Note: Within nine months of the publication of the mention of the grant of the European patent in the European Patent Bulletin, any person may give notice to the European Patent Office of opposition to that patent, in accordance with the Implementing Regulations. Notice of opposition shall not be deemed to have been filed until the opposition fee has been paid. (Art. 99(1) European Patent Convention).
Description

BACKGROUND OF THE INVENTION

1. Field of the Invention

[0001] The present invention generally relates to a technology generically referred to as a micro-reactor and a micro-chemical process for controlling a fluid with a flow passage structure of several micrometers to several millimeters to stably realize a chemical engineering unit operation or an analysis within a short period of time. More particularly, the present invention relates to a microscopic flow passage structure, a microscopic liquid droplet generating method, a microscopic liquid droplet generating system, particles, and microcapsules.

[0002] The present invention can be applied to particles, particle dispersions and manufacturing methods thereof used for image display elements (polymerized toner, microcapsules used for cataphoresis type paperlike displays), emulsions used for drug delivery, coating, cosmetics, other emulsions, and photoconductive materials.

2. Description of the Related Art

[0003] Particles obtained by solidifying emulsions or dispersed phases in emulsions and capsule particles obtained by solidifying liquid droplet surfaces are used for various industrial purposes.

[0004] Presently, the following methods are used for industrial production of emulsified dispersions.


The technologies of (1) and (2) are used for obtaining an emulsified dispersion from a substance. The substance to be dispersed into particles corresponds to a dispersed phase, and the dispersed phase is injected into a continuous phase. Shear force is repeatedly applied to the dispersed phase with mechanical actions. However, the shear force applied to the dispersed phase is inconsistent according to the emulsifying position, and therefore, the resultant particles become polydispersive (i.e., having inconsistent sizes).

(3) Patent Document 3, etc., discloses a method of emulsification by using a porous glass film. Conversely, in the method of (3), the dispersed phase and the continuous phase are partitioned with a porous glass film, the dispersed phase is pushed against the continuous phase so that the dispersed phase passes through the porous glass film and contacts the continuous phase, surface tension serves as the shear force, and the dispersed phase is finally turned into particles, thereby obtaining an emulsified dispersion. However, sizes of the generated particles are affected by inconsistencies in the pore sizes, and therefore, the resultant particles become polydispersive.

There is a method of manufacturing highly monodispersive emulsified dispersions and particles as follows.

(4) Patent Document 4, etc., discloses a method of micro-channel emulsification. A film for dividing a dispersed phase and a continuous phase is artificially made to have a uniform structure. Therefore, considerably monodisperse particles can be obtained, where the standard deviation of diameters of particles/average diameter of particles is 0.03 or less. However, the sizes of the channels are smaller than the desired particle size. Therefore, when a liquid including particles is used as the dispersed phase, the channels may be clogged.

(5) Patent Document 5 discloses a method of generating monodispersive micro liquid droplets by using plural Y-type microchannels. However, it is difficult to form multiple flow passages and to guide the same amount of fluid into all of the flow passages. Further, the number of liquid droplets generated from one channel per one unit of time is several thousand, and therefore, yielding ability is low.


SUMMARY OF THE INVENTION

[0005] The present invention provides a microscopic flow passage structure, a microscopic liquid droplet generating method and a microscopic liquid droplet generating system in which one or more of the above-described disadvantages is eliminated.

[0006] A preferred embodiment of the present invention provides a microscopic flow passage structure, a microscopic liquid droplet generating method and a microscopic liquid droplet generating system that can generate highly monodisperse microscopic liquid droplets and significantly increase production by employing a microchannel method having a simple structure.

[0007] An embodiment of the present invention provides a microscopic flow passage structure for generating microscopic liquid droplets, the microscopic flow passage structure including fluid introduction flow passages configured to individually introduce plural types of fluids that are incompatible with each other; a merged flow passage in which the fluids merge; a common outlet through which the fluids can be discharged from the merged flow passage; wherein the merged flow passage includes a region in which a cross-sectional area of the merged flow passage changes cyclically in a fluid flow direction, wherein a cyclic wavelength of the changes in a cross-sectional area of the region is 3.0 to 10.0 times a circle-equivalent diameter of a fluid to be turned into the liquid droplets immediately before the fluids merge.

[0008] An embodiment of the present invention provides a microscopic liquid droplet generating method including the steps of: (a) merging plural types of fluids that are incompatible with each other; (b) inducing a disturbance at an interface between the fluids by causing the fluids to flow through a merged flow passage having a cross-sectional area that changes cyclically in a fluid flow direction; and (c) turning one of the types of the fluids into liquid droplets, wherein a cyclic wavelength of the changes in a cross-sectional area of the region is 3.0 to 10.0 times a circle-equivalent diameter of a fluid to be turned into the liquid droplets immediately before the fluids merge.

BRIEF DESCRIPTION OF THE DRAWINGS

[0009] Other objects, features and advantages of the present invention will become more apparent from the following detailed description when read in conjunction with the accompanying drawings, in which:

FIG. 1 is a schematic diagram of a microscopic flow passage structure according to a first embodiment of the present invention;
FIGS. 2A - 2C are detailed diagrams of the microscopic flow passage structure shown in FIG. 1; FIG. 2A is a plan view of a top glass plate, FIG. 2B is a plan view of a bottom glass plate, and FIG. 2C is a cut-away side view;
FIG. 3 is a microscopic liquid droplet generating system including the microscopic flow passage structure shown in FIG. 1;
FIGS. 4A - 4C are detailed diagrams of a microscopic flow passage structure according to a second embodiment of the present invention; FIG. 4A is a plan view of a top glass plate, FIG. 4B is a plan view of a bottom glass plate, and FIG. 4C is a cut-away side view;
FIGS. 5A - 5C are detailed diagrams of a microscopic flow passage structure according to a third embodiment of the present invention; FIG. 5A is a plan view of a top glass plate, FIG. 5B is a plan view of a bottom glass plate, and FIG. 5C is a cut-away side view;
FIGS. 6A - 6C are detailed diagrams of a microscopic flow passage structure according to a fourth embodiment of the present invention; FIG. 6A is a plan view of a top glass plate, FIG. 6B is a plan view of a bottom glass plate, and FIG. 6C is a cut-away side view;
FIG. 7 is a plan view illustrating how fluids are introduced into the microscopic flow passage structure according to the fourth embodiment;
FIG. 8 is a schematic diagram illustrating how liquid droplets are typically formed;
FIGS. 9A, 9B are schematic diagrams for illustrating how liquid droplets are formed by protruding/depressed structures;
FIG. 10 is a schematic diagram for illustrating how liquid droplets are formed by protruding/depressed cyclic structures;
FIG. 11 is a schematic diagram for illustrating how liquid droplets are formed by a protruding/depressed cyclic structure.
structure in a system including dispersed phases and continuous phases arranged alternately;

FIG. 12 is a schematic diagram illustrating a shape of a protruding/depressed cyclic structure;
FIGS. 13A - 13C are schematic diagrams illustrating other shapes of protruding/depressed cyclic structures;
FIG. 14 is a schematic diagram illustrating yet another shape of a protruding/depressed cyclic structure;
FIG. 15 illustrates a process for forming a film on a surface of a liquid droplet according to a fifth embodiment of the present invention;
FIG. 16 is a schematic diagram illustrating a granulation process (microscopic liquid droplet generating system);
FIG. 17 is a schematic diagram of a system for performing large-scale production by arranging microscopic flow passage structures in parallel;
FIGS. 18A - 18C are detailed diagrams of a microscopic flow passage structure according to a sixth embodiment of the present invention; FIG. 18A is a plan view of a top glass plate, FIG. 18B is a plan view of a bottom glass plate, and FIG. 18C is a cut-away side view;
FIGS. 19A - 19C are detailed diagrams of a microscopic flow passage structure according to a seventh embodiment of the present invention; FIG. 19A is a plan view of a top glass plate, FIG. 19B is a plan view of a bottom glass plate, and FIG. 19C is a cut-away side view;
FIG. 20 is a plan view of relevant parts of a microscopic flow passage structure according to an eighth embodiment of the present invention;
FIG. 21 is a schematic diagram of a microscopic liquid droplet generating system including a Y-type flow passage according to a comparative example;
FIGS. 22A - 22C illustrate a basic configuration of a microscopic flow passage structure with protruding/depressed structures arranged in the depth direction;
FIGS. 23A - 23C illustrate a basic configuration of a microscopic flow passage structure with protruding/depressed structures arranged in the width direction;
FIG. 24 illustrates simulation results of a liquid droplet forming operation in a structure with protruding/depressed structures arranged in the depth direction and when the aspect ratio is 1.5;
FIG. 25 illustrates simulation results of a liquid droplet forming operation in a structure with protruding/depressed structures arranged in the depth direction and when the aspect ratio is 0.5;
FIG. 26 illustrates simulation results of a liquid droplet forming operation in a structure with protruding/depressed structures arranged in the width direction and when the aspect ratio is 4;
FIG. 27 illustrates simulation results of a liquid droplet forming operation in a structure with protruding/depressed structures arranged in the width direction and when the aspect ratio is 0.5;
FIGS. 28A, 28B are schematic diagrams of a merging operation of fluids in the configuration shown in FIG. 24; FIG. 28A illustrates a status before merging, and FIG. 28B illustrates a status after merging; and FIGS. 29A, 29B are schematic diagrams of a merging operation of fluids in the configuration shown in FIG. 25; FIG. 29A illustrates a status before merging and FIG. 29B illustrates a status after merging.

DETAILED DESCRIPTION OF THE PREFERRED EMBODIMENTS

A description is given, with reference to the accompanying drawings, of embodiments of the present invention.

First, referring to FIG. 3, an outline of a microscopic liquid droplet generating system 1 according to the first embodiment is described. The microscopic liquid droplet generating system 1 includes a microscopic flow passage structure 2, fluid tanks 3, 4 storing fluid to be supplied to the microscopic flow passage structure 2, pumps 5, 6 for sending out fluid from the fluid tanks 3, 4, and a recovery tank 7 for recovering microscopic liquid droplets generated by the microscopic flow passage structure 2.

As shown in FIGS. 1, 2A - 2C, the flow passage structure 2 having flow passage patterns is formed by bonding together two PYREX (registered trademark) glass plates 8, 9. In FIGS. 1, 2A - 2C, the cross-hatching area indicates a fusion bonded surface between the glass plates. In FIG. 1, the top glass plate 9 is indicated by dashed lines for visual purposes.

The bottom glass plate 8 is patterned with flow passages by glass etching. As shown in FIG. 1, a fluid introduction flow passage 10 for introducing a fluid (dispersed phase) is formed in the middle of the glass plate 8. On both sides of the fluid introduction flow passage 10 are provided fluid introduction flow passages 11 for introducing a fluid (continuous phase) that is incompatible with the fluid introduced by the fluid introduction flow passage 10 (including fluids that are substantially incompatible).

"Incompatible fluids" refers to two fluids having a solubility less than or equal to 10% with respect to each other.

"Dispersed phase" refers to a fluid that is turned into liquid droplets by a liquid droplet forming unit according to an embodiment of the present invention. "Continuous phase" refers to a fluid serving as a medium for dispersing the liquid droplets generated by the liquid droplet forming unit according to an embodiment of the present invention.
[0017] A merged flow passage 12 is in communication with the fluid introduction flow passages 10, 11; the fluid introduction flow passages 10, 11 merge in the merged flow passage 12. A protruding structure 14 is provided at a position downstream in a direction of the fluid flow (indicated by arrow F) of a merge starting point 12a of the merged flow passage 12. The protruding structure 14 includes two trapezoidal parts facing each other protruding inward from corresponding sides of the merged flow passage 12, so as to change the cross-sectional area of the merged flow passage 12 (in this case, to narrow the passage).

[0018] The protruding structure 14 is provided in an area where liquid columns of the dispersed phase become unstable and turn into liquid droplets. Specifically, the protruding structure 14 is positioned so as to control the process of forming liquid droplets from dispersed phase. The effect that the protruding structure 14 has on forming microscopic liquid droplets is described in subsequent embodiments. The structure for changing the cross-sectional area of the merged flow passage 12 can be a depressed structure.

[0019] As shown in FIGS. 2A - 2C, the top glass plate 9 has a fluid introduction inlet 15 in communication with the fluid introduction flow passage 10, fluid introduction inlets 16 in communication with the fluid introduction flow passages 11, and a common outlet 13.

[0020] Materials of the flow passages are not limited to silica glass; the flow passage can be made of resin that is insoluble to the dispersed phase and the continuous phase. The passage size depends on how the final product is to be used; effects of the present invention are realized when the width and the depth of the flow passage are in a range of 10 μm-1,000 μm.

[0021] When flow passages are formed by glass etching, the protruding/depressed structure for changing the cross-sectional area of the flow passage at the protruding/depressed structure can be formed on the side surfaces of the flow passage as described above, on the bottom, or on the ceiling of the flow passage. When the protruding/depressed structure is formed on the bottom of the flow passage, it can be formed by two-stage etching.

[0022] The cross section of the flow passage is preferably circular; however, depending on conditions in processing the member configuring the flow passage, the cross section can be polygonal such as a rectangle, or substantially polygonal. The cross-sectional area preferably changes in a range of 10.0 μm² - 10,000 μm². The cross-sectional area of the flow passage at the protruding/depressed structure can be larger or smaller than that at the fluid introduction inlet.

[0023] Further, liquid (fluid) is preferably supplied by a pressure liquid feeding method using a pump. As shown in FIG. 3, a liquid droplet dispersion 17 is recovered into the recovery tank 7 from the common outlet 13 positioned at the downstream edge of the merged flow passage 12.

[0024] FIGS. 4A - 4C illustrate a second embodiment according to the present invention. In FIGS. 4A - 4C, elements corresponding to those in FIGS. 1A - 1C are denoted by the same reference numbers, configurations and functions described above are not described unless necessary, and only chief parts are described (likewise in subsequent embodiments).

[0025] In the second embodiment, a protruding structure 18 for changing the cross-sectional area of the merged flow passage 12 is formed on the ceiling of the flow passage. Thus, the protruding structure 18 is formed on the top glass plate 9.

[0026] Accordingly, in the microscopic flow passage structure 2, the protruding structure 18, the fluid introduction inlets 15, 16, and the common outlet 13 are formed on the glass plate 9, and the glass plate 9 is bonded to the bottom glass plate 8.

[0027] FIGS. 5A - 5C illustrate a third embodiment according to the present invention. In the third embodiment, plural liquid columns of a fluid to be turned into liquid droplets are introduced simultaneously, thus enhancing productivity of forming microscopic liquid droplets.

[0028] Plural fluid introduction flow passages 10 for the dispersed phase and plural fluid introduction flow passages 11 for the continuous phase are alternately arranged by fluid type, in parallel (side-by-side) in a direction orthogonal to the direction of the flow of the fluid (width direction of the glass plate). The parallel passages are arranged such that the fluid in contact with the walls of the merged flow passage 12 is a continuous phase.

[0029] When there are plural introduction flow passages, the protruding/depressed structure for changing the cross-sectional areas of the flow passages is preferably formed on the bottom or the ceiling of the merged flow passage 12, in such a manner that the cross-sectional areas of the plural flow passages change by the same amount.

[0030] In consideration of the above, in the third embodiment, a protruding structure 20 is formed on the top glass plate 9. A single protruding structure 20 is provided for changing the cross-sectional area of three flow passages by the same amount (microscopic liquid droplet forming effect).

[0031] FIGS. 6A - 6C and FIG. 7 illustrate a fourth embodiment according to the present invention. In the fourth embodiment, the merged flow passage 12 includes an area 21 in which the cross-sectional area cyclically changes in the direction of the fluid flow.

[0032] As shown in FIGS. 6A - 6C, the area 21 includes plural protruding structures 20, which are equally spaced apart. As a result, the protruding structures 20 configure a cyclic structure including two or more protruding/depressed structures. The cycle is preferably repeated as many times as possible; the cycle is preferably repeated more than or equal to 10 times and less than 100 times to ensure that liquid droplets are stably formed. However, the number of
cycles repeated is not limited thereto.

[0033] As shown in FIG. 7, the flow passages are arranged so that fluids 23 (continuous phases) in contact with walls 22 of the merged flow passage 12 at the edge of in the microscopic flow passages are always continuous phases.

[0034] Next, a detailed description is given of the effects of forming microscopic liquid droplets by changing the cross-sectional area of the merged flow passage 12.

[0035] According to an embodiment of the present invention, a destabilizing phenomenon in a columnar fluid in a microscopic flow passage is efficiently realized. A typical liquid droplet forming phenomenon is described with reference to FIG. 8.

(Liquid droplet forming phenomenon)

[0036] Non-patent literature 1 describes a uniform liquid droplet forming phenomenon occurring in a liquid column. An infinitely long liquid column of fluid is provided in a still fluid. The liquid column is incompatible with the still fluid. A wavelength condition \( \lambda \) at which the liquid column is most unstable is expressed by the following formula (1), where the liquid column diameter is \( d(jet) \).

\[
\lambda = 4.5d(jet) \cdots \cdots (1)
\]

[0037] This formula is satisfied not only for the infinitely long liquid column, but also for liquid flowing at an initial velocity discharged from a through hole into a still fluid or flowing fluid. A frequency \( f \) of a disturbance phenomenon can be expressed by the following formula (2), where the velocity of the liquid column is \( v \). Due to this instability, the liquid column can no longer maintain its shape, and the liquid column turns into liquid droplets having a volume corresponding to each cycle of (1).

\[
f = \frac{v}{\lambda} \cdots \cdots (2)
\]

[0038] As described in non-patent literature 2, experiments were conducted to find conditions under which uniform particles can be stably formed. It was found that uniform particles are stably formed under the conditions expressed by the formula (3).

\[
3.5 < \frac{\lambda}{d(jet)} < 7.0 \cdots \cdots (3)
\]

The upper limit (3.5) and the lower limit (7.0) in the formula (3) may vary depending on the type of fluid. The condition may be satisfied with the upper limit being 3.0 and the lower limit being 10.0; however, the ranges between 3.0 and 3.5 and between 7.0 and 10.0 are transitional, and liquid droplet diameters became uneven in some cases in such ranges.

[0039] As described in non-patent literature 3, based on conservation of energy, the minimum jet velocity \( V(min) \) at which liquid discharged from through holes forms liquid columns is expressed by the following formula (4).

\[
V(min) = \left( \frac{8\sigma}{\rho d(jet)} \right)^{1/2} \cdots (4)
\]

In formula (4), \( \sigma \) is surface tension of the liquid, \( \rho \) is liquid density, and \( d(jet) \) is the diameter of a liquid column. The condition formulas (1) through (4) are useful for estimating conditions for reproducing the above described phenomenon; however, the inventors of the present invention have confirmed that these formulas may vary according to the type of liquid substance, the compound, the dispersion, etc.

[0040] When liquid droplets are formed based on conditions estimated by the above condition formulas, the state of a single liquid column may change with time, or states of adjacent liquid columns may be different from each other. As a result, the status of destabilization may vary as time passes; therefore, it is considerably difficult to produce a large amount of microscopic liquid droplets having uniform diameters.

[0041] In an embodiment of the present invention, the cross-sectional area of the merged flow passage 12 is changed to intentionally induce uniform disturbances at interfaces of the two fluids. This prevents effects of subtle changes in the conditions, so that liquid droplet diameters are consistently uniform.

[0042] That is, in an embodiment of the present invention, the destabilizing phenomenon of the columnar fluid is used to prevent diameters from becoming uneven.
FIGS. 9A, 9B are cross-sectional views of a flow passage taken along a normal line in the direction of the fluid flow in the flow passage. The velocity at which a fixed flow quantity of fluid passes through a flow passage is inversely proportional to the cross-sectional area of the flow passage. Accordingly, the flow velocity is fast at cross section A, and the flow velocity is slow at cross section B.

When there are protruding/depressed structures on the wall of a flow passage such that cross-sectional areas of the flow passage vary at different positions, and a fixed flow quantity of a fluid is made to pass through the flow passage, the velocity of the fluid changes depending on the position.

As shown in FIG. 9B, when a dispersed phase fluid and a continuous phase fluid that are incompatible with each other are made to flow through a microscopic flow passage having the protruding/depressed structures, the velocities of both fluids change due to changes in the cross-sectional area, as described above.

An embodiment of the present invention takes advantage of this feature in order to stably and efficiently realize a destabilizing phenomenon of the columnar fluid.

This feature is applied to the configuration of the fourth embodiment, as shown in FIG. 10. A dispersed phase introduced into a flow passage having a cyclic structure forms a liquid column in the continuous phase. A protruding/depressed cyclic structure in the area 21 induces disturbances at interfaces of the two fluids (phases). Accordingly, a dispersed phase 24 is turned into liquid droplets, thereby generating microscopic liquid droplets 25.

In the model illustrated in FIGS. 9A, 9B, the protruding/depressed cyclic structure is provided symmetrically along the direction of the fluid flow. However, the disturbance phenomenon can be induced by the changes in the cross-sectional areas of the flow path even if the protruding/depressed cyclic structure is provided only on one side, on the bottom, or on the ceiling of the merged flow passage 12.

Further, the same phenomenon can be induced by providing the protruding/depressed cyclic structure in a system where dispersed phases and continuous phases are arranged alternately, as shown in FIG. 11.

The protruding/depressed cyclic structure provides equal effects on plural columnar fluids, so that the microscopic liquid droplets generated from the dispersed phases have uniform diameters.

The diameters of the microscopic liquid droplets are less uniform when a single protruding structure or a single depressed structure is employed, compared to the case of employing a cyclic structure including plural protruding/depressed structures. However, even with a single protruding/depressed structure, effects of the above-described "intentionally induced uniform disturbances" can be provided to stably produce uniform liquid droplets.

According to Rayleigh's instability principle, a columnar fluid becomes most unstable when the wavelength of the disturbance is approximately 4.5 times the diameter of the columnar fluid of the dispersed phase. Further, liquid droplets are stably formed when the wavelength of the disturbance is between 3 to 10 times the diameter of the columnar fluid of the dispersed phase. The diameter of the columnar fluid can be decreased or increased by changing the speed of the continuous phase.

Therefore, the wavelength of the cyclic structure is preferably in a range of 3.0 - 10.0 times the narrowest portion of the dispersed phase (circle-equivalent diameter immediately before merging), more preferably in a range of 3.5 - 7.0 times.

The shape of the protruding/depressed cyclic structure can be a rectangular wave shape in which the angle between two adjacent sides is a right angle as shown in FIG. 12, or other types of wave shapes as shown in FIGS. 13A - 13C. These shapes are appropriately determined so that this area is not affected by satellites (microscopic particles) generated at a certain speed.

The wave shapes include shapes in which the angle between two adjacent sides is not a right angle (e.g., FIG. 13A), such as a triangular wave shape (e.g., FIG. 13B) or a curved wave shape (e.g., FIG. 13C).

The wave shape can be a combination of two or more arbitrary shapes, as shown in FIG. 14. In this example, a combination 28 of two arbitrary shapes 26, 27 form one cycle in the cyclic structure. These shapes are also appropriately determined so that this area is not affected by satellites generated at a certain speed.

FIG. 15 illustrates a fifth embodiment according to the present invention. The fifth embodiment requires a process of forming a film on the liquid droplet surface at the end of the liquid droplet forming flow passage (downstream in the direction of the fluid flow). Specifically, a fluid including a film forming material 31 is introduced into a main flow passage 29 from a branch flow passage 30. It is possible to employ a film forming method of depositing the material onto the liquid droplet surface or causing a chemical reaction on the liquid droplet surface.

The branch flow passage 30 is provided downstream of the cyclic structure in the embodiments described above. A unit for forming a film on a liquid droplet surface includes the branch flow passage 30 and a not shown means for supplying the film forming material 31.

The film forming process is not necessarily performed by using a flow path system; it is possible to perform a
typical encapsulating process on an emulsion recovered at the outlet of the flow passage.

[0062] Particles obtained by solidifying microscopic liquid droplets formed by the liquid droplet forming method can be used as toner. If the variance is 5% or less, a toner having excellent image granularity and excellent powder fluidity can be achieved.

[0063] Microcapsules having a variance of 5% or less formed by the liquid droplet forming method according to the fifth embodiment can be used as cataphoresis microcapsules. As particle diameters are uniform, an electronic paper having a high level of whiteness can be provided.

[0064] Preferable guidelines concerning configurations and materials are given below for implementing the present invention.

(Flow passage structure, fluid introduction inlet, fluid discharge outlet)

[0065] Flow passage patterns are formed on one PYREX (registered trademark) glass plate by a regular glass etching method. Another PYREX (registered trademark) glass plate having an inlet/outlet for fluid is put on the first plate. After being positioned, the two plates are bonded together by heat sealing, thereby forming a flow passage having an inlet/outlet.

(Fluid conveying pump)

[0066] Means for conveying fluid include, but are not limited to, a syringe pump, a pressurized liquid sending method using inert gas, a peristaltic pump, and a diaphragm pump. To introduce liquid into a flow passage of micrometer order, a pump that can pressurize for a pressure drop of approximately 100 kPa - 1,000 kPa is used.

[0067] In the following practical examples, a pressurized liquid sending method using inert gas was employed. The inert gas is to be inert to the chemical liquid used. In the following practical examples, nitrogen gas was used.

(Piping, connection)

[0068] The pipe is to be inert to the chemical liquid and insoluble to the chemical liquid. Furthermore, the pipe is to be pressure resistant. The material of the pipe depends on the type of the chemical liquid, including, but not limited to, Teflon (registered trademark), various metals, and PEEK. A pipe made of PEEK was employed in the following practical examples.

[0069] When connecting two pipes, connecting a pipe and a liquid tank, or connecting a pipe and a flow passage structure, the connection part is to be inert to the chemical liquid used, insoluble to the chemical liquid, and leakproof. Furthermore, the connection part needs to be pressure resistant. Therefore, the connection method and material are selected depending on usage conditions.

[0070] The connection part can be connected with, but not limited to, a rubber O ring, a Teflon (registered trademark) resin-made O ring, an adhesive, or a flange. The material of the connection part includes, but is not limited to, Teflon (registered trademark), various metals, and PEEK. The material depends on the type of chemical liquid. In the following practical examples, the connection part was made of PEEK, which was connected by a flange. Generally available components were used, such as those employed in high pressure liquid chromatography.

(Granulation process overview)

[0071] FIG. 16 is an overall diagram of a granulation process performed in the following practical examples. Continuous phases and dispersed phases are introduced to a microscopic flow passage structure 122 by pressurized liquid sending mechanisms 121 using dry nitrogen. A pipe 123 and the microscopic flow passage structure 122 are connected by a connector.

[0072] Generated emulsion is collected in a recovery container 124. Needle valves 125 are provided in pipes of the pressurized liquid sending mechanisms 121 to adjust the flow amounts of the continuous phases and the dispersed phases. Pressure gauges 126 are provided for confirming the pressure applied on the fluids. Further, a temperature adjusting mechanism can adjust the temperature of the microscopic flow passage structure 122, the pipe 123, and a storage tank, between -20 °C and 150 °C.

(Materials of dispersed phase and continuous phase, dispersing agent, surfactant)

[0073] The method according to an embodiment of the present invention can be implemented if the dispersed phase and the continuous phase have a compatibility of 10% or less, more preferably 1% or less. Typical dispersing agents and surfactant are not required for forming liquid droplets. However, the dispersed phase and the continuous phase
may be combined when they are recovered. Therefore, such agents are preferably included at a critical concentration or higher, or added in a subsequent process.

[0074] The dispersing agent can be made from gelatin or polyvinyl alcohol. Examples of surfactant are anionic surfactant such as alkyl benzene sulfonate, alpha olefin sulfonate, phosphate; amine salt type surfactant such as alkylamine salt, amino alcohol fatty acid derivative, polyamine fatty acid derivative, imidazoline; quaternary ammonium salt type cationic surfactant such as alkyl trimethyl ammonium salt, dialkyl dimethyl ammonium salt, alkyl dimethyl benzylammonium salt, pyridinium salt, alkyl isoquinolinium salt, benzethonium chloride; nonionic surfactant such as fatty acid amide derivative, polyalcohol derivative; and amphoteric surfactant such as alanine, dodecyl di (aminoethyl) glycine, di (octyl-aminoethyl) glycine, N-alkyl-N, N-dimethyl ammonium betaine.

[0075] By using surfactant having a fluoro alkyl group, effects can be enhanced with an extremely small amount. Preferable examples from the anion surfactant having a fluoro alkyl group include fluoro alkyl carboxylic acid with carbon number from 2 to 10 or metallic salts thereof; perfluoro octane sulfonate glutamic acid disodium, 3-[omega-fluoro alkyl (C6-C11) oxy]-1-alkyl (C3-C4) sodium sulfonate, 3-[omega-fluoro alkanoyl (C6-C8)-N-ethyl amino]-1-propane sulfonic acid, sodium fluoro alkyl (C11-C20) carboxylic acid or metallic salts thereof; perfluoro alkyl carboxylic acid (C7-C13) or metallic salts thereof; perfluoro alkyl (C4-C12) sulfonic acid or metallic salts thereof; perfluoro octane sulfonic acid diethanol amide, N-propyl-N-(2 hydroxyethyl)perfluoro octane sulfone amide, perfluoro alkyl (C6-C10) sulfone amide propyl trimethyl ammonium salt, perfluoro alkyl (C6-C10)-N-ethyl sulfonate glycine salt, and monoperfluoroalkyl (C6-C16) ethyl phosphate ester.

[0076] As a commercial item of the anion surfactant having a fluoro alkyl group, for example, Surflon S-111, S-112, and S-113 (manufactured by Asahi Glass Co., Ltd.), Fluorac FC-93, FC-95, FC-98, and FC-129 (manufactured by Sumitomo 3M), Unidyne DS-101 and DS-102 (manufactured by Daikin Industries, Ltd.), Megafase F-110, F-120, F-113, F-191, F-812, and F-833 (manufactured by Dainippon Ink), Eftop EF-102, 103, 104, 105, 112, 123A, 123B, 306A, 501, 201, and 204 (manufactured by JEMCO Inc.), FTERGENT F-100 and F150 (manufactured by Neos) can be cited.

[0077] Examples of the cation surfactant having a fluoro alkyl group include fatty series quaternary ammonium salt such as perfluoro alkyl (C6-C10) sulfone amide propyl trimethyl ammonium salt, benzal conium salt, benzethonium chloride, pyridinium salt, and imidazolium salt. As a commercial item of the cation surfactant having a fluoro alkyl group, for example, Surflon S-121 (manufactured by Asahi Glass Co., Ltd.), Fluorad FC-135 (manufactured by Sumitomo 3M), Unidyne DS-202 (manufactured by Daikin Industries, Ltd.), Megafase F-150 and F-824 (manufactured by Dainippon Ink), Eftop EF-132 (manufactured by JEMCO Inc.), FTERGENT F-300 (manufactured by Neos) can be cited.

[0078] Further, it is possible to stably disperse toner constituents in water using a polymeric protection colloid. Specific examples of such protection colloids include polymers and copolymers prepared using monomers such as acids (e.g., acrylic acid, methacrylic acid, alpha-cyanoacrylic acid, alpha-cyanomethacrylic acid, itaconic acid, crotonic acid, fumaric acid, maleic acid and maleic anhydride), (meta)acrylic monomers having a hydroxyl group (e.g., beta-hydroxyethyl acrylate, beta-hydroxyethyl methacrylate, beta-hydroxypropyl acrylate, beta-hydroxypropyl methacrylate, gamma-hydroxypropyl acrylate, gamma-hydroxypropyl methacrylate, 3-chloro-2-hydroxypropyl acrylate, 3-chloro-2-hydroxypropyl methacrylate, diethylene glycol monomethacrylic acid esters, diethylene glycol monomethacrylic acid ethers, glycerinmonomethacrylic acid esters, glycerinmonomethacrylic acid esters, N-methylolacrylamide and N-methylolmethacrylamide), vinyl alcohol and its ethers (e.g., vinyl methyl ether, vinyl ethyl ether and vinyl propyl ether), esters of vinyl alcohol with a compound having a carboxyl group (e.g., vinyl acetate, vinyl propionate and vinyl butyrate); acrylic amides (e.g., acrylamide, methacrylamide and diacetoneacrylamide) and their methylol compounds, acid chlorides (e.g., acrylic acid chloride and methacrylic acid chloride), and monomers having a nitrogen atom or an alicyclic ring having a nitrogen atom (e.g., vinyl pyridine, vinyl pyrrolidone, vinyl imidazole and ethylene imine). In addition, polymers such as polyoxyethylene compounds (e.g., polyoxyethylene, polyoxypropylene, polyoxyethylenealkyl amines, polyoxypropylenealkyl amines, polyoxyethylenealkyl amides, polyoxypropylenealkyl amides, polyoxyethylene nonylphenyl ethers, polyoxyethylene laurylphenyl ethers, polyoxyethylene stearylphenyl esters, and polyoxyethylene nonylphenyl esters); and cellulose compounds such as methyl cellulose, hydroxyethyl cellulose and hydroxypropyl cellulose, can also be used as the polymeric protective colloid.

[0079] When a dispersing agent is used, the dispersing agent can be left adhering on liquid droplet surfaces. However, in a case where it is important to maintain insulation of liquid droplets, the dispersing agent can be removed.

(Encapsulating process)

[0080] A coacervation method or an ultraviolet curing method, etc., can be employed for an encapsulating process. In the following practical examples, a simple coacervation method was employed. Specifically, emulsion discharged from a flow passage structure was introduced into a 10% gelatin water solution heated to 45 degrees Celsius, and ethanol was added by small portions. When the ethanol concentration is approximately 50%, the solution is cooled, so that the colloid is micro-capsulated.
(Liquid droplet solidifying process)

[0081] Liquid droplets can be solidified by including a monomer capable of a crosslinking reaction and a radical polymerization initiator in the dispersed phase, and initiating polymerization by ultraviolet rays or heat.

(Particle diameter evaluation means)

[0082] Some of the recovered liquid droplets or capsules were extracted, and observed on a preparation with a microscope. An image analysis method was employed to obtain the average particle diameter of 1,000 particle images and a dispersion value (standard deviation of diameter)/(average diameter).

(Parallel arrangement)

[0083] When multiple microscopic flow passage structures 122 are operated in parallel as illustrated in FIG. 17, flow amount adjusting mechanisms 132 are provided immediately before introduction parts of the microscopic flow passage structures 122, so that equal amounts of fluid are sent into the microscopic flow passage structures 122.

[0084] As a result of further studies, it was found that formation of liquid droplets is affected by the shape of the outlet of the dispersed phase flow passage, where the flow passages of the dispersed phase and the continuous phase merge.

[0085] Therefore, the cross-sectional shape of the outlet of the flow passage of the dispersed phase (shape of boundary between the merged flow passage 12) is determined as follows. Specifically, assuming that a width direction is the direction in which the flow passages of the dispersed phases and the continuous phases are arranged, and a depth direction is the direction orthogonal to the width direction, the shape of the outlet is made to satisfy a condition that "length in the depth direction/length in the width direction” (aspect ratio) is greater than or equal to one. When the aspect ratio is greater than or equal to one, liquid droplets can be formed properly.

[0086] FIGS. 22A - 22C illustrate an example where a dispersed phase is introduced into the fluid introduction flow passage 10 that is a first flow passage, continuous phases are introduced into the fluid introduction flow passages 11 that are second and third flow passages, and the protruding/depressed structures are arranged in the depth direction. FIGS. 23A - 23C illustrate an example where a dispersed phase is introduced into the fluid introduction flow passage 10, continuous phases are introduced into the fluid introduction flow passages 11, and the protruding/depressed structures are arranged in the width direction. The outlet shape where the flow passage of the dispersed phase and the flow passages of the continuous phases merge is important in both cases shown in FIGS. 22A - 22C and 23A - 23C. Specifically, the ratio of the outlet shape in the depth direction and in the width direction is important.

[0087] Simulations were conducted to study differences in liquid droplet formation depending on this ratio (aspect ratio). The simulations were performed by using thermal-liquid analysis software called FLUENT.

[0088] Examples of results of the simulations are shown in FIGS. 24 - 27. In the examples shown in FIGS. 24, 25, the protruding/depressed structures shown in FIG. 22B are arranged in the depth direction. The aspect ratio is 1.5 in the example shown in FIG. 24, and the aspect ratio is 0.5 in the example shown in FIG. 25.

[0089] In the examples shown in FIGS. 26, 27, the protruding/depressed structures shown in FIG. 23B are arranged in the width direction. The aspect ratio is 4 in the example shown in FIG. 26, and the aspect ratio is 0.5 in the example shown in FIG. 27.

[0090] As shown in FIGS. 25, 27, when the aspect ratio is 0.5, the liquid droplets 25 are not formed. As shown in FIGS. 24, 26, when the aspect ratio is large enough, the liquid droplets 25 are formed.

[0091] Results of the simulations say that liquid droplets are formed when the aspect ratio is large. The reason is described with reference to the models shown in FIG. 24, 25.

[0092] As shown in FIG. 28A, in the fluid introduction flow passage 10, the cross-sectional shape of the dispersed phase 24 is the same as the fluid passage. However, as shown in FIG. 28B, when the dispersed phase 24 merges with the continuous phase 23, a layered flow is formed. Accordingly, the cross-sectional shape of the dispersed phase 24 becomes circular due to surface tension, and the dispersed phase 24 finally becomes a cylinder. In the example shown in FIG. 24, the cross-sectional shape of the dispersed phase 24 becomes vertically long, corresponding to a high aspect ratio, and gaps are formed at the top and the bottom in the depth00 direction. The continuous phase 23 enters these gaps.

[0093] In the example shown in FIG. 25, the cross-sectional shape of the dispersed phase 24 becomes horizontally long, corresponding to a low aspect ratio, and gaps are not formed at the top and the bottom in the depth direction. Accordingly, there are no spaces for the continuous phase 23 to enter.

[0094] As shown in FIG. 28B, in the example shown in FIG. 24, the dispersed phase 24 is covered by the continuous phase 23 as shown in FIG. 28B. In the example shown in FIG. 25, the dispersed phase 24 is not totally covered by the continuous phase 23 as shown in FIG. 29B. The dispersed phase 24 needs to be totally covered by the continuous phase 23 in order to form liquid droplets.

[0095] The same relationship as described above applies to the examples shown in FIGS. 26, 27.
It is important to determine the level of the aspect ratio necessary for forming liquid droplets. Simulations were performed to find relationships between aspect ratios and formation of liquid droplets. Results are shown in Tables 1, 2. Table 1 includes results for a case where the protruding/depressed structures are provided in the width direction. Table 2 includes results for a case where the protruding/depressed structures are provided in the depth direction. Both tables indicate whether liquid droplets were formed under different aspect ratios and viscosities of the continuous phase.

### Table 1

Formation of liquid droplets with protruding/depressed structures provided in width direction

<table>
<thead>
<tr>
<th>Aspect ratio</th>
<th>Viscosity of continuous phase (cSt)</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.1</td>
<td>1 1.5 2 10</td>
</tr>
<tr>
<td>1</td>
<td>✗ o o o</td>
</tr>
<tr>
<td>1.5</td>
<td>o</td>
</tr>
<tr>
<td>2</td>
<td>o</td>
</tr>
<tr>
<td>4</td>
<td></td>
</tr>
</tbody>
</table>

Note: Blank cells: Not calculated

### Table 2

Formation of liquid droplets with protruding/depressed structures provided in depth direction

<table>
<thead>
<tr>
<th>Aspect ratio</th>
<th>Viscosity of continuous phase (cSt)</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.1</td>
<td>1 1.5 2 10</td>
</tr>
<tr>
<td>1</td>
<td>o ✗ o o</td>
</tr>
<tr>
<td>1.5</td>
<td>o</td>
</tr>
<tr>
<td>2</td>
<td></td>
</tr>
<tr>
<td>4</td>
<td></td>
</tr>
</tbody>
</table>

Note: Blank cells: Not calculated

A description is given below of the case of Table 1 in which protruding/depressed structures are provided in the width direction. The results of Table 1 say that liquid droplets are not formed when the aspect ratio is 0.5. Further, when the aspect ratio is one, liquid droplets are formed more successfully as the viscosity of the continuous phase increases. The viscosity of the continuous phase never falls below the viscosity of water (1 cSt).

Surfactant, etc., is mixed with water or a liquid similar to water in order to facilitate the formation of liquid droplets, and therefore, the viscosity often becomes high. When the viscosity becomes high, the liquid does not flow well in the flow passage. Accordingly, an appropriate viscosity of the continuous phase is in a range of 1 cSt - 10 cSt in the simulation. When the aspect ratio is larger than one, liquid droplets can be formed regardless of the viscosity of the continuous phase.

A description is given below of the case of Table 2 in which protruding/depressed structures are provided in the depth direction. Based on the results shown in Table 1, it can be estimated that when the aspect ratio is 0.5 and the viscosity of the continuous phase is high, liquid droplets cannot be formed. When the aspect ratio is larger than one, it can be estimated that liquid droplets can be formed regardless of the viscosity of the continuous phase.

The relationship between the aspect ratio and the viscosity of the continuous phase is indicated in the tables. In addition, simulations were performed to study the relationship between the aspect ratio and the viscosity of the dispersed phase and the relationship between flow speeds of the dispersed phase and the continuous phase. It has been confirmed that the viscosity of the continuous phase slightly affects formation of liquid droplets when the protruding/depressed structures are provided in the width direction, but the viscosity of the dispersed phase and the flow speeds of the dispersed phase and the continuous phase do not affect formation of liquid droplets.

When actually forming liquid droplets, water is not used as the continuous phase. Surfactant is dissolved in...
water or a liquid other than water is used as the continuous phase, and therefore, the viscosity is higher than 1 cSt. Accordingly, whether the protruding/depressed structures are formed in the width direction or the depth direction, liquid droplets are formed as long as the aspect ratio is larger than one.

[0102] Due to the above reasons, the aspect ratio is defined as being larger than one in claim 2 (nineth embodiment).

[0103] With regard to the maximum aspect ratio, the aspect ratio is preferably as large as possible, so that gaps are formed at the top and the bottom in the depth direction into which the continuous phase can enter. In reality, it is difficult to fabricate a flow passage having a high aspect ratio, so the maximum aspect ratio corresponds to the largest possible aspect ratio that can actually be manufactured.

(Working example 1)

[0104] Working example 1 employed a PYREX (registered trademark) glass plate with flow passages formed by etching and a single protruding structure having a substantially rectangular wave shape (see FIGS. 4A - 4C) to form liquid droplets. The cross-sectional shape of the dispersed phase at the introduction inlet was 20 μm in the width direction and 40 μm in the depth direction. The protruding structure was 4.9 μm in height and 8.2 μm in width. The merging part of the continuous phase and the dispersed phase was 100 μm in width and 40 μm in depth.

[0105] A liquid obtained by dispersing a pigment in ISOPAR G (registered trademark; Exxon Mobil Corporation) was used as the dispersed phase. A liquid including 2% gelatin dissolved in ion-exchange water was used as the continuous phase. These were the compositions of the dispersed phase and the continuous phase of the working examples and comparative example described herein. Pressure applied to the dispersed phase fluid was 150 kPa. Pressure applied to the continuous phase fluid was 320 kPa. After granulation was continued for five hours, the liquid droplets obtained had an average diameter of 48.0 μm, a dispersion value of 8.2%, and the yield was 21 g.

(Working example 2)

[0106] Working example 2 employed a PYREX (registered trademark) glass plate with flow passages formed by etching and a cyclic structure having substantially rectangular wave shapes (see FIGS. 18A - 18C; sixth embodiment) to form liquid droplets. The cross-sectional shape of the dispersed phase at the introduction inlet was 20 μm in the width direction and 40 μm in the depth direction. The cyclic structure had a cycle of 100 μm. The merging part of the continuous phase and the dispersed phase was 100 μm in width and 40 μm in depth.

[0107] Pressure applied to the dispersed phase fluid was 150 kPa. Pressure applied to the continuous phase fluid was 320 kPa. After granulation was continued for five hours, the liquid droplets obtained had an average diameter of 48.0 μm, a dispersion value of 1.2%, and the yield was 25 g.

(Working example 3)

[0108] Working example 3 employed a PYREX (registered trademark) glass plate with flow passages formed by etching and a cyclic structure having substantially rectangular wave shapes (see FIGS. 6A - 6C) to form liquid droplets. The cross-sectional shape of the dispersed phase at the introduction inlet was 20 μm in the width direction and 40 μm in the depth direction. The cyclic structure had a cycle of 100 μm. The merging part of the continuous phase and the dispersed phase was 100 μm in width and 40 μm in depth.

[0109] Pressure applied to the dispersed phase fluid was 150 kPa. Pressure applied to the continuous phase fluid was 320 kPa. After granulation was continued for five hours, the liquid droplets obtained had an average diameter of 45.0 μm, a dispersion value of 1.8%, and the yield was 142 g.

(Working example 4)

[0110] Working example 4 employed ten PYREX (registered trademark) glass plates arranged in parallel (see FIG. 17) to form liquid droplets, each plate having flow passages formed by etching and a cyclic structure having substantially rectangular wave shapes (see FIGS. 6A - 6C). The cross-sectional shape of the dispersed phase at the introduction inlet was 20 μm in the width direction and 40 μm in the depth direction. The cyclic structure had a cycle of 100 μm. The merging part of the continuous phase and the dispersed phase was 100 μm in width and 40 μm in depth.

[0111] Pressure applied to the dispersed phase fluid was 250 kPa. Pressure applied to the continuous phase fluid was 480 kPa. After granulation was continued for five hours, the liquid droplets obtained had an average diameter of 52.0 μm, a dispersion value of 4.8%, and the yield was 512 g.
(Working example 5)

[0112] Working example 5 employed a PYREX (registered trademark) glass plate with flow passages formed by etching and a cyclic structure having substantially rectangular wave shapes (see FIGS. 19A - 19C; seventh embodiment) to form liquid droplets. The cross-sectional shape of the dispersed phase at the introduction inlet was 20 μm in the width direction and 40 μm in the depth direction. The cyclic structure had a cycle of 100 μm. The merging part of the continuous phase and the dispersed phase was 100 μm in width and 40 μm in depth. The depressed parts were 5.0 μm in depth and 12 μm in length. The protruding parts were 8.0 μm in height and 88 μm in length.

[0113] Pressure applied to the dispersed phase fluid was 110 kPa. Pressure applied to the continuous phase fluid was 300 kPa. After granulation was continued for five hours, the liquid droplets obtained had an average diameter of 42.0 μm, a dispersion value of 0.5%, and the yield was 20 g.

(Comparative example 1) Y-shaped flow passage

[0114] Comparative example 1 employed a PYREX (registered trademark) glass plate with flow passages formed by etching and a Y-shaped flow passage structure 50 (see FIG. 21) to form liquid droplets. The cross-sectional area of the dispersed phase at the introduction inlet had a circle-equivalent diameter of 60 μm. The merging part of the continuous phase and the dispersed phase was 100 μm in width and 40 μm in depth.

[0115] Pressure applied to the dispersed phase fluid was 100 kPa. Pressure applied to the continuous phase fluid was 120 kPa. After granulation was continued for five hours, the liquid droplets obtained had an average diameter of 74.0 μm, a dispersion value of 1.5%, and the yield was 0.6 g.

[0116] Table 3 includes particle diameters, dispersion values, yield, etc., of the liquid droplets obtained in working examples 1 - 5 and comparative example 1.

| Table 3 |
|---|---|---|---|---|
| | Particle diameter [μm] | Dispersion value [%] | Yield [g] | No. of liquid columns [dispersed phase] |
| Working example 1 | 48 | 8.2 | 21 | single |
| Working example 2 | 48 | 1.2 | 25 | single |
| Working example 3 | 45 | 1.8 | 142 | plural |
| Working example 4 | 52 | 4.8 | 512 | plural |
| Working example 5 | 42 | 0.5 | 20 | single |
| Comparative example 1 | 74 | 1.5 | 0.6 | none |

[0117] The following can be understood from the results shown in Table 3.

(1) Particle diameters (microscopic liquid droplet diameters) can be made smaller when a protruding/depressed structure, which induces a disturbance at the interface of the fluids, is provided (working examples 1 - 5), compared to when a protruding/depressed structure is not provided (comparative example 1).

(2) Dispersion values are smaller and particle diameters are more uniform when a cyclic protruding/depressed structure is provided, compared to when the protruding/depressed structure is provided singularly (working example 1).
(3) Particlediametersareevenmoreuniformwhendifferentsubstantiallyrectangularwavelikepartsareprovided
incombinationinthecyclicstructure(workingexample5).
(4) Yield(productivity)ishigherwhenaprotruding/depressedstructure,whichinducesadisturbanceattheinterface
ofthefluids,ispromoted(workingexamples1-5),comparedtowhenaprotruding/depressedstructureisnotprovided
(comparativeexample1).
(5) By providing acyclicstructure and increasing thenumberofliquidcolumns, a largeryieldofliquiddroplets having
small and highlyuniformdiameterscanbeobtained(large-scaleproduction).

(Working example 6) Microcapsulation

[0118] The particles formed in working example 5 were micro-capaculated by forming films by the above-described
coacervationmethod. The particle diameter of the microcapsules was 42 μm, and the dispersion value was 1.5%. By
applyingthesemicrocapsulesonafilmsubstrate, ahiglydensecapsulefilmhavingafillingstructurewasformed.

[0119] In the above embodiments, the protruding/depressed structure is permanently provided on the walls of the
mergedflowpassage12. However, the protruding/depressedstructurecanappearonlywhennecessary. Forexample,
the flowpassagestructurecanbemadeofarubbermaterial,thewallsoftemergedflowpassage12canhaveportions
diffferenthardnesslevelswithintervalstherebetweenintheflowdirectionofthefluid, and theportionswithlowhardness
levelscanbedepressedduetopressureofthefluid. The protruding/depressed cyclic structure can berealized in this
manner.

[0120] In an eighth embodiment shown in FIG. 20, soft parts 40a having lowhardnesslevels are provided along a
wall of a flowpassagestructure40, and movable rods 41 for controlling the soft parts 40a can press the soft parts 40a
todepressthewallinward.

[0121] By adjusting the strokes of the movable rods 41, the depths of the protruding/depressedstructurecanbefinely
controlled.

[0122] Accordingtooneembodimentofthepresentinvention,itispossibletoinduceformationofliquiddropletswith
asimplestructurewithoutusingapietoelectricelementtoapplymechanicaloscillations. Accordingly, liquiddroplets
withuniformdiameterscanbegenerated.

[0123] Further, according to one embodiment of the present invention, monodispersive liquid droplets can be stably
generated from all flow passages.

[0124] Further, according to one embodiment of the present invention, liquid droplets can be stably and continuously
generated at a high-speed cycle.

[0125] Further, according to one embodiment of the present invention, satellites are prevented from being generated.

[0126] Further, according to one embodiment of the present invention, microcapsules with uniform particle diameters
canbegenerated. By usingthesemicrocapsulesascataphoresistypedisplaydevices, quality of the reproduced image
isenhanced.

[0127] Further, according to one embodiment of the present invention, when microscopic particles obtained by solid-
ifying the liquid droplets are used as display particles such as toner, highly precise image quality can be achieved.

[0128] The present invention is not limited to the specifically disclosed embodiment, and variations and modifications
may be made without departing from the scope of the present invention.

Claims

1. A microscopic flow passage structure (2) for generating microscopic liquid droplets, the microscopic flow passage
structure comprising:

- fluid introduction flow passages (10, 11) configured to individually introduce plural types of fluids that are in-
  compatible with each other;
- a merged flow passage (12) in which the fluids merge;
- a common outlet (13) through which the fluids can be discharged from the merged flow passage,
wherein the merged flow passage (12) includes a region in which a cross-sectional area of the merged flow
passage changes cyclically in a fluid flow direction (F), characterized in that
- a cyclic wavelength (λ) of the changes in a cross-sectional area of the region is 3.0 to 10.0 times a circle-
equivalent diameter (d(jet)) of a fluid to be turned into the liquid droplets immediately before the fluids merge.

2. The microscopic flow passage structure according to claim 1, wherein the fluid introduction flow passages (10, 11)
include at least one passage configured to introduce a fluid to be turned into the liquid droplets,
the passage configured to introduce the fluid to be turned into the liquid droplets includes an outlet having a cross-
sectional shape with a length in a depth direction and a length in a width direction, the width direction being a direction in which the fluid introduction flow passages are arranged and the depth direction being a direction orthogonal to the width direction, and a value of “the length in the depth direction/the length in the width direction” is one or more.

3. The microscopic flow passage structure according to claim 2, wherein either a length in the depth direction or a length in the width direction of the cross-sectional area of the said region in the merged flow passage (12) changes.

4. The microscopic flow passage structure according to any one of claims 1 to 3, wherein the fluid introduction flow passages (10, 11) are arranged in such a manner that two or more different types of the fluids are arranged alternately in parallel in a direction orthogonal to the fluid flow direction (F).

5. The microscopic flow passage structure according to claim 4, wherein among the fluids in the merged flow passage (12), fluids (23) in contact with walls (22) of the merged flow passage are continuous phases.

6. The microscopic flow passage structure according to any one of claims 1 to 5, wherein the said region includes a cyclic protruding/depressed structure (14, 18, 20).

7. The microscopic flow passage structure according to claim 6, wherein the cyclic protruding/depressed structure (14, 18, 20) has a rectangular wave shape or a rectangular-like wave shape.

8. The microscopic flow passage structure according to claim 6, wherein the cyclic protruding/depressed structure (14, 18, 20) has a shape that is a combination of different rectangular wave shapes or rectangular-like wave shapes.

9. The microscopic flow passage structure according to claim 6, wherein walls of the merged flow passage (12) include portions of different hardness levels with intervals therebetween in the fluid flow direction, and the cyclic protruding/depressed structure (14, 18, 20) appears when portions of low hardness levels are depressed by pressure of the fluids.

10. A microscopic liquid droplet generating system comprising the microscopic flow passage structure (2) according to any one of claims 1 to 9.

11. The microscopic liquid droplet generating system according to claim 10, further comprising:

   a unit (30) configured to form a film on each of the liquid droplets generated by the microscopic flow passage structure (2).

12. A microscopic liquid droplet generating method with the structure of claim 1, comprising the steps of:

   (a) merging plural types of fluids that are incompatible with each other;
   (b) inducing a disturbance at an interface between the fluids by causing the fluids to flow through a merged flow passage (12) having a cross-sectional area that changes cyclically in a fluid flow direction (F); and
   (c) turning one of the types of the fluids into liquid droplets, characterized in that a cyclic wavelength (λ) of the changes in a cross-sectional area of the merged flow passage (12) is 3.0 to 10.0 times a circle-equivalent diameter (d(jet)) of a fluid to be turned into the liquid droplets immediately before the fluids merge.

13. The microscopic liquid droplet generating method according to claim 12, wherein the step (a) includes introducing the fluids in such a manner that two or more different types of the fluids are arranged alternately in parallel in a direction orthogonal to a fluid flow direction (F), before merging the fluids.

14. The microscopic liquid droplet generating method according to claim 12 or 13, further comprising a step of:

   (d) forming a film on each of the liquid droplets at a downstream side of the merged flow passage in a fluid flow direction (F).

15. A particle forming method comprising the step of solidifying the liquid droplets generated by the microscopic liquid droplet generating method according to claim 12 or 13.
16. A microcapsule generating method comprising the step of forming microcapsules from the liquid droplets generated by the microscopic liquid droplet generating method according to claim 14.

patentansprüche

1. Mikroskopisches Strömungsdurchgangselement (2) zum Erzeugen mikroskopischer Flüssigkeitströpfchen, wobei das mikroskopische Strömungsdurchgangselement Folgendes umfasst:

   Fluideinführungsströmungsdurchgänge (10, 11), die für das einzelne Einführen mehrere Typen von Fluiden konfiguriert sind, die miteinander unverträglich sind;
   einen Zusammenführungströmungsdurchgang (12), in dem die Fluide zusammengeführt werden;
   einen gemeinsamen Auslass (13), durch die die Fluide aus dem Zusammenführungsströmungsdurchgang abgelassen werden können;
   wobei der Zusammenführungströmungsdurchgang (12) eine Region umfasst, in der ein Querschnittsbereich des Zusammenführungsströmungsdurchgangs sich zyklisch in einer Fluidströmungsrichtung (F) ändert, dadurch gekennzeichnet, dass
   eine zyklische Wellenlänge ($\lambda$) der Änderungen in einem Querschnittsbereich der Region das 3,0- bis 10,0-fache eines kreisäquivalenten Durchmessers (d Strahl) eines Fluids beträgt, das direkt bevor die Fluide zusammengeführt werden in die Flüssigkeitströpfchen umgewandelt werden soll.

2. Mikroskopisches Strömungsdurchgangselement nach Anspruch 1, wobei die Fluideinführungsströmungsdurchgänge (10, 11) mindestens einen Durchgang umfassen, der zum Einführen eines Fluids konfiguriert ist, das in die Flüssigkeitströpfchen umgewandelt werden soll, der Durchgang, der zum Einführen des Fluids konfiguriert ist, das in die Flüssigkeitströpfchen umgewandelt werden soll, einen Auslass umfasst, der eine Querschnittsgestalt mit einer Länge in Tiefenrichtung und einer Länge in Breitenrichtung aufweist, wobei die Breitenrichtung eine Richtung ist, in der die Fluideinführungsströmungsdurchgänge angeordnet sind, und die Tiefenrichtung eine Richtung ist, die orthogonal zur Breitenrichtung liegt und ein Wert "der Länge in Tiefenrichtung/der Länge in Breitenrichtung" eins oder mehr beträgt.

3. Mikroskopisches Strömungsdurchgangselement nach Anspruch 2, wobei entweder eine Länge in Tiefenrichtung oder eine Länge in Breitenrichtung des Querschnittsbereichs der Region sich im Zusammenführungströmungsdurchgang (12) ändert.

4. Mikroskopisches Strömungsdurchgangselement nach einem der Ansprüche 1 bis 3, wobei die Fluideinführungsströmungsdurchgänge (10, 11) derart angeordnet sind, das zwei oder mehrere verschiedene Typen der Fluide abwechselnd parallel in einer Richtung angeordnet sind, die orthogonal zur Fluidströmungsrichtung (F) liegt.


6. Mikroskopisches Strömungsdurchgangselement nach einem der Ansprüche 1 bis 5, wobei die Region ein zyklisch hervorspringendes/eingedrücktes Element (14, 18, 20) umfasst.

7. Mikroskopisches Strömungsdurchgangselement nach Anspruch 6, wobei das zyklisch hervorspringende/eingedrückte Element (14, 18, 20) eine rechteckige Wellengestalt oder eine rechteckähnliche Wellengestalt aufweist.

8. Mikroskopisches Strömungsdurchgangselement nach Anspruch 6, wobei das zyklisch hervorspringende/eingedrückte Element (14, 18, 20) eine Gestalt aufweist, die eine Kombination verschiedener rechteckiger Wellengestalten oder rechteckähnlicher Wellengestalten ist.

9. Mikroskopisches Strömungsdurchgangselement nach Anspruch 6, wobei Wände des Zusammenführungströmungsdurchgangs (12) Teile verschiedener Härteniveaus mit Abständen dazwischen in Fluidströmungsrichtung umfassen und das zyklisch hervorspringende/eingedrückte Element (14, 18, 20) erscheint, wenn Teile geringer Härteniveaus durch den Druck der Fluide heruntergedrückt werden.
10. Mikroskopische Flüssigkeitströpfchen erzeugendes System umfassend das mikroskopische Strömungsdurchgangselement (2) nach einem der Ansprüche 1 bis 9.

11. Mikroskopische Flüssigkeitströpfchen erzeugendes System nach Anspruch 10, des Weiteren umfassend:

   eine Einheit (30), die zum Bilden eines Films auf jedem der Flüssigkeitströpfchen konfiguriert ist, die durch das mikroskopische Strömungsdurchgangselement (2) erzeugt werden.

12. Verfahren zum Erzeugen mikroskopischer Flüssigkeitströpfchen mit dem Element nach Anspruch 1, umfassend die Schritte des:

   (a) Zusammenführens mehrerer Typen von Fluiden, die miteinander unverträglich sind;
   (b) Einführens einer Störung an der Grenzfläche zwischen den Fluiden durch Verursachen, dass die Fluide durch einen Zusammenführungsströmungsdurchgang (12) strömen, der einen Querschnittsbereich aufweist, der sich zyklisch in Fluidströmungsrichtung (F) ändert; und
   (c) Umwandlens eines der Typen der Fluide in Flüssigkeitströpfchen, dadurch gekennzeichnet, dass eine zyklische Wellenlänge (λ) der Änderungen in einem Querschnittsbereich des Zusammenführungsdurchgangs (12) das 3,0-bis 10,0-fache eines kreisäquivalenten Durchmessers (d(jet)) eines Fluids beträgt, das direkt bevor die Fluide zusammengeführt werden in Flüssigkeitströpfchen umgewandelt werden soll.

13. Verfahren zum Erzeugen mikroskopischer Flüssigkeitströpfchen nach Anspruch 12, wobei der Schritt (a) das Einführen der Fluide auf derartige Weise umfasst, dass zwei oder mehrere verschiedene Typen der Fluide vor dem Zusammenführen der Fluide abwechselnd parallel in einer Richtung angeordnet sind, die orthogonal zu einer Fluidströmungsrichtung (F) liegt.

14. Verfahren zum Erzeugen mikroskopischer Flüssigkeitströpfchen nach Anspruch 12 oder 13, des Weiteren einen Schritt umfassend des:

   (d) Bildens eines Films auf jedem der Flüssigkeitströpfchen auf einer stromabwärts liegenden Seite des Zusammenführungsdurchgangs in einer Fluidströmungsrichtung (F).

15. Verfahren zum Bilden von Teilchen umfassend den Schritt des Verfestigens der Flüssigkeitströpfchen, die durch das Verfahren zum Erzeugen mikroskopischer Flüssigkeitströpfchen nach Anspruch 12 oder 13 erzeugt werden.


Revendications

1. Structure pour le passage d’un écoulement microscopique (2) permettant de produire des gouttelettes microscopiques de liquide, la structure pour le passage d’un écoulement microscopique comprenant:

   des passages d’écoulement pour l’introduction d’un fluide (10, 11) configurés afin d’introduire individuellement plusieurs types de fluides qui sont incompatibles les uns avec les autres;
   un passage d’écoulements fusionnés (12) dans lequel les fluides fusionnent;
   un orifice de sortie commun (13) par lequel les fluides peuvent être évacués depuis le passage d’écoulements fusionnés,

   dans laquelle le passage des écoulements fusionnés (12) comprend une région dans laquelle une zone transversale du passage des écoulements fusionnés change de manière cyclique dans un sens d’écoulement des fluides (F), caractérisée en ce que
   une longueur d’onde cyclique (λ) des changements dans une zone transversale de la région est 3,0 à 10,0 fois un diamètre équivalent au cercle (d(jet)) d’un liquide qui doit être transformé en les gouttelettes de liquide immédiatement avant que les fluides ne fusionnent.

2. Structure pour le passage d’un écoulement microscopique selon la revendication 1, dans laquelle les passages des écoulements pour l’introduction des fluides (10, 11) comprennent au moins un passage configuré pour introduire
un liquide qui doit être transformé en les gouttelettes de liquide,
le passage configuré pour introduire le liquide qui doit être transformé en les gouttelettes de liquide comprend un
orifice de sortie ayant une forme transversale d’une longueur dans un sens de la profondeur et d’une longueur dans
un sens de la largeur, le sens de la largeur étant un sens dans lequel les passages des écoulements pour l’introduction
des liquides sont disposés et le sens de la profondeur étant un sens orthogonal au sens de la largeur, et
une valeur de « la longueur dans le sens de la profondeur/la longueur dans le sens de la largeur » est de un ou plus.

3. Structure pour le passage d’un écoulement microscopique selon la revendication 2, dans laquelle l’une ou l’autre
d’une longueur dans le sens de la profondeur ou d’une longueur dans le sens de la largeur de la zone transversale
de ladite région dans le passage des écoulements fusionnés (12) change.

4. Structure pour le passage d’un écoulement microscopique selon l’une quelconque des revendications 1 à 3, dans
laquelle les passages des écoulements pour l’introduction de liquide (10, 11) sont disposés d’une manière telle que
deux ou plusieurs types différents de liquides sont disposés alternativement en parallèle dans un sens orthogonal
au sens d’écoulement des liquides (F).

5. Structure pour le passage d’un écoulement microscopique selon la revendication 4, dans laquelle parmi les fluides
dans le passage des écoulements fusionnés (12), les liquides (23) en contact avec les parois (22) du passage des
écoulements fusionnés sont des phases continues.

6. Structure pour le passage d’un écoulement microscopique selon l’une quelconque des revendications 1 à 5, dans
laquelle ladite région comprend une structure cyclique faisant saillie/déprimée (14, 18, 20).

7. Structure pour le passage d’un écoulement microscopique selon la revendication 6, dans laquelle la structure
cyclique faisant saillie/déprimée (14, 18, 20) a une forme d’onde rectangulaire ou une forme d’onde de type rect-
tangulaire.

8. Structure pour le passage d’un écoulement microscopique selon la revendication 6, dans laquelle la structure
cyclique faisant saillie/déprimée (14, 18, 20) a une forme qui est une combinaison de différentes formes d’onde
rectangulaire ou formes d’onde de type rectangulaire.

9. Structure pour le passage d’un écoulement microscopique selon la revendication 6, dans laquelle les parois du
passage des écoulements fusionnés (12) comprennent des portions de niveau de dureté différente avec des inter-
valles entre elles dans le sens d’écoulement des liquides, et
la structure cyclique faisant saillie/déprimée (14, 18, 20) apparaît lorsque les portions de niveau de faible dureté
sont déprimées sous la pression des liquides.

10. Système de production de gouttelettes microscopiques de liquide comprenant la structure pour le passage d’un
écoulement microscopique (2) selon l’une quelconque des revendications 1 à 9.

11. Système de production de gouttelettes microscopiques de liquide selon la revendication 10, comprenant en outre:
une unité (30) configurée pour former un film sur chacune des gouttelettes de liquide produites par la structure
pour le passage d’un écoulement microscopique (2).

12. Procédé de production de gouttelettes microscopiques de liquide et de la structure selon la revendication 1 com-
prenant les étapes de:

(a) fusion de plusieurs types de liquides qui sont incompatibles les uns avec les autres;
(b) induction d’une perturbation à l’interface entre les liquides en faisant s’écouler les liquides à travers un
passage d’écoulements fusionnés (12) ayant une zone transversale qui change de manière cyclique dans le
sens d’écoulement des liquides (F); et
(c) transformer l’un des types de liquides en gouttelettes de liquide, caractérisé en ce qu’une longueur d’onde
cyclique (λ) des changements dans une zone transversale du passage d’écoulements fusionnés (12) est 3,0
à 10,0 fois un diamètre équivalent à un cercle (d(jet)) d’un liquide à transformer en gouttelettes de liquide
immédiatement avant que les liquides ne fusionnent.

13. Procédé de production de gouttelettes microscopiques de liquide selon la revendication 12, dans laquelle l’étape
(a) comprend l’introduction des liquides d’une manière telle que deux ou plusieurs types différents de liquides soient disposés alternativement en parallèle dans un sens orthogonal au sens d’écoulement des liquides (F), avant la fusion des liquides.

14. Procédé de production de gouttelettes microscopiques de liquide selon la revendication 12 ou 13, comprenant en outre une étape de:

(d) formation d’un film sur chacune des gouttelettes de liquide du côté situé en aval du passage des écoulements fusionnés dans un sens d’écoulement des liquides (F).

15. Procédé de formation de particules comprenant l’étape de solidification des gouttelettes de liquide produites par le procédé de production de gouttelettes microscopiques de liquide selon la revendication 12 ou 13.

16. Procédé de production de microcapsules comprenant l’étape de formation de microcapsules à partir des gouttelettes de liquide produites par le procédé de production de gouttelettes microscopiques de liquide selon la revendication 14.
FIG. 23A

WIDTH OF FLOW PASSAGE

FIG. 23B

DEPTH OF FLOW PASSAGE

FIG. 23C
FIG. 24

(WIDTH DIRECTION

ASPECT RATIO 1.5)

FIG. 25

(DEPTH DIRECTION

ASPECT RATIO 0.5)
FIG.26

(ASPECT RATIO 4)

FIG.27

(ASPECT RATIO 0.5)
REFERENCES CITED IN THE DESCRIPTION

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Patent documents cited in the description

- JP 3476223 B [0004]
- JP 3218445 B [0004]
- JP 2733729 B [0004]
- JP 2000273188 A [0004]
- JP 2004358386 A [0004]
- JP 2004243308 A [0004]
- JP 2004197083 A [0004]
- JP 2005185877 A [0004]
- JP 2005213334 A [0004]
- JP 2005238118 A [0004]
- JP 2005238120 A [0004]

Non-patent literature cited in the description