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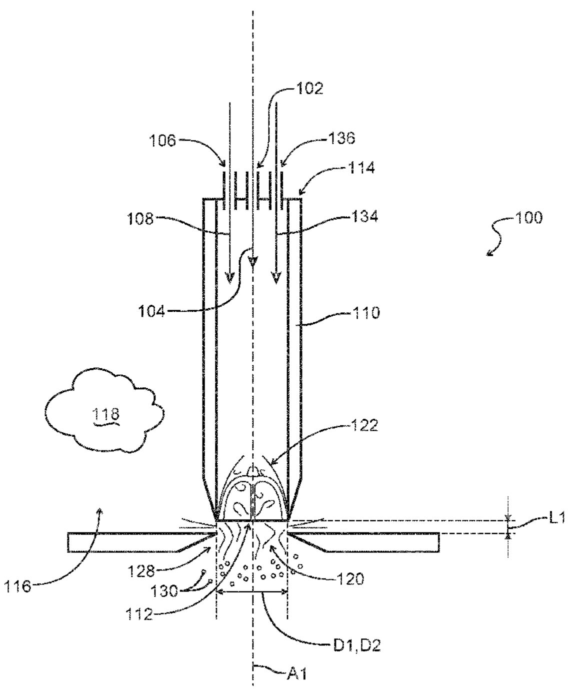

FIG. 1

(57) Abstract: A method of mixing at least two liquids includes providing a nozzle 100 including a feeding tube 110 having a feeding tube outlet 112. The feeding tube is positioned in a pressure chamber 116 containing a pressurized gas 118. The pressure chamber includes a pressure chamber exit orifice 120 substantially coaxial with the feeding tube outlet downstream of the feeding tube outlet. The feeding tube outlet is axially offset from the pressure chamber exit orifice by an axial gap. The method also includes introducing a first stream 104 of a first liquid and a second stream 108 of a second liquid into the feeding tube upstream from the feeding tube outlet. The method further includes operating the nozzle in a flow blurring mode such that a reflux cell is formed in the feeding tube upstream from the feeding tube outlet. A turbulent mixing of the first and second liquids is thereby provided in the reflux cell.

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APPARATUS AND METHOD OF MIXING AT LEAST TWO LIQUIDS

BACKGROUND

[0001] The present disclosure relates generally to a apparatus and a method of mixing at least two liquids. More particularly, the present disclosure pertains to micromixing of liquids to obtain emulsions.

[0002] The production of multiphase systems at a small scale is very interesting in many applications in pharmacy, food, agronomic, and scientific industries. Among these multiphase systems, we can find emulsions, foams, or aerosols. Their production by purely fluid dynamic processes, particularly by pneumatic means, allows very different applications and developments in industry, technology, science, and daily life. Aerosols have been used in various technological fields, particularly as a means to treat respiratory diseases through nebulization of liquid medicines. The administration of medicines through inhalation using aerosols allows appropriate concentrations of medicine to be obtained in the respiratory system, minimizing side effects. In the same way, applications in the agronomic field are very well known, such as spraying pest-control substances as a part of a treatment of protection against insects. To do this, manual or automatic equipment are used which allow a targeted delivery and the capacity to control the size of drops, whose diameter usually varies between 100 and 500 microns. When drop sizes are inferior, between 50 and 100 microns, the term “nebulization” is usually used. When applying pest-control substances, it increases not only the capacity of flotation of the preparation but also the covered area when deposition of drops takes place.

[0003] Current aerosol technologies include invasion of the feeding tube by an invading stream of gas. The essential feature of the technology is the production of a reflux cell, where scales of turbulence are created ensuring in this way a closer interaction between the gas and liquid. As such, an invading gas may mix with an intercepted fluid. The exiting flow of the intercepted fluid meets an approximately perpendicular stream of gas directed radially and centripetally to the axis of the exiting flow.

DESCRIPTION

[0004] Briefly, the present disclosure relates, in one embodiment, to a method of mixing at least two liquids. The method includes providing a nozzle including a feeding tube having a feeding tube outlet. The feeding tube is positioned in a pressure chamber containing a pressurized gas. The pressure chamber includes a pressure chamber exit orifice substantially coaxial with the feeding tube outlet downstream of the feeding tube outlet. The feeding tube outlet is axially offset from the pressure chamber exit orifice by an axial gap. The method also includes introducing a first stream of a first liquid and a second stream of a second liquid into the feeding tube upstream from the feeding tube outlet. The method further includes operating the nozzle in a flow blurring mode such that a reflux cell is formed in the feeding tube upstream from the feeding tube outlet. A turbulent mixing of the first and second liquids is thereby provided in the reflux cell.

[0005] An alternative embodiment includes ejecting a mixture of the first and second liquids out of the pressure chamber exit orifice.

[0006] Still another embodiment includes the mixture being in a form of droplets entrained in the gas.

[0007] Yet another embodiment includes each droplet including an emulsion of the first and second liquids.

[0008] Another embodiment includes drying the droplets to create dried mixture particles.

[0009] A further embodiment includes at least one of the first and second liquids including solids, and the dried mixture particles including the solids.

[0010] A further still embodiment includes forming an emulsion of the first and second liquids in the reflux cell and breaking the emulsion into droplets as the emulsion is ejected out of the pressure chamber exit orifice.

[0011] Yet another embodiment includes the first liquid and the second liquid including incompatible reagents. The nozzle operation also includes forming a mixture of the first and second liquids in the reflux cell and breaking the mixture into droplets as the mixture is ejected out of the pressure chamber exit orifice. The embodiment further includes reacting the incompatible reagents with each other inside each droplet so that each droplet acts as a microreactor.

[0012] Still another embodiment includes each of the first and second liquids being introduced into the feeding tube at separate inlets upstream of the reflux cell.

[0013] An even further embodiment includes the feeding tube being an outer feeding tube, and the feeding tube outlet being an outer feeding tube outlet. The introduction of the first and second liquid streams further comprises introducing the first liquid into the outer feeding tube via a concentric inner feeding tube having an inner feeding tube outlet, and introducing the second liquid into the outer feeding tube at a second inlet upstream of the inner feeding tube outlet.

[0014] Another embodiment includes the inner feeding tube outlet being located upstream of the outer feeding tube outlet by a recess distance.

[0015] Still another embodiment includes the recess distance being configured to allow the reflux cell to form uninterrupted by the inner feeding tube.

[0016] One embodiment includes the recess distance being at least as great as an outlet diameter of the outer feeding tube outlet.

[0017] A further embodiment includes the nozzle operation including the reflux cell being formed in the outer feeding tube but not in the inner feeding tube.

[0018] A further still embodiment includes the first liquid having enough low viscosity to allow the first liquid to be introduced through the inner feeding tube.

[0019] An even further embodiment includes the introduction of the first and second liquid streams further including introducing a third stream of a third liquid into the feeding tube upstream from the feeding tube outlet.

[0020] The present disclosure also relates, in one embodiment, to a nozzle apparatus. The nozzle apparatus includes a first inlet for receiving a first liquid stream, a second inlet for receiving a second liquid stream, and an outer feed tube. The outer feed tube is configured to receive the first and second liquid streams and has an outer feed tube axis. The outer feed tube further includes an outer feed tube outlet having an outer feed tube outlet diameter. The nozzle apparatus further includes a pressure chamber surrounding the outer feed tube outlet for containing a pressurized gas. The pressure chamber includes a pressure chamber exit orifice. The pressure chamber exit orifice is coaxial with the outer feed tube axis and has an exit orifice diameter. The pressure chamber exit orifice is spaced from the outer feed tube outlet to define an axial gap between the pressure chamber exit orifice and the outer feed tube outlet. The outer feed tube outlet diameter, the exit orifice diameter, and the axial gap are configured such that a reflux cell can form inside the outer feed tube when the first and second liquid streams are forced through the outer feed tube and the gas is forced through the axial gap.

[0021] A further embodiment includes an inner feed tube concentrically disposed in the outer feed tube, the first inlet being communicated with the inner feed tube.

[0022] Another embodiment includes the inner feed tube including an inner feed tube outlet, the inner feed tube outlet being recessed from the outer feed tube outlet by a recess distance.

[0023] Some embodiments include the recess distance configured to allow the reflux cell to form uninterrupted by the inner feed tube.

[0024] Still another embodiment includes the recess distance being at least as great as the outer feed tube outlet diameter.

[0025] Other embodiments include the recess distance being sufficient that the reflux cell forms in the outer feed tube but not in the inner fed tube during use.

[0026] Some other embodiments include the outer feed tube outlet diameter being in a range of from 70 microns to 8 millimeters.

[0027] Yet another embodiment includes the outer feed tube outlet diameter being in a range of from 200 to 5000 microns. The exit orifice diameter is in a range of from 90 to 120 percent of the outer feed tube outlet diameter. The inner feed tube includes an inner feed tube outlet having an inner feed tube outlet diameter. The axial gap is no greater than one fourth of the exit orifice diameter.

[0028] Yet another embodiment includes the outer feed tube outlet diameter being in a range of from 500 to 2000 microns.

[0029] Other embodiments include the inner feed tube including an inner feed tube outlet having an inner feed tube outlet diameter.

[0030] Still other embodiments include the exit orifice diameter in a range of from 90 to 120 percent of the outer feed tube outlet diameter.

[0031] Further embodiments include the exit orifice diameter substantially equal to the outer feed tube outlet diameter.

[0032] Another further embodiment includes the outer feed tube outlet diameter being in a range of from 70 microns to 8 millimeters.

[0033] Yet further embodiments include the outer feed tube outlet diameter in a range of from 200 to 5000 microns.

[0034] Yet further embodiment includes the outer feed tube outlet diameter being in a range of from 500 to 2000 microns.

[0035] Still further embodiments include the axial gap no greater than one half of the exit orifice diameter.

[0036] Another embodiment includes the axial gap being no greater than one fourth of the exit orifice diameter.

[0037] Still another embodiment includes the outer feed tube including an inlet end opposite from the outer feed tube outlet. Both of the first and second inlets are communicated with the outer feed tube closer to the inlet end than to the outer feed tube outlet.

[0038] One advantage of the present invention is that it provides a technique for mixing of very viscous liquids, which are otherwise very difficult to mix. Mixing of liquids having viscosities up to 300,000 cP is possible by the techniques disclosed herein.

BRIEF DISCRIPTION OF THE DRAWINGS

[0039] Fig. 1 is a cross-sectional side view of one embodiment of a nozzle apparatus.

[0040] Fig. 2 is a cross-sectional side view of another embodiment of a nozzle apparatus.

[0041] Fig. 3A is a photo of an example of poorly mixed liquids. It is noted that all of Figs. 3A-10B are actually photographs of dried samples of the liquid droplets which are representative of the mixing of the liquids in the droplets.

[0042] Fig. 3B is a photo of the example in Fig. 3A under a black light so as to show the fluorochrome introduced in one of the two liquids.

[0043] Fig. 4A is a photo of an example of substantially homogeneous liquid mixture achieved by the current disclosure.

[0044] Fig. 4B is a photo of the example in Fig. 4A under a black light so as to show the fluorochrome introduced in one of the two liquids.

[0045] Fig. 5A is a photo of another example of substantially homogeneous liquid mixture achieved by the current disclosure.

[0046] Fig. 5B is a photo of the example in Fig. 5A under a black light so as to show the fluorochrome introduced in one of the two liquids.

[0047] Fig. 6A is a photo of yet another example of substantially homogeneous liquid mixture achieved by the current disclosure.

[0048] Fig. 6B is a photo of the example in Fig. 6A under a black light so as to show the fluorochrome introduced in one of the two liquids.

[0049] Fig. 7A is a photo of still another example of substantially homogeneous liquid mixture achieved by the current disclosure.

[0050] Fig. 7B is a photo of the example in Fig. 7A under a black light so as to show the fluorochrome introduced in one of the two liquids.

[0051] Fig. 8A is a photo of a further example of substantially homogeneous liquid mixture achieved by the current disclosure.

[0052] Fig. 8B is a photo of the example in Fig. 8A under a black light so as to show the fluorochrome introduced in one of the two liquids.

[0053] Fig. 9A is a photo of a further still example of substantially homogeneous liquid mixture achieved by the current disclosure.

[0054] Fig. 9B is a photo of the example in Fig. 9A under a black light so as to show the fluorochrome introduced in one of the two liquids.

[0055] Fig. 10A is a photo of yet a further example of substantially homogeneous liquid mixture achieved by the current disclosure.

[0056] Fig. 10B is a photo of the example in Fig. 10A under a black light so as to show the fluorochrome introduced in one of the two liquids.

[0057] Fig. 11A is a droplet size distribution chart of the results of a first experiment achieved by the current disclosure.

[0058] Fig. 11B is a droplet size distribution chart of the results of a second experiment under the same experimental conditions than 11A but with a liquid with higher viscosity achieved by the current disclosure.

[0059] Fig. 12A is a droplet size distribution chart of the results of a third experiment achieved by the current disclosure.

[0060] Fig. 12B is a droplet size distribution chart of the results of a fourth experiment under the same experimental conditions than 12A but with a liquid with higher viscosity achieved by the current disclosure.

[0061] Fig. 13A is a droplet size distribution chart of the results of a fifth experiment achieved by the current disclosure.

[0062] Fig. 13B is a droplet size distribution chart of the results of a sixth experiment with the same liquids than 13A but different pressure achieved by the current disclosure.

[0063] Fig. 13C is a droplet size distribution chart of the results of a seventh experiment with the same liquids than 13A and 13B but different pressure achieved by the current disclosure.

[0064] Fig. 14 is a droplet size distribution chart of the results of an eighth experiment achieved by the current disclosure as shown in Figs. 10A and 10B.

[0065] Fig. 15 is a schematic representation of an exemplary droplet achieved by the current disclosure. Fig. 15 is also representative of a dried particle made from a liquid droplet.

DETAILED DESCRIPTION

[0066] Reference will now be made in detail to embodiments of the present disclosure, one or more drawings of which are set forth herein. Each drawing is provided by way of explanation of the present disclosure and is not a limitation. In fact, it will be apparent to those skilled in the art that various modifications and variations can be made to the teachings of the present disclosure without departing from the scope of the disclosure. For instance, features illustrated or described as part of one embodiment can be used with another embodiment to yield a still further embodiment.

[0067] Thus, it is intended that the present disclosure covers such modifications and variations as come within the scope of the appended claims and their equivalents. Other objects, features, and aspects of the present disclosure are disclosed in, or are obvious from, the following detailed description. It is to be understood by one of ordinary skill in the art that the present discussion is a description of exemplary embodiments only and is not intended as limiting the broader aspects of the present disclosure.

[0068] The words “connected”, “attached”, “joined”, “mounted”, “fastened”, and the like should be interpreted to mean any manner of joining two objects including, but not limited to, the use of any fasteners such as screws, nuts and bolts, bolts, pin and clevis, and the like allowing for a stationary, translatable, or pivotable relationship; welding of any kind such as traditional MIG welding, TIG welding, friction welding, brazing, soldering, ultrasonic welding, torch welding, inductive welding, and the like; using any resin, glue, epoxy, and the like; being integrally formed as a single part together; any mechanical fit such as a friction fit, interference fit, slidable fit, rotatable fit, pivotable fit, and the like; any combination thereof; and the like.

[0069] Unless specifically stated otherwise, any part of the apparatus of the present disclosure may be made of any appropriate or suitable material including, but not limited to, metal, alloy, polymer, polymer mixture, wood, composite, or any combination thereof.

[0070] Referring to Fig. 1, a nozzle apparatus **100** is shown. The nozzle apparatus **100** may include a first inlet **102** for receiving a first liquid stream **104** and a second inlet **106** for receiving a second liquid stream **108**. An outer feed tube **110** of the nozzle apparatus **100** may be configured to receive the first liquid stream **104** and second liquid stream **108**. The outer feed tube **110** may include an outer feed tube axis **A1**, an outer feed tube outlet **112**, and an inlet end **114** opposite from the outer feed tube outlet. The outer feed tube outlet **112** may include an outer feed tube outlet diameter **D1**. Both the first inlet **102** and the second inlet **106** may be communicated with the outer feed tube **110** nearer the inlet end **114** than the outer feed tube outlet **112**.

[0071] A pressure chamber **116** may surround the outer feed tube outlet **112**. The pressure chamber **116** may be configured to contain a pressurized gas **118**, such as oxygen, nitrogen, carbon dioxide, air, and the like, and may include a pressure chamber exit orifice **120**. The pressure chamber exit orifice **120** may be coaxial with the outer feed tube axis **A1** and may include an exit orifice diameter **D2**. The outer feed tube outlet **112** and the pressure chamber exit orifice **120** may be spaced from each other to define an axial gap **L1** between the pressure chamber exit orifice and the outer feed tube outlet. The exit orifice diameter **D2** may be in a range of from 90% to 120% of the outer feed tube outlet diameter **D1**. Various embodiments may include the exit orifice diameter **D2** being substantially equal to the outer feed tube outlet diameter **D1**. Many embodiments may include the outer feed tube outlet diameter **D1** being in the range of from about 70 microns to about 8 millimeters. Some embodiments may include the outer feed tube outlet diameter **D1** being in the range of from about 200 microns to about 5000 microns. Some embodiments may include the outer feed tube outlet diameter **D1** being in the range of from about 500 microns to about 2000 microns. In some embodiments, the axial gap **L1** may be no greater than one half of the exit orifice diameter **D2**. In other embodiments, the axial gap **L1** may be no greater than one fourth of the exit orifice diameter **D2**. These proportions may provide for a proper reflux cell **122** to be formed in the outer feed tube **110** for appropriate pressures and other conditions.

[0072] The outer feed tube outlet diameter **D1**, the exit orifice diameter **D2**, and the axial gap **L1** may be configured such that a reflux cell **122** may form inside the outer feed tube **110** when the first liquid stream **104** and the second liquid stream **108** are forced through the outer feed tube and the gas **118** is forced through the axial gap.

[0073] In some embodiments, as shown in Fig. 2, the nozzle apparatus **100** may further include an inner feed tube **124**. The inner feed tube **124** may be concentrically disposed in the outer feed tube **110**. In such embodiments, the first inlet **102** may be communicated with the inner feed tube **124**. The inner feed tube **124** may include an inner feed tube outlet **126** with an inner feed tube outlet diameter **D3**. The inner feed tube outlet diameter **D3** may be any appropriate size. Some embodiments may include the inner feed tube outlet diameter **D3** in a range from about one quarter to about one half of the outer feed tube outlet diameter **D1**. The inner feed tube outlet **126** may be recessed from the outer feed tube outlet **112** by a recess distance **L2**. Generally, the recess distance **L2** may allow for the reflux cell **122** to form without interruption by the inner feed tube **124**. In some embodiments, the recess distance **L2** may be at least as great as the outer feed tube outlet diameter **D1**. In other embodiments the recess distance **L2** may be up to about five times as great as the outer feed tube outlet diameter **D1.** These dimensions may allow for a proper reflux cell **122** to be formed in the outer feed tube **110**. Many embodiments include the recess distance **L2** being sufficient that the reflux cell **122** may form in the outer feed tube **110** but not in the inner feed tube **124**.

[0074] The present disclosure also relates to a method of mixing at least two liquids. The method may include providing a nozzle **100** including a feeding tube **110** having a feeding tube outlet **112**. The feeding tube **110** may be positioned in a pressure chamber **116** containing a pressurized gas **118**. The pressure chamber **116** may include a pressure chamber exit orifice **120** substantially coaxial with the feeding tube outlet **112** downstream of the feeding tube outlet. The feeding tube outlet **112** may be axially offset from the pressure chamber exit orifice **120** by an axial gap **L1**. The method may also include introducing a first stream **104** of a first liquid and a second stream **108** of a second liquid into the feeding tube **110** upstream from the feeding tube outlet **112**. The nozzle **100** may be operated in a flow blurring mode such that a reflux cell **122** is formed in the feeding tube **110** upstream from the feeding tube outlet **112**. A turbulent mixing of the first and second liquids may thereby be provided in the reflux cell **122**.

[0075] The method may further include ejecting a mixture **128** of the first and second liquids **104**, **108** out of the pressure chamber exit orifice **120**. The mixture **128** may be in the form of droplets **130** entrained in the gas **118**. As seen in Fig. 15 each droplet **130** may include an emulsion of the first liquid **104** and the second liquid **108**. In Fig. 15 the second liquid **108** forms a matrix or external phase in which are dispersed minute droplets of the first liquid **104** which is the internal phase of the emulsion. Some embodiments of the method may include drying the droplets **130** to create dried mixture particles **132**. The schematic representation of droplet **130** in Fig. 15 is also representative of a dried mixture particle **132**. Many embodiments may include at least one of the first and second liquids **104**, **108** including solids that are then a part of the dried mixture particles **132**. The liquids **104**, **108** may be highly viscous in some embodiments. Other embodiments may include the liquids **104**, **108** of any viscosity. These liquids **104**, **108** may be of any nature and may interact to form a solution instead of an emulsion.

[0076] The method may also include, in the introduction of the first stream **104** and second stream **108**, forming an emulsion of the first and second liquids in the reflux cell **122**. The emulsion may be broken into droplets **130** as the emulsion is ejected out of the pressure chamber exit orifice **120**.

[0077] Some embodiments of the method may include the first liquid **104** and the second liquid **108** including reagents that are incompatible with each other. In such embodiments, the operation of the nozzle **100** in a flow blurring mode step may further include forming a mixture **128** of the first and second liquids **104**, **108** in the reflux cell **122**. The mixture **128** may be broken into droplets **130** as the mixture is ejected out of the pressure chamber exit orifice **120**. The incompatible reagents may react with each other inside each droplet **130** so that each droplet acts as a microreactor.

[0078] The method may also include introducing each of the first and second liquids **104**, **108** into the feeding tube **110** at separate inlets **102**, **106** upstream of the reflux cell **122**. Some embodiments may also include introducing a third stream **134** of a third liquid into the feeding tube **110** upstream from the feeding tube outlet **112**. The third liquid **134** may be introduced via one of the first and second inlets **102**, **106** or may be introduced via a third inlet **136**.

[0079] The feeding tube **110** may be an outer feeding tube in many embodiments of the method. In such embodiments, the feeding tube outlet **112** is an outer feeding tube outlet. These embodiments may also include the introduction of the first liquid stream **104** into the inner feeding tube **124** at a first inlet **102** and introducing the second liquid stream **108** into the outer feeding tube **110** at a second inlet **106** upstream of the inner feeding tube outlet **126**. The inner feeding tube outlet **126** may be located upstream of the outer feeding tube outlet **112** by a recess distance **L2**. The recess distance **L2** may allow for the reflux cell **122** to form without interference from the inner feeding tube **124**. In some embodiments, the recess distance **L2** may be at least as great as an outlet diameter **D1** of the outer feeding tube outlet **112**. The reflux cell **122** in the flow blurring step may be formed in the outer feeding tube **110** but not in the inner feeding tube **124**. When the first liquid is to be introduced through the inner feeding tube **124** the first liquid **104** may the proper viscosity in order to promote flow through the smaller inner tube **124**. Other embodiments may include the second liquid **108** having a greater viscosity than the first liquid **104**. Even further embodiments may include the liquids **104**, **108** having the same or substantially similar characteristics, such as viscosity, especially when the single tube embodiment of Fig. 1 is used. It is also possible in some embodiments to allow the reflux cell to also form in the inner feeding tube if the inner feeding tube is of sufficient size.

[0080] Turning now to the photographs of Figs. 3A-10B. It is noted that all of Figs. 3A-10B are actually photographs of dried samples of the liquid droplets which are representative of the mixing of the liquids in the droplets. The photos are taken through a confocal microscope. The easiest way to observe and measure the mixture quality of the first and second liquids is to produce droplets through the claimed method and apparatus, dry the droplets, and put them on slides to be viewed under a microscope. The hydrophobic liquid is supplemented with a fluorochrome to highlight the mixture quality. Once the droplets were dried, the fluorochrome remained and was observable under a microscope. In each photographic example the “A” suffix figure is a photograph taken in normal light. Each “B” suffix photograph is taken with a fluorescence microscope after exciting the fluorochrome with the proper exciting wavelength so that the fluorochrome contained in the particles resulting from the first liquid is readily apparent. Actually the particles resulting from the first fluid containing the fluorochrome are so small that individual fluorescent dots are sometimes not very visible, and what is often seen in the “B” photographs is that the entire dried droplet appears fluorescent.

[0081] As shown in Figs. 3A and 3B, emulsifications can undesirably have a poor mixture of the two liquids. In this example the hydrophobic liquid to which flurochrome was added, shown as the darker portions of Fig. 3A, did not mix homogeneously with the hydrophilic liquid. This result is shown by the representative dried sample shown in Figs. 3A and 3B.

[0082] Turning now to Figs. 4A-10B, the results of various successful experiments with the currently disclosed apparatus and method are shown. These results indicate a substantially homogeneous mixture is achieved in each droplet. These homogeneous mixtures can be achieved without premixing of the two liquids prior to feeding the liquids through the currently disclosed apparatus. As is shown in the following examples the current disclosure allows for mixing of very viscous fluids including liquids up to about 300,000 cP. Such high viscosity liquids can be very difficult to mix by prior art procedures.

[0083] The experiments were performed with controlled variables and subsequent results as shown in the tables below. The following comments are generally applicable to the starting liquids that were mixed and dried to form the indirect indication of good results shown in Figs. 5A-7B (Tables 1-2). Many of the experiments included an aqueous polymeric solution and a hydrophobic solution to be mixed. As shown in Fig. 15, the resulting droplet of the mixing process and apparatus may include the aqueous polymeric solution surrounding the very small droplets of the hydrophobic solution. Depending on the case, Solution 1 could be both, a non-viscous oily flavor or a very viscous oily phase natural extract to which fluorochrome has been added. Solution 2 is an aqueous phase showing different examples for polymeric matrixes. The polymeric matrixes used in these examples are Iota Carrageenan (CGI), Shellac (Sh), Methyl Cellulose (MC), Sodium alginate (Algogel), Gelatin and Nutrateric. It would be very difficult to pre-prepare emulsions of these liquids due to their high viscosities.

[0084] The ṁ of gas/ṁ of liquids data shows the ratio between the mass flow rate of the gas and the mass flow rate of the liquid (sum of liquids). The "Entrapment" data shows the amount of ingredient (solution 1 here; flavor or natural extract) in the dried sample. The "encapsulation efficiency" data shows the real amount of ingredient introduced in the dried sample times 100 divided by the theoretical amount of ingredient to be introduced into the dried sample. The different experimental conditions illustrate different flow rate and air pressure combinations.

[0085] Table 1 is shown below:

TABLE 1

Figure(s)	5A and 5B	6A and 6B	7A and 7B
Nozzle	2 Entrances, single tube Exit orifice D2: 700μm	2 Entrances, single tube Exit orifice D2: 700μm	2 Entrances, single tube Exit orifice D2: 700μm
Solution 1	Flavour	Natural extract	Natural extract
Viscosity 1 (cP)	1.15	7160	7160
Density 1 (Kg/m^3)	846	1206	1206
Flow rate 1 (mL/min)	0.09	0.15	0.15
Solution 2 % (w/w)	Nutrateric: 8% CGI: 1% MC: 4%	Gelatin: 10% CGI: 5% Algogel: 3%	Gelatin: 7% CGI: 3.5% Algogel: 2.1%
Density 2 (Kg/m^3)	1080	1010	1010
Viscosity 2 (cP)	9024	8520	45000
Flow rate 2 (mL/min)	3	3	3
P (mbar)	4000	1500	1500
ṁ of gas/ṁ of liquids	5.7	3.53	3.52
Particle size (μm)	1-19	4 - 34	2 - 20
Entrapment (%)	15%	22%	27%
Encapsulation Efficiency (%)	100%	100%	95%

[0086] Additional experimental test parameters and results are shown in Table 2. No photographic results are shown for these tests. Table 2 is shown below:

TABLE 2

Nozzle	2 Entrances, single tube Exit orifice D2: 700μm	2 Entrances, single tube Exit orifice D2: 700μm	2 Entrances, single tube Exit orifice D2: 700μm
Solution 1	Natural extract	Natural extract	Flavor
Viscosity 1 (cP)	7160	7160	1.15
Density 1 (Kg/m^3)	1206	1206	846
Flow rate 1 (mL/min)	0.05	0.133	0.22
Solution 2 % (w/w)	Nutrateric: 6% CGI: 1.5% MC: 5%	Gelatin: 8.5% MC: 4% Sh: 5% CGI: 0.3%	Nutrateric: 8% CGI: 1% MC: 4%
Density 2 (Kg/m^3)	1010	1010	1080
Viscosity 2 (cP)	52280	--	9024
Flow rate 2 (mL/min)	3	5.3	5.3
P (mbar)	1500	2000	2000
ṁ of gas/ṁ of liquids	3.52	2.37	2.5
Particle size (μm)	--	1 - 24	1-21
Entrapment (%)	11.2%	14%	20%
Encapsulation Efficiency (%)	100.0%	100%	100%

[0087] The following comments are generally applicable to the results shown in Figs. 8A-10B (Table 3). In each of these tests a concentric tube arrangement as shown in Fig. 2 is utilized.

[0088] Solution 1 is a relatively low viscosity liquid which is able to be introduced through the inner tube **124**. In these examples it is a flavor or an organic oil to which the fluorochrome has been added. Solution 2 is an aqueous phase, for which different examples are shown for polymeric matrixes. The polymeric matrixes used in these examples are Iota Carrageenan (CGI), Shellac (Sh), Methyl Cellulose (MC), Maltodextrin (MD), Nutriose, and modified Starch (OSS).

[0089] Table 3 is shown below:

TABLE 3

Figure(s)	8A and 8B	9A and 9B	10A and 10B

Nozzle	Concentric tubes Exit orifice D2: 700μm	Concentric tubes Exit orifice D2: 700μm	Concentric tubes Exit orifice D2: 700μm
Solution 1	Flavor	Organic Oil	Flavor
Viscosity 1 (cP)	1.15	4.6	1.15
Density 1 (Kg/m^3)	846	843	846
Flow rate 1 (mL/min)	1	0.55	0.25
Solution 2 % (w/w)	Sh: 5% MC: 5% MD: 30%	MD: 56%	Nutriose: 10% OSS: 10% CGI: 1%
Viscosity 2 (cP)	145	310	12640
Concentration Solution 2 (%)	40	56	21
Density 2 (Kg/m^3)	1150	1270	1090
Flow rate 2 (mL/min)	3	3	5.3
P (mbar)	2000	3500	2000
ṁ of gas/ṁ of liquids	3.53	4.82	1.77
Particle size (μm)	1 - 10.65	1 - 8.03	1 - 10
Entrapment (%)	37%	17%	13.00%
Encapsulation Efficiency (%)	98%	96%	88%

[0090] Additional experimental test parameters and results are shown in Tables 4-5. The polymeric matrixes used in these examples are Iota Carrageenan (CGI), Shellac (Sh), Methyl Cellulose (MC), Maltodextrin (MD), Nutrateric, Xanthan gum (Xan) and Hydroxypropylcellulose (KEF). No photographic results are shown for these tests. Table 4 is shown below:

TABLE 4

Nozzle	Concentric tubes Exit orifice D2: 700μm	Concentric tubes Exit orifice D2: 700μm	Concentric tubes Exit orifice D2: 700μm
Solution 1	Flavor	Flavor	Flavor
Viscosity 1 (cP)	1.15	1.15	1.15
Density 1 (Kg/m^3)	846	846	846
Flow rate 1 (mL/min)	0.8	0.5	0.09

Solution 2 % (w/w)	Sh: 5% KEF: 5% MD: 30%	Sh: 5% KEF: 5% MD: 30%	Nutrateric: 8% CGI: 1% MC: 4%
Viscosity 2 (cP)	64	64	2276
Concentration Solution 2 (%)	40	40	15.75
Density 2 (Kg/m^3)	1140	1140	1080
Flow rate 2 (mL/min)	6	4	3
P (mbar)	2000	3500	2000
ṁ of gas/ṁ of liquids	2.52	4.01	3.25
Particle size (μm)	1 – 14.3	1 – 6.49	--
Entrapment (%)	20.72%	19%	15.00%
Encapsulation Efficiency (%)	100%	100%	96%

[0091] Table 5 is shown below:

TABLE 5

Nozzle	Concentric tubes Exit orifice D2: 700μm	Concentric tubes Exit orifice D2: 700μm
Solution 1	Flavor	Flavor
Viscosity 1 (cP)	1.15	1.15
Density 1 (Kg/m^3)	846	846
Flow rate 1 (mL/min)	0.140	0.260
Solution 2	MD: 15% XAN: 0.75%	MD: 15% XAN: 0.75%
Viscosity 2 (cP)	37400	37400
Concentration Solution 2 (%)	21	15.75
Density 2 (Kg/m^3)	--	--
Flow rate 2 (mL/min)	3	5.3
P (mbar)	2000	2000
ṁ of gas/ṁ of liquids	3.46	1.95
Particle size (μm)	1 – 17	1 – 25
Entrapment (%)	19.00%	22.00%

Encapsulation Efficiency (%)	93%	100%

[0092] Figs. 11A-14 (Tables 6-9) show some examples of how droplet size distribution behaves. The droplet size was measured with a Sympatec HELOS laser diffraction sensor. Dv10 indicates the maximum particle diameter below which 10% of the sample volume exists. Dv50 indicates the maximum particle diameter below which 50% of the sample volume exists. Dv90 indicates the maximum particle diameter below which 90% of the sample volume exists. The polymeric matrixes used in these examples are Gelatin, Iota Carrageenan (CGI), Shellac (Sh), Methyl Cellulose (MC), Arabic Gum (GA), Starch (Clariaplus), modified Starch (OSS), Sodium alginate (Algogel) and Nutriose. Note that Figs. 11A and 11B (Table 6) show the effect of different viscosities of the aqueous matrix for similar experimental conditions. Figs. 12A and 12B (in Table 7) show the effect of different viscosities of the aqueous matrix for another set of experimental conditions.

[0093] Table 6 is shown below:

TABLE 6

Figure(s)	11A	11B
Nozzle	2 Entrances, single tube Exit orifice D2: 700μm	2 Entrances, single tube Exit orifice D2: 700μm
Solution 1	Natural extract	Natural extract
Viscosity 1 (cP)	296000	296000
Density 1 (Kg/m^3)	1408	1408
Flow rate 1 (mL/min)	0.5	0.5
Solution 2 % (w/w)	Gelatin: 2% CGI: 0.5% Clariaplus: 5%	Sh: 6% MC: 2% GA: 2% CGI: 0.4% Algogel: 0.4%
Viscosity 2 (cP)	2.95	274
Density 2 (Kg/m3)	1019	1020
Qext (mL/min)	3	3
P (mbar)	1500	1500
ṁ of gas/ṁ of liquids	2.86	2.83

Dv10 (μm)	1.98	2.12
Dv50 (μm)	7.75	8
Dv90 (μm)	63.36	42.68

[0094] Table 7 is shown below:

TABLE 7

Figure(s)	12A	12B
Nozzle	2 Entrances, single tube Exit orifice D2: 700μm	2 Entrances, single tube Exit orifice D2: 700μm
Solution 1	Natural extract	Natural extract
Viscosity 1 (cP)	296000	296000
Density 1 (Kg/m^3)	1408	1408
Flow rate 1 (mL/min)	0.7	0.7
Solution 2	Gelatin: 2% CGI: 0.5% Clariaplus: 5%	Sh: 6% MC: 2% GA: 2% CGI: 0.4% Algogel: 0.4%
Viscosity 2 (cP)	2.95	274
Density 2 (Kg/m^3)	1019	1020
Flow rate 2 (mL/min)	3	3
P (mbar)	1500	1500
ṁ of gas/ṁ of liquids	2.64	2.63
Dv10 (μm)	2.07	2.17
Dv50 (μm)	8.23	8.23
Dv90 (μm)	57.22	45.99

[0095] Figs. 13A-13C (Table 8) can be compared to show the influence of pressure on droplet size. Table 8 is shown below:

TABLE 8

Figure(s)	13A	13B	13C
Nozzle	Concentric tubes Exit orifice D2: 700μm	Concentric tubes Exit orifice D2: 700μm	Concentric tubes Exit orifice D2: 700μm
Solution 1	Organic oil	Organic oil	Organic oil
Viscosity 1 (cP)	4.6	4.6	4.6
Density 1 (Kg/m^3)	841	841	841
Flow rate 1 (mL/min)	0.4	0.4	0.4
Solution 2 % (w/w)	Nutriose: 10% OSS: 10% CGI: 0.75%	Nutriose: 10% OSS: 10% CGI: 0.75%	Nutriose: 10% OSS: 10% CGI: 0.75%
Viscosity 2 (cP)	1035	1035	1035
Density 2 (Kg/m^3)	1090	1090	1090
Flow rate 2 (mL/min)	5	5	5
P (mbar)	2000	3000	4000
$\dot{m}$ of gas/$\dot{m}$ of liquids	1.90	2.66	3.36
Dv10 (μm)	2.11	1.79	1.45
Dv50 (μm)	7.92	6.72	5.55
Dv90 (μm)	33.5	23.10	17.99

[0096] Table 9 is shown below:

TABLE 9

Figure(s)	10A, 10B, and 14
Nozzle	Concentric tubes Exit orifice D2: 700μm
Solution 1	Flavor
Viscosity 1 (cP)	1.15
Density 1 (Kg/m^3)	846
Flow rate 1 (mL/min)	0.25
Solution 2	Nutriose: 10% OSS: 10% CGI: 1%
Viscosity 2 (cP)	12640
Density 2 (Kg/m^3)	1087
Flow rate (mL/min)	5.3
P (mbar)	2000
$\dot{m}$ of gas/$\dot{m}$ of	1.77

liquids	
Dv10 (μm)	3.78
Dv50 (μm)	12.93
Dv90 (μm)	81.71

[0097] The present disclosure also relates to the introduction of incompatible reagents to each other such that each droplet is a microreactor. One embodiment includes the one step production of zinc carbonate crystals using two solutions that are very reactive when both contact each other. Both solutions may be nebulized using a single tube with two entrances. Air may be the pressuring gas. The droplets are obtained in air and may be recovered in a bath. As the droplets fly through the air, the mixture of the salts in the droplets start reacting and producing the zinc carbonate. In the recovery bath a suspension of $ZnCO_3$ is recovered. These suspension results are not to be confused with the dried product described in the previous Figures and Tables. Experimental results of the suspension are shown in Table 10 below:

TABLE 10

Nozzle	2 Entrances, single tube Exit orifice D2: 700μm	2 Entrances, single tube Exit orifice D2: 700μm	2 Entrances, single tube Exit orifice D2: 700μm
Solution 1	$Zn(NO_3)_2$ (15%) + $Mg(NO_3)_2$ (3%)	$Zn(NO_3)_2$ (15%) + $Mg(NO_3)_2$ (3%)	$Zn(NO_3)_2$ (15%) + $Mg(NO_3)_2$ (3%)
Flow Rate 1 (mL/min)	1	1	1
Solution 2	Na_2CO_3 (15%)	Na2CO3 (15%)	Na2CO3 (15%)
Flow rate 2 (mL/min)	1	1	1
P (mbar)	2000	3000	1500
Recovery bath	Water	Water	PVA 1%
Solid particle size Dv50 (μm)	12.8	13.8	10.5
Solid particle size Dv90 (μm)	25.9	27.1	24.2
Span (Malvern 2000)	1.6	1.6	1.9

[0098] Table 10 is just one example of potential results obtained by the current disclosure. The mixed first and second liquids may form droplets, but they may also be converted into powder, granular particles, dust, and the like through any known treatment methods such as that described above with regard to Table 10.

[0099] This written description uses examples to disclose the invention and also to enable any person skilled in the art to practice the invention, including making and using any devices or systems. The patentable scope of the invention is defined by the claims, and may include other examples that occur to those skilled in the art. Such other examples are intended to be within the scope of the claims if they have structural elements that do not differ from the literal language of the claims, or if they include equivalent structural elements with insubstantial differences from the literal language of the claims.

[0100] Although embodiments of the disclosure have been described using specific terms, such description is for illustrative purposes only. The words used are words of description rather than limitation. It is to be understood that changes and variations may be made by those of ordinary skill in the art without departing from the spirit or the scope of the present disclosure, which is set forth in the following claims. In addition, it should be understood that aspects of the various embodiments may be interchanged in whole or in part. While specific uses for the subject matter of the disclosure have been exemplified, other uses are contemplated. Therefore, the spirit and scope of the appended claims should not be limited to the description of the versions contained herein.

CLAIMS

1. A method of mixing at least two liquids, the method comprising:

(a) providing a nozzle including a feeding tube having a feeding tube outlet, the feeding tube positioned in a pressure chamber containing a pressurized gas, the pressure chamber including a pressure chamber exit orifice substantially coaxial with the feeding tube outlet downstream of the feeding tube outlet, the feeding tube outlet being axially offset from the pressure chamber exit orifice by an axial gap;

(b) introducing a first stream of a first liquid and a second stream of a second liquid into the feeding tube upstream from the feeding tube outlet; and

(c) operating the nozzle in a flow blurring mode such that a reflux cell is formed in the feeding tube upstream from the feeding tube outlet, thereby providing a turbulent mixing of the first and second liquids in the reflux cell.

2. A method according to claim 1, characterized in that it further comprises:

ejecting a mixture of the first and second liquids out of the pressure chamber exit orifice.

3. A method according to any one of the precedent claims , characterized in that:

the mixture is in a form of droplets entrained in the gas.

4. A method according to any one of the precedent claims, characterized in that:

each droplet includes an emulsion of the first and second liquids.

5. A method according to any one of the precedent claims, characterized in that it further comprises:

drying the droplets to create dried mixture particles.

6. A method according to claim 5, characterized in that at least one of the first and second liquids includes solids, and the dried mixture particles include the solids.

7. A method according to any of the precedent claims, characterized in that step (b) further comprises:

forming an emulsion of the first and second liquids in the reflux cell; and

breaking the emulsion into droplets as the emulsion is ejected out of the pressure chamber exit orifice.

8. A method accoding to any of the precedent claims, characterized in that it further comprises:

the first liquid and the second liquid including incompatible reagents;

step (c) further including:

(c)(1) forming a mixture of the first and second liquids in the reflux cell; and

(c)(2) breaking the mixture into droplets as the mixture is ejected out of the pressure chamber exit orifice; and

reacting the incompatible reagents with each other inside each droplet so that each droplet acts as a microreactor.

9. A method according to any of the preceding claims, characterized in that:

each of the first and second liquids is introduced into the feeding tube at separate inlets upstream of the reflux cell.

10. A method according to any of the preceding claims, characterized in that:

the feeding tube is an outer feeding tube, and the feeding tube outlet is an outer feeding tube outlet; and

step (b) further comprises introducing the first liquid into the outer feeding tube via a concentric inner feeding tube having an inner feeding tube outlet, and introducing the second liquid into the outer feeding tube at a second inlet upstream of the inner feeding tube outlet.

11. A method according to claim 10, characterized in that:

the inner feeding tube outlet is located upstream of the outer feeding tube outlet by a recess distance.

12. A method according to any of the claims 10-11, characterized in that:

the recess distance is configured to allow the reflux cell to form uninterrupted by the inner feeding tube.

13. A method according to any of the claims 10-12, characterized in that the recess distance is at least as great as an outlet diameter of the outer feeding tube outlet.

14. A method according to any of the claims 10-13, characterized in that:

in step (c) the reflux cell is formed in the outer feeding tube but not in the inner feeding tube.

15. A method according to any of the preceding claims, characterized in that:

step (b) further includes introducing a third stream of a third liquid into the feeding tube upstream from the feeding tube outlet.

16. An apparatus, characterized in that said apparatus comprises:

a first inlet for receiving a first liquid stream;

a second inlet for receiving a second liquid stream;

an outer feed tube configured to receive the first and second liquid streams and having an outer feed tube axis, the outer feed tube including an outer feed tube outlet having an outer feed tube outlet diameter;

a pressure chamber surrounding the outer feed tube outlet for containing a pressurized gas, the pressure chamber including a pressure chamber exit orifice, the pressure chamber exit orifice being coaxial with the outer feed tube axis and having an exit orifice diameter, the pressure chamber exit orifice being spaced from the outer feed tube outlet to define an axial gap between the pressure chamber exit orifice and the outer feed tube outlet; and

wherein the outer feed tube outlet diameter, the exit orifice diameter, and the axial gap are configured such that a reflux cell can form inside the outer feed tube when the first and second liquid streams are forced through the outer feed tube and the gas is forced through the axial gap.

17. An apparatus according to claim 16, characterized in that it further comprises:

an inner feed tube concentrically disposed in the outer feed tube, the first inlet being communicated with the inner feed tube.

18. An apparatus according to claim 17, characterized in that:

the inner feed tube includes an inner feed tube outlet, the inner feed tube outlet being recessed from the outer feed tube outlet by a recess distance.

19. An apparatus according to any of the claims 17- 18, characterized in that:

the recess distance is configured to allow the reflux cell to form uninterrupted by the inner feed tube.

20. An apparatus according to any of the claims 17-19, characterized in that:
the recess distance is at least as great as the outer feed tube outlet diameter.

21. An apparatus according to any of the claims 17-20, characterized in that:
the recess distance is sufficient that the reflux cell forms in the outer feed tube but not in the inner feed tube during use.

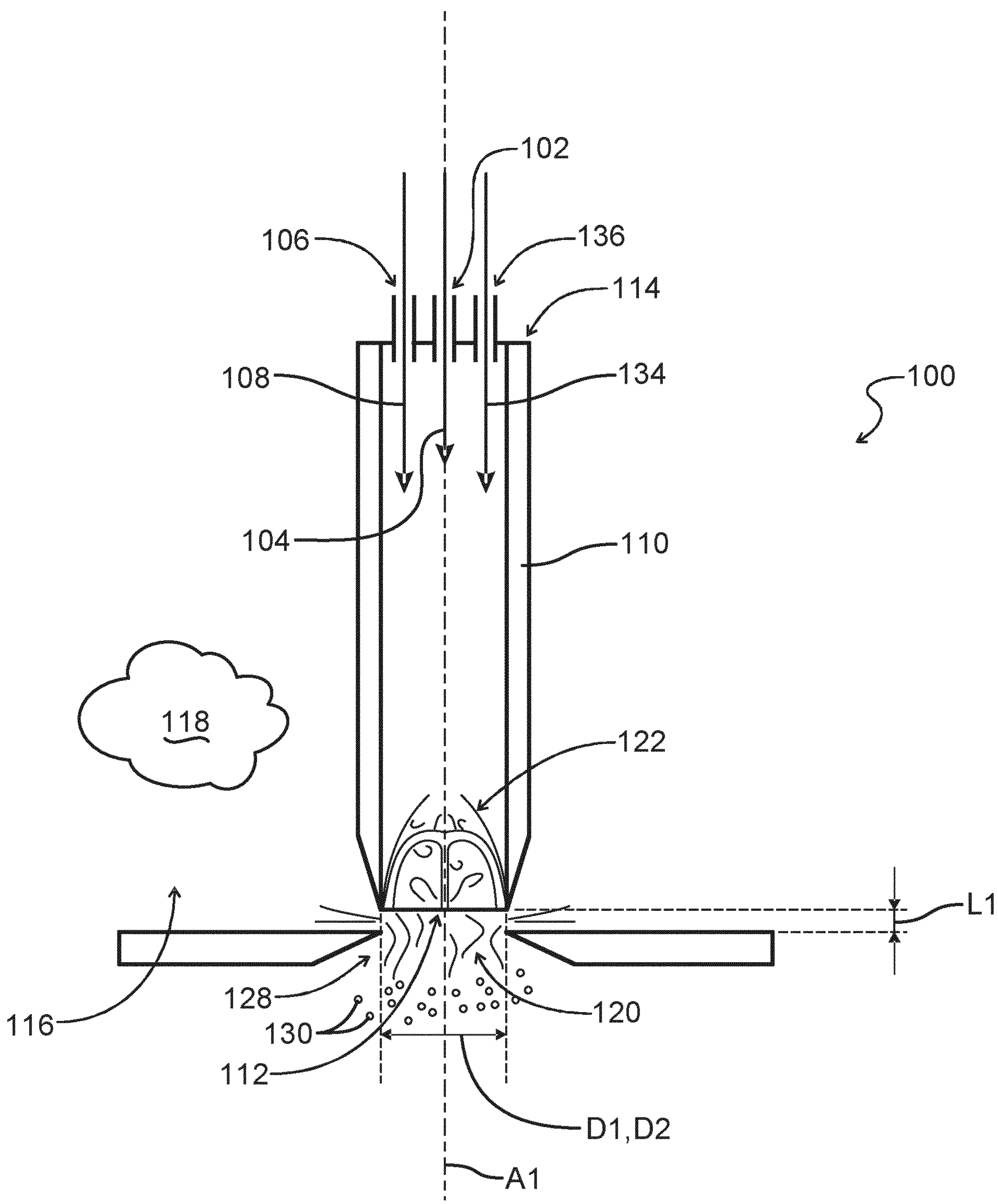
22. An apparatus according to any of the claims 16-21, characterized in that:
the exit orifice diameter is in a range of from 90 to 120 percent of the outer feed tube outlet diameter.

23. An apparatus according to any of the claims 16-22, characterized in that:
the exit orifice diameter is substantially equal to the outer feed tube outlet diameter.

24. An apparatus according to any of the claims 16-23, characterized in that:
the outer feed tube outlet diameter is in a range of from 70 microns to 8 millimeters more preferably from 200 to 5000 microns, more preferably 500 to 2000 microns..

25. An apparatus according to any of the claims 16-24, characterized in that:
the axial gap is no greater than one half of the exit orifice diameter, preferably no greater than one fourth of the exit orifice diameter.

26. An apparatus according to any of the claims 16-25, characterized in that:
the outer feed tube includes an inlet end opposite from the outer feed tube outlet, and both of the first and second inlets are communicated with the outer feed tube closer to the inlet end than to the outer feed tube outlet.


102
106
136
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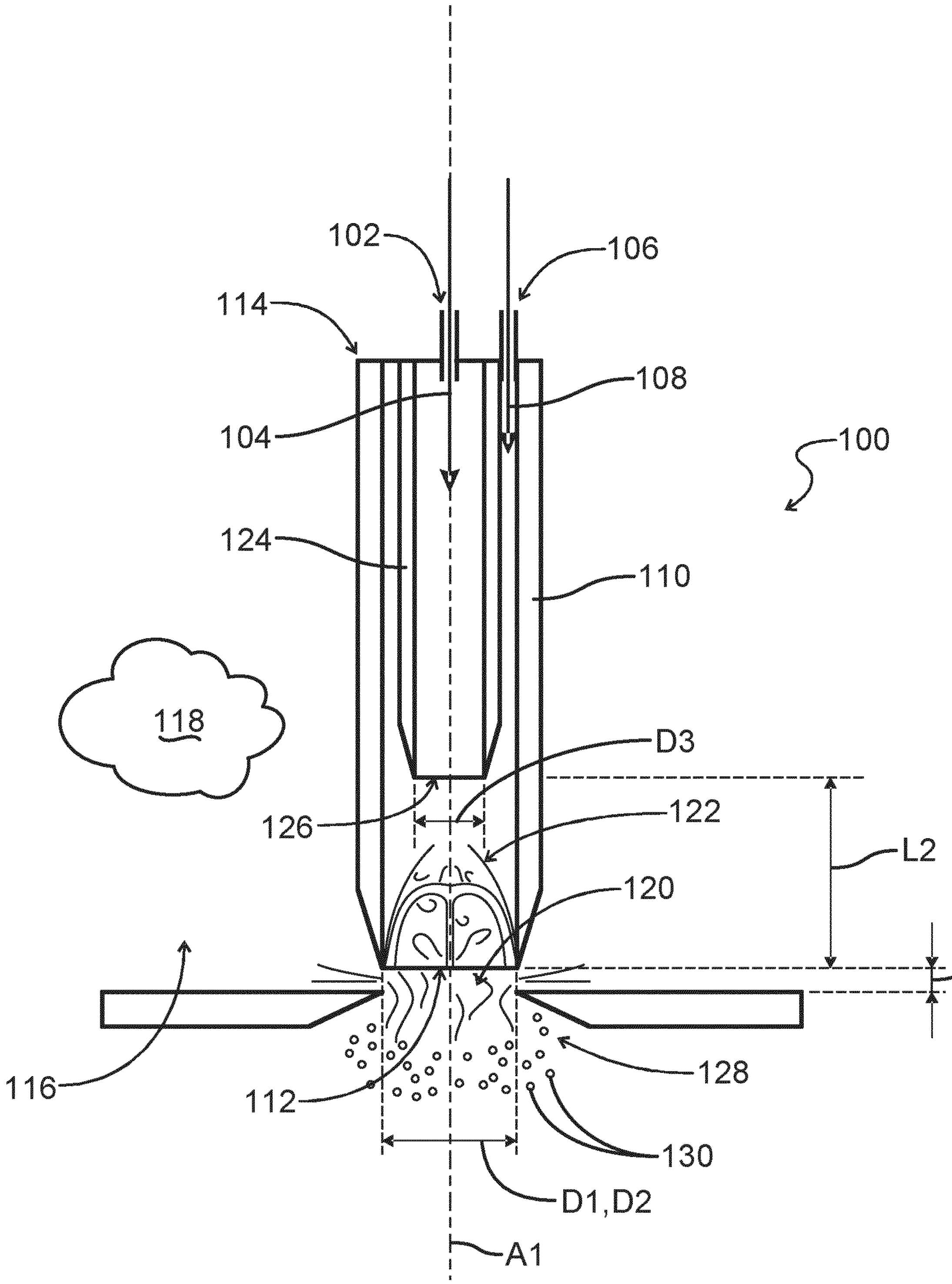
D1,D2
A1

FIG. 1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A1

FIG. 2

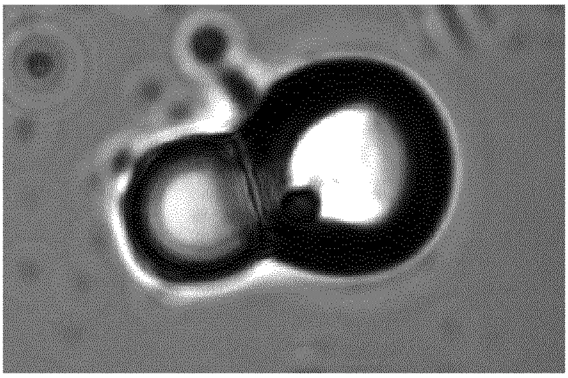


FIG. 3A

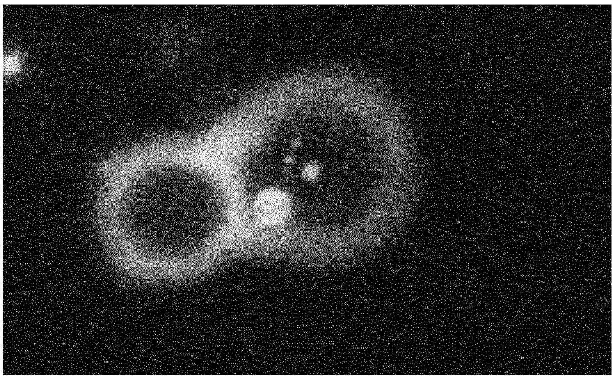


FIG. 3B

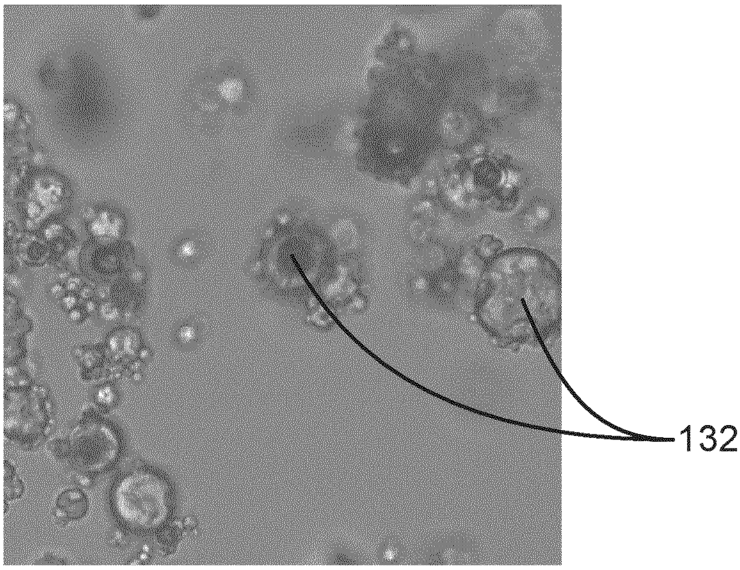


FIG. 4A

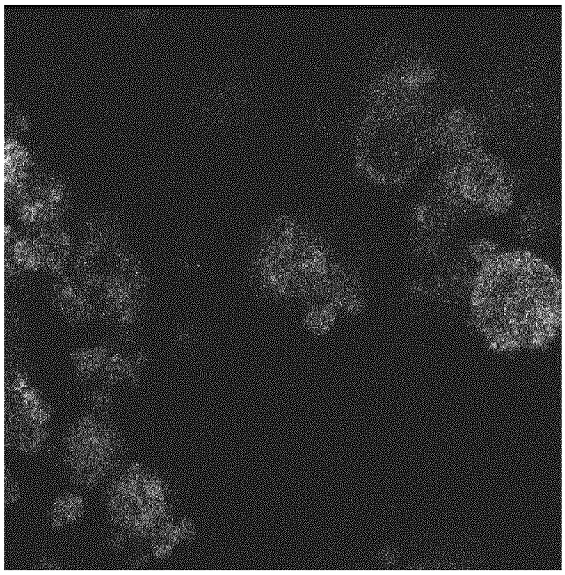


FIG. 4B

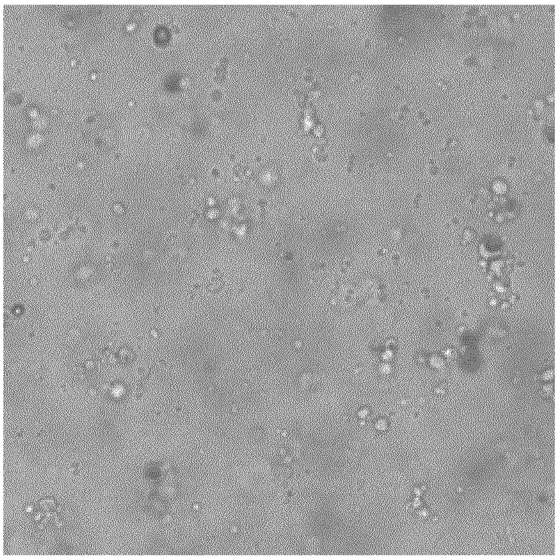


FIG. 5A

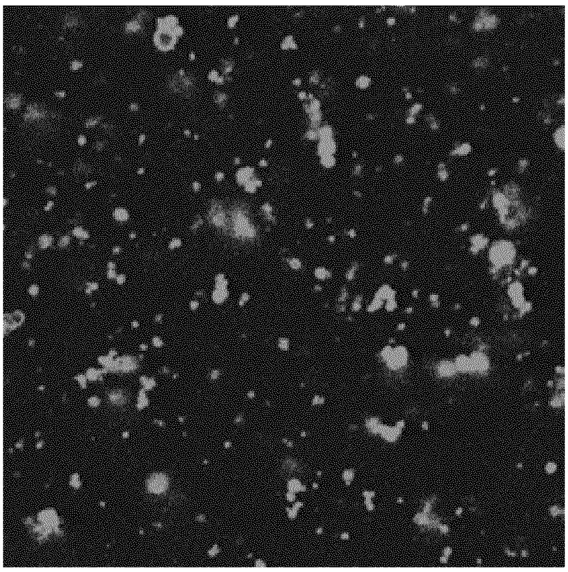


FIG. 5B

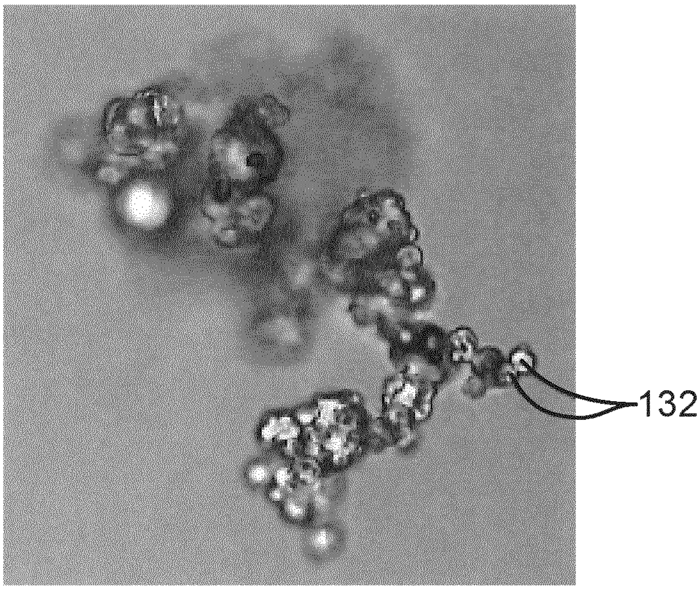


FIG. 6A

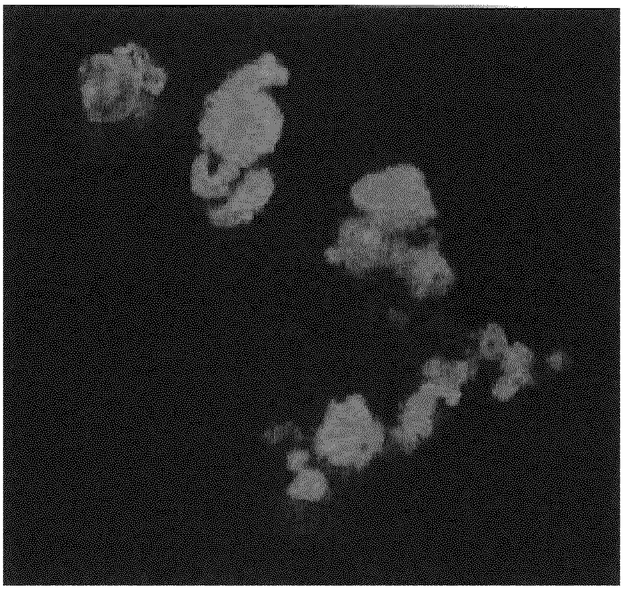


FIG. 6B

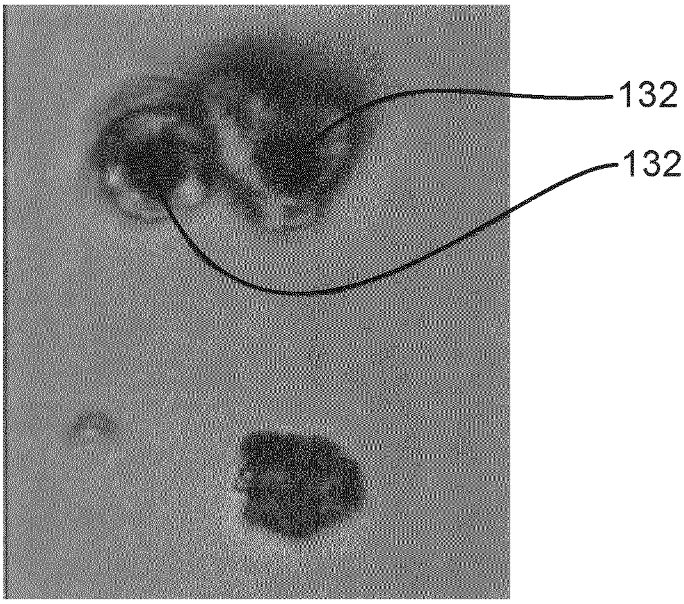


FIG. 7A

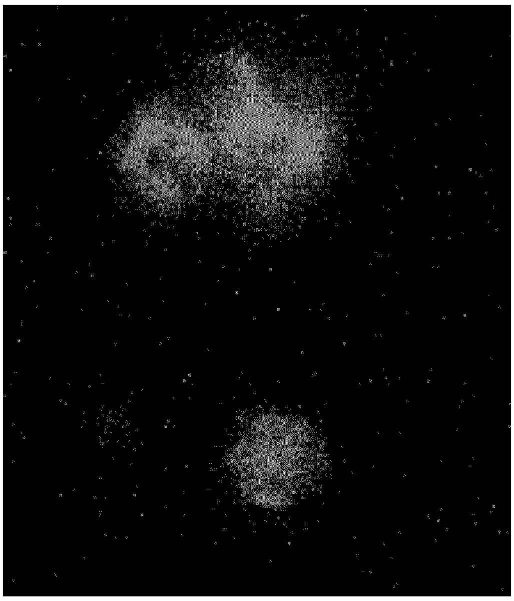


FIG. 7B

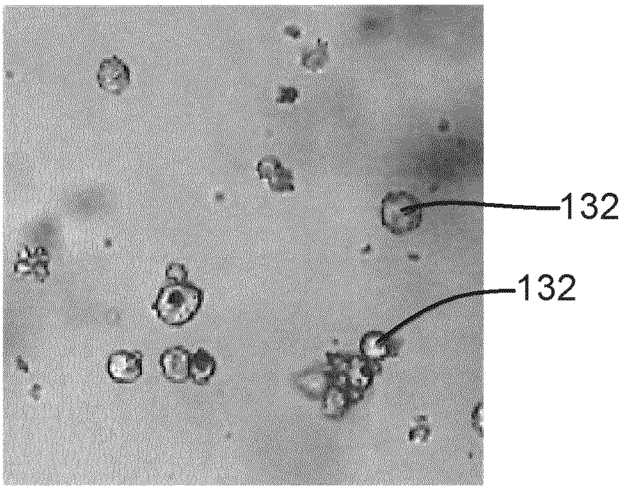


FIG. 8A

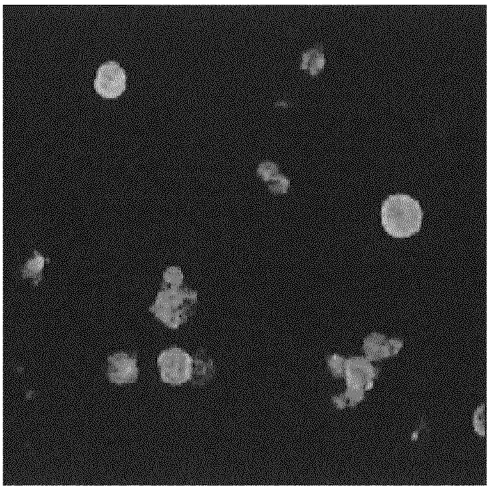


FIG. 8B

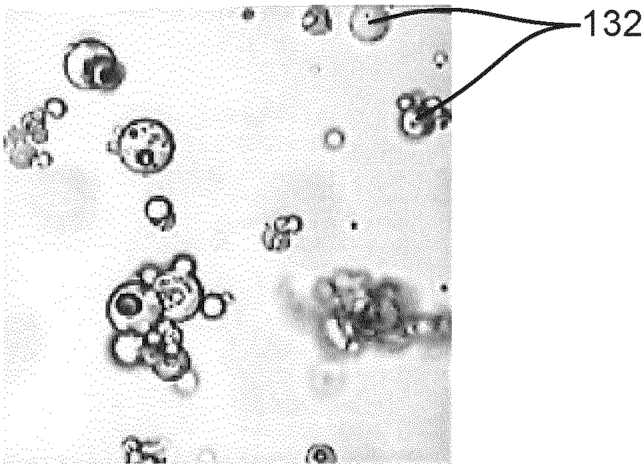


FIG. 9A

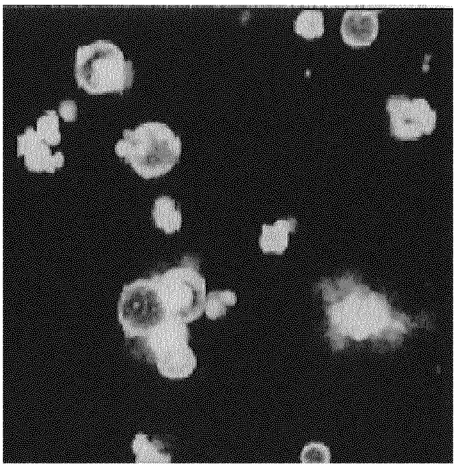


FIG. 9B

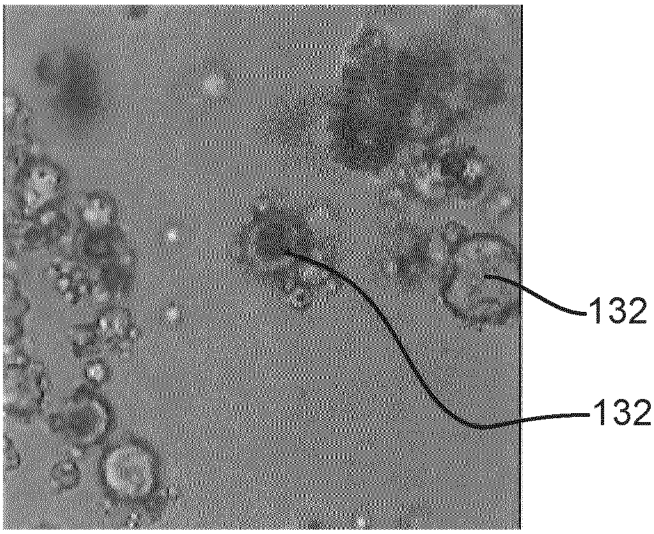


FIG. 10A

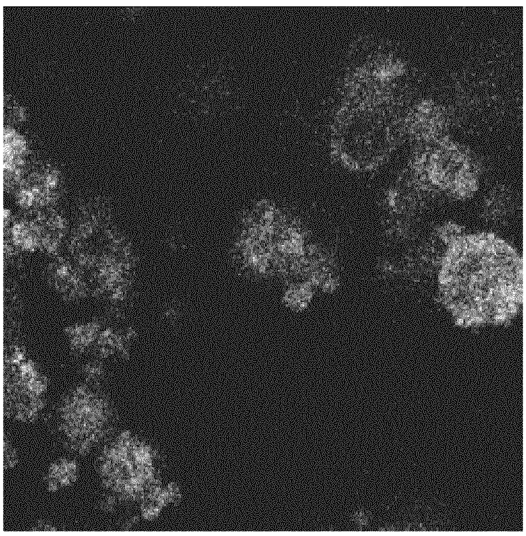
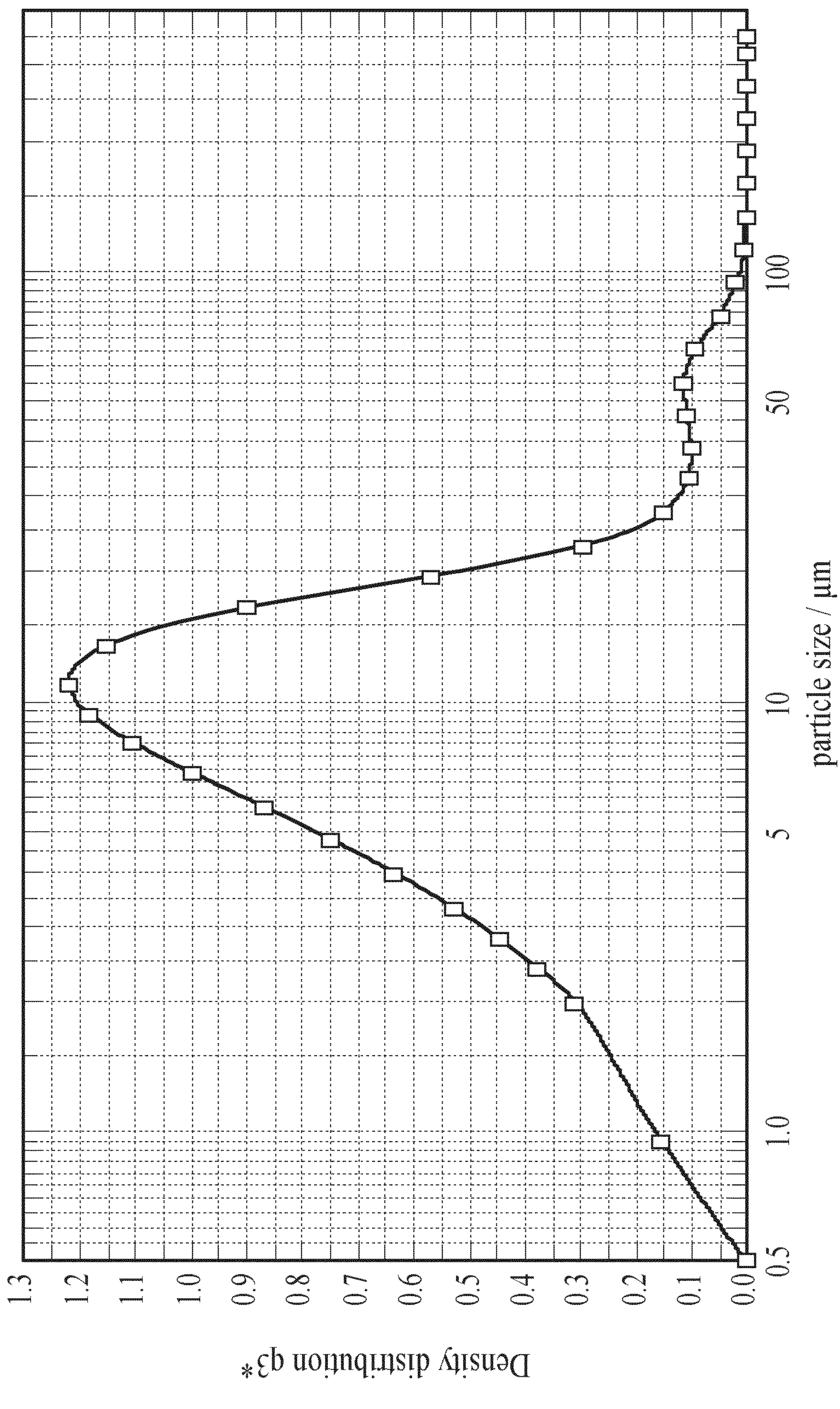


FIG. 10B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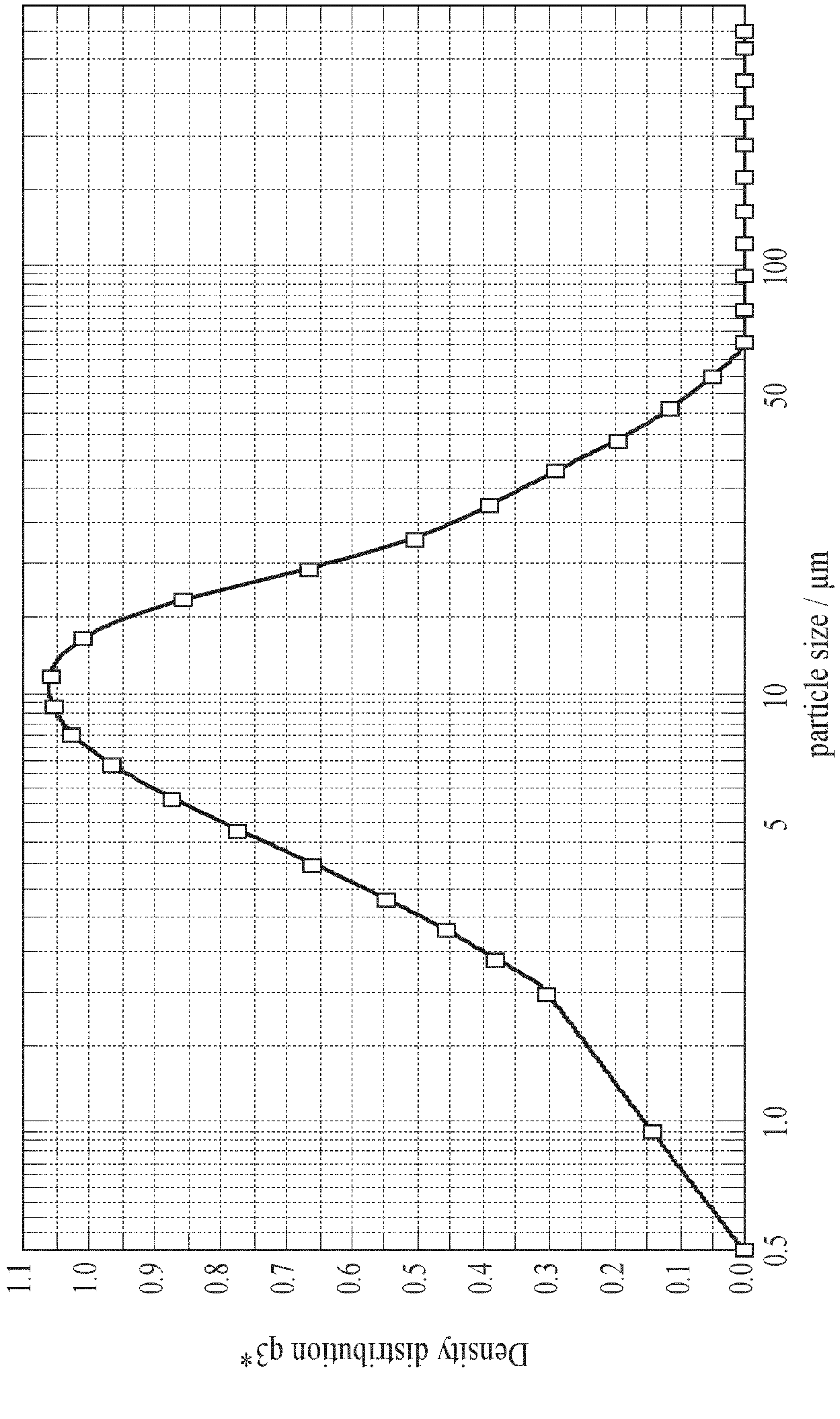
0.2
0.3
0.4
0.5
0.6
0.7
0.8
0.9
1.0
1.1
1.2
1.3
0.5
1.0
5
10
50
100
particle size / µm

FIG. 11A


Density distribution q3*
particle size / µm
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0.2
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0.4
0.5
0.6
0.7
0.8
0.9
1.0
1.1
0.5
1.0
5
10
50
100

FIG. 11B

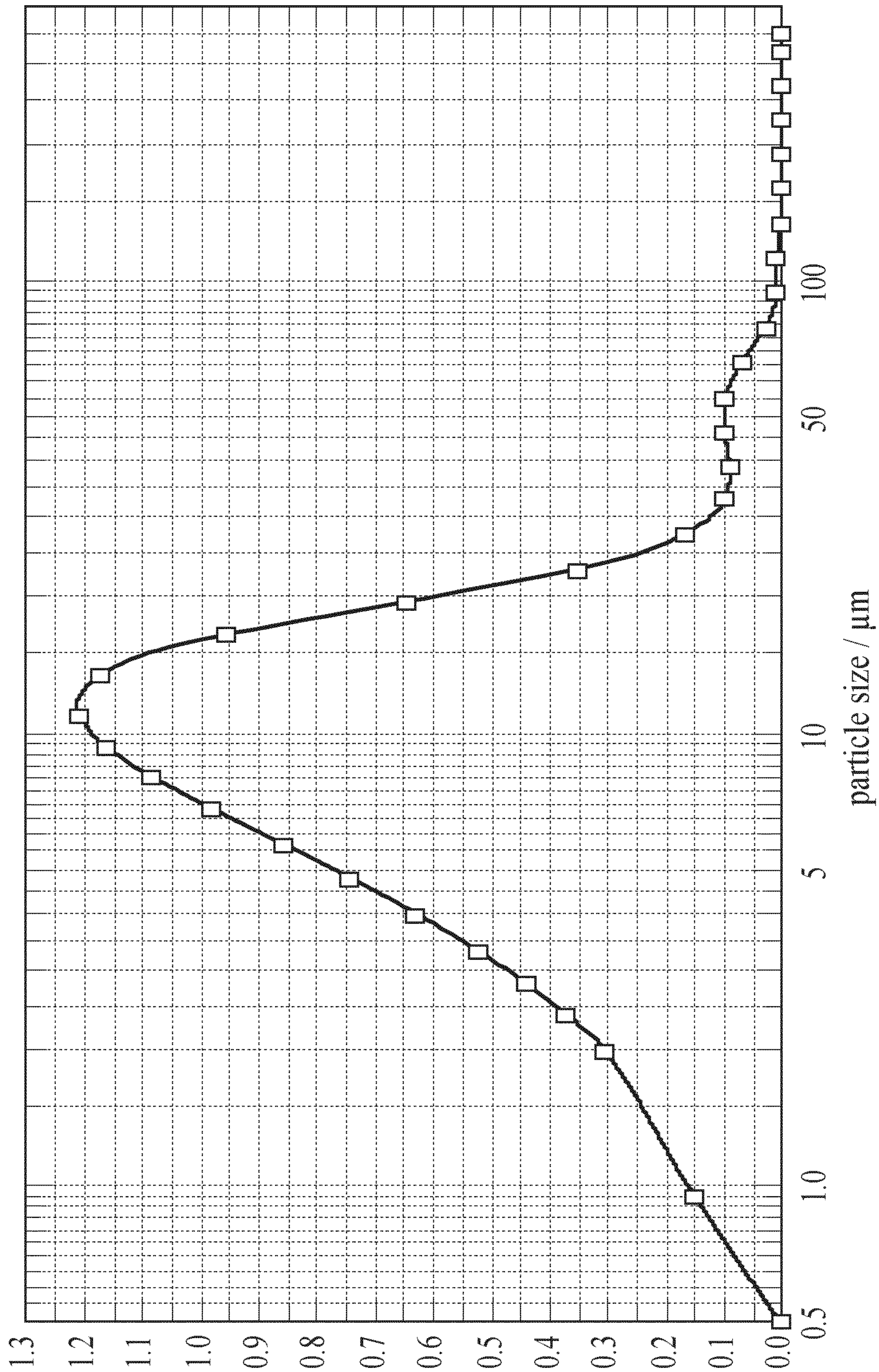
Density distribution q3*
0.00
0.1
0.2
0.3
0.4
0.5
0.6
0.7
0.8
0.9
1.0
1.1
1.2
1.3
0.5
1.0
5
10
50
100
particle size / µm

FIG. 12A

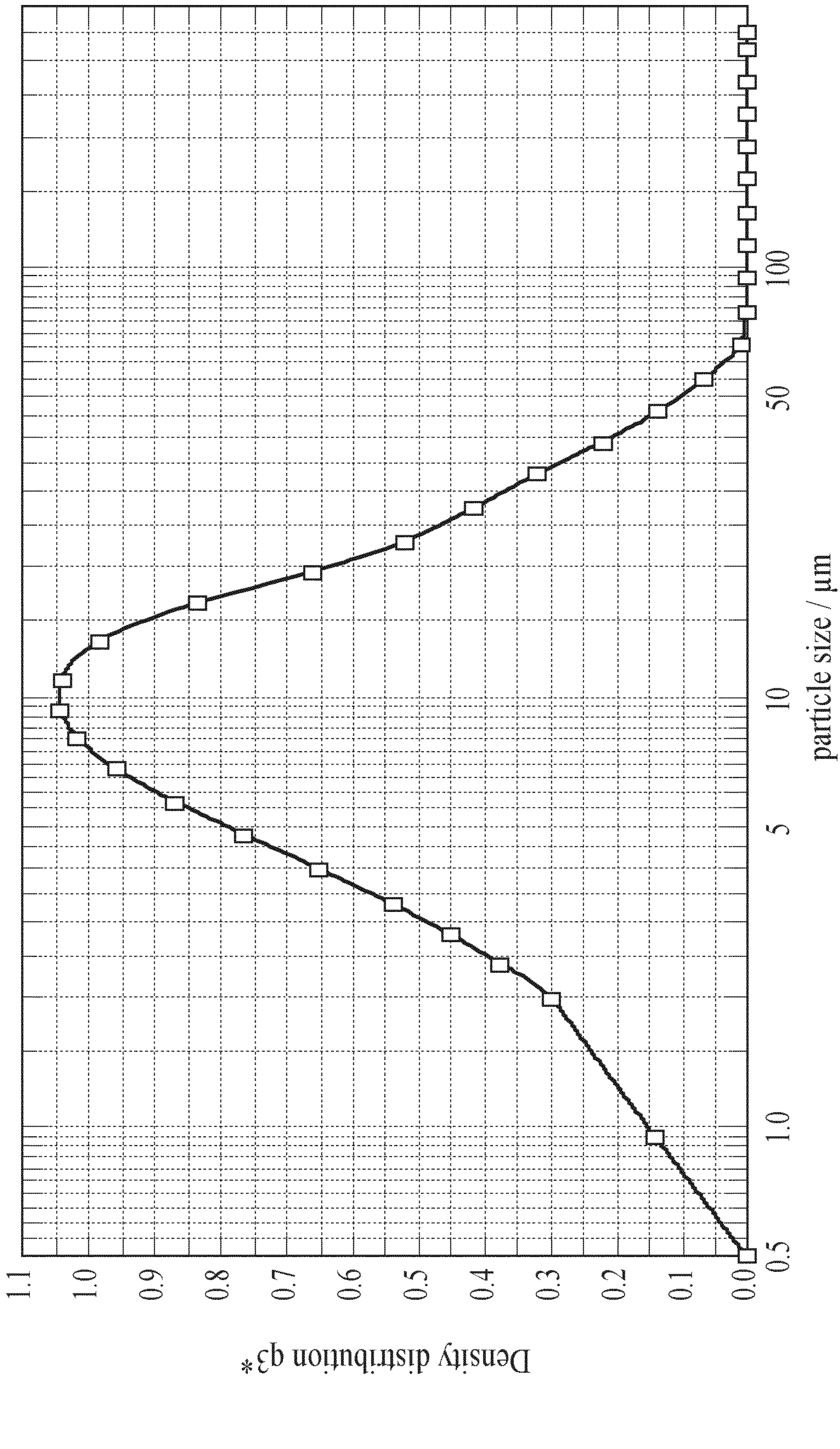
Density distribution q3*
1.1
1.0
0.9
0.8
0.7
0.6
0.5
0.4
0.3
0.2
0.1
0.00
0.5
1.0
5
10
50
100
particle size / µm

FIG. 12B

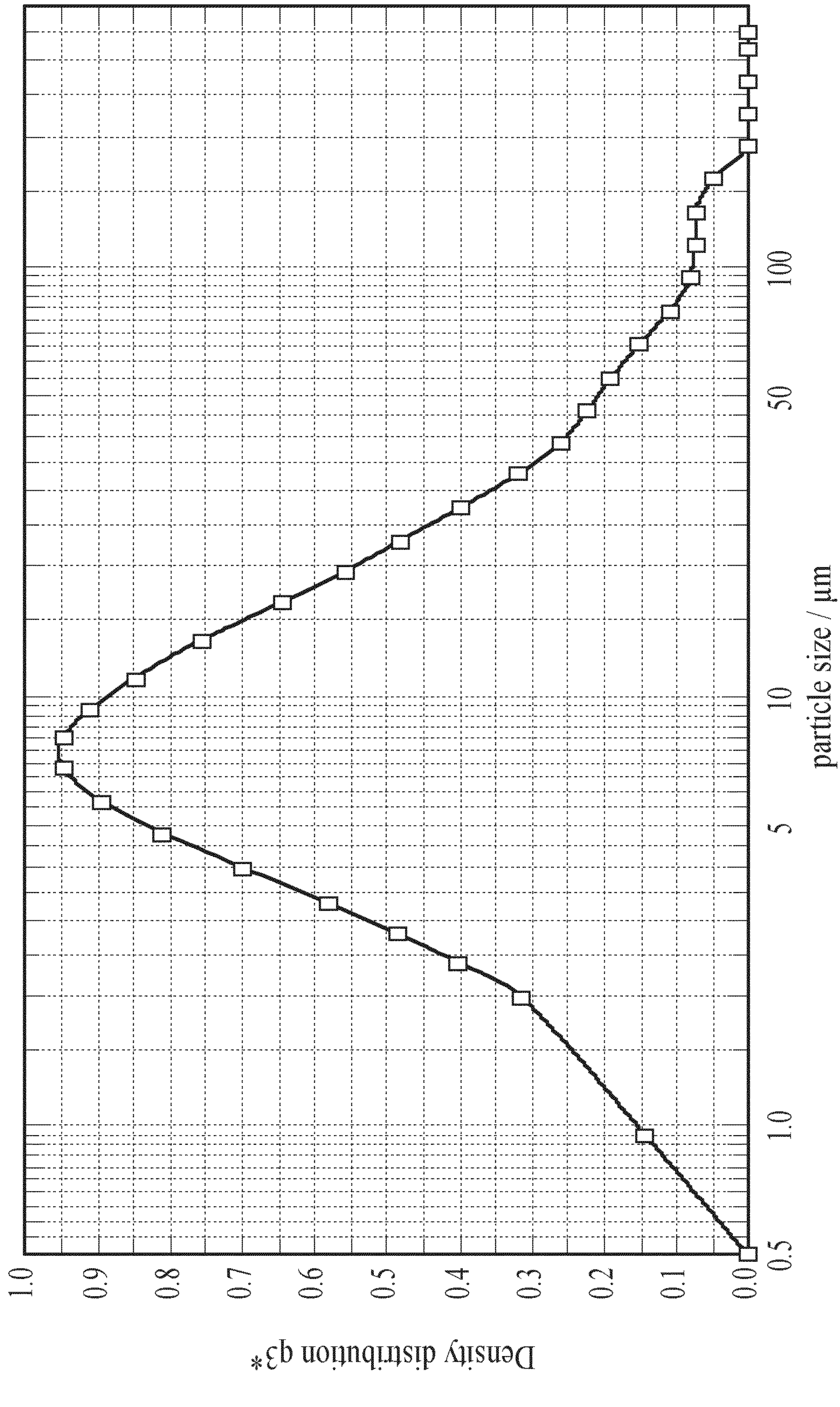
Density distribution q3*
0.0
0.1
0.2
0.3
0.4
0.5
0.6
0.7
0.8
0.9
1.0
0.5
1.0
5
10
50
100
particle size / µm

FIG. 13A

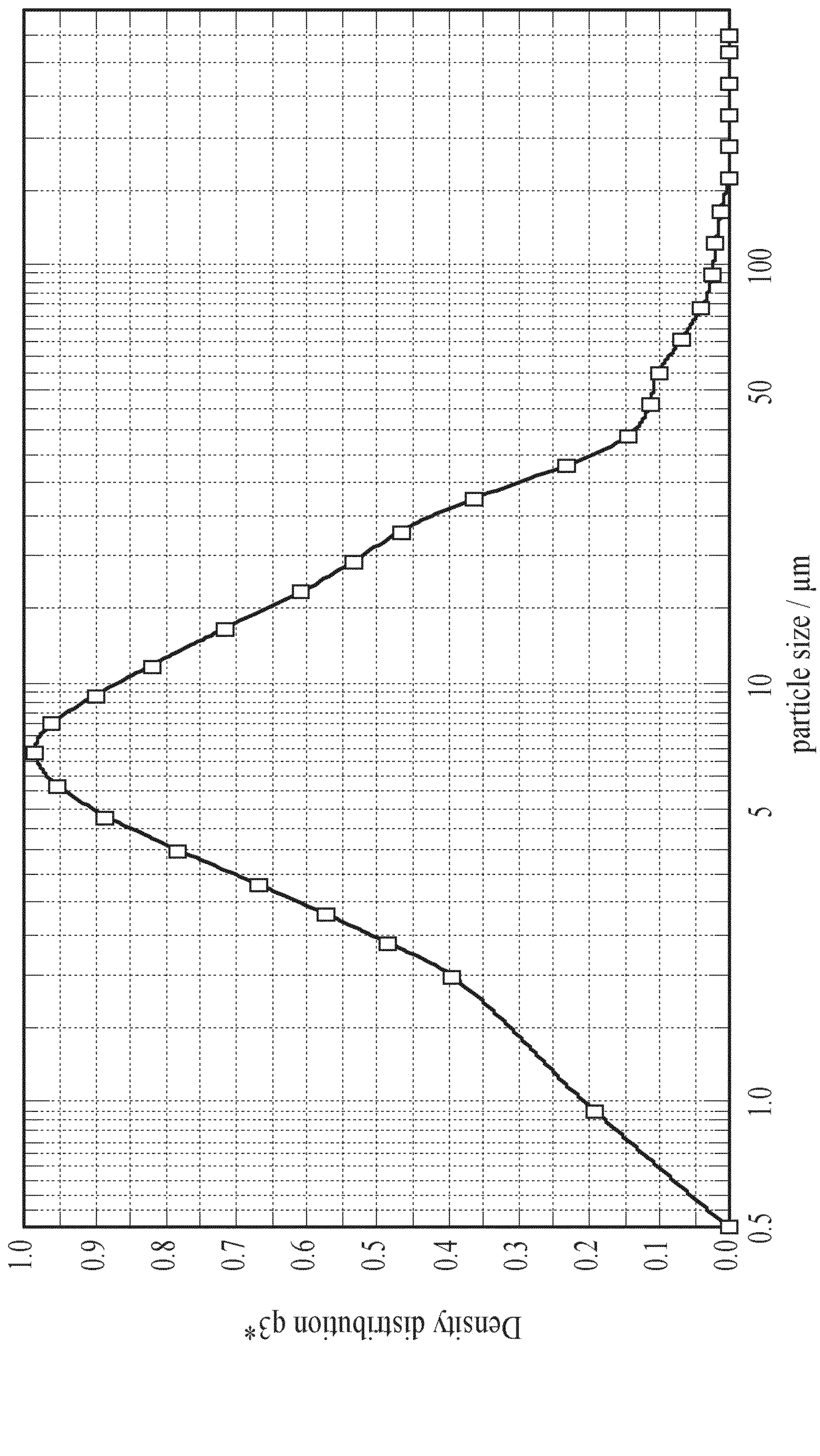
Density distribution q3*
1.0
0.9
0.8
0.7
0.6
0.5
0.4
0.3
0.2
0.1
0.0
0.5
1.0
5
10
50
100
particle size / µm

FIG. 13B

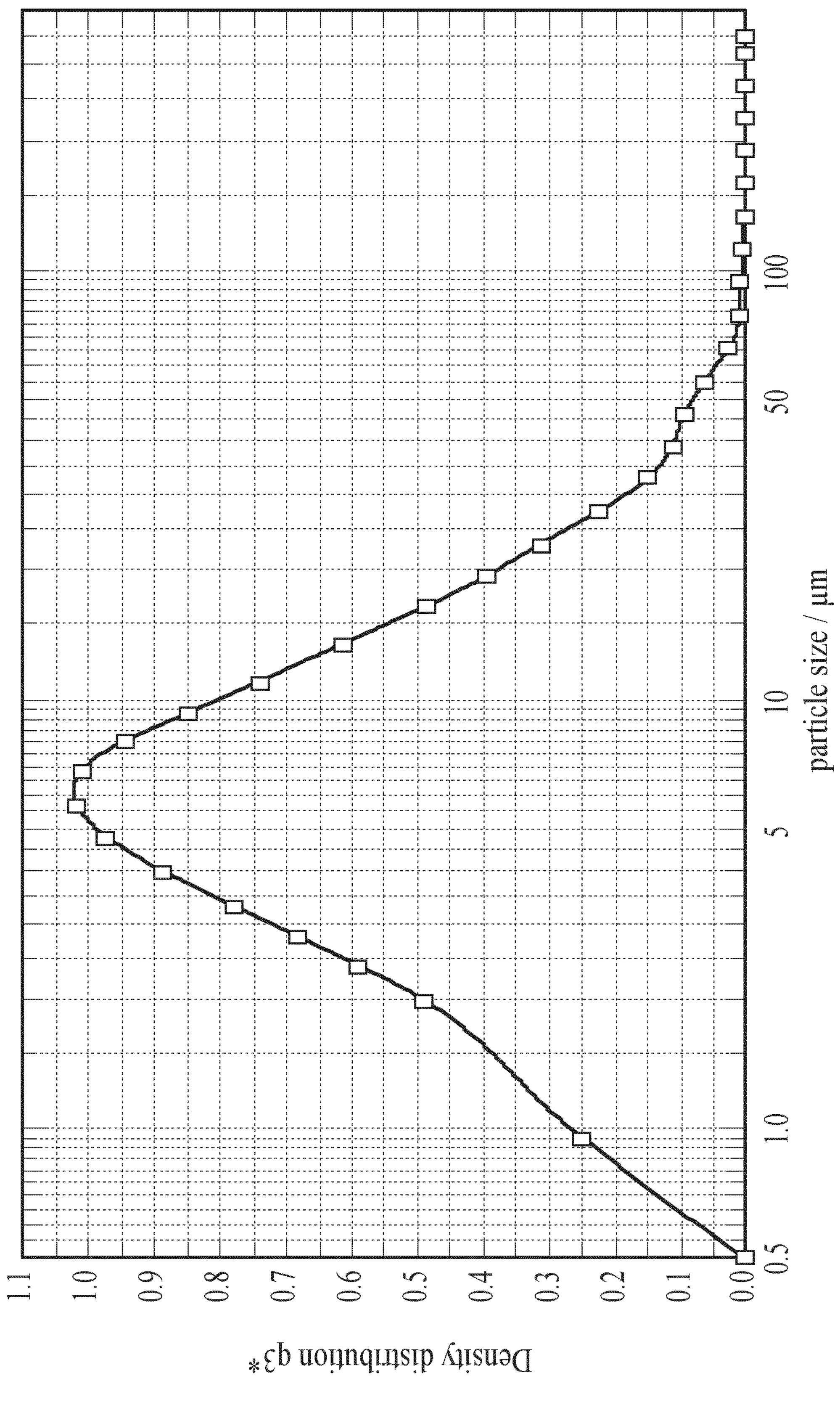
Density distribution q3*
1.1
1.0
0.9
0.8
0.7
0.6
0.5
0.4
0.3
0.2
0.1
0.0
0.5
1.0
5
10
50
100
particle size / µm

FIG. 13C

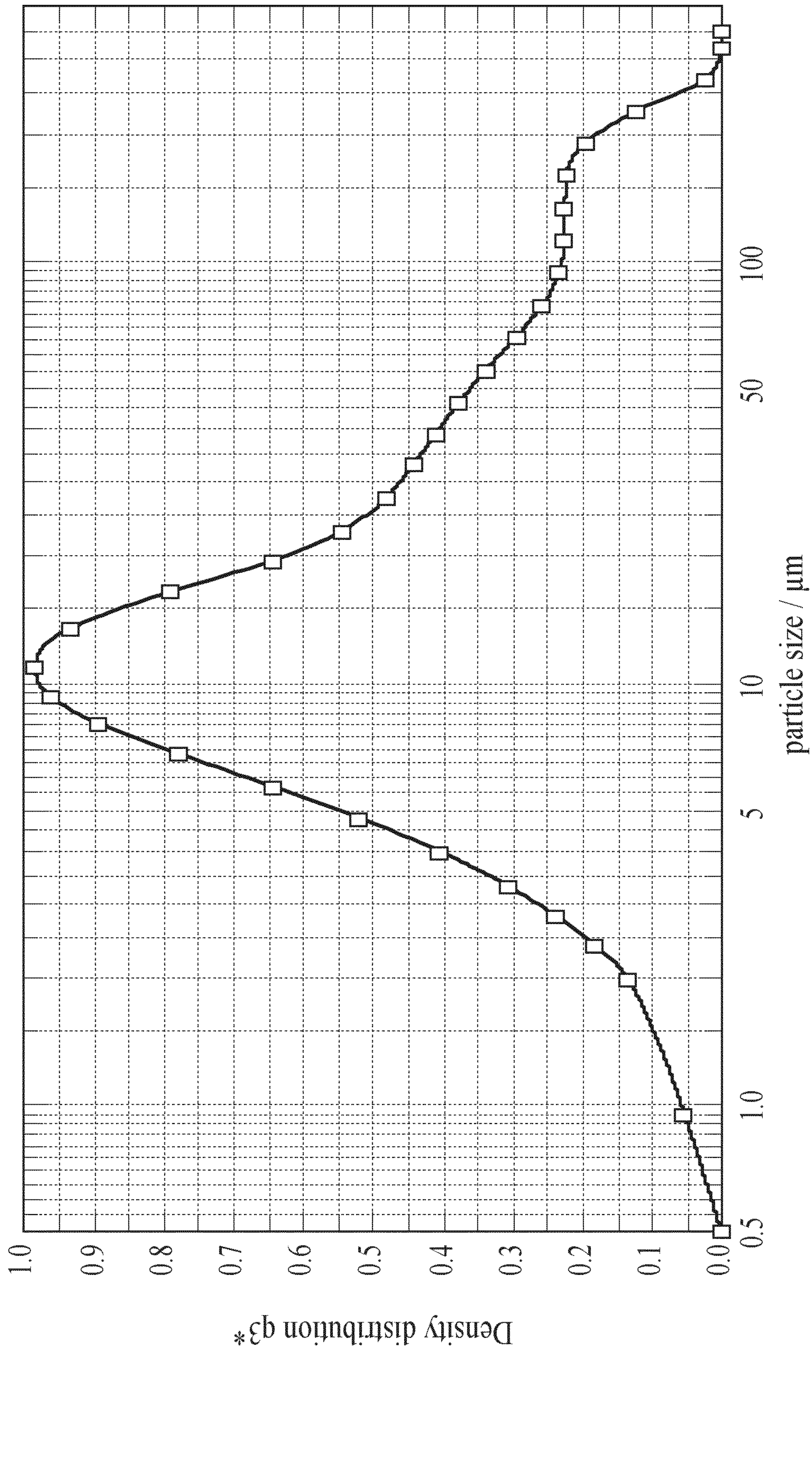
Density distribution q3*
0.0
0.1
0.2
0.3
0.4
0.5
0.6
0.7
0.8
0.9
1.0
0.5
1.0
5
10
50
100
particle size / µm

FIG. 14

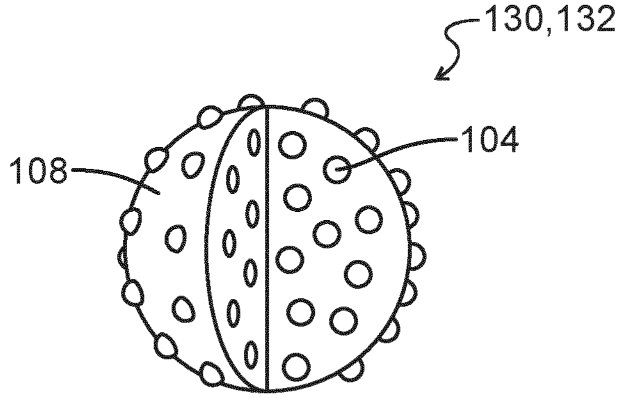

FIG. 15

INTERNATIONAL SEARCH REPORT

International application No
PCT/EP2017/075829

A. CLASSIFICATION OF SUBJECT MATTER
INV. B01F3/08 B01F13/02 B01F5/04 B05B7/04
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)
B01F B05B

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

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

EPO-Internal

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	US 5 484 107 A (HOLMES MICHAEL J [US]) 16 January 1996 (1996-01-16)	1-12, 14-19, 21-26
A	abstract figure 1 column 3, line 7 - line 36 -----	13,20
X	WO 2016/040243 A1 (WEIERSTALL UWE [US]; WANG DINGJIE [US]; SPENCE JOHN [US]) 17 March 2016 (2016-03-17)	1-12, 14-19, 21,24-26
A	abstract figures 1,2 paragraphs [0033] - [0034] -----	13,20, 22,23
	-/--	

[X] Further documents are listed in the continuation of Box C.

[X] See patent family annex.

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Date of the actual completion of the international search	Date of mailing of the international search report
3 January 2018	15/01/2018

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

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INTERNATIONAL SEARCH REPORT

International application No
PCT/EP2017/075829

C(Continuation). DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X A	WO 02/02210 A1 (EBARA CORP [JP]; IZUTSU MASAHIRO [JP]) 10 January 2002 (2002-01-10) abstract figure 2	16-21, 24-26 1-15,22, 23
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X A	WO 2006/126987 A1 (TRONOX LLC [US]; FLYNN HARRY E [US]; MARTIN ROBERT O [US]; NATALIE CHA) 30 November 2006 (2006-11-30) figure 1 abstract	16-26 1-15
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Information on patent family members

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