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(54) **Title:** COMPOSITIONS COMPRISING STRUCTURED NON-AQUEOUS LIQUID PHASE

(57) **Abstract:** The present invention has as an objective to provide non-aqueous liquids non-aqueous liquids that are structured by lipophilic fibrous materials. This is required to develop consumer products like foods or personal care products, which are healthy, or have beneficial sensory benefits. The non-aqueous liquid phases may be oils or other lipophilic compounds. These non-aqueous liquid phases may be incorporated as ingredients of products such as oil-in-water emulsions or water-in-oil emulsions. This objective can be met non-aqueous liquid phases which are structured by a lipophilic fibrous material comprising a polymer, and wherein the fibrous material has been prepared by a method involving spinning.



COMPOSITIONS COMPRISING STRUCTURED NON-AQUEOUS LIQUID PHASE

The present invention relates to a composition comprising a non-aqueous liquid phase, that is structured by fibres that are produced by a spinning method.

5

BACKGROUND OF THE INVENTION

The structure and morphology of a consumer product is essential for the properties and the appreciation of the product. For example a food product like a margarine should not be too soft and not be too hard, and should be spreadable under all normal household
10 conditions and should melt at in body temperature when consumed. This can be achieved by using a correct ratio of saturated and unsaturated fats and oils in the formulation of the product. Similarly a deodorant stick should keep its consistency during storage, nevertheless should deliver its constituents when applied to the skin. These required properties may lead to contradictory requirements in product development.

15

It is well known that thickeners and fibres can be used to create useful structures, both in foods as well as in cosmetic or personal care products. Numerous fibrous materials have been described, and several methods have been disclosed to produce fibrous materials. The production of fibres out of vegetable or dairy proteins has been described, in order to
20 use these fibres as meat replacers. Additionally fibres made by electrospinning are used in medical applications, especially as wound dressing materials.

It is well known that thickeners and fibres can be used to create useful structures, both in foods as well as in cosmetic or personal care products. Numerous fibrous materials have
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30 WO 2007/068344 A1 discloses fibres like microcrystalline cellulose, that have been modified to give them surface-active properties, and that are used as stabiliser for aerated food products and emulsions.

WO 89/10068 discloses microfragmented ionic polysaccharide/protein complex aqueous
35 dispersions that are used for nutritious bulking, viscosity or texture control agents (also fat

replacer) in food products. These materials may be formed in the form of fibres, and the method may involve a fragmentation step by homogenisation.

WO 01/54667 relates to an electrospun pharmaceutical composition comprising an active
5 agent, and a polymeric carrier for use in therapy. The carrier may be water-soluble or water-insoluble.

WO 2006/136817 A1 discloses various polymers which may be used as source to create
10 fibres by electrospinning.

US 4,287,219 discloses fibres made from proteins, with fat containing phase in the core of
the fibrous materials. These are used as meat replacers.

EP 1 743 975 A1 discloses liquids containing fibres containing thermoplastic polymers.
15

Fernandez A. et al. (Food Hydrocolloids, vol. 23, 2009, 1427–1432) disclose
encapsulation of beta-carotene in electrospun fibres of zein prolamin, to protect the beta-
carotene from oxidation.

20 Li Y. et al. (Journal of Food Science, vol. 74, 2009, C233-C240) discloses electrospun
zein fibres as carriers to stabilise (-)-epigallocatechin gallate.

Kriegel C.-A. et al. (Critical Reviews in Food Science and Nutrition, vol. 48, 2008, 775-
779) disclose the use of electrospun fibres in food products as ingredients.
25

Wongsasulak S. et al. (Journal of Food Engineering, vol. 98, 2010, p. 370–376) discloses
electrospinning of food-grade nanofibres from cellulose acetate and egg albumen blends.
These can be used for controlled delivery of nutraceuticals or pharmaceuticals to the
gastro-intestinal tract.

30 Schiffman J.D. et al. (Polymer Reviews, vol. 48, 2008, p. 317–352) disclose various
combinations of cellulose materials and other polymers to create fibres by electrospinning.
They also describe that proteins can be used to create fibres by means of electrospinning.

35

SUMMARY OF THE INVENTION

In spite of these disclosures, there still is a need to produce new compositions containing non-aqueous liquids that are structured by lipophilic fibrous materials. This is required to develop consumer products like foods or personal care products, which are healthy, or
5 have beneficial sensory benefits. The non-aqueous liquid phases may be oils or other lipophilic compounds. These non-aqueous liquid phases may be incorporated as ingredients of products such as oil-in-water emulsions or water-in-oil emulsions.

We have now determined that this objective can be met non-aqueous liquid phases which
10 are structured by a lipophilic fibrous material comprising a polymer, wherein the fibrous material comprises one or more compounds chosen from the group of prolamins and lipophilic cellulose derivatives, and wherein the fibrous material has been prepared by a method involving spinning. These fibres are very efficient structurants of non-aqueous liquids, such as vegetable oil in a food product, or lipid compounds in personal care
15 products such as skin creams.

Using these non-aqueous liquid structured by lipophilic fibres has the advantage that in case of structuring food products, less saturated fats are required to structure the food product. Nevertheless similar sensory and in-use physical properties can be achieved, like
20 rheology, spreadability, storage stability, and chemical stability. Reducing the amounts of saturated fat in a product, makes a food product healthier. When applied in personal care products, new structures can be made which are liked by consumers. Examples of this are a superior sensory feeling such as silky feel (like in skin care cream), or delivery of actives on the skin (like in skin cleansing product). Also improved temperature stability
25 can be achieved.

Hence in a first aspect the present invention provides a composition comprising a non-aqueous liquid phase, wherein the liquid phase is structured by a lipophilic fibrous material comprising a polymer, wherein the fibrous material comprises one or more compounds
30 chosen from the group of prolamins and lipophilic cellulose derivatives, and wherein the fibrous material has been prepared by a method involving spinning.

In a second aspect the present invention provides a method for production of a composition according to the first aspect of the invention, comprising the steps:
35 a) spinning of a fibrous material from a polymer, wherein the polymer is in liquid form during the spinning; and

- b) dispersing the fibrous material obtained from step a) in a non-aqueous liquid; and
 - c) homogenising the mixture from step b), to fragment the fibrous material to an average length from 1 micrometer to 10 millimeter; and
 - d) bringing the mixture obtained from step c) into contact with one or more other
- 5 ingredients of the composition.

DETAILED DESCRIPTION

Unless defined otherwise, all technical and scientific terms used herein have the same
10 meaning as commonly understood by one of ordinary skill in the art.

All percentages, unless otherwise stated, refer to the percentage by weight. The abbreviation 'wt%' refers to percentage by weight.

15 In the context of the present invention, an average particle diameter is generally expressed as the $d_{3,2}$ value, which is the Sauter mean diameter, unless stated otherwise. The Sauter mean diameter is the diameter of a sphere that has the same volume/surface area ratio as a particle of interest. In case of fibrous material of which the cross-section may not be completely circular, the diameter of the fibre as expressed herein, is the
20 diameter of a circle having the same surface area as the cross-section of the fibre. Also the $d_{4,3}$ value, which is the volume weighted mean diameter, may be used herein. The volume based particle size equals the diameter of the sphere that has same the same volume as a given particle.

25 In case a range is given, the given range includes the mentioned endpoints.

By the term 'fibre' or 'fibrous material', we mean any water-insoluble structure wherein the ratio between the length and the diameter ranges from about 10 to infinite. Here, the diameter means the largest distance of the cross-section. The materials of the "fibre"
30 substance can be organic, inorganic, polymeric and macromolecular. The cross-sectional area of the fibre may be not completely circular, and may be in the form of an oval or the like.

A 'non-aqueous liquid phase' as used in this context relates to a liquid at ambient
35 conditions (temperature about 20°C, atmospheric pressure), and where said liquid has a tendency to flow, as determined by having a loss modulus G'' larger than the storage

modulus G' at shear rates $\dot{\gamma}$ (gamma) ranging from 1 per second to 500 per second. The non-aqueous character is defined as the material not being able to dissolve more than 10% by weight in water under ambient conditions, preferably less than 5% by weight, preferably less than 1% by weight, preferably less than 0.5% by weight, preferably less than 0.2% by weight.

Composition comprising structured non-aqueous liquid phase

In a first aspect the present invention provides a composition comprising a non-aqueous liquid phase, wherein the liquid phase is structured by a lipophilic fibrous material comprising a polymer, wherein the fibrous material comprises one or more compounds chosen from the group of prolamins and lipophilic cellulose derivatives, and wherein the fibrous material has been prepared by a method involving spinning.

Spinning is a process that can be used to create fibres of polymeric materials. A preferred way of performing a spinning process, is by pressing a polymer in a liquid form through for example one or more nozzles or other orifices, to form continuous filaments. Usually the pressing through the nozzle may be done using an extruder, and there may be multiple nozzles to create parallel filaments, like in a spinneret to form multiple continuous filaments. The polymer may be brought in liquid form by melting, or by dissolving in a suitable solvent. By pressing the molten polymer through the nozzle it may solidify by cooling (melt spinning). If the polymer is dissolved in a solvent, it may solidify by precipitation in a liquid bath (wet spinning), or may solidify by evaporation of the solvent (dry spinning). Examples of the spinning process are shear-driven spinning, centrifugation spinning, jet spinning, and electrospinning.

In the context of the present invention, a lipophilic fibrous material is considered to be a fibre which preferably has a three-phase contact angle between a drop of non-aqueous liquid, and a film of the fibrous material, and air of less than 70° at 20°C . Here a non-aqueous liquid preferably comprises sunflower oil or silicon oil, or derivatives of silicon oil. Preferably the contact angle is less than 50° , more preferred less than 40° .

In the present context, the contact angle is measured as the angle in the droplet, as schematically depicted in Figure 1.

Preferably the non-aqueous liquid phase is chosen from the group consisting of vegetable oil or fat, dairy oil or fat, fish oil or fat, mineral oil or mineral oil derivative, petrolatum or petrolatum derivative, silicon oil or silicon oil derivative. Also other animal oils like lard and tallow may be within the scope of the present invention.

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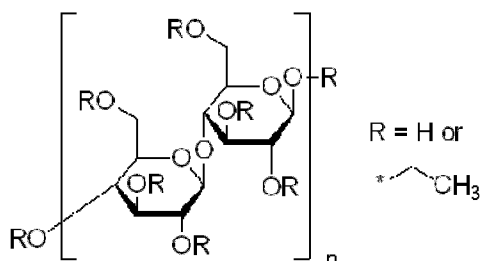
Lipophilic cellulose derivatives

- In order to achieve good structuring capacity, a fibre should have a good compatibility or adhesion to a continuous non-aqueous liquid phase. A poor compatibility causes
- 10 agglomeration of the fibres and weak interaction with the continuous phase, which may induce a reduction of mechanical properties. The preferential route is to use fibres that are compatible with the continuous phase, which are either made from appropriate materials or modified chemically or physically during the process of their production.
- 15 The compatibility between fibre and non-aqueous liquid can be estimated by measuring the fibre wetting by the non-aqueous liquid. Measure for this is the three phase contact angle of non-aqueous liquid or water droplet in air placed on the substrate made from the same material the fibres are made from. Alternatively the contact angle of non-aqueous liquid droplet in water (or other way around) on the substrate can be measured as well.
- 20 Here the implicit assumption is that both fibre and substrate have the comparable surface roughness and that line tension effects can be neglected. A better non-aqueous liquid wetting (or poorer water wetting) are indicative of better compatibility between the fibre and the non-aqueous liquid phase. Therefore one can convert the problem of compatibility between the fibre and non-aqueous liquid phase to a problem of preparing fibres with
- 25 optimal lipophilicity measured via the contact angle.

- The fibrous material comprises one or more compounds chosen from the group of prolamins and lipophilic cellulose derivatives. The lipophilic cellulose derivative is defined as a cellulose derivative wherein preferably the three-phase contact angle between
- 30 sunflower oil, and a film of the lipophilic cellulose derivative, and air is less than 70° at 20°C. Preferably the angle is less than 50°, most preferred less than 40°.

- The contact angle can be measured using standard equipment like the Drop shape analysis DSA100 (Krüss GmbH, Neunkirchen am Brand, Germany). This technique is
- 35 common in the art.

Preferably the lipophilic cellulose derivative comprises an alkylated cellulose. Examples of such alkylated celluloses are methyl-ethylcellulose, ethylcellulose, propylcellulose or butylcellulose. Another preferred lipophilic cellulose derivative is cellulose diacetate. Also combinations of these compounds are within the scope of the present invention. Most
 5 preferred lipophilic cellulose derivative comprises ethylcellulose. The general structural formula of ethylcellulose is:



The degree of substitution of the ethylcellulose preferably used in the present invention is
 10 preferably from 2 to 3, more preferably about 2.5. The average number of hydroxyl groups substituted per anhydroglucose unit (the 'monomer') is known as the 'degree of substitution' (DS). If all three hydroxyls are replaced, the maximum theoretical DS of 3 results.

15 Suitable sources and types of the ethylcellulose preferably used in the present invention are supplied by for example Ashland (formerly Hercules), Aldrich, and Dow Chemicals. Suitable ethylcellulose preferably has a viscosity ranging from 5 to 300 cP at a concentration of 5 % in toluene/ethanol 80:20, more preferably from 100 to 300 cP at these conditions.

20

Prolamins

Prolamins are a group of plant storage proteins having a high proline content and are found in the seeds of cereal grains. Examples of these grains are wheat (protein gliadin), barley (protein hordein), rye (protein secalin), corn (protein zein) and as a minor protein,
 25 avenin in oats. The prolamins are characterised by a high glutamine and proline content and are generally soluble only in strong alcohol solutions.

Preferably the prolamin is chosen from the group of zein, gliadin, hordein, secalin, and avenin. Also combinations of these compounds are within the scope of the present
 30 invention.

Zein is the alcohol-soluble protein of corn and is classified as a prolamin. Biologically, zein is a mixture of proteins varying in molecular size and solubility. These proteins can be separated by differential solubilities and their related structures into four distinct types: alpha, beta, gamma, and delta. Alpha-zein is by far the most abundant, accounting for
5 about 70% of the total. The next most abundant zein is gamma-zein, contributing to about 20% of the total.

Gluten is a storage protein from wheat and comprises two major protein groups, namely the gliadins (molecular weight 30,000–80,000) and glutenin polymers (molecular weight
10 higher than 100,000). It is classified as prolamins due to the presence of aqueous alcohol soluble gliadin groups.

Gliadin is a glycoprotein present in wheat and several other cereals within the grass genus *Triticum*. Gliadins are prolamins and are separated on the basis of electrophoretic
15 mobility and isoelectric focusing. Together with glutenin it forms an important component of wheat gluten. Hordein is a major storage protein from barley. It is a glycoprotein also classified as prolamin based on its solubility characteristics. Secalin, a storage protein found in rye, with high glutamine and proline content and low lysine content is also classified as prolamin.

20

Lipids

Preferably the fibrous material comprises one or more lipid compounds. Lipid compounds in the context of the present invention are lipophilic materials which often are from natural
25 origin, but may also be a synthetic compound. Preferably the lipid compound comprises lecithin, fatty acid, monoglyceride, diglyceride, triglyceride, phytosterol, phytostanol, phytosteryl-fatty acid ester, phytostanyl-fatty acid ester, wax, fatty alcohol, carotenoid, oil-soluble colourant, oil-soluble vitamin, oil soluble flavour, or oil soluble fragrance. Also combinations of these compounds are within the scope of the present invention.

30

Oils and fats such as dairy fats, or vegetable oils are a common source for monoglycerides, diglycerides, and triglycerides. Examples of fat-soluble vitamins are vitamin A, vitamin D2, vitamin D3, vitamin E, and vitamin K. These vitamins include all compounds which function as the respective vitamin. The carotenoids include alpha-
35 carotene, beta-carotene, lycopene, lutein, zeaxanthin.

Also materials like mineral oils, petrolatum, and silicon oils, and derivatives of these compounds are examples of compounds which could be used in the fibres of the invention as the lipid compound in the context of the present invention.

- 5 Lecithin: is a general term for a mixture which may originate from plant origin (e.g. soy bean) or animal origin (e.g. egg yolk), and is used as emulsifier. The most important compounds in lecithin are phosphatidylcholine, phosphatidylethanolamine, and phosphatidylinositol. In commercially available lecithins also free fatty acids, triglycerides and mono- and diglycerides can be present. The nature of the phosphoric group and said
10 fatty acids determine the emulsification properties of lecithin.

Fatty acid: fatty acids suitable in the present invention are C3 fatty acids and longer chains, preferably at least C12, up to preferably C26. The aliphatic tail may be saturated or unsaturated. The chain can be unbranched or have branches like a hydroxy, methyl- or ethyl group. The fatty acid suitable in the present invention consists of minimum 3 carbon
15 atoms and a maximum of 26.

Monoglyceride: an ester of glycerol and one fatty acid, wherein the fatty acid may be as described above.

Diglyceride: an ester of glycerol and two fatty acids, wherein the fatty acids may be as described above.

- 20 Triglyceride: a glycerol which is esterified with three fatty acids, as described above. The fatty acids may be saturated, or monounsaturated or polyunsaturated. In the context of the present invention, triglycerides are understood to be edible oils and fats. As used herein the term 'oil' is used as a generic term for oils and fats either pure or containing compounds in solution. Oils can also contain particles in suspension.

- 25 As used herein the term 'fats' is used as a generic term for compounds containing more than 80% triglycerides. They can also contain diglycerides, monoglycerides and free fatty acids. In common language, liquid fats are often referred to as oils but herein the term fats is also used as a generic term for such liquid fats. Fats include: plant oils (for example: allanblackia oil, apricot kernel oil, arachis oil, arnica oil, argan oil, avocado oil, babassu oil,
30 baobab oil, black seed oil, blackberry seed oil, blackcurrant seed oil, blueberry seed oil, borage oil, calendula oil, camelina oil, camellia seed oil, castor oil, cherry kernel oil, cocoa butter, coconut oil, corn oil, cottonseed oil, evening primrose oil, grapefruit oil, grape seed oil, hazelnut oil, hempseed oil, illipe butter, jojoba oil, lemon seed oil, lime seed oil, linseed oil, kukui nut oil, macadamia oil, maize oil, mango butter, meadowfoam oil, melon seed oil,
35 moringa oil, mowrah butter, mustard seed oil, olive oil, orange seed oil, palm oil, palm kernel oil, papaya seed oil, passion seed oil, peach kernel oil, plum oil, pomegranate seed

oil, poppy seed oil, pumpkins seed oil, rapeseed (or canola) oil, red raspberry seed oil, rice bran oil, rosehip oil, safflower oil, seabuckthorn oil, sesame oil, shea butter, soy bean oil, strawberry seed oil, sunflower oil, sweet almond oil, walnut oil, wheat germ oil); fish oils (for example: sardine oil, mackerel oil, herring oil, cod-liver oil, oyster oil); animal oils
5 (for example: butter or conjugated linoleic acid, lard or tallow); or any mixture or fraction thereof. The oils and fats may also have been modified by hardening, fractionation, chemical or enzymatical interesterification or by a combination of these steps.

Phytosterol: a group of steroid alcohols, phytochemicals naturally occurring in plants. At room temperature they are white powders with mild, characteristic odor, insoluble in water
10 and soluble in alcohols. They can be used to decrease the LDL-cholesterol level in plasma in humans.

Phytostanol: similar to the phytosterol, a group of steroid alcohols, phytochemicals naturally occurring in plants. They may also be obtained by hardening a phytosterol.

Phytosteryl-fatty acid ester: a phytosterol which has been modified by esterifying it with a
15 fatty acid.

Phytostanyl-fatty acid ester: a phytostanol which has been modified by esterifying it with a fatty acid.

Waxes: a wax is a non-glyceride lipid substance having the following characteristic properties: plastic (malleable) at normal ambient temperatures; a melting point above
20 approximately 45°C; a relatively low viscosity when melted (unlike many plastics); insoluble in water but soluble in some organic solvents; hydrophobic. Waxes may be natural or artificial, but natural waxes, are preferred. Beeswax, carnauba (a vegetable wax) and paraffin (a mineral wax) are commonly encountered waxes which occur naturally. Some artificial materials that exhibit similar properties are also described as wax
25 or waxy. Chemically speaking, a wax may be an ester of ethylene glycol (ethane-1,2-diol) and two fatty acids, as opposed to fats which are esters of glycerol (propane-1,2,3-triol) and three fatty acids. It may also be a combination of fatty alcohols with fatty acids, alkanes, ethers or esters. Preferred waxes are one or more waxes chosen from carnauba wax, shellac wax or beeswax or their synthetic equivalents. Also paraffin-based synthetic
30 waxes are within the scope of the present invention.

Fibres used in the compositions of the invention

Fibres which are used in the composition according to the invention are fibres which
35 comprise a lipophilic cellulose derivative, or a prolamin, or a combination of a lipophilic cellulose derivative and a prolamin. These classes of compound have been defined herein

before. The fibres additionally may contain a lipid compound as defined herein before. Preferably the fibre comprises a prolamin and a lipid compound; or a lipophilic cellulose derivative and a lipid compound; or a prolamin, a lipophilic cellulose, and a lipid compound.

5

Preferably the fibre used in the composition according to the invention has a length from 1 micrometer to 10 millimeter, wherein the fibre has a diameter from 30 nanometer to 50 micrometer, and wherein the aspect ratio of the fibre is larger than 10. Preferably the fibre has a length from 1 micrometer to 1,000 micrometer, preferably from 2 micrometer to 500 micrometer. Preferably the fibre has a length from 5 micrometer to 300 micrometer.

Preferably the fibre used in the composition according to the invention has a diameter from 50 nanometer to 40 micrometer, preferably from from 100 nanometer to 25 micrometer, preferably from 200 nanometer to 25 micrometer, preferably from 300 nanometer to 10 micrometer, more preferably from 500 nanometer to 5 micrometer.

Preferably the fibre used in the composition according to the invention has an aspect ratio of larger than 50, preferably larger than 100, or preferably even larger than 200 or 500. The aspect ratio is defined as the ratio between the length and the diameter of an individual fibre.

The cross-sectional area of the fibres used in the composition according to the invention may be not completely circular, and may be in the form of an oval or the like. This may mean that for instance the cross-section of a fibre according to the invention may have a longest dimension of 2 to 5 microns, while the shortest dimension may be less than 1 micrometer. In that case the diameter of the fibre as expressed herein, is the diameter of a circle having the same surface area as the cross-section of the fibre.

In one preferred embodiment the fibrous material used in the composition according to the invention comprises a lipophilic cellulose derivative, and a prolamin. In that case preferably the fibre comprises from 1% by weight to 99% by weight of a lipophilic cellulose derivative and from 1% by weight to 99% by weight of a prolamin. Preferably the fibre comprises from 10% by weight to 90% by weight of a lipophilic cellulose derivative and from 10% by weight to 90% by weight of a prolamin. Preferably the fibre comprises from 20% by weight to 80% by weight of a lipophilic cellulose derivative and from 20% by weight to 80% by weight of a prolamin.

In another preferred embodiment the fibrous material used in the composition according to the invention comprises a lipophilic cellulose derivative, and a lipid material. In that case preferably the fibre comprises from 10% by weight to 99.9% by weight of lipophilic

5 cellulose derivative and from 0.7% by weight to 90% by weight of a lipid compound.

Preferably the fibre comprises from 70% by weight to 99% by weight of lipophilic cellulose derivative and from 1% by weight to 30% by weight of a lipid compound. Preferably the fibre comprises from 90% by weight to 98% by weight of lipophilic cellulose derivative and from 2% by weight to 10% by weight of a lipid compound.

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In another preferred embodiment the fibrous material used in the composition according to the invention comprises a prolamin, and a lipid material. In that case preferably the fibre comprises from 10% by weight to 99.9% by weight of a prolamin and from 0.7% by weight to 90% by weight of a lipid compound. Preferably the fibre comprises from 70% by weight to 99% by weight of a prolamin and from 1% by weight to 30% by weight of a lipid compound. Preferably the fibre comprises from 90% by weight to 98% by weight of a prolamin and from 2% by weight to 10% by weight of a lipid compound.

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Preferably the fibrous material prepared used in the composition according to the invention comprises the three mentioned classes of compounds. In that case preferably the fibre comprises 1% by weight to 98.9% by weight of a lipophilic cellulose derivative and from 1% by weight to 98.9% by weight of a prolamin, and from 0.1% by weight to 90% by weight of a lipid compound. Preferably the fibre comprises from 10% by weight to 89% by weight of a lipophilic cellulose derivative and from 10% by weight to 89% by weight of a prolamin, and from 1% by weight to 30% by weight of a lipid compound. Preferably the fibre comprises from 10% by weight to 88% by weight of a lipophilic cellulose derivative and from 10% by weight to 88% by weight of a prolamin, and from 2% by weight to 10% by weight of a lipid compound.

20

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30 Examples of such composite fibres are the following: ethylcellulose-zein composite; ethylcellulose-lecithin composite; ethylcellulose-triglyceride composite; zein- triglyceride composite; ethylcellulose-phytosterol composite; zein-phytosterol composite; ethylcellulose-phytosterol ester composite; and zein-phytosterol ester composite.

35 The functions of lipid or other lipophilic materials in the fibres can be used to tune the lipophilicity of fibres for oil structuring; and/or to tune the meltdown property of fibre

structured oil; and/or to tune the mechanical strength of fibres; and/or to tune the mechanical strength of the fibre network. This behaviour makes it possible to modify the properties of a product containing such structured non-aqueous liquids, for example to create a nice melting emulsion, or a skin cream with favourable properties to apply to the skin.

In principle long fibre lengths lead to good structuring properties when used to structure a non-aqueous liquid phase. On the other hand long hairy structures are often not desired in food products or personal care products. By using fibres having a relatively short length for structuring as compared to longer fibres, the lengths as defined in the claims lead to shear alignment. This means that under shear forces the fibres can align, and therewith give the impression to the consumer that a solid-liquid transition is obtained. This can be perceived to be analogous to a melting curve of a solid fat which melts upon chewing in the mouth or applying to the skin, and gives a positive impression to the consumer.

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Electrospinning

Preferably the fibrous material has been prepared by a method involving electrospinning. In an electrospinning process (as described by Schiffman J.D. et al., Polymer Reviews, vol. 48, 2008, p. 317–352) a molten or dissolved polymer is pressed through for example a capillary, to be collected on a collector. An electric field is applied between the capillary and the collector. Alternatively a system may be used that does not utilise nozzles or capillaries to create cones or jets of polymeric material. An example of such a system is the Nanospider™ technology from Elmarco (Liberec, Czech Republic). A cylinder is partly submerged in a bath of liquid polymer (solution). When the cylinder rotates, a thin layer of polymer is carried on the cylinder surface and exposed to a high voltage electric field. If the voltage exceeds a critical value, a number of electrospinning jets are generated from the polymer bath towards a collector. The jets are distributed over the electrode surface with periodicity. Both the spinning and electrospinning methods are known in the art.

25
30

Preferably the electrospinning process which uses a capillary uses the following settings and parameters. The nozzle from which the solution of the compounds is pressed preferably has an internal diameter of at least 0.1 millimeter. The upper diameter is preferably less than 2 millimeter. During the electrospinning process cone is formed at the bottom and fibers are formed from the tip of this cone. The cone diameter usually is much smaller than the nozzle diameter. The nozzle play an indirect role as it is used as

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electrode as well that it influences electric field gradients. The flow rate from the nozzle preferably is from 0.1 to 1,000 milliliter per hour, preferably from 1 to 100 milliliter per hour. These flow rates are per nozzle; multiple nozzles can be applied to create parallel flows. Preferably the metal collector is placed from 1 to 100 centimeter from the tip of the nozzle, preferably from 10 to 18 centimeter. The collector preferably is a copper mesh covering on a stainless steel mandrel, for example having about 12 cm internal diameter and a length of about 30 cm. The positive lead from a high DC voltage supply is attached to the nozzle metal portion, and the collector is grounded. The voltage between the nozzle and the collector preferably is from 1 kV to 100 kV, preferably from 12 kV to 25 kV. The mandrel may rotate to create an evenly distributed mat during the spinning process, preferably at a rotational speed from 10 to 200 rpm, preferably from 70 to 130 rpm. The temperature and pressure that are applied during the process preferably is from 5°C to 60°C, preferably from 20°C to 40°, preferably from 20°C to 25°C. The pressure may be at atmospheric pressure, but may also be reduced to facilitate the evaporation of the solvent. A mat of electrospun fibres is formed on the grounded copper mesh during the process.

Method for production of fibrous material

In a second aspect the present invention provides a method for production of a composition according to the first aspect of the invention, comprising the steps:

- a) spinning of a fibrous material from a polymer, wherein the polymer is in liquid form during the spinning; and
- b) dispersing the fibrous material obtained from step a) in a non-aqueous liquid; and
- c) homogenising the mixture from step b), to fragment the fibrous material to an average length from 1 micrometer to 10 millimeter; and
- d) bringing the mixture obtained from step c) into contact with one or more other ingredients of the composition.

Step a) of the method comprises the spinning of the fibrous material from a polymer, wherein the polymer is in liquid form during the spinning. The general principles of a spinning process have been described herein before. Especially preferred in step a) the spinning process is an electrospinning process. Also electrospinning has been described herein before.

The polymer being in liquid form should be understood to mean the following. The polymer from which the lipophilic fibre is spun may be used in a molten state when being

spun. Alternatively the polymer is preferably dissolved in a suitable solvent when being spun. When the polymer is pressed through a nozzle, the polymer may solidify, e.g. by cooling, or alternatively the polymer may form a solid fibrous material by evaporation of the solvent.

5

In case of producing composite fibres, the compounds used for making the fibres may be dissolved in a suitable solvent separately, and after dissolving the separate solutions may be combined, before being pressed through the nozzle to be collected on the collector. Alternatively the various compounds may be dissolved in the solvent simultaneously in
10 order to make a mixture of compounds to be pressed through the nozzle. This way fibres are made with a fixed composition.

Alternatively multiple parallel solutions can be made, which are mixed in a micro chamber or junction formed between different channels in line, just before its being pressed through
15 a nozzle. Each solution may have its own pump and consequently its own flow rate. For example one solution contains the lipophilic cellulose derivative, while another solution contains a prolamin. Both solutions are pumped to a three way valve where they mix, and subsequently they are pressed through the nozzle, and a fibrous material is collected on the collector. This has the advantage that the composition of the fibre can be varied during
20 the preparation process, by adjusting the flow rate of one of the pumps relative to the other. Additionally one of the solutions may contain a second compound (e.g. a lipid compound), or a third solution may be coupled in line, parallel to the other two solutions.

The solvent in the method according to the invention is a solvent in which the polymer can
25 be dissolved. Preferred polymers to be used in the method according to the invention are chosen from lipophilic cellulose derivatives and prolamins, or combinations of these.

These materials have been described herein before. A lipid compound as herein described before, may be mixed with the one or more polymers to be spun together with the polymers. Examples of these solvents are alcohols, preferably ethanol, ethyl acetate,
30 acetic acid, acetone, *N,N*-dimethylformamide (DMF), or any suitable combination of these solvents. The concentration of the compounds in the solvent is preferably from 5% by weight to 50% by weight, preferably between 10% by weight to 30% by weight. When the solution is released from the nozzle, the solvent evaporates.

35 In one preferred embodiment the fibrous material used in the composition according to the invention comprises a lipophilic cellulose derivative and a prolamin. In that case the

solution in the spinning step preferably comprises in addition to the solvent from 1% by weight to 99% by weight of a lipophilic cellulose derivative and from 1% by weight to 99% by weight of a prolamin. Preferably the solution comprises from 10% by weight to 90% by weight of a lipophilic cellulose derivative and from 10% by weight to 90% by weight of a prolamin. Preferably the solution comprises from 20% by weight to 80% by weight of a lipophilic cellulose derivative and from 20% by weight to 80% by weight of a prolamin. Here this is all based on the weight of the compounds in the solvent.

In another preferred embodiment the fibrous material used in the composition according to the invention comprises a prolamin and a lipid material. In that case the solution in the spinning step preferably comprises in addition to the solvent from 10% by weight to 99.9% by weight of prolamin and from 0.7% by weight to 90% by weight of a lipid compound. Preferably the solution comprises from 70% by weight to 99% by weight of prolamin and from 1% by weight to 30% by weight of a lipid compound. Preferably the solution comprises from 90% by weight to 98% by weight of prolamin and from 2% by weight to 10% by weight of a lipid compound.

In another preferred embodiment the fibrous material used in the composition according to the invention comprises a lipophilic cellulose derivative and a lipid material. In that case the solution in the spinning step preferably comprises in addition to the solvent from 10% by weight to 99.9% by weight of lipophilic cellulose derivative and from 0.7% by weight to 90% by weight of a lipid compound. Preferably the solution comprises from 70% by weight to 99% by weight of lipophilic cellulose derivative and from 1% by weight to 30% by weight of a lipid compound. Preferably the solution comprises from 90% by weight to 98% by weight of lipophilic cellulose derivative and from 2% by weight to 10% by weight of a lipid compound.

Preferably the fibrous material used in the composition according to the invention comprises the three mentioned classes of compounds. In that case the solution in the spinning step preferably comprises in addition to the solvent from 1% by weight to 98.9% by weight of a lipophilic cellulose derivative and from 1% by weight to 98.9% by weight of a prolamin, and from 0.1% by weight to 90% by weight of a lipid compound. Preferably the solution comprises from 10% by weight to 89% by weight of a lipophilic cellulose derivative and from 10% by weight to 89% by weight of a prolamin, and from 1% by weight to 30% by weight of a lipid compound. Preferably the solution comprises from 10% by weight to 88% by weight of a lipophilic cellulose derivative and from 10% by weight to

88% by weight of a prolamin, and from 2% by weight to 10% by weight of a lipid compound.

The method according to the invention further comprises the steps:

- 5 b) dispersing the fibrous material obtained from step a) in a non-aqueous liquid; and
- c) homogenising the mixture from step b), to fragment the fibrous material to an average length from 1 micrometer to 10 millimeter; and

This way the correct length of fibrous material is obtained. Preferably the length of the
10 fibre that is obtained is from 1 micrometer to 1,000 micrometer, preferably from 2 micrometer to 500 micrometer. Preferably the fibre has a length from 5 micrometer to 300 micrometer.

Preferably in step b) the non-aqueous liquid comprises a vegetable oil, for example
15 sunflower oil, palm oil, olive oil, rapeseed oil, or any other suitable oil or combinations of oils. The oil may be liquid at room temperature, or alternatively may be solid at room temperature, in which case the oil should be melted first by increasing the temperature. A fat or oil from animal origin, such as fish oil, dairy fat, lard, or tallow, may be used as well. Such a vegetable or animal oil obtained from step b) may be used as an ingredient of food
20 products.

The non-aqueous liquid in step b) may also be chosen from materials like mineral oils, petrolatum, and silicon oils, and derivatives of these compounds, and combinations of these. In that case the structured non-aqueous liquid obtained from step b) may be used
25 as an ingredient of personal care products.

In step c) the homogenisation preferably is carried out by subjecting the mixture of fibrous material and non-aqueous liquid to high shear. This high shear can be created by methods common in the art. These methods include rotor-stator systems, e.g. the Ultra-
30 Turrax® (IKA Werke GmbH & Co. KG, Staufen, Germany), or a Silverson mixer (Silverson Machines Ltd., Chesham, Bucks, UK). Another method is high pressure homogenisation. An example of such a high pressure homogeniser is the Microfluidizer® (Microfluidics International Corporation, MA-Newton, USA). Also sonication, a colloid mill, and a ball mill may be used to homogenise the mixture.

In case of a rotor-stator system, e.g. the Ultra-Turrax[®], the rotational speed preferably ranges from 1,000 to 30,000 rpm. The system is preferably homogenised during a period from 15 seconds to 60 minutes. This way a homogeneous mixture of cut fibres in oil can be achieved.

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The amount of fibre to be added to the non-aqueous liquid in step b) of the method ranges from 0.01% by weight to 50% by weight, preferably from 0.1% by weight to 40% by weight, more preferred from 0.2% by weight to 25% by weight, more preferred from 0.5% by weight to 10% by weight. The mixture of homogenised non-aqueous liquid and fibrous material may be used as an ingredient of a food product or a personal care product, as applicable. In that case it may be brought into contact with other ingredients of such product. Alternatively the homogenised non-aqueous liquid from step b) is diluted first with a non aqueous liquid, before being brought into contact with the other ingredients of the product.

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After the homogenisation step the material obtained in step c) may need to be cooled, as the temperature may have risen due to the homogenisation operation.

By the homogenisation step two possible fragmenting operations take place. First, if the fibrous material has been obtained from a spinning process and a mat of fibrous material has been formed, then the homogenisation first leads to break up of the mat. Individual fibres are obtained. Second the long fibres which are formed are broken into smaller pieces, leading to reduction of the length of the fibre. These two steps may take place simultaneously, such that while the fibrous mat is broken into pieces, also long individual fibres are broken into shorter fibres.

In step d) of the method according to the invention, the mixture obtained from step c) is brought into contact with one or more other ingredients of the composition. This way the products can be made which comprise the structured non-aqueous liquid phase. The structured structured non-aqueous liquid phase can be used in the manufacturing of the composition according to the invention in any method which is commonly used for preparing such product.

Compositions containing the structured non-aqueous liquid

Preferred products that may be structured by the composition in the form of fibre according to the invention are food products or personal care products. Food products may be water-in-oil emulsions or oil-in-water emulsions. Personal care products, such as
5 skin creams, may be oil-in-water emulsions. The compositions of the invention may also be double emulsions and multiple emulsions (like oil-in-water-in-oil and water-in-oil-in-water emulsions), of which the non-aqueous liquid phase can be structured by the fibrous material.

10 Hence preferably the composition is a water-in-oil emulsion, containing between 1% by weight and 99% by weight of non-aqueous liquid phase. In the case of food products, which are water-in-oil emulsions such as margarines, butter, and other spreads, the lipid phase can be considered to be the continuous vegetable oil phase or butter fat phase, as applicable.

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The amount of non-aqueous liquid phase in such products may range from 1% by weight to 99% by weight of the product, depending on the product. For example a shortening may contain 99% by weight of edible oil or fat. A margarine contains about 80% edible oils and fats. A water-in-oil spread may contain from 20 to 70% by weight of edible oils and
20 fats.

In another preferred embodiment the composition is an oil-in-water emulsion, containing between 1% by weight and 95% by weight of non-aqueous liquid phase. Examples of oil-in-water emulsions are dressings and mayonnaise-type products, dairy spreads, and body
25 lotions and skin creams.

In the case of food products, the non-aqueous liquid phase can be a lipid phase, for example droplets of a dairy fat or sunflower oil dispersed in an aqueous phase to form an oil-in-water emulsion (like a dressing or a dairy spread). A dressing or mayonnaise may
30 contain from about 5% by weight up to 80% by weight of non-aqueous lipid phase. A dairy spread may contain about 20 to 30% by weight of edible oils and fats.

In case of personal care products, the non-aqueous liquid phase may be chosen from materials like mineral oils, petrolatum, and silicon oils, and derivatives of these
35 compounds, and combinations of these. The concentration of non-aqueous lipid phase may range from 1% by weight up to 50% by weight, or higher.

Preferably the concentration of fibrous material is between 0.01% and 50% by weight, based on the amount of non-aqueous liquid phase, preferably between 0.5% and 10% by weight, based on the amount of non-aqueous liquid phase., preferably from 0.2% by weight to 25% by weight, more preferred from 0.5% by weight to 10% by weight, more preferred maximally 5% by weight.

The fibrous material leads to structuring of the non-aqueous liquid phase. By rheology measurements it can be shown that the physical behaviour of the structured lipids is such that it resembles lipid phases that are structured by solid triglycerides (for example like in butter and margarine), for example in meltdown behaviour upon increase of temperature. Also extended temperature stability can be obtained.

By rheology measurements is meant that storage modulus G' (in Pa) and loss modulus G'' (in Pa) are determined. The elasticity of the system is determined by these rheology measurements. G' and G'' can be determined as function of the strain γ (gamma, in %) at a fixed temperature, and as function of temperature at a fixed strain. When measured as function of temperature, the meltdown behaviour is determined. This determines how structured non-aqueous liquid behaves under the influence of temperature.

20

In general, the storage modulus (G') describe how a material behave like solid, and the loss modulus (G'') describes how a material behave like liquid. If G' is larger than G'' , it means that the oil behaves like a solid or semi-solid, and otherwise, like a liquid. The point where G' equals G'' is a characteristic one: beyond this point the material starts to flow.

25

Temperature-dependent behaviour can characterize the meltdown process of sample. Meltdown is an important property for margarine, but also for other products an appropriate temperature dependency is important. 5°C is to mimic temperature in a refrigerator, and higher value indicates good stability of margarine in the fridge. 25°C is to mimic temperature when margarine is taken out of fridge and in use. A value at 25°C should be slightly lower than that at 5°C, but not too much. 37°C is to mimic mouth temperature. A lower value indicates more thorough meltdown in the mouth. An ideal temperature curve should be high enough from 5° C to 25° C, then decrease gradually with temperature increasing, finally reach a very low level.

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The meltdown index is introduced to determine how much structured non-aqueous liquid loses its structure upon temperature increase. The meltdown index is calculated by:

$$\text{meltdown index} = \log_{10} (G'_{5^{\circ}\text{C}} / G'_{37^{\circ}\text{C}})$$

- 5 A meltdown index of more than 1 is considered to be good, and the higher the meltdown index, the better the structuring behaviour connected with acceptable properties of the structured non-aqueous liquid for the consumer. For example a food emulsion structured by the fibrous network shows similar behaviour as a standard margarine wherein the oil is structured by solid fat crystals. These crystals melt upon consumption and/or use, leading
10 to favourable properties.

Also the viscosity (in Pa.s) of a structured non-aqueous liquid can be determined as function of the shear rate (in 1/s) can be determined in order to compare for example a margarine (structured by solid (saturated) fat crystals) and structured non-aqueous liquid.

15

- Here the temperature influences the interaction within the fibrous network, by increase of temperature the interactions between fibres becomes less, generally leading to softer non-aqueous liquids at increased temperature. The relatively short fibres that structure the non-aqueous liquid may align, to soften the structured non-aqueous liquid. This way the
20 meltdown behaviour of vegetable edible oils that are structured by saturated fat crystals can be mimicked, while not using the saturated fats.

- The advantage of using the fibrous materials according to the invention is that the amount of saturated triglycerides that is required to structure triglycerides can be reduced, which
25 leads to a healthier triglyceride profile of foods containing such structured lipid phase.

The food products of the invention may be all kinds of food products, for instance marinades, sauces, seasonings, butter, spray products, spreads, liquid shallow frying products, seasonings, dressings, mayonnaise, low-fat mayonnaise, and ice cream.

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- Preferably, food products according to the invention are spreads (water-in-oil emulsions or oil-in-water emulsions), margarines (water-in-oil emulsions), dairy products such as butter (water-in-oil emulsion), or liquid water-in-oil emulsions or liquid oil-in-water emulsions designed for shallow frying.

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Many food emulsions are stabilised by solid fat particles, especially margarine-type of emulsions and spreads. These are water-in-oil emulsions. The solid fat usually is mainly a saturated fat, which is considered to be unhealthy when consumed in large amounts.

Therefore replacing saturated fats by the fibrous materials has the advantage that the
5 amount of saturated fat can be reduced, and has a beneficial health effect for the consumer. Nevertheless similar sensory and in-use physical property can be achieved, like rheology, spreadability, storage stability, and chemical stability.

In another preferred embodiment, the present invention provides a personal care product.
10 In this case the personal care product is for example a skin cream, a body lotion, bodywash, handwash, facial foam, shampoo, or hair conditioner. When applied in personal care products, new structures can be made which are liked by consumers. Examples of this are a superior sensory feeling such as silky feel (like in skin care cream), or deliver actives on the skin (like in skin cleansing product).

15 The non-aqueous liquid phase in personal care products as described in here is preferably chosen from materials like mineral oils, petrolatum, and silicon oils, and derivatives of these compounds, and combinations of these.

20

DESCRIPTION OF FIGURES

Figure 1: Schematic representation of the contact angle as defined herein. Droplet is dark, on the surface of a film. The contact angle as indicated is the angle in the droplet between the surface and the tangential line hitting the droplet.

25 **Figure 2:** Three scanning electron microscope pictures of fibrous mat produced by electrospinning ethylcellulose, as described in example 2. From top to bottom: A magnification 80× (scale width 500 micrometer); B magnification 1,000× (scale width 50 micrometer); C magnification 10,000× (scale width 5 micrometer).

Figure 3: Optical microscopy image of ethylcellulose fibre after homogenising. Bar width
30 is 100 micrometer.

Figure 4: Image of the dispersion of ethylcellulose fibre in oil by means of phase contrast microscopy, bar width 100 micrometer; from example 2.

Figure 5: Rheology profile (G' (closed squares and triangles) and G'' (open squares and triangles) (in Pa) as function of temperature (in °C), at fixed strain of 0.1% and frequency
35 of 1 Hz) of ethylcellulose fibre structured oil (squares) and margarine (triangles); from example 2.

Figure 6: Viscosity η (eta, in Pa.s) as function of shear rate $\dot{\gamma}$ (gamma, in 1/s) of various compositions at 25°C, from example 2:

Curve A: Margarine; B: sunflower oil; C: sunflower oil structured with 0.625% ethylcellulose fibre; D: sunflower oil structured with 2% ethylcellulose fibre; E: sunflower oil structured with 5% ethylcellulose fibre.

Figure 7 Two scanning electron microscope pictures of fibrous mat produced by electrospinning zein, as described in example 3. A magnification 2,000 $\times$ (scale width 20 micrometer); B magnification 5,000 $\times$ (scale width 10 micrometer).

Figure 8: Image of the dispersion of zein fibre in oil by means of phase contrast microscopy; bar width 100 micrometer; from example 3.

Figure 9: Rheology profile (G' (closed triangles) and G'' (open triangles) (in Pa) as function of temperature (in °C), at fixed strain of 0.1% and frequency of 1 Hz) of zein fibre structured oil, from example 3.

Figure 10: Viscosity η (eta, in Pa.s) as function of shear rate $\dot{\gamma}$ (gamma, in 1/s) of various compositions at 25°C, from example 3:

Curve A: Margarine; B: sunflower oil; C: sunflower oil structured with 2% zein fibre; D: sunflower oil structured with 5% zein fibre; E: sunflower oil structured with 10% zein fibre.

Figure 11: Two scanning electron microscope pictures of fibrous mat produced by electrospinning zein-inES48, as described in example 4. A: magnification 100 $\times$ (scale width 20 micrometer); B: magnification 10,000 $\times$ (scale width 10 micrometer).

Figure 12: Image of the dispersion of zein-inES48 fibre in oil by means of phase contrast microscopy; bar width 100 micrometer; from example 4.

Figure 13: Rheology profile (G' (closed squares and triangles) and G'' (triangles) (in Pa) as function of temperature (in °C), at fixed strain of 0.1% and frequency of 1 Hz) of zein-inES48 fibre structured oil (triangles) and zein-fibre structured oil (squares), from example 4.

Figure 14: Scanning electron microscope picture of fibres obtained by electrospinning ethylcellulose-zein mixture, as described in example 5; scale width 10 micrometer.

Figure 15: Rheology profile of oil structured with ethylcellulose-zein fibre (G' (closed squares and triangles) and G'' (open squares and triangles) (in Pa) as function of strain $\dot{\gamma}$ (gamma, in %), at 25°C; from example 5. Squares: margarine; triangle: ethylcellulose-zein structured oil.

Figure 16, Left a visual image of a emulsion A structured with zein-inES48 composite fibre, and right a light microscopy image of the structured emulsion; from example 6. Bar size in right hand image is 100 micrometer. The size of the dispersed water droplets was less than 5 micrometer.

Figure 17: Rheology profile (G' (closed squares, triangles, and circles) and G'' (open squares, triangles, and circles) (in Pa) as function of temperature (in °C), at fixed strain of 0.1% and frequency of 1 Hz) of emulsions structured with either zein-inES48 fibres (squares), or ethylcellulose-zein fibres (triangles), or zein fibres (circles); from example 6.

5

EXAMPLES

The following non-limiting examples illustrate the present invention.

10 *Raw materials:*

Ethylcellulose: Aqualon® Ethylcellulose (type N 100) was purchased from Hercules (Widnes, UK). Ethoxyl content was 48.0-49.5%, and degree of substitution was 2.46-2.57. Viscosity was 80-105 mPa.s (at 5% and 25°C in 80/20 toluene/ethanol).

Ethanol (95%) supplied by Shanghai Dongfeng reagent (China)

15 Sunflower oil, (100%), brand: Duoli, supplied by Shanghai Jia Ge Food Co., Ltd (China).

Dimethicone: Polydimethylsiloxanem, trademark & product name: PMX-200 Fluid, 50 cPs, molecular weight: 3,200; refractive index: 1.402, specific gravity: 0.960, manufacturer: Dow Corning (Midland MI, USA).

Demineralised water was obtained from a Millipore filter system.

20 Zein from corn was obtained from Sigma-Aldrich (Schnelldorf, Germany).

inES48 triglyceride: inES48 is an interesterified mixture of 65% dry fractionated palm oil stearin with an iodine value of 14 and 35% palm kernel oil. inES48 contains about 86.2% saturated fatty acids, about 11.5% mono-unsaturated fatty acids, and about 2.3% poly-unsaturated fatty acids, and is free from trans fatty acids. Obtained from Unimills

25 (Zwijndrecht, Netherlands).

PGPR (polyglycerol polyricinoleate, water-in-oil emulsifier) was obtained from Danisco Shanghai (China).

Rheology analysis

30 Rheological analysis was performed in a controlled stress rheometer (Anton Paar, Physica MCR501, Austria) with a parallel-plate (PP 25) measuring system. This configuration has been chosen because of some particles contained in the each sample. Prior to each experiment, samples were left to equilibrate for the same time after loading PP25. Also viscosity experiments were performed using this device.

35

Example 1 – Measurement of Contact Angle

The contact angle of ethylcellulose and zein was determined using a Drop shape analysis DSA100 (Krüss GmbH, Neunkirchen am Brand, Germany). In the present context, the contact angle is measured as the angle in the droplet, as schematically depicted in Figure

5 1. The method applied was the following:

- dissolve ethylcellulose or zein in a solvent to make homogenous solution;
- case a few drops of a solution onto a whole glass slide, under slowly spinning the glass slide to evenly spread the drops on the glass slide;
- dry, smooth and even film formed after solvent has evaporated;
- 10 - 5 microliter drop of demineralised sunflower oil or dimethicone is brought onto the surface, at ambient pressure, humidity and temperature;

Measured contact angles:

Sunflower oil - ethylcellulose film: 37°.

15 of Sunflower - zein film: 29°.

Dimethicone - ethylcellulose film 24°.

Dimethicone - zein film: 31°.

20 Example 2 – Standard Procedure for making Fibres and Structuring*Preparation of electrospun ethylcellulose fibre*

This example shows the basic procedure for making fibres by electrospinning. 1 gram ethylcellulose was dissolved in 9 gram aqueous ethanol solution (90 wt%). The solution was loaded onto a 10 mL syringe plunger. A blunt end stainless steel adapter with outer
25 diameter of 0.9 mm was then placed on the syringe to act as the electrospinning nozzle and charging point for the contained ethylcellulose solution. The filled syringe was placed in a cole-parmer syringe pump with flowing rate of 1.25 ml/hr. The positive lead from the high DC voltage supply was attached to the adapter metal portion. The voltage was set at 19 kV. The grounded collector was a copper mesh covering on a stainless steel mandrel
30 (12 cm internal diameter and 30 cm length) placed 10 cm from the tip of the adapter. The mandrel was rotated at 100 rpm during the spinning process. In the experiment, 10 ml of the ethylcellulose solution was electrospun to form a nice, white mat on the grounded copper mesh. After electrospinning, the ethylcellulose mat was removed from the copper mesh and process for scanning electron microscopy evaluation. The results of fibrous mat
35 can be seen in Figure 2 (magnification 80×, 1,000×, and 10,000×, respectively). The

average diameter of ethylcellulose fibre varied from 100 nanometer to 500 nanometer. The thickness of ethylcellulose mat was approximately 500 micrometer.

Preparation of ethylcellulose fibre structured oil

5 2 gram inES48 (triglyceride hardstock) was melted in 17.6 g sunflower oil used as lipid phase. 0.6 gram of ethylcellulose mat was torn into small pieces first, and then dispersed into oil phase under homogenization at the rate of $10,000 \text{ min}^{-1}$ for 5 min using an Ultra Turrax IKA T-25 digital (IKA Werke GmbH & Co. KG, Staufen, Germany). The concentration of fibre in the lipid phase was 3 % by weight. After totally dispersing the oil
10 phase containing ethylcellulose fibre continued homogenizing at the rate of $14,000 \text{ min}^{-1}$ for another 5 min. The resultant oil was put in the ethanol/water bath where temperature is -20°C , then fast stir using spatula to transfer heat as quick as possible. The cooling rate can vary from -15°C/min to -20°C/min . When temperature decreased to 5°C , the structured emulsion was stored at 5°C for 48 hr. Through this process ethylcellulose mat
15 was broken down into single and short ethylcellulose fibre dispersing evenly in oil (see Figure 3), and the fibre structured oil showed good rheological behavior and temperature response. Figure 4 shows the dispersion of ethylcellulose fibre in oil by means of phase contrast microscopy. Figure 5 shows rheological response (G' and G'' as function of temperature, at fixed strain of 0.1% and frequency of 1 Hz) profile of fibre structured oil
20 and of commercial margarine (as a comparison for proof of principle). The margarine was a commercially available Flora margarine (ex Unilever, bought in a local shop in Shanghai, China), containing 70% fat, of which 25% is saturated fats.

G' and G'' of the fibre structured oil (squares in Figure 5) both show a plateau first upon
25 increase of temperature from 5°C to about 20°C , and upon reaching higher temperatures the structured oil starts to loose its structure (G' and G'' decrease). The meltdown index of ethylcellulose structured oil was 2.9.

The curve of margarine (triangles in Figure 5) as comparison for proof of principle also shows a plateau until a temperature of 25°C , and then drop of G' and G'' upon higher
30 temperatures. The meltdown index in the present case is larger than 2.9.

The similarities between the curves of the structured oil and of the margarine is remarkable, both in absolute values of G' and G'' , as well in decrease of G' and G'' upon increase of temperature, leading to softening of the structured oil or margarine.

This experiment shows that the rheology profile of margarine can be mimicked by the oil
35 structured by the lipophilic fibres. And the amount of saturated fat is reduced from 25% to 17%, which makes the product healthier for consumption.

Viscosity profile of structured sunflower oil

Sunflower oil was structured as described above, at three concentrations of ethylcellulose fibre: 0.625%, 2% and 5% by weight. The viscosity η (eta, in Pa.s) as function of shear rate $\dot{\gamma}$ (gamma, in 1/s) of these compositions was determined at 25°C, and compared to pure sunflower oil and with margarine. The margarine was a commercially available Flora margarine (ex Unilever, bought in a local shop in Shanghai, China), containing 70% fat, of which 25% is saturated fats. Figure 6 shows the result; curve A: Margarine; B: sunflower oil; C: sunflower oil structured with 0.625% ethylcellulose fibre; D: sunflower oil structured with 2% ethylcellulose fibre; E: sunflower oil structured with 5% ethylcellulose fibre. This shows that the sunflower oil structured with 5% ethylcellulose fibre has the same viscosity profile as function of shear stress at 25°C as the commercially available margarine. This shows that by using the lipophilic fibres the amount of saturated fats in a margarine can be reduced, while keeping the same viscosity profile.

15

Example 3 - Preparation of electrospun zein fibre

The method for this example is the same as the example 2 except for the electrospun solution. In this case, the spinning solution consisted of 2 gram zein dissolved in 8 gram aqueous ethanol solution (80 wt%). In this experiment, 10 ml of yellow, clear solution was spun to form off-white mat. The results are shown by SEM pictures in Figure 7 (magnification 2,000 $\times$ and 5,000 $\times$, respectively). The fibres showed a flattened, ribbon-like structure. The average diameter of zein fibre varied from 500 nanometer to 2 micrometer, the thickness of the fibres was less than 1 micrometer. The thickness of the zein mat was approximately 800 micrometer.

25

Preparation of zein fibre structured oil

The process for this example is the same as the example 2 except for the electrospun fibre. In this case the electrospun fibre was zein as described above. The microscopy and rheology results are shown in Figure 8, Figure 9. The meltdown index of zein fibre structured oil was 2.2.

G' and G'' show a kind of plateau when the temperature is raised from 5 to about 20°C; and subsequently loses structure rapidly when the temperature is raised further. Slow melting as shown here is a positive property, as such melting behaviour is similar to liquid oils stabilised by solid fat, as in margarine.

35

Determination of average length of the fibres after fragmentation

1g Zein fibre mat was dispersed in 9g sunflower oil under homogenisation (using an Ultra Turrax IKA T-25 digital) at the rate of $10,000 \text{ min}^{-1}$ for 5 min, then continue homogenising at $22,000 \text{ min}^{-1}$ for another 5 min. The average length of the fibres after fragmentation was also determined using the method described herein before. The number average length of the fibres was 47.6 micrometer. Distribution: 53.8% of the fibres was in the range of 20-50 micrometer.

Viscosity profile of structured sunflower oil

10 Sunflower oil was structured as described above, at three concentrations of zein fibre: 2%, 5%, and 10% by weight. The viscosity η (eta, in Pa.s) as function of shear rate $\dot{\gamma}$ (gamma, in $1/\text{s}$) of these compositions was determined at 25°C , and compared to pure sunflower oil and compared to margarine. The margarine was a commercially available Flora margarine (ex Unilever, bought in a local shop in Shanghai, China), containing 70% fat, of which
15 25% is saturated fats. Figure 10 shows the result; Curve A: Margarine; B: sunflower oil; C: sunflower oil structured with 2% zein fibre; D: sunflower oil structured with 5% zein fibre; E: sunflower oil structured with 10% zein fibre.

This shows that the viscosity of sunflower oil structured with zein fibre can be modified by
20 modifying the concentration of zein fibre. When also the results from example 2 are taken into account, it shows that by choosing the type and concentration of fibre the rheology behaviour can be tuned.

25 Example 4 - Preparation of electrospun zein-triglyceride composite fibre

The method for this example is the same as the examples 2 and 3 except for the electrospun solution. In this case, the spinning solution consisted of 1 gram zein, 0.05 gram inES48 dissolved in 9 gram aqueous ethanol solution (90 wt%) at 53°C . In this experiment, 10 ml of solution was spun to form off-white mat. The results are shown by
30 means of SEM pictures in Figure 11 (magnification $100\times$, $10,000\times$). The diameter of zein-inES48 composite fibre varied from 500 nanometer to 2 micrometer. The thickness of Zein-inES48 composite fibre mat was approximately 500 micrometer.

Preparation of zein-inES48 composite fibre structured oil

35 The process for this example is similar as in example 2 except for the composition of oil. In this case the structured oil consisted of sunflower oil, containing 5% inES48 triglyceride,

and either 5% zein-inES48 fibre as structurant, or 5% zein fibre as structurant. The microscopy, rheology results (G' and G'' as function of temperature) of structured oil are shown in Figure 12, and Figure 13.

- 5 Meltdown index of zein-inES48 composite fibre structured oil (triangle) was 1.
Meltdown index of zein fibre structured oil (square) is 0.6.

This show that meltdown behavior can be effectively improved by addition of triglyceride (inES48 here) into zein fibre. The absolute value of G' and G'' of the oil stuctured by zein
10 fibre, was higher than that structured by zein-inES48 fibre.

Example 5 - Preparation of ethylcellulose-zein composite fibre

Preparation of ethylcellulose-zein composite fibres with two syringes:

- 15 1. Ethylcellulose was dissolved in 90 wt% ethanol/water to prepare 10% ethylcellulose solution.
2. Zein was dissolved in 80 wt% ethanol/water to prepare 23% zein solution.
3. Ethylcellulose and zein solution were loaded into two 10 ml syringes, respectively.
4. The syringes were fixed onto one pump and the two solutions were mixed in a
20 three valve connector at a volume ratio of 1 to 1, then the mixture was spun from one spinneret.
5. The electrospinning parameters were: 1) voltage: 18-20 kV; 2) distance from the tip of needle to collector: 10 cm; 3) flowing rate: 1.25 ml/hr.
6. Ethylcellulose/zein composite fibres were collected on the rotary copper mesh.
- 25 7. The fibres were separated from the copper mesh and torn into small pieces.

A SEM image of the composite fibre is shown in Figure 14. The diameter of ethylcellulose-zein composite fibre varied from 500 nanometer to 3 micrometer.

30 *Preparation of ethylcellulose-zein composite fibre structured oil*

The procedure applied for this example is the same as in example 3. The concentration of ethylcellulose-zein composite fibre was 5% by weight, and the composition also contained 5% by weight of inES48 triglyceride, in addition to the fibre and the sunflower oil.

- 35 Figure 15 shows the rheology profile of oil structured with the ethylcellulose-zein fibre (G' and G'' (in Pa) versus strain γ (gamma in %), at temperature 25°C. The rheology profile

was compared to the rheology profile of margarine. The margarine was a commercially available Flora margarine (ex Unilever, bought in a local shop in Shanghai, China), containing 70% fat, of which 25% is saturated fats. This graph shows that the rheology profile of oil structured with ethylcellulose-zein composite fibres is comparable with
 5 margarine. An oil phase structured by lipophilic fibres can be designed such that the viscosity profile of margarine is effectively mimicked.

Example 6 - Preparation of composite fibre structured emulsions

10 Three structured water-in-oil emulsion were prepared according to the following method. Three types of fibre, zein-inES48 composite fibre (from example 4), ethylcellulose-zein composite fibre (from example 5), and zein fibre (from example 3) were used in this experiment. 0.54 gram of each fibre were torn into small pieces and dispersed in sunflower oil, then fragmented using Ultra Turrax at 14,000 rpm. It can be defined as fibre
 15 slurry. 1.8 gram inES48 (triglyceride hardstock) was melted in 10 gram sunflower oil, using as oil phase. Then 0.3 gram PGPR and 12 gram water were added into the oil phase and homogenised using Ultra Turrax at 1,000 rpm, defined as pre-emulsion. Then each of the three fibre slurry was added into pre-emulsion using Ultra Turrax at 1, 000 rpm and then continued homogenizing with Ultra Turrax at 14,000 rpm for 5 min. The resultant
 20 emulsions were put in an ethanol/water bath at a temperature of -20°C, then fast stirred using spatula to transfer heat as quick as possible. The cooling rate varied from 15°C/min to 20°C/min. When the temperature decreased to 5°C, the structured oil was stored at 5°C for 48 hr.

25 The resultant emulsions had the following compositions:

Table 1 Compositions of emulsions

	A	B	C
zein-inES48 composite fibre (from example 4)	1.8%		
ethylcellulose-zein composite fibre (from example 5)		1.8%	
zein fibre (from example 3)			1.8%
inES48	5.9%	5.9%	5.9%
sunflower oil	51.7%	51.7%	51.7%
PGPR	1.0%	1.0%	1.0%
water	39.6%	39.6%	39.6%

A photograph of emulsion A is shown in Figure 16, left a visual image of the structured emulsion, and right a light microscopy image of the structured emulsion. The size of the dispersed water droplets was less than 5 micrometer.

- 5 The rheology profile (G' and G'' (in Pa)) was determined as function of temperature (at fixed strain of 0.1% and frequency of 1 Hz), see Figure 17.

The meltdown index of the emulsion A was: 2.8.

The meltdown index of the emulsion B was: 1.2.

The meltdown index of the emulsion C was: 2.3.

10

This shows that in this system, at the indicated concentrations of the fibres, the system containing zein-inES48 fibres has a higher meltdown index than the systems containing zein fibres and ethylcellulose-zein fibres. It also shows that the meltdown index of all emulsions is larger than 1, meaning that this is a good result perse. It shows that by

- 15 structuring the oil phase by the fibres, favourable rheology properties of an emulsion can be obtained, and this rheology can be tuned by the type of fibres and the concentration of the fibres. Food products and personal care products can be designed which show good melting behaviour, meaning not too rapid and not too slow melting. This behaviour can be controlled by modification of the composition of the fibres.

20

Example 7 - Preparation of emulsions structured with ethylcellulose fibre

Droplet Size and Droplet Size Distribution

In the context of the present invention, the average particle or droplet diameter is

- 25 generally expressed as the $d_{3,3}$ value, which is the volume weighted geometric mean particle or droplet diameter. The normal terminology for nuclear magnetic resonance (NMR) is used to measure the parameters $d_{3,3}$ and sigma (or alternatively $\exp(\sigma)$) of a log-normal oil droplet size distribution. Sigma is the standard deviation of the logarithmic of the droplet diameter $d_{3,3}$.

30

Droplet size and droplet size distribution are determined using standardised NMR equipment. A Bruker magnet with a field of 0.47 Tesla (20 MHz proton frequency) with an air gap of 25 mm is used (NMR Spectrometer Bruker Minispec MQ20 Grad, ex Bruker Optik GmbH, Germany) The NMR signal (echo height) of the protons of the water in a

35 water -in-oil emulsion are measured using a sequence of 4 radio frequency pulses in the presence (echo height E) and absence (echo height E^*) of two magnetic field gradient

pulses as a function of the gradient power. The oil protons are suppressed in the first part of the sequence by a relaxation filter. The ratio ($R=E/E^*$) reflects the extent of restriction of the translational mobility of the water molecules in the water droplets and thereby is a measure of the water droplet size. By a mathematical procedure (which uses the log-normal droplet size distribution) the parameters of the water droplet size distribution $d_{3,3}$ (volume weighted geometric mean diameter) and sigma or $\exp(\sigma)$ (measures for distribution width) are calculated.

Hardness and spreadability

- 10 Stevens values give an indication about the hardness (also called firmness) of a product. The Stevens value is determined according to the following protocol.

Freshly prepared products are stabilized at 5 degrees Celsius. The hardness of the product is measured with a Stevens penetrometer (Brookfield LFRA Texture Analyser
15 (LFRA 1500), ex Brookfield Engineering Labs, UK) equipped with a stainless steel probe with a diameter of 6.35 mm and operated in "normal" mode. The probe is pushed into the product at a speed of 2 mm/s, a trigger force of 5 gram from a distance of 10 mm. The force required is read from the digital display and is expressed in grams.

- 20 Three water-in-oil emulsion spreads were prepared using lab-scale margarine processing line (microvotator):
- (i) a medium-fat spread (variant with relatively low content of saturated fats), this can be considered to be a standard spread, a benchmark for good quality.
 - (ii) a reduced saturated fat spread, and
 - 25 (iii) a reduced saturated fat spread, which is structured with ethylcellulose fibre, according to the invention.

The composition of these margarines is listed in the following table:

Table 2 *Composition of water-in-oil emulsion spreads produced.*

<i>Description</i>	<i>Spread i)</i>	<i>Spread ii)</i>	<i>Spread iii)</i>
Linseed/Sunflower oil mixture 24 wt%/ 76wt%	52.02%	56.75%	55.40%
inES48 hardstock (as in example 6)	7.57%	2.84%	2.99%
Ethylcellulose fibre (as from example 2)	0.00%	0.00%	1.20%
Emulsifier Dimodan HP (ex Danisco)	0.27%	0.27%	0.27%
Emulsifier Lecithin Sunlec Z	0.07%	0.07%	0.07%
Colourant beta-carotene 0.4%	0.08%	0.08%	0.08%
Water	40.00%	40.00%	40.00%
	100.00%	100.00%	100.00%

Spreads ii) and iii) have 30% less saturated fat than Spread i).

- 5 Ethylcellulose was introduced as an ethylcellulose fibre premix, prepared as follows: 36 gram of fibre was torn into small pieces and dispersed at ambient temperature in 414 gram oil using handy blender Philips HR1372 at highest speed, then fragmented using Ultra Turrax at 14,000 rpm.
- 10 All compositions were processed as follows:
Fat phase was made by heating the oil to 65°C and adding the inES48, emulsifier, lecithin, beta-carotene and ethylcellulose fibre premix. Water at 80°C was added and stirring continued to create a coarse pre-emulsion using Ultra Turrax at 10,000 rpm at 65°C.
- 15 The pre-emulsion was processed over an AAAC votator setup. A-units are surface scraped heat exchangers and C-units are pin-stirred crystalliser units.

Exit temperatures of the A-units were set at 18, 10 and 2°C. Stirring speed of the 75 mL C-unit was set at 100 rpm. The product exit temperature was around 5°C. Throughput was
20 set at 5 kg/h.

Plastic tubs were filled with approximately 150 grams of spread and stored at 5°C prior to further analysis. In total 3 kg was produced of each spread.

One day after production, hardness measured by Stevens texture analyser with 6.35 mm probe was: 12g for Spread ii), 21g for Spread iii) and 24g for Spread i) (benchmark).

After two weeks of storage, the hardness was: 52 g for Spread i) (benchmark), 30 for Spread iii) and 15 for Spread ii).

5

This shows that Spread iii) (containing ethylcellulose fibres) has much more firmness than Spread ii) at the same level of saturated fatty acids, whilst also a more consistent firmness over time compared with the benchmark Spread i).

- 10 Droplet size of spreads is an important parameter for microbiological safety and sensory perception. The $d_{3,3}$ describes the average droplet size, while the $\exp(\sigma)$ describes the width of the size distribution. In both cases, lower values are usually preferred. In the table, $d_{3,3}$ and $\exp(\sigma)$ for the freshly made samples, as well as samples after 2 weeks storage, are presented.

15

Table 3 Results on dispersed phase droplet size of water-in-oil emulsion spreads produced.

	$d_{3,3}$ (micrometer)	$\exp(\sigma)$ (-)
Fresh Spread i)	6.7	2.00
Fresh Spread iii)	8.6	2.11
Fresh Spread ii)	16.0	3.00
2 week Spread i)	8.4	2.50
2 week Spread iii)	10.9	2.31
2 week Spread ii)	24.2	2.20

- This shows that Spread iii) (with ethylcellulose fibre) had droplet sizes close to the
 20 benchmark Spread i) and was not sensitive to ageing, whilst Spread ii) had unacceptably large droplets and showed coarsening of water droplets upon ageing (which is a negative property).

- A temperature-cycling test was carried out under the following conditions: the three
 25 samples were stored at 30°C for 24 hours, then transferred to 5°C storage overnight. Hardness and droplet size after each cycling temperature were measured and listed in the following table.

Table 4 *Results on dispersed phase droplet size of water-in-oil emulsion spreads after temperature cycling test*

	<i>After 30°C</i>			<i>After 5°C</i>		
	<i>d_{3,3}</i> <i>(micrometer)</i>	<i>exp</i> <i>(sigma)</i> <i>(-)</i>	<i>hardness</i> <i>(g)</i>	<i>d_{3,3}</i> <i>(micrometer)</i>	<i>exp</i> <i>(sigma)</i> <i>(-)</i>	<i>hardness</i> <i>(g)</i>
Spread i)	14.9	0.78	<5	16.2	2.21	19
Spread iii)	13.0	0.72	<5	15.8	2.04	15
Spread ii)	30.9	0.78	<5	30.4	2.18	<5

This shows that Spread iii) (ethylcellulose fibre-containing spread) had droplet size and hardness close to the benchmark Spread i), while Spread ii) demonstrated substantial instability after cycling test. Furthermore, Spread ii) became liquid after 30°C storage; this process was irreversible, as the spread did not recover the texture after 5°C storage. This cycling test showed that Spread iii) (ethylcellulose fibre-containing spread) survived from very tough temperature variation and had visibly better thermal stability, which would be very important for sale in tropical areas with ambient product distribution.

CLAIMS

1. A composition comprising a non-aqueous liquid phase, wherein the liquid phase is structured by a lipophilic fibrous material comprising a polymer, wherein the fibrous material comprises one or more compounds chosen from the group of prolamins and lipophilic cellulose derivatives, and wherein the fibrous material has been prepared by a method involving spinning.
2. A composition according to claim 1, wherein the non-aqueous liquid phase is chosen from the group consisting of vegetable oil or fat, dairy oil or fat, fish oil or fat, mineral oil or mineral oil derivative, petrolatum or petrolatum derivative, silicon oil or silicon oil derivative.
3. A composition according to claim 1 or 2, wherein the composition is a water-in-oil emulsion, containing between 1% by weight and 99% by weight of non-aqueous liquid phase.
4. A composition according to claim 1 or 2, wherein the composition is an oil-in-water emulsion, containing between 1% by weight and 95% by weight of non-aqueous liquid phase.
5. A composition according to any of claims 1 to 4, wherein the concentration of fibrous material is between 0.01% and 50% by weight, based on the amount of non-aqueous liquid phase, preferably between 0.5% and 10% by weight, based on the amount of non-aqueous liquid phase.
6. A composition according to any of claims 1 to 5, wherein the three-phase contact angle between sunflower oil, and a film of the lipophilic cellulose derivative, and air is less than 70° at 20°C.
7. A composition according to any of claims 1 to 6, wherein the lipophilic cellulose derivative comprises ethylcellulose.
8. A composition according to any of claims 1 to 7, wherein the prolamin is chosen from the group of zein, gliadin, hordein, secalin, and avenin.

9. A composition according to any of claims 1 to 8, wherein the fibrous material comprises one or more lipid compounds.
10. A composition according to claim 9, wherein the lipid compound comprises lecithin, fatty acid, monoglyceride, diglyceride, triglyceride, phytosterol, phytostanol, phytosteryl-fatty acid ester, phytostanyl-fatty acid ester, wax, fatty alcohol, carotenoid, oil-soluble colourant, oil-soluble vitamin, oil soluble flavour, or oil soluble fragrance.
11. A composition according to any of claims 1 to 10, wherein the fibre has a length from 1 micrometer to 10 millimeter,
wherein the fibre has a diameter from 30 nanometer to 50 micrometer,
and wherein the aspect ratio of the fibre is larger than 10.
12. A composition according to any of claims 1 to 11, wherein the fibrous material has been prepared by a method involving electrospinning.
13. A method for production of a composition according to any of claims 1 to 12, comprising the steps:
- a) spinning of a fibrous material from a polymer, wherein the polymer is in liquid form during the spinning; and
 - b) dispersing the fibrous material obtained from step a) in a non-aqueous liquid; and
 - c) homogenising the mixture from step b), to fragment the fibrous material to an average length from 1 micrometer to 10 millimeter; and
 - d) bringing the mixture obtained from step c) into contact with one or more other ingredients of the composition.
14. A method according to claim 13, wherein in step a) the spinning process is an electrospinning process.

Figure 1

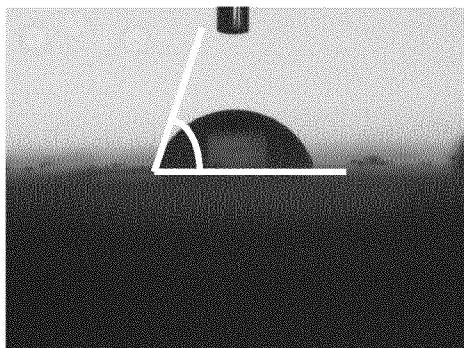


Figure 2

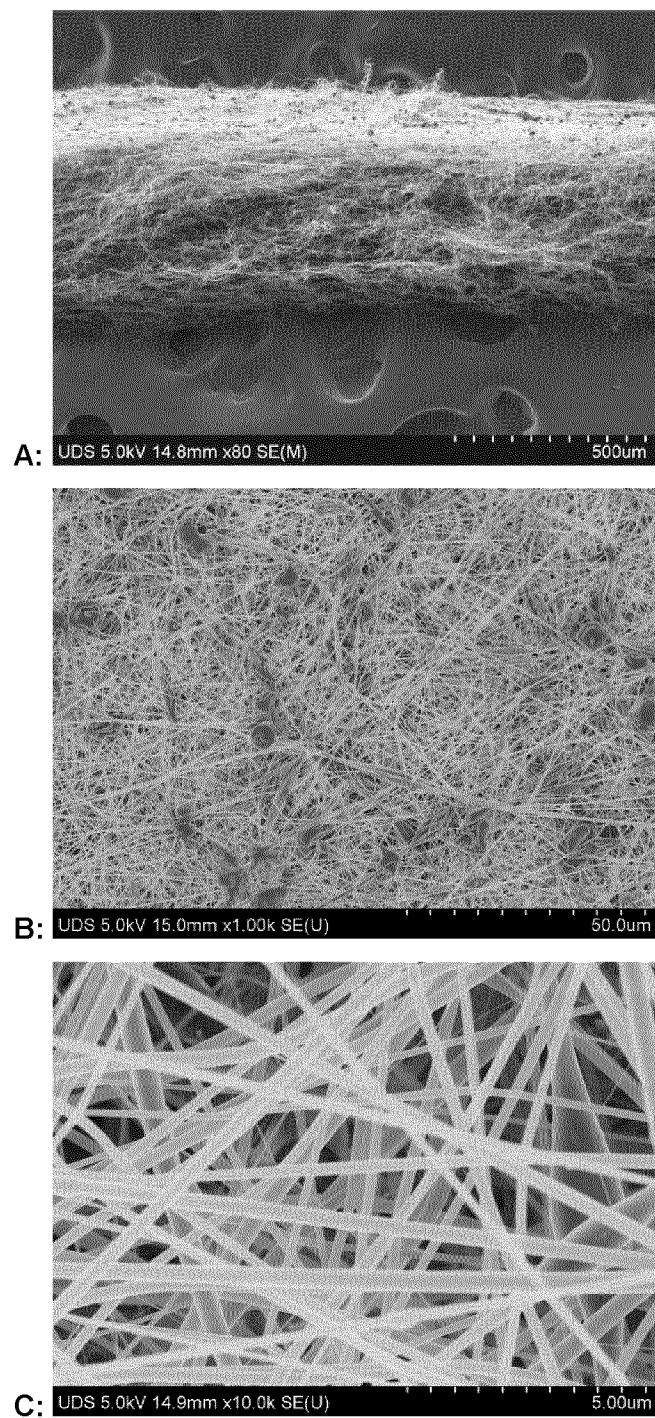


Figure 3

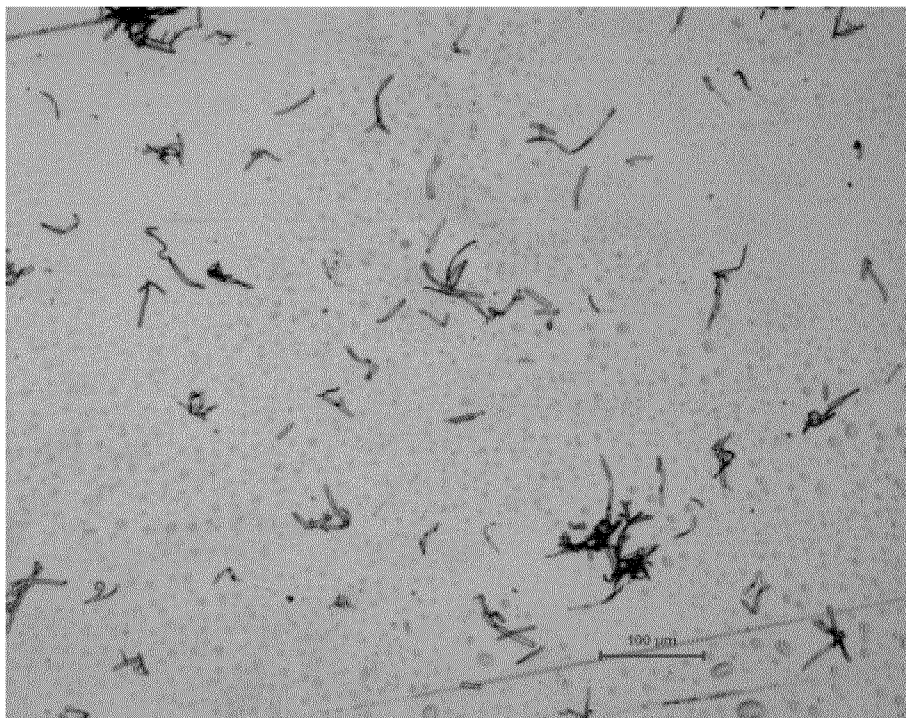


Figure 4

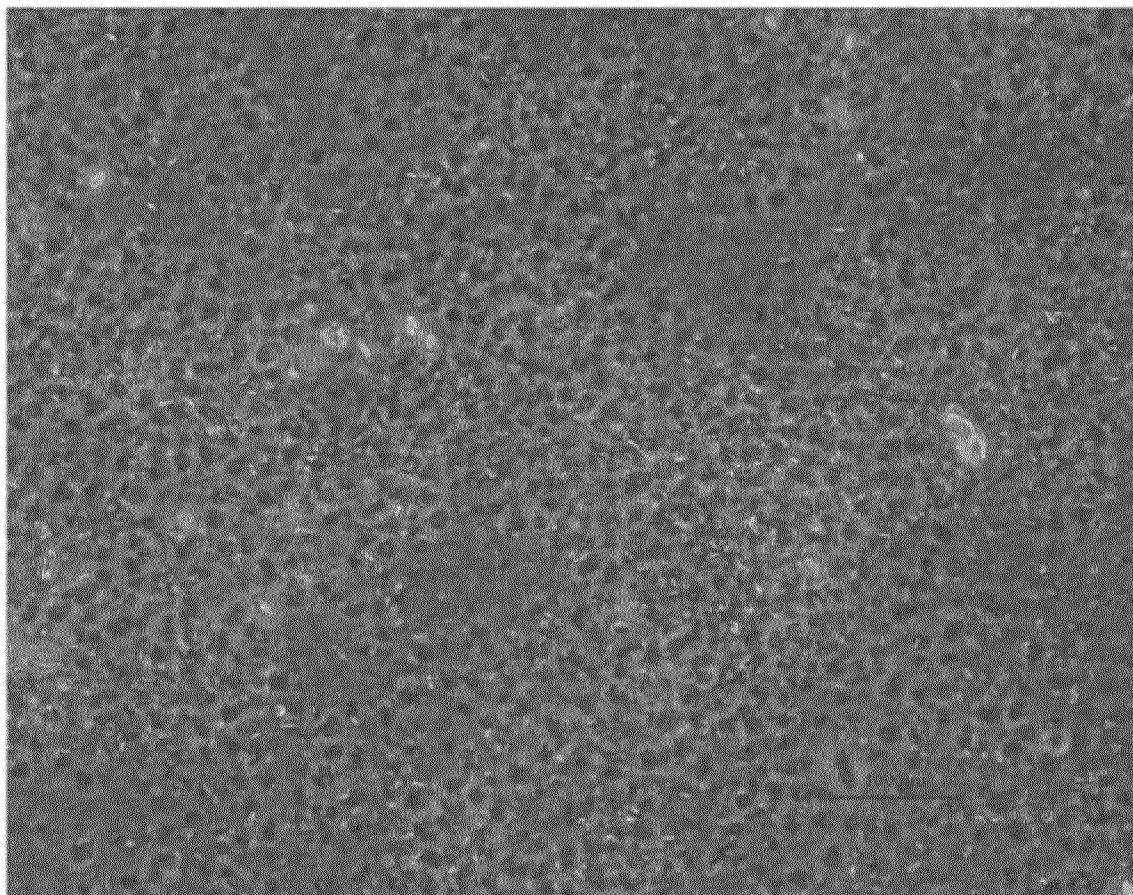


Figure 5

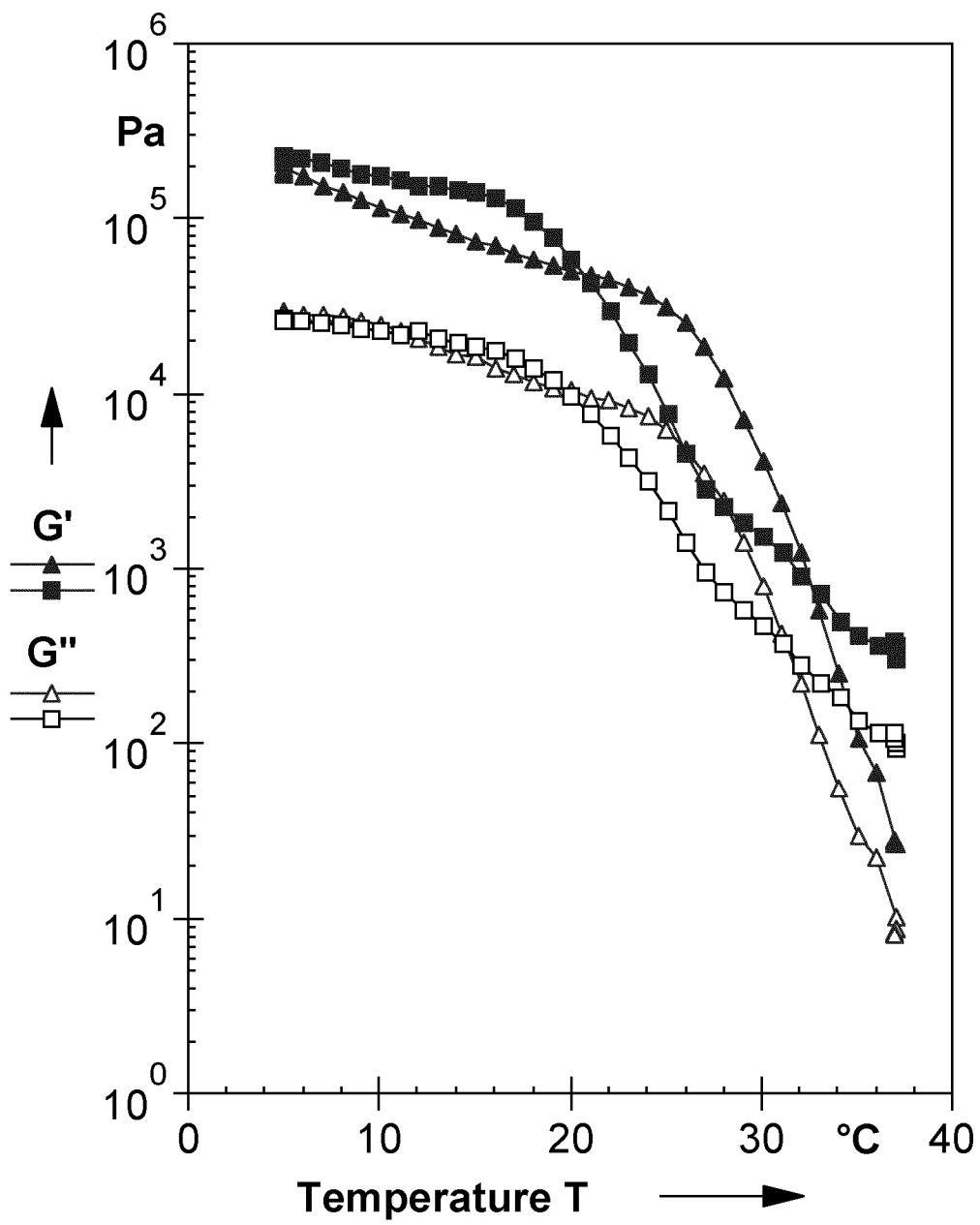


Figure 6

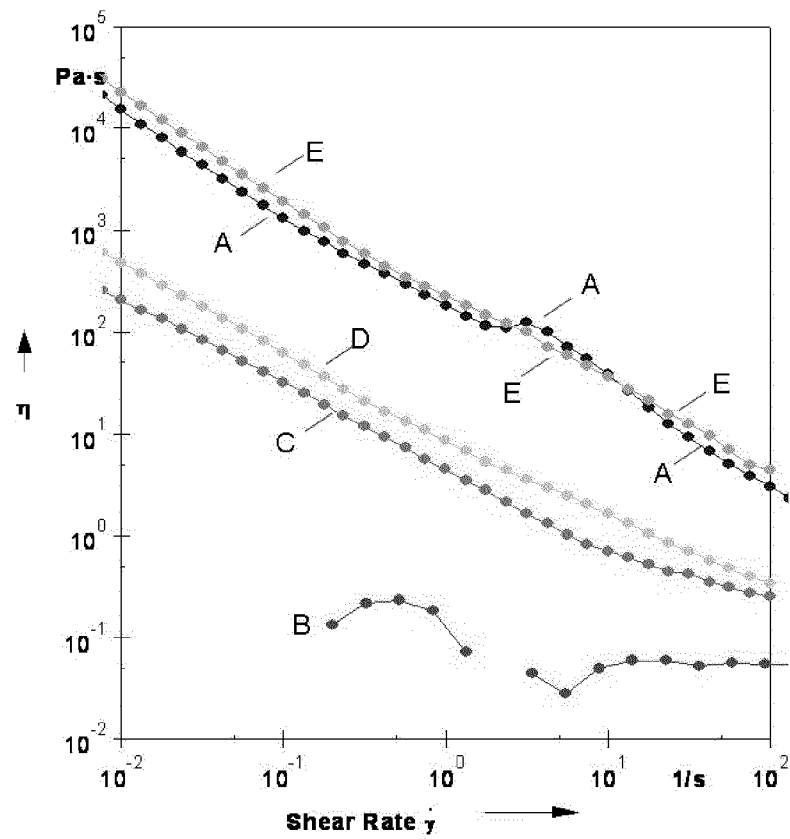


Figure 7

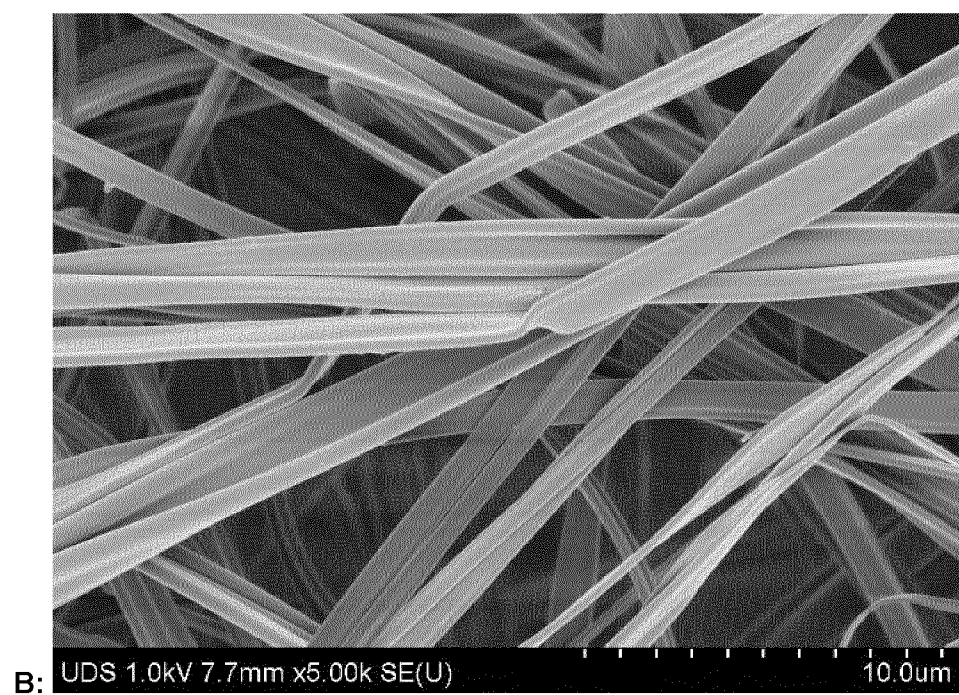
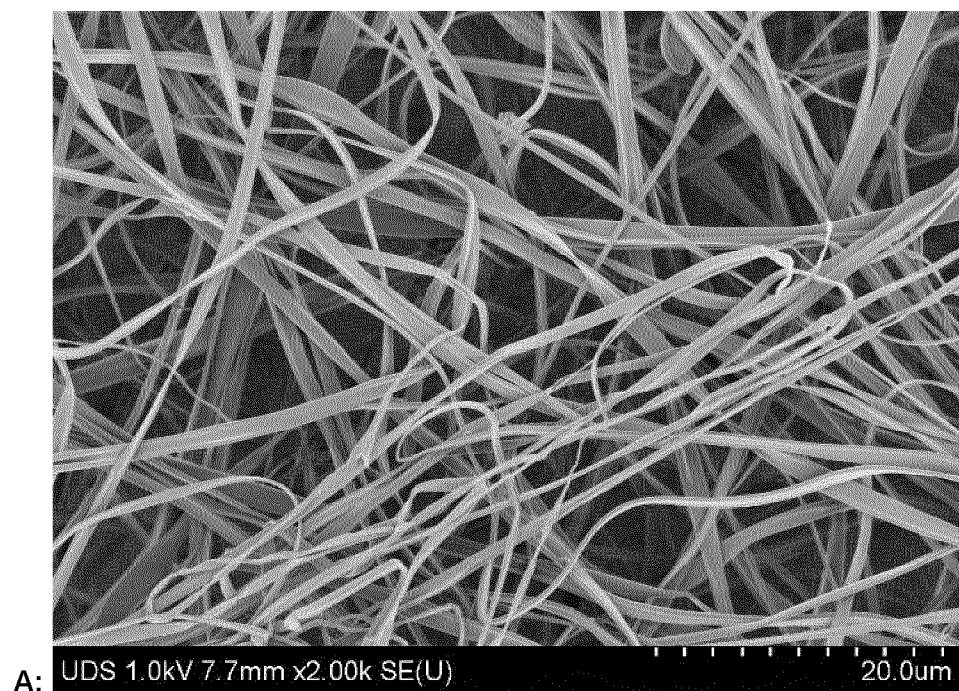


Figure 8

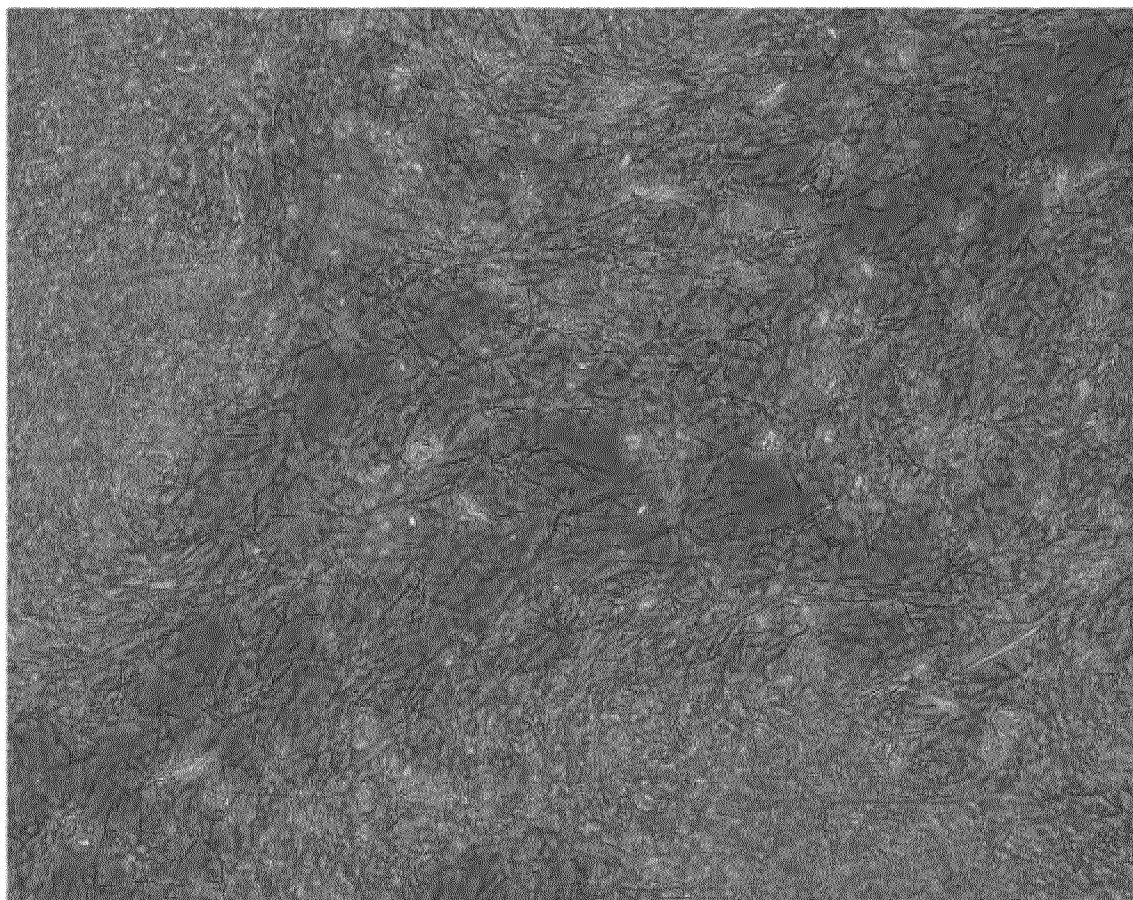


Figure 9

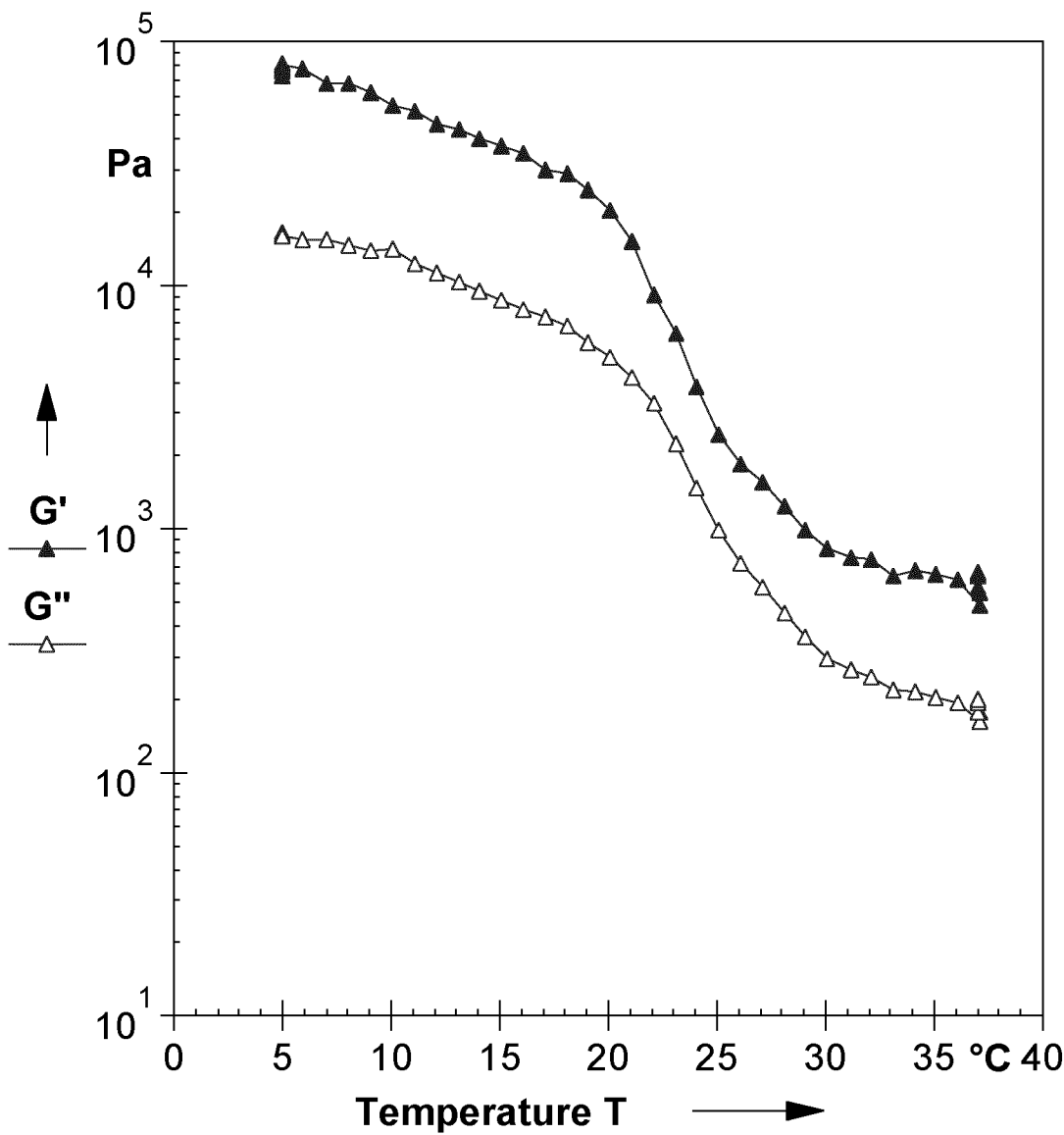


Figure 10

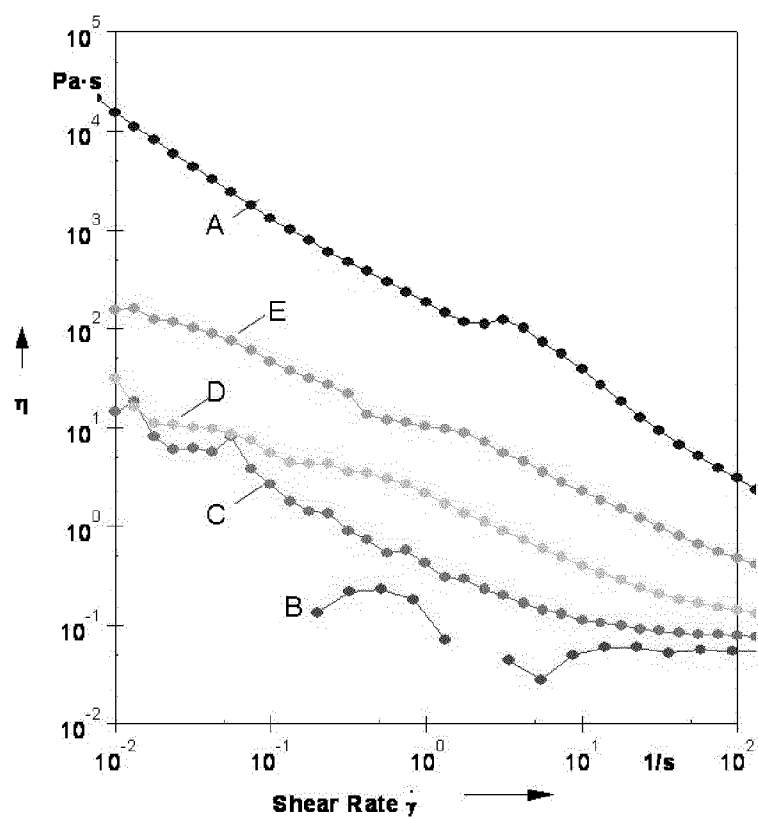


Figure 11

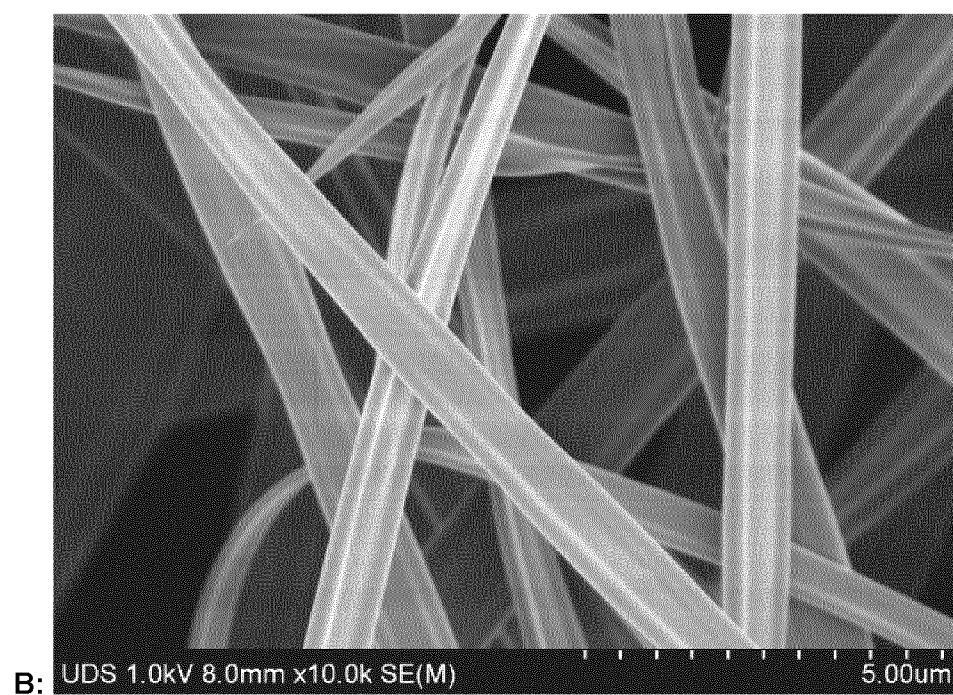
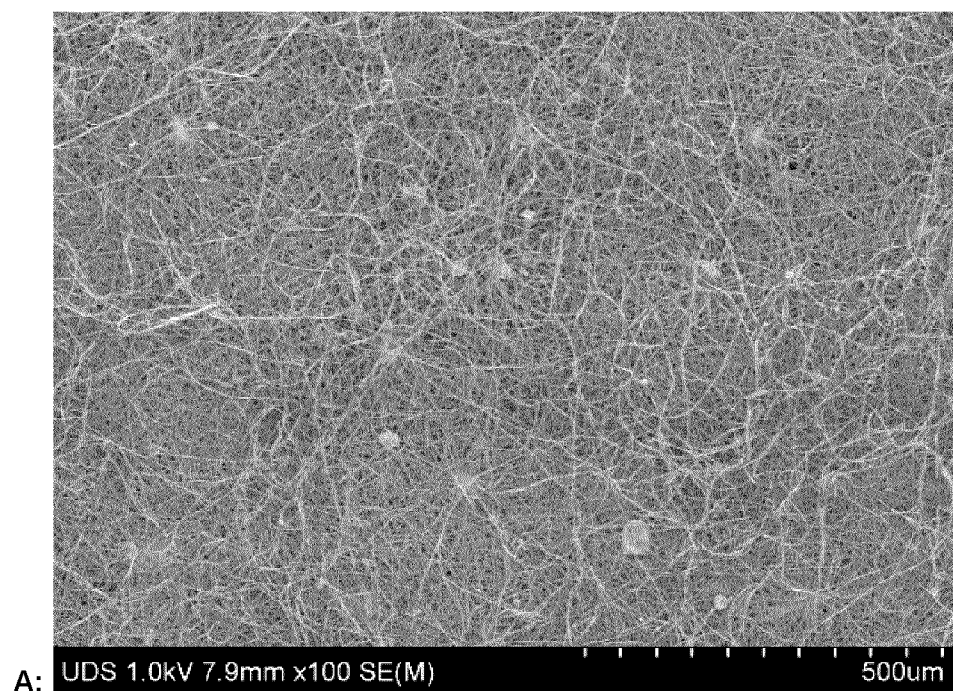


Figure 12

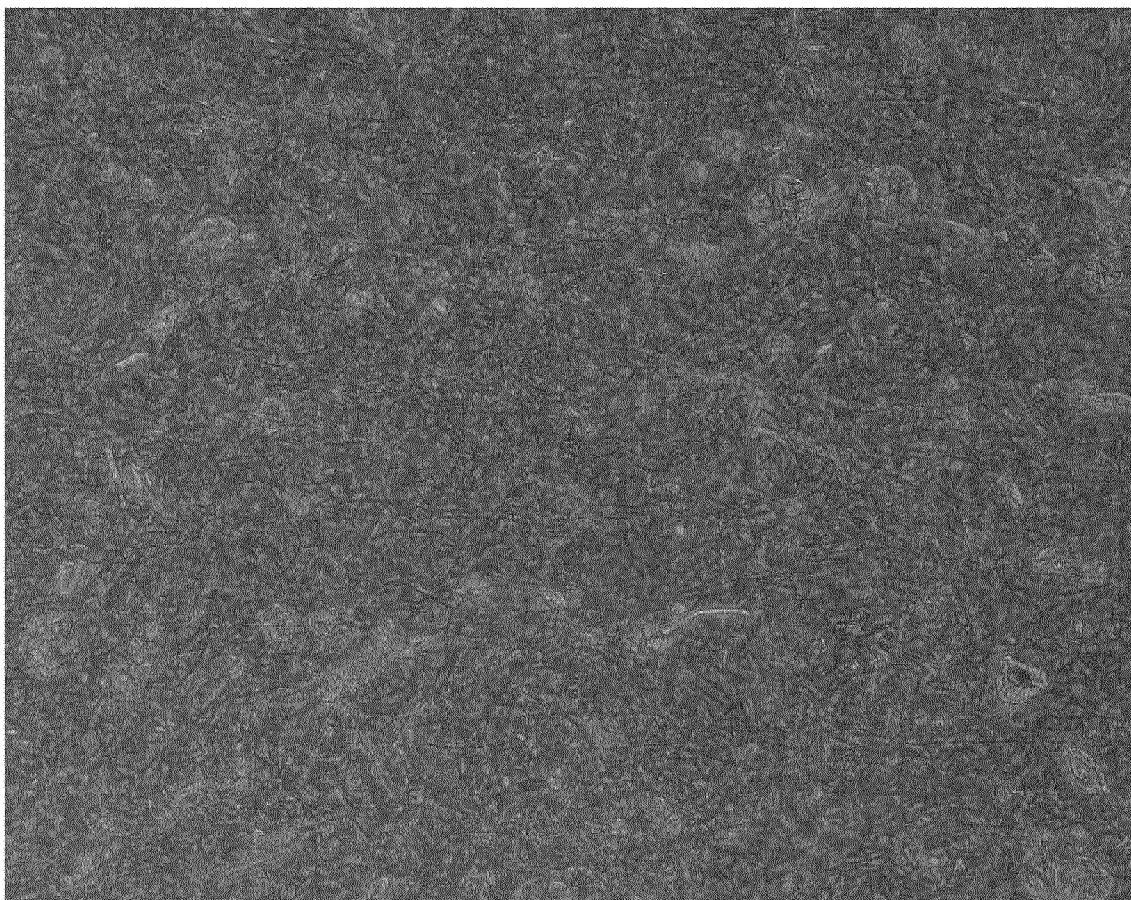


Figure 13

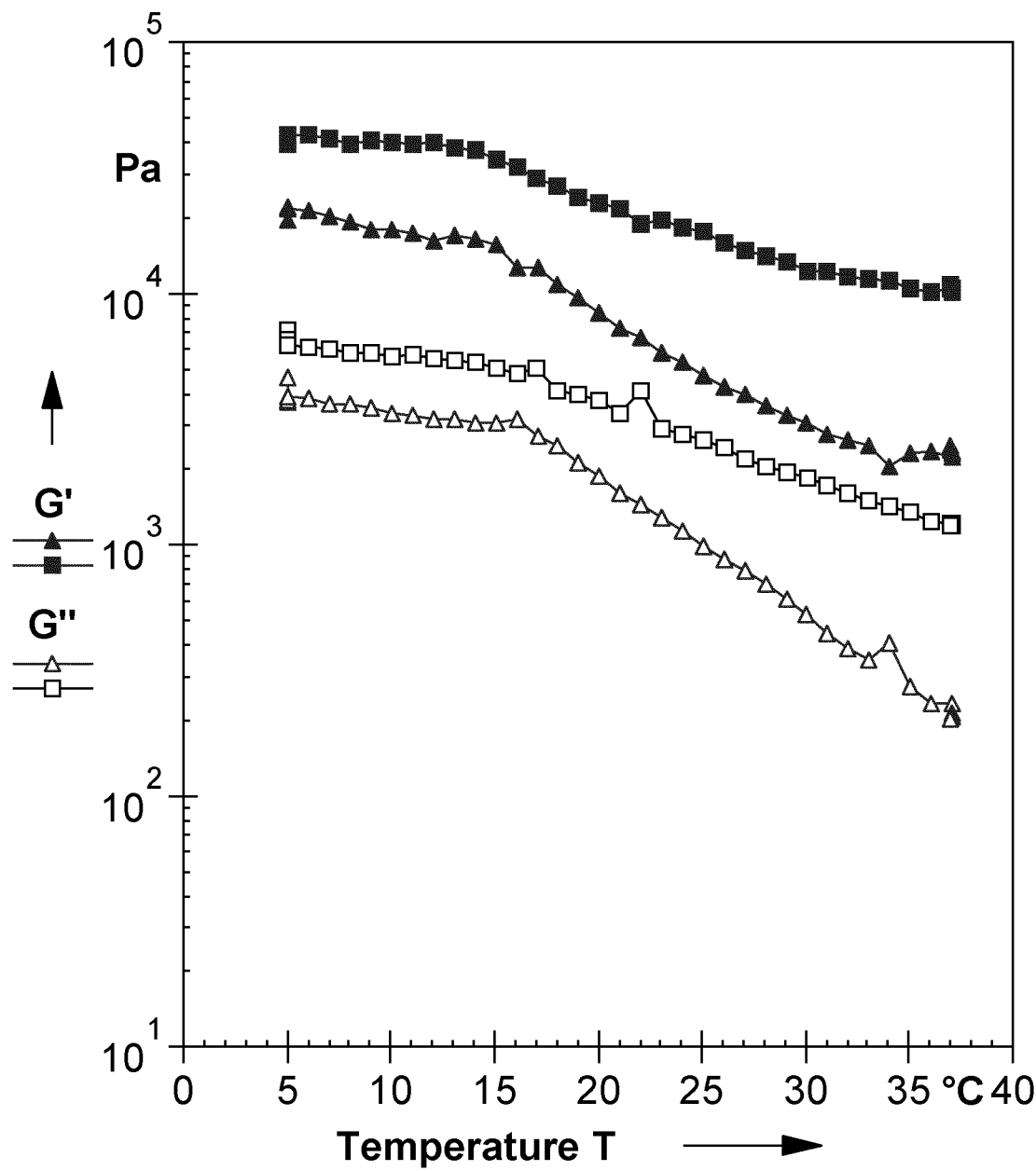


Figure 14

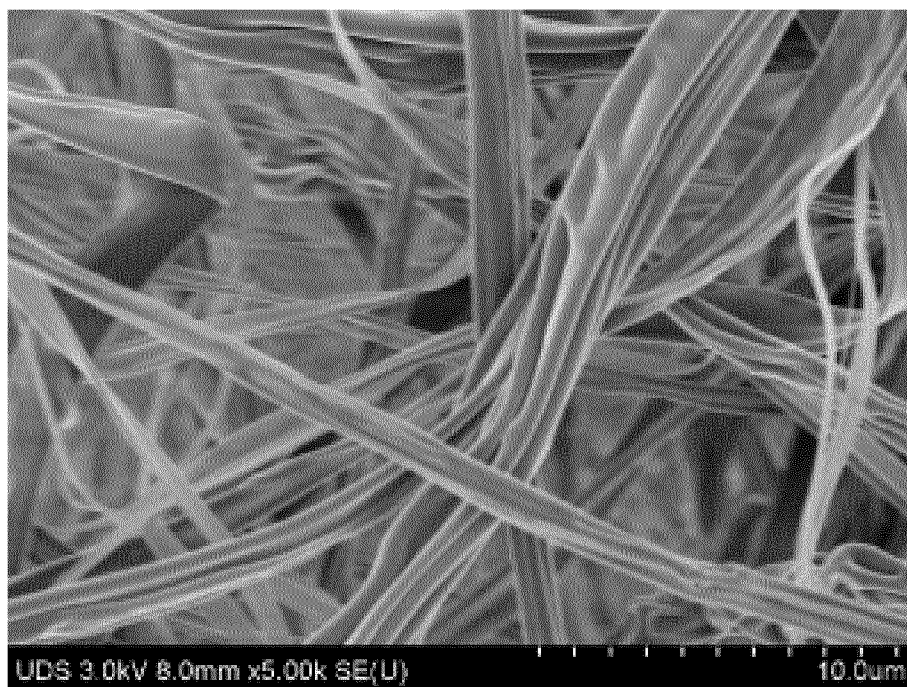


Figure 15

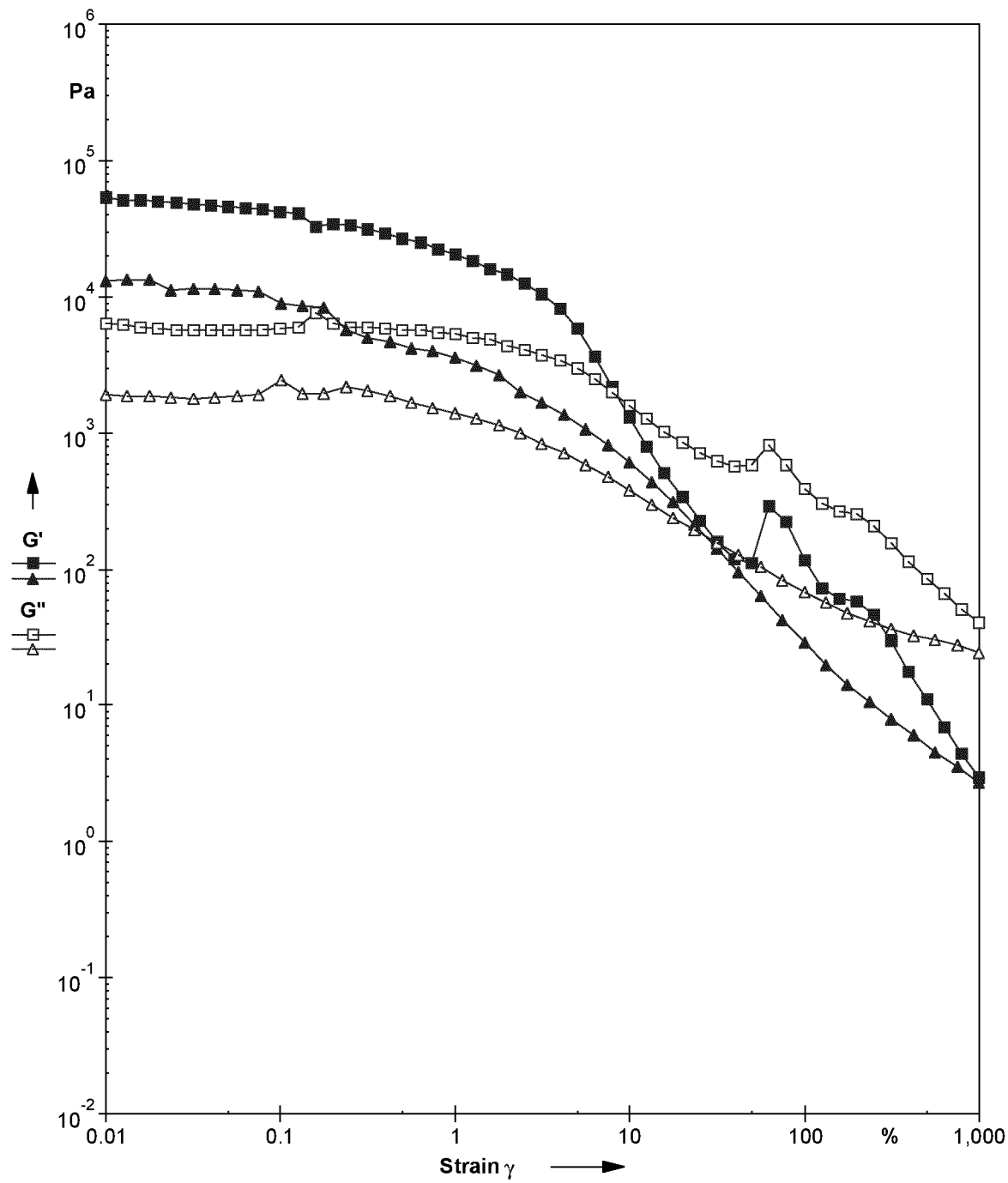


Figure 16

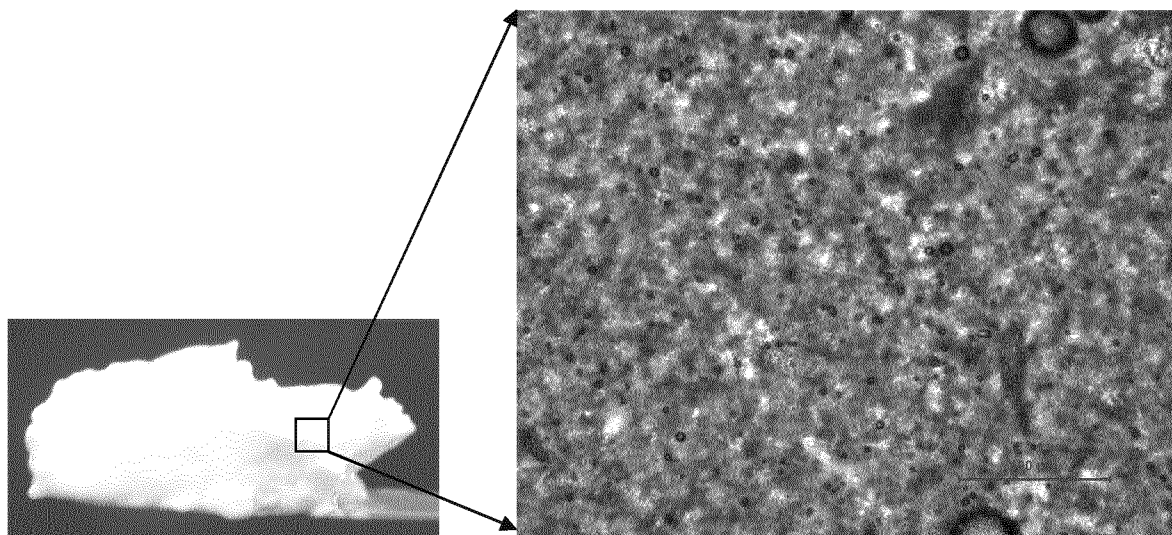
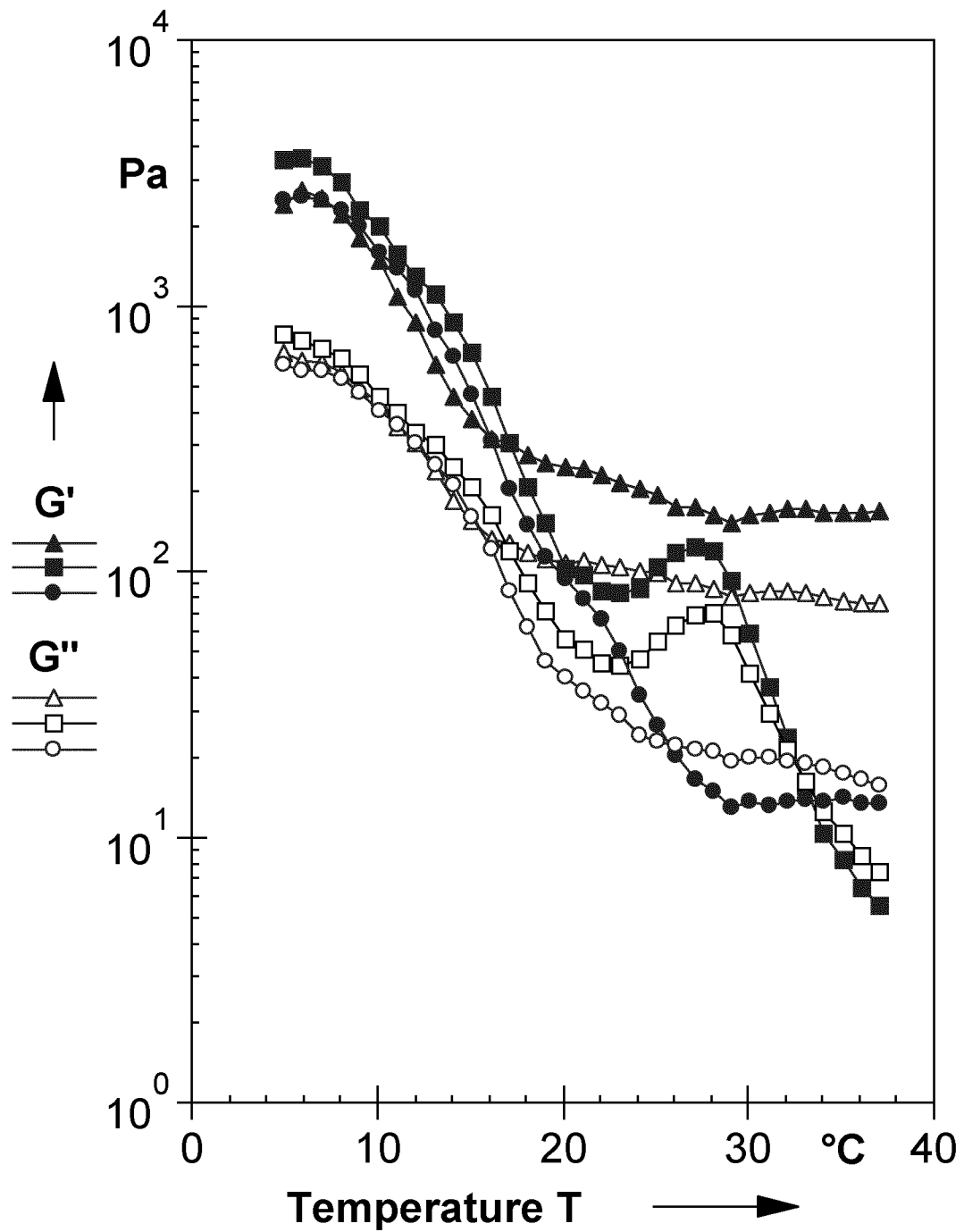


Figure 17



INTERNATIONAL SEARCH REPORT

International application No
PCT/EP2011/071264

A. CLASSIFICATION OF SUBJECT MATTER INV. A23L1/00 A23L1/0534 A23L1/10 A61K8/64 A61K8/73 A23D7/005 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) A23L A61K A23D 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 practical, search terms used) EPO-Internal, WPI Data																				
C. DOCUMENTS CONSIDERED TO BE RELEVANT <table border="1"> <thead> <tr> <th>Category*</th> <th>Citation of document, with indication, where appropriate, of the relevant passages</th> <th>Relevant to claim No.</th> </tr> </thead> <tbody> <tr> <td>X</td> <td>WO 2010/121492 A1 (UNILEVER NV [NL]; UNILEVER PLC [GB]; UNILEVER HINDUSTAN [IN]; CAO JIAN) 28 October 2010 (2010-10-28)</td> <td>1-5,7,9-12</td> </tr> <tr> <td>A</td> <td>page 6, line 25 - page 10, line 20; claims 1-15; examples 1-7</td> <td>13,14</td> </tr> <tr> <td>X</td> <td>EP 2 230 294 A1 (UNILEVER NV [NL]; UNILEVER PLC [GB]) 22 September 2010 (2010-09-22) cited in the application claims 1-15</td> <td>1,2,6</td> </tr> <tr> <td>X</td> <td>US 5 271 961 A (MATHIOWITZ EDITH [US] ET AL) 21 December 1993 (1993-12-21) claims 1-20</td> <td>1,8</td> </tr> <tr> <td></td> <td style="text-align: center;">----- -/-</td> <td></td> </tr> </tbody> </table>			Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.	X	WO 2010/121492 A1 (UNILEVER NV [NL]; UNILEVER PLC [GB]; UNILEVER HINDUSTAN [IN]; CAO JIAN) 28 October 2010 (2010-10-28)	1-5,7,9-12	A	page 6, line 25 - page 10, line 20; claims 1-15; examples 1-7	13,14	X	EP 2 230 294 A1 (UNILEVER NV [NL]; UNILEVER PLC [GB]) 22 September 2010 (2010-09-22) cited in the application claims 1-15	1,2,6	X	US 5 271 961 A (MATHIOWITZ EDITH [US] ET AL) 21 December 1993 (1993-12-21) claims 1-20	1,8		----- -/-	
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A	page 6, line 25 - page 10, line 20; claims 1-15; examples 1-7	13,14																		
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X	US 5 271 961 A (MATHIOWITZ EDITH [US] ET AL) 21 December 1993 (1993-12-21) claims 1-20	1,8																		
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☒ Further documents are listed in the continuation of Box C. ☒ See patent family annex.																				
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Date of the actual completion of the international search 14 March 2012		Date of mailing of the international search report 23/03/2012																		
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