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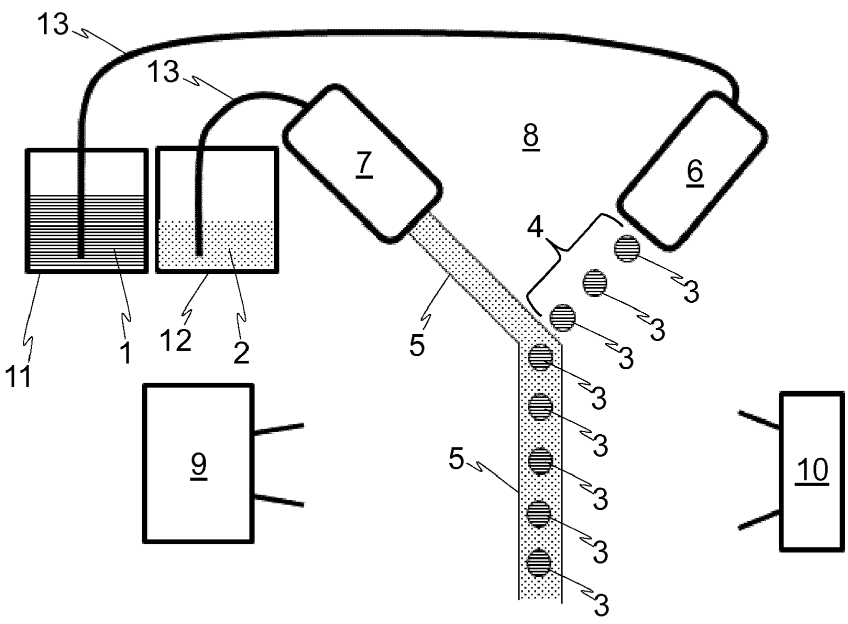
(54) **METHOD FOR PRODUCING A REGULAR ARRANGEMENT OF DROPLETS OF A FIRST LIQUID IN A CONTINUOUS JET OF A SECOND LIQUID**

(57) Method for producing a regular arrangement of droplets (3) of at least one first liquid (1) in a continuous jet (5) of a second liquid (2), wherein the first liquid (1) and the second liquid (2) are immiscible and chosen such that a surface tension (σ_d) of the first liquid (1) is greater than the sum of a surface tension (σ_j) of the second liquid (2) and an interfacial tension (σ_{dj}) between the first liquid (1) and the second liquid (2), wherein at least one regular stream (4) of the droplets (3) is produced using at least one first nozzle (6) and the continuous jet (5) is produced using a second nozzle (7), wherein the nozzles (6, 7) are adjusted such that the continuous jet (5) and the at least one regular stream (4) of droplets (3) are in a common plane (8), collide and the continuous jet (5) of the second liquid (2) encapsulates the regular arrangement of the droplets (3) of the at least one first liquid (1) after the collision.

Fig. 1



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Description

FIELD OF THE INVENTION

[0001] The present invention relates to a method for producing a regular arrangement of droplets of at least one first liquid in a continuous jet of a second liquid, which can be further used for producing a fibre.

[0002] Furthermore, the present invention relates to a cylindrical fibre.

STATE OF THE ART

[0003] Drops can be encapsulated in a liquid continuous phase in the form of emulsions, which is well-known. A regular arrangement of drops, particularly a regular spatial arrangement of monodisperse drops, is, however, hardly obtainable. Existing approaches try to achieve that goal by means of guiding the continuous phase through pipes or channels, which prevents, however, the formation of fibres.

[0004] Although core-shell nanofibres can be produced using electrospinning, it is not possible to produce a regular arrangement of inclusion and encapsulated objects, respectively, see e.g. Md. Fazley Elahi et al., "Core-shell Fibers for Biomedical Applications-A Review", Journal of Bioengineering & Biomedical Sciences 2013, Volume 3, Issue 1, 1000121. Moreover, fibres with encapsulated objects are usually not cylindrical, but exhibit constrictions between the encapsulated objects, which can be disadvantageous for some applications.

OBJECTIVE OF THE INVENTION

[0005] It is therefore an objective of the present invention to provide a method for producing a regular arrangement of droplets in a continuous second phase, which overcomes the above mentioned problems. Particularly, the method should allow for the subsequent production of fibres, more particularly of cylindrical fibres, with regularly arranged encapsulated objects.

SUMMARY OF THE INVENTION

[0006] In order to solve the above-mentioned problem, according to the invention a method for producing a regular arrangement of droplets of at least one first liquid in a continuous jet of a second liquid is provided, wherein the first liquid and the second liquid are immiscible and chosen such that a surface tension (σ_d) of the first liquid is greater than the sum of a surface tension (σ_j) of the second liquid and an interfacial tension (σ_{dj}) between the first liquid and the second liquid, wherein at least one regular stream of the droplets is produced using at least one first nozzle and the continuous jet is produced using a second nozzle, wherein the nozzles are adjusted such that the continuous jet and the at least one regular stream of droplets are in a common plane, collide and the continuous jet of the second liquid encapsulates the regular arrangement of the droplets of the at least one first liquid after the collision.

[0007] This means that a well-controllable regular drop stream is combined with a well-controllable regular continuous jet and the relative values of the surface and interfacial tensions lead to a total encapsulation of the drop liquid (the first liquid) in portions of the jet liquid (the second liquid). Due to the usage of a one continuous jet and one droplet stream the collision process enables the perfectly controlled formation of liquid structures (i.e. of droplets in a jet) which can be of interest for materials sciences as well as for biological and pharmaceutical applications.

[0008] Typically, both liquids have densities in the range from 700 kg/m^3 to 2000 kg/m^3 and viscosities in the range from 0,5 mPa s to 5 Pa s. The choice of the surface tensions and interfacial tension ($\sigma_d > \sigma_j + \sigma_{dj}$) ensures the thermodynamic stability of the full encapsulation of the drops by the jet. In other words, due to surface tensions of the two liquids and the interfacial tension between them the first liquid can be totally wetted by the second liquid, ensuring full encapsulation of the droplets in the jet.

[0009] For example, the above condition can be met employing silicon oils as second liquid, i.e. for the jet, and aqueous glycerol solutions for the first liquid, i.e. for the stream of droplets. Other suitable couples of liquids can be easily found. Moreover, adding surfactants in the aqueous phase, e.g. didodecyldimethylammonium bromide with an appropriate oil like isopropyl myristate, enables the exploration of cases where the encapsulating jet is aqueous. This means, however, that in practice the surface and interfacial tensions of the liquids can be routinely adjusted by means of surfactants, in order to fulfil the above-mentioned relation, making possible an almost arbitrary choice of the liquids.

[0010] "Regular stream of droplets" means in particular that the spatial arrangement of the droplets within the stream is regular, e.g. due to a periodic spatial arrangement.

[0011] The liquids are supplied to the nozzles by well-known means and liquid supplying system, respectively, e.g. by means of pressurised tanks, with each liquid being stored in an own pressurised tank. Naturally, also any kind of pump can be used instead of a pressurised tank, e.g. a peristaltic pump, a syringe pump, etc.

[0012] The adjustment of the nozzles is done not only with respect to the orientation and position of the nozzles, but in general with respect to collision parameters, like velocities and (spatial) periodicities of the continuous jet and the stream of droplets, respectively. Thereby, the typical distance between nozzle orifices is in the range from 1 mm to 20 cm, particularly from 5 mm to 10 cm, and the typical distances between a point or region of collision and the nozzle orifices is in the range from 1 mm to 20 cm, particularly from 1 cm to 10 cm.

[0013] That the continuous jet and the stream of droplets are in the same (common) plane particularly means that a trajectory of the continuous jet and a trajectory of the droplets and the stream of droplets, respectively, are in said plane.

[0014] Note that it is in principal possible to have several streams of droplets.

[0015] Preferably, it is possible to have several stream of droplets of several first liquids, i.e. the droplets of each of these streams are of a different first liquid (instead of the same first liquid), with each first liquid being immiscible with the second liquid and fulfilling the above-mentioned condition ($\sigma_d > \sigma_j + \sigma_{dj}$).

[0016] Each stream of droplets can be provided by a corresponding first nozzle.

[0017] Each first liquid can be supplied to each first nozzle by means of, e.g. an own pressurised tank or an own pump.

[0018] Hence, a regular arrangement of droplets of the several first liquids in the continuous jet of the second liquid can be obtained after the collisions of the droplet streams with the continuous stream.

[0019] Preferably, the different streams of droplets - of the same first liquid or of several first liquids - collide with the continuous jet at different positions or in different regions.

[0020] Accordingly, in a preferred embodiment of the method according to the present invention, it is provided that several regular streams of droplets, preferably of several first liquids, are provided, wherein the several regular streams of the droplets are produced using several first nozzles.

[0021] Preferably, only the continuous jet (of the second liquid) encapsulating the regular arrangement of droplets (of the at least one first liquid) remains after the collision, i.e. downstream of the point or region where the collision(s) take(s) place. This can be fostered by tuning the angle under which the continuous jet and the stream of droplets collide. Correspondingly, in a preferred embodiment of the method according to the present invention, it is provided that the continuous jet and the at least one stream of droplets enclose an angle in the common plane, which angle is in the range from 1° to 170°, preferably from 5° to 90°.

[0022] In a preferred embodiment of the method according to the present invention, it is provided that a diameter of an orifice of the at least one first nozzle and a diameter of an orifice of the second nozzle are adjusted in the range from 10 μm to 1500 μm. This means that the nozzles can have variable orifices, wherein such nozzles are known in the art. The given range of orifice diameters guarantee a particularly reliable production of the continuous jet of the second liquid encapsulating the regular arrangement of the droplets of the first liquid in the course of the collision of the continuous jet and the stream of droplets.

[0023] Note that the droplet diameter can be precisely tuned by choosing the nozzle diameter, but typically is not equal to the latter. E.g. a nozzle diameter from 30 to 1300 μm can cause droplet diameters from 50 μm to 2700 μm. On the other hand, the nozzle diameter directly corresponds to the jet diameter, i.e. a nozzle diameter from 30 μm to 1500 μm causes identical jet diameters from 30 μm to 1500 μm.

[0024] Ideally, the jet diameter is chosen larger than the droplet diameter.

[0025] The choice of the diameters of the nozzle orifices directly affects flow rates of the liquids through the nozzles. For both liquids, typical flow rates are in the range from 5 μl/s to 10 ml/s, fostering the desired result of the continuous jet of the second liquid encapsulating the regular arrangement of the droplets of the first liquid.

[0026] Since the droplet diameter can be precisely tuned, as already mentioned above, it is possible to control the diameter of the droplets in the stream of droplets and consequently also the diameter of the droplets encapsulated in the continuous jet of the second liquid, i.e. of those droplets that form the encapsulated regular arrangement in the continuous jet. In this case "regular" does not exclusively relate to the spatial arrangement of the droplets, but also to their size.

[0027] Preferably, it is thus possible to produce the regular stream of droplets with the droplets being monodisperse. This leads to a regular arrangement of the droplets of the at least one first liquid encapsulated in the continuous jet of the second liquid, with the encapsulated droplets being monodisperse.

[0028] Accordingly, in a preferred embodiment of the method according to the present invention, it is provided that the at least one regular stream of the droplets is produced with the size of the droplets being deliberately adjusted, preferably with the droplets being monodisperse, in order to achieve the regular arrangement of the droplets of the at least one first liquid encapsulated by the continuous jet of the second liquid with the size of the droplets being deliberately adjusted, preferably with the droplets being monodisperse.

[0029] In a preferred embodiment of the method according to the present invention, it is provided that micro stages are used for adjusting orientations and positions of the nozzles. Micro stages as such are well-known. Typically, with respect to an initial position each nozzle can be precisely translated in all three directions in space in the range from -5 mm to +5 mm and can be precisely rotated in the common plane in the range from 0° to 170° by means of the micro stages.

[0030] In a preferred embodiment of the method according to the present invention, it is provided that the at least one regular stream of droplets is produced with a droplet production frequency in the range from 1 Hz to 100 kHz, preferably from 5 kHz to 50 kHz. Accordingly, the regular arrangement of the droplets is an easily produced periodic spatial arrangement as a function of to the production frequency. In order to produce droplets with said production frequencies, the at least one first nozzle can be equipped with a piezo actuator, for example, which is known in the art.

[0031] It should be noted, however, that there are many other well-known means and droplet generators, respectively, for producing such drop streams, particularly systems that are typically referred to as "print head" of continuous or drop-on-demand type. They can be operated based on different principles, e.g. with a piezo element that squeezes a chamber and eject a controlled volume of liquid, with heat that locally brings the liquid (particularly an ink) above its boiling temperature for producing a bubble that leads to the ejection of a liquid drop of more or less the bubble volume, with a valve that is quick enough (solenoid valve) to let the pressurized liquid flow for a very short time period, etc.

[0032] The resulting regularity of the droplet stream provides perfectly reproducible collisions. The latter can be imaged using stroboscopic illumination (e.g. by means of LEDs) at the droplet production frequency, allowing for an elegant way of in-situ controlling the production of the continuous jet of the second liquid encapsulating the regular arrangement of the droplets of the first liquid.

[0033] In a preferred embodiment of the method according to the present invention, it is provided that the at least one regular stream of droplets has a velocity $\mathbf{u}_d$, the continuous jet has a velocity $\mathbf{u}_j$, the droplets in the regular stream are spaced at a spatial period l_d, the continuous jet has a diameter D_j, and the following relation holds

$$l_d/D_j * |\mathbf{u}_j|/|\mathbf{u}_d| < 2,$$

preferably

$$l_d/D_j * |\mathbf{u}_j|/|\mathbf{u}_d| < 1,8.$$

[0034] In doing so, an extremely well-defined continuous jet of the second liquid encapsulating the regular arrangement of the droplets of the first liquid is produced in the course of the collision of the continuous jet and the stream of droplets. Note that the velocities $\mathbf{u}_j$ and $\mathbf{u}_d$ are defined in the laboratory frame of reference. Absolute values of these velocities are typically in the range from 1 m/s to 20 m/s.

[0035] The above-described method also allows for an easy production of fibres with encapsulated objects that are regularly arranged within the fibres. Therefore, the continuous jet of the second liquid encapsulating the regular arrangement of the droplets of the first liquid (also referred to as "continuous jet with droplets" in the following) only needs to be hardened. Thus, a method for producing a fibre is provided, wherein it is provided according to the present invention that a regular arrangement of droplets of at least one first liquid in a continuous jet of a second liquid is produced using a method according to the present invention and that the continuous jet of the second liquid encapsulating the regular arrangement of the droplets of the at least one first liquid is hardened. Thereby, it has to be pointed out that the encapsulated objects and inclusions, respectively in the so-produced fibre, which objects are built by the droplets, not only can be spatially regularly arranged in the fibre, but also their sizes can be deliberately adjusted, since the size of the droplets can be deliberately adjusted, as described above. Particularly, fibres with encapsulated monodisperse objects can thus be produced.

[0036] The resulting fibres can be of particular interest for materials sciences, for example, since the inclusions can provide specific properties to the fibres. Moreover, the contrast between the properties of the hardened continuous jet of the second liquid and of the inclusions as well as the regularity of their spatial distribution in the fibre can be of interest in optics, since the inclusions can act as an array of lenses, for example. Furthermore the resulting fibres can be of interest for biological and pharmaceutical applications where encapsulation is a very active field. Encapsulating active biological elements such as cells or micro-organisms opens the routes to several therapeutic and non-therapeutic applications including, for example, tissue engineering, regenerative medicine, oral delivery of vaccines, etc. In all these applications, the difficulties lie in keeping the encapsulated cells or microorganisms (referred to as "reservoirs" in the following) alive during the encapsulation process but also after the encapsulation process for a longer period which depends on the specific application. Particularly, it should be able that nutriments diffuse through the encapsulating phase and it is therefore essential to achieve a spatial distribution of the encapsulated reservoirs which allows equal supply to all of them. Encapsulating each reservoir individually is unsatisfactory because of tremendous difficulties in manipulating and "dosing" the reservoirs, particularly in case of medical applications. The production method according to the invention, on the other hand, which exploits collisions of immiscible liquids in the form of at least one stream of droplets and a continuous jet, does offer the possibility to control and tune the distribution of the encapsulated objects via the formation of regular structures, i.e. via the formation of a regular arrangement of the droplets of at least one first liquid in the continuous jet of the second liquid, leading to regularly arranged objects in the fibres. Note that if several first liquids are used, fibres with regularly arranged encapsulated objects of different species can be produced, which can be of particular interest for medical applications.

[0037] In order to achieve the above-mentioned hardening, plenty of well-known mechanisms can be applied. For example, hardening can be achieved by exploiting a sol-gel transition.

[0038] As another example, hardening can also be achieved by cooling down the continuous jet with droplets at least below the solidification temperature of the second liquid. This can be done utilising the naturally occurring cooling down of the continuous jet with droplets along its trajectory in air (or in another chosen ambient gas in a certain production set-up). When the continuous jet with droplets has e.g. an initial temperature of 60°C (which is above the solidification temperature of the second liquid in this example) and cools down along its trajectory downstream the region of the collision to a temperature of 30° (which is at least below the solidification temperature of the second liquid, preferably also below the solidification temperature of the first liquid in this example).

[0039] Hence, in a preferred embodiment of the method according to the present invention, it is provided that the hardening is achieved by cooling down the continuous jet of the second liquid encapsulating the regular arrangement of the droplets of the at least one first liquid below a solidification temperature of the second liquid. If the temperature is reduced below the temperature of the at least one first liquid too, a fibre is obtained with encapsulated regularly arranged objects that are solid.

[0040] However, if the temperature is not reduced below the temperature of the first liquid, a solid fibre with liquid inclusions can be obtained. Similarly, also by other known hardening methods - e.g. by exploiting a sol-gel transition, where only the continuous jet is hardened, but not the droplets - a solid fibre with liquid inclusions can be obtained. Accordingly, in a preferred embodiment of the method according to the present invention, it is provided that only the second liquid is solidified during the hardening.

[0041] It has to be pointed out that using the method according to the present invention solid fibres containing regularly arranged inclusions can be obtained that have a perfectly cylindrical shape and a more or less uniform outer diameter. The latter means that the fibres exhibit practically no constrictions that are undesirable in many situations. Correspondingly, a cylindrical fibre is provided, which according to the present invention is obtainable by a method according to the present invention. Note that also the size of the inclusions can be deliberately adjusted, as already mentioned above. Preferably, monodisperse inclusions can be achieved.

[0042] Naturally, this includes that also a regular arrangement of droplets of a first liquid in a continuous jet of a second liquid is provided, which is obtainable by a method according to the present invention. Naturally, the size of the droplets of the regular arrangement in the continuous jet can be deliberately adjusted, as described above. Preferably, the droplets of the regular arrangement in the continuous jet can be monodisperse.

BRIEF DESCRIPTION OF FIGURES

[0043] The invention will be explained in closer detail by reference to a preferred embodiment, with

Fig. 1 showing a schematic view of a set-up for carrying out a method for producing a regular arrangement of droplets of a first liquid in a continuous jet of a second liquid according to the invention

Fig. 2 showing a detailed view of the continuous jet and a stream of droplets in Fig. 1

Fig. 3 showing a detailed view of the continuous jet and the stream of droplets just before and after a collision

Fig. 4 showing a fibre according to the present invention

WAYS FOR CARRYING OUT THE INVENTION

[0044] Fig. 1 shows a schematic view of a set-up for carrying out a method for producing a regular arrangement of droplets 3 of a first liquid 1 in a continuous jet 5 of a second liquid 2 according to the present invention. The first liquid 1 and the second liquid 2 are immiscible and chosen such that a surface tension σ_d of the first liquid 1 is greater than the sum of a surface tension σ_j of the second liquid 2 and an interfacial tension σ_{dj} between the first liquid 1 and the second liquid 2, i.e. the following relation holds:

$$\sigma_d > \sigma_j + \sigma_{dj}.$$

[0045] Hence, the first liquid 1 can be totally wetted by the second liquid 2.

[0046] A regular stream 4 of the droplets 3 is produced using at a first nozzle 6 and the continuous jet 5 is produced using a second nozzle 7. For producing the droplets 3 the first nozzle is equipped with a piezo actuator (not shown), which is known in the art. Hence, the droplets can be produced with a droplet production frequency in the range from 1 Hz to 100 kHz, e.g. with a droplet production frequency of 10 kHz.

[0047] The resulting regularity of the droplets 3 in the stream 4 provides perfectly reproducible collisions. The latter can be imaged using stroboscopic illumination by means of an LED (light-emitting diode) 10 at the droplet production frequency, allowing for an elegant way of in-situ controlling the production of the continuous jet 5 of the second liquid 2 encapsulating the regular arrangement of the droplets 3 of the first liquid 1. In Fig. 1 the LED 10 is arranged downstream the point or region where the collision takes place. The LED 10 is positioned at one side of the continuous jet 5 with the encapsulated droplets 3 and a camera 9 is positioned on the other side, opposite the LED 10 for taking the corresponding images, preferably with an integer multiple of the droplet production frequency.

[0048] The nozzles 6, 7 are adjusted such that the continuous jet 5 and the regular stream 4 of droplets 3 are in a common plane 8, collide and the continuous jet 5 of the second liquid 2 encapsulates the regular arrangement of the droplets 3 of the at least one first liquid 1 after the collision. Thereby, the above-mentioned relation ensures full encapsulation of the droplets 3 in the jet 5.

[0049] In the shown embodiment, the common plane

8 is parallel to spatial directions x, y that are standing perpendicular to each other as well as to spatial direction z, cf. Fig. 2.

[0050] The distance between nozzle orifices (not shown) is in the range from 5 mm to 5 cm and the distances between a point or region of collision and the nozzle orifices is in the range from 1 cm to 10 cm, typically. For adjusting the orientation (+/- 90°) and position (+/- 5 mm) of the nozzles 6, 7 very precisely, micro stages (not shown) are used.

[0051] The orifices have diameters that can be varied and are preferably adjusted in the range from 10 μm to 1500 μm.

[0052] Typically, both liquids 1, 2 have densities in the range from 700 kg/m^3 to 2000 kg/m^3 and viscosities in the range from 0,5 mPa s to 5 Pa s. For example, the first liquid 1 is an aqueous solution of glycerol with 50 wt.% glycerol and 50 wt.% water and the second liquid 2 is a silicon oil.

[0053] In the shown embodiment, the orifices of the nozzles 6, 7 are adjusted such that a Diameter D_j of the continuous jet 5 (of the silicon oil) is 300 μm and a diameter D_d of the droplets 3 (of the aqueous solution of glycerol) is 192 μm. In the shown example, this leads to a flow rate of 348 μl/s for the continuous jet 5 of the second liquid 2 and to a flow rate of 44 μl/s for the regular stream 4 of droplets 3.

[0054] The liquids 1, 2 are supplied to the nozzles 6, 7 by means of pressurised tanks 11, 12 that are independent from each other. Thereby, the first liquid 1 is stored in the pressurised tank 11 and the second liquid 2 is stored in the pressurised tank 12. Supply tubes 13 connect each of the pressurised tanks 11, 12 with the respective nozzle 6, 7.

[0055] As shown in Fig. 1 only the continuous jet 5 of the second liquid 5 encapsulating the regular arrangement of the droplets 3 of the first liquid 1 remains after the collision, i.e. downstream of the point or region where the collision takes place. For fostering this result, the continuous jet 5 and the stream 4 of droplets 3 enclose an angle α in the common plane 8, which is in the range from 1° to 170°, preferably from 5° to 90°, more preferably from 10° to 90°.

[0056] The angle α can be seen in the detailed view of Fig. 2. Moreover, it is illustrated that the regular stream 4 of droplets 3 has a velocity $\mathbf{u}_d$ (note that bold letters indicate vectors) and the continuous jet 5 has a velocity $\mathbf{u}_j$ in the laboratory frame. Absolute values $|\mathbf{u}_j|$, $|\mathbf{u}_d|$ are typically in the range from 1 m/s to 20 m/s.

[0057] The resulting relative velocity **U** (indicated by the vectors for **U**/2 in Fig. 2) is given by $\mathbf{u}_d - \mathbf{u}_j$. In Fig. 2 the Cartesian coordinate system spanned by spatial directions x, y, z is rotated such that the spatial direction y is parallel to **U**.

[0058] The droplets 3 in the regular stream 4 are spaced at a spatial period l_d. From the periodic character of the collisions it follows that a spatial period l_j can be attributed to the continuous jet 5, with

$$l_j = l_d * |\mathbf{u}_j|/|\mathbf{u}_d|.$$

[0059] In order to further increase the quality of the resulting continuous jet 5 of the second liquid 2 encapsulating the regular arrangement of the droplets 3 of the first liquid 1, in a preferred embodiment of the method according to the present invention it is provided that

$$l_d/D_j * |\mathbf{u}_j|/|\mathbf{u}_d| < 2,$$

preferably

$$l_d/D_j * |\mathbf{u}_j|/|\mathbf{u}_d| < 1,8,$$

holds.

[0060] Fig. 3 further illustrates how perfectly regular the achieved arrangement of the droplets 3 in the continuous jet 5 after the collision is. This means that the continuous jet 5 contains regularly embedded droplets 3, with the regular arrangement of the droplets 3 encapsulated in the continuous jet 5 being determined by the regular arrangement of the droplets 3 in the stream 4 just before the collision. Said regular arrangement of the droplets 3 encapsulated in the continuous jet 5 manifests in a spatial period l_i.

[0061] The resulting continuous jet 5 encapsulating the regular arrangement of the droplets 3 can be hardened, e.g. by cooling, in order to produce a (solid) fibre 14 containing a regular arrangement of inclusions. Particularly, said regular arrangement of the inclusions can (but in general does not have to) manifest in the spatial period l_i of the droplets 3 encapsulated in the continuous jet 5.

[0062] If cooling is done only below the solidification temperature of the second liquid 2, but not below the solidification temperature of the first liquid, the fibre 14 contains liquid inclusions, i.e. the regularly arranged inclusions are the regularly arranged droplets 3 of the first liquid 1. Fig. 4 shows such a fibre 14, wherein a solidified second liquid 15 forms a body of the fibre 14. Within the solidified second liquid 15 and the body of the fibre 14, respectively, the droplets 3 of the first liquid 1 are regularly arranged along the fibre 14, i.e. the droplets 3 of the first liquid 1 form the regularly arranged inclusions.

[0063] Note that the body of the fibre 14 is perfectly cylindrical, i.e. a diameter of the body is essentially constant along the fibre 14, which is advantageous for many applications. Thereby, the perfectly cylindrical shape can be achieved for solid inclusions as well - for example, when cooling down is done also below the solidification temperature of the first liquid 1.

List of reference signs

[0064]

1	First liquid
2	Second liquid
3	Droplet (of the first liquid)
4	Regular stream of droplets
5	Continuous jet (of the second liquid)
6	First nozzle
7	Second nozzle
8	Plane
9	Camera
10	Light emitting diode (LED)
11	Pressurised tank for the first liquid
12	Pressurised tank for the second liquid
13	Supply tube
14	Fibre
15	Solidified second liquid
σ_d	Surface tension of the first liquid
σ_j	Surface tension of the second liquid
σ_{dj}	interfacial tension
$\mathbf{u}_d$	Velocity of the regular stream of droplets
$\mathbf{u}_j$	Velocitiy of the continuous jet
$\mathbf{U}$	Relative velocitiy ($\mathbf{u}_d$ - $\mathbf{u}_j$)
l_d	Spatial period of the droplets in the regular stream of droplets
l_j	Spatial period of the continuous jet
l_i	Spatial period of the droplets encapsulated in the continuous jet and of the inclusions in the fibre, respectively
D_j	Diameter of the continuous jet
D_d	Droplet diameter
α	Angle enclosed by the continuous jet and the droplet stream
x, y, z	Spatial direction

Claims

1. Method for producing a regular arrangement of droplets (3) of at least one first liquid (1) in a continuous jet (5) of a second liquid (2), wherein the first liquid (1) and the second liquid (2) are immiscible and chosen such that a surface tension (σ_d) of the first liquid (1) is greater than the sum of a surface tension (σ_j) of the second liquid (2) and an interfacial tension (σ_{dj}) between the first liquid (1) and the second liquid (2),
wherein at least one regular stream (4) of the droplets (3) is produced using at least one first nozzle (6) and the continuous jet (5) is produced using a second nozzle (7), wherein the nozzles (6, 7) are adjusted such that the continuous jet (5) and the at least one regular stream (4) of droplets (3) are in a common plane (8), collide and the continuous jet (5) of the second liquid (2) encapsulates the regular arrangement of the droplets (3) of the at least one first liquid (1) after the collision.

2. Method according to claim 1, **characterised in that** the at least one regular stream (4) of the droplets (3) is produced with the size of the droplets (3) being deliberately adjusted, preferably with the droplets (3) being monodisperse, in order to achieve the regular arrangement of the droplets (3) of the at least one first liquid (1) encapsulated by the continuous jet (5) of the second liquid (2) with the size of the droplets (3) being deliberately adjusted, preferably with the droplets (3) being monodisperse.

3. Method according to any one of claims 1 to 2, **characterised in that** the continuous jet (5) and the at least one stream (4) of droplets (3) enclose an angle (α) in the common plane (8), which angle (α) is in the range from 1° to 170°, preferably from 5° to 90°.

4. Method according to any one of claims 1 to 3, **characterised in that** a diameter of an orifice of the at least one first nozzle (6) and a diameter of an orifice of the second nozzle (7) are adjusted in the range from 10 μm to 1500 μm.

5. Method according to any one of claims 1 to 4, **characterised in that** micro stages are used for adjusting orientations and positions of the nozzles (6, 7).

6. Method according to any one of claims 1 to 5, **characterised in that** the at least one regular stream (4) of droplets (3) is produced with a droplet production frequency in the range from 1 Hz to 100 kHz, preferably from 5 kHz to 50 kHz.

7. Method according to any one of claims 1 to 6, **characterised in that** the at least one regular stream (4) of droplets (3) has a velocity $\mathbf{u}_d$, the continuous jet (5) has a velocity $\mathbf{u}_j$, the droplets (3) in the regular stream (4) are spaced at a spatial period l_d, the continuous jet has a diameter D_j, and the following relation holds

$$l_d/D_j * |\mathbf{u_j}|/|\mathbf{u_d}| < 2,$$

preferably

$$l_d/D_j * |\mathbf{u_j}|/|\mathbf{u_d}| < 1,8.$$

8. Method according to any one of claims 1 to 7, **characterised in that** several regular streams (4) of droplets (3), preferably of several first liquids (1), are provided, wherein the several regular streams (4) of the droplets (3) are produced using several first nozzles (6) .

9. Method for producing a fibre, **characterised in that** a regular arrangement of droplets (3) of at least one first liquid (1) in a continuous jet (5) of a second liquid (2) is produced using a method according to any one

of claims 1 to 8
and **in that** the continuous jet (5) of the second liquid (2) encapsulating the regular arrangement of the droplets (3) of the at least one first liquid (1) is hardened.

10. Method according to claim 9, **characterised in that** the hardening is achieved by cooling down the continuous jet (5) of the second liquid (2) encapsulating the regular arrangement of the droplets (3) of the at least one first liquid (1) below a solidification temperature of the second liquid (2).

11. Method according to any one of claims 9 to 10, **characterised in that** only the second liquid (2) is solidified during the hardening.

12. Cylindrical fibre obtainable by a method according to any one of claims 9 to 11.

Amended claims in accordance with Rule 137(2) EPC.

1. Method for producing a regular arrangement of droplets (3) of at least one first liquid (1) in a continuous jet (5) of a second liquid (2), wherein the first liquid (1) and the second liquid (2) are immiscible and chosen such that a surface tension (σ_d) of the first liquid (1) is greater than the sum of a surface tension (σ_j) of the second liquid (2) and an interfacial tension (σ_{dj}) between the first liquid (1) and the second liquid (2),
wherein at least one regular stream (4) of the droplets (3) is produced using at least one first nozzle (6) and the continuous jet (5) is produced using a second nozzle (7), wherein the nozzles (6, 7) are adjusted such that the continuous jet (5) and the at least one regular stream (4) of droplets (3) are in a common plane (8), collide and the continuous jet (5) of the second liquid (2) encapsulates the regular arrangement of the droplets (3) of the at least one first liquid (1) after the collision.

2. Method according to claim 1, **characterised in that** the at least one regular stream (4) of the droplets (3) is produced with the size of the droplets (3) being deliberately adjusted, preferably with the droplets (3) being monodisperse, in order to achieve the regular arrangement of the droplets (3) of the at least one first liquid (1) encapsulated by the continuous jet (5) of the second liquid (2) with the size of the droplets (3) being deliberately adjusted, preferably with the droplets (3) being monodisperse.

3. Method according to any one of claims 1 to 2, **characterised in that** the continuous jet (5) and the at least one stream (4) of droplets (3) enclose an angle (α) in the common plane (8), which angle (α) is in the range from 1° to 170°, preferably from 5° to 90°.

4. Method according to any one of claims 1 to 3, **characterised in that** a diameter of an orifice of the at least one first nozzle (6) and a diameter of an orifice of the second nozzle (7) are adjusted in the range from 10 μm to 1500 μm.

5. Method according to any one of claims 1 to 4, **characterised in that** micro stages are used for adjusting orientations and positions of the nozzles (6, 7).

6. Method according to any one of claims 1 to 5, **characterised in that** the at least one regular stream (4) of droplets (3) is produced with a droplet production frequency in the range from 1 Hz to 100 kHz, preferably from 5 kHz to 50 kHz.

7. Method according to any one of claims 1 to 6, **characterised in that** the at least one regular stream (4) of droplets (3) has a velocity $\mathbf{u_d}$, the continuous jet (5) has a velocity $\mathbf{u_j}$, the droplets (3) in the regular stream (4) are spaced at a spatial period l_d, the continuous jet has a diameter D_j, and the following relation holds

$$l_d/D_j * |\mathbf{u_j}|/|\mathbf{u_d}| < 2,$$

preferably

$$l_d/D_j * |\mathbf{u_j}|/|\mathbf{u_d}| < 1,8.$$

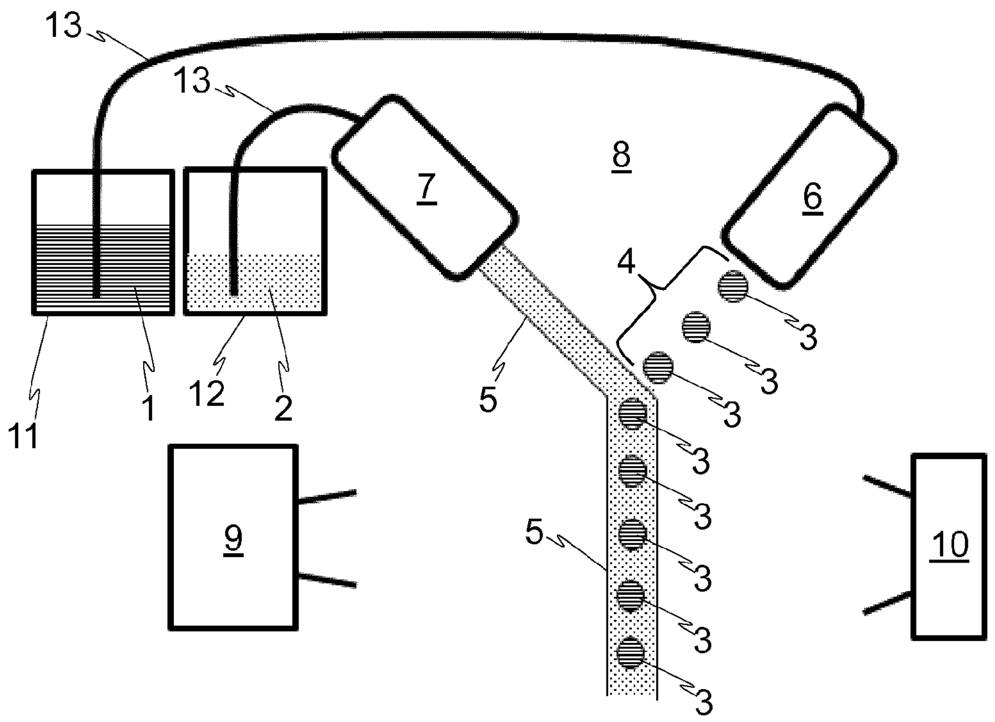
8. Method according to any one of claims 1 to 7, **characterised in that** several regular streams (4) of droplets (3), preferably of several first liquids (1), are provided, wherein the several regular streams (4) of the droplets (3) are produced using several first nozzles (6).

9. Method for producing a fibre, **characterised in that** a regular arrangement of droplets (3) of at least one first liquid (1) in a continuous jet (5) of a second liquid (2) is produced using a method according to any one of claims 1 to 8
and **in that** the continuous jet (5) of the second liquid (2) encapsulating the regular arrangement of the droplets (3) of the at least one first liquid (1) is hardened.

10. Method according to claim 9, **characterised in that** the hardening is achieved by cooling down the continuous jet (5) of the second liquid (2) encapsulating the regular arrangement of the droplets (3) of the at least one first liquid (1) below a solidification temperature of the second liquid (2).

11. Method according to any one of claims 9 to 10, **characterised in that** only the second liquid (2) is solidified during the hardening.

Fig. 1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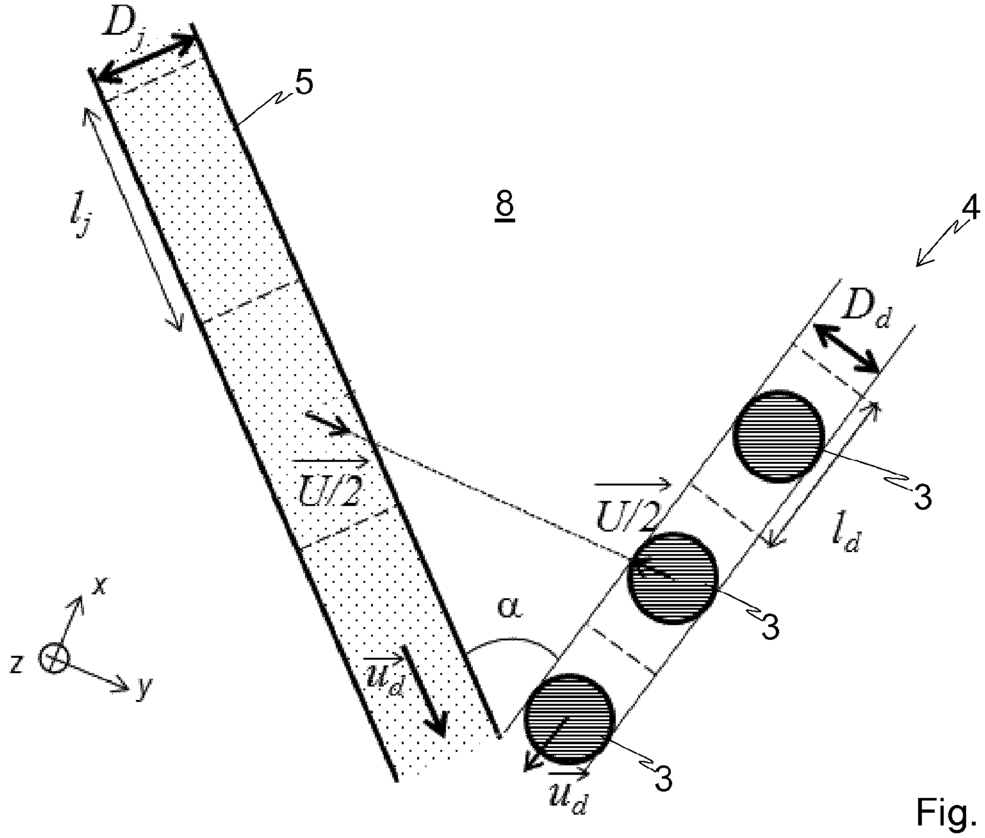
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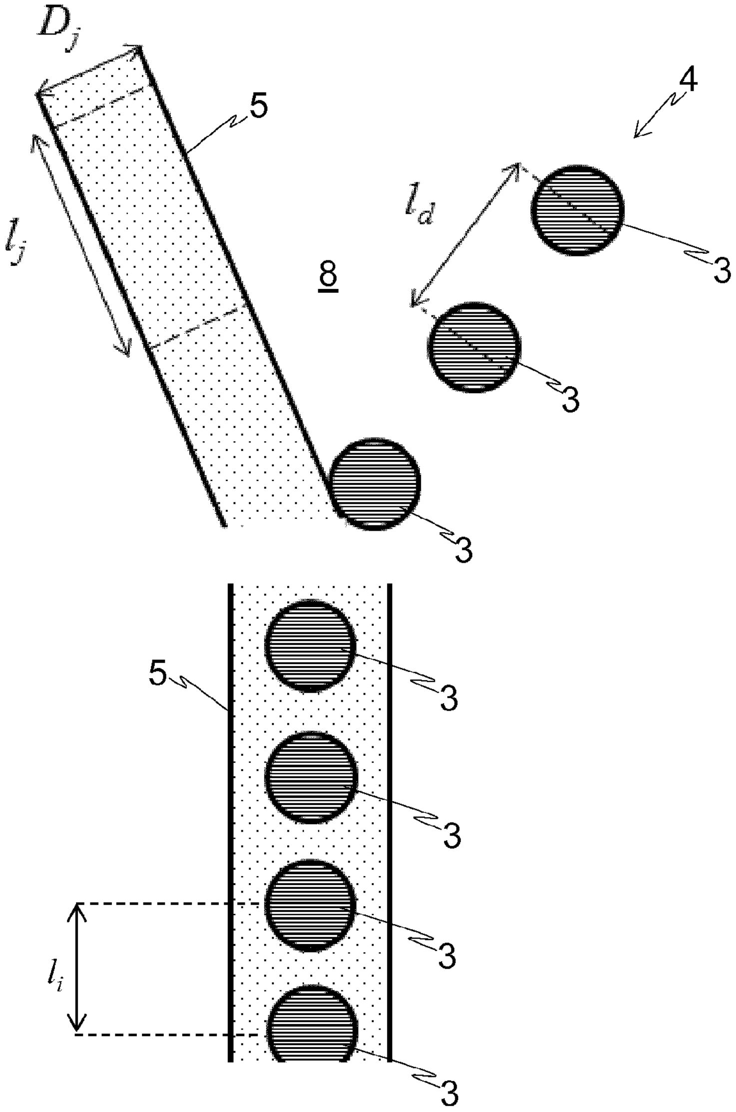
Fig. 2

Fig. 3



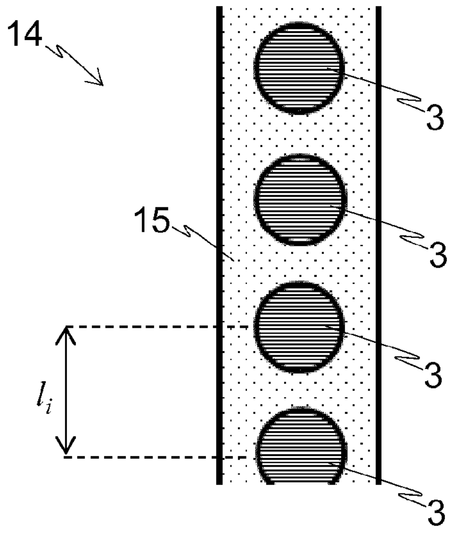


Fig. 4

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EUROPEAN SEARCH REPORT

Application Number
EP 17 17 5132

DOCUMENTS CONSIDERED TO BE RELEVANT

Category	Citation of document with indication, where appropriate, of relevant passages	Relevant to claim	CLASSIFICATION OF THE APPLICATION (IPC)
A	US 2007/054119 A1 (GARSTECKI PIOTR [PL] ET AL) 8 March 2007 (2007-03-08) * paragraphs [0003], [0032], [0050], [0078]; figures 1A,1B; example 1 * -----	1-12	INV. D01D5/00 B05B7/04 B05B17/04 D01D5/36
A	WO 2014/120321 A2 (NDSU RES FOUNDATION [US]) 7 August 2014 (2014-08-07) * page 15, line 17 - page 16, line 2; figure 9 * * page 18, lines 1-9; figures 13,14 * -----	1-12	

TECHNICAL FIELDS SEARCHED (IPC)
D01D
B05B

The present search report has been drawn up for all claims

3

Place of search	Date of completion of the search	Examiner
The Hague	18 October 2017	Van Beurden-Hopkins

CATEGORY OF CITED DOCUMENTS

X : particularly relevant if taken alone
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EP 17 17 5132

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18-10-2017

Patent document cited in search report	Publication date	Patent family member(s)	Publication date
US 2007054119 A1	08-03-2007	NONE	
WO 2014120321 A2	07-08-2014	NONE	

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For more details about this annex : see Official Journal of the European Patent Office, No. 12/82

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