

DTU Library

Polysiloxane-based elastomers and methods of producing such

Sønderbæk-Jørgensen, René; Daugaard, Anders E; Ladegaard Skov, Anne; Dam-Johansen, Kim

Publication date: 2023

Document Version
Publisher's PDF, also known as Version of record

Link back to DTU Orbit

Citation (APA):

Sønderbæk-Jørgensen, R., Daugaard, A. E., Ladegaard Skov, A., & Dam-Johansen, K. (2023). Polysiloxane-based elastomers and methods of producing such. (Patent No. *WO2023139218*).

General rights

Copyright and moral rights for the publications made accessible in the public portal are retained by the authors and/or other copyright owners and it is a condition of accessing publications that users recognise and abide by the legal requirements associated with these rights.

- Users may download and print one copy of any publication from the public portal for the purpose of private study or research.
- You may not further distribute the material or use it for any profit-making activity or commercial gain
- You may freely distribute the URL identifying the publication in the public portal

If you believe that this document breaches copyright please contact us providing details, and we will remove access to the work immediately and investigate your claim.

(12) INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

(19) World Intellectual Property **Organization**

International Bureau

27 July 2023 (27.07.2023)





(10) International Publication Number WO 2023/139218 A1

(51) International Patent Classification:

C08G 77/62 (2006.01) C08L 83/04 (2006.01) C08L 83/16 (2006,01) C08G 77/16 (2006,01)

(21) International Application Number:

PCT/EP2023/051379

(22) International Filing Date:

20 January 2023 (20.01.2023)

(25) Filing Language: **English**

(26) Publication Language: English

(30) Priority Data:

22152707.0 21 January 2022 (21.01.2022)

- (71) Applicant: DANMARKS TEKNISKE UNIVERSITET [DK/DK]; Anker Engelunds Vej 101, 2800 Kongens Lyngby (DK).
- (72) Inventors: SØNDERBÆK-JØRGENSEN, René; c/o Danmarks Tekniske Universitet, Anker Engelunds Vej 101, 2800 Kongens Lyngby (DK). DAUGAARD, Anders E.; c/o Danmarks Tekniske Universitet, Anker Engelunds Vej 101, 2800 Kongens Lyngby (DK). LADEGAARD SKOV, Anne; c/o Danmarks Tekniske Universitet, Anker Engelunds Vej 101, 2800 Kongens Lyngby (DK). DAM-JOHANSEN, Kim; c/o Danmarks Tekniske Universitet, Anker Engelunds Vej 101, 2800 Kongens Lyngby (DK).
- (74) Agent: HØIBERG P/S; Adelgade 12, 1304 Copenhagen K (DK).
- (81) Designated States (unless otherwise indicated, for every kind of national protection available): AE, AG, AL, AM, AO, AT, AU, AZ, BA, BB, BG, BH, BN, BR, BW, BY, BZ, CA, CH, CL, CN, CO, CR, CU, CV, CZ, DE, DJ, DK, DM, DO, DZ, EC, EE, EG, ES, FI, GB, GD, GE, GH, GM, GT, HN, HR, HU, ID, IL, IN, IQ, IR, IS, IT, JM, JO, JP, KE, KG, KH, KN, KP, KR, KW, KZ, LA, LC, LK, LR, LS, LU, LY, MA, MD, MG, MK, MN, MW, MX, MY, MZ, NA, NG, NI, NO, NZ, OM, PA, PE, PG, PH, PL, PT, QA, RO, RS, RU, RW, SA, SC, SD, SE, SG, SK, SL, ST, SV, SY, TH, TJ, TM, TN, TR, TT, TZ, UA, UG, US, UZ, VC, VN, WS, ZA, ZM, ZW.
- (84) Designated States (unless otherwise indicated, for every kind of regional protection available): ARIPO (BW, CV, GH, GM, KE, LR, LS, MW, MZ, NA, RW, SD, SL, ST, SZ, TZ, UG, ZM, ZW), Eurasian (AM, AZ, BY, KG, KZ, RU, TJ, TM), European (AL, AT, BE, BG, CH, CY, CZ, DE, DK, EE, ES, FI, FR, GB, GR, HR, HU, IE, IS, IT, LT, LU, LV, MC, ME, MK, MT, NL, NO, PL, PT, RO, RS, SE, SI, SK, SM, TR), OAPI (BF, BJ, CF, CG, CI, CM, GA, GN, GQ, GW, KM, ML, MR, NE, SN, TD, TG).

Declarations under Rule 4.17:

of inventorship (Rule 4.17(iv))

Published:

- with international search report (Art. 21(3))
- in black and white; the international application as filed contained color or greyscale and is available for download from PATENTSCOPE





(57) Abstract: The disclosure provides for elastomers prepared at least from a composition comprising a polysiloxane based binder system, wherein said binder system comprises one or more hydroxyl functional polysiloxanes and one or more organic polysilozanes. The disclosure also provides for methods of producing such elastomers.

POLYSILOXANE-BASED ELASTOMERS AND METHODS OF PRODUCING SUCH

Technical field

The present disclosure relates to an elastomeric material comprising a polysiloxane based binder system, wherein said binder system comprises one or more hydroxyl functional polysiloxanes and one or more organic polysilazanes. The present disclosure also relates to methods of producing said elastomer, as well as products comprising said elastomer.

10 Background

5

15

20

25

Silicone elastomers are widely used materials due to their specific properties. These include low thermal conductivity, flexible mechanical properties, low chemical reactivity. low toxicity, thermal stability, their ability to form watertight seals and repel water, have strong adhesiveness to certain materials but do not adhere to others, do not support microbial growth, and resistance to oxidation, ozone, and UV light. These properties have lead to their widespread use in the electrical field (e.g. insulation), household (e.g., sealants and cooking utensils and containers), automobile (e.g. gaskets), airplane (e.g., seals), office machines (e.g. keyboard pads), medicine and dentistry (e.g. tooth impression moulds), textiles and paper. Accordingly, new polysiloxane based materials with favourable properties, for example with regards to the manufacture of articles using said materials, would be beneficial. For elastomeric polysiloxane based materials, curing speed and workability typically require the use of tin-based, or other metal-based catalysts and/or high temperatures. These may be unwanted due to health and safety considerations for the people working with the systems. Thus, there is also a need for development of new curing processes for polysiloxane based materials improving the safety conditions during handling and application.

Summary

The present disclosure provides for elastomeric materials having advantageous curing properties: the elastomeric materials cure without the need of metal-based catalysts, and without the need for elevated temperatures.

One aspect of the present disclosure provides an elastomer prepared at least from a composition comprising a polysiloxane based binder system, wherein said binder system comprises:

- a. one or more carbinol functional polysiloxanes; and
- b. one or more organic polysilazanes, wherein said one or more organic polysilazanes constitutes 0.5-25 parts (weight) relative to 100 parts (weight) of said one or more carbinol functional polysiloxanes, wherein the elastomer is self-supported.
- On aspect of the present disclosure provides for an elastomer prepared at least from a composition comprising a polysiloxane based binder system, wherein said binder system comprises:
 - a. one or more hydroxyl functional polysiloxanes; and
 - b. one or more organic polysilazanes;
- wherein said one or more organic polysilazanes constitutes 0.5-25 parts (weight) relative to 100 parts (weight) of said one or more hydroxyl functional polysiloxanes, wherein the elastomer is self-supported.

One aspect of the present disclosure provides for a kit of parts comprising

- a. a first composition comprising one or more hydroxyl functional polysiloxanes as disclosed herein; and
 - b. a second composition comprising one or more organic polysilazane as disclosed herein;

wherein said kit is for preparation of the elastomer as disclosed herein.

25

Another aspect of the present disclosure provides for a kit of parts comprising

- a. a first composition comprising one or more hydroxyl functional polysiloxanes as disclosed herein; and one or more organic polysilazanes as disclosed herein; and
- b. a second composition comprising one or more catalysts as disclosed herein;
- wherein said kit is for preparation of the elastomer as disclosed herein.

One aspect of the present disclosure provides for a product comprising the elastomer as disclosed herein.

Another aspect of the present disclosure provides for a method of preparing the self-supported elastomer of the disclosure, said method comprising mixing one or more of the carbinol functional polysiloxanes according to the disclosure and one or more of the organic polysilazanes according to the disclosure, wherein said one or more organic polysilazanes constitutes 0.5-25 parts (weight) relative to 100 parts (weight) of said one or more carbinol functional polysiloxanes.

Another aspect of the present disclosure provides for a method of preparing the self-supported elastomer as disclosed herein, said method comprising mixing one or more of the hydroxyl functional polysiloxanes as disclosed herein and one or more of the organic polysilazanes as disclosed herein, wherein said one or more organic polysilazanes constitutes 0.5-25 parts (weight) relative to 100 parts (weight) of said one or more hydroxyl functional polysiloxanes.

15 **Description of Drawings**

Figure 1: Storage modulus of each elastomer composition from rheology measurements.

Figure 2: Tensile measurements of selected elastomer compositions. The tested embodiments could be stretched 75-200 % before break.

20

25

30

5

10

Detailed description

Definitions

As used herein "elastomer" or "elastomeric" refer to materials that can reversibly extend. For example, the elastomer or elastomeric material can reversibly extend at least 5%, 10%, 15%, 20%, 25%, 30%, 40%, 50%, 60%, 70%, 80%, such as at least 90%. Most preferably, the elastomer or elastomeric material can reversibly extend at least 90%. When stretched to these extents the elastomers recover almost all of the original dimension, e.g. to within $\pm 20\%$, $\pm 15\%$, $\pm 10\%$, or $\pm 5\%$ of the original dimensions, e.g. original length or original width. Such recovery may occur over a relatively short time span, such as in less than 30 seconds.

As used herein, "self-supported" relates to object that is able to substantially maintain its shape without a container or a supporting element, such as an element onto which the object is adhered. In a specific embodiment, "self-supported" is taking to mean an

object that is not adhered onto a surface. In one embodiment, the self-supported object is a free-standing object, i.e. an object not adhered or attached to any other element. However, the self-supported object can after production be adhered to other elements. In the context of the invention, the self-supported object is not a coating, e.g. it is not a (thin) coating cast or cured onto the surface of another element, such as a thin coating cast or cured onto the surface of another element. The self-support is achieved through the specific mechanical properties of the elastomer.

As used herein, expressions such as "x to y" means at least x and at most y, that is, not more than y.

5

20

25

30

35

By "hydroxyl functional" is meant a chemical compound, e.g. a polymer such as a polysiloxane, comprising one or more hydroxyl (-OH) functional groups.

By "polysilazane" is mean a polymer compound that includes an Si-N bond, such as for example repeating units of Si-N.

In the present context, "optionally substituted" means that the indicated moiety may or may not be substituted, typically on carbon atoms, and when substituted, a carbon atom may be mono-, di-, or tri-substituted when valency allows. It is understood that where no substituents are indicated for an "optionally substituted" moiety, then the position is held by a hydrogen atom.

The term "2-30-membered linear or branched saturated or unsaturated chain" refers to a linear (i.e. unbranched) or branched hydrocarbon having in the chain from 2 up to 30 atoms, inclusive; and optionally having one or more double bonds. The chain generally consists of hydrocarbons and may optionally contain one or more linkers selected independently from -O-, -S-, -N-, -N-C(O)-, -O-C(O)-, -N-C(O)-N- and -N-C(O)-O-. In one sense, such linkers can be considered to have replaced a carbon atom in the chain. Chains solely consisting of hydrocarbon include linear or branched C_2 - C_{30} alkyl and linear or branched C_2 - C_{30} alkenyl. Examples of chains containing one or more linkers include for example ethers and polyethers.

The term "3-30-membered saturated or unsaturated ring system" refers to a monocyclic ring system having from 3 up to 30 ring atoms, inclusive; and optionally having one or

more double bonds. The ring generally consists of hydrocarbons and may optionally contain one or more linkers selected independently from -O-, -S-, -N-, -N-C(O)-, -O-C(O)-, -N-C(O)-N- and -N-C(O)-O-. Ring systems solely consisting of hydrocarbon include C_3 - C_{30} cycloalkyl; examples include (but are not limited to) monocyclic rings, fused ring systems, bridged ring systems and spirocyclic ring systems.

The term "aromatic ring system" in the present context refers to both monocyclic and fused C₆-C₃₀ membered aromatic rings. Examples include (but are not limited to) monocyclic aromatic rings such as for example benzene and bicyclic fused aromatic rings such as for example naphthalene.

The term "heteroaromatic ring system" in the present context refers to both monocyclic and fused C_5 - C_{30} membered aromatic rings containing at least one ring atom selected from O, N or S. Examples include (but are not limited to) monocyclic heteroaromatic rings such as for example furan, imidazole and pyridine and bicyclic fused aromatic rings such as for example indole, benzimidazole and quinoline.

The terms "halo" and "halogen" are used interchangeably and typically refer to fluorine, chlorine, bromine or iodine.

20

5

10

15

The terms "carbinol" and "silanol" are frequently used terms in connection with silicone chemistry. "carbinol" or "carbinol functional group "refers to a hydroxyl group bound to a carbon atom (C-OH) while "silanol" or "silanol functional group" refers to a hydroxyl group bound to a silicon atom (Si-OH).

25

In the general formulas (Ia), (Ib) and (II) used herein it should be understood that all R groups are independently selected, also within the repeating units indicated by n, m and p. Thus for example in formula (Ia) R_1 substituents may vary in between the repeating units (see also the illustrated polysiloxanes and polysilazanes used in the examples).

30

35

The term 'polysiloxane' is known in the art and refers to polymers having a backbone in which atoms of silicon and oxygen alternate, and which is devoid of carbon atoms (The New Encyclopedia Britannica in 30 volumes micropaedia volume IX. 1975 defining polysiloxane by referral to silicone). A polysiloxane backbone with organic (i.e. carbon-

based) substituent on the silicon atom may be referred to as a polyorganosiloxane. A commonly known polyorganosiloxane is polydimethyl- siloxane (PDMS).

The use of the terms "a" and "an" and "the" and similar referents in the context of describing the disclosure are to be construed to cover both the singular and the plural, unless otherwise indicated herein or clearly contradicted by context. For example, the phrase "the composition" is to be understood as referring to various "compositions" of the disclosure or particular described aspect, unless otherwise indicated.

The description herein of any aspect or aspect of the disclosure using terms such as "comprising", "having," "including" or "containing" with reference to an element or elements is intended to provide support for a similar aspect or aspect of the disclosure that "consists of", "consists essentially of" or "substantially comprises" that particular element or elements, unless otherwise stated or clearly contradicted by context (e.g., a composition described herein as comprising a particular element should be understood as also describing a composition consisting of that element, unless otherwise stated or clearly contradicted by context).

The terms "elastomer" and "elastomer composition" may be used interchangeably herein, unless otherwise specified.

Elastomers and polysiloxane based binder systems

5

20

25

30

35

The present disclosure relates to polysiloxane based elastomers. The disclosure also provides for a curing process with advantages over the typically used curing processes for polysiloxane based elastomer compositions.

The curing methodology disclosed herein relates to the use of an organic polysilazane curing agent which decomposes to condensate hydroxyl functional polysiloxanes. The curing reaction spontaneously proceeds at ambient temperature. This also allows for curing of elastomers without the use of a conventional catalyst, which would otherwise be incorporated in the elastomeric material.

Although the reaction is spontaneous, the curing rate can be further increased by addition of a catalyst, should the intended use of the elastomeric material formed therefrom allow this.

It has presently been found that non-metal, organic catalysts can increase the rate of curing. In this regard, the present inventors found that primary and secondary amines increase the curing of the presently claimed elastomers. One such amine is benzotriazole (BTZ). These catalysts are especially effective, even in small amounts, and do not suffer from the same environmental or health impact as other metal-based catalysts. The organic catalysts disclosed herein are particularly useful in curing of alcohol functional siloxanes.

The presently claimed elastomers are suitable as self-supported materials, that is, for production of articles that do not require a support to retain structural integrity.

The disclosed elastomers can be produced as bulk materials, allowing for casting of object with specific three-dimensional structures.

15

20

25

5

Some coating systems (e.g. not self-supported elastomers) combining silicone resins and polysilazane are known in the art. One example is JP2012153849 disclosing coating compositions comprising a polysilazane and a modified silicone oil and/or a curable silicone resin, for use in particular for vehicles, aircrafts, furniture, and outer walls of buildings. The polysilazane applied in the system is an inorganic polysilazane. These are generally known for being very reactive and thus, difficult to handle (see e.g. Barosso et al., Polysilazane-Based Coatings with Anti-Adherent Properties for Easy Release of Plastics and Composites from Metal Molds, Adv. Mater. Interfaces (2020) 7, 1901952). Organic polysilazanes applied in coatings have been disclosed for example in EP 3733801 and US 9676946. Both discloses coating compositions comprising the combination of polysilazane and silicone resins with polysilazane being the major component.

The elastomer composition of the current disclosure mainly constitute a polysiloxanebased binder system wherein organic polysilazane, used for promoting curing of the
system, only constitutes a minor amount of the composition. The elastomer
composition may be prepared from a range of different combinations of reactive
polysiloxanes and polysilazanes.

One embodiment of the present disclosure provides for an elastomer prepared at least from a composition comprising a polysiloxane based binder system, wherein said binder system comprises:

- a. one or more carbinol functional polysiloxanes; and
- b. one or more organic polysilazanes, wherein said one or more organic polysilazanes constitutes 0.5-25 parts (weight) relative to 100 parts (weight) of said one or more carbinol functional polysiloxanes, wherein the elastomer is self-supported.
- In one embodiment, the elastomer further comprises one or more silanol functional polysiloxanes.

One embodiment of the present disclosure provides for an elastomer prepared at least from a composition comprising a polysiloxane based binder system, wherein said binder system comprises:

a. one or more hydroxyl functional polysiloxanes; and

15

20

25

b. one or more organic polysilazanes;
 wherein said one or more organic polysilazanes constitutes 0.5-25 parts (weight)
 relative to 100 parts (weight) of said one or more hydroxyl functional polysiloxanes,
 wherein the elastomer is self-supported.

In one embodiment, said hydroxyl functional polysiloxane has the structure of formula (I):

$$R_{1} \longrightarrow Si \longrightarrow O \longrightarrow Si \longrightarrow O \longrightarrow Si \longrightarrow R_{1}$$

$$R_{1} \longrightarrow R_{1}$$

$$R_{1} \longrightarrow R_{1}$$

$$R_{1} \longrightarrow R_{1}$$
formula (I).

In embodiment, said hydroxyl functional polysiloxane has the structure of formula (Ia), (I):

5

10

15

20

25

30

wherein n is an integer from 5 to 7500; and wherein each R_1 are independently selected from

methyl; or

a 2-30-membered linear or branched saturated or unsaturated chain consisting of carbon atoms, optionally wherein one or more of said carbon atoms are replaced with a moiety independently selected from the group consisting of -O-, -S-, -N-, -N-C(O)-, -O-C(O)-, -N-C(O)-N- and -N-C(O)-O-; or

a 3-30-membered saturated or unsaturated ring system consisting of carbon atoms, optionally wherein one or more of said carbon atoms are replaced with a moiety independently selected from the group consisting of -O-, -S-, -N-, -N-C(O)-, -O-C(O)-, -N-C(O)-N- and -N-C(O)-O-; or an aromatic or heteroaromatic ring system;

wherein each R_1 may optionally be substituted one or more times with one or more substituents selected from halogen, hydroxyl, oxo and R_2 ; wherein each R_2 are independently selected from

methyl; or

a 2-30-membered linear or branched saturated or unsaturated chain consisting of carbon atoms, optionally wherein one or more of said carbon atoms are replaced with a moiety independently selected from the group consisting of -O-, -S-, -N-, -N-C(O)-, -O-C(O)-, -N-C(O)-N- and -N-C(O)-O-; or

a 3-30-membered saturated or unsaturated ring system consisting of carbon atoms, optionally wherein one or more of said carbon atoms are replaced with a moiety independently selected from the group consisting of -O-, -S-, -N-, -N-C(O)-, -O-C(O)-, -N-C(O)-N- and -N-C(O)-O-; or an aromatic or heteroaromatic ring system;

wherein said R₂ may optionally be substituted one or more times with halogen, hydroxyl and oxo; and

wherein each optionally substituted R_1 is having a Mw of <500 g/mol; wherein at least one R_1 and/or R_2 has at least one hydroxyl functional group.

In one embodiment, said hydroxyl functional polysiloxane has a molecular weight in the range of 750 to 500,000 g/mol.

In the context of the present disclosure, the polysiloxane binder system comprises one or more hydroxyl functional polysiloxane. The hydroxyl functional polysiloxane typically comprises one or more carbinol functional polysiloxane and/or one or more silanol functional polysiloxane. In a preferred embodiment, said polysiloxane comprises at least one or more carbinol functional polysiloxane.

In one embodiment, said one or more hydroxyl functional polysiloxanes comprise:

- a. one or more carbinol functional polysiloxanes, and/or
- b. one or more silanol functional polysiloxanes.

10

15

20

25

30

35

5

In one embodiment, said one or more hydroxyl functional polysiloxane comprises a1) one or more carbinol functional polysiloxane, and/or

a2) one or more silanol functional polysiloxane.

In a further embodiment said one or more hydroxyl functional polysiloxane comprises

- a1) one or more carbinol functional polysiloxane, and optionally
- a2) one or more silanol functional polysiloxane.

In one embodiment of the present disclosure, the binder system comprises one or more hydroxyl functional polysiloxanes in an amount of 75.0 to 99.5 % by dry weight. In one embodiment, the binder system comprises one or more hydroxyl functional polysiloxanes in an amount of 75 to 76 %, such as 76 to 77 %, such as 77 to 78 %, such as 78 to 79 %, such as 79 to 80 %, such as 80 to 81 %, such as 81 to 82 %, such as 82 to 83 %, such as 83 to 84 %, such as 84 to 85 %, such as 85 to 86 %, such as 86 to 87 %, such as 87 to 88 %, such as 88 to 89 %, such as 89 to 90 %, such as 90 to 91 %, such as 91 to 92 %, such as 92 to 93 %, such as 93 to 94 %, such as 94 to 95 %, such as 95 to 96 %, such as 96 to 97 %, such as 97 to 98 %, such as 98 to 99 %, such as 99 to 99.5 %.

In one embodiment, the binder system comprises one or more organic polysilazanes in an amount of 0.5 to 20.0 % by dry weight. In one embodiment, the binder system comprises one or more organic polysilazanes in an amount of 0.5 to 1.0 %, such as 1.0 to 1.5 %, such as 1.5 to 2.0 %, such as 2.0 to 2.5 %, such as 2.5 to 3.0 %, such as 3.0 to 3.5 %, such as 3.5 to 4.0 %, such as 4.0 to 4.5 %, such as 4.5 to 5.0 %, such as 5.0 to 5.5. %, such as 5.5 to 6.0 %, such as 6.0 to 6.5 %, such as 6.5 to 7.0 %, such as 7.0 to 7.5 %, such as 7.5 to 8.0 %. such as 8.0 to 8.5 %, such as 8.5 to 9.0 %, such as 9.0

to 9.5 %, such as 9.5 to 10.0 %, such as 10.0 to 11.0 %, such as 11.0 to 12.0 %, such as 12.0 to 13.0 %, such as 13.0 to 14.0 %, such as 15.0 to 16.0 %, such as 16.0 to 17.0 %, such as 17.0 to 18.0 %, such as 18.0 to 19.0 %, such as 19.0 to 20.0 %.

In one embodiment, the binder system comprises one or more carbinol functional polysiloxanes in an amount of 50 to 99.5 % by dry weight. In one embodiment, the binder system comprises one or more carbinol functional polysiloxanes in an amount of 75 to 99.5 % by dry weight. In one embodiment, the binder system comprises one or more carbinol functional polysiloxanes in an amount of 30 to 35 %, such as 35 to 40 %, such as 40 to 45 %, such as 45 to 50 %. such as 50 to 55 %, such as 55 to 60 %, such as 60 to 65 %, such as 65 to 70 %, such as 70 to 75 %, such as 75 to 80 %, such as 80 to 85 %, such as 85 to 90 %, such as 90 to 95 %, such as 95 to 99.5 %.

In one embodiment, the binder system comprises one or more hydroxyl functional polysiloxanes in an amount of 50 to 99.5 % by dry weight. In one embodiment, the binder system comprises one or more hydroxyl functional polysiloxanes in an amount of 75 to 99.5 % by dry weight. In one embodiment, the binder system comprises one or more hydroxyl functional polysiloxanes in an amount of 50 to 55 %, such as 55 to 60 %, such as 60 to 65 %, such as 65 to 70 %, such as 70 to 75 %, such as 75 to 80 %, such as 80 to 85 %, such as 85 to 90 %, such as 90 to 95 %, such as 95 to 99.5 %.

In one embodiment, said one or more organic polysilazane constitutes 1-25 parts (weight), such as 1-20 parts (weight), such as 1-15 parts (weight), such as 1-10 parts (weight), such as 2-10 parts (weight), such as 3-10 parts (weight), such as 4-10 parts (weight), such as or 0.5-9 parts (weight), such as 0.5-8 parts (weight), or 0.5-7 parts (weight), or 1-9 parts (weight), or 1-8 parts (weight), or 1-7 parts (weight) or 2-9 parts (weight), preferably 2-8 parts (weight), or 2-7 parts (weight) relative to 100 parts (weight) of said one or more hydroxyl functional polysiloxane.

In one embodiment, the binder system comprises one or more polysiloxanes in amount of 75 to 99.5 % by dry weight, and one or more organic polysilazanes in an amount of 0.5 to 10.0 % by dry weight. In one embodiment, the binder system comprises one or more polysiloxanes in amount of 75 to 99.5 % by dry weight, and one or more organic polysilazanes in an amount of 0.5 to 20.0 % by dry weight.

15

20

Commercially available hydroxyl functional polysiloxanes suitable for preparation of the elastomer of the disclosure include polydimethylsiloxanes (PDMS) from Gelest, such as DMS S14, S15, S21 and S31 from Gelest and Siltech S-701, S-5000 and S-20000 from Siltech.

5

10

Carbinols

In one embodiment, said one or more organic polysilazanes constitute 1-25 parts (weight), such as 1-20 parts (weight), such as 1-15 parts (weight), such as 1-10 parts (weight), such as 2-10 parts (weight), such as 3-10 parts (weight), such as 4-10 parts (weight), such as 0.5-9 parts (weight), such as 0.5-8 parts (weight), such as 0.5-7 parts (weight), such 1-9 parts (weight), such as 1-8 parts (weight), such as 1-7 parts (weight) such 2-9 parts (weight), such as 2-8 parts (weight), such as 2-7 parts (weight) relative to 100 parts (weight) of said one or more hydroxyl functional polysiloxanes.

In one embodiment, said carbinol functional polysiloxane has the structure of formula (Ia):

$$R_{1} \longrightarrow Si \longrightarrow O \longrightarrow Si \longrightarrow O \longrightarrow Si \longrightarrow R_{1}$$

$$R_{1} \longrightarrow R_{1}$$

$$R_{1} \longrightarrow R_{1}$$

$$R_{1} \longrightarrow R_{1}$$
formula (Ia).

In embodiment, said carbinol functional polysiloxane has the structure of formula (Ia), (Ia):

$$R_{1} \longrightarrow Si \longrightarrow O \longrightarrow Si \longrightarrow R_{1}$$

$$R_{1} \longrightarrow Si \longrightarrow O \longrightarrow Si \longrightarrow R_{1}$$

$$R_{1} \longrightarrow R_{1} \longrightarrow R_{1}$$

$$R_{1} \longrightarrow R_{1}$$
formula (Ia).

wherein n is an integer from 5 to 7500; and

wherein each R₁ are independently selected from

methyl; or

25

20

a 2-30-membered linear or branched saturated or unsaturated chain consisting of carbon atoms, optionally wherein one or more of said carbon atoms are replaced with a moiety independently selected from the group

5

15

20

25

consisting of -O-, -S-, -N-, -N-C(O)-, -O-C(O)-, -N-C(O)-N- and -N-C(O)-O-; or

a 3-30-membered saturated or unsaturated ring system consisting of carbon atoms, optionally wherein one or more of said carbon atoms are replaced with a moiety independently selected from the group consisting of -O-, -S-, -N-, -N-C(O)-, -O-C(O)-, -N-C(O)-N- and -N-C(O)-O-; or an aromatic or heteroaromatic ring system;

wherein each R₁ may optionally be substituted one or more times with one or more substituents selected from halogen, hydroxyl, oxo and R₂;

10 wherein each R₂ are independently selected from

methyl; or

a 2-30-membered linear or branched saturated or unsaturated chain consisting of carbon atoms, optionally wherein one or more of said carbon atoms are replaced with a moiety independently selected from the group consisting of -O-, -S-, -N-, -N-C(O)-, -O-C(O)-, -N-C(O)-N- and -N-C(O)-O-; or

a 3-30-membered saturated or unsaturated ring system consisting of carbon atoms, optionally wherein one or more of said carbon atoms are replaced with a moiety independently selected from the group consisting of -O-, -S-, -N-, -N-C(O)-, -O-C(O)-, -N-C(O)-N- and -N-C(O)-O-; or an aromatic or heteroaromatic ring system;

wherein said R₂ may optionally be substituted one or more times with halogen, hydroxyl and oxo; and

wherein each optionally substituted R₁ is having a Mw of <500 g/mol;

wherein at least one R₁ and/or R₂ has at least one carbinol functional group.

In one embodiment, said carbinol functional polysiloxane has a molecular weight in the range of 750 to 500,000 g/mol.

In one embodiment, the molecular weight of the compound having the structure of formula (Ia) is in the range of 1000 to 500,000 g/mol, such as 1000 to 100,000 g/mol, 5000 to 100,000 g/mol or 5000 to 50,000 g/mol.

In one embodiment, each optionally substituted R_1 is having a molecular weight of less than 450 g/mol, such as less than 400 g/mol, such as less than 350 g/mol, such as less than 300 g/mol.

In one embodiment, n is an integer from 5 to 5000; such as from 5 to 4000, such as from 5 to 3000, such as from 5 to 2000, preferably from 5 to 1000, such as from 20 to 1000, such as from 20 to 500.

In one embodiment, each R₁ is independently selected from unsubstituted methyl or an unsubstituted 2-10 membered linear or branched saturated or unsaturated chain consisting of carbon atoms, optionally wherein one or more of said carbon atoms are replaced with a moiety independently selected from the group consisting of -O-, -S-, -N-C(O)-, -C-O-C(O)- and -N-C(O)-O-. In one embodiment, said linker is -O-.

10

15

20

25

30

In one embodiment, each R_1 are independently selected from methyl; or a 2-30-membered linear or branched saturated or unsaturated chain consisting of carbon atoms and optionally containing one or more linkers selected independently from -O-, -S-, -N-C(O)-, -C-O-C(O)- and -N-C(O)-O-; or a 3-30-membered saturated or unsaturated ring system consisting of carbon atoms and optionally containing one or more linkers selected independently from -O-, -S-, -N-C(O)-; -C-O-C(O)- and -N-C(O)-O-; wherein said methyl and said 2-30-membered linear or branched saturated or unsaturated chain and said 3-30-membered saturated or unsaturated ring may optionally be substituted one or more times with one or more R_2 ; wherein each R_2 is independently selected from halogen, hydroxyl and oxo; with the proviso that at least one R_1 and/or R_2 has at least one carbinol functional group.

In a further embodiment, said carbinol functional polysiloxane is represented by formula (Ia') below having a molecular weight in the range of 750 to 500,000 g/mol:

$$R_{1} \xrightarrow{R_{1}} S_{i} \xrightarrow{R_{1}} O \xrightarrow{R_{1}} O \xrightarrow{R_{1}} S_{i} \xrightarrow{R_{1}} O \xrightarrow{R_{1}}$$

wherein (n+n'+n") is an integer from 5 to 7500; and

wherein (n'+n") is an integer from 0-50,

10

15

20

25

30

35

R₁ and R₂ are as defined for compound (Ia); and

any R_2 substituent on R_1 ' is denoted R_2 ', and R_1 ' and R_2 ' are as defined R_1 and R_2 in compound (Ia)

with the proviso that at least one of R_1 and/or R_2 is having at least one carbinol functional group and none of R_1 and R_2 can have a carbinol functional group.

In one embodiment, the molecular weight of the compound of formula (la') is in the range of 1000 to 500,000 g/mol, such as 1000 to 100,000 g/mol, 5000 to 100,000 g/mol or 5000 to 50,000 g/mol.

In one embodiment, (n+n'+n") is an integer from 5 to 5000; such as from 5 to 4000, such as from 5-3000, such as from 5-2000, preferably from 5 to 1000, more preferably from 20 to 1000 or from 20 to 500; and (n'+n") is an integer from 0-40, such as from 0-30, such as from 0-20, preferably from 0-10.

In the compound of formula (Ia') in one embodiment, at least two of R_1 ' and/or R_2 ' are having at least one carbinol functional group, or at least three of R_1 ' and/or R_2 ' is having at least one carbinol functional group, or at least one of R_1 ' and/or R_2 ' is having at least two carbinol functional groups, or at least one of R_1 ' and/or R_2 ' are having at least three carbinol functional groups, or at least two of R_1 ' and/or R_2 ' are having with at least two carbinol functional groups. In a further embodiment, the remaining of R_1 ' and/or R_2 ' are all selected from unsubstituted C_1 - C_6 alkyl, preferably unsubstituted methyl. In a further embodiment, R_1 are all selected independently from unsubstituted C_1 - C_6 alkyl, preferably all R_1 are unsubstituted methyl.

In one embodiment of the present disclosure:

groups; or

at least two of R_1 and/or R_2 are having at least one carbinol functional group; or at least three of R_1 and/or R_2 are having at least one carbinol functional group; or at least one of R_1 and/or R_2 are having at least two carbinol functional groups; or at least one of R_1 and/or R_2 are having at least three carbinol functional at least one of R_1 and/or R_2 are having at least three carbinol functional

at least two of R_1 and/or R_2 are having at least two carbinol functional groups.

In one embodiment, the compound having the structure of formula (Ia) has at least two carbinol functional groups, such as at least three carbinol functional groups, such as at least four carbinol functional groups.

In one embodiment, the number of carbinol groups in the carbinol functional polysiloxane is between 1 and 100, such as between 1 and 50, such as between 1 and 25, such as between 1 and 10, such as between 2 and 10, such as between 3 and 10.

<u>Silanols</u>

5

10

15

20

25

In one embodiment of the present disclosure, said silanol functional polysiloxane is represented by formula (lb):

In one embodiment of the present disclosure, said silanol functional polysiloxane is represented by formula (lb):

wherein m is an integer from 5 to 7500;

wherein each R_3 are independently selected from the group consisting of hydroxyl, methyl or linear or branched C_1 - C_{30} alkyl, linear or branched C_2 - C_{30} alkenyl, C_3 - C_{30} cycloalkyl or aryl;

wherein at least two R₃ groups are hydroxyl groups.

In one embodiment, said silanol functional polysiloxane has a molecular weight in the range of 750 to 500,000 g/mol.

In one embodiment, the molecular weight of the compound of formula (lb) is in the range of 1000 to 500,000 g/mol, such as 1000 to 100,000 g/mol, such as 5000 to 100,000 g/mol, such as 5000 to 50,000 g/mol.

- In one embodiment of the present disclosure, m is an integer from 5 to 5000; such as from 5 to 4000, such as from 5 to 3000, such as from 5 to 2000, such as from 5 to 1000, such as from 20 to 1000, such as from 20 to 500.
- In a preferred embodiment, the molecular weight of the compound of formula (lb) is in the range of 1000 to 500,000 g/mol such as 1000 to 100,000 g/mol, 5000 to 100,000 g/mol or 5000 to 50,000 g/mol, and wherein m is an integer from 5 to 5000; such as from 5 to 4000, such as from 5-3000, such as from 5-2000, preferably from 5 to 1000, more preferably from 20 to 1000 or from 20 to 500.
- In one embodiment, at least two of R₃ are hydroxyl. In one embodiment, at least two of R₃ are hydroxyl and the remaining R₃ are selected independently from linear or branched C₁-C₆ alkyl, C₂-C₆ alkenyl, C₃-C₆ cycloalkyl, or aryl.
- In a specific embodiment, at least two of R_3 are hydroxyl and the remaining R_3 are methyl.

In a further embodiment, said silanol functional polysiloxane is represented by formula (Ib') below having a molecular weight in the range of 750 to 500,000 g/mol:

wherein (m+m'+m") is an integer from 5 to 7500; and wherein (m'+m") is an integer from 0-50, and each R₃ and each R₃' are independently selected from hydroxyl, methyl or linear or branched C₁-C₃₀ alkyl, linear or branched C₂-C₃₀ alkenyl, C₃-C₃₀ cycloalkyl or aryl; R₃ is as defined for compound (lb); with the proviso that at least two R₃' groups are hydroxyl groups.

In one embodiment, the molecular weight of the compound of formula (lb') is in the range of 1000 to 500,000 g/mol, such as 1000 to 100,000 g/mol, 5000 to 100,000 g/mol or 5000 to 50,000 g/mol.

- In one embodiment, (m+m'+m") is an integer from 5 to 5000; such as from 5 to 4000, such as from 5-3000, such as from 5-2000, preferably from 5 to 1000, more preferably from 20 to 1000 or from 20 to 500; and (m'+m") is an integer from 0-40, such as from 0-30, such as from 0-20, preferably from 0-10.
- In the compound of formula (lb'), in one embodiment at least two R₃' are hydroxyl, and the remaining R₃' are selected independently from linear or branched C₁-C₆, preferably all the remaining R₃' are methyl, and all R₃ are selected independently from C₁-C₆ alkyl, preferably all R₃ are methyl.
- In the most preferred embodiment, two R_3 ' are hydroxyl and the remaining R_3 ' and all R_3 are all methyl.

In a preferred embodiment, the number of silanol groups in the silanol functional polysiloxane is between 1 and 100, such as between 1 and 50, such as between 1 and 25 or 1 and 10. Most preferably, the number of silanol groups is between 1 and 5, such as 1, 2, 3, or 4, such as 2.

Organic polysilazanes

20

25

30

The organic polysilazanes as disclosed herein are characterised in having Si-N bonds, specifically, the polysilazane comprise alternating silicon and nitrogen atoms in the backbone. The remaining positions on the silicium and nitrogen atoms may be substituted with organic moieties and/or hydrogen. In one embodiment, the organic polysilazane the silicium and/or nitrogen atom is substituted with at least one organic moiety.

In one embodiment of the present disclosure, said one or more organic polysilazane comprises a compound having the structure of formula (II), wherein the compound has a linear or branched or cyclic structure:

$$\begin{bmatrix} R_4 \\ -S_1 & N_- \\ -N_- & N_- \end{bmatrix}$$

$$\begin{bmatrix} R_4 & R_5 \end{bmatrix}^p \text{ formula (II)}$$

wherein p is an integer from 5 to 100;

wherein each R_4 are independently selected from hydrogen, linear or branched C_1 - C_6 alkenyl; and

5 each R₅ are independently selected from hydrogen, linear or branched C₁-C₆ alkyl or linear or branched C₁-C₆ alkenyl.

By linear, branched, or cyclic structures is meant that a plurality of the moieties of formula (II) are connected as to form a linear backbone, a branched backbone, or a cyclic backbone, or a combination of any of these three structures.

A compound in which all of R_4 and R_5 represent hydrogen atoms is generally referred to as an inorganic polysilazane or a perhydropolysilazane, while compounds containing at least one organic group as any one of R_4 and R_5 are generally referred to as organic polysilazanes or organopolysilazanes. In one embodiment, the polysilazane is an organic polysilazane. In a preferred embodiment, at least one of R_4 are selected from methyl, ethyl, and vinyl. In another preferred embodiment, at least one of R_4 is a vinyl. In a preferred embodiment, R_5 is hydrogen.

The polysilazane may be appropriately selected according to characteristics required for a cured product. Mixtures of one or more polysilazane may be used.

Commercially available polysilazanes suitable for the present disclosure include organic polysilazanes: Durazane 1033, Durazane 1066, Durazane 1800 from Merck, IOTA-OPSZ-9150 and IOTA-OPSZ-9108 from IOTA Silicone Oil (Anhui) Co. Ltd., and Octamethylcyclotetrasilazane from Merck, Germany.

Catalysts

10

15

25

30

In one embodiment of the present disclosure, the composition further comprises one or more catalysts. The catalyst is applied to accelerate the reaction between the one or more organic polysiloxanes and the one or more polysilazanes.

In one embodiment, said one or more catalysts comprise an organic amine.

In one embodiment, said one or more organic amines are primary or secondary amines.

5

10

15

In one embodiment, the catalyst is selected from inorganic acid compounds, organic acid compounds, inorganic base compounds and organic base compounds. In one embodiment, the catalyst is selected from the group consisting of hydrochloric acid, phosphoric acid, and sulfuric acid. In one embodiment, the catalyst is selected from the group consisting of p-toluenesulfonic acid, oxalic acid, acetic acid. In one embodiment, the catalyst is selected from the group consisting of ammonia, and sodium hydroxide. In one embodiment, the catalyst is selected from the group consisting of trimethylamine, triethylamine, tributylamine, 1,5-diazabicyclo[4.3.0]nonene-5 (DBN), 1,8-diazabicyclo[5.4.0]undecene-7 (DBU), 1,1,3,3-tetramethylguanidine, dimethylaminopyridine (DMAP), an azole, imidazole, an imidazole derivative, benzotriazole or a benzotriazole derivative. In one embodiment, the catalyst is a conjugated amine, such as a cyclic, conjugated amine. In one embodiment, the catalyst is a strained amine, e.g. ring-strain, such as a cyclic strained amine, such as a bicyclic strained amine, such as DBU.

20

25

As used herein, "azole" is a compound comprising a 5-membered ring comprising a nitrogen atom, said ring being saturated or unsaturated. In on embodiment, the azole is imidazole or an imidazole derivative. By "imidazole derivative" is mean imidazole substituted with one or more substituents. In one embodiment the azole is benzotriazole or a benzotriazole derivative. By "benzotriazole derivative" is meant benzotriazole substituted with one or more substituents.

30

In one embodiment, the catalyst is an organic base compound, for example an amine compound. In one preferred embodiment, said catalyst is a basic catalyst, more preferably an azole, such as imidazole or an imidazole derivative, such as benzotriazole or a benzotriazole derivative; or a cyclic amine such as DBU; or di-methyl amino pyridine (DMAP).

35

In one embodiment of the present disclosure, said one or more catalysts comprise a metal catalyst.

In one embodiment, said one or more metal catalysts is an organic metal catalyst.

5

10

15

20

25

30

35

In one embodiment of the present disclosure, the organic metal catalyst is selected from the group consisting of organic tin compounds, organic zinc compounds, organic titanium compounds, organic bismuth compounds, organic zirconium compounds, organic aluminum compounds, and organic nickel compounds. In one embodiment, the organic tin compounds is selected from the group consisting of dibutyltin dilaurate, dibutyltin dioctate, dibutyltin diacetate, dioctyltin dilaurate, dioctyltin dioctate, dioctyltin diacetate, dibutyltin bisacetylacetate. In one embodiment, the organic zinc compound is selected from the group consisting of zinc acetylacetonate, zinc-2-ethylhexoate, zinc naphthenate, zinc stearate. In one embodiment of the present disclosure, the organic bismuth compound is selected from the group consisting of bismuth 2-ethylhexanoate. bismuth octanoate, and bismuth neodecanoate. In one embodiment, the organic titanium compound is selected from the group consisting of tetrabutyl titanate, tetranonyl titanate, tetrakis ethylene glycol methyl ether titanate, and titanium diisopropoxide bis(acetylacetonate). In one embodiment, the zirconium compound is selected from the group consisting of zirconium tetraacetylacetonate, zirconium tributoxy acetylacetonate, zirconium dibutoxy diacetylacetonate, zirconium tetra propoxide, zirconium tetra butoxide, and zirconium octoate. In one embodiment, the organic aluminum compound is selected from the group consisting of aluminum salts such as aluminum octylate, aluminum triacetate, and aluminum tristearate; aluminum alkoxides such as aluminum trimethoxide, aluminum triethoxide, aluminum ethoxybis(acetylacetonate), aluminum butoxybis(ethylacetoacetate), aluminum dimethoxy (ethylacetoacetate), aluminum diisopropoxy (ethylacetoacetate), aluminum tris(ethylacetoacetate), and aluminum tris(acetylacetonate). In one embodiment, the nickel compound is selected from the group consisting of nickel (II) acetylacetonate, nickel (II) hexafluoroacetylacetonate hydrate.

The catalyst may be used alone or as combination of two or more catalysts. The amount of catalyst to be used is depending on the reactivity of the catalyst and the cross-linker(s) and desired curing time.

In one embodiment, the one or more catalysts constitute 0.001 to 0.5 parts relative to 100 parts (weight) of said one or more hydroxyl functional polysiloxanes.

In one embodiment, the one or more catalysts constitute 0.01 to 0.1 parts relative to 100 parts (weight) of said one or more hydroxyl functional polysiloxanes.

In one embodiment, the catalyst concentration is between 0.0001-5.0 %, e.g. 0.0001-2.0 %, or 0.005-5 %, or 0.01-4.0 %, or 0.1-3.0 %, by dry weight of the polysiloxane binder system; or in an amount of 0.0001-0.001%, or 0.0001-0.01% or 0.0001-1.0% by dry weight of the polysiloxane binder system. In one embodiment, the catalyst concentration is between 0.0001 and 0.001 %, such as between 0.001 and 0.01 %, such as between 0.001 and 0.01 %, such as between 0.1 and 0.5 %, such as between 0.5 and 2 % of the polysiloxane binder system.

A catalyst is preferably included when the hydroxyl functional polysiloxane mainly comprises silanol functional polysiloxane. In one embodiment, the hydroxyl functional polysiloxane is a silanol functional polysiloxane and the elastomer is prepared from a composition comprising a catalyst.

In one embodiment, said polysiloxane-based binder system constitutes at least 40 % by dry weight, such as 40-98 % by dry weight, such 45-95 % by dry weight, such as 50-95 % by dry weight, such as 50-90 % by dry weight, such as 55-90 % by dry weight, such as 60-90 % by dry weight, of the elastomer.

As shown in the disclosed examples, the curing of the elastomer can occur without the aid of a catalyst. Thus, in one embodiment of the present disclosure, said elastomer is free of catalyst. In one embodiment, said elastomer is free of metal catalyst. For certain applications, it is beneficial that the elastomer does not contain metal such as metal ions. In one embodiment, said elastomer is free of metal such as metal ions.

Fillers, pigments, additives, and solvents

15

20

25

The elastomers of the present disclosure may further comprise fillers, pigments, additives, and/or solvents. Fillers can assist in lowering the cost of materials while retaining the properties of the elastomer, or they may tune the mechanical or chemical properties of the elastomer.

Pigments and fillers are in the present context viewed in conjunction as constituents that may be included in the elastomer composition to further modify its properties. "Pigments" are normally characterised in that they render the final elastomer non-transparent and non-translucent, whereas "fillers" normally are characterised in that they do not render the elastomer non-translucent.

Examples of pigments are grades of titanium dioxide, red iron oxide, zinc oxide, carbon black, graphite, yellow iron oxide, red molybdate, yellow molybdate, zinc sulfide, antimony oxide, sodium aluminium sulfosilicates, quinacridones, phthalocyanine blue, phthalocyanine green, black iron oxide, indanthrone blue, cobalt aluminium oxide, carbazole dioxazine, chromium oxide, isoindoline orange, bis-acetoacet-o-tolidiole, benzimidazolon, quinaphtalone yellow, isoindoline yellow, tetrachloroisoindolinone, quinophthalone yellow.

Examples of fillers are calcium carbonate such as silica (including hydrophilic and hydrophobic silica), calcite, dolomite, talc, mica, feldspar, barium sulfate, kaolin, nephelin, perlite, magnesium oxide, and quartz flour.

In one embodiment, the filler silicone resin of type M, D, T, or Q, or combinations thereof.

Fillers and pigments may also be added in the form of nanotubes or fibers, thus, apart from the before-mentioned examples of fillers, the elastomer composition may also comprise fibers, e.g. those generally and specifically described in WO 00/77102.

Any pigments and/or fillers typically constitute 0-60 %, such as 0-50 %, preferably 5-45 %, such as 5-40 %, or 5-35 %, or 0.5-25 %, or 1-20 %, by dry weight of the elastomer. Taking into account the density of any pigments and/or fillers, such constituents typically constitute 0.2-20 %, such as 0.5-15 % by solids volume of the elastomer composition. In one embodiment, the elastomer comprises 5 to 30 %, such as 10 to 20 % of dry weight of the filler. In a specific embodiment, the filler is silica. In a further embodiment, the elastomer composition comprises hydrophobic silica in an amount of 0.5-30% by dry weight of the elastomer composition.

35 Examples of additives are:

5

10

20

25

- (i) non-reactive fluids such as non-reactive organopolysiloxanes; for example polydimethylsiloxane, methylphenyl polysiloxane; petroleum oils and combinations thereof;
- (ii) surfactants such as derivatives of propylene oxide or ethylene oxide such as alkylphenol-ethylene oxide condensates (alkylphenol ethoxylates); ethoxylated monoethanolamides of unsaturated fatty acids such as ethoxylated monoethanolamides of linoleic acid; sodium dodecyl sulfate; and soya lecithin.

Any additives typically constitute 0-30 %, such as 0-15 %, by dry weight of the elastomer composition.

Moreover, the elastomer composition may be supplemented with one or more solvents. Examples of solvents are aliphatic, cycloaliphatic and aromatic hydrocarbons such as white spirit, cyclohexane, toluene, xylene and naphtha solvent, esters such as methoxypropyl acetate, n-butyl acetate and 2-ethoxyethyl acetate; and mixtures thereof. Alternatively, the solvent system may include water or be water-based (>50% water in the solvent system). Preferably the solvents have a boiling point of 110 °C or more. The solvents, if any, typically constitute 5-50 % by volume of the elastomer composition.

20

15

5

Kits

The polysiloxane based binder system of the present disclosure may feasibly be provided as a kit of two or more parts, wherein when said parts are mixed, the elastomer forms.

25

30

35

One embodiment provides for a kit of parts comprising:

- a. a first composition comprising one or more carbinol functional polysiloxanes as disclosed herein; and
- a second composition comprising one or more organic polysilazane as disclosed herein;

wherein said kit is for preparation of the elastomer as disclosed herein.

One embodiment provides for a kit of parts comprising:

 a. a first composition comprising one or more hydroxyl functional polysiloxanes as disclosed herein; and b. a second composition comprising one or more organic polysilazanes as disclosed herein;

wherein said kit is for preparation of the elastomer disclosed herein.

5 In one embodiment of the present disclosure:

- a. said first composition further comprises one or more catalysts as disclosed herein;
- b. said kit further comprises a third composition comprising one or more catalysts as disclosed herein.

10

One embodiment provides for a kit of parts comprising:

- a. a first composition comprising one or more carbinol functional polysiloxane as disclosed herein; and one or more organic polysilazanes as disclosed herein; and
- b. a second composition comprising one or more catalysts as disclosed herein;
- wherein said kit is for preparation of the elastomer as disclosed herein.

One embodiment of the disclosure provides for a kit of parts comprising:

- a first composition comprising one or more hydroxyl functional polysiloxane as disclosed herein; and one or more organic polysilazanes as disclosed herein; and
- b. a second composition comprising one or more catalysts as disclosed herein; wherein said kit is for preparation of the elastomer as disclosed herein.

One embodiment of the present disclosure provides for a product comprising the elastomer disclosed herein.

25

35

The first composition, the second composition, and/or the third composition as described herein may be provided in a first container, a second container, and/or a third container, respectively.

30 Methods

The elastomer of the present disclosure can be manufactured using the methods disclosed herein.

The elastomer composition may be prepared by any suitable technique that is commonly used within the field of elastomer production. Thus, the various constituents

may be mixed together utilizing a mixer, a high speed disperser, a ball mill, a pearl mill, a grinder, a three-roll mill etc. The elastomer compositions may be prepared and shipped as two- or three-component systems that should be combined and thoroughly mixed immediately prior to use. Thus, the disclosure also relates to a kit of parts comprising two or three containers comprising components for the elastomer composition. In a separate embodiment, the elastomer composition is a one-component composition. Whether shipment must be performed in separate containers depends on the exact combination of polysiloxane and polysilazane.

- The elastomer compositions according to the disclosure may be filtrated using bag filters, patron filters, wire gap filters, wedge wire filters, metal edge filters, EGLM turnoclean filters (ex. Cuno), DELTA strain filters (ex. Cuno), and Jenag Strainer filters (ex. Jenag), or by vibration filtration.
- An example of a suitable preparation method is described in the Examples.

5

20

25

The elastomer composition is typically prepared by mixing two or more components e.g. two pre-mixtures, one pre-mixture comprising the one or more reactive polysiloxane and the optional catalyst, and one pre-mixture comprising the one or more polysilazane. Both pre-mixtures may optionally comprise components selected from solvents, additives, fillers and pigments.

It should be understood that when reference is made to the "elastomer composition", it refers to the mixed composition comprising all constituents including the polysiloxane and the polysilazanes and optional catalyst and solvent, additives, fillers, pigments. Furthermore, all amounts stated as % by dry weight of the elastomer composition should be understood as % by dry weight of the mixed elastomer composition ready to be applied, i.e. the weight apart from the solvents (if any).

One embodiment of the present disclosure provides for a method of preparing the selfsupported elastomer disclosed herein, said method comprising mixing one or more of the carbinol functional polysiloxanes disclosed herein and one or more of the organic polysilazanes disclosed herein, wherein said one or more organic polysilazanes constitutes 0.5-25 parts (weight) relative to 100 parts (weight) of said one or more carbinol functional polysiloxanes.

One embodiment of the present disclosure provides for a method of preparing the self-supported elastomer disclosed herein, said method comprising mixing one or more of the hydroxyl functional polysiloxanes disclosed herein and one or more of the organic polysilazanes disclosed herein, wherein said one or more organic polysilazanes constitutes 0.5-25 parts (weight) relative to 100 parts (weight) of said one or more hydroxyl functional polysiloxanes.

One embodiment of the present disclosure provides for a method of preparing a self-supported elastomer, said method comprising mixing one or more carbinol functional polysiloxanes as disclosed herein and one or more of organic polysilazanes as disclosed herein, wherein said one or more organic polysilazanes constitutes 0.5-25 parts (weight) relative to 100 parts (weight) of said one or more carbinol functional polysiloxanes.

15

20

25

30

10

5

One embodiment provides for a method of preparing a self-supported elastomer, said method comprising mixing one or more hydroxyl functional polysiloxanes as disclosed herein and one or more of organic polysilazanes as disclosed herein, wherein said one or more organic polysilazanes constitutes 0.5-25 parts (weight) relative to 100 parts (weight) of said one or more hydroxyl functional polysiloxanes.

In one embodiment, said one or more organic polysilazanes constitute 1-25 parts (weight), such as 1-20 parts (weight), such as 1-15 parts (weight), such as 1-10 parts (weight), such as 2-10 parts (weight), such as 3-10 parts (weight), such as 4-10 parts (weight), such as 0.5-9 parts (weight), such as 0.5-8 parts (weight), such as 0.5-7 parts (weight), such 1-9 parts (weight), such as 1-8 parts (weight), such as 1-7 parts (weight) such 2-9 parts (weight), such as 2-8 parts (weight), such as 2-7 parts (weight) relative to 100 parts (weight) of said one or more hydroxyl functional polysiloxanes.

In one embodiment, said method further comprises adding a catalyst as disclosed herein.

In one embodiment, said method does not comprise adding a catalyst, such as a catalyst disclosed herein.

In one embodiment, said method further comprises adding a solvent.

In one embodiment, said solvent is selected from the group consisting of aliphatic solvents, cycloaliphatic solvents, and aromatic hydrocarbons, such as white spirit, cyclohexane, toluene, xylene and naphtha solvent, esters such as methoxypropyl acetate, n-butyl acetate and 2-ethoxyethyl acetate, water, and mixtures thereof.

In one embodiment of the present disclosure, said solvent constitutes 5 to 50 % by volume of the mixture.

10

15

20

35

5

In one embodiment, said method further comprises adding a filler, a pigment, an additive, or a solvent as disclosed herein.

In one embodiment, the elastomer is cured in a mould. In one embodiment, said method further comprises a step of liberating the elastomer from the mould. In one embodiment, the method comprises using a mould-release agent to facilitate separation of the elastomer from the mould.

In one embodiment, the elastomer is cured in contact with another article to which it adheres during the curing.

In one embodiment, the elastomer is cured at a temperature between 0 and 220 °C. In one embodiment, the elastomer is cured at a temperature between 0 and 80 °C.

- In one embodiment, the elastomer is cured without heating the composition. However, it is to be understood that the composition can itself develop heat from the curing reaction. In one embodiment, "without heating" is understood as not providing any additional heat, such as by heating with a heating element or a flame.
- In one embodiment, the method comprises a first curing period and a second curing period.

In one embodiment, the method comprises the steps of:

 carrying out a first curing of the mixture for a first curing period to obtain a semicured elastomer,

- b. stretching or compressing the semi-cured elastomer, and
- c. carrying out a second curing of the semi-cured elastomer for a second curing period.
- In one embodiment, the first curing is carried out at a first temperature, and wherein the second curing is carried out at a second temperature, and wherein the second temperature is higher than the first temperature.

10 Examples

Example 1: Elastomer preparation and curing

Materials and methods

Tables 1 and 2 below indicate the identity of the polysiloxanes and polysilazanes applied in the examples.

Table 1: Polysiloxanes applied in the examples (available from Gelest, Siltech and Wacker).

Structure	Mw (g/mol)	Trade name	
$\begin{array}{c ccccccccccccccccccccccccccccccccccc$	5000	DMS-C21 ex. Gelest	
$\begin{array}{c ccccccccccccccccccccccccccccccccccc$	30000	Silmer OHT Di-400 ex. Siltech	
CH_3	30000	Silmer OH Di- 400 ex. Siltech	
$\begin{array}{c ccccccccccccccccccccccccccccccccccc$	11600	Silmer OH C50 ex. Siltech	

$\begin{array}{c ccccccccccccccccccccccccccccccccccc$	6100	Silmer OHT E13 ex. Siltech
H ₃ C—O CH ₃ CH ₃ CH ₃ O—CH ₃ O—Si—C Si—O—Si—O—Si—C Si—O CH ₃ CH ₃ CH ₃ O—CH ₃ H ₃ C O—CH ₃ CH ₃ CH ₃ CH ₃ CH ₃ O—CH ₃	1100	Silmer TMS Di-10 ex. Siltech
$\begin{array}{c ccccccccccccccccccccccccccccccccccc$	4200	DMS-S21 ex. Gelest
$\begin{array}{c ccccccccccccccccccccccccccccccccccc$	n.a.	C2T ex. Wacker

Table 2: Polysilazanes applied in the examples (available from Merck, Germany).

Structure	Trade name
$ \begin{array}{c c} CH_3 \\ N \\ N \\ N \\ M \end{array} $ $ \begin{array}{c} CH_3 \\ N \\ M \\ M \end{array} $ $ \begin{array}{c} CH_3 \\ N \\ M \\ M \end{array} $	Durazane 1800
Si—NH HN Si NH HN—Si HN—Si	1,2,3,4,5,6,7,8- octamethylcyclotetrasilazane

Pigments, fillers and solvents applied in the compositions are listed below:

5 <u>Pigments:</u>

Carbon black, Titanium dioxide, Iron oxide.

Fillers:

Calcium carbonate, Hydrophobic silica (HDK H2000 ex. Wacker),

PCT/EP2023/051379

Solvents:

Xylene, Butyl Acetate.

5 Catalysts:

Benzotriazole, ex. Merck

Triethylamine, ex. Merck

Diethylaminepropylamine, ex. Merck

Dimethylaminepropylamine, ex. Merck

10 Diethylaminopyridine, ex. Merck

Dimethylaminopyridine, ex. Merck

Diazabicyclo(5,4,0)undec-7-ene, ex. Merck

1-ethyl-1H-imidiazole, ex. Merck

1-methyl-1H-benzo(d)(1,2,3)triazole, ex. Merck

15 4,N,N-trimethylaniline, ex. Merck

Hexylamine, ex. Merck

Pyridine, ex. Merck

Propylamine, ex. Merck

2-ethylimidazole, ex. Merck

20

30

Preparation of the pure elastomer compositions:

Polysiloxanes were speed-mixed with solvent and catalyst for one minute at 3500 rpm. Thereafter polysilazane was mixed in by speed-mixing another 30 seconds.

25 Preparation of filled elastomer compositions:

Polysiloxane(s) were weighed in a large speed-mixing cup along with the hydrophobic silica filler. Filler to be added in increments, if above 10%. The two components were mixed at 3500rpm for 2 min to homogenize. This was followed by the addition of xylene and mixing for 1 min. Next step involved the addition of polysilazane and mixing for 30 sec. Next more xylene was added (0.5 mL) along with the catalyst Benzotriazole and mixing for 30 seconds. The composition is to be cast immediately after.

Application (casting) of the elastomer:

A clean Teflon mould was kept on a level surface, determined by the spirit level. The freshly prepared composition was poured into the mould and allowed to take its shape. The mould was covered with a tissue cut out to cover the top and secured by tape on four sides. The composition was kept under the fume hood for 48 hours. After this, the formed film was de-moulded and transferred onto a Teflon film before being placed in an oven at 80°C for 24 hours.

Drying time:

10 Drying time for the compositions was measured according to ASTM D5895-20.

Effect of catalyst

Composition D presented below in Table 3 was prepared following the description under preparation of coating compositions.

15

5

Table 3: Composition D

	D
Silmer OHT Di-400 (g)	1.00
Durazane 1800 (g)	0.05
Xylene (g)	0.5

Catalysts in different levels were added to composition D and the drying time (hours) was recorded. The results are shown in table 4.

Table 4: drying time with various catalysts.

		Amount of catalyst added						
	150 ppm*	1* 1200 ppm 3000 ppm		6000 ppm [–]				
Catalyst	Stage III (h)	Stage III (h)	Stage III (h)	Stage III (h)				
Benzotriazole	1.1	0.2		_				
Triethylamine		20.8	17.5	_				
Diethylaminepropylamine		14.4	1.9	_				
Dimethylaminepropylamine			13.2	12				
Dimethylaminopyridine		7.2	1.8	_				
Diazabicyclo(5,4,0)undec-7-	3.6	1.1						
ene				_				
1-ethyl-1H-imidiazole			6.4	-				
1-methyl-1H-			22.2	15.6				
benzo(d)(1,2,3)triazole				-				

4,N,N-trimethylaniline			12.9	_
Hexylamine		17.6	1.3	_
Pyridine		22.5	16.9	13.9
Propylamine		18.5	5.5	_
2-ethylimidazole		3.2	1.5	_
No catalyst	21	21	21	21

^{*}ppm: weight ppm relative to composition D

Table 4 indicates that it is possible to adjust the curing speed by varying the type and amount of catalyst and thereby obtain a curable composition that can meet the various demands.

Observation of curing

Tables 5a-f illustrate a variety of coating compositions. The coating compositions follow the general preparation of pure binder and coating compositions previously described.

Curing after 24 hours (yes/no) was evaluated by inspecting the films visually and by touch. A yes was given to coherent films, a no as given to compositions still in a liquid or viscous state.

Table 5a: Exemplified elastomer compositions.

Component		E	F	G	Н	I	J	K	L
Polysiloxane	Silmer OHT	3.05	3.025	3.045	3.02	2.131	2.121		
(g)	Di-400								
	Silmer OHT					0.0734			
	E13								
	Silmer OH						1.206	0.54	0.54
	C50								
	DMS-C21							0.45	0.45
Polysilazane	Durazane	0.084	0.151	0.3	0.454	0.126	0.169	0.03	0.05
(g)	1800								
Solvent (g)	Xylene							0.5	0.5
Catalyst (g)	Benzotriazole								
Cured	Y/N	Υ	Υ	Υ	Υ	Υ	Υ	Υ	Υ
within 24									
hours									

Table 5b: exemplified compositions

Component		М	N	0	Р	Q	R	S	Т
Polysiloxan	Silmer		1.0	1.0	1.0	1.0	1.0		
e (g)	OHT								
	Di-400								
	Silmer	0.54							
	ОН								
	C50								
	DMS-	0.45							
	C21								
	DMS-							1.01	1.03
	S21								
Polysilazan	Duraza	0.07	0.25	0.05	0.1	0.15	0.15		
e (g)	ne								
	1800								
Solvent (g)	Xylene	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5
Catalyst (g)	Benzo-							0.00015	0.00045
	triazole								
Cured within	Y/N	Υ	Υ	Υ	Υ	Υ	Υ	Υ	Υ
24 hours									

Tables 5a and 5b shows that the various hydroxyl functional siloxane polymers can be used to obtain the curable polysiloxane composition of this disclosure.

Table 5c: exemplified compositions

Component		U	V	Х	Υ	Z	AA	AB
Polysiloxane	Silmer OHT	1.04	1.05	1.02	1.01	1.02	1.06	1.04
(g)	Di-400							
Polysilazane	Durazane	0.06	0.06	0.06	0.06	0.05	0.05	0.05
(g)	1800							
Solvent (g)	Xylene	0.31	0.45	0.3	0.3	4.81		0.9
	Butyl acetate						4.22	
Filler (g)	CaCO3				0.339			

	Hydrophilic silica							0.4
	Hydrophobic silica					0.4	0.4	
Pigment (g)	TiO2			0.197	0.237	0.25	0.23	0.24
	Fe2O3	0.19			0.063			
		2						
	FeOOH					0.067	0.061	0.05
								9
	Carbon black		0.11					
Cured within	Y/N	Υ	Υ	Υ	Υ	Υ	Υ	Υ
24 hours								

Table 5d: exemplified compositions

Component		AC	AD	AE	AF	AG	АН
Polysiloxane	Silmer OHT Di-	1.01	1.01	1.04	1	1	1
(g)	400						
Polysilazane	Durazane 1800	0.05	0.05	0.05	0.06	0.06	0.06
(g)							
Solvent (g)	Xylene		0.3				
	Butyl acetate	1.7		0.3	0.6	0.5	0.4
Filler (g)	CaCO3		0.39	0.42			
	Hydrophilic	0.399			0.1	0.2	0.3
	silica						
Pigment (g)	TiO2	0.25	0.28	0.24	0.2	0.2	0.2
	FeOOH	0.066	0.061	0.061	0.06	0.06	0.06
Cured within	Y/N	Υ	Υ	Υ	Υ	Υ	Υ
24 hours							

Table 5e: exemplified elastomer compositions

Component		Al	AJ	AK	AL	AM	AN	AO
Polysiloxane	Silmer OHT Di-	1	1	1	1	1	1	2
(g)	400							
Polysilazane	Durazane 1800	0.06	0.06	0.06	0.06	0.06	0.06	0.1
(g)								
Solvent(g)	Xylene							1
	Butyl acetate	0.6	0.5	0.4	0.6	0.5	0.4	

Catalyst (g)	Benzotriazole							0.0003
Filler (g)	CaCO3				0.1	0.2	0.3	
	Hydrophobic	0.1	0.2	0.3				0.02
	silica							
Pigment (g)	TiO2	0.2	0.2	0.2	0.2	0.2	0.2	
	FeOOH	0.06	0.06	0.06	0.06	0.06	0.06	
	Carbon black							0.01
Cured within	Y/N	Υ	Υ	Υ	Υ	Υ	Υ	Υ
24 hours								

Tables 5c, 5d, and 5e indicates that multiple variations in common coating ingredients that can be used without compromising the curable polysiloxane-based composition.

5 Table 5f: Comparison of polysilazanes

Component		BB	BC -
Polysiloxane (g)	Silmer OH C50	2	2 -
	DMS-C21	1.68	1.68
Polysilazane (g)	Durazane 1800		0.168
	1,2,3,4,5,6,7,8-	0.17	
	OCTAMETHYLCYCLOTETRASILAZANE		_
Solvent (g)	Xylene	1.5	1
Catalyst (g)	Benzotriazole	0.00045	0.00045
Cured within 24	Y/N	Υ	Υ
hours			-

Table 5f indicates that various polysilazanes can be used to obtain a curable polysiloxane composition containing polysilazane.

10 Conclusion

These results demonstrate the compositional variety and curing properties of the compositions disclosed herein. While some compositions above have been cured onto a support, it is contemplated that the compositions can also be cured in a mould or similar to form a self-supported elastomer.

15

Example 2: Test and evaluation of elastomer properties

Materials and methods

Samples were prepared according to the description under preparation of coating compositions.

Rheology measurements

Samples were cured at ambient conditions in a Teflon mould (size) and cut in 25 mm discs of approx. 500 µm thickness. Measurements was obtained on a TA Instruments Discovery HR-1 Hybrid Rheometer with a 25 mm parallel steel plate geometry on a Peltier temperature controlled plate. Temperature was set for 25 °C and gap was software controlled to a constant axial force of 3 N. LVE region was determined with a strain sweep at 2.5 Hz between 0.1 to 10%. Frequency sweep was recorded in a logarithmic sweep between 100 and 0.01 Hz. Strain was set at 2.5% for softer samples and 0.5% for harder samples.

10

15

5

Tensile samples

After mixing the solution was cast in a Teflon mould. Samples were left to cure for 2 h, and then matured for 16 h at 100 °C. Samples were cut in dog bone shapes with a thickness of 60 mm length, 6 mm width and 0.5 mm before testing. Sample tensile stress-strain behavior was tested using an Instron 3340 materials testing system at room temperature, initially separated by a distance of 30 mm. The test specimen was elongated uniaxially at 20 mm min⁻¹.

Results

Tables 6a and 6b provide an overview of the studies elastomer compositions.

Table 6a: exemplified compositions

Component		E	F	G	Н	I	J
Polysiloxane	Silmer OHT	3.05	3.025	3.045	3.02	2.131	2.121
(g)	Di-400						
	Silmer OHT					0.0734	
	E13						
	Silmer OH						1.206
	C50						
	DMS-C21						
Polysilazane	Durazane	0.084	0.151	0.3	0.454	0.126	0.169
(g)	1800						
Solvent (g)	Xylene						
Catalyst (g)	Benzotriazole						

Cured within	Y/N	Υ	Υ	Υ	Υ	Υ	Υ
24 hours							

Table 6b: exemplified compositions

Composition		BD
Polysiloxane	Silmer OHT Di-400	3.019
	Silmer OHT E13	
	Silmer OH C50	
Polysilazane	Durazane 1800	0.155
Filler	Hydrophilic silica	0.151
Cure within	Y/N	Υ
24 hour		

Figure 1 shows the storage modulus of each elastomer composition from rheology measurements. Figure 2 shows tensile measurements of selected elastomer compositions. The compositions and the storage moduli before and after post curing of the four samples at 180°C for one hour are shown in table 7 below.

Table 7: Storage modulus for various compositions

Component		AX	AY	AZ	BA
					(comparative)
Polysiloxane (g)	Silmer OHT	7.3	7.3	7.3	6.0
	Di-400				
Polysilazane (g)	Durazane	0.3	0.6	0.9	3.6
	1800				
Solvent (g)	Xylene	3.65	3.65	3.65	3.0
Catalyst (g)	Benzotriazole	0.0005	0.0005	0.0005	0.002
Parts		4.1	8.2	12.3	60
polysilazane per					
100 parts					
polysiloxane					
(weight)					

Storage modulus	Before	15008	15513	19891	39209
G' (Pa)	heating				
	After heating	17830	25253	46196	194312
	Increase (%)	19	63	132	396

Preparation of elastomer compositions:

Polysiloxane(s) were weighed in a large speed-mixing cup along with the hydrophobic silica filler. Filler to be added in increments, if above 10%. The two components were mixed at 3500rpm for 2 min to homogenize. This was followed by the addition of xylene and mixing for 1 min. Next step involved the addition of polysilazane and mixing for 30 sec. Next more xylene was added (0.5 mL) along with the catalyst Benzotriazole and mixing for 30 seconds. The composition is to be cast immediately after.

Application (casting) of the elastomer

5

10 A clean Teflon mould was kept on a level surface, determined by the spirit level. The freshly prepared composition was poured into the mould and allowed to take its shape. The mould was covered with a tissue cut out to cover the top and secured by tape on four sides. The composition was kept under the fume hood for 48 hours. After this, the formed film was de-moulded and transferred onto a Teflon film before being placed in 15 an oven at 80°C for 24 hours.

Die punch was used to prepare dumbbell samples for tensile testing. Compositions are shown in Tables 8-10.

Table 8: Filled elastomer compositions

Component		B4	B5	B6	B7
Polysiloxane (g)	Silmer OHT Di-	6.86	6.87	6.86	6.89
	400				
	Silmer OH J10	-			
	Gelest DMS S31	-			
Polysilazane (g)	Durazane 1800	0.34	0.34	0.34	0.34
Solvent (ml)	Xylene	6.8	6.8	6.8	3.4
Silica (g)	Hydrophobic	0.686	0.686	0.514	0.343
	silica				
Catalyst (mg)	BTZ	0	0.343	0.343	0.343

Component		B8	B10	B11	B12	B13
Polysiloxane (g)	Silmer OHT Di-	6.86	6.86	6.86	5.7	3.6
	400					
	Silmer OH J10	-	-	-	-	-
	Gelest DMS	-	-	-	1.52	3.703
	S32					
Polysilazane (g)	Durazane 1800	0.34	0.34	0.34	0.316	0.257
Solvent (ml)	Xylene	3.2	9.5	9.5	9.5	9.5
Silica (g)	Hydrophobic	0.171	1.37	2.06	1.44	1.46
	silica					
Catalyst (mg)	BTZ	0.343	0.343	0.343	0.343	0.343

Table 10: Mechanical properties of various elastomer compositions

#	Modulus	Std	EB	Std	UT	Std
	(MPa)	(MPa)	(%)	(%)	(MPa)	(MPa)
B4	1.20	0.16	133.20	33.00	1.28	0.29
B5	1.36	0.19	156.02	34.83	1.55	0.36
B6	1.01	0.07	139.42	19.46	0.95	0.13
B7	0.66	0.07	162.84	29.86	0.67	0.13
B8	0.75	0.05	99.04	13.16	0.74	0.10
B10	2.38	0.18	179.72	40.73	3.67	0.83
B11	3.62	0.25	131.18	19.84	3.32	0.50
B12	1.44	0.13	147.52	19.44	1.95	0.23
B13	0.81	0.08	182.40	30.88	1.27	0.30

Conclusion

5

The tested compositions show elastomeric properties and that the chemical composition can be used to vary the mechanical properties of the elastomers. Similarly filled systems have the expected effects as reinforcing agents and can be applied as for normal silicone systems.

In addition, Table 7 shows that higher amounts of the polysilazane in the composition can lead to a two-stage curing profile – compare AX, AY, and AZ. This is particularly beneficial for curing of elastomers, where a two-stage curing can be exploited to perform and fix a pre-stretch of the elastomer. Specifically, initial curing allows handling and stretching of the material, while a subsequent second stage curing of the stretched elastomer at higher temperature (or alternatively allowing ambient atmosphere post-curing) will add a second level of crosslinks to the elastomer. This system, which is reliant on only a single curing agent (polysilazane) can prove advantageous over other known systems, which may rely on two different curing agents to achieve curing in two

41

Items

stages.

- 1. An elastomer prepared at least from a composition comprising a polysiloxane based binder system, wherein said binder system comprises:
 - a. one or more carbinol functional polysiloxanes; and
 - b. one or more organic polysilazanes,

wherein said one or more organic polysilazanes constitutes 0.5-25 parts (weight) relative to 100 parts (weight) of said one or more carbinol functional polysiloxanes, wherein the elastomer is self-supported.

20

25

30

35

5

10

15

- 2. The elastomer according to item 1, wherein the elastomer further comprises one or more silanol functional polysiloxanes.
- 3. An elastomer prepared at least from a composition comprising a polysiloxane based binder system, wherein said binder system comprises:
 - a. one or more hydroxyl functional polysiloxanes; and
 - b. one or more organic polysilazanes;
 wherein said one or more organic polysilazanes constitutes 0.5-25 parts
 (weight) relative to 100 parts (weight) of said one or more hydroxyl functional polysiloxanes,

wherein the elastomer is self-supported.

- 4. The elastomer according to any one of the preceding items, wherein said one or more hydroxyl functional polysiloxanes comprise:
 - a. one or more carbinol functional polysiloxanes, and/or

5

10

15

25

30

- b. one or more silanol functional polysiloxanes.
- 5. The elastomer according to any one of the preceding items, wherein said one or more organic polysilazanes constitute 1-25 parts (weight), such as 1-20 parts (weight), such as 1-15 parts (weight), such as 1-10 parts (weight), such as 2-10 parts (weight), such as 3-10 parts (weight), such as 4-10 parts (weight), such as 0.5-9 parts (weight), such as 0.5-8 parts (weight), such as 0.5-7 parts (weight), such 1-9 parts (weight), such as 1-8 parts (weight), such as 1-7 parts (weight) such 2-9 parts (weight), such as 2-8 parts (weight), such as 2-7 parts (weight) relative to 100 parts (weight) of said one or more hydroxyl functional polysiloxanes.
- 6. The elastomer according to any one of the preceding items, wherein said carbinol functional polysiloxane has the structure of formula (Ia) having a molecular weight in the range of 750 to 500,000 g/mol:

 $R_{1} \longrightarrow Si \longrightarrow O \longrightarrow Si \longrightarrow O \longrightarrow Si \longrightarrow R_{1}$ $R_{1} \longrightarrow R_{1} \longrightarrow R_{1} \longrightarrow R_{1}$ $R_{1} \longrightarrow R_{1} \longrightarrow R_{1}$ formula (Ia),

wherein n is an integer from 5 to 7500; and

wherein each R₁ are independently selected from

methyl; or

a 2-30-membered linear or branched saturated or unsaturated chain consisting of carbon atoms, optionally wherein one or more of said carbon atoms are replaced with a moiety independently selected from the group consisting of -O-, -S-, -N-, -N-C(O)-, -O-C(O)-, -N-C(O)-N- and -N-C(O)-

O- ; or

a 3-30-membered saturated or unsaturated ring system consisting of carbon atoms, optionally wherein one or more of said carbon atoms are replaced with a moiety independently selected from the group consisting of -O-, -S-, -N-, -N-C(O)-, -O-C(O)-, -N-C(O)-N- and -N-C(O)-O-; or an aromatic or heteroaromatic ring system;

wherein each R_1 may optionally be substituted one or more times with one or more substituents selected from halogen, hydroxyl, oxo and R_2 ; wherein each R_2 are independently selected from

methyl; or

5

10

15

20

25

a 2-30-membered linear or branched saturated or unsaturated chain consisting of carbon atoms, optionally wherein one or more of said carbon atoms are replaced with a moiety independently selected from the group consisting of -O-, -S-, -N-, -N-C(O)-, -O-C(O)-, -N-C(O)-Nand -N-C(O)-O-; or

a 3-30-membered saturated or unsaturated ring system consisting of carbon atoms, optionally wherein one or more of said carbon atoms are replaced with a moiety independently selected from the group consisting of -O-, -S-, -N-, -N-C(O)-, -O-C(O)-, -N-C(O)-N- and -N-C(O)-O-; or an aromatic or heteroaromatic ring system;

wherein said R₂ may optionally be substituted one or more times with halogen, hydroxyl and oxo; and wherein each optionally substituted R₁ is having a Mw of <500 g/mol; wherein at least one R₁ and/or R₂ has at least one carbinol functional group.

- 7. The elastomer according to any one of the preceding items, wherein the molecular weight of the compound having the structure of formula (Ia) is in the range of 1000 to 500,000 g/mol, such as 1000 to 100,000 g/mol, 5000 to 100,000 g/mol or 5000 to 50,000 g/mol.
- 8. The elastomer according to any one of the preceding items, wherein each optionally substituted R₁ is having a molecular weight of less than 450 g/mol, such as less than 400 g/mol, such as less than 350 g/mol, such as less than 300 g/mol.
- 9. The elastomer according to any one of the preceding items, wherein n is an integer from 5 to 5000; such as from 5 to 4000, such as from 5 to 3000, such as from 5 to 2000, preferably from 5 to 1000, such as from 20 to 1000, such as from 20 to 500.
- 30 10. The elastomer according to any one of the preceding items, wherein each R₁ is independently selected from unsubstituted methyl or an unsubstituted 2-10 membered linear or branched saturated or unsaturated chain consisting of carbon atoms, optionally wherein one or more of said carbon atoms are replaced with a moiety independently selected from the group consisting of -O-, -S-, -N-C(O)-, -C-35 O-C(O)- and -N-C(O)-O-.

11. The elastomer according to any one of the preceding items, wherein

5

10

15

20

25

30

at least two of R_1 and/or R_2 are having at least one carbinol functional group; or

at least three of R_1 and/or R_2 are having at least one carbinol functional group; or

at least one of R_1 and/or R_2 are having at least two carbinol functional groups; or

at least one of R₁ and/or R₂ are having at least three carbinol functional groups; or

at least two of R_1 and/or R_2 are having at least two carbinol functional groups.

- 12. The elastomer according to any one of the preceding items, wherein the compound having the structure of formula (Ia) has at least two carbinol functional groups, such as at least three carbinol functional groups, such as at least four carbinol functional groups.
- 13. The elastomer according to any one of the preceding items, wherein the number of carbinol groups in the carbinol functional polysiloxane is between 1 and 100, such as between 1 and 50, such as between 1 and 25, such as between 1 and 10.
 - 14. The elastomer according to any one of the preceding items, wherein said silanol functional polysiloxane is represented by formula (lb) below having a molecular weight in the range of 750 to 500,000 g/mol:

$$R_3$$
 R_3
 R_3

wherein m is an integer from 5 to 7500;

wherein each R_3 are independently selected from the group consisting of hydroxyl, methyl or linear or branched C_1 - C_{30} alkyl, linear or branched C_2 - C_{30} alkenyl, C_3 - C_{30} cycloalkyl or aryl;

wherein at least two R₃ groups are hydroxyl groups.

WO 2023/139218 PCT/EP2023/051379

15. The elastomer according to any one of the preceding items, wherein the molecular weight of the compound of formula (Ib) is in the range of 1000 to 500,000 g/mol, such as 1000 to 100,000 g/mol, such as 5000 to 100,000 g/mol, such as 5000 to 50,000 g/mol.

5

- 16. The elastomer according to any one of the preceding items, wherein m is an integer from 5 to 5000; such as from 5 to 4000, such as from 5 to 3000, such as from 5 to 2000, such as from 5 to 1000, such as from 20 to 1000, such as from 20 to 500.
- 17. The elastomer according to any one of the preceding items, wherein at least two of R₃ are hydroxyl and the remaining R₃ are selected independently from linear or branched C₁-C₆ alkyl, C₂-C₆ alkenyl, C₃-C₆ cycloalkyl, or aryl.
 - 18. The elastomer according to any one of the preceding items, wherein at least two of R₃ are hydroxyl and the remaining R₃ are methyl.
 - 19. The elastomer according to any one of the preceding items, wherein said one or more organic polysilazane comprises a compound having the structure of formula (II), wherein the compound has a linear or branched or cyclic structure:

20

25

15

wherein p is an integer from 5 to 100;

wherein each R_4 are independently selected from hydrogen, linear or branched C_1 - C_6 alkyl or linear or branched C_1 - C_6 alkenyl; and

each R_5 are independently selected from hydrogen, linear or branched C_1 - C_6 alkyl or linear or branched C_1 - C_6 alkenyl;

wherein at least one R₄ is not hydrogen.

20. The elastomer according to any one of the preceding items, wherein the composition further comprising one or more catalysts.

30

21. The elastomer according to any one of the preceding items, wherein said one or more catalysts comprise an organic amine.

- 22. The elastomer according to any one of the preceding items, wherein said one or more organic amines are primary or secondary amines.
- 5 23. The elastomer according to any one of the preceding items, wherein said organic amine is selected from the group consisting of trimethylamine, triethylamine, tributylamine, 1,5-diazabicyclo[4.3.0]non-5-ene (DBN), 1,8-diazabicyclo[5.4.0]undec-7-ene (DBU), 1,1,3,3-tetramethylguanidine, 4-dimethylaminopyridine (DMAP), an azole, imidazole, an imidazole derivative, benzotriazole, and a benzotriazole derivative.
 - 24. The elastomer according to any one of the preceding items, wherein said one or more catalysts comprise a metal catalyst.
- 15 25. The elastomer according to any one of the preceding items, wherein said one or more metal catalysts is an organic metal catalyst.
 - 26. The elastomer according to any one of the preceding items, wherein said metal catalyst is selected from the group consisting of organic tin compounds, organic zinc compounds, organic titanium compounds, organic zirconium compounds, organic aluminum compounds, and organic nickel compounds.
 - 27. The elastomer according to any one of the preceding items, wherein said elastomer is free of catalyst.
 - 28. The elastomer according to any one of the preceding items, wherein said elastomer is free of metal catalyst.
 - 29. The elastomer according to any one of the preceding items, wherein said elastomer is free of metal.
 - 30. The elastomer according to any one of the preceding items, wherein the one or more catalysts constitute 0.001 to 0.5 parts relative to 100 parts (weight) of said one or more hydroxyl functional polysiloxanes.

20

25

- 31. The elastomer according to any one of the preceding items, wherein the one or more catalysts constitute 0.01 to 0.1 parts relative to 100 parts (weight) of said one or more hydroxyl functional polysiloxanes.
- 5 32. The elastomer according to any one of the preceding items, wherein said polysiloxane-based binder system constitutes at least 40 % by dry weight, such as 40-98 % by dry weight, such 45-95 % by dry weight, such as 50-95 % by dry weight, such as 50-90 % by dry weight, such as 55-90 % by dry weight, such as 60-90 % by dry weight, of the elastomer.

10

- 33. The elastomer according to any one of the preceding items, wherein the elastomer further comprises one or more of a filler, a pigment, an additive, or a solvent.
- 34. The elastomer according to any one of the preceding items, wherein the elastomer comprises 5 to 30 %, such as 10 to 20 % of dry weight of the filler.
 - 35. The elastomer according to any one of the preceding items, wherein the filler is silica.

20 36. A kit of parts comprising:

- a. a first composition comprising one or more carbinol functional polysiloxanes according to any one of the preceding items; and
- b. a second composition comprising one or more organic polysilazane according to any one of the preceding items;
- wherein said kit is for preparation of the elastomer according to any one of the preceding items.

37. A kit of parts comprising:

30

25

- a first composition comprising one or more hydroxyl functional polysiloxanes
 according to any one of the preceding items; and
- b. a second composition comprising one or more organic polysilazane according to any one of the preceding items;

wherein said kit is for preparation of the elastomer according to any one of the preceding items.

- 38. The kit according to any one of the preceding items, wherein:
 - said first composition further comprises one or more catalysts according to any one of the preceding items; or
 - b. said kit further comprises a third composition comprising one or more catalysts according to any one of the preceding items.

39. A kit of parts comprising

- a first composition comprising one or more carbinol functional polysiloxane according to any one of the preceding items; and one or more organic polysilazanes according to any one of the preceding items; and
- a second composition comprising one or more catalysts according to any one of the preceding items;

wherein said kit is for preparation of the elastomer according to any one of the preceding items.

15

20

30

35

10

5

40. A kit of parts comprising

- a first composition comprising one or more hydroxyl functional polysiloxane according to any one of the preceding items; and one or more organic polysilazanes according to any one of the preceding items; and
- a second composition comprising one or more catalysts according to any one of the preceding items;

wherein said kit is for preparation of the elastomer according to any one of the preceding items.

- 25 41. A product comprising the elastomer according to any one of the preceding items.
 - 42. A method of preparing the self-supported elastomer according to any one of the preceding items, said method comprising mixing one or more of the carbinol functional polysiloxanes according to any one of the preceding items and one or more of the organic polysilazanes according to any one of the preceding items, wherein said one or more organic polysilazanes constitutes 0.5-25 parts (weight) relative to 100 parts (weight) of said one or more carbinol functional polysiloxanes.
 - 43. A method of preparing the self-supported elastomer according to any one of the preceding items, said method comprising mixing one or more of the hydroxyl

functional polysiloxanes according to any one of the preceding items and one or more of the organic polysilazanes according to any one of the preceding items, wherein said one or more organic polysilazanes constitutes 0.5-25 parts (weight) relative to 100 parts (weight) of said one or more hydroxyl functional polysiloxanes.

5

10

15

- 44. A method of preparing a self-supported elastomer, said method comprising mixing one or more carbinol functional polysiloxanes according to any of the preceding items and one or more of organic polysilazanes according to any one of the preceding items, wherein said one or more organic polysilazanes constitutes 0.5-25 parts (weight) relative to 100 parts (weight) of said one or more carbinol functional polysiloxanes.
- 45. A method of preparing a self-supported elastomer, said method comprising mixing one or more hydroxyl functional polysiloxanes according to any one of the preceding items and one or more of organic polysilazanes according to any one of the preceding items, wherein said one or more organic polysilazanes constitutes 0.5-25 parts (weight) relative to 100 parts (weight) of said one or more hydroxyl functional polysiloxanes.
- 46. The method according to any one of the preceding items, wherein said one or more organic polysilazanes constitute 1-25 parts (weight), such as 1-20 parts (weight), such as 1-15 parts (weight), such as 1-10 parts (weight), such as 2-10 parts (weight), such as 3-10 parts (weight), such as 4-10 parts (weight), such as 0.5-9 parts (weight), such as 0.5-8 parts (weight), such as 0.5-7 parts (weight), such 1-9 parts (weight), such as 1-8 parts (weight), such as 1-7 parts (weight) such 2-9 parts (weight), such as 2-8 parts (weight), such as 2-7 parts (weight) relative to 100 parts (weight) of said one or more hydroxyl functional polysiloxanes.
 - 47. The method according to any one of the preceding items, said method further comprising adding a catalyst according to any one of the preceding items.
 - 48. The method according to any one of the preceding items, said method not comprising adding a catalyst according to any one of the preceding items.

WO 2023/139218 PCT/EP2023/051379

- 49. The method according to any one of the preceding items, said method further comprising adding a solvent.
- 50. The method according to any one of the preceding items, wherein said solvent is selected from the group consisting of aliphatic solvents, cycloaliphatic solvents, and aromatic hydrocarbons, such as white spirit, cyclohexane, toluene, xylene and naphtha solvent, esters such as methoxypropyl acetate, n-butyl acetate and 2-ethoxyethyl acetate, water, and mixtures thereof.
- 10 51. The method according to any one of the preceding items, wherein said solvent constitutes 5 to 50 % by volume of the mixture.
 - 52. The method according to any one of the preceding items, said method further comprising adding a filler, a pigment, an additive, or a solvent.
 - 53. The method according to any one of the preceding items, wherein the elastomer is cured in a mould.
- 54. The method according to any one of the preceding items, said method further comprising a step of liberating the elastomer from the mould.

15

- 55. The method according to any one of the preceding items, wherein the elastomer is cured at a temperature between 0 and 220 °C.
- 25 56. The method according to any one of the preceding items, wherein the elastomer is cured at a temperature between 0 and 80 °C.
 - 57. The method according to any one of the preceding items, wherein the elastomer is cured without heating the composition.
 - 58. The method according to any one of the preceding items, wherein the method comprises a first curing period and a second curing period.
- 59. The method according to any one of the preceding items, wherein the method comprises the steps of:

- a. carrying out a first curing of the mixture for a first curing period to obtain a semi-cured elastomer,
- b. stretching or compressing the semi-cured elastomer, and

5

- c. carrying out a second curing of the semi-cured elastomer for a second curing period.
- 60. The method according to any one of the preceding items, wherein the first curing is carried out at a first temperature, and wherein the second curing is carried out at a second temperature, and wherein the second temperature is higher than the first temperature.

Claims

1. An elastomer prepared at least from a composition comprising a polysiloxane based binder system, wherein said binder system comprises:

52

- a. one or more carbinol functional polysiloxanes; and
- b. one or more organic polysilazanes,

wherein said one or more organic polysilazanes constitutes 0.5-25 parts (weight) relative to 100 parts (weight) of said one or more carbinol functional polysiloxanes, wherein the elastomer is self-supported.

10

5

- 2. The elastomer according to claim 1, wherein the elastomer further comprises one or more silanol functional polysiloxanes.
- 3. The elastomer according to any one of claims 1 or 2, wherein said one or more organic polysilazanes constitute 1-10 parts (weight), such as 2-10 parts (weight), such as 3-10 parts (weight), such as 4-10 parts (weight), such as 0.5-9 parts (weight), such as 0.5-8 parts (weight), such as 0.5-7 parts (weight), such 1-9 parts (weight), such as 1-8 parts (weight), such as 1-7 parts (weight) such 2-9 parts (weight), such as 2-8 parts (weight), such as 2-7 parts (weight) relative to 100 parts (weight) of said one or more carbinol functional polysiloxanes.
 - 4. The elastomer according to any one of the preceding claims, wherein said carbinol functional polysiloxane has the structure of formula (la) having a molecular weight in the range of 750 to 500,000 g/mol:

25

$$R_1 \longrightarrow Si \longrightarrow O \longrightarrow Si \longrightarrow R_1$$

$$R_1 \longrightarrow Si \longrightarrow O \longrightarrow Si \longrightarrow R_1$$

$$R_1 \longrightarrow R_1$$

$$R_1 \longrightarrow R_1$$

$$R_1 \longrightarrow R_1$$
formula (la

wherein n is an integer from 5 to 7500; and

wherein each R₁ are independently selected from

methyl; or

30

a 2-30-membered linear or branched saturated or unsaturated chain consisting of carbon atoms, optionally wherein one or more of said carbon atoms are replaced with a moiety independently selected from the group

consisting of -O-, -S-, -N-, -N-C(O)-, -O-C(O)-, -N-C(O)-N- and -N-C(O)-O- ; or

a 3-30-membered saturated or unsaturated ring system consisting of carbon atoms, optionally wherein one or more of said carbon atoms are replaced with a moiety independently selected from the group consisting of -O-, -S-, -N-, -N-C(O)-, -O-C(O)-, -N-C(O)-N- and -N-C(O)-O-; or an aromatic or heteroaromatic ring system;

wherein each R_1 may optionally be substituted one or more times with one or more substituents selected from halogen, hydroxyl, oxo and R_2 ;

wherein each R₂ are independently selected from

methyl; or

a 2-30-membered linear or branched saturated or unsaturated chain consisting of carbon atoms, optionally wherein one or more of said carbon atoms are replaced with a moiety independently selected from the group consisting of -O-, -S-, -N-, -N-C(O)-, -O-C(O)-, -N-C(O)-N- and -N-C(O)-O-; or

a 3-30-membered saturated or unsaturated ring system consisting of carbon atoms, optionally wherein one or more of said carbon atoms are replaced with a moiety independently selected from the group consisting of -O-, -S-, -N-, -N-C(O)-, -O-C(O)-, -N-C(O)-N- and -N-C(O)-O-; or an aromatic or heteroaromatic ring system;

wherein said R_2 may optionally be substituted one or more times with halogen, hydroxyl and oxo; and

wherein each optionally substituted R_1 is having a Mw of <500 g/mol; wherein at least one R_1 and/or R_2 has at least one carbinol functional group.

5. The elastomer according to claim 4, wherein

at least two of R_1 and/or R_2 are having at least one carbinol functional group; or

at least three of R_1 and/or R_2 are having at least one carbinol functional group; or

at least one of R_1 and/or R_2 are having at least two carbinol functional groups; or

at least one of R_1 and/or R_2 are having at least three carbinol functional groups; or

10

5

15

20

30

25

at least two of R₁ and/or R₂ are having at least two carbinol functional groups.

6. The elastomer according to claim 2, wherein said silanol functional polysiloxane is represented by formula (lb) below having a molecular weight in the range of 750 to 500,000 g/mol:

$$R_3$$
 R_3
 R_3

wherein m is an integer from 5 to 7500;

5

10

15

20

wherein each R₃ are independently selected from the group consisting of hydroxyl, methyl or linear or branched C₁-C₃₀ alkyl, linear or branched C₂-C₃₀ alkenyl, C₃-C₃₀ cycloalkyl or aryl;

wherein at least two R₃ groups are hydroxyl groups.

7. The elastomer according to any one of claims 1 and 3 wherein said one or more organic polysilazane comprises a compound having the structure of formula (II), wherein the compound has a linear or branched or cyclic structure:

wherein p is an integer from 5 to 100;

wherein each R₄ are independently selected from hydrogen, linear or branched C₁-C₆ alkyl or linear or branched C₁-C₆ alkenyl; and each R₅ are independently selected from hydrogen, linear or branched C₁-C₆ alkyl or linear or branched C₁-C₆ alkenyl; wherein at least one R₄ is not hydrogen.

25 8. The elastomer according to any one of the preceding claims, wherein the composition further comprising one or more catalysts.

- 9. The elastomer according to any one of the preceding claims, wherein said catalyst is an organic amine selected from the group consisting of trimethylamine, triethylamine, tributylamine, 1,5-diazabicyclo[4.3.0]non-5-ene (DBN), 1,8-diazabicyclo[5.4.0]undec-7-ene (DBU), 1,1,3,3-tetramethylguanidine, 4-dimethylaminopyridine (DMAP), an azole, imidazole, an imidazole derivative, benzotriazole, and a benzotriazole derivative.
- 10. The elastomer according to any one of the preceding claims, wherein said elastomer is free of metal.
- 11. The elastomer according to any one of claims 1 to 9, wherein said catalyst is one or more metal catalysts that is one or more organic metal catalysts.
- 12. The elastomer according to any one of the preceding claims, wherein said metal catalyst is selected from the group consisting of organic tin compounds, organic zinc compounds, organic titanium compounds, organic zirconium compounds, organic aluminum compounds, and organic nickel compounds.

13. A kit of parts comprising:

20

25

5

10

15

- a. a first composition comprising one or more carbinol functional polysiloxanes according to any one of the preceding claims; and
- b. a second composition comprising one or more organic polysilazane according to any one of the preceding claims;
- wherein said kit is for preparation of the elastomer according to any one of the preceding claims.
- 14. A method of preparing a self-supported elastomer, said method comprising mixing one or more carbinol functional polysiloxanes and one or more of organic polysilazanes, wherein said one or more organic polysilazanes constitutes 0.5-25 parts (weight) relative to 100 parts (weight) of said one or more hydroxyl functional polysiloxanes.
- 15. The method according to claim 14, wherein the elastomer is cured without heating the composition.

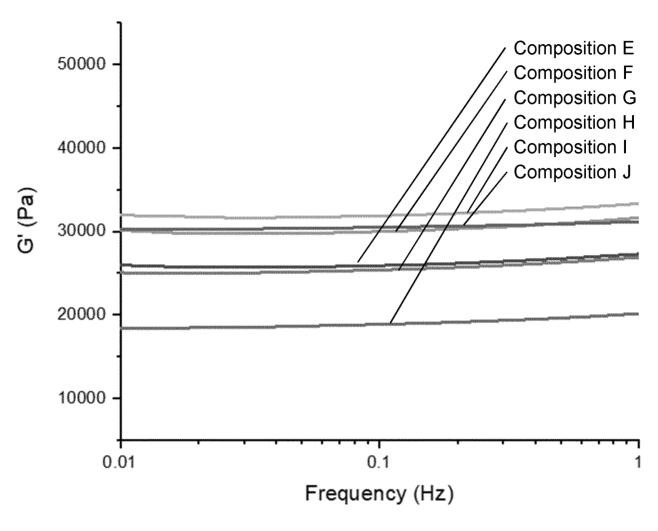


FIG. 1

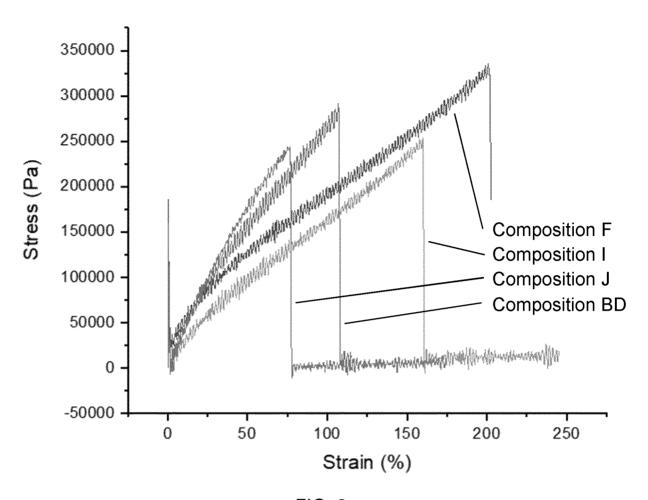


FIG. 2

INTERNATIONAL SEARCH REPORT

International application No
PCT/EP2023/051379

A. CLASSIFICATION OF SUBJECT MATTER INV. C08L83/04 C08L83/16 C08G77/62 C08G77/16 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) C08G C09D C08T 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 Relevant to claim No. Category* Citation of document, with indication, where appropriate, of the relevant passages EP 0 384 183 A2 (GEN ELECTRIC [US]) Х 1-15 29 August 1990 (1990-08-29) example 6 table 6 Х US 4 987 155 A (INOUE YOSHIO [JP] ET AL) 1-15 22 January 1991 (1991-01-22) example 1 See patent family annex. Further documents are listed in the continuation of Box C. Special categories of cited documents: "T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention "A" document defining the general state of the art which is not considered to be of particular relevance "E" earlier application or patent but published on or after the international "X" document of particular relevance;; the claimed invention cannot be considered novel or cannot be considered to involve an inventive filing date "L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other step when the document is taken alone document of particular relevance;; the claimed invention cannot be special reason (as specified) considered to involve an inventive step when the document is combined with one or more other such documents, such combination "O" document referring to an oral disclosure, use, exhibition or other means being obvious to a person skilled in the art document published prior to the international filing date but later than the priority date claimed "&" document member of the same patent family Date of the actual completion of the international search Date of mailing of the international search report 22 March 2023 30/03/2023 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, Queste, Sébastien Fax: (+31-70) 340-3016

INTERNATIONAL SEARCH REPORT

Information on patent family members

International application No
PCT/EP2023/051379

Patent document cited in search report		Publication date	Patent family member(s)		Publication date	
EP 0384183	A 2	29-08-1990	AT	59668	T	15-01-1991
			AT	125848	T	15-08-1995
			ΑU	9124582	A	08-09-1983
			BR	8208064	A	07-02-1984
			CA	1256227	A	20-06-1989
			DE	3280470	T2	04-04-1996
			DK	478783	A	17-10-1983
			EP	0104179	A1	04-04-1984
			EP	0384183	A2	29-08-1990
			ES	8503015	A1	01-02-1985
			FI	833580	A	03-10-1983
			IT	1175914	В	12-08-1987
			JP	H0413382	B2	09-03-1992
			JP	S59500219	A	16-02-1984
			KR	840003664	A	15-09-1984
			US	4417042	A	22-11-1983
			WO	8302948	A1	01-09-1983
			ZA	827750	В	28-12-1983
US 4987155	A	22-01-1991	JP	н068363	в2	02-02-1994
			JP	н0356542	A	12-03-1991
			US	4987155	A	22-01-1991