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(71) Applicant: **SIKA TECHNOLOGY AG** [CH/CH]; Zugerstrasse 50, 6340 Baar (CH).

(72) Inventors: **BANDERA, Davide**; Wallisellenstrasse 4, 8050 Zürich (CH). **HOFSTETTER, David**; Tösstalstrasse 244a, 8405 Winterthur (CH). **MENNECKE, Klaas**; Joseph-Weisshaarstrasse 13, 79807 Lottstetten (CH).

(74) Agent: **SIKA PATENT ATTORNEYS**; c/o Sika Technology AG, Corp. IP Dept., Tüffenwies 16, 8048 Zürich (CH).

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(54) Title: THERMALLY CONDUCTIVE CURABLE COMPOSITION

(57) Abstract: The invention relates to multi-component curable composition, comprising - at least one organic polymer PO containing alkoxysilane groups; - at least one liquid epoxy resin EP; - at least one polyamine PA; - more than 70 wt.-%, based on the total composition, of at least one thermally conductive filler Fl; - water; - optionally additives selected from curing catalysts, accelerators, plasticizers, organosilanes, stabilizers, and colorants; wherein said liquid epoxy resin EP and said polyamine PA are not being present in the same component, and in that said organic polymer PO and said water are not present in the same component. The multi-component curable compostion shows improved mechanical properties and facile applicability alongside with excellent thermal conductivity and is particularly suitable as adhesive, gap filler, or sealant suitable for e-coat applications and battery bonding especially in electric automobile assembly.

THERMALLY CONDUCTIVE CURABLE COMPOSITION

Technical field

The invention relates to multi-component compositions which are curable at room temperature and are based on a combination of silane group-containing polymer and epoxy resin, and their use as adhesives, sealants, coatings, gap fillers or potting resins.

State of the art

Curable compositions based on polymers containing reactive groups play an important role in many industrial applications, for example as adhesives, sealants, coatings, or potting resins. Such compositions chemically cure to cross-linked plastics with a broad possible range of mechanical, chemical, and physical properties. Such chemically curing compositions can be formulated as one-component, ready-to-use compositions by employing reactive polymers that only cure when exposed to external influences, such as heat or humidity, in order to ensure sufficient storage stability inside their package prior to use. Another possibility is to use multi-component compositions (in most cases two-component compositions) that are mixed with each other right before or during application. The latter allows for more reactive systems with spontaneous, often much more rapid curing and they do not require any external influences but cure spontaneously once mixed and applied. Due to their faster and more controlled curing behavior, multi-component compositions are preferred in industrial applications, as they allow for higher process control and shorter production cycles. Also, highly skilled industrial applicators or automatic application equipment do not require the benefit of an easy, fail-safe application a one-component composition better provides.

Known and broadly used two-component curable compositions are for example two-component epoxy resin adhesives. These adhesives attain very high strengths but are normally not tough and elastic, instead being brittle and barely stretchable, and hence unsuitable for numerous applications within structural adhesive bonding or other industrial assembly operations.

Also known are curable materials based on silane-functional polymers. These materials are mostly one-component formulations which crosslink at room temperature by reaction with atmospheric moisture. They are notable for blister-free curing and for high adhesion forces but cure relatively slowly and are very limited in terms of maximum attainable strength. Oftentimes they also have low tear resistance, being therefore

decidedly brittle. Furthermore, they are relatively unstable thermally. Hence, they are poorly suited to use as tough elastic adhesive or in thermally demanding applications. Also known are curable materials based on a combination of silane-functional polymers with epoxy resins, from EP 0 186 191 and EP 0 370 464, for example. Such materials are commonly in two-component form and attain greater strength and toughness than those based on silane-functional polymers alone but are still in need of improvement in this regard. Another example of such a composition is disclosed in CN 115 926 719 A. Therein, two-component compositions based on silane-terminated polyurethane resins and epoxy resins are disclosed. More specifically, their first component contains, inter alia, 50-70 parts of silane-terminated polyurethane resin and 10-20 parts of a first flame-retardant filler, for example alumina. Their second component contains, inter alia, 40-60 parts epoxy resin and also 10-20 parts of a second flame-retardant filler, for example polyphosphate.

With the increasing use of batteries and electronic equipment nowadays, the demand for suitable adhesives, sealants, gap fillers, and potting resins that are in contact with these electronic elements or devices has risen significantly. Especially in electric automobile assembly, large batteries and a multitude of electronic parts require large amounts of such sealants and adhesives that cover, seal, or bond electronics and batteries directly. A particular requirement for these materials is a high heat conductivity, since batteries and electronics generate significant amounts of heat while operating that must be dissipated efficiently to prevent detrimental heat accumulation on the batteries and electronic parts. This aspect has proven to be a severe problem for traditionally used curable compositions, e.g., based on silane-functional polymers or epoxy resins. Commonly known such compositions possess a poor heat conductivity and rather act as thermal insulators than efficient heat conductors. There have been several attempts to formulate curable compositions with high heat conductivity. The current state of the art most commonly uses silicone-based compositions with special fillers. These marketed compositions are curable and show adequate thermal conductivity. However, silicone-based compositions are notoriously difficult to paint or coat and possibly create volatile siloxanes that could deposit on battery electronics, which is a critical problem in the rather young electric automobile industry. Silicones are especially problematic for electrodeposition coating (“e-coat” or cathode-dipping) processes, which are a common and often indispensable process step in automotive manufacturing. Many automotive manufacturers strictly try to avoid silicones in their plants to prevent interference with their e-coat processes.

So far, commercially available thermally conductive compositions are mainly limited to silicones and thus have limited applicability in automotive, especially electric vehicle, manufacturing. Other proposed curable compositions, based on organic silane-functional polymers, do not interfere with the e-coat process, but so far did not prove to be suitable because of their limited thermal conductivity. Recently, thermally conductive compositions based on silane-functional polymers having high content of thermally conductive fillers have been developed. However, they are mechanically weak due to their low polymer content and can most suitably be used as gap fillers only. Also epoxy-based such highly filled, thermally conductive compositions have been developed, but they suffer from high viscosity and intrinsic brittleness, which makes their application cumbersome and reduces their suitability as potting resin or adhesive with exposure to vibrations or other mechanical impacts.

There is thus an increasing demand for a curable composition that exhibits low viscosity and pumpability and outstanding mechanical properties and thus is suitable as adhesive, gap filler, sealant, or potting resin especially for battery and electronics assembly and that furthermore possesses high thermal conductivity but at the same time does not interfere with coating or painting processes, in particular e-coat processes.

Summary of the invention

It is therefore an object of the present invention to provide a storage-stable, low viscosity and pumpable curable composition that exhibits high thermal conductivity, in particular of at least 1.5 W/mK according to ASTM D5470, while at the same time exhibiting excellent mechanical properties, especially tensile strength, laps shear strength, and elongation at break, and that hence is suitable as gap-filler, sealant, potting resin, or adhesive for thermal management in batteries and electronic equipment.

The present invention achieves these objects with the features of independent claim 1.

Further aspects of the invention are the subject of further independent claims. Particularly preferred embodiments of the invention are the subject of the dependent claims.

Ways of executing the invention

The present invention relates in a first aspect to multi-component curable composition, comprising

- at least one organic polymer **PO** containing alkoxysilane groups;
- at least one liquid epoxy resin **EP**;
- at least one polyamine **PA**;
- more than 70 wt.-%, based on the total composition, of at least one thermally conductive filler **FI**;
- water;
- optionally additives selected from curing catalysts, accelerators, plasticizers, organosilanes, stabilizers, and colorants;

wherein said liquid epoxy resin **EP** and said polyamine **PA** are not being present in the same component, and in that said organic polymer **PO** and said water are not present in the same component.

In the present document, the term “reactive silane group” refers to a silyl group that is bonded to an organic radical and has one to three, especially two or three, hydrolyzable substituents or hydroxyl groups on the silicon atom. Particularly useful hydrolyzable substituents are alkoxy groups. These silane groups are also referred to as “alkoxysilane groups”. Reactive silane groups may also be in partly or fully hydrolyzed form, for example as silanols.

“Hydroxysilane”, “isocyanatosilane”, “aminosilane” and “mercaptosilane” refer respectively to organoalkoxysilanes having one or more hydroxyl, isocyanato, amino or mercapto groups on the organic radical in addition to the silane group.

“Organofunctional compound” refers to a compound that contains a functional group that is bound via a carbon atom. For example, “aminofunctional compound” is a compound having an aminoalkyl group.

“Primary amino group” refers to an NH_2 group that is bonded to an organic radical, and “secondary amino group” refers to an NH group that is bonded to two organic radicals which may also together be part of a ring, and “tertiary amino group” refers to an N group that is bonded to three organic radicals, two or three of which together may also be part of one or more rings. Accordingly, “primary aminosilanes” are aminosilanes comprising a primary amino group and “secondary aminosilanes” are aminosilanes comprising a secondary amino group. The latter also encompasses compounds having both a primary and a secondary amino group.

"Polyoxyalkylene radical" refers to a linear or branched hydrocarbyl radical which contains ether groups and contains more than two repeat units of the (O-R) type in succession, where R is a linear or branched alkylene radical, as for example from the polyaddition of ethylene oxide or 1,2-propylene oxide onto starter molecules having two active hydrogen atoms.

Substance names beginning with "poly", such as polyol or polyisocyanate, refer to substances containing, in a formal sense, two or more of the functional groups that occur in their name per molecule.

The term "organic polymer" encompasses a collective of macromolecules that are chemically homogeneous but differ in relation to degree of polymerization, molar mass and chain length, which has been prepared by a poly reaction (polymerization, polyaddition, polycondensation) and has a majority of carbon atoms in the polymer backbone, and reaction products of such a collective of macromolecules. Polymers having a polyorganosiloxane backbone (commonly referred to as "silicones") are not organic polymers in the context of the present document.

The term "polyether containing reactive silane groups" also encompasses organic polymers which contain silane groups and which, in addition to polyether units, may also contain urethane groups, urea groups or thiourethane groups. Such polyethers containing reactive silane groups may also be referred to as "polyurethanes containing reactive silane groups".

"Molecular weight" is understood in the present document to mean the molar mass (in grams per mole) of a molecule or part of a molecule, also referred to as "radical". The term "radical" is used in this document in a formal sense, meaning a molecular rest bound to an atom by a covalent bond, while the bond is formally "cut" to describe the molecular rest attached to it. Molecular weight of polymers is understood as the average molecular weight of their chain length distribution. "Average molecular weight" is understood herein to mean the number-average M_n of an oligomeric or polymeric mixture of molecules or radicals, which is typically determined by means of gel permeation chromatography (GPC) against polystyrene as standard.

"Weight percent" or "percentage by weight", and its abbreviation "wt.-%" refer to the weight percentage of a certain compound in a total composition, if not otherwise defined. The terms "weight" and "mass" are used interchangeably in this document and refer to the mass as a property of a physical body and commonly measured in kilograms (kg).

"Storage-stable" or "storable" refers to a substance or composition when it can be stored at room temperature in a suitable container over a prolonged period, typically at least 3 months up to 6 months or more, without any change in its application or use properties, especially in the viscosity and crosslinking rate, to a degree of relevance for the use thereof as a result of the storage.

"Room temperature" refers to a temperature of 23°C.

"Thermal conductivity" is defined as ability of material to transmit heat and it is measured in watts per metre-kelvin (W/(m·K)), herein denoted as "W/mK". Thermal conductivity of materials disclosed in this document are measured according to ASTM D5470-06, if not otherwise specified.

All industrial standards and norms cited in this document refer to the respective edition in force on the time of filing of the first application of this invention, if not otherwise defined.

A dotted line in the formulae in this document in each case represents the bond between a substituent and the corresponding molecular radical.

The term "does not contain polydiorganosiloxanes" means that no such compounds have been added during formulation of the composition. If traces of such species are unknowingly and/or unavoidably present, for example stemming from the synthesis of the silane-functional polymer or due to condensation reactions of silane-functional diorganosilane compounds possibly present in the composition, these are not considered as polydiorganosiloxanes in the meaning of this term. In simpler words, the term "does not contain polydiorganosiloxanes" means that no silicone oils or reactive silicone polymers, in particular polydimethylsiloxanes, were added during formulation of the composition.

<u>Organic polymer **PO** containing alkoxysilane groups</u>

The composition according to the present invention comprises at least one organic polymer **PO** containing alkoxysilane groups.

In preferred embodiments, the composition comprises composition comprises between 3 wt.-% and 10 wt.-%, preferably between 3.5 wt.-% and 7.5 wt.-%, in particular between 4 wt.-% and 6 wt.-%, based on the total composition, of said organic polymer **PO**.

The organic polymer **PO** containing alkoxysilane groups is in particular a polyurethane, polyolefin, polyester, polycarbonate, polyamide, poly(meth)acrylate or polyether or a mixed form of these polymers, each of which bears one or preferably more than one reactive alkoxysilane group. The alkoxysilane groups may be in pendant positions in the chain or in terminal positions and are bonded to the organic polymer via a carbon atom.
More preferably, the organic polymer **PO** containing alkoxysilane groups is a polyolefin containing alkoxysilane groups or a polyurethane containing alkoxysilane groups or a polyether containing alkoxysilane groups or a mixed form of these polymers.
Most preferably, the organic polymer containing reactive silane groups is a polyether containing alkoxysilane groups.

The organic polymer **PO** containing alkoxysilane groups preferably has an average of 1.3 to 4, especially 1.5 to 3, more preferably 1.7 to 2.8, alkoxysilane groups per molecule. The silane groups are preferably terminal.
The organic polymer **PO** containing alkxysilane groups preferably has an average molecular weight, determined by means of GPC against a polystyrene standard, in the range from 1'000 to 30'000 g/mol, especially from 2'000 to 20'000 g/mol. The organic polymer **PO** containing alkoxysilane groups preferably has a silane equivalent weight of 300 to 25'000 g/eq, especially of 500 to 15’000 g/eq.

The organic polymer **PO** may be solid or liquid at room temperature. It is preferably liquid at room temperature.
Most preferably, the organic polymer **PO** containing alkoxysilane groups is liquid at room temperature, wherein the silane groups are especially dialkoxysilane groups and/or trialkoxysilane groups, more preferably trimethoxysilane groups, methyldimethoxysilane groups, or triethoxysilane groups.

Processes for preparing organic polymers **PO** containing alkoxysilane groups are known to the person skilled in the art.

In a preferred process, organic polymers containing alkoxysilane groups are obtainable from the reaction of organic polymers containing allyl groups with hydrosilanes, optionally with chain extension using, for example, diisocyanates.

In a further preferred process, polyethers containing alkoxysilane groups are obtainable from the copolymerization of alkylene oxides and epoxysilanes, optionally with chain extension using, for example, diisocyanates.

In a further preferred process, organic polymers containing reactive silane groups are obtainable from the reaction of organic polyols with isocyanatosilanes, optionally with chain extension using diisocyanates.

In a further preferred process, polyethers containing alkoxysilane groups are obtainable from the reaction of organic polymers containing isocyanate groups, especially NCO-terminated urethane polymers from the reaction of polyols with a superstoichiometric amount of polyisocyanates, with aminosilanes, hydroxysilanes or mercaptosilanes. This process enables the use of a multitude of inexpensive starting materials of good commercial availability, by means of which it is possible to obtain different polymer properties, for example high extensibility, high strength, low modulus of elasticity, low glass transition point or high weathering resistance.

In some preferred embodiments, the organic polymer **PO** containing alkoxysilane groups is obtainable from the reaction of NCO-terminated urethane polyethers with aminosilanes, mercaptosilanes, or hydroxysilanes. Suitable NCO-terminated urethane polymers are obtainable from the reaction of polyols, especially polyether polyols, in particular polyoxyalkylenediols or polyoxyalkylenetriols, preferably polyoxypropylenediols or polyoxypropylenetriols, with a superstoichiometric amount of polyisocyanates, especially diisocyanates. Also, other polyols, such as poly(meth)acrylate polyols, polyhydrocarbon polyols, in particular polybutadiene polyols, polyhdroxy functional fats or oils, polycarbonate polyols, polyester polyols and polyhydroxy functional acrylonitrilie/butadiene copolymers are suitable. Furthermore, small amounts of low molecular weight dihydric or polyhydric alcohols, such as diols, glycols, and sugar alcohols may be used as additives.

Preferably, the reaction between the polyisocyanate and the polyol is conducted with exclusion of moisture at a temperature of 50°C to 160°C, optionally in the presence of suitable catalysts, with metered addition of the polyisocyanate in such a way that the isocyanate groups thereof are present in a stoichiometric excess in relation to the hydroxyl groups of the polyol. More particularly, the excess of polyisocyanate is chosen such that a content of free isocyanate groups of 0.1% to 5% by weight, preferably 0.2%

to 4% by weight, more preferably 0.3% to 3% by weight, based on the overall polymer, remains in the resulting urethane polymer after the reaction of all hydroxyl groups. Preferred diisocyanates are selected from the group consisting of hexamethylene 1,6-diisocyanate (HDI), 1-isocyanato-3,3,5-trimethyl-5-isocyanatomethylcyclohexane (= isophorone diisocyanate or IPDI), tolylene 2,4- and 2,6-diisocyanate and any desired mixtures of these isomers (TDI) and diphenylmethane 4,4'-, 2,4'- and 2,2'-diisocyanate and any desired mixtures of these isomers (MDI). Particular preference is given to IPDI or TDI. Most preferred is IPDI. In this way, polyethers containing reactive silane groups with particularly good lightfastness are obtained.

Especially suitable as polyether polyols are polyoxyalkylenediols or polyoxyalkylenetriols having a degree of unsaturation lower than 0.02 meq/g, especially lower than 0.01 meq/g, and a mean molecular weight in the range from 400 to 25'000 g/mol, especially 1000 to 20'000 g/mol.

As well as polyether polyols, it is also possible to use portions of other polyols, especially polyacrylate polyols, and low molecular weight diols or triols.

Suitable aminosilanes for the reaction with an NCO-terminated urethane polyether are primary and secondary aminosilanes. Preference is given to 3-aminopropyltrimethoxysilane, 3-aminopropyldimethoxymethylsilane, 4-aminobutyltrimethoxysilane, 4-amino-3-methylbutyltrimethoxysilane, 4-amino-3,3-dimethylbutyltrimethoxysilane, N-butyl-3-aminopropyltrimethoxysilane, N-phenyl-3-aminopropyltrimethoxysilane, adducts formed from primary amino-silanes such as 3-aminopropyltrimethoxysilane, 3-aminopropyldimethoxy-methylsilane or N-(2-aminoethyl)-3-aminopropyltrimethoxysilane and Michael acceptors such as acrylonitrile, (meth)acrylic esters, (meth)acrylamides, maleic or fumaric diesters, citraconic diesters or itaconic diesters, especially dimethyl or diethyl N-(3-trimethoxysilylpropyl)aminosuccinate. Likewise suitable are analogs of the aminosilanes mentioned with ethoxy or isopropoxy groups in place of the methoxy groups bonded to the silicon atom.

Suitable hydroxysilanes for the reaction with an NCO-terminated urethane polyether are especially obtainable from the addition of aminosilanes onto lactones or onto cyclic carbonates or onto lactides.

Aminosilanes suitable for the purpose are especially 3-aminopropyltrimeth-oxysilane, 3-aminopropyltriethoxysilane, 4-aminobutyltrimethoxysilane, 4-aminobutyltriethoxysilane, 4-amino-3-methylbutyltrimethoxysilane, 4-amino-3-methylbutyltriethoxysilane, 4-amino-3,3-dimethylbutyltrimethoxysilane, 4-amino-3,3-dimethylbutyltriethoxysilane, 2-aminoethyltrimethoxysilane or 2-aminoethyltriethoxysilane. Particular preference is given to 3-aminopropyl-trimethoxysilane, 3-aminopropyltriethoxysilane, 4-amino-3,3-dimethylbutyl-trimethoxysilane or 4-amino-3,3-dimethylbutyltriethoxysilane.

Suitable lactones are especially γ-valerolactone, γ-octalactone, δ-decalactone, and ε-decalactone, especially γ-valerolactone.

Suitable cyclic carbonates are especially 4,5-dimethyl-1,3-dioxolan-2-one, 4,4-dimethyl-1,3-dioxolan-2-one, 4-ethyl-1,3-dioxolan-2-one, 4-methyl-1,3-dioxolan-2-one or 4-(phenoxymethyl)-1,3-dioxolan-2-one.

Suitable lactides are especially 1,4-dioxane-2,5-dione (lactide formed from 2-hydroxyacetic acid, also called “glycolide”), 3,6-dimethyl-1,4-dioxane-2,5-dione (lactide formed from lactic acid, also called “lactide”) and 3,6-diphenyl-1,4-dioxane-2,5-dione (lactide formed from mandelic acid).

Preferred hydroxysilanes which are obtained in this way are N-(3-triethoxysilylpropyl)-2-hydroxypropanamide, N-(3-trimethoxysilylpropyl)-2-hydroxypropanamide, N-(3-triethoxysilylpropyl)-4-hydroxypentanamide, N-(3-triethoxysilylpropyl)-4-hydroxyoctanamide, N-(3-triethoxysilylpropyl)-5-hydroxydecanamide and N-(3-triethoxysilylpropyl)-2-hydroxypropyl carbamate.

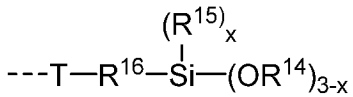
In addition, suitable hydroxysilanes are also obtainable from the addition of aminosilanes onto epoxides or from the addition of amines onto epoxysilanes.

Preferred hydroxysilanes which are obtained in this way are 2-morpholino-4(5)-(2-trimethoxysilylethyl)cyclohexan-1-ol, 2-morpholino-4(5)-(2-triethoxysilyl-ethyl)cyclohexan-1-ol or 1-morpholino-3-(3-(triethoxysilyl)propoxy)propan-2-ol.

Further suitable polyethers containing reactive silane groups are commercially available products, especially the following: MS Polymer™ (from Kaneka Corp.; especially the S203H, S303H, S227, S810, MA903 and S943 products); MS Polymer™ or Silyl™ (from Kaneka Corp.; especially the SAT010, SAT030, SAT200, SAX350, SAX400, SAX725, MAX450, MAX951 products); Excestar® (from Asahi Glass Co. Ltd.; especially the S2410, S2420, S3430, S3630 products); SPUR+* (from Momentive Performance Materials; especially the 1010LM, 1015LM, 1050MM products); Vorasil™ (from Dow

Chemical Co.; especially the 602 and 604 products); Desmoseal® (from Bayer MaterialScience AG; especially the S XP 2458, S XP 2636, S XP 2749, S XP 2774 and S XP 2821 products), TEGOPAC® (from Evonik Industries AG; especially the Seal 100, Bond 150, Bond 250 products), Polymer ST (from Hanse Chemie AG/Evonik Industries AG, especially the 47, 48, 61, 61LV, 77, 80, 81 products); Geniosil® (from Wacker Chemie AG; especially the STP-E10, STP-E15, STP-E30, STP-E35, WP1, and WP2 products). Furthermore, suitable is the silylated polyether available under the trade name Si-PolyU 5013 (from PolyU GmbH).

Particularly preferred organic polymers **PO** containing alkoxysilane groups have end groups of the formula (VIII)



(VIII)

where

R^{16} is a linear or branched divalent hydrocarbyl radical which has 1 to 12 carbon atoms and optionally has cyclic and/or aromatic moieties and optionally one or more heteroatoms, especially one or more nitrogen atoms;

T is a divalent radical selected from –O–, –S–, –N(R^{17})–, –O–CO–N(R^{17})–, –N(R^{17})–CO–O– and –N(R^{17})–CO–N(R^{17})–,

where R^{17} is a hydrogen radical or a linear or branched hydrocarbyl radical which has 1 to 20 carbon atoms and optionally has cyclic moieties, and which optionally has an alkoxysilane, ether or carboxylic ester group; and

R^{14}, R^{15} and x have the definitions already given.

Preferably, R^{16} is methylene, 1,3-propylene or 1,4-butylene, where butylene may be substituted by one or two methyl groups.

In an especially preferred embodiment, R^{16} is a methylene radical;

Preferably, T is a divalent radical selected from –O–, –S–, –N(R^{17})–, –O–CO–N(R^{17})–, –N(R^{17})–CO–O– and –N(R^{17})–CO–N(R^{17})–,

where R^{17} is a hydrogen radical or a linear or branched hydrocarbyl radical which has 1 to 20 carbon atoms and optionally has cyclic moieties;

R^{14} is in particular a methyl or ethyl radical;

R^{15} is preferably a methyl radical; and

x is preferably 0 or 1.

Liquid epoxy resin **EP**

The multi-component composition further comprises at least one liquid epoxy resin **EP**. Suitable liquid epoxy resin **EP** comprises customary technical epoxy resins which are fluid at room temperature and have a glass transition temperature of below 25°C. They are obtained conventionally, more particularly from the glycidylization of compounds having at least two active hydrogen atoms, more particularly polyphenols, polyols or amines, by reaction with epichlorohydrin.

Suitability as liquid epoxy resin **EP** are in particular aliphatic or cycloaliphatic epoxy resins, more particularly

- glycidyl ethers of saturated or unsaturated, branched or unbranched, cyclic or open-chain di-, tri- or tetra-functional C_2 to C_{30} alcohols, more particularly ethylene glycol, propylene glycol, butylene glycol, hexanediol, octanediol, polypropylene glycols, dimethylolcyclohexane, neopentyl glycol, dibromoneopentyl glycol, castor oil, trimethylolpropane, trimethylolethane, pentaerythritol, sorbitol or glycerol, or alkoxylated glycerol or alkoxylated trimethylolpropane;
- glycidyl ethers of hydrogenated bisphenol A, F or A/F, or ring-hydrogenated liquid bisphenol A, F or A/F resins;
- N-glycidyl derivatives of hydantoins, amides or heterocyclic nitrogen bases, such as, in particular, triglycidyl cyanurate or triglycidyl isocyanurate.

Preferred as liquid epoxy resin **EP** are aromatic epoxy resins, more particularly

- glycidyl ethers of polyphenols, more particularly of resorcinol, hydroquinone, pyrocatechol, 2,2-bis(4-hydroxyphenyl)propane (bisphenol A), bis(hydroxyphenyl)methane (bisphenol F), bisphenol A/F, bis(4-hydroxy-3-methylphenyl)methane, 2,2-bis(4-hydroxy-3-methylphenyl)propane (bisphenol C), bis(3,5-dimethyl-4-hydroxyphenyl)methane, 2,2-bis(3,5-dimethyl-4-hydroxyphenyl)propane, 2,2-bis(3,5-dibromo-4-hydroxyphenyl)propane, 2,2-bis(4-hydroxy-3-tert-butylphenyl)propane, 2,2-bis(4-hydroxyphenyl)butane (bisphenol B), 3,3-bis(4-hydroxyphenyl)pentane, 3,4-bis(4-hydroxyphenyl)hexane, 4,4-bis(4-hydroxyphenyl)heptane, 2,4-bis(4-hydroxyphenyl)-2-methylbutane, 2,4-bis(3,5-dimethyl-4-hydroxyphenyl)-2-methylbutane, 1,1-bis(4-hydroxyphenyl)cyclohexane (bisphenol Z)) 1,1-bis(4-hydroxyphenyl)-3,3,5-trimethylcyclohexane (bisphenol TMC), 1,1-bis(4-hydroxyphenyl)-1-phenylethane, 1,4-bis[2-(4-hydroxyphenyl)-2-propyl]benzene (bisphenol P), 1,3-bis[2-(4-hydroxyphenyl)-2-propyl]benzene

(bisphenol M), 4,4'-dihydroxybiphenyl (DOD), 4,4'-dihydroxybenzophenone, bis(2-hydroxynaphth-1-yl)methane, bis(4-hydroxynaphth-1-yl)methane, 1,5-dihydroxynaphthalene, tris(4-hydroxyphenyl)methane, 1,1,2,2-tetrakis(4-hydroxyphenyl)ethane, bis(4-hydroxyphenyl) ether or bis(4-hydroxyphenyl) sulfone;

- glycidyl ethers of condensation products of phenols with aldehydes, obtained under acidic conditions, more particularly glycidyl ethers of phenol-formaldehyde novolacs or cresol-formaldehyde novolacs;
- glycidylization products of aromatic amines, more particularly of aniline, toluidine, 4-aminophenol, 4,4'-methylenediphenyldiamine, 4,4'-methylenediphenyldi(N-methyl)amine, 4,4'-[1,4-phenylenebis(1-methylethylidene)]bisaniline (bisaniline P) or 4,4'-[1,3-phenylenebis(1-methylethylidene)]bisaniline (bisaniline M).

Particularly preferred as liquid epoxy resin **EP** are diglycidyl ethers of bisphenol A or bisphenol F or bisphenol A/F, as are available commercially, for example, from Dow, Huntsman or Momentive. These liquid epoxy resins have readily manageable viscosity and allow high strengths and resistance properties.

The composition preferably comprises between 2 wt.-% and 10 wt.-%, preferably between 2.5 wt.-% and 7.5 wt.-%, in particular between 3 wt.-% and 6 wt.-%, based on the total composition, of said liquid epoxy resin **EP**.

A composition of this kind exhibits high strength in conjunction with good stretchability and tough elastic properties. Particularly surprising here is the circumstance that such materials display good stretchability even in combination with very high strengths.

Polyamine **PA**

The multi-component composition further comprises at least one polyamine **PA**. This polyamine **PA** serves as hardener for epoxy resin **EP** and should be suitable for this purpose. Thus, polyamine **PA** should contain at least 2 amine hydrogens that can react with epoxy resins in order to be suitable as hardener for epoxy resins. Amine hydrogens are part of primary and secondary amino groups. Polyamines containing exclusively tertiary amino groups and thus no amine hydrogens are hence not suitable as polyamine **PA**.

Suitable as polyamine **PA** are in particular compounds or polymers with at least two amine hydrogens, including at least two primary and/or secondary amino groups. Preferably, polyamine **PA** has at least one primary amino group, more preferably at least two primary amino groups.

Examples of suitable polyamines **PA** include, for example

– aliphatic, cycloaliphatic or arylaliphatic primary diamines,

e.g., ethylenediamine, 1,2-propanediamine, 1,3-propanediamine, 2-methyl-1,2-propanediamine, 2,2-dimethyl-1,3-propanediamine, 1,3-butanediamine, 1,4-butanediamine, 1,3-pentanediamine (DAMP), 1,5-pentanediamine, 1,5-diamino-2-methylpentane (MPMD), 2-butyl-2-ethyl-1,5-pentanediamine (C11-neodiamine), 1,6-hexanediamine, 2,5-dimethyl-1,6-hexanediamine, 2,2,4- and 2,4,4-trimethylhexamethylenediamine (TMD), 1,7-heptanediamine, 1,8-octanediamine, 1,9-nonanediamine, 1,10-decanediamine, 1,11-ecanediamine, 1,12-dodecanediamine, 1,2-, 1,3- and 1,4-diaminocyclohexane, bis-(4-aminocyclohexyl)methane (H_{12}-MDA), bis-(4-amino-3-methylcyclohexyl)methane, bis-(4-amino-3-ethylcyclohexyl)methane, bis-(4-amino-3,5-dimethylcyclohexyl)methane, bis-(4-amino-3-ethyl-5-methylcyclohexyl)methane (M-MECA), 1-amino-3-aminomethyl-3,5,5-trimethylcyclohexane (= isophoronediamine or IPDA), 2- and 4-methyl-1,3-diaminocyclohexane and mixtures thereof, 1,3- and 1,4-bis-(aminomethyl)cyclohexane, 2,5(2,6)-bis-(aminomethyl)-bicyclo[2.2.1]heptane (NBDA), 3(4),8(9)-bis-(aminomethyl)-tricyclo[5.2.1.$0^{2,6}$]decane, 1,4-diamino-2,2,6-trimethylcyclohexane (TMCDA), 1,8-menthanediamine, 3,9-bis-(3-aminopropyl)-2,4,8,10-tetraoxaspiro[5.5]undecane and 1,3- and 1,4-xylylenediamine;

– aliphatic primary diamines containing ether groups,

e.g., bis(2-aminoethyl)ether, 3,6-dioxaoctane-1,8-diamine, 4,7-dioxadecane-1,10-diamine, 4,7-dioxadecane-2,9-diamine, 4,9-dioxadodecane-1,12-diamine, 5,8-dioxadodecane-3,10-diamine, 4,7,10-tgrioxatridecane-1,13-diamine and higher oligomers of these diamines, bis-(3-aminopropyl)polytetrahydrofuranes and other polytetrahydrofuranediamines having molecular weights ranging, e.g., from 350 to 2000, as well as polyoxyalkylenediamines. Typically, the latter are products of the amination of polyoxyalkylene diols and can, for example, be obtained under the name Jeffamine® (from Huntsman), under the name Polyetheramin (from BASF) or under the name PC Amine® (from Nitroil). Particularly suitable polyoxyalkylenediamines are Jeffamine® D-230, Jeffamine® D-400, Jeffamine® D-2000, Jeffamine® XTJ-511, Jeffamine® ED-600, Jeffamine® ED-900, Jeffamine® ED-2003, Jeffamine® XTJ-568, Jeffamine® XTJ-569,

Jeffamine® XTJ-523, Jeffamine® XTJ-536, Jeffamine® XTJ-542, Jeffamine® XTJ-559, Jeffamine® EDR-104, Jeffamine® EDR-148, Jeffamine® EDR-176; Polyetheramin D 230, Polyetheramin D 400, and Polyetheramin D 2000, PC Amine® DA 250, PC Amine® DA 400, PC Amine® DA 650, and PC Amine® DA 2000;

– Polyamines having secondary amino groups,

e.g., diethylenetriamine (DETA), dipropylenetriamine (DPTA), bishexamethylenetriamine (BHMT), 3-(2-aminoethyl)aminopropylamine, N3-(3-aminopentyl)-1,3-pentanediamine, N5-(3-aminopropyl)-2-methyl-1,5-pentanediamine, N5-(3-amino-1-ethylpropyl)-2-methyl-1,5-pentanediamine, N,N'-dibutylethylenediamine; N,N'-di-tert.butyl-ethylenediamine, N,N'-diethyl-1,6-hexanediamine, 1-(1-methylethylamino)-3-(1-methylethylaminomethyl)-3,5,5-trimethylcyclohexane (Jefflink® 754 from Huntsman), N4-cyclohexyl-2-methyl-N2-(2-methylpropyl)-2,4-pentanediamine, N,N'-dialkyl-1,3-xylylenediamine, bis-(4-(N-alkylamino)cyclohexyl)methane, 4,4'-trimethylenedipiperidine, N-alkylated polyetheramines, e.g., the Jeffamine® types SD-231, SD-401, SD-404, and SD-2001 (from Huntsman);

– amine/polyepoxide addition products,

in particular additions products of the mentioned polyamines with diepoxides with a molar ratio of at least 2/1, in particular with a molar ration from 2/1 to 6/1;

– Polyamidoamines,

which are the reaction products of a mono- or polybasic carboxylic acid or the esters or anhydrides thereof, in particular the reaction products of a dimer fatty acid, and a aliphatic, cycloaliphatic or aromatic polyamine used in a stoichiometric excess, in particular a polyalkyleneamine such as, e.g., DETA or triethylenetetramine (TETA), in particular the commercially available polyamidoamines Versamid® 100, 125, 140, and 150 (from Cognis), Aradur® 223, 250, and 848 (from Huntsman), Euretek® 3607, Euretek® 530 (from Huntsman), Beckopox® EH 651, EH 654, EH 655, EH 661, and EH 663 (from Cytec);

– Polyethyleneimines (PEI).

These are branched polymeric amines derived from the polymerization of ethyleneimine. A suitable polyethyleneimine typically has an average molecular weight in the range from 250 to 25,000 g/mol and contains tertiary, secondary, and primary amino groups. Polyethyleneimines can be obtained, for example, under the trade name Lupasol® (from BASF), for example, the types Lupasol® FG, Lupasol® G20, and Lupasol® PR 8515.

– Cashew nutshell based amines

These are reaction products from cardanol, the main component of cashew nutshell liquid (CNSL) and amines, leading to phenalkamine structures. These are rewewable raw materials with very good properties for use as polyamine **PA**.

Preferably, polyamine **PA** contains at least two primary amino groups. Among those, especially preferred are aliphatic and cycloalphatic polyamines, in particular diamines, and polyetheramines, in particular polyether diamines.

In preferred embodiments, polyamine **PA** comprises or consists of at least one polyetheramine.

Suitable polyetheramines are polyoxyalkylenes or polyoxyalkylated compounds having terminal amino groups, of the kind available commercially, for example, under the tradenames Jeffamine® (from Huntsman), Polyetheramine (from BASF) or PC Amine® (from Nitroil), more particularly the following:

- polyetherdiamines having terminal 2-aminopropyl or 2-aminobutyl groups, more particularly Jeffamine® D-230, Jeffamine® D-400 or Jeffamine® D-2000, Jeffamine® D-4000, Jeffamine® XTJ-582, Jeffamine® XTJ-578, Jeffamine® HK-511, Jeffamine® ED-600, Jeffamine® ED-900, Jeffamine® ED-2003, Jeffamine® XTJ-568, Jeffamine® XTJ-569, Jeffamine® THF-100, Jeffamine® THF-140, Jeffamine® THF-230, Jeffamine® XTJ-533 or Jeffamine® XTJ-536 (all from Huntsman).
- Polyetherdiamines having terminal 4-aminobutyl groups from the amination of poly(tetramethylene ether) glycols, more particularly Jeffamine® THF-170 (from Huntsman).
- Polyetherdiamines from the polyalkoxylation of diols, more particularly propoxylated 1,4-dimethylolcyclohexane such as Jeffamine® RFD-270 (from Huntsman).
- Polyethertriamines, more particularly Jeffamine® T-403, Jeffamine® T-3000, Jeffamine® T-5000 or Jeffamine® XTJ-566 (all from Huntsman).
- Polyetheramines having secondary amino groups, more particularly Jeffamine® SD-231, Jeffamine® SD-401, Jeffamine® SD-2001 or Jeffamine® ST-404 (all from Huntsman).
- Aminopropylated polyetheramines, as obtainable by reaction of polyetheramines with acrylonitrile and subsequent hydrogenation.

The polyetheramine used as polyamine **PA** preferably has an average molecular weight in the range from 200 to 500 g/mol.

Particularly preferred polyetheramines are polyetherdiamines or –triamines having primary amino groups and having an average molecular weight in the range from 200 to 500 g/mol, more particularly Jeffamine® D-230, Jeffamine® D-400 Jeffamine® XTJ-582, Jeffamine® HK-511, Jeffamine® XTJ-568, Jeffamine® T-403 or Jeffamine® XTJ-566 (all from Huntsman), or corresponding grades from BASF or from Nitroil.
Most preferred are Jeffamine® D-230 or Jeffamine® D-400 or Jeffamine® T-403 (all from Huntsman), or corresponding grades from BASF or from Nitroil.
Particularly high strengths are obtained with the preferred polyetheramines.

Preferably, the multi-component composition comprises comprises between 0.2 wt.-% and 2.5 wt.-%, preferably between 0.25 wt.-% and 2.0 wt.-%, in particular between 0.5 wt.-% and 1.5 wt.-%, based on the total composition, of said polyamine **PA**.
A composition of this kind exhibits high strength in conjunction with good stretchability and tough elastic properties.

Water

The multi-component curable composition according to present invention comprises water. Water should however not be present in the same component as moisture-reactive ingredients, such as polymer **PO**, in sufficient amounts to initiate pre-mature cross-linking or curing of these ingredients within the sealaed component before mixing with the other components.

The addition of water, especially within a component of the multicomponent composition that does not contain polymer **PO**, accelerates the curing of the mixed two-component composition without the need to rely on moisture from air. This is especially advantageous in cases where a fast curing of the composition is required, or in cases where diffusion of moisture into the composition is hindered, such as narrow gaps with only a small portion of the applied composition exposed to air. However, especially in cases where non-dried fillers **FI** are used, the naturally present amount of adsorbed water in those fillers may be sufficient for curing of the low amounts of polymer **PO** used. Addition of free water is thus preferred in cases where an especially fast curing is intended or in cases where the naturally present content of free or chemically accessible water within the composition (e.g., absorbed on fillers **FI**) is too low to effect curing of the polymers **PO**.

Water may be added in free form (as liquid water), or in otherwise chemically accessible form. This is not particularly limited, as long as the water remains accessible and chemically active to react with moisture-reactive ingredient of the composition after mixing of the components of the multi-component curable composition. This may also be the case when the used fillers **FI** contain significant amounts of chemically accessible (in particular adsorbed) water, which might in such cases be sufficient for enabling proper curing of the polymers **PO**. Nevertheless, in order to ensure a sufficient availability of water, It is preferred that water is added in free form. However, as water is often poorly miscible with some other ingredients of the multi-component curable composition (due to differences in polarity), it might be advantageous to add water as a mixture with a suitable emulsifier. Such suitable emulsifiers include common surfactants, for example ethoxylated linear fatty alcohols. Suitable such emulsifiers are for example available under the trade name Disponil® by BASF.

The multi-component curable composition preferably comprises between 0.1 wt.-% and 2.0 wt.-%, in particular between 0.1 wt.-% and 1.5 wt.-%, preferably between 0.15 wt.-% and 1.0 wt.-%, particularly between 0.15 wt.-% and 0.5 wt.-%, preferably between 0.2 wt.-% and 0.4 wt.-%, in particular between 0.25 wt.-% and 0.35 wt.-%, based on the total composition, of water in liquid or chemically accessible form.

Accelerator **AC**

The multi-component curable composition optionally but preferably comprises an accelerator **AC** for accelerating the reaction between liquid epoxy resin **EP** and polyamine **PA**. Inclusion of an accelerator **AC** has the advantage that the composition cures faster after mixing of its components, especially at lower temperatures (e.g., room temperature or below).

Suitable accelerators **AC** are substances which accelerate the reaction between amino groups and epoxide groups, in particular acids or compounds hydrolyzable to acids, in particular organic carboxylic acids such as acetic acid, benzoic acid, salicylic acid, 2-nitrobenzoic acid, lactic acid, organic sulfonic acids such as methanesulfonic acid, p-toluenesulfonic acid or 4-dodecylbenzenesulfonic acid, sulfonic acid esters, other organic or inorganic acids such as in particular phosphoric acid, or mixtures of the abovementioned acids and acid esters; Tertiary amines such as in particular 1,4-

diazabicyclo [2.2.2] octane, triethanolamine, imidazoles such as in particular N-methylimidazole, N-vinylimidazole or 1,2-dimethylimidazole, salts of such tertiary amines, quaternary ammonium salts, in particular benzyltrimethylammonium chloride, amidines, in particular 1,8-diazabicyclo[5.4.0]undec-7-enes, guanidines, in particular 1,1,3,3-tetramethylguanidine, phenols, in particular bisphenols, phenol-resins or Mannich bases such as in particular 2,4,6-tris(dimethylaminomethyl) phenol or 2,4,6-tris (N, N-dimethyl-4-amino-2-azabutyl)phenol, phosphites such as in particular di- or triphenyl phosphites, or mercapto-containing compounds. Preferred as accelerators are acids, tertiary amines or Mannich bases.

Most preferred as accelerator **AC** among those is salicylic acid or 2,4,6-tris(dimethylaminomethyl)phenol or 2,4,6-tris(N,N-dimethyl-4-amino-2-azabutyl) phenol or a combination thereof.

The multi-component curable composition thus preferably comprises an accelerator **AC** for the reaction between epoxides and amines, in particular with an amount of between 0.1 wt.-% and 1.5 wt.-%, preferably between 0.2 wt.-% and 1.0 wt.-%, more preferably between 0.25 wt.-% and 0.75 wt.-% of said accelerator **AC** based on the total composition.

Accelerator **AC** is preferably included in a different component than liquid epoxy resin **EP**.

Catalyst **CA**

The multi-component curable composition optionally but preferably comprises at least one catalyst **CA** for the curing of silane-functional polymers. The addition of catalyst **CA** is advantageous to ensure a sufficient curing rate given the fact that low amounts of reactive polymers are used. However, a catalyst is not in every case necessary. In particular when using a highly reactive polymer **PO**, for example one with a methylene group between the reactive silane group and the organic linker group (R^{16} = methylene in Formula (VIII)), a catalyst may be omitted.

Suitable catalysts are especially metal compounds and/or basic nitrogen or phosphorus compounds.

Suitable metal compounds are especially compounds of tin, titanium, zirconium, aluminum or zinc, especially diorganotin(IV) compounds such as, in particular,

dibutyltin(IV) diacetate, dibutyltin(IV) dilaurate, dibutyltin(IV) dineodecanoate or dibutyltin(IV) bis(acetylacetonate) and dioctyltin(IV) dilaurate, and also titanium(IV) or zirconium(IV) or aluminum(III) or zinc(II) complexes, especially with alkoxy, carboxylate, 1,3-diketonate, 1,3-ketoesterate or 1,3-ketoamidate ligands.

Suitable basic nitrogen or phosphorus compounds are especially imidazoles, pyridines, phosphazene bases, secondary or tertiary amines, hexahydrotriazines, biguanides, guanidines, or amidines.

Nitrogen-containing compounds suitable as catalysts **CA** are in particular amines, especially N-ethyl-diisopropylamine, N,N,N',N'-tetramethylalkylenediamines, 1,4-diazabicyclo[2.2.2]octane; amidines such as especially 1,8-diazabicyclo[5.4.0]undec-7-ene (DBU), 1,5-diazabicyclo[4.3.0]non-5-ene (DBN), 6-dibutylamino1,8-diazabicyclo-[5.4.0]undec-7-ene; guanidines such as especially tetramethylguanidine, 2-guanidino-benzimidazole, acetylacetone-guanidine, 3-di-o-tolyl-guanidine, 2-tert-butyl-1,1,3,3-tetramethyl guanidine; biguanides such as biguanide, 1-butylbiguanide, 1,1-dimethylbiguanide, 1-butylbiguanide, 1-phenylbiguanide or 1-(o-tolyl)biguanide, and imidazoles, in particular N-(3-trimethoxysilylpropyl)-4,5-dihydroimidazole and N-(3-triethoxysilylpropyl)-4,5-dihydroimidazole.

It is possible and might be advantageous to include a mixture of these compounds as catalyst **CA**, for example a metal compound and a nitrogen-containing compound.

The multi-component curable composition thus preferably comprises a catalyst **CA** for for the curing of silane-functional polymers, in particular with an amount of between 0.005 wt.-% and 0.5 wt.-%, preferably between 0.01 wt.-% and 0.25 wt.-%, more preferably between 0.025 wt.-% and 0.15 wt.-% of said catalyst **CA** based on the total composition.

Catalyst **CA** is preferably included in a different component than polymer **PO**.

<u>Thermally conductive filler **FI**</u>

The composition according to the present invention comprises more than 70 wt.-%, based on the total composition, of at least one thermally conductive filler **FI**.

Preferably, the composition comprises up to 95 wt.-%, in particular up to 92 wt.-%, preferably between 75 wt.-% and 90 wt.-%, more preferably between 80 wt.-% and 85 wt.-%, based on the total composition, of said thermally condicutive filler **FI**.

The thermally conductive filler **FI** is not particularly limited.
Suitable thermally conductive fillers are in particular those that have a coefficient of thermal conductivity that is greater than 5 W/mK, preferably greater than 10 W/mK, more preferably greater than 15 W/mK.
Examples of thermally conductive fillers include alumina, alumina trihydrate or aluminum trihydroxide, silicon carbide, boron nitride, aluminium nitride, zinc oxide, aluminosilicate, diamond, and graphite, or mixtures thereof. Particularly preferred are aluminium trihydroxide (ATH), and aluminium oxide, with ATH being the most preferred.

The filler **FI** should be in powder form or at least particulate, in order to ensure homogeneous compounding in its high amounts.

In a preferred embodiment, the thermally conductive filler **FI** has a broad particle size distribution characterized by a ratio of D90/D50 of at or about 3 or more. Particularly preferably the thermally conductive filler is ATH or aluminium oxide having a broad particle size distribution characterized by a ratio of D90/D50 of at or about 3 or more, most preferably ATH. Also preferred are thermally conductive fillers having a bimodal particle size distribution. A bimodal distribution is when, for example, the ratio D90/D50 is at or about 3 or more, more preferably at or about 5 or more, more particularly preferably at or about 9 or more. For example, particles having a D50 of 5 to 20 microns and a D90 of 70 to 90 microns, particularly a D50 of 7-9 microns and a D90 of 78-82 microns. Particle size can be determined using laser diffraction. Preferred are aluminium oxide and ATH having a bimodal distribution, particularly ATH.

In a preferred embodiment of the moisture-curable composition according to present invention, the filler **FI** comprises or consists of aluminium oxide and aluminium hydroxide. This combination has the advantage that it shows especially good thermal conductivity and at the same time excellent flame-retardant properties. This makes it especially suitable for battery assemblies.

It is preferred in some embodiments to use more than one type of filler **FI**, for example aluminium oxide and aluminium hydroxide, with different particle sizes. For example, it is advantageous to use a bimodal or multimodal particle size range for the employed fillers. When at least one filler with relatively large particles and at least another one with relatively small particles are used together, a higher density of close packing of spheres may be achieved, which is beneficial for heat conductivity and compounding, especially when using small amounts of polymer **PO** and high amounts of filler **FI**.

The thermally conductive filler **FI** is preferably present in the final adhesive at a concentration that gives a thermal conductivity of at or about 1.5 W/mK or more, preferably at or about 2.0 or more, more preferably at or about 2.5 or more.

The thermally conductive filler **FI** may be present in any component of the multi-component curable composition according to the present invention. In case of a two-component composition that consists of two individual components **A** and **B** that are stored in separated containers and mixed before or during applications to yield the fully mixed curable composition, filler **FI** may be present in component **A**, in component **B**, or, preferably, in both components **A** and **B**. In a preferred embodiment thermally conductive filler **FI** is present in in both components **A** and **B**, as this reduces the amount of mixing required to properly distribute the thermally conductive filler **FI** within the composition when components **A** and **B** are mixed. Furthermore, this allows for a broader range of mixing ratios between the components while ensuring a sufficiently high amount of filler **FI** within the composition.

Plasticizer **PL**

The multi-component composition according to the present invention preferably comprises at least one plasticizer **PL**. The composition may be formulated without plasticizer, but it is preferred that small amounts (e.g., from 1 wt.-% up to 10 wt.-%, 12 wt.-%, or 15 wt.-%, based on the total composition) of plasticizer are used for ease of compounding and resulting application and mechanical properties.
Preferably, the composition additionally comprises between 1.5 wt.-% and 12.5 wt.-%, in particular between 2.5 wt.-% and 10 wt.-%, preferably between 3 wt.-% and 9 wt.-%, in particular between 4 wt.-% and 8 wt.-%, based on the total composition, of at least one plasticizer **PL**.

The plasticizer **PL** may be any of the plasticizers commonly used in compositions based on silane-functional polymers or mixtures of different such plasticizers. These include, for example, carboxylic esters such as phthalates, especially dioctyl phthalate, bis(2-ethylhexyl) phthalate, bis(3-propylheptyl) phthalate, diisononyl phthalate or diisodecyl phthalate, diesters of ortho-cyclohexane-dicarboxylic acid, especially diisononyl 1,2-cyclohexanedicarboxylate, adipates, especially dioctyl adipate, bis(2-ethylhexyl) adipate, azelates, especially bis(2-ethylhexyl) azelate, sebacates, especially bis(2-ethylhexyl) sebacate or diisononyl sebacate, glycol ethers, glycol esters, organic phosphoric or sulfonic esters, sulfonamides, polybutenes, or fatty acid methyl or ethyl esters derived from natural fats or oils, also called “biodiesel”.

Furthermore suitable are polymeric plasticizers. These have the advantage of lower migration tendency into surrounding areas and lower contribution to VOC levels.
The term “polymeric plasticizer” herein means a polymeric additive that is liquid at room temperature and contains no hydrolyzable silane groups. In contrast to traditional plasticizers, such as phthalates, the polymeric plasticizers generally have a higher molecular weight.
Preferably, the polymeric plasticizer has an average molecular weight M_n of 500 to 12’000 g/mol, in particular 1’000 to 10’000 g/mol, more preferably 2’500 to 5’000 g/mol.

Suitable polymeric plasticizers include polyols, such as those suitable for the production of the organic polymers **PO** mentioned there, as long as they are liduid at room temperature, and polyols where the OH-groups have been reacted to chemically inert functional groups. Preferred polyols suitable as polymeric plasticizers include polyether polyols, polyester polyols, polyhydrocarbon polyols, polybutadiene polyols, and poly(meth)acrylate polyols. Particularly preferred are polyether polyols, especially those with an average molecular weight of M_n of 500 to 12’000 g/mol, especially 1’000 to 10’000 g/mol, more preferably 2’500 to 5’000 g/mol.

Especially preferred plasticizers **PL** are low molecular weight diesters of polyether diols, in particular tri(ethylenglycol)bis-2-ethylhexanoate, which is available under the trade name Eastman® TEG-EH by Eastman.

Further preferred plasticizers **PL** for the multi-component curable composition according to the present invention comprise or consist of a trialkyl and/or triaryl phosphate, for

example triethyl phosphate, tricresyl phosphate, triphenyl phosphate, diphenyl cresyl phosphate, isodecyl diphenyl phosphate, tris(1,3-dichloro-2-propyl) phosphate, tris(2-chloroethyl) phosphate, tris(2-ethylhexyl) phosphate, tris(chloroisopropyl) phosphate, tris(chloropropyl) phosphate, isopropylated triphenyl phosphate, mono-, bis- or tris(isopropylphenyl) phosphates. Preferred trialkyl and/or triaryl phosphate plasticizers **PL** are tris-(2-ethylhexyl)-phosphate (sold under the trade name Disflamoll® TOF by Lanxess), cresyl diphenyl phosphate, tricresyl phosphate, and triphenyl phosphate (all sold under the trade name range Disflamoll® by Lanxess). Trialkyl and/or triaryl phosphate plasticizers have the advantage that they improve the flame-retardant properties of the composition.

Plasticizers **PL** may be present in one or several components of the multi-component curable composition according to the invention. Preferably, plasticizer **PL** is present in all components of the present invention, especially if filler **FI** is also present, as plasticzer **PL** facilitates a homogeneous compounding of highly filled components.

Dispersion additive

In some mebodiments, the multi-component curable composition furthermore may comprise at least one dispersion additive **D**. Dispersion additives are well known in the field of coating formulations containing fillers and pigments. They are also known as dispersants or wetting agents and facilitate the compounding of solids into a liquid matrix. In the context of the present invention, dispersion additive **D** may facilitate incorporation of very high amounts (i.e., more than 85 wt.-% based on the total composition, or higher) of filler **FI** within the composition. In principle all common dispersion additives are suitable, and their suitable amount for a given formulation depends on the type of dispersion additive and relative amount of filler **FI** and other consituents, such as polymer **PO**. In preferred embodiments, the composition comprises said dispersion additive **D** with an amount of between 0.5 and 5.0 wt.-%, preferably between 0.7 and 4.0 wt.-%, in particular between 1.0 and 3.0 wt.-%, based on the total composition.

An especially preferred dispersion additive **D** is an ammonium salt or alkyl ammonium salt of a polymer or compolymer containing carboxylate and/or phosphate groups, preferably a polyether and/or polyester polymer containing carboxylate and/or phosphate groups.

Suitable such dispersion additives **D** are, for example, Byk-W 996, Byk-W 969, Byk-W 985 (available from Altana) and Disparlon DA-234, DA-325, and DA-375 (available from King Industries).

Organosilanes

The multi-component curable composition according to the present invention furthermore preferably comprises organosilanes or oligomers of organosilanes, in particular monomeric or oligomeric organofunctional alkoxysilanes.

Preferably, the composition comprises between 0.1 wt.-% and 2.5 wt.-%, preferably between 0.2 wt.-% and 1.5 wt.-%, preferably between 0.25 wt.-% and 1.0 wt.-%, in particular between 0.5 wt.-% and 0.75 wt.-%, based on the total composition, of organosilanes or oligomers of organosilanes, in particular comprising or consisting of aminosilanes or oligomers thereof.

Organosilanes have various advantages. For example, they may act as desciccant or drying agent, in particular vinyl trimethoxysilane and/or propyl trimethoxysilane.

Other organosilanes have co-catalytic activity, in particular aminosilanes such as 3-aminopropyl trimethoxysilane, and/or they act as adhesion promotors, such as 3-glycidoxypropyl trimethoxysilane. Nevertheless, in some embodiments the composition does not contain adhesion-promoting silanes, such as amino silanes or glycidoxy silanes. A composition free of adhesion-promoting silanes, in particular amino-functional and/or glycidoxy-functional silanes, has the advantage that the composition may be easily removed from the substrate when the object bonded with the composition needs to be disassembled, for example during a battery replacement operation.

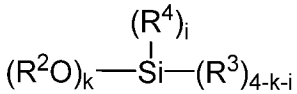
Preferred organosilanes acting, for example, as adhesion promoters and/or crosslinkers are in particular aminosilanes, mercaptosilanes, epoxysilanes, (meth)acryloylsilanes, anhydridosilanes, carbamatosilanes, alkylsilanes or iminosilanes, oligomeric forms of these silanes, adducts formed from primary aminosilanes with epoxysilanes or (meth)acryloylsilanes or anhydridosilanes, amino-functional alkylsilsesquioxanes, 3-glycidoxypropyltrimethoxysilane, 3-glycidoxypropyltriethoxysilane or 3-ureidopropyltrimethoxysilane, or oligomeric forms of these silanes.

Preferred organosilanes acting as desiccants or drying agents are in particular tetraethoxysilane, vinyltrimethoxysilane, vinyltriethoxysilane, propyltrimethoxysilane, or organoalkoxysilanes having a functional group in the α-position to the silane group, especially N-(methyldimethoxysilylmethyl)-O-methylcarbamate, (methacryloyloxymethyl)silanes, and methoxymethylsilanes.

Preferred organosilanes acting as co-catalysts are in particular aminosilanes.

The organosilanes preferably comprised in the composition may be monomeric organosilanes or oligomeric organosilanes.

Examples of suitable monomeric organosilanes are shown in formula (II),



(II)

where

R^2 is a monovalent alkyl radical having 1 to 6 carbon atoms, preferably 1 or 2 carbon atoms, most preferably a methyl radical;

R^3 is a monovalent, linear or branched hydrocarbon radical having 1 to 20 carbon atoms and optionally containing olefinic, aromatic and/or cyclic moieties and optionally containing heteroatoms selected from O, N, S, and Si;

R^4 is a monovalent linear, branched, or cyclic alkyl or arakyl radical having 1 to 12 carbon atoms, preferably 1 to 8 carbon atoms, most preferably a methyl radical;

index i is an integer with a value of 0 or 1, preferably 0;

index k is an integer with a value of 2 or 3, with the provisio that if i = 1 then k = 2.

In preferred embodiments, R^3 is an aminoalkyl radical selected from $–C_pH_{2p}-NH_2$, $–C_pH_{2p}-NH-R^5$, $–C_pH_{2p}-NH-C_dH_{2d}-NH_2$, $–C_pH_{2p}-NH-C_dH_{2d}-NH-C_eH_{2e}-NH_2$, $–C_pH_{2p}-NH-C_dH_{2d}-NH-R^5$, and $–C_pH_{2p}-NH-C_dH_{2d}-NH-C_eH_{2e}-NH-R^5$;

where

R^5 is a monovalent linear, branched, or cyclic alkyl or arakyl radical having 1 to 12 carbon atoms, preferably 1 to 6 carbon atoms;

index p is an integer with a value of 1 to 6;

indices d and e are independently integers with a value of 2 to 6.

In other preferred embodiments, R^3 is a glycidoxyalkyl radical.

In further preferred embodiments, R^3 is a mercapto- or hydroxyalkyl radical.

Suitable oligomeric silanes are linear, cyclic, or branched oligomers of the aforementioned monomeric organosilanes. They may be oligomers of one or more than one type of organosilane.

One advantage of using oligomeric organosilanes is that lower VOC levels can be achieved when employing them in greater amounts compared to purely monomeric silanes.

Preferred monomeric or oligomeric aminofunctional alkoxysilanes include N-(n-butyl)-3-aminopropyltrimethoxysilane, 3-aminopropyltrimethoxysilane, 3-aminopropyldimethoxymethyl-silane, N-(2-aminoethyl)-3-aminopropyltrimethoxysilane, N-(2-aminoethyl)-3-aminopropyldimethoxymethylsilane, N-(2-aminoethyl)-N'-[3-(trimethoxysilyl)-propyl]ethylenediamine and oligomers obtained from the condensation of the mentioned aminosilanes, optionally oligomerized together with alkylalkoxysilanes, in particular methyltrimethoxysilane, ethyltrimethoxysilane, propyltrimethoxysilane, vinyltrimethoxysilane, n-butyltrimethoxysilane, isobutyltrimethoxysilane, phenyltrimethoxysilane, and octyltrimethoxysilane.

Also preferred are monomeric or oligomeric or the analogs thereof with ethoxy in place of methoxy groups.

Preferred glycidoxyfunctional organosilanes are glycidoxypropyltrimethoxysilane and glycidoxypropyltriethoxysilane and oligomers containing glycidoxypropyl groups, stemming from monomeric glycidoxypropylalkxoysilanes condensated with the aforementioned organosilanes or with themselves.

Organsilanes are preferably present in the same component of the multi-component composition as polymer **PO**.

Further additives

The multi-component composition may comprise further constituents, especially the following auxiliaries and additives:

- additional desiccants or drying agents, especially orthoformic esters, calcium oxide or molecular sieves;

- additional plasticizers, especially reactive plasticizers in the form of monofunctional organic polymers or silanes, i.e. those that are silane-reactive only at one end;
- reactive diluents, in particular mono- or difunctional glycidyl ethers;
- solvents;
- further inorganic or organic fillers, especially baryte (heavy spar), talcs, quartz flours, quartz sand, dolomites, chalk, wollastonites, kaolins, calcined kaolins, mica (potassium aluminum silicate), molecular sieves, magnesium hydroxide, silicas including finely divided silicas from pyrolysis processes, industrially produced carbon blacks, metal powders such as iron or steel, PVC powder or hollow spheres;
- fibers, especially glass fibers, carbon fibers, metal fibers, ceramic fibers or polymer fibers such as polyamide fibers or polyethylene fibers;
- dyes;
- pigments, especially titanium dioxide or iron oxides;
- rheology modifiers, in particular thickeners or thixotropy additives, especially sheet silicates such as bentonites, derivatives of castor oil, hydrogenated castor oil, polyamides, polyurethanes, urea compounds, fumed (pyrogenic) silicas, cellulose ethers or hydrophobically modified polyoxyethylenes;
- stabilizers against oxidation, heat, light or UV radiation;
- natural resins, fats or oils such as rosin, shellac, linseed oil, castor oil or soybean oil;
- non-reactive polymers that are preferably solid at room temperature such as, in particular, homo- or copolymers of unsaturated monomers, especially from the group comprising ethylene, propylene, butylene, isobutylene, isoprene, vinyl acetate or alkyl (meth)acrylates, especially polyethylenes (PE), polypropylenes (PP), polyisobutylenes, ethylene-vinyl acetate copolymers (EVA) or atactic poly-α-olefins (APAO), polyesters, and polycarbonates;
- flame-retardant substances, especially the already mentioned filler aluminum hydroxide, magnesium hydroxide, or, in particular, the already mentioned organic phosphoric esters such as, in particular, triethyl phosphate, tricresyl phosphate, triphenyl phosphate, diphenyl cresyl phosphate, isodecyl diphenyl phosphate, tris(1,3-dichloro-2-propyl) phosphate, tris(2-chloroethyl) phosphate, tris(2-ethylhexyl) phosphate, tris(chloroisopropyl) phosphate, tris(chloropropyl) phosphate, isopropylated triphenyl phosphate, mono-, bis- or tris(isopropylphenyl) phosphates of different degrees of isopropylation, resorcinol bis(diphenyl phosphate), bisphenol A bis(diphenyl phosphate) or ammonium polyphosphates;

– surface-active substances, especially leveling agents, deaerating agents or defoamers;
– biocides, especially algicides, fungicides or substances that inhibit fungal growth;

and other substances customarily used in curable compositions. It may be advisable to chemically or physically dry certain constituents before mixing them into the composition, especially if they are stored in the same component as polymer **PO**.

In preferred embodiments of the multi-component curable composition according to the invention, the composition does not contain polydiorganosiloxanes.

The composition is preferably produced and stored with exclusion of moisture, especially regarding the component containing polymer **PO**. Typically, all components of the multi-component composition are storage-stable with exclusion of moisture in a suitable package or arrangement, such as, more particularly, a bottle, a canister, a pouch, a bucket, a vat or a cartridge.

The composition is a multi-component, especially two-component, composition.

In the present document, "two-component" refers to a composition in which the constituents of the composition are present in two different components which are stored in separate containers. Normally, the reactive species, i.e. polymer **PO** and organosilane, are stored in the first component **A**, and water is present in the second component **B**. Liquid epoxy resin **EP** and polyamine **PA** may be included in either one of components **A** and **B**, as long as they are not in the same component. However, preferably, polyamine **PA** is included in component **A** and epoxy resin **EP** in component **B**. Only shortly before or during the application of the composition are the two components mixed with one another, whereupon polymer **PO** in the mixed composition cures under the action of moisture from air humidity and/or from the second component. Epoxy resin **EP** and polyamine **PA** also cure when mixed with each other.
Any second or optionally further components is/are mixed with the first component prior to or on application, especially by means of a static mixer or by means of a dynamic mixer.
In the case of a two-component composition, it is still preferable to dry the filler **FI** in the first component **A** in order to ensure storage stability. The fillers of the second component **B**, however, do not have this restriction.

In preferred embodiments of the moisture-curable composition according to the present invention, the composition therefore consists the composition is a two-component composition that consists of two individual components **A** and **B** that are stored in separated containers and mixed before or during applications to yield the fully mixed curable composition, wherein said component **A** contains polymer **PO**, polyamine **PA**, and optionally organosilanes and/or other optional additives; and said component **B** contains water, epoxy resin **EP** and optionally further additives; and wherein thermally conductive filler **FI** and all further optional consitutents of the composition are present in either one or both of the components **A** and **B**.

An especially preferred embodiment of a two-component moisture-curable composition according to the present invention consists of a component **A** and a component **B**, wherein said component **A** comprises, based on component **A**,

- between 3 wt.-% and 15 wt.-%, preferably between 5 wt.-% and 10 wt.-% of said polymer **PO**;
- between 0.5 wt.-% and 7 wt.-%, preferably between 1 wt.-% and 5 wt.-% of said polyamine **PA**;
- between 0.5 and 4.0 wt.-%, preferably between 1.0 wt.-% and 4.0 wt.-% of at least one organosilane or oligomer thereof;
- optionally between 1 wt.-% and 15 wt.-%, preferably between 2 wt.-% and 10 wt.-% of said plasticizer **PL**;
- between 70 and 92 wt.-%, preferably between 80 wt.-% and 90 wt.-% of said thermally conductive filler **FI**;
- optionally between 0.25 wt.-% and 1.5 wt.-%, preferably between 0.5 wt.-% and 1.0 wt.-% of said accelerator **AC**;

and said component **B** comprises, based on component **B**,

- between 3 wt.-% and 15 wt.-%, preferably between 5 wt.-% and 12 wt.-% of said epoxy resin **EP**;
- between 70 and 92 wt.-%, preferably between 70 wt.-% and 90 wt.-% of said thermally conductive filler **FI**;
- between 1 wt.-% and 10 wt.-%, preferably between 2 wt.-% and 7.5 wt.-% of said plasticizer **PL**;
- between 0.1 wt.-% and 2.5 wt.-%, preferably between 0.2 wt.-% and 1.5 wt.-% of water;

- optionally between 0.005 and 0.5 wt.-%, preferably between 0.01 wt.-% and 0.25 wt.-% of said catalyst **CA**.

A preferred volumetric mixing ratio of component **A** to component **B** for above two-component composition is between 1:2 and 2:1, preferably between 1:1.5 and 1.5:1, in particular between 1:1.2 and 1.2:1, most preferably 1:1.

The water mentioned for component **B** may be added accordingly, or it may be present from the beginning as impurity in not previously dried fillers **FI**. Fillers **FI** may contain, if not dried, up to 1 wt.-% or more of water bound to their surface or within pores. This water is normally not chemically bound but only physisorbed and is able to promote the crosslinking of the polymers **PO** when the composition is mixed. Depending on the type and dryness of fillers **FI** used, however, it may be advantageous to add water to component **B**, preferably in the above specified amounts.

All the preferred embodiments described for the individual consituents, e.g., polymer **PO** or organosilane, of above two-component composition are the same as described in a more general manner further above in the description.

The curable composition according to the present invention is preferably applied at ambient temperature, such as room temperature, preferably within a temperature range between 0°C and 45°C, especially 5°C to 35°C, and cures under these conditions. However, it can also cure in an accelerated manner at temperatures up to 80°C.
On application, the crosslinking reaction of the silane groups commences, if appropriate under the influence of moisture. Silane groups present can condense with silanol groups present to give siloxane groups (Si-O-Si groups). Silane groups present can also be hydrolyzed on contact with moisture to give silanol groups (Si-OH groups) and form siloxane groups (Si-O-Si groups) through subsequent condensation reactions. As a result of these reactions, the composition ultimately cures. The catalysts **CA** described further above accelerate this curing mechanism catalytically.
Likewise, on mixing of the components, the reaction of epoxide groups on liquid epoxy resin **EP** with amino groups on polyamine **PA** commences, also leading to further cross-linking and curing of the composition. The accelerators **AC** described further above accelerate this curing mechanism catalytically.

In summary, a two-fold curing mechanism occurs that leads to a mechanically favorable cured composition that exhibits better properties than compositions only based on a single one of these curing mechanisms.

The multi-component composition according to the invention is in principle suitable for a multitude of uses, for example as a molding, elastomer, film or membrane, as a potting compound, sealant, gap filler, adhesive, covering, or coating for construction and industrial applications, for example as a seam seal, cavity seal, electrical insulation compound, assembly adhesive, bodywork adhesive, seal, or gap filler. The composition is particularly suitable as an adhesive, gap filler, potting resin, and/or sealant, especially in automotive manufacturing, for batteries, electronic elements, engine control units, anti-lock breaking and electronic stability control and safety systems, DC/DC converter of hybrid electric vehicles, advanced driver-assistance systems, sensors, or control units.

For an application as adhesive, gap filler, or sealant, the composition preferably has a pasty consistency with structurally viscous properties. Such a pasty sealant or adhesive is especially applied to a substrate from standard cartridges which are operated manually, by means of compressed air or with a battery, or from a vat or hobbock by means of a delivery pump or an extruder, optionally by means of an application robot.

On application, the composition is preferably applied to at least one substrate. Suitable substrates are especially

- glass, glass ceramic, concrete, mortar, brick, tile, gypsum and natural rocks such as limestone, granite or marble;
- metals and alloys such as aluminum, iron, steel and nonferrous metals, and also surface-finished metals and alloys such as galvanized or chromed metals or surface coated metals, such as Kynar®- or Duranar®-coated aluminum, and electrolytically nickel-plated steel, for example available under the trade name Hilumin® from Tata Steel;
- leather, textiles, paper, wood, woodbase materials bonded with resins, for example phenolic, melamine or epoxy resins, resin-textile composites and further polymer composites;
- plastics such as polyvinyl chloride (rigid and flexible PVC), acrylonitrile-butadiene-styrene copolymers (ABS), polycarbonate (PC), polyamide (PA), polyesters,

poly(methyl methacrylate) (PMMA), epoxy resins, polyurethanes (PUR), polyoxymethylene (POM), polyolefins (PO), polyethylene (PE) or polypropylene (PP), ethylene/propylene copolymers (EPM) and ethylene/propylene/diene terpolymers (EPDM), and also fiber-reinforced plastics such as carbon fiber-reinforced plastics (CFP), glass fiber-reinforced plastics (GFP) and sheet molding compounds (SMC), where the plastics may have been surface-treated by means of plasma, corona or flames;

- coated substrates such as powder-coated metals or alloys;
- electrocoated (e-coat) surfaces coated by electrophoretic painting processes;
- paints or varnishes, especially automotive topcoats;
- electrically isolating foils (such as PET foils) and dielectric castings.

If required, the substrates can be pretreated prior to the application of the composition, especially by chemical and/or physical cleaning methods or by the application of an adhesion promoter, an adhesion promoter solution or a primer.
In general, it is not required to pre-treat the surfaces prior to application of the composition. The composition shows in preferred embodiments an excellent adhesion profile on a large variety of unprimed, non-pretreated, and even uncleaned materials. These compositions preferably do not contain adhesion promoting organosilanes as described further above.

It is possible to bond or seal two identical or two different substrates, especially the aforementioned substrates.

After the curing of the composition involving the two chemical cross-linking reactions between polymers **PO** and between epoxy resin **EP** and polyamine **PO**, a cured composition is obtained.

The compositions disclosed herein possess in preferred embodiments excellent thermal conductivity, in particular of ≥ 1.5 W/mK, in preferred embodiments of ≥ 2.0 W/mK, according to ASTM D5470. Other preferred embodiments may have lower thermal conductivity, but instead have higher mechanical properties, e.g., tensile strength and elongation at break.

The compositions disclosed herein generally possess low viscosities of less than 200 Pa s at a shear rate of 10 s^{-1} in their mixed state. Likewise, they possess a low squeeze flow of less than 500 N, in particular less than 250 N, more preferably less than 150 N at a gap with of 0.8 mm according to the measurement specified in the experimental section below.

Furthermore, the compositions disclosed herein after curing have the advantage of good mechanical properties, including high tensile strength, high elongation at break, and high lap shear strength.

Another aspect of the present invention is the use of a multi-component curable composition as described herein to adhesively bond, coat, or seal substrates or as a filler or potting resin for hollow bodies.

In preferred embodiments of said use, at least one of the substrates or hollow bodies is part of a battery or an electronic device.

The use of the composition gives rise to an article that was bonded, sealed, filled, or coated with the composition according to the invention. The article is especially a built structure, especially a structure built by structural engineering or civil engineering, an industrially manufactured good or a consumable good, especially a domestic appliance or a mode of transport such as, more particularly, an automobile, a bus, a truck, a rail vehicle, a ship, an aircraft, a drone, or a helicopter; or the article may be an installable component thereof, in particulate a battery box or electronic part.

Another aspect of the present invention is thus a substrate, adhesively sealed, coated, or bonded by a cured multi-component composition described herein, as well as a hollow body filled with a cured multi-component composition described herein.

Examples

Adduced hereinafter are working examples which are intended to elucidate the invention described in detail. It will be appreciated that the invention is not restricted to these described working examples.

The term “standard climatic conditions” refers to a temperature of 23±1°C and a relative air humidity of 50±5%.

Test methods:

Viscosity was determined on a MCR 302 rheometer (Anton Paar) according to ISO 3219. Measurement parameters were: Rotation 0.1 – 10 s-1, measurement point at 10 s-1, temperature 20°C, gap 0.5 mm. Several compositions **A** and compositions **B** were measured individually before mixing (Table 7).

Thermal conductivity ("TC") was determined according to ASTM D5470-12 on samples cured during 7 days under standard climatic conditions. For the measurements, a TIM (thermal interface material) testing device (Zentrum für Wärmemanagement, Stuttgart, Germany) using the stationary cylinder method was used. Sample dimensions were: Diameter 30 mm, thickness 2 mm. The pressure parameter of the measurements were 1, 2, 3, 5, 7, 10 bar.

The **tensile strength ("TS")**, the **elongation at break ("EOB")**, and the modulus of elasticity at 0.5 to 5.0% elongation (**E-modulus**, "**E-mod**") were determined in accordance with DIN EN 53504 (tensioning speed 10 mm/min) on dumbbells with a length of 75 mm, with a bar length of 30 mm and a bar width of 4 mm, which were produced by punching from films with a thickness of around 2 mm, these films being films of the composition cured after a storage time of 7 days under standard conditions.

For the measurement of the **lap shear strength ("LSS")**, test specimens were produced by applying the composition for 1 minute in each case after the end of the mixing time, between two aluminium sheets (aluminium sheet thickness 1.5 mm) degreased with isopropanol, in a layer thickness of 1 mm, on an overlapping bond area of 10 x 25 mm. The lap shear strength was determined on these test specimens at room temperature in accordance with DIN EN 1465, the test specimens having been cured prior to measurement for 7 days under standard conditions.

Squeeze flow measurements were performed on a Zwick machine properly equipped to determine the forces occurring during the measurements. The test compositions were applied on a 60 mm diameter pressure plate and pressed by a 40 mm pressure stamp, wherein the surfaces of the stamp and the plate were parallel. The measurements were performed under standard climate (23°C, 50% r.h.). In particular, the measurements were performed as follows: After application of the test material on the 60 mm plate, the

40 mm pressure stamp was driven vertically downwards at a speed of 1 mm/s until a gap of 5 mm between the stamp and the plate were reached. At this point the test material must cover the entire surface of the 40 mm pressure stamp. After this, the pressure stamp was further driven down until a 0.3 mm gap was reached, with a speed of 1 mm/s. The required force was evaluated in Newton (N) at a defined gap, e. g. 0.5 mm, 0.8 mm, or 1 mm, according to the specification in the table.

Compounds used:

Name	Description, trade name
STP-E15 (polymer **PO**)	Polyether polymer having trimethoxysilylpropyl-carbamate end groups (Geniosil® STP-E15, Wacker)
Organosilane 1	N-(n-Butyl)-3-aminopropyltrimethoxysilane (Dynasylan® 1189, Evonik)
Organosilane 2	3-Aminopropyltrimethoxysilane (Dynasylan® 1110, Evonik)
Organosilane 3	N-Aminoethyl-3-aminopropyltrimethoxysilane (Dynasylan® 1120, Evonik)
Organosilane 4	Propyltrimethoxysilane (Dynasylan® PTMO, Evonik)
TEG-EH (plasticizer **PL**)	Triethylene glycol bis(2-ethylhexanoate) (Eastman® TEG-EH, Eastman)
D-230 (polyamine **PA**)	Polyetherdiamine, ca. 230 g/mol, AHEW about 60 g (Jeffamine® D-230, Huntsman)
T-3000 (polyamine **PA**)	Polyethertriamine, ca. 3000 g/mol, (Jeffamine® T-3000, Huntsman)
THF-100 (polyamine **PA**)	Polyetherdiamine, ca. 1000 g/mol, based on a PTMEG [poly(tetramethylene ether glycol)]/PPG (polypropylene glycol) copolymer (Jeffamine® THF-100, Huntsman)
A-1769 (polyamine **PA**)	Hydroxyalkylated polyamine having only secondary amino groups (Ancamine® 1769, Evonik)
IPDA (polyamine **PA**)	Isophorone diamine (Vestamin® IPD, Evonik)
ATH (filler **FI**)	Aluminium hydroxide ($Al(OH)_3$), surface-coated, with bimodal particle size distribution; residual water content ≤ 0.25 wt.-%.
Chalk	Ground natural calcium carbonate (Omyacarb® 5, Omya)
K54 (accelerator **AC**)	Tris-(dimethylaminomethyl) phenol (Ancamine® K54, Evonik)

BADGE (epoxy resin **EP**)	Liquid Bisphenol A diglycidyl ether (Epikote® 828 LVEL, Westlake Epoxy)
Water	Deionized water
DBTDL (catalyst **CA**)	Dibutyltin(IV) dilaurate
Silica	Pyrogenic silica (HDK® H18, Wacker)
Reactive diluent	Diglycidylether of 1.6-hexanediol; reactive diluent (Araldite® DY-H, Huntsman)

Table 1: Compounds used for the example compositions.

Example compositions:

Comparative examples (not according to the present invention) are identified in the following tables by "(Ref.)".

A series of example two-component compositions was prepared by mixing the ingredients of each respective component **A** and **B** shown in Tables 2, 5, 8, and 11 (components **A**) and Tables 3, 6, 9, and 12 (components **B**) in the indicated sequence as listed in the table under nitrogen atmosphere in a vacuum mixer until homogeneous pastes were obtained. The individual components **A** and **B** were filled into internally coated aluminum spreading piston cartridges that were closed airtight and stored under standard climate conditions for at least 24 h until the testing protocol (e.g, viscosity) was employed (see above for details). For testing of the cured compositions, the respective components **A** and **B** of the compositions were mixed in a volume ratio of 1:1 and subsequently left for curing during 7 days under standard climate (if not otherwise specified for the respective test method). The test results of these 1:1 mixtures are shown in Tables 4, 7, 10, ans 13 for selected compositions.

Composition (components A)	**C1-A** (Ref.)	**C2-A**	**C3-A**	**C4-A**	**C5-A**	**C6-A**	**C7-A**	**C26-A** (Ref.)	**C27-A** (Ref.)
STP-E15 (polymer **PO**)	11	7.7	7.7	7.7	7.7	7.7	7.7	10.0	14.0
Organosilane 1	0.57	0.6	0.6	0.6	0.6	0.6	0.6	0.8	0.8
Organosilane 3	1.02	0.99	0.99	0.99	0.99	0.99	0.99	1.3	0.9
TEG-EH (plasticizer **PL**)	8.41	7.28	6.61	5.58	5.21	6.11	5.01	8.4	1.8
D-230 (polyamine **PA**)	0	0.43	1.1	2.13	0.7	1.6	2.7	4.7	2.3
THF-100 (polyamine **PA**)	0	0	0	0	1.8	0	0	0	0
T-3000 (polyamine **PA**)	0	0	0	0	0	0	0	11.8	0
K54 (accelerator **AC**)	0	0	0	0	0	0	0	1.0	0
Silica	0	0	0	0	0	0	0	2.0	0
Chalk	0	0	0	0	0	0	0	0	16.6
ATH (filler **FI**)	79	83	83	83	83	83	83	60	63.4
Organosilane 4	0	0	0	0	0	0	0	0	0.2

Table 2: Example compositions components **A** (all numbers in wt.-%, based on the total individual composition **A**).

Composition (components B)	**C1-B** (Ref.)	**C2-B**	**C3-B**	**C4-B**	**C5-B**	**C6-B**	**C7-B**	**C26-B** (Ref.)	**C27-B** (Ref.)
BADGE (epoxy resin **EP**)	0	4.5	6.5	6.5	6.5	8	14	25	8
Water	0.26	0.5	0.5	0.5	0.5	0.5	0.5	1.5	0.7
TEG-EH (plasticizer **PL**)	11.67	10.95	8.95	8.95	8.95	7.45	1.45	11.42	9.23
DBTDL (catalyst **CA**)	0.09	0.05	0.05	0.05	0.05	0.05	0.05	0.08	0.07
TM3810 (filler **FI**)	88	84	84	84	84	84	84	60	65.4
Chalk	0	0	0	0	0	0	0	0	16.6
Silica	0	0	0	0	0	0	0	2	0

Table 3: Example compositions components **B** (all numbers in wt.-%, based on the total individual composition **B**).

Test results compositions A+B (1:1 volume mixture)	**C1-A + C1-B** (Ref.)	**C2-A + C2-B**	**C3-A + C3-B**	**C4-A + C4-B**	**C5-A + C5-B**	**C6-A + C6-B**	**C7-A + C7-B**	**C26-A + C26-B** (Ref.)	**C27-A + C27-B** (Ref.)
LSS [MPa]	1.11	1.5	1.97	1.93	2.03	3.06	4.98	3.8	n/p
EOB [%]	22.8	17.5	9.6	11.6	11.1	7.3	3.5	10.0	n/p
E-mod [MPa]	6.63	14.2	32.3	26.5	30.5	65	n/m	56.0	n/p
TS [MPa]	1.14	1.62	2.36	2.24	2.4	3.77	5.81	4.46	n/p
Squeeze flow (0.8 mm) [N]	898	258	193	203	225	205	421	104	n/p
TC [W/mK]	n/m	n/m	n/m	n/m	n/m	n/m	n/m	0.6	n/p

Table 4: Testing results of the investigated samples. “n/m” means not measured. “n/p” means that the samples were so highly viscous that no homogeneous mixture could be produced and hence no measurements could be performed.

Composition (components A)	**C8-A**	**C9-A**	**C10-A**	**C11-A**	**C12-A**	**C13-A**	**C14-A**
STP-E15 (polymer **PO**)	10	10	10	10	10	6	10
Organosilane 1	0.9	0.9	0.9	0.9	0.9	0.9	0.9
Organosilane 2	0.9	0.9	0.9	0.9	0.9	0.9	0.9
TEG-EH (plasticizer **PL**)	3	3.7	4	2	2.5	6	2.5
D-230 (polyamine **PA**)	3.7	0	0	3.7	3.7	3.7	3.7
A-1769 (polyamine **PA**)	0	3	0	0	0	0	0
IPDA (polyamine **PA**)	0	0	1.1	0	0	0	0
K54 (accelerator **AC**)	0	0	0	1	0.5	0	0.5
ATH (filler **FI**)	81	81	81	81	81	82	81
Organosilane 4	0.5	0.5	0.5	0.5	0.5	0.5	0.5

Table 5: Example compositions components **A** (all numbers in wt.-%, based on the total individual composition **A**).

Composition (components B)	**C8-B**	**C9-B**	**C10-B**	**C11-B**	**C12-B**	**C13-B**	**C14-B**
BADGE (epoxy resin **EP**)	11	11	11	11	11	11	9
Reactive diluent	0	0	0	0	0	0	2
Water	0.7	0.7	0.7	0.7	0.7	0.7	0.7
TEG-EH (plasticizer **PL**)	6.23	6.23	6.23	6.23	6.23	6.23	6.23
DBTDL (catalyst **CA**)	0.07	0.07	0.07	0.07	0.07	0.07	0.07
ATH (filler **FI**)	82	82	82	82	82	82	82

Table 6: Example compositions components **B** (all numbers in wt.-%, based on the total individual composition **B**).

Test results compositions A+B (1:1 volume mixture)	**C8-A + C8-B**	**C9-A + C9-B**	**C10-A + C10-B**	**C11-A + C11-B**	**C12-A + C12-B**	**C13-A + C13-B**	**C14-A + C14-B**
LSS [MPa]	4.4	2.6	4.2	5.9	5.7	4.4	5.5
EOB [%]	3.1	3.3	2.2	1.8	3	3	3.6
E-mod [MPa]	370	80	246	663	327	244	283
TS [MPa]	6.2	3.3	4.6	10	8.2	5.7	7.2
Viscosity **A** (10 s^{-1})	57	148	160	96	60	40	60
Viscosity **B** (10 s^{-1})	80	80	80	80	80	80	56
TC [W/mK]	n/m	1.86	1.85	1.88	n/m	n/m	n/m

Table 7: Testing results of the investigated samples. “n/m” means not measured.

Composition (components A)	**C15-A**	**C16-A**	**C17-A**	**C18-A**	**C19-A**	**C20-A**
STP-E15 (polymer **PO**)	8	8	8	8	8	4
Organosilane 1	0.9	0.9	0.9	0.9	0.9	0.8
Organosilane 3	0.9	0.9	0.9	0.9	0.9	0.9
TEG-EH (plasticizer **PL**)	6.8	6.4	3.7	3.4	2.6	11.8
D-230 (polyamine **PA**)	1.2	1.6	4.3	4.6	5.4	2.3
ATH (filler **FI**)	82	82	82	82	82	80
Organosilane 4	0.2	0.2	0.2	0.2	0.2	0.2

Table 8: Example compositions components **A** (all numbers in wt.-%, based on the total individual composition **A**).

5

Composition (components B)	**C15-B**	**C16-B**	**C17-B**	**C18-B**	**C19-B**	**C20-B**
BADGE (epoxy resin **EP**)	5	6	14	15	17	8
Water	0.7	0.7	0.7	0.7	0.7	0.7
TEG-EH (plasticizer **PL**)	12.23	11.23	3.23	2.23	0.23	9.23
DBTDL (catalyst **CA**)	0.07	0.07	0.07	0.07	0.07	0.07
ATH (filler **FI**)	82	82	82	82	82	82

Table 9: Example compositions components **B** (all numbers in wt.-%, based on the total individual composition **B**).

Test results compositions A+B (1:1 volume mixture)	C15-A + C15-B	C16-A + C16-B	C17-A + C17-B	C18-A + C18-B	C19-A + C19-B	C20-A + C20-B
LSS [MPa]	1.3	1.6	5.3	5.3	5.4	1.5
EOB [%]	8	8	1	1	0.5	7
E-mod [MPa]	18	30	n/m	n/m	n/m	23
TS [MPa]	1.2	1.6	7.7	9.5	9.8	1.2
Squeeze flow (0.5 mm) [N]	214	850	575	681	799	87
Squeeze flow (0.8 mm) [N]	67	268	188	225	262	28
TC [W/mK]	1.95	1.94	1.92	1.95	1.96	1.88

Table 10: Testing results of the investigated samples. “n/m” means not measured.

Composition (components A)	C21-A	C22-A	C23-A	C24-A	C25-A
STP-E15 (polymer **PO**)	5	6	14	15	16
Organosilane 1	0.8	0.8	0.8	0.8	0.8
Organosilane 3	0.9	0.9	0.9	0.9	0.9
TEG-EH (plasticizer **PL**)	10.8	9.8	1.8	0.8	0
D-230 (polyamine **PA**)	2.3	2.3	2.3	2.3	2.3
ATH (filler **FI**)	80	80	80	80	80
Organosilane 4	0.2	0.2	0.2	0.2	0.2

Table 11: Example compositions components **A** (all numbers in wt.-%, based on the total individual composition **A**).

5

Composition (components B)	C21-B	C22-B	C23-B	C24-B	C25-B
BADGE (epoxy resin **EP**)	8	8	8	8	8
Water	0.7	0.7	0.7	0.7	0.7
TEG-EH (plasticizer **PL**)	9.23	9.23	9.23	9.23	9.23
DBTDL (catalyst **CA**)	0.07	0.07	0.07	0.07	0.07
ATH (filler **FI**)	82	82	82	82	82

Table 12: Example compositions components **B** (all numbers in wt.-%, based on the total individual composition **B**).

Test results compositions A+B (1:1 volume mixture)	**C21-A + C21-B**	**C22-A + C22-B**	**C23-A + C23-B**	**C24-A + C24-B**	**C25-A + C25-B**
LSS [MPa]	1.8	1.9	2.8	3	2.9
EOB [%]	7	6	7	5	8
E-mod [MPa]	32	37	75	84	76
TS [MPa]	1.6	1.6	2.8	2.8	2.9
Squeeze flow (0.5 mm) [N]	117	143	677	1240	n/m
Squeeze flow (0.8 mm) [N]	39	45	217	418	n/m
TC [W/mK]	1.86	1.85	1.82	1.86	1.84

Table 13: Testing results of the investigated samples. “n/m” means not measured.

Claims

1. A multi-component curable composition, comprising
   - at least one organic polymer **PO** containing alkoxysilane groups;
   - at least one liquid epoxy resin **EP**;
   - at least one polyamine **PA**;
   - more than 70 wt.-%, based on the total composition, of at least one thermally conductive filler **FI**;
   - water;
   - optionally additives selected from curing catalysts, accelerators, plasticizers, organosilanes, stabilizers, and colorants;

   wherein said liquid epoxy resin **EP** and said polyamine **PA** are not being present in the same component, and in that said organic polymer **PO** and said water are not present in the same component.

2. The multi-component curable composition as claimed in claim 1, characterized in that the composition comprises between 3 wt.-% and 10 wt.-%, preferably between 3.5 wt.-% and 7.5 wt.-%, in particular between 4 wt.-% and 6 wt.-%, based on the total composition, of said organic polymer **PO**.

3. The multi-component curable composition as claimed in claim 1 or 2, characterized in that the composition comprises between 2 wt.-% and 10 wt.-%, preferably between 2.5 wt.-% and 7.5 wt.-%, in particular between 3 wt.-% and 6 wt.-%, based on the total composition, of said liquid epoxy resin **EP**.

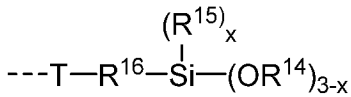
4. The multi-component curable composition as claimed in any of claims 1 to 3, characterized in that the thermally conductive filler **FI** comprises or consists of aluminium oxide and/or aluminium hydroxide.

5. The multi-component curable composition as claimed in any of claims 1 to 4, characterized in that the composition comprises between 0.2 wt.-% and 2.5 wt.-%, preferably between 0.25 wt.-% and 2.0 wt.-%, in particular between 0.5 wt.-% and 1.5 wt.-%, based on the total composition, of said polyamine **PA**.

6. The multi-component curable composition as claimed in any of claims 1 to 5, characterized in that the composition comprises between 0.1 wt.-% and 2.0 wt.-%, based on the total composition, of water in liquid or chemically accessible form.

7. The multi-component curable composition as claimed in any of claims 1 to 6, characterized in that the composition additionally comprises between 1.5 wt.-% and 12.5 wt.-%, in particular between 4 wt.-% and 8 wt.-%, based on the total composition, of at least one plasticizer **PL**.

8. The multi-component curable composition as claimed in any of claims 1 to 7, characterized in that said organic polymer **PO** has end groups of the formula (VIII)



(VIII)

wherein

R^{16} is a linear or branched divalent hydrocarbyl radical having 1 to 12 carbon atoms, preferably a methylene radical;

T is a divalent radical selected from –O–, –S–, –N(R^{17})–, –O–CO–N(R^{17})–, –N(R^{17})–CO–O– and –N(R^{17})–CO–N(R^{17})–, where R^{17} is a hydrogen radical or a linear or branched hydrocarbyl radical which has 1 to 20 carbon atoms and optionally has cyclic moieties;

R^{14} is a methyl or ethyl radical;

R^{15} is a methyl radical; and

x is 0 or 1.

9. The multi-component curable composition as claimed in any of claims 1 to 8, characterized in that the composition furthermore comprises between 0.1 wt.-% and 2.5 wt.-%, in particular between 0.5 wt.-% and 0.75 wt.-%, based on the total composition, of organosilanes or oligomers of organosilanes, in particular comprising or consisting of aminosilanes or oligomers thereof.

10. The multi-component curable composition as claimed in any of claims 1 to 9, characterized in that the composition additionally comprises a catalyst **CA** for the hydrolysis and/or condensation of alkoxysilane groups and preferably an accelerator **AC** for the reaction between epoxides and amines.

11. The multi-component curable composition as claimed in any of claims 1 to 10, characterized in that the composition comprises up to 92 wt.-%, preferably between 75 wt.-% and 90 wt.-%, based on the total composition, of said thermally condicutive filler **FI**.

12. The multi-component curable composition as claimed in any of claims 1 to 11, characterized in that the composition is a two-component composition that consists of two individual components **A** and **B** that are stored in separated containers and mixed before or during applications to yield the fully mixed curable composition, wherein said component **A** contains polymer **PO**, polyamine **PA**, and optionally organosilanes and/or other optional additives; and said component **B** contains water, epoxy resin **EP** and optionally further additives; and wherein thermally conductive filler **FI** and all further optional consitutents of the composition are present in either one or both of the components **A** and **B**.

13. The two-component curable composition as claimed in claim 12, characterized in that said component **A** comprises, based on component **A**,
- between 3 wt.-% and 15 wt.-% of said polymer **PO**;
- between 0.5 wt.-% and 7 wt.-% of said polyamine **PA**;
- between 0.5 and 4.0 wt.-% of at least one organosilane or oligomer thereof;
- optionally between 1 wt.-% and 15 wt.-% of said plasticizer **PL**;
- between 70 and 92 wt.-% of said thermally conductive filler **FI**;
- optionally between 0.25 wt.-% and 1.5 wt.-% of said accelerator **AC**;

and said component **B** comprises, based on component **B**,
- between 3 wt.-% and 15 wt.-% of said epoxy resin **EP**;
- between 70 and 92 wt.-% of said thermally conductive filler **FI**;
- between 1 wt.-% and 10 wt.-% of said plasticizer **PL**;
- between 0.1 wt.-% and 2.5 wt.-% of water;
- optionally between 0.005 and 0.5 wt.-% of said catalyst **CA**.

14. Use of a multi-component curable composition according to any of claims 1 to 13 to adhesively bond, coat, or seal substrates or as a filler or potting resin for hollow bodies.

15. The use according to claim 14, characterized in that at least one of the substrates or hollow bodies is part of a battery or an electronic device.

INTERNATIONAL SEARCH REPORT

International application No
PCT/EP2024/065431

A. CLASSIFICATION OF SUBJECT MATTER

INV. C08G59/50 C08G65/336 C09J171/02 C08K3/22 C08L63/00
C08L71/02 C09D163/00 C09D171/02 C09J163/00

ADD.

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

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

C08K C09J C08G

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

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)

EPO-Internal

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X,P	WO 2023/149328 A1 (THREE BOND CO LTD [JP]) 10 August 2023 (2023-08-10) claims 1-9 table 1	1-15

X	CN 115 926 719 A (ZHEJIANG DUOBANG NEW MAT CO LTD) 7 April 2023 (2023-04-07) Examples claims 1-10	1-15

A	US 2022/073738 A1 (MENNECKE KLAAS [DE] ET AL) 10 March 2022 (2022-03-10) claims 1-15 Examples	1-15

A	EP 1 695 989 A1 (KANEKA CORP [JP]) 30 August 2006 (2006-08-30) claims 1-6 Examples	1-15

☐ Further documents are listed in the continuation of Box C.

☒ See patent family annex.

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"&" document member of the same patent family

Date of the actual completion of the international search	Date of mailing of the international search report
19 June 2024	03/07/2024

Name and mailing address of the ISA/	Authorized officer
European Patent Office, P.B. 5818 Patentlaan 2 NL - 2280 HV Rijswijk Tel. (+31-70) 340-2040, Fax: (+31-70) 340-3016	Pouilley, Delphine

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INTERNATIONAL SEARCH REPORT

Information on patent family members

International application No
PCT/EP2024/065431

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