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(71) Applicant: **UNIVERSITÉ LAVAL** [CA/CA]; Pavillon
des Sciences de l'Éducation, 2320 rue des Bibliothèques, lo-
cal 1434, Québec, Québec G1V 0A6 (CA).

(72) Inventors: **SHARIFISHOURABI**, Gholamali; 307-825
rue Beaugard, Québec, Québec G1V 4L7 (CA). **RO-
DRIGUE**, Denis; 1310 Pierre-Beaudin, Québec, Québec
G2K 2H9 (CA).

(74) Agent: **BERESKIN & PARR LLP/S.E.N.C.R.L., S.R.L.**;
40 King Street West, 40th Floor, Toronto, Ontario M5H
3Y2 (CA).

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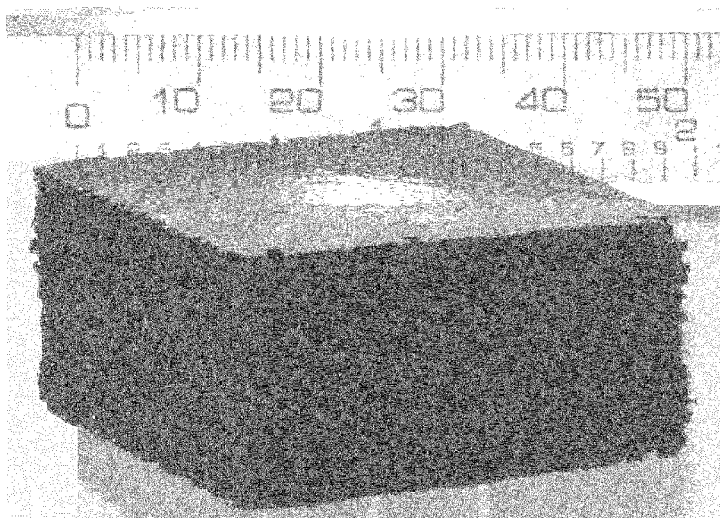


FIG. 1

(57) Abstract: The present disclosure relates to elastomeric composites and provides methods for producing elastomeric composites and more particularly to elastomeric open cell and/or closed cell foams. This may be achieved by dispersing insoluble particles, as foaming agent, into a reactive elastomer mixture. The insoluble particles have at least one of compressive yield point, decomposition temperature point, melting point and density lower than at least one of compressive yield point, decomposition temperature point, melting point and density of the elastomer. The insoluble particles remain in the composite, however they can be shrunk down or remained intact according to the application.



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ELASTOMERIC COMPOSITES AND METHODS FOR PRODUCING SAME

CROSS-REFERENCE TO RELATED APPLICATIONS

[0001] The present application claims priority to United States Application 62/776,145, filed December 6, 2018, the content of which is herein incorporated
5 by reference in its entirety.

FIELD

[0002] The present disclosure relates to elastomeric composites and in particular to methods of producing elastomeric open cell and/or closed cell foams.

BACKGROUND

10 [0003] Porous polymer composites, with and without particles, are increasingly being used in various fields because of their large surface area and unique mechanical, electrical and thermal properties. Among them, silicone foams, due to their desirable properties such as thermal resistance, low shrinkage and good electrical insulation, good flexibility at low temperatures and high resistance
15 against aging and weathering, have found a broad range of applications in different engineering fields, especially in mass transit vehicle industries such as aerospace, rail and marine industries.

[0004] Generally, silicone foams are divided into two main categories based on their processing method and the method used to introduce the porosity: blowing
20 agents or rigid hollow particles (syntactic foams). Blowing agents can be classified as physical or chemical agents based on their mechanisms. They release gases in the matrix prior to curing resulting in a cellular structure. Nitrogen (N₂) and carbon dioxide (CO₂) are good candidates as physical blowing agents due to their broad availability, as well as limited health and safety hazard, however, hydrogen
25 is also produced during a chemical reaction of additives prior to curing. Chemical blowing agents are still very common as the direct use of CO₂ and N₂ is very challenging (gas loss) because, under normal conditions, they are in the gas state

which makes them difficult to handle and they need optimized conditions like temperature and pressure.

[0005] Syntactic foams are a class of composite materials produced by filling a matrix with hollow particles like glass microspheres (cenospheres).

5 Recently, syntactic foams have attracted a great deal of attention due to their better morphology and thus better properties compared to other closed cell porous materials. But the previous syntactic silicone foams were not successfully marketed because the embedded common hollow particles negatively affected the viscoelastic properties of the final product. Common hollow particles, for example
10 glass microspheres, are crushable/brittle in nature and not well suited to produce viscoelastic silicone foams.

[0006] On the other hand, processing of composite silicone foams using a blowing agent is complex and becomes even more difficult, in the case of high filler content. The amount of blowing agent, pressure, temperature and viscosity of the
15 mixture during curing must be precisely controlled to avoid slow/rapid/over expansion of the foaming product causing the composite to have weak cell walls. Decrease in blowing agent limits the foam expansion, while an excessive amount of it causes the cell boundaries to coalesce resulting in the production of few very large cells leading to a poor foam structure (inhomogeneous) with low strength.
20 Recent developments of silicone foam formulations have been mostly based on the addition of different fillers to improve performance and properties such as hardness, thermal/electrical conductivity, and resistance against fire, aging and weathering. But the filler addition directly affects the rheological properties of the reactive mixture. Any change in composition and thus rheology will affect the
25 curing kinetics and properties of the final product, and especially the morphology of a foamed product. Fillers negatively affect the size and shape of the pores preventing a proper development of the voids resulting in non-spherical and disordered pores, and thus anisotropic structure and reduced properties.

[0007] The morphology of highly filled silicone foams, excluding syntactic silicone foams, is poor (non-spherical and disordered pores) resulting in anisotropy and inhomogeneity. On the other hand, the available syntactic silicone foams are not 100% viscoelastic, because of the nature of the used microspheres
5 (crushable/brittle hollow particles).

SUMMARY

[0008] In a first aspect, there is provided herein a syntactic elastomeric composite comprising an elastomer and a plurality of insoluble particles embedded
10 therein, wherein the insoluble particles have at least one of compressive yield point, decomposition temperature point, melting point and density lower than at least one of compressive yield point, decomposition temperature point, melting point and density of the elastomer.

[0009] In another aspect, there is provided herein an elastomeric composite
15 comprising an elastomer and a plurality of insoluble particles embedded therein, wherein the insoluble particles have at least one of compressive yield point, decomposition temperature point, melting point and density lower than at least one of compressive yield point, decomposition temperature point, melting point and density of the elastomer.

20 [0010] In another aspect described herein, there is provided a method of producing a syntactic elastomeric composite, comprising:

preparing a mixture comprising an elastomer; insoluble particles; and optionally a filler and/or an additive; the insoluble particles having at least one of compressive yield point, decomposition temperature point, melting
25 point and density lower than at least one of compressive yield point, decomposition temperature point, melting point and density of the elastomer;

optionally driving the mixture into a cavity;

curing the mixture to obtain the composite; and

optionally reducing the size of the insoluble particles.

[0011] In another aspect described herein, there is provided a method of producing an elastomeric composite, comprising:

5 preparing a mixture comprising an elastomer; insoluble particles; and optionally a filler and/or an additive; the insoluble particles having at least one of compressive yield point, decomposition temperature point, melting point and density lower than at least one of compressive yield point, decomposition temperature point, melting point and density of the
10 elastomer;

optionally driving the mixture into a cavity;

curing the mixture to obtain the composite; and

optionally reducing the size of the insoluble particles.

[0012] In a further aspect disclosed herein there is provided a method of
15 producing a layered syntactic elastomeric composite, comprising:

producing a plurality of layers of syntactic elastomeric composite according to the method described herein,

wherein the elastomer used in each of the layers is the same;

and wherein

20 each layer has a different relative density compared to the relative density of an adjacent layer,

each layer has a different average cell size compared to the average cell size of an adjacent layer, and/or

25 each layer comprises insoluble particles that are different, optionally having a different size, diameter, shape and/or composition, compared to the insoluble particles comprised in an adjacent layer.

[0013] In a further aspect disclosed herein there is provided a method of producing a layered elastomeric composite, comprising:

producing a plurality of layers of elastomeric composite according to the method described herein,

5 wherein the elastomer used in each of the layers is the same;

and wherein

each layer has a different relative density compared to the relative density of an adjacent layer,

10 each layer has a different average cell size compared to the average cell size of an adjacent layer, and/or

each layer comprises insoluble particles that are different, optionally having a different size, diameter, shape and/or composition, compared to the insoluble particles comprised in an adjacent layer.

[0014] In yet another aspect, there is provided herein a method of preparing
15 a syntactic cell-graded elastomeric composite, comprising:

preparing a first mixture comprising an elastomer and first insoluble particles;

20 preparing a second mixture comprising the elastomer and second insoluble particles, the second insoluble particles having a larger size and/or diameter compared to the first insoluble particles;

optionally preparing a third mixture comprising the elastomer and third insoluble particles, the third insoluble particles having a larger size and/or diameter compared to the second insoluble particles;

25 optionally preparing a fourth mixture comprising the elastomer and fourth insoluble particles, the fourth insoluble particles having a larger size and/or diameter compared to the third insoluble particles;

pouring the first mixture into a cavity;

- pouring the second mixture into the cavity;
optionally pouring the third mixture into the cavity;
optionally pouring the fourth mixture into the cavity;
optionally pressing down the combined mixtures;
- 5 curing the mixture to obtain the composite; and
reducing the size of the insoluble particles,
- wherein the insoluble particles have at least one of compressive yield point,
decomposition temperature point, melting point and density lower than at least one
of compressive yield point, decomposition temperature point, melting point and
10 density of the elastomer.
- [0015] In yet another aspect, there is provided herein a method of preparing
a cell-graded elastomeric composite, comprising:
- preparing a first mixture comprising an elastomer and first insoluble
particles;
- 15 preparing a second mixture comprising the elastomer and second insoluble
particles, the second insoluble particles having a larger size and/or diameter
compared to the first insoluble particles;
- optionally preparing a third mixture comprising the elastomer and third
insoluble particles, the third insoluble particles having a larger size and/or
20 diameter compared to the second insoluble particles;
- optionally preparing a fourth mixture comprising the elastomer and fourth
insoluble particles, the fourth insoluble particles having a larger size and/or
diameter compared to the third insoluble particles;
- pouring the first mixture into a cavity;
- 25 pouring the second mixture into the cavity;
optionally pouring the third mixture into the cavity;

optionally pouring the fourth mixture into the cavity;
optionally pressing down the combined mixtures;
curing the mixture to obtain the composite; and
reducing the size of the insoluble particles,

- 5 wherein the insoluble particles have at least one of compressive yield point, decomposition temperature point, melting point and density lower than at least one of compressive yield point, decomposition temperature point, melting point and density of the elastomer.

[0016] The elastomeric composite obtained using the methods herein
10 disclosed provides advantages over the conventional elastomer (e.g. silicone) foams, including desirable isotropic properties due to morphology, tunable properties, less chemical use (e.g. no solvent requirement), viscoelastic or viscoelastic-viscoplastic properties, no or little reaction with matrix; preserved properties (e.g. hardness and fire resistance) and energy absorption capacity.

15 **BRIEF DESCRIPTION OF THE DRAWINGS**

[0017] In the following drawings, which represent by way of example only, various embodiments of the disclosure:

[0018] FIG. 1 is a photograph of a viscoelastic-viscoplastic syntactic silicone foam having a relative density of 0.32;

20 [0019] FIG. 2 is a graph showing the compressive stress strain curves of the viscoelastic-viscoplastic syntactic silicone foam of FIG. 1;

[0020] FIG. 3 is a graph showing the compressive stress strain of a viscoelastic syntactic silicone foam having a relative density of 0.23; and

[0021] FIG. 4 is a photograph of a cell-size graded syntactic silicone foam
25 having a thickness of 20 mm and a relative density of 0.34.

DETAILED DESCRIPTION

[0022] Unless otherwise indicated, the definitions and examples described herein are intended to be applicable to all embodiments and aspects of the present disclosure herein described for which they are suitable as would be understood by
5 a person skilled in the art.

[0023] As used herein, “relative density” means the density of the elastomeric composite (e.g. elastomer in which insoluble particles are embedded) compared to the density of the elastomer alone.

[0024] As used herein, the term “porous” means a composite with a cellular
10 structure (e.g. a foam) comprising insoluble particles that have been either reduced in size, e.g. by applying pressure and/or heat, or that have not been reduced in size and thus are left substantially intact.

[0025] As used herein, the term “substantially” means that the specified term is modified to a degree of 10% or less, preferably 5% or less or more
15 preferably 1% or less, in a way that is recognized by a person skilled in the art as being reasonable and typical.

[0026] In understanding the scope of the present disclosure, the term “comprising” and its derivatives, as used herein, are intended to be open ended terms that specify the presence of the stated features, elements, components,
20 groups, integers, and/or steps, but do not exclude the presence of other unstated features, elements, components, groups, integers and/or steps. The foregoing also applies to words having similar meanings such as the terms, “including”, “having” and their derivatives.

[0027] As used herein, the term “about” means a reasonable amount of
25 deviation of the modified term such that the end result is not significantly changed. This term of degree should be construed as including a deviation of at least $\pm 5\%$ or at least $\pm 10\%$ of the modified term if this deviation would not negate the meaning of the word it modifies.

[0028] The term “% wt.” or “wt. %” as used herein when describing an ingredient present in a mixture or composite, refers to the weight % of this ingredient based on the total weight of the mixture or composite.

[0029] The term “insoluble particles” as used herein means particles that are insoluble in the elastomer matrix and behave as mechanical foaming agent. It will be understood that the insoluble particles may be of any shape e.g. spherical, cylindrical, beads, and can be regularly shaped or irregularly shaped. Various materials may be used for the insoluble particles so long as they have at least one of compressive yield point, decomposition point, melting point and/or density lower than that of the elastomer. For example, the insoluble particles are expanded polystyrene (EPS) beads.

[0030] The present disclosure provides a new method to produce viscoelastic elastomeric composites such as viscoelastic syntactic elastomeric composites including silicone sponges with relative density ranging from about 0.07 to about 0.8. It is a foamed-in-place method which enables foam filling of different cavities and production of isotropic silicone foam with different densities and cell sizes, ranging from less than 1 mm to over 1 cm in size. This is achieved by dispersing insoluble particles such as EPS beads, as the foaming agent, into a reactive mixture like a silicone resin. The EPS beads remain in the foam, however they can be shrunk down or remained intact depending on the application.

[0031] Generally, the method may comprise some or all of the following steps:

- a) Providing a one-part, two-part or more parts elastomer (e.g. silicone) resin system with desired properties (such as fire resistance, hardness, conductivity, etc.) according to the application.
- b) Providing insoluble particles (e.g. EPS beads) with desired properties (such as size density) according to the application.

- c) Mixing together the different parts of the elastomer system and optionally other additives (e.g. filler or solvent).
- d) Embedding the insoluble particles in the liquid mixture (resin(s) and additive(s)), within a container or the final mold, using a syringe like device or using a vacuum resin infusion system or any other suitable method.
- e) Driving the mixture into a cavity (e.g. casting, pouring, injecting or plunging the mixture).
- f) Curing the mixture.
- g) Post-curing the mixture.

[0032] One or more of the following steps can also be applied to control the density or any other properties (e.g. mechanical, thermal, chemical, electrical and optical properties) of the final porous product:

- h) Modification of the liquid mixture (e.g. using an additive like a solvent).
- i) Modification of the particle type.
- j) Addition of reinforcing/modifying fillers including hollow or compact particles.
- k) Controlling the reaction rate using additives and/or by controlling the temperature and/or pressure during curing.
- l) Applying positive or negative pressure during curing.

[0033] One or both of the following steps can also be applied following curing and post-curing steps to tune the properties of the final porous product:

- m) Treatment under heat.
- n) Treatment under pressure.

[0034] The method was used to produce high performance silicone foams with different relative densities.

[0035] A person skilled in the art will understand that the density of the elastomeric foam may be modified according to the amount of insoluble particles as well as fillers (e.g. reinforcing fibers) and additives (e.g. solvent) comprised in the elastomer mixture. The density is also dependent on the size/diameter of the insoluble particles.

[0036] For example, the insoluble particles have a compressive yield point lower than the compressive yield point of the elastomer.

10 [0037] For example, the insoluble particles have a decomposition temperature point lower than the decomposition temperature point of the elastomer.

[0038] For example, the insoluble particles have a melting point lower than the melting point of the elastomer.

15 [0039] For example, the insoluble particles have a density lower than the density of the elastomer.

[0040] For example, the insoluble particles are chosen from chemical particles, optionally polymers, and natural particles, optionally wood.

20 [0041] For example, the insoluble particles are chosen from expanded polystyrene (EPS), extruded polystyrene (Styrofoam), polypropylene, polyethylene and mixtures thereof.

[0042] For example, the insoluble particles are substantially spherical.

[0043] For example, the insoluble particles have an average diameter of about 0.05 mm to about 2 cm. For example, the insoluble particles have an average diameter of about 0.05 mm to about 0.5 mm. For example, the insoluble particles have an average diameter of about 0.75 mm to about 0.25 mm. For example, the insoluble particles have an average diameter of about 1 mm.

[0044] For example, the size of the insoluble particles is reduced and the composite is porous.

[0045] For example, the size of the insoluble particles is reduced by applying heat.

5 [0046] For example, the size of the insoluble particles is reduced by applying heat at a temperature of about 130°C to about 200°C.

[0047] For example, the heat is applied for about 30 minutes to about 60 minutes.

[0048] For example, the size of the insoluble particles is reduced by
10 applying pressure. The applying of pressure may for example be carried out in less than a second (e.g. by impact) or over several hours (e.g. by quasi-static compression).

[0049] For example, about 0.15 MPa to about 0.5 MPa of pressure is applied to the composite.

15 [0050] For example, the composite comprises a plurality of cells having an average size of about 0.05 mm to about 2 cm. For example, the composite comprises a plurality of cells having an average size of about 0.05 mm to about 0.5 mm. For example, the composite comprises a plurality of cells having an average size of about 0.75 mm to about 0.25 mm. For example, the composite
20 comprises a plurality of cells having an average size of about 1 mm.

[0051] For example, the insoluble particles are substantially intact.

[0052] For example, the composite has an open cell structure. For example, the composite comprises open cells.

[0053] For example, the elastomer is chosen from silicone, vinyl ester,
25 polyester, epoxy, polyurethane, natural rubber, melamine, urea, phenol, formaldehyde resins and mixtures thereof.

[0054] For example, the elastomer is silicone.

[0055] For example, the composite further comprises a filler chosen from silica, carbon, glass, metal, metal oxide, metal hydroxide and mixtures thereof.

[0056] For example, the composite comprises about 0.1 wt % to about 50 wt. % of the filler. For example, the composite comprises about 1 wt % to about 40 wt. % of the filler. For example, the composite comprises about 1 wt % to about 30 wt. % of the filler. For example, the composite comprises about 1 wt % to about 20 wt. % of the filler. For example, the composite comprises about 1 wt % to about 10 wt. % of the filler.

[0057] For example, the composite further comprises an additive chosen from thixotropes, fire retardants, suppressants, inhibitors, stabilizers, solvents, blowing agents, catalysts, pigments, coloring agents, conductive additives and mixtures thereof.

[0058] For example, the composite comprises about 0.1 wt % to about 50 wt. % of the additive. For example, the composite comprises about 1 wt % to about 40 wt. % of the additive. For example, the composite comprises about 1 wt % to about 30 wt. % of the additive. For example, the composite comprises about 1 wt % to about 20 wt. % of the additive. For example, the composite comprises about 1 wt % to about 10 wt. % of the additive.

[0059] For example, the fire retardant is chosen from carbon-based particles, metal oxide, metal hydroxide, red phosphorus, boron compounds and mixtures thereof. For example, the metal hydroxide is aluminum hydroxide.

[0060] For example, the additive is a solvent, optionally chosen from ethanol, acetone, toluene and mixtures thereof. For example, the solvent is ethanol.

[0061] For example, the composite has a relative density of about 0.07 to about 0.8. For example, the composite has a relative density of about 0.1 to about 0.8. For example, the composite has a relative density of about 0.1 to about 0.6. For example, the composite has a relative density of about 0.1 to about 0.5. For

example, the composite has a relative density of about 0.2 to about 0.5. For example, the composite has a relative density of about 0.2 to about 0.4. For example, the composite has a relative density of about 0.2 to about 0.35.

[0062] For example, the composite is isotropic.

- 5 [0063] For example, the composite is viscoelastic. For example, the composite is viscoelastic-viscoplastic.

[0064] For example, the composite comprises at least two integrally connected layers.

- 10 [0065] For example, the at least two integrally connected layers are substantially parallel relative to one another.

[0066] For example, the at least two integrally connected layers comprise the same elastomer.

[0067] For example, each layer has a different relative density compared to the relative density of an adjacent layer.

- 15 [0068] For example, each layer has a different average cell size compared to the average cell size of an adjacent layer.

[0069] For example, each layer comprises insoluble particles that have a different size, diameter, shape and/or composition, compared to the size, diameter, shape and/or composition of the insoluble particles comprised in an adjacent layer.

- 20 [0070] For example, the composite comprises three integrally connected layers.

[0071] For example, the two outer layers have a relative density greater than the relative density of the middle layer.

[0072] For example, the two outer layers are integral skins.

- 25 [0073] For example, the composite comprises a plurality of macropores and a plurality of micropores. For example, the macropores have an average cell size

of about 50 microns to about 990 microns and/or the micropores have an average cell size of about 1 mm to about 20 mm.

[0074] For example, the mixture is driven prior to the curing.

[0075] For example, the driving mixture comprises casting, pouring,
5 injecting and/or plunging the mixture into the cavity.

[0076] For example, the mixture is cured at room temperature. For example, the mixture is cured at a temperature of about 0°C to about 110°C. For example, the mixture is cured at a temperature of about 20°C to about 100°C. For example, the mixture is cured at a temperature of about 40°C to about 90°C. For example,
10 the mixture is cured at a temperature of about 50°C to about 90°C. For example, the mixture is cured at a temperature of about 60°C to about 80°C.

[0077] For example, the method further comprises post-curing the composite. It will be understood that post-curing is carried out at a suitable temperature above room temperature. For example, the post-curing may be
15 carried out at a temperature nearing that of the decomposition point of the silicone matrix which, as the person skilled will readily understand, differs depending on the silicone resin used.

[0078] For example, the size of the insoluble particles is reduced by applying heat to the composite. For example, the heat is applied at a temperature
20 of about 130°C to about 200°C. For example, the heat is applied for about 30 minutes to about 60 minutes.

[0079] For example, the size of the insoluble particles is reduced by applying pressure to the composite. For example, about 0.15 MPa to about 0.5 MPa of pressure is applied to the composite. For example, the pressure is applied
25 to the composite using a calender. For example, the pressure is applied to the composite using an InstronTM universal testing machine.

[0080] For example, the mixture is prepared using a syringe like device, a vacuum infusion system, a plunger, an injector or an extruder.

[0081] For example, the insoluble particles are chosen from chemical particles, optionally polymers, and natural particles, optionally wood.

[0082] For example, the insoluble particles are chosen from expanded polystyrene (EPS), extruded polystyrene (Styrofoam), polypropylene, polyethylene
5 and mixtures thereof.

[0083] For example, the insoluble particles are coated. For example, the insoluble particles are coated with an electrically conductive material. For example, all or substantially all of the insoluble particles are coated with an electrically conductive material.

10 [0084] For example, the electrically conductive material comprises or is carbon based and/or a metal based fibers. For example the carbon based fibers are nickel coated carbon fibers.

[0085] For example, the composite comprises about 0.1 wt % to about 50 wt. % of the electrically conductive material. For example, the composite
15 comprises about 1 wt % to about 40 wt. % of the electrically conductive material. For example, the composite comprises about 1 wt % to about 30 wt. % of the electrically conductive material. For example, the composite comprises about 1 wt % to about 20 wt. % of the electrically conductive material. For example, the composite comprises about 1 wt % to about 10 wt. % of the electrically conductive
20 material.

[0086] For example, the composite has a volume resistivity of about 0.1 Ohms to about 1 Megaohm. For example, the composite has a volume resistivity of about 0.1 Ohms to about 5 Ohms. For example, the composite has a volume resistivity of about 0.1 Ohms to about 3 Ohms.

25 [0087] For example, the insoluble particles are substantially spherical.

[0088] For example, the insoluble particles have an average diameter of about 0.05 mm to about 2 cm. For example, the insoluble particles have an average diameter of about 0.05 mm to about 0.5 mm. For example, the insoluble

particles have an average diameter of about 0.75 mm to about 0.25 mm. For example, the insoluble particles have an average diameter of about 1 mm.

[0089] For example, the composite is porous.

[0090] For example, the composite comprises a plurality of cells having an average size of about 0.05 mm to about 2 cm. For example, the composite comprises a plurality of cells having an average size of about 0.05 mm to about 0.5 mm. For example, the composite comprises a plurality of cells having an average size of about 0.75 mm to about 0.25 mm. For example, the composite comprises a plurality of cells having an average size of about 1 mm.

10 [0091] For example, the insoluble particles are substantially intact.

[0092] For example, the elastomer is chosen from silicone, vinyl ester, polyester, epoxy, polyurethane, natural rubber, melamine, urea, phenol formaldehyde resins and mixtures thereof.

[0093] For example, the elastomer is silicone.

15 [0094] For example, the method further comprises adding to the mixture a filler chosen from silica, carbon, glass, metal, metal oxide, metal hydroxide and mixtures thereof.

[0095] For example, the method further comprises adding to the mixture an additive chosen from thixotropes, fire retardants, suppressants, inhibitors, 20 stabilizers, solvents, blowing agents, catalysts, pigments, coloring agents, conductive additives and mixtures thereof.

[0096] For example, the fire retardant is chosen from carbon-based particles, metal oxide, metal hydroxide, red phosphorus, boron compounds and mixtures thereof. For example, the metal hydroxide is aluminum hydroxide.

25 [0097] For example, the additive is a solvent, optionally chosen from ethanol, acetone, toluene and mixtures thereof. For example, the solvent is ethanol.

[00098] For example, the method further comprises adding one or more of a blowing agent, surfactant or additional insoluble particles, optionally hollow or compact insoluble particles, to the mixture.

[00099] For example, the composite has a relative density of about 0.1 to about 0.8. For example, the composite has a relative density of about 0.1 to about 0.5. For example, the composite has a relative density of about 0.2 to about 0.5. For example, the composite has a relative density of about 0.2 to about 0.4. For example, the composite has a relative density of about 0.2 to about 0.35.

[00100] For example, the composite is isotropic.

10 [00101] For example, the composite is viscoelastic.

[00102] For example, the composite is viscoelastic-viscoplastic.

[00103] For example, the method comprises:

preparing a first mixture and curing the first mixture to obtain a first layer;
preparing a second mixture, pouring the second mixture adjacent to the first
15 layer and curing the second mixture to obtain a second layer; and
optionally preparing a third mixture, pouring the third mixture adjacent to the first or second layer, and curing the third mixture to obtain a third layer.

[00104] The examples detailed below are non-limitative and are used to better exemplify the methods of the present disclosure.

20 **EXAMPLES**

Example 1 – Viscoelastic-viscoplastic syntactic silicone foam with relative density of 0.32

[00105] Using the presently disclosed method, a syntactic silicone foam displaying viscoelastic-viscoplastic behaviour was produced. The foam has a pore
25 size of about 1 mm and a relative density of 0.32 (as shown in FIG. 1) and can withstand very high compressive stresses (of at least 0.4 MPa) at least 25% strain (as shown in FIG. 2). The foam was achieved by controlled curing of a silicone

system with embedded expanded polystyrene (EPS) beads, through steps a)-g) and l) described as follows. A two parts room temperature vulcanizing silicone system with high degree of hardness and mechanical properties (M4370) was provided (step a). EPS beads having a diameter of about 1 mm were provided (step b). Parts A and B of the silicone system were combined together at a mixing ratio of 9:1 using a dual hand mixer (step c). Subsequently, about 3 wt. % EPS beads were manually added to about 97 wt. % of the silicone resin using a stick (step d). Then, a sufficient amount of the mixture was poured into a container until the container became completely full (step e). The lid of container was forcibly closed to push back down the buoyant EPS beads (step l) and left at room temperature to cure (step f). The product was then post-cured to complete the cross-linkage process (step g). The resulting foam had a relative density of 0.32.

[00106] As shown in FIG. 2, under cyclic compression, the silicone material shows a combination of viscoelastic-viscoplastic behavior, similar to shape memory foams, on the Cycle 1. This behavior is attributed to the properties of the EPS beads. On the next cycles (Cycle 2, Cycle 3 and Cycle 8), as the beads are shrunk down, the behavior of the material becomes more viscoelastic, i.e. the behavior of conventional silicone foams. As can be seen, Cycles 2, 3 and 8 are similar to one another. Loading (upper) portions of Cycles 1, 2, 3 and 8 are labelled.

20 **Example 2 – Viscoelastic syntactic silicone foam with relative density of 0.32**

[00107] In terms of compression behavior, a completely different material was achieved through heat treatment of the foam sample described in Example 1. A part of the same sample of Example 1 was cut and placed in an oven for 1 hour at 130°C (step m) to tune its compression properties to be viscoelastic only. The intent was to heat up the EPS beads to make them shrink down so as to transform the viscoelastic-viscoplastic material of Example 1 into a viscoelastic silicone foam. As the EPS beads were still left in the foam, the foam density remained the same i.e. the foam had a relative density of 0.32. The compression properties of the produced foam were verified to be viscoelastic, as shown in FIG. 3.

[00108] As shown in FIG. 3, in the absence of the initial EPS beads, the produced foam displays viscoelastic behavior just like conventional silicone foams during all cycles (subsequent cycles not shown but similar to the cycle shown in FIG. 3). Therefore, depending on the application, the compression behavior of the
5 produced foam may be tuned to be according to FIG. 3 or any of cycles of FIG. 2. For example, a foam which behaves like cycle 1 of FIG. 2 is stiff and has better energy absorption capacity and flexural behavior, which makes it suitable for applications where strength, deflection and energy absorption capability of material is critical, like for example in packaging applications. However, a foam
10 with behavior similar to FIG. 3 is flexible and suitable for example for vibration dampening applications.

Example 3 – Viscoelastic syntactic silicone foam with relative density of 0.27

[00109] Using the presently disclosed method, a viscoelastic syntactic
15 silicone foam was produced with pore size of about 1 mm and relative density of 0.27 through steps a)-g), h), and I) described as follows. A two parts room temperature vulcanizing silicone system with high degree of hardness and mechanical properties (M4370) was provided (step a). EPS beads with a diameter of about 1 mm were provided (step b). Parts A and B of the silicone system were
20 combined together at a mixing ratio of 9:1 using a dual hand mixer (step c). Subsequently, the mixture comprising about 4 wt. % EPS and about 96 wt. % silicone resin was diluted with 10 wt. % ethanol (step h) to obtain a silicone with lower density compared to the silicone foam described in Example 1. The EPS beads were then manually added to the mixture using a stick (step d). Then, a
25 sufficient amount of the mixture was poured into a mold until the mold became completely full (step e). The lid of the mold was forcibly closed to push down back the buoyant EPS beads (step I) and left at room temperature to cure (step f). The product was then post-cured in an oven at 150°C for about 2 hours to complete the cross-linkage process (step g). The resulting foam had a relative density of
30 0.27.

Example 4 – Graded syntactic silicone foam with relative density of 0.34

[00110] Using the presently disclosed method, a cell-size graded syntactic
5 silicone foam was produced with thickness of 20 mm and relative density of 0.34.
In the foam, the size of cells varies from about 0.6 mm to about 1.9 mm through
the thickness as shown in FIG. 4. The foam was achieved by controlled curing of
a silicone system with embedded EPS beads, through steps a)-g), l) and k)
described as follows. A two parts room temperature vulcanizing silicone system
10 with high degree of hardness and mechanical properties (M4370) was provided
(step a). Four groups of EPS beads with same weights, but different diameter sizes
(as shown in Table 1) were provided (step b). Parts A and B of the silicone system
were combined together at a mixing ratio of 9:1 using a dual hand mixer (step c).
Subsequently, each group of EPS beads was manually added to a quarter of the
15 silicone system mixture using a stick to prepare four different mixtures with same
weight ratio of bead to silicone (step d). Each of the four different mixtures
comprised about 4 wt. % EPS and about 96 wt. % silicone resin. Then, an equal
weight of each bead/silicone mixtures was poured into a mold and leveled, one
after another, in the order of bead size and starting from the mixture having smaller
20 beads (step e). The four-layer material was then covered with a rigid lid and
subjected to about 150 kPa compression to push back down the buoyant EPS
beads (step l). The mold was subsequently left in the oven at 70°C (step k) for
three hours to cure the materials (step f), and to form a single-layer foam with four
different cell distributions. The product was then post-cured in an oven at 150°C
25 for about 2 hours to complete the cross-linkage process (step g).

Table 1. The diameter sizes of the four groups of EPS beads

Group No.	Mean Bead diameter (mm)	Beads diameter range (mm)
1	0.8	0.6-1.0
2	1.1	0.9-1.3
3	1.4	1.2-1.6
4	1.7	1.5-1.9

Example 5 – One-part silicone foam

- 5 [00111] Using the presently disclosed method, different viscoelastic silicone foams with a pore size of about 1 mm and densities above 100 kg/m³ (corresponding to a relative density of greater than about 0.09) were made using one-part silicone polymer (e.g. acetoxysilicone produced by DAP Products Inc.) through the following steps: Silicone (about 85 wt. %) and EPS beads with a diameter of about 1 mm (about 15 wt. %) were weighed and mixed together. The mixture was subsequently molded in a cylindrical mold, the lid of the mold was forcibly closed to push down back the buoyant EPS beads and the mixture was left at room temperature for 24 hours to cure. The material was then demolded and put in oven at 140°C for 30 minutes to shrink the EPS beads. Silicone foams with relative densities ranging from less than 0.1 to above 0.5 can be produced using this method.

Example 6 – Open cell silicone foam

- [00112] Using the presently disclosed method, different open cell silicone foams with a cell size of about 0.9 mm and densities above 100 kg/m³ (corresponding to a relative density greater than about 0.09) were made using a two-part liquid silicone polymer (LSR). The open cell foams were achieved using hot compression molding. Silicone (about 85 wt. %) and EPS beads with a diameter of about 1 mm (about 15 wt. %) were weighed and mixed together. The

mixture was subsequently molded in a cylindrical mold and cured under 2 psi compression at 70°C for 24 hours. The cured material was then demolded and put in oven at 180°C for 30 minutes to shrink the EPS beads. Depending on the beads content and the compression load, different combinations of closed and open cells
5 can be achieved. Silicone foams with relative densities ranging from less than 0.1 to above 0.5 can be produced using this method.

Example 7. Electrically and thermally conductive silicone foam

[00113] Using the presently disclosed method, an electrically conductive silicone foam was produced using nickel coated carbon fibers and a thermally
10 conductive silicone resin. The foam has a pore size of about 0.9 mm and density of 250 kg/m³ (corresponding to a relative density of about 0.17). The volume resistivity of the matrix material of the foam was measured to be less than 3 Ohms.

[00114] The fabrication process includes the following steps: Firstly, a two-part room temperature vulcanizing silicone system with a high degree of thermal
15 conductivity (M4370) was selected. Parts A and B of the silicone system were then mixed together at a mixing ratio of 9:1 using a dual hand mixer. Most of the EPS beads (with a diameter of around 1 mm) were impregnated with the active silicone resin and then coated with the nickel coated carbon fibers, through manually mixing with a spatula in a container. The mixture comprised about 15 wt.
20 % nickel coated fibers; about 5 wt. % EPS and about 80 wt. % silicone resin. Subsequently, the EPS beads were put in a compression mold. Then the mold was subjected to a compression load of about 2 psi for six hours at room temperature to cure the material. The product was then post-cured in an oven at 150°C for about 2 hours to complete the polymerization process, and to shrink the EPS
25 beads. Depending on the desired level of electrical conductivity, different electrically conductive fillers (e.g. carbon based and metal-based particles) with different concentrations can be used.

Example 8. Auxetic silicone foam

[00115] Using the presently disclosed method, an auxetic silicone foam was produced using a two-part platinum cure silicone resin system (M4370) and EPS beads. The foam has a pore size of about 0.9 mm and a density of 140 kg/m³ (corresponding to a relative density of about 0.09). The foam shows negative Poisson's ratio under compression. The auxetic foams was achieved using compression molding through the following steps. Silicone (about 90 wt. %) and EPS beads with a diameter of about 1 mm (about 10 wt. %) were weighed and mixed together. The mixture was subsequently molded in a cylindrical mold and put under about 2 psi compression and cured at room temperature for 6 hours. The cured material was then demolded and put in an oven at 140°C for 30 minutes to shrink the EPS beads. Auxetic silicone foams with relative densities ranging from about 0.08 to about 0.6 can be produced using this method.

Example 9. Fire resistant silicone foam

15 [00116] Using the presently disclosed method, a fire resistant silicone foam was produced using a high temperature resistant silicone resin system (M4370) and aluminum hydroxide microparticles through the following steps: Firstly, parts A and B of the silicone system were mixed together at a mixing ratio of 9:1 using a dual hand mixer. Then the aluminum hydroxide particles were added using the same mixer. Subsequently, the silicone/aluminum hydroxide mixture was poured in a container full of EPS beads. The mixture comprised about 15 wt. % aluminum hydroxide; about 5 wt. % EPS and about 80 wt. % silicone resin. The lid of the container was then closed to push back down the buoyant EPS beads, and the material was left at room temperature to cure. The product was then post-cured in an oven at 150°C for about 2 hours to complete the polymerization process, and to shrink the EPS beads. The foam has a pore size of about 1 mm and a density of 350 kg/m³ (corresponding to a relative density of about 0.23). To test its fire resistant properties, the resulting silicone foam was lit for about 30 seconds and found to be non-flammable and fire resistant. The foam was also

tested for its degree of temperature resistance. It was put in an oven at a temperature of about 300°C for 10 minutes, and then the foam was tested for compression properties after cooling down to room temperature. It was found that the foam can withstand high temperatures up to 300°C without significant loss in compression properties.

[00117] Depending on the desired level of temperature and fire resistance, different fire resistance/flame retardant particles (e.g. carbon based particles, metal oxides, metal hydroxide, red phosphorus, and boron compounds) with different concentrations can be used.

10 [00118] While a description was made with particular reference to the specific embodiments, it will be understood that numerous modifications thereto will appear to those skilled in the art.

[00119] The scope of the claims should not be limited by specific embodiments and examples provided in the present disclosure and accompanying drawings, but should be given the broadest interpretation consistent with the disclosure as a whole.

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CLAIMS:

1. A syntactic elastomeric composite comprising an elastomer and a plurality of insoluble particles embedded therein, wherein the insoluble particles have at least one of compressive yield point, decomposition temperature point, melting point and density lower than at least one of compressive yield point, decomposition temperature point, melting point and density of the elastomer.
2. An elastomeric composite comprising an elastomer and a plurality of insoluble particles embedded therein, wherein the insoluble particles have at least one of compressive yield point, decomposition temperature point, melting point and density lower than at least one of compressive yield point, decomposition temperature point, melting point and density of the elastomer.
3. The composite of claim 1 or 2, wherein the mixture comprises about 50 wt. % to about 99 wt. % of the elastomer.
4. The composite of claim 1 or 2, wherein the mixture comprises about 60 wt. % to about 99 wt. % of the elastomer.
5. The composite of claim 1 or 2, wherein the mixture comprises about 70 wt. % to about 99 wt. % of the elastomer.
6. The composite of claim 1 or 2, wherein the mixture comprises about 80 wt. % to about 99 wt. % of the elastomer.
7. The composite of claim 1 or 2, wherein the mixture comprises about 90 wt. % to about 99 wt. % of the elastomer.
8. The composite of any one of claims 1 to 7, wherein the mixture comprises about 1 wt. % to about 40 wt. % of the insoluble particles.
9. The composite of any one of claims 1 to 7, wherein the mixture comprises about 1 wt. % to about 30 wt. % of the insoluble particles.

10. The composite of any one of claims 1 to 7, wherein the mixture comprises about 1 wt. % to about 20 wt. % of the insoluble particles.
11. The composite of any one of claims 1 to 7, wherein the mixture comprises about 1 wt. % to about 15 wt. % of the insoluble particles.
12. The composite of any one of claims 1 to 7, wherein the mixture comprises about 1 wt. % to about 10 wt. % of the insoluble particles.
13. The composite of any one of claims 1 to 7, wherein the mixture comprises about 1 wt. % to about 5 wt. % of the insoluble particles.
14. The composite of any one of claims 1 to 13, wherein the insoluble particles are chosen from chemical particles, optionally polymers, and natural particles, optionally wood.
15. The composite of any one of claims 1 to 14, wherein the insoluble particles are chosen from expanded polystyrene (EPS), extruded polystyrene (Styrofoam), polypropylene, polyethylene and mixtures thereof.
16. The composite of any one of claims 1 to 15, wherein the insoluble particles are substantially spherical.
17. The composite of any one of claims 1 to 16, wherein the insoluble particles have an average diameter of about 0.05 mm to about 2 cm.
18. The composite of any one of claims 1 to 16, wherein the insoluble particles have an average diameter of about 0.05 mm to about 0.5 mm.
19. The composite of any one of claims 1 to 18, wherein the size of the insoluble particles is reduced.
20. The composite of any one of claim 1 to 19, wherein the size of the insoluble particles is reduced by applying heat.

21. The composite of any one of claims 1 to 20, wherein the size of the insoluble particles is reduced by applying heat at a temperature of about 130°C to about 200°C.
22. The composite of claim 20 or 21, wherein the heat is applied for about 30 minutes to about 60 minutes.
23. The composite of any one of claims 1 to 22, wherein the size of the insoluble particles is reduced by applying pressure.
24. The composite of claim 23, wherein about 0.15 MPa to about 0.5 MPa of pressure is applied to the composite.
25. The composite of any one of claims 1 to 24, wherein the composite comprises a plurality of cells having an average size of about 0.05 mm to about 2 cm.
26. The composite of any one of claims 1 to 24, wherein the composite comprises a plurality of cells having an average size of about 0.05 mm to about 0.5 mm.
27. The composite of any one of claims 1 to 18, wherein the insoluble particles are substantially intact.
28. The composite of any one of claims 1 to 27, wherein the composite comprises open cells.
29. The composite of any one of claims 1 to 28, wherein the elastomer is chosen from silicone, vinyl ester, polyester, epoxy, polyurethane, natural rubber, melamine, urea, phenol, formaldehyde resin, and mixtures thereof.
30. The composite of any one of claims 1 to 28, wherein the elastomer is silicone, optionally a one-part or a two-part silicone.

31. The composite of claim 30, wherein the two-part silicone is a two-part platinum silicone.

32. The composite of any one of claims 1 to 31, further comprising about 0.1 wt % to about 50 wt. % of a filler chosen from silica, carbon, glass, metal, metal oxide, metal hydroxide, and mixtures thereof.

33. The composite of any one of claims 1 to 32, further comprising about 0.1 wt % to about 50 wt. % of an additive chosen from thixotropes, fire retardants, suppressants, inhibitors, stabilizers, solvents, blowing agents, catalysts, pigments, coloring agents, conductive additives, and mixtures thereof.

34. The composite of claim 33, wherein the additive is a solvent, optionally chosen from ethanol, acetone, toluene and mixtures thereof.

35. The composite of claim 34, wherein the solvent is ethanol.

36. The composite of any one of claims 1 to 35, wherein the insoluble particles are coated.

37. The composite of any one of claims 1 to 35, wherein the insoluble particles are coated with an electrically conductive material, optionally chosen from carbon based fibers, metal based fibers and mixtures thereof.

38. The composite of claim 37, wherein the composite comprises about 0.1 wt. % to about 50 wt. % of the electrically conductive material.

39. The composite of claim 37 or 38, wherein the electrically conductive material is nickel coated carbon fibers.

40. The composite of any one of claims 1 to 39, having a volume resistivity of about 0.1 Ohms to about 1 Megaohm.

41. The composite of any one of claims 1 to 39, having a volume resistivity of about 0.1 Ohms to about 3 Ohms.
42. The composite of any one of claims 1 to 41, having a density of about 70 kg/m³ to about 700 kg/m³.
43. The composite of any one of claims 1 to 41, having a density of about 100 kg/m³ to about 500 kg/m³.
44. The composite of any one of claims 1 to 41, having a density of about 100 kg/m³ to about 400 kg/m³.
45. The composite of any one of claims 1 to 44, having a relative density of about 0.07 to about 0.8.
46. The composite of any one of claims 1 to 44, having a relative density of about 0.1 to about 0.8.
47. The composite of any one of claims 1 to 44, having a relative density of about 0.1 to about 0.5.
48. The composite of any one of claims 1 to 44, having a relative density of about 0.2 to about 0.5.
49. The composite of any one of claims 1 to 44, having a relative density of about 0.2 to about 0.4.
50. The composite of any one of claims 1 to 44, having a relative density of about 0.2 to about 0.35.
51. The composite of any one of claims 1 to 50, wherein the composite is isotropic.
52. The composite of any one of claims 1 to 51, wherein the composite is viscoelastic.

53. The composite of any one of claims 1 to 52, wherein the composite is viscoelastic-viscoplastic.
54. The composite of any one of claims 1 to 53, wherein the composite comprises at least two integrally connected layers.
55. The composite of claim 54, wherein the at least two integrally connected layers are substantially parallel relative to one another.
56. The composite of claim 54 or 55, wherein the at least two integrally connected layers comprise the same elastomer.
57. The composite of any one of claims 54 to 56, wherein each layer has a different relative density compared to the relative density of an adjacent layer.
58. The composite of any one of claims 54 to 56, wherein each layer has a different average cell size compared to the average cell size of an adjacent layer.
59. The composite of any one of claims 54 to 56, wherein each layer comprises insoluble particles that have a different size, diameter, shape and/or composition, compared to the size, diameter, shape and/or composition of the insoluble particles comprised in an adjacent layer.
60. The composite of any one of claims 1 to 54, wherein the composite comprises three integrally connected layers.
61. The composite of claim 60, wherein the two outer layers have a relative density greater than the relative density of the middle layer.
62. The composite of claim 60, wherein the two outer layers are integral skins.
63. The composite of any one of claims 1 to 62, comprising a plurality of micropores and a plurality of macropores, optionally wherein the micropores have

and average cell size of about 50 microns to about 990 microns and/or the macropores have an average cell size of about 1 mm to about 20 mm.

64. A method of producing a syntactic elastomeric composite, comprising:

preparing a mixture comprising an elastomer, insoluble particles, and optionally a filler and/or an additive, the insoluble particles having at least one of compressive yield point, decomposition temperature point, melting point and density lower than at least one of compressive yield point, decomposition temperature point, melting point and density of the elastomer;

optionally driving the mixture into a cavity;

curing the mixture to obtain the composite; and

optionally reducing the size of the insoluble particles.

65. A method of producing an elastomeric composite, comprising:

preparing a mixture comprising an elastomer, insoluble particles, and optionally a filler and/or an additive, the insoluble particles having at least one of compressive yield point, decomposition temperature point, melting point and density lower than at least one of compressive yield point, decomposition temperature point, melting point and density of the elastomer;

optionally driving the mixture into a cavity;

curing the mixture to obtain the composite; and

optionally reducing the size of the insoluble particles.

66. The method of claim 64 or 65, wherein the mixture is driven prior to the curing.

67. The method of claim 64 or 65, wherein the driven mixture comprises casting, pouring, injecting and/or plunging the mixture into the cavity.
68. The method of any one of claims 64 to 66, wherein the mixture is cured at room temperature.
69. The method of any one of claims 64 to 66, wherein the mixture is cured at a temperature of about 0°C to about 110°C.
70. The method of any one of claims 64 to 66, wherein the mixture is cured at a temperature of about 20°C to about 100°C.
71. The method of any one of claims 64 to 70, further comprising post-curing the composite.
72. The method of any one of claims 64 to 71, wherein the size of the insoluble particles is reduced by applying heat to the composite.
73. The method of claim 72, wherein the heat is applied at a temperature of about 130°C to about 200°C.
74. The method of claim 72 or 73, wherein the heat is applied for about 30 minutes to about 60 minutes.
75. The method of any one of claims 64 to 74, wherein the size of the insoluble particles is reduced by applying pressure to the composite.
76. The method of claim 75, wherein about 0.15 MPa to about 0.5 MPa of pressure is applied to the composite.
77. The method of claim 75 or 76, wherein the pressure is applied to the composite using a calender.
78. The method of claim 75 or 76, wherein the pressure is applied to the composite using an InstronTM universal testing machine.

79. The method of any one of claims 64 to 78, wherein the mixture is prepared using a syringe like device, a vacuum infusion system, a plunger, an injector or an extruder.
80. The method of any one of claims 64 to 79, wherein the insoluble particles are chosen from chemical particles, optionally polymers, and natural particles, optionally wood.
81. The method of any one of claims 64 to 71, wherein the insoluble particles are chosen from expanded polystyrene (EPS), extruded polystyrene (Styrofoam), polypropylene, polyethylene and mixtures thereof.
82. The method of any one of claims 64 to 81, wherein the insoluble particles are substantially spherical.
83. The method of any one of claims 64 to 82, wherein the insoluble particles have an average diameter of about 0.05 mm to about 2 cm.
84. The method of any one of claims 64 to 82, wherein the insoluble particles have an average diameter of about 0.05 mm to about 0.5 mm.
85. The method of any one of claims 64 to 84, wherein the composite is porous.
86. The method of any one of claims 64 to 85, wherein the composite comprises a plurality of cells having an average size of about 0.05 mm to about 2 cm.
87. The method of any one of claims 64 to 86, wherein the composite comprises a plurality of cell having an average size of about 0.05 mm to about 0.5 mm.
88. The method of any one of claims 64 to 87, wherein the insoluble particles are substantially intact.

89. The method of any one of claims 64 to 88, wherein the elastomer is chosen from silicone, vinyl ester, polyester, epoxy, polyurethane, natural rubber, melamine, urea, phenol, formaldehyde resins and mixtures thereof.
90. The method of any one of claims 64 to 88, wherein the elastomer is silicone, optionally a one-part or a two-part silicone.
91. The method of claim 90, wherein the two-part silicone is a two-part platinum silicone.
92. The method of any one of claims 64 to 91, further comprising adding to the mixture a filler chosen from silica, carbon, glass, metal, metal oxide, metal hydroxide and mixtures thereof.
93. The method of any one of claims 64 to 92, further comprising adding to the mixture an additive chosen from thixotropes, fire retardants, suppressants, inhibitors, stabilizers, solvents, blowing agents, catalysts, pigments, coloring agents, conductive additives and mixtures thereof.
94. The method of claim 93, wherein the additive is a solvent, optionally chosen from ethanol, acetone, toluene and mixtures thereof.
95. The method of claim 94, wherein the solvent is ethanol.
96. The method of any one of claims 64 to 95, further comprising adding one or more of a blowing agent, surfactant or additional insoluble particles, optionally hollow or compact insoluble particles, to the mixture.
97. The method of any one of claims 64 to 96, wherein prior to preparing the mixture, the method comprises coating the insoluble particles with an electrically conductive material, optionally chosen from carbon based fibers, metal based fibers and mixtures thereof.

98. The method of claim 97, wherein the electrically conductive material is nickel coated carbon fibers.

99. The method of claim 97 or 98, wherein the composite has a volume resistivity of about 0.1 Ohms to about 1 Megaohm.

100. The method of claim 97 or 98, wherein the composite has a volume resistivity of about 0.1 Ohms to about 3 Ohms.

101. The method of any one of claims 64 to 100, having a density of about 70 kg/m³ to about 700 kg/m³.

102. The method of any one of claims 64 to 100, having a density of about 100 kg/m³ to about 500 kg/m³.

103. The method of any one of claims 64 to 100, having a density of about 100 kg/m³ to about 400 kg/m³.

104. The method of any one of claims 64 to 103, wherein the composite has a relative density of about 0.07 to about 0.8.

105. The method of any one of claims 64 to 104, wherein the composite has a relative density of about 0.1 to about 0.8.

106. The method of any one of claims 64 to 104, wherein the composite has a relative density of about 0.1 to about 0.5.

107. The method of any one of claims 64 to 104, wherein the composite has a relative density of about 0.2 to about 0.5.

108. The method of any one of claims 64 to 104, wherein the composite has a relative density of about 0.2 to about 0.4.

109. The method of any one of claims 64 to 104, wherein the composite has a relative density of about 0.2 to about 0.35.

110. The method of any one of claims 64 to 109, wherein the composite is isotropic.

111. The method of any one of claims 64 to 110, wherein the composite is viscoelastic.

112. The method of any one of claims 64 to 110, wherein the composite is viscoelastic-viscoplastic.

113. A method of producing a layered syntactic elastomeric composite, comprising:

producing a plurality of layers of elastomeric composite according to the method of any one of claims 64 to 112,

wherein the elastomer used in each of the layers is the same;

and wherein

each layer has a different relative density compared to the relative density of an adjacent layer,

each layer has a different average cell size compared to the average cell size of an adjacent layer, and/or

each layer comprises insoluble particles that are different, optionally having a different size, diameter, shape and/or composition, compared to the insoluble particles comprised in an adjacent layer.

114. A method of producing a layered elastomeric composite, comprising:

producing a plurality of layers of elastomeric composite according to the method of any one of claims 64 to 112,

wherein the elastomer used in each of the layers is the same;

and wherein

each layer has a different relative density compared to the relative density of an adjacent layer,

each layer has a different average cell size compared to the average cell size of an adjacent layer, and/or

each layer comprises insoluble particles that are different, optionally having a different size, diameter, shape and/or composition, compared to the insoluble particles comprised in an adjacent layer.

115. The method of claim 113 or 113, comprising:

preparing a first mixture and curing the first mixture to obtain a first layer;

preparing a second mixture, pouring the second mixture adjacent to the first layer and curing the second mixture to obtain a second layer; and

optionally preparing a third mixture, pouring the third mixture adjacent to the first or second layer, and curing the third mixture to obtain a third layer.

116. A method of preparing a syntactic cell-graded elastomer composite, comprising:

preparing a first mixture comprising an elastomer and first insoluble particles;

preparing a second mixture comprising the elastomer and second insoluble particles, the second insoluble particles having a larger size and/or diameter compared to the first insoluble particles;

optionally preparing a third mixture comprising the elastomer and third insoluble particles, the third insoluble particles having a larger size and/or diameter compared to the second insoluble particles;

optionally preparing a fourth mixture comprising the elastomer and fourth insoluble particles, the fourth insoluble particles having a larger size and/or diameter compared to the third insoluble particles;

pouring the first mixture into a cavity;

pouring the second mixture into the cavity;

optionally pouring the third mixture into the cavity;

optionally pouring the fourth mixture into the cavity;

optionally pressing down the combined mixtures;

curing the mixture to obtain the composite; and

reducing the size of the insoluble particles.

wherein the insoluble particles have at least one of compressive yield point, decomposition temperature point, melting point and density lower than at least one of compressive yield point, decomposition temperature point, melting point and density of the elastomer.

117. A method of preparing a cell-graded elastomer composite, comprising:

preparing a first mixture comprising an elastomer and first insoluble particles;

preparing a second mixture comprising the elastomer and second insoluble particles, the second insoluble particles having a larger size and/or diameter compared to the first insoluble particles;

optionally preparing a third mixture comprising the elastomer and third insoluble particles, the third insoluble particles having a larger size and/or diameter compared to the second insoluble particles;

optionally preparing a fourth mixture comprising the elastomer and fourth insoluble particles, the fourth insoluble particles having a larger size and/or diameter compared to the third insoluble particles;

pouring the first mixture into a cavity;

pouring the second mixture into the cavity;

optionally pouring the third mixture into the cavity;

optionally pouring the fourth mixture into the cavity;

optionally pressing down the combined mixtures;

curing the mixture to obtain the composite; and

reducing the size of the insoluble particles.

wherein the insoluble particles have at least one of compressive yield point, decomposition temperature point, melting point and density lower than at least one of compressive yield point, decomposition temperature point, melting point and density of the elastomer.

118. The method of any one of claims 64 to 117, wherein the composite comprises open cells.

119. The composite of any one of claims 1 to 63 or the method of any one of claims 64 to 118, wherein the insoluble particles have a compressive yield point lower than the compressive yield point of the elastomer.

120. The composite of any one of claims 1 to 63 or the method of any one of claims 64 to 118, wherein the insoluble particles have a decomposition temperature point lower than the decomposition temperature point of the elastomer.

121. The composite of any one of claims 1 to 63 or the method of any one of claims 64 to 118, wherein the insoluble particles have a melting point lower than the melting point of the elastomer.

122. The composite of any one of claims 1 to 63 or the method of any one of claims 64 to 118, wherein the insoluble particles have a density lower than the density of the elastomer.

123. The composite of any one of claims 1 to 63 or the method of any one of claims 64 to 118, wherein the composite has an open cell structure.

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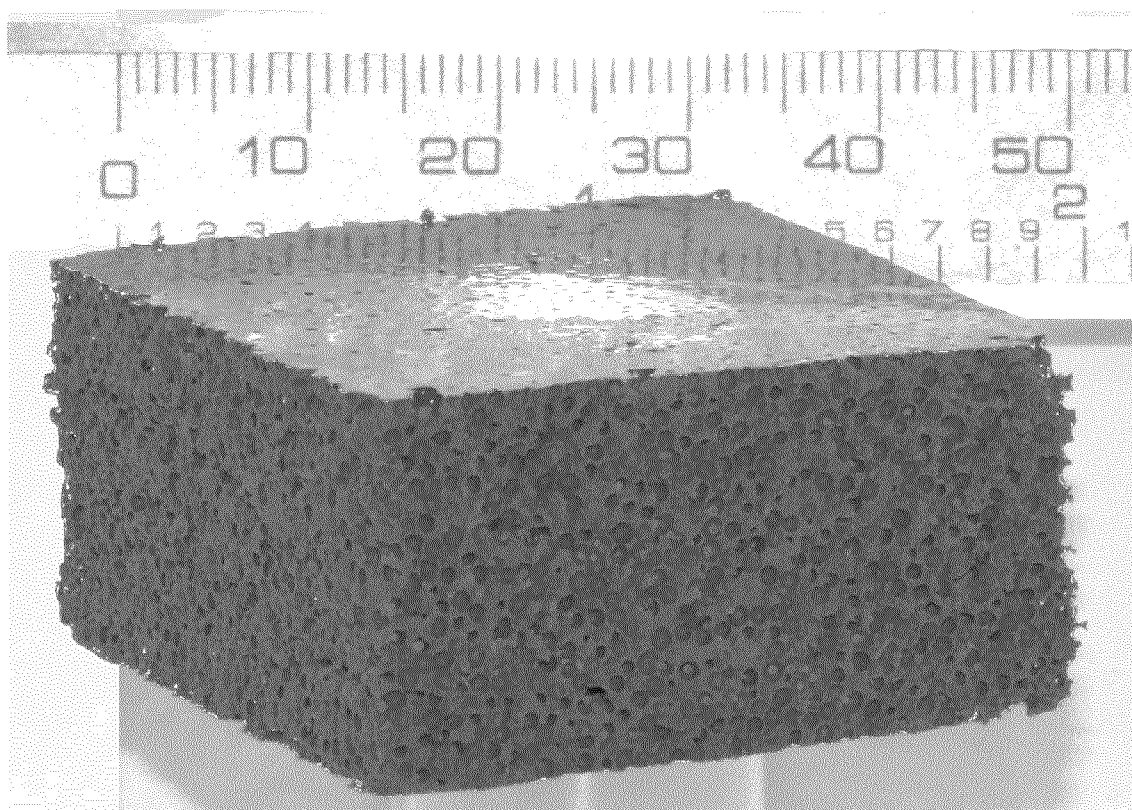


FIG. 1

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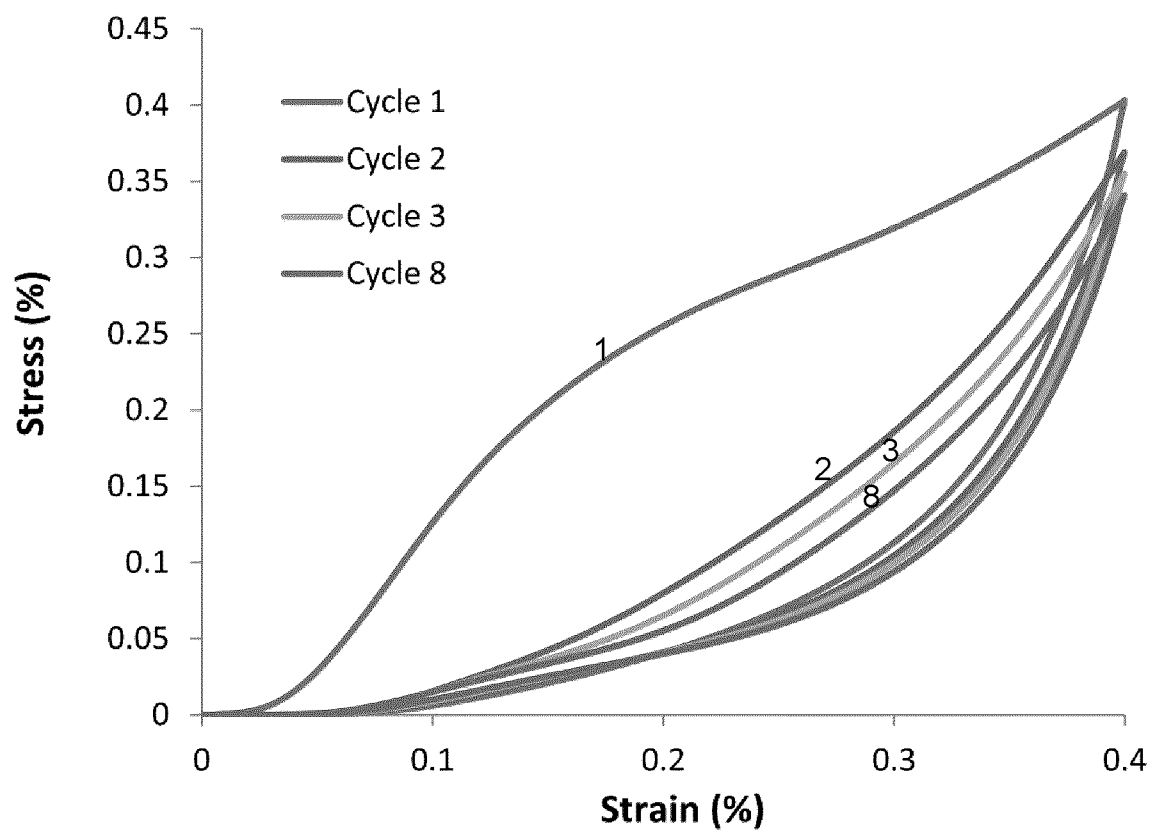


FIG. 2

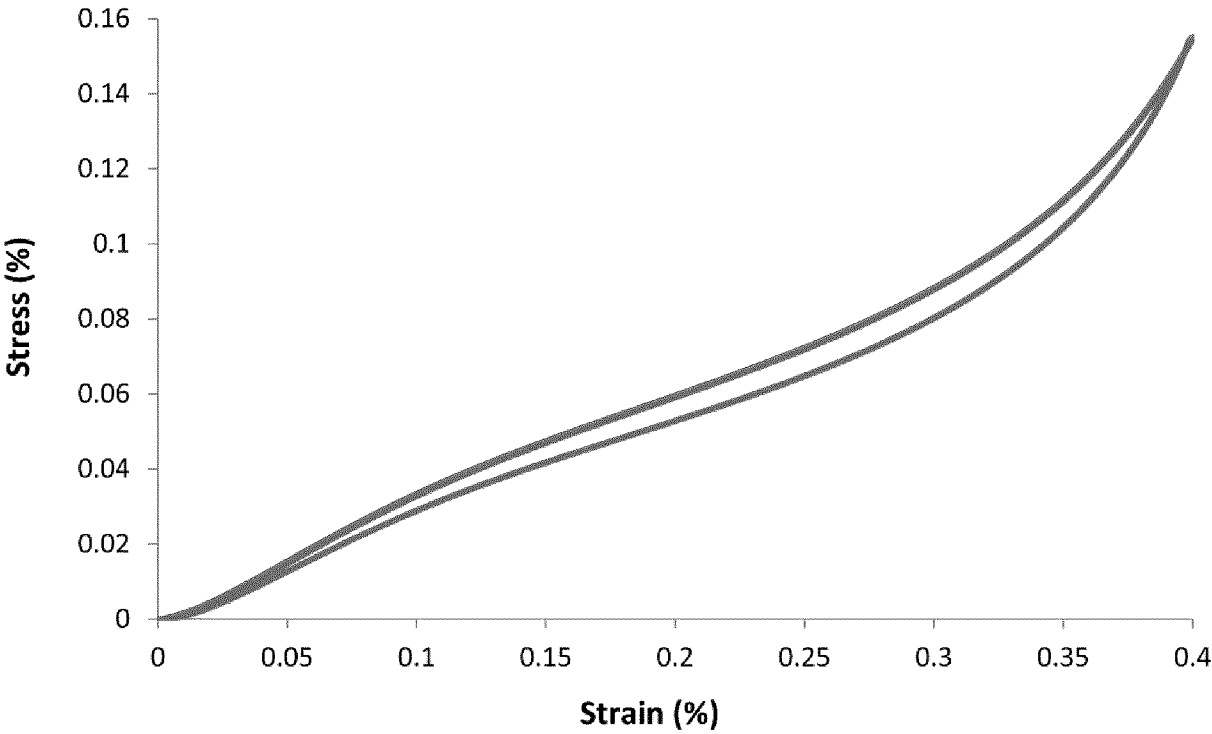


FIG. 3

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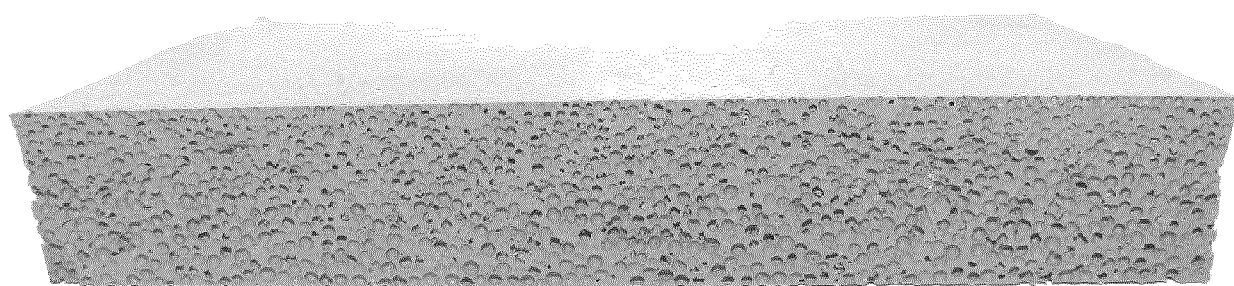


FIG. 4

INTERNATIONAL SEARCH REPORT

International application No.

PCT/CA2019/051747

A. CLASSIFICATION OF SUBJECT MATTER

IPC: **C08L 101/12** (2006.01), **B32B 7/022** (2019.01), **C08J 3/20** (2006.01), **C08K 11/00** (2006.01), **C08K 9/10** (2006.01), **C08L 83/04** (2006.01)

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)

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

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

Questel Orbit, CIPO library discovery tool

Key words: melamine, urea, phenol formaldehyde, silicone, polysiloxane, vinyl ester, polyester, epoxy, polyurethane, natural rubber, expanded polystyrene, particles, beads, pores, porous, composite, elastomer, syntactic foam, shrink, shrunk, compressed, reduced, deflated

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	ADAM, M. et al., " <i>Hierarchically ordered foams derived from polysiloxanes with catalytically active coatings</i> ". Journal of the European Ceramic Society, 17 January 2014 (17-01-2014), Vol. 34, pp. 1715-1725. *see entire document	1-123
X	MALEWSKA, E. et al., " <i>Physical and mechanical properties of rigid polyurethane foams modified with polystyrene beads</i> ". e-Polymers, 10 July 2012 (10-07-2012), Vol. 055, pp. 1-10. *see entire document	1-20, 28, 29, 64-72, 80-87, 89
X	WO2006015440 (QUEVAUVILLIERS, E.) 16 February 2006 (16-02-2006) *see entire document	1-16, 27, 29, 30, 32, 33, 42-44

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

☒ See patent family annex.

* "A" "D" "E" "L" "O" "P"	Special categories of cited documents: document defining the general state of the art which is not considered to be of particular relevance document cited by the applicant in the international application earlier application or patent but published on or after the international filing date document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified) document referring to an oral disclosure, use, exhibition or other means document published prior to the international filing date but later than the priority date claimed	"T" "X" "Y" "&"	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 document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art document member of the same patent family
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Date of the actual completion of the international search
24-01-2020 (24 January 2020)

Date of mailing of the international search report
17 February 2020 (17-02-2020)

Name and mailing address of the ISA/CA
Canadian Intellectual Property Office
Place du Portage I, C114 - 1st Floor, Box PCT
50 Victoria Street
Gatineau, Quebec K1A 0C9
Facsimile No.: 819-953-2476

Authorized officer

Erin Todd (819) 639-9402

INTERNATIONAL SEARCH REPORT

International application No.

PCT/CA2019/051747

C (Continuation). DOCUMENTS CONSIDERED TO BE RELEVANT		
Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	DE102013007999 (FORERO, S.) 14 November 2013 (14-11-2013) *see entire document *computer translation	1, 2, 14, 15, 64, 65, 81, 82 1-16, 29, 32, 33, 42-44, 64-71, 80-82, 96
X	CA2438303A1 (QUIST, P.I. et al.) 22 August 2002 (22-08-2002) *see entire document	

INTERNATIONAL SEARCH REPORT
Information on patent family members

International application No.
PCT/CA2019/051747

Patent Document Cited in Search Report	Publication Date	Patent Family Member(s)	Publication Date
WO2006015440A1	16 February 2006 (16-02-2006)	CN101128303A TW200613412A	20 February 2008 (20-02-2008) 01 May 2006 (01-05-2006)
DE102013007999A1	14 November 2013 (14-11-2013)	None	
CA2438303A1	22 August 2002 (22-08-2002)	CA2438303C AP1627A AT377041T AU755917B2 AU755917C BR0207250A BR0207250B1 CN1491254A CN1249134C CY1107869T1 CZ20032175A3 DE60223228D1 DE60223228T2 DK1368421T3 EA200300881A1 EA006962B1 EP1368421A1 EP1368421A4 EP1368421B1 ES2296904T3 HK1060364A1 HU0303275A2 HU0303275A3 HU226899B1 IL157212A IL157212D0 JP2004521975A JP4168418B2 KR20030081444A KR100714373B1 MXPA03007230A NO20033533D0 NO20033533L NO333855B1 NZ527196A OA12447A PL365347A1 PL206957B1 PT1368421E SK10262003A3 SK288044B6 UA75637C2 US2004115455A1 US2009214853A1 WO02064672A1 ZA200306005B	02 February 2010 (02-02-2010) 06 July 2006 (06-07-2006) 15 November 2007 (15-11-2007) 02 January 2003 (02-01-2003) 12 June 2003 (12-06-2003) 06 July 2004 (06-07-2004) 07 February 2012 (07-02-2012) 21 April 2004 (21-04-2004) 05 April 2006 (05-04-2006) 19 June 2013 (19-06-2013) 12 May 2004 (12-05-2004) 13 December 2007 (13-12-2007) 14 August 2008 (14-08-2008) 25 February 2008 (25-02-2008) 25 December 2003 (25-12-2003) 30 June 2006 (30-06-2006) 10 December 2003 (10-12-2003) 11 October 2006 (11-10-2006) 31 October 2007 (31-10-2007) 01 May 2008 (01-05-2008) 09 May 2008 (09-05-2008) 29 December 2003 (29-12-2003) 28 November 2005 (28-11-2005) 28 January 2010 (28-01-2010) 03 December 2007 (03-12-2007) 19 February 2004 (19-02-2004) 22 July 2004 (22-07-2004) 22 October 2008 (22-10-2008) 17 October 2003 (17-10-2003) 02 May 2007 (02-05-2007) 14 February 2005 (14-02-2005) 11 August 2003 (11-08-2003) 08 October 2003 (08-10-2003) 30 September 2013 (30-09-2013) 26 March 2004 (26-03-2004) 23 May 2006 (23-05-2006) 27 December 2004 (27-12-2004) 29 October 2010 (29-10-2010) 14 January 2008 (14-01-2008) 06 April 2004 (06-04-2004) 04 February 2013 (04-02-2013) 15 October 2003 (15-10-2003) 17 June 2004 (17-06-2004) 27 August 2009 (27-08-2009) 22 August 2002 (22-08-2002) 01 September 2004 (01-09-2004)