a building structure, the method comprising steps of
- i) Providing a single- or multiple-component composition according to any of claims 1-11,
 - ii) Mixing the single- or multiple-component composition with water such that the weight ratio of the total amount of the water-soluble (meth)acrylic compounds to water in the mixture is in the range of 0.2 to 3,
 - iii) Applying the mixture to the site to be sealed/filled in the building structure and letting the mixture to form (meth)acrylic hydrogel.