(19) Europäisches Patentamt
European Patent Office
Office européen des brevets

(11) EP 2 266 691 A1

(12) # EUROPEAN PATENT APPLICATION
published in accordance with Art. 153(4) EPC

(43) Date of publication:
29.12.2010 Bulletin 2010/52

(21) Application number: **09723884.4**

(22) Date of filing: **24.03.2009**

(51) Int Cl.:
B01J 19/00 (2006.01) ***B01F 3/08*** (2006.01)
B01F 5/00 (2006.01) ***B01J 13/00*** (2006.01)
B81B 1/00 (2006.01)

(86) International application number:
PCT/JP2009/055816

(87) International publication number:
WO 2009/119578 (01.10.2009 Gazette 2009/40)

(84) Designated Contracting States:
AT BE BG CH CY CZ DE DK EE ES FI FR GB GR HR HU IE IS IT LI LT LU LV MC MK MT NL NO PL PT RO SE SI SK TR
Designated Extension States:
AL BA RS

(30) Priority: **25.03.2008 JP 2008079375**

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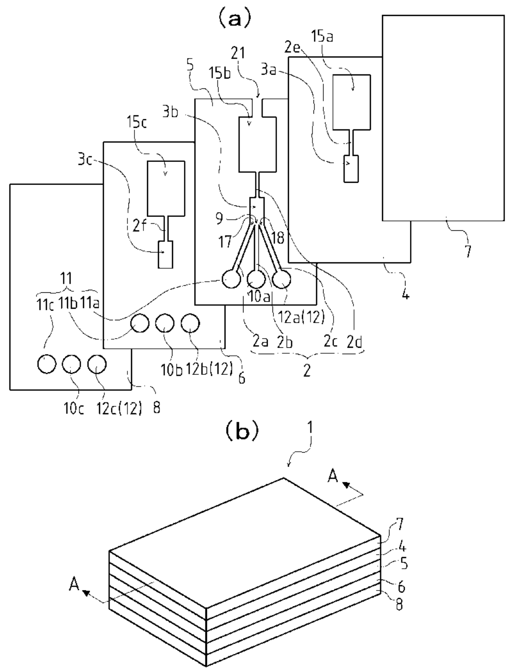
(54) **MICRO-DROPLET DISPENSING DEVICE**

(57) There is provided a microreactor that has a simple structure and prepares stable monodisperse droplets over a long period of time, with simple modifications to a conventional microreactor.

A microreactor 1 that includes flow channels 2 formed by the lamination of a plurality of substrates so as to allow fluids to flow therethrough, and prepares droplets by causing a continuous phase fluid to shear a dispersed phase fluid, has an integrated lamination of: a flow-channel substrate 5 including the flow channels 2 formed open, through which the continuous phase fluid and the dispersed phase fluid flow, and also including a merge section 3b formed open where the flow channels 2 merge; spacer substrates 4 and 6, either or both joined to one side or respective sides of the flow-channel substrate 5 and including space portions 3a and 3c each formed so as to have a predetermined space in a corresponding one of the vertical directions with respect to the merge section 3b of the flow-channel substrate 5; and cover substrates 7 and 8 joined to respective outer sides of the lamination of: the spacer substrates 4 and 6 and the flow-channel substrate 5, the spacer substrates 4 and 6 joined to the respective sides of the flow-channel substrate 5; or the flow-channel substrate 5 and the one of the spacer substrates 4 and 6, the spacer substrate joined to the one side of the flow-channel substrate 5, so as to close both outer sides.

Fig.1



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Description

Technical Field

[0001] The present invention relates to a microdroplet preparation device used for emulsification employing micro flow channels having microstructures.

Background Art

[0002] Conventionally, it has been shown in many cases that it is effective to use micro flow channels for emulsification, such as the dispersion of oil into water and the dispersion of water into oil. Among those cases, reported as a conventional technique is an emulsification technique where Y-shaped, or T-shaped, two-dimensional flow channels are engraved on a substrate, so as to prepare monodisperse droplets by the shear force between two fluids. Further, also known as a conventional technique is microchannel emulsification where micropores termed "microchannels" are provided for a similar purpose, so as to prepare monodisperse droplets (see Patent Literature 1, for example).

Among such techniques, one drawing attention is an emulsification technique (so-called microchannel branching emulsification) using a microreactor (microdroplet preparation device) including Y-shaped, or T-shaped, micro flow channels. This is because the technique has a smaller pressure loss than microchannel emulsification, and obtains monodisperse droplets smaller than the width of each of the flow channels.

Patent Literature 1: Japanese Laid-Open Patent Publication No. 2001-181309

Disclosure of Invention

Problems to Be Solved By the Invention

[0003] It is considered that a Y-shaped, or T-shaped, microreactor prepares droplets by causing a continuous phase fluid to shear a dispersed phase fluid near the part where the two fluids merge. When the continuous phase is an aqueous phase and the dispersed phase is an oil phase, it may be impossible to form droplets, depending on the properties of the dispersed oil phase. The cause is assumed to be the adsorption (attachment) of the oil-phase ingredients to the wall surfaces of the flow channels. In response to this, a method is studied of preventing oil-phase ingredients from adsorbing to the wall surfaces of the flow channels, by performing hydrophilic treatment, such as ozone gas treatment, on the entire wall surfaces of the flow channels in order to obtain stable droplets. However, the hydrophilization of the wall surfaces of the flow channels by normal surface treatment is effective only for a short period of time (for about a couple of days), and therefore is not effective in the continuous operation of an emulsification device, where emulsification is continuously performed using a microreactor.

[0004] In addition, a conventional microreactor including flow channels has a structure where a substrate including the flow channels is sandwiched between cover substrates closing the top and bottom, respectively, of the substrate, and therefore, an oil phase may flow while becoming attached to the rear surface (the ceiling portion of the flow channels) of the top cover substrate or the rear surface (the bottom portion of the flow channels) of the bottom cover substrate. This makes it difficult to prepare monodisperse droplets.

[0005] In view of the above problems, it is an object of the present invention to provide a microdroplet preparation device that has a simple structure and prepares stable monodisperse droplets over a long period of time, with simple modifications to a microreactor.

Means of Solving the Problems

[0006] A microdroplet preparation device according to the present invention is a microdroplet preparation device that prepares droplets by causing a continuous phase fluid to shear a dispersed phase fluid, wherein

both or at least one of a ceiling portion and a bottom portion of flow channels where the continuous phase fluid and the dispersed phase fluid merge are formed so as to have a predetermined distance from a flow of the fluid formed by the merger of the continuous phase fluid and the dispersed phase fluid.

[0007] In the microdroplet preparation device according to the present invention,

the flow channels are formed by an integrated lamination of:

- a flow-channel substrate including flow channels formed open through which the continuous phase fluid and the dispersed phase fluid flow, and also including a merge section formed open where the flow channels merge;
- at least one spacer substrate joined to one side or respective sides of the flow-channel substrate and including a space portion formed so as to have a predetermined space in a corresponding one of vertical directions with respect to the merge section of the flow-channel substrate; and
- cover substrates joined to respective outer sides of the lamination of: the spacer substrates and the flow-channel substrate, the spacer substrates joined to the respective sides of the flow-channel substrate; or the flow-channel substrate and the one of the spacer substrates, the spacer substrate joined to the one side of the flow-channel substrate, so as to close both outer sides.

[0008] In the microdroplet preparation device according to the present invention,

upstream of the flow channels which are formed open in the flow-channel substrate and through which the con-

tinuous phase fluid and the dispersed phase fluid flow, predetermined openings are provided in communication with the respective flow channels,
one of the spacer substrates joined to the respective sides of the flow-channel substrate, and the cover substrate joined to the outer side of the spacer substrate so as to close the outer side, each include openings so as to correspond in position to the predetermined openings, and
the flow-channel substrate, the spacer substrate including the corresponding openings, and the cover substrate including the corresponding openings are laminated, so as to form continuous phase supply sections for supplying a continuous phase and a dispersed phase supply section for supplying a dispersed phase, such that the continuous phase supply sections and the dispersed phase supply section are in communication with the respective flow channels.

[0009] In the microdroplet preparation device according to the present invention,
the space portions of the spacer substrates joined to the respective sides of the flow-channel substrate are placed so as to correspond in position to respective outer sides of the merge section formed open in the flow-channel substrate.

[0010] In the microdroplet preparation device according to the present invention,
the continuous phase shears the dispersed phase within and on an upstream side of the merge section of the flow-channel substrate.

[0011] In the microdroplet preparation device according to the present invention,
each of the space portions formed in the spacer substrates joined to the respective sides of the flow-channel substrate is a recess excavated, or an opening formed open, so as to correspond in position to the merge section formed open in the flow-channel substrate.

[0012] In the microdroplet preparation device according to the present invention,
the space portions formed in the spacer substrates joined to the respective sides of the flow-channel substrate have the same shape.

[0013] In the microdroplet preparation device according to the present invention,
the spacer substrates joined to the respective sides of the flow-channel substrate have the same thickness.

[0014] In the microdroplet preparation device according to the present invention,
each of the spacer substrates joined to the respective sides of the flow-channel substrate and including the respective space portions each formed so as to have a predetermined space in the corresponding one of the vertical directions with respect to the merge section of the flow-channel substrate is integrated with the corresponding one of the cover substrates each joined to the corresponding outer side of the corresponding spacer substrate so as to close both outer sides, and
the space portions are provided so as to correspond in position to the respective outer sides of the merge section of the flow-channel substrate, and an integrated lamination is formed so as to sandwich the flow-channel substrate, between both the integrated substrates, from the respective sides of the flow-channel substrate such that positions of the space portions match a position of the merge section of the flow-channel substrate.

Effect of the Invention

[0015] With the microdroplet preparation device according to the present invention, it is possible to prevent droplets from becoming attached to the wall surfaces of the flow channels, and therefore possible to prepare stable monodisperse droplets over a long period of time.

[0016] With the microdroplet preparation device according to the present invention, it is possible to emulsify even a dispersed phase containing ingredients having a high viscosity or a high hydrophobicity.

[0017] With the microdroplet preparation device according to the present invention, it is possible to achieve stable emulsification semipermanently without performing surface treatment on the wall surfaces of the flow channels of the microdroplet preparation device.

Brief Description of Drawings

[0018]

Fig. 1 is a diagram showing the structure of a microreactor according to an embodiment of the present invention: (a) is a plan view showing the shapes of substrates; and (b) is a perspective view of the microreactor.

Fig. 2 is a perspective view showing the state where spacer substrates are placed on and under a substrate.

Fig. 3 is a cross-sectional view along an arrow A-A in Fig. 1(b).

Fig. 4 is a cross-sectional view showing another embodiment of the microreactor.

Fig. 5 is a photograph showing monodisperse droplets prepared by the microreactor.

Fig. 6 is a photograph showing a part of a flow-channel substrate according to a comparative example.

Fig. 7 is a photograph showing a dispersed phase according to the comparative example, where droplets are not formed.

Description of the Reference Characters

[0019]

1	microreactor (microdroplet preparation device)
2	flow channel
2a, 2c	continuous phase flow channel
2b	dispersed phase flow channel
3b	merge section

3a, 3c	space portion
4, 6	spacer substrate
5	flow-channel substrate
7, 8	cover substrate

The Best Mode for Carrying Out the Invention

[0020] With reference to the drawings, a description is given below of the best embodiments of the present invention. It is noted that common components in all the figures are denoted by the same reference characters, and are not redundantly described. It is also noted that the lamination direction is defined as the up-down direction.

A microdroplet preparation device according to the present invention is a device including micro flow channels, such as a microreactor, that prepares microdroplets by causing a continuous phase fluid to shear a dispersed phase fluid, and is hereinafter referred to as a "microreactor" for convenience.

Fig. 1 is a diagram showing the structure of a microreactor according to an embodiment of the present invention: (a) is an exploded plan view showing the shapes of substrates; and (b) is a perspective view of the microreactor. Fig. 2 is a perspective view showing the state where spacer substrates are placed on and under a flow-channel substrate. Fig. 3 is a cross-sectional view along an arrow A-A in Fig. 1(b). Fig. 4 is a cross-sectional view showing another embodiment of the microreactor. Fig. 5 is a photograph showing monodisperse droplets prepared by the microreactor. Fig. 6 is a photograph showing a part of a flow-channel substrate according to a comparative example. Fig. 7 is a photograph showing a dispersed phase according to the comparative example, where droplets are not formed.

[0021] The microreactor according to the present invention is a microreactor that prepares droplets by causing a continuous phase fluid to shear a dispersed phase fluid. In flow channels where the continuous phase fluid and the dispersed phase fluid merge, both or at least one of the ceiling portion and the bottom portion of the flow channels are formed so as to have a predetermined distance from the flow of the fluid formed by the merger of the continuous phase fluid and the dispersed phase fluid.

[0022] That is, the microreactor according to the present invention includes predetermined flow channels formed to prepare droplets by causing a continuous phase fluid to shear a dispersed phase fluid. The microreactor includes flow channels formed so as to have a predetermined distance from the flow of a dispersed phase (monodisperse phase), which is a fluid prepared by: introducing a continuous phase fluid and a dispersed phase fluid through a plurality of branching flow channels, correspondingly; causing the introduced fluids to merge by the arrangement of the plurality of flow channels at predetermined angles from one another; and using the shear force acting when the fluids merge.

That is, in the microreactor according to the present invention, in a flow-channel-forming portion, which is the sections where the fluids are caused to merge as described above, both or at least one of the ceiling portion and the bottom portion of the flow-channel-forming portion are formed so as to have a predetermined distance from the flow of the fluid formed by the merger of the fluids. This prevents the dispersed phase droplets from becoming attached to the wall surfaces of the flow channels (the wall surfaces of the flow-channel-forming portion), and therefore enables the preparation of stable monodisperse droplets over a long period of time.

It is noted that the shapes of the ceiling portion and the bottom portion are not particularly limited, so long as predetermined spaces can be formed such that each of the ceiling portion and the bottom portion has a predetermined distance from the flow of the monodisperse phase prepared by the merger of the fluids, so that the monodisperse phase does not become attached to the ceiling portion or the bottom portion. A detailed description is given below of a specific embodiment.

[0023] In the present embodiment, the descriptions are given taking as a specific example the process of preparing a monodisperse O/W emulsion, which contains minute droplets of the same size, using a lamination-type microreactor 1 (hereinafter referred to as a "microreactor 1") that includes micro flow channels of shapes as shown in Fig. 1(a), and prepares a droplet dispersion system (prepares an O/W emulsion).

[0024] With reference to Figs. 1, 2, and 3, a description is given of the embodiment of the microreactor according to the present invention.

[0025] As shown in Fig. 1(a) and (b), the microreactor 1 is a microreactor that includes flow channels 2 formed by the lamination of a plurality of substrates so as to allow fluids to flow therethrough, and prepares droplets by causing a continuous phase fluid to shear a dispersed phase fluid. As shown in Fig. 1(b), the microreactor 1 has an integrated lamination of: a flow-channel substrate 5 including a merge section 3b where the continuous phase fluid and the dispersed phase fluid merge, and also including the flow channels 2 formed open; a first spacer substrate 4 joined to one side (the top side, in the present embodiment) of the flow-channel substrate 5 and including a space portion 3a described later that is formed so as to have a predetermined space (distance) vertically on the merge section 3b; a second spacer substrate 6 joined to the other side (the bottom side, in the present embodiment) of the flow-channel substrate 5 and including a space portion 3c described later that is formed so as to have a predetermined space (distance) vertically under the merge section 3b; and cover substrates 7 and 8 joined to the respective outer sides of the first spacer substrate 4 and the second spacer substrate 6, i.e., the top side of the first spacer substrate 4 and the bottom side of the second spacer substrate 6, respectively, so as to close both outer sides. A detailed description is given below of the structure of the microreactor 1.

It is noted that in the present embodiment, for convenience, of the inner wall surfaces of the flow channels 2 formed by the lamination of all the substrates, the surface placed on the inner side of the top cover substrate 7 is defined as a ceiling portion 7a, and the surface placed on the inner side of the bottom cover substrate is defined as a bottom portion 8a (see Fig. 3).

In addition, in the present embodiment, the first spacer substrate 4 and the second spacer substrate 6 are joined to the respective sides of the flow-channel substrate 5; however, the present invention is not particularly limited to this. The structure may be such that either one of the first spacer substrate 4 and the second spacer substrate 6 is joined to one side of the flow-channel substrate 5, and the cover substrate 7 (8) is joined to the other side.

[0026] The flow-channel substrate 5 is a plate-like member formed of thin stainless steel (50 μm thick), and includes the flow channels 2 of predetermined patterns that are open. The flow channels 2 include: a dispersed phase flow channel 2b (a flow channel width of 50 μm), through which a dispersed phase (an oil phase, in the present embodiment) flows; two continuous phase flow channels 2a and 2c (a flow channel width of 70 μm), through which a continuous phase (an aqueous phase, in the present embodiment) flows and which are placed on the respective sides of the dispersed phase flow channel 2b; the merge section 3b described later; and a droplet-forming flow channel 2d described later. One end of the dispersed phase flow channel 2b is in communication with a dispersed phase supply section 10a, which is a predetermined opening placed upstream of the dispersed phase flow channel 2b and formed open in a circular shape in plan view. The other end of the dispersed phase flow channel 2b is in communication with the merge section 3b of a substantially quadrangular shape in plan view, placed downstream of the dispersed phase flow channel 2b. Further, one end of each of the continuous phase flow channels 2a and 2c is in communication with a corresponding one of continuous phase supply sections 11a and 12a, which are predetermined openings placed upstream of the respective continuous phase flow channels 2a and 2c and formed open in circular shapes in plan view. The continuous phase flow channels 2a and 2c are placed on the respective sides of the dispersed phase flow channel 2b such that the further downstream, the closer the continuous phase flow channels 2a and 2c are to the dispersed phase flow channel 2b. The other ends of the continuous phase flow channels 2a and 2c are in communication with the merge section 3b of a substantially quadrangular shape in plan view, placed downstream of the dispersed phase flow channel 2b. The other end of the dispersed phase flow channel 2b includes a dispersed phase discharge outlet 9, through which the oil phase is discharged into the merge section 3b. On the respective horizontal sides of the dispersed phase discharge outlet 9, continuous phase discharge outlets 17 and 18 included in the other ends of the respective continuous phase flow channels 2a and 2c are placed adjacent to the dispersed phase discharge outlet 9. Further, via the droplet-forming flow channel 2d that forms microdroplets, the merge section 3b is in communication with a reservoir section 15b of a quadrangular shape in plan view that stores the droplets. Furthermore, a droplet ejection section 21 is provided downstream of the reservoir section 15b.

It is noted that the shapes and the arrangement pattern of the flow channels 2a, 2b, 2c, and 2d included in the flow channels 2 are not limited to those of the present embodiment, and may be appropriately changed depending on the preparation conditions of emulsification or the like. For example, the shapes of the flow channels 2 may not only be linear, but may also be crank-shaped, or meandering. The arrangement pattern of the flow channels 2 may be Y-shaped, T-shaped, or the like.

[0027] The first spacer substrate 4 is a plate-like member formed of thin stainless steel (50 μm thick), and as shown in Fig. 1(a), includes openings of predetermined patterns so as to correspond in vertical position to the merge section 3b, the droplet-forming flow channel 2d, and the reservoir section 15b of the flow-channel substrate 5. That is, the openings of the predetermined patterns of the first spacer substrate 4 include: a space portion 3a of a quadrangular shape in plan view, formed open so as to have a predetermined space vertically on the merge section 3b of the flow-channel substrate 5; a reservoir section 15a of a quadrangular shape in plan view, formed open so as to have a predetermined space vertically on the reservoir section 15b of the flow-channel substrate 5; and a droplet-forming flow channel 2e formed open so as to have a predetermined space vertically on the droplet-forming flow channel 2d of the flow-channel substrate 5. Further, the space portion 3a is in communication with the reservoir section 15a via the droplet-forming flow channel 2e.

[0028] The second spacer substrate 6 is a plate-like member formed of thin stainless steel (50 μm thick), and as shown in Fig. 1(a), includes openings of predetermined patterns so as to correspond in vertical position to the merge section 3b, the droplet-forming flow channel 2d, the reservoir section 15b, the dispersed phase supply section 10a, and the continuous phase supply sections 11a and 12a of the flow-channel substrate 5. That is, the openings of the predetermined patterns of the second spacer substrate 6 include: a space portion 3c of a quadrangular shape in plan view, formed open so as to have a predetermined space vertically under the merge section 3b of the flow-channel substrate 5; a reservoir section 15c of a quadrangular shape in plan view, formed open so as to have a predetermined space vertically under the reservoir section 15b of the flow-channel substrate 5; a droplet-forming flow channel 2f formed open so as to have a predetermined space vertically under the droplet-forming flow channel 2d of the flow-channel substrate 5; and a dispersed phase supply section 10b and continuous phase supply sections 11b and 12b of circular shapes in plan view, formed open so as to correspond in position to the dispersed phase supply section 10a and the continuous phase supply sections 11a and 12a, respectively, when the second spacer substrate 6 is joined to the bottom side of the flow-channel substrate 5. Further, the space portion 3c is in communication with the reservoir section 15c via the droplet-forming flow channel 2f.

[0029] The cover substrates 7 and 8 are plate-like members formed of stainless steel (5 mm thick) or quartz glass, and as shown in Fig. 1(a), are joined to the respective outer sides, i.e., the top and bottom sides, respectively, of the sequential lamination of the first spacer substrate 4, the flow-channel substrate 5, and the second spacer substrate 6, so as to close both outer sides. The cover substrate 7 does not include any openings, while the cover substrate 8 includes a dispersed phase supply section 10c and continuous phase supply sections 11c and 12c of circular shapes in plan view, formed open so as to correspond in position to the dispersed phase supply section 10b and the continuous phase supply sections 11b and 12b, respectively, when the cover substrate 8 is joined to the bottom side of the second spacer substrate 6. The lamination of the substrates 5, 6, and 8 forms a dispersed phase supply section 10 and continuous phase supply sections 11 and 12, so as to include: the dispersed phase supply section 10c and the continuous phase supply sections 11c and 12c; the dispersed phase supply section 10a and the continuous phase supply sections 11a and 12a included in the flow-channel substrate 5 described above; and the dispersed phase supply section 10b and the continuous phase supply sections 11b and 12b included in the second spacer substrate 6 described above. The dispersed phase supply section 10 and the continuous phase supply sections 11 and 12 are connected to supply means, not shown, that supplies a dispersed phase and a continuous phase.

It is noted that in the present embodiment, the dispersed phase supply section 10 and the continuous phase supply sections 11 and 12 include the openings of circular shapes in plan view, formed in the second spacer substrate 6 and the cover substrate 8, so that supply means, not shown, supplies a dispersed phase and a continuous phase through the bottom side of the microreactor 1; however, the present invention is not particularly limited to this. A dispersed phase supply section and continuous phase supply sections may include openings of circular shapes in plan view, formed in each of the first spacer substrate 4 and the cover substrate 7 so as to correspond in position to the dispersed phase supply section 10a and the continuous phase supply sections 11a and 12a, so that supply means, not shown, supplies a dispersed phase and a continuous phase through the top side of the microreactor 1.

[0030] The flow-channel substrate 5, the first spacer substrate 4, and the second spacer substrate 6 are thus laminated as shown in Fig. 2. The structure is such that the lamination is formed so as to: match the positions of the openings of the supply sections 10a, 11a, and 12a, the supply sections 10b, 11b, and 12b, and the supply sections 10c, 11c, and 12c, respectively, of the substrates 5, 6, and 8; match the positions of the openings of the merge section 3b and the space portions 3a and 3c of the substrates 4, 5, and 6; match the positions of the openings of the droplet-forming flow channels 2d, 2e, and 2f of the substrates 4, 5, and 6; and match the positions of the openings of the reservoir sections 15a, 15b, and 15c of the substrates 4, 5, and 6.

[0031] As shown in Fig. 1(b), the microreactor 1 has the lamination of the flow-channel substrate 5, the first spacer substrate 4, the second spacer substrate 6, and the cover substrates 7 and 8, so as to join the substrates 4, 5, 6, 7, and 8 to one another. The substrates 4, 5, 6, 7, and 8 of the microreactor 1 thus having the integrated lamination of the substrates 4, 5, 6, 7, and 8 are firmly attached to one another in planar contact. When the periphery of the microreactor 1 is additionally fixed with a fixing member, not shown, the flow channels 2 function as micro flow channels such that even when fluids are caused to flow through the flow channels 2, an oil phase or an aqueous phase supplied through the supply sections 10, 11, and 12 does not seep through the interfaces between the substrates 4, 5, 6, 7, and 8.

[0032] Next, with reference to Fig. 3, a description is given of the state of the microreactor 1 having the lamination of the substrates 4, 5, 6, 7, and 8. Fig. 3 is a cross-sectional view along the arrow A-A, taken at the center of the short side direction of the microreactor 1 shown in Fig. 1(b).

[0033] As shown in Fig. 3, in the state of the microreactor 1 having the lamination of the substrates 4, 5, 6, 7, and 8, the space portion 3a of the first spacer substrate 4, the merge section 3b of the flow-channel substrate 5, and the space portion 3c of the second spacer substrate 6 form a merge section 3, which is a minute space, at a substantially central portion within the microreactor 1. That is, the merge section 3 is formed by placing the space portion 3a of the first spacer substrate 4 and the space portion 3c of the second spacer substrate 6 so as to correspond in position to the respective sides, i.e., the top and bottom sides, respectively, of the merge section 3b formed open in the flow-channel substrate 5. The space portion 3a formed open in the first spacer substrate 4 also forms an upper space of the merge section 3. The space portion 3c formed open in the second spacer substrate 6 also forms a lower space of the merge section 3. Further, the droplet-forming flow channel 2e of the first spacer substrate 4, the droplet-forming flow channel 2d of the flow-channel substrate 5, and the droplet-forming flow channel 2f of the second spacer substrate 6 form a droplet-forming flow channel 19. Furthermore, the reservoir section 15a of the first spacer substrate 4, the reservoir section 15b of the flow-channel substrate 5, and the reservoir section 15c of the second spacer substrate 6 form a reservoir section 15, which is a minute space, within the microreactor 1. The droplet-forming flow channel 19 is a flow channel that enables the communication between the merge section 3, which is a minute space, and the reservoir section 15, which is a minute space placed downstream of the merge section 3.

In addition, in the state of the substrates 5, 6, and 8 laminated, the dispersed phase supply section 10a and the continuous phase supply sections 11a and 12a of the flow-channel substrate 5, the dispersed phase supply section 10b and the continuous phase supply sections 11b and 12b of the second spacer substrate 6, and the dispersed phase supply section 10c and the continuous phase supply sections 11c and 12c of the cover substrate 8 form the dispersed phase supply section 10 for supplying a dispersed phase and the continuous phase supply sections 11 and 12 for supplying a continuous phase. The dispersed phase supply section 10 is in communication with the merge section 3, which is the minute space described above, via the dispersion supply phase section 10a, which is an upper space of the dispersed phase supply section 10, and also via the dispersed phase flow channel 2b. Further, the continuous phase supply sections 11 and 12 are in communication with the merge section 3, which is the minute space described above, via the continuous phase supply sections 11a and 12a, which are upper spaces of the continuous phase supply sections 11 and 12, and also via the continuous phase flow channels 2a and 2c, respectively.

It is noted that the cross-sectional view of the microreactor 1 shown in Fig. 3 is a cross-sectional view along the arrow A-A, taken at the center of the short side direction of the microreactor 1 shown in Fig. 1(b), and therefore shows cross sections of the dispersed phase supply section 10 and the dispersed phase flow channel 2b.

[0034] The lamination thus formed so as to join the first spacer substrate 4 and the second spacer substrate 6 to the top and bottom sides, respectively, of the flow-channel substrate 5 provides the upper space portion 3a having a predetermined space vertically on the merge section 3b of the flow-channel substrate 5, and also provides the lower space portion 3c having a predetermined space vertically under the merge section 3b of the flow-channel substrate 5. That is, the provision of the upper space portion 3a and the lower space portion 3c forms, as shown in Fig. 3, stepped portions 4a and 6a, respectively, near the discharge outlet 9 of the dispersed phase flow channel 2b. In other words, the shape is such that the space becomes wider outwardly than the opening of the dispersed phase discharge outlet 9 by a predetermined distance (50 μm, which is the thickness of each of the substrates 4 and 6, in the present embodiment) from the upper and lower edges of the opening.

[0035] In addition, supply means, not shown, that supplies a dispersed phase and a continuous phase is connected to the dispersed phase supply section 10 and the continuous phase supply sections 11 and 12. This supply means includes a syringe pump having a plurality of syringes, and the syringes of the syringe pump are connected to the supply sections 10, 11, and 12 via tubes, respectively. This fills each syringe with a continuous phase fluid or a dispersed phase fluid, and supplies the corresponding fluid to the corresponding supply sections 10, 11, and 12 of the microreactor 1 at a predetermined flow rate and a predetermined flow velocity by causing a controller to drive the pump, not shown.

[0036] Next, with reference to Fig. 4, a description is given of another embodiment using a microreactor similar in function to the microreactor 1 described above.

[0037] As shown in Fig. 4, a microreactor 30 has an integrated lamination of: a first spacer substrate 14 having the integration of the first spacer substrate 4 and the cover substrate 7 included in the microreactor 1 described above; a second spacer substrate 16 having the integration of the second spacer substrate 6 and the cover substrate 8 included in the microreactor 1 described above; and the flow-channel substrate 5. The integrated lamination is formed so as to sandwich the flow-channel substrate 5, between a first spacer substrate 14 and a second spacer substrate 16, from the respective sides of the flow-channel substrate 5 such that the first spacer substrate 14 and the second spacer substrate 16 correspond in position to the merge section 3b of the flow-channel substrate 5.

[0038] The first spacer substrate 14 is a plate-like member formed of stainless steel (5 mm thick), and as shown in Fig. 4, includes recesses excavated in predetermined patterns so as to correspond in vertical position to the merge section 3b, the droplet-forming flow channel 2d, and the reservoir section 15b of the flow-channel substrate 5. That is, the recesses of the predetermined patterns of the first spacer substrate 14 include: a space portion 13a of a quadrangular shape in plan view, excavated so as to have a predetermined space vertically on the merge section 3b of the flow-channel substrate 5; a reservoir section 25a of a quadrangular shape in plan view, excavated so as to have a predetermined space vertically on the reservoir section 15b of the flow-channel substrate 5; and a droplet-forming flow channel 20e excavated so as to have a predetermined space vertically on the droplet-forming flow channel 2d of the flow-channel substrate 5. Further, the space portion 13a is in communication with the reservoir section 25a via the droplet-forming flow channel 20e. Furthermore, a depth d1 of each of the recesses, i.e., the space portion 13a, the droplet-forming flow channel 20e, and the reservoir section 25a, is 50 μm.

[0039] The second spacer substrate 16 is a plate-like member formed of stainless steel (5 mm thick), and as shown in Fig. 4, includes recesses excavated in predetermined patterns so as to correspond in vertical position to the merge section 3b, the droplet-forming flow channel 2d, the reservoir section 15b, the dispersed phase supply section 10a, and the continuous phase supply sections 11a and 12a of the flow-channel substrate 5. That is, the recesses of the predetermined patterns of the second spacer substrate 16 include: a space portion 13c of a quadrangular shape in plan view, excavated so as to have a predetermined space vertically under the merge section 3b of the flow-channel substrate 5; a reservoir section 25c of a quadrangular shape in plan view, exca-

vated so as to have a predetermined space vertically under the reservoir section 15b of the flow-channel substrate 5; a droplet-forming flow channel 20f excavated so as to have a predetermined space vertically under the droplet-forming flow channel 2d of the flow-channel substrate 5; and a dispersed phase supply section 10d and continuous phase supply sections (not shown) of circular shapes in plan view, formed open so as to correspond in position to the dispersed phase supply section 10a and the continuous phase supply sections 11a and 12a, respectively, when the second spacer substrate 16 is joined to the bottom side of the flow-channel substrate 5. Further, the space portion 13c is in communication with the reservoir section 25c via the droplet-forming flow channel 20f. Furthermore, a depth d2 of each of the recesses, i.e., the space portion 13c, the droplet-forming flow channel 20f, and the reservoir section 25c, is 50 μm. That is, the space portion 13a formed in the first spacer substrate 14 and the space portion 13c formed in the second spacer substrate 16 have the same shape.
It is noted that the cross-sectional view of the microreactor 30 shown in Fig. 4 is a cross-sectional view taken at the center of the short side direction of the microreactor 30, and therefore shows cross sections of the dispersed phase supply section 10 and the dispersed phase flow channel 2b.

[0040] The lamination thus formed so as to join the first spacer substrate 14 and the second spacer substrate 16 to the top and bottom sides, respectively, of the flow-channel substrate 5 provides the upper space portion 13a having a predetermined space vertically on the merge section 3b of the flow-channel substrate 5, and also provides the lower space portion 13c having a predetermined space vertically under the merge section 3b of the flow-channel substrate 5. That is, the provision of the upper space portion 13a and the lower space portion 13c forms, as shown in Fig. 4, stepped portions 14a and 16a, respectively, near the discharge outlet 9 of the dispersed phase flow channel 2b. In other words, the shape is such that the space becomes wider outwardly than the opening of the dispersed phase discharge outlet 9 by a predetermined distance (50 μm, which is the depth of each of the recesses excavated in the respective substrates 14 and 16, in the present embodiment) from the upper and lower edges of the opening.

[0041] As shown in Fig. 4, the microreactor 30 has the lamination of the flow-channel substrate 5, the first spacer substrate 14, and the second spacer substrate 16, so as to join the substrates 14, 5, and 16 to one another. The internal structure (internal space) of the microreactor 30 is the same in shape as that of the microreactor 1, and the functions of the microreactor 30 are the same as those of the microreactor 1. Thus, the internal structure and the functions of the microreactor 30 are not described here.
It is noted that the lamination may not only be formed so as to sandwich the flow-channel substrate 5, between the first spacer substrate 14 and the second spacer substrate 16, from the respective sides of the flow-channel substrate 5, but may also be formed so as to place the first spacer substrate 4 and the cover substrate 7 on the top side of the flow-channel substrate 5, and place the second spacer substrate 16 on the bottom side of the flow-channel substrate 5. That is, the shape of the space portion formed in each of the first spacer substrate and the second spacer substrate may be either a recess excavated, or an opening formed open, so as to correspond in position to the merge section 3b formed open in the flow-channel substrate 5.

[0042] In addition, it is also possible to provide the microreactor 1 with heating means or cooling means near the flow channels 2.

[0043] The above structure of the microreactor 1 enables the preparation of uniform and minute droplets. A description is given below of an example where droplets were prepared using the microreactor 1 described above.

[Example]

[0044] An oil phase (a dodecane solution obtained by dissolving a polymer, in the present example), which is a dispersed phase, is introduced by the supply means described above, not shown, through the dispersed phase supply section 10 of the microreactor 1, and flows through the dispersed phase flow channel 2b at a predetermined flow rate and a predetermined flow velocity. An aqueous phase (a solution containing 1% by weight of sodium dodecyl sulfate, in the present example), which is a continuous phase, is introduced by the supply means described above, not shown, through the continuous phase supply sections 11 and 12 of the microreactor 1, and flows through the continuous phase flow channels 2a and 2c at a predetermined flow rate and a predetermined flow velocity. The oil phase is discharged through the discharge outlet 9 of the flow channel 2b, while the aqueous phase is discharged through the continuous phase discharge outlets 17 and 18 placed on the respective sides of the discharge outlet, and diagonally shears the oil phase discharged through the discharge outlet 9. That is, the continuous phase shears the dispersed phase within and on the upstream side of the merge section 3b of the flow-channel substrate 5. At this time, the provision of the upper space portion 3a and the lower space portion 3c (the stepped portions 4a and 6a), formed respectively by the spacer substrates 4 and 6, causes the oil phase to move to the droplet-forming flow channels 2d, 2e, and 2f while the oil phase is formed into thread-like droplets such that the outside of the oil phase is covered by the aqueous phase (so-called oil in water: O/W), and unlike in a conventional manner, does not cause the oil phase to flow in contact with the ceiling portion 7a, which is the inner surface of the cover substrate 7, or with the bottom portion 8a, which is the inner surface of the cover substrate 8. Consequently, uniform and minute droplets (emulsion) are discharged into the reservoir section 15 (15a, 15b, 15c) through the discharge outlet of the droplet-forming flow channel 19 (2d, 2e, 2f). The droplets thus stored in the reservoir section 15 (15a, 15b, 15c) are ejected through the droplet ejection section 21, and are sent to a storage container, not shown. When the droplets thus prepared using the microreactor 1 according to the present example were poured into a Petri dish and observed under a stereomicroscope (optical microscope), and the droplet sizes and the degree of dispersion (CV value) were measured, it was confirmed that as shown in Fig. 5, uniform droplets having a minimum droplet size of 14.2 μm and a degree of dispersion of 8.51 % were prepared.

[0045] As described above, the microreactor 1 according to the present example prepares stable droplets, because the sandwiching of the flow-channel substrate 5 between the first spacer substrate 4 and the second spacer substrate 6 causes a dispersed phase solution (oil phase solution), discharged through the discharge outlet 9 of the dispersed phase flow channel 2b, to be sheared by an aqueous phase, which is a continuous phase, such that the dispersed phase solution does not contact the ceiling portion 7a or the bottom portion 8a of the flow channels 2. As a matter of course, the microreactor 30 described above, which has the same shape as that of the microreactor 1, also prepares stable droplets in a similar manner.

[Comparative Example]

[0046] Fig. 6 shows a micro-flow-channel substrate, used in the present comparative example, that prepares monodisperse droplets in a normal manner.
With reference to Figs. 6 and 7, a description is given of a comparative example where droplets were prepared using a microreactor including: a flow-channel substrate, shown in Fig. 6, including openings of substantially similar patterns to those of the substrate 5 described above; and the cover substrates 7 and 8 directly closing the top and bottom, respectively, of the flow-channel substrate without using the spacer substrates 4 and 6 of example 1.

[0047] The case is where droplets were prepared, using the microreactor described above, from a solution composition (the oil phase: a dodecane solution obtained by dissolving a polymer; the aqueous phase: a solution containing 1% by weight of sodium dodecyl sulfate) similar to that of example 1, on operating conditions similar to those of example 1. In this case, even when the preparation was made on optimal flow rate conditions, droplets discharged through the droplet-forming flow channel 2d were in a continuous state, as indicated by the arrow in Fig. 7. Consequently, uniform droplets as shown in Fig. 5 failed to be formed. A possible cause of the failure is that with the microreactor used in the present comparative example, the dispersed phase solution (oil phase) discharged through the discharge outlet 9 of the dispersed phase flow channel 2b was, while in contact with the inner surfaces of the cover substrates 7 and 8 of the microreactor 1, sheared by the aqueous phase, which was a continuous phase entering the dispersed phase solution from each side of the dispersed phase flow channel 2b through the continuous phase flow channels 2a and 2c. Thus, the dispersed phase solution failed to be successfully sheared due to the properties of the dispersed phase, and therefore was conducted along the inner surfaces of the cover substrates 7 and 8.

[0048] As is clear by comparing the above example to the comparative example, the microreactor 1 prepares stable monodisperse droplets for the following reason. The spacer substrates 4 and 6 are provided on the top and bottom, respectively, of the flow-channel substrate 5, so as to place the upper space portion 3a and the lower space portion 3c vertically on and under the merge section 3b, i.e., so as to place the stepped portions 4a and 6a near the dispersed phase discharge outlet 9. This prevents, as described above, an oil phase from contacting the inner surfaces (the ceiling portion 7a and the bottom portion 8a) of the cover substrates 7 and 8 when the oil phase is sheared by the aqueous phase. Further, the introduction of the spacer substrates 4 and 6 enables the preparation of stable monodisperse droplets without performing, for example, surface treatment, such as ozone treatment, on the wall surfaces of the flow channels.

[0049] As described above, a microreactor 1 includes flow channels 2 formed by the lamination of a plurality of substrates so as to allow fluids to flow therethrough, and prepares droplets by causing a continuous phase fluid to shear a dispersed phase fluid. The microreactor 1 has an integrated lamination of: a flow-channel substrate 5 including flow channels 2a, 2b, and 2c formed open, through which the continuous phase fluid and the dispersed phase fluid flow, and also including a merge section 3b formed open where the flow channels 2a, 2b, and 2c merge; a first spacer substrate 4 joined to one side of the flow-channel substrate 5 and including a space portion 3a formed so as to maintain a predetermined distance in the corresponding one of the vertical directions with respect to the merge section 3b; a second spacer substrate 6 joined to the other side of the flow-channel substrate 5 and including a space portion 3c formed so as to maintain a predetermined distance in the other vertical direction with respect to the merge section 3b; and cover substrates 7 and 8 joined to the respective outer sides of the first spacer substrate 4 and the second spacer substrate 6 so as to close both outer sides. This makes it possible to prevent droplets from becoming attached to the wall surfaces of the flow channels, and therefore possible to prepare stable monodisperse droplets over a long period of time. This also makes it possible to emulsify even a dispersed phase containing ingredients having a high viscosity or a high hydrophobicity. It is possible to achieve stable emulsification semipermanently without performing surface treatment on the wall surfaces of the flow channels of the microreactor 1. Further, the manufacture of the microreactor 1 is facilitated by its laminated structure, and the maintenance of the microreactor 1 is also facilitated by its easy disassembly.

[0050] In addition, according to the present invention, with simple modifications, it is possible to emulsify even a material composition that is conventionally difficult to emulsify. Further, it is possible to design a microreactor having the lamination of flat plates, taking into account a three-dimensional flow state.

[0051] In addition, according to the present invention, it is possible to prepare stable monodisperse droplet over a long period of time without performing surface treatment, such as ozone treatment, on the surface walls of the flow channels.

[0052] As a matter of course, it is also possible to prepare a W/O (water-in-oil) emulsion by causing an aqueous phase, as a dispersed phase, to flow through the dispersed phase flow channel 2b of the flow channels 2, through which a dispersed phase flows, and causing an oil phase, as a continuous phase, to flow through the continuous phase flow channels 2a and 2c, through which a continuous phase flows.

[0053] In addition, with simple modifications including the insertion of spacer substrates into an existing microreactor as in the present invention, it is possible to provide a microreactor that has a simple structure and prepares stable monodisperse droplets over a long period of time, while maintaining the required reduction in size of a microreactor.

[0054] In addition, in the present embodiment, for ease of understanding, the descriptions are given of a single microreactor 1; however, the present invention is not particularly limited to this. It is also possible to provide a high-lamination-type microreactor by stacking (numbering up) a plurality of microreactors 1 so as to be, for example, 10 to 20 stories. In this case, it is possible to prepare a large number of droplets, and therefore possible to mass-produce droplets.

[0055] It is noted that the present invention is not limited to the embodiments described above, and other variations can be devised based on the intent of the present invention, and such variations are not excluded from the scope of the present invention.

Industrial Applicability

[0056] The present invention is widely applicable to: an emulsification device using a microfluidic device having a microstructure, as typified by a μTAS, a lab-on-a-chip, and a microreactor; synthesis of monodisperse particles and synthesis of gel particles, using microdroplets; a device including micro flow channels, such as a microreactor, that disperses two fluids into each other; and the like.

Claims

1. A microdroplet preparation device that prepares droplets by causing a continuous phase fluid to shear a dispersed phase fluid, wherein both or at least one of a ceiling portion and a bottom portion of flow channels where the continuous phase fluid and the dispersed phase fluid merge are formed so as to have a predetermined distance from a flow of the fluid formed by the merger of the continuous phase fluid and the dispersed phase fluid.

2. The microdroplet preparation device according to claim 1, wherein
the flow channels are formed by an integrated lamination of:

a flow-channel substrate including flow channels formed open through which the continuous phase fluid and the dispersed phase fluid flow, and also including a merge section formed open where the flow channels merge;
at least one spacer substrate joined to one side or respective sides of the flow-channel substrate and including a space portion formed so as to have a predetermined space in a corresponding one of vertical directions with respect to the merge section of the flow-channel substrate; and
cover substrates joined to respective outer sides of the lamination of: the spacer substrates and the flow-channel substrate, the spacer substrates joined to the respective sides of the flow-channel substrate; or the flow-channel substrate and the one of the spacer substrates, the spacer substrate joined to the one side of the flow-channel substrate, so as to close both outer sides.

3. The microdroplet preparation device according to claim 1 or 2, wherein
upstream of the flow channels which are formed open in the flow-channel substrate and through which the continuous phase fluid and the dispersed phase fluid flow, predetermined openings are provided in communication with the respective flow channels,
one of the spacer substrates joined to the respective sides of the flow-channel substrate, and the cover substrate joined to the outer side of the spacer substrate so as to close the outer side, each include openings so as to correspond in position to the predetermined openings, and
the flow-channel substrate, the spacer substrate including the corresponding openings, and the cover substrate including the corresponding openings are laminated, so as to form continuous phase supply sections for supplying a continuous phase and a dispersed phase supply section for supplying a dispersed phase, such that the continuous phase supply sections and the dispersed phase supply section are in communication with the respective flow channels.

4. The microdroplet preparation device according to any one of claims 1 to 3, wherein the space portions of the spacer substrates joined to the respective sides of the flow-channel substrate are placed so as to correspond in position to respective outer sides of the merge section formed open in the flow-channel substrate.

5. The microdroplet preparation device according to any one of claims 1 to 4, wherein the continuous phase shears the dispersed phase within and on an upstream side of the merge section of the flow-channel substrate.

6. The microdroplet preparation device according to any one of claims 1 to 5, wherein each of the space portions formed in the spacer substrates joined to the respective sides of the flow-channel substrate is a recess excavated, or an opening formed open, so as to correspond in position to the merge section formed open in the flow-channel substrate.

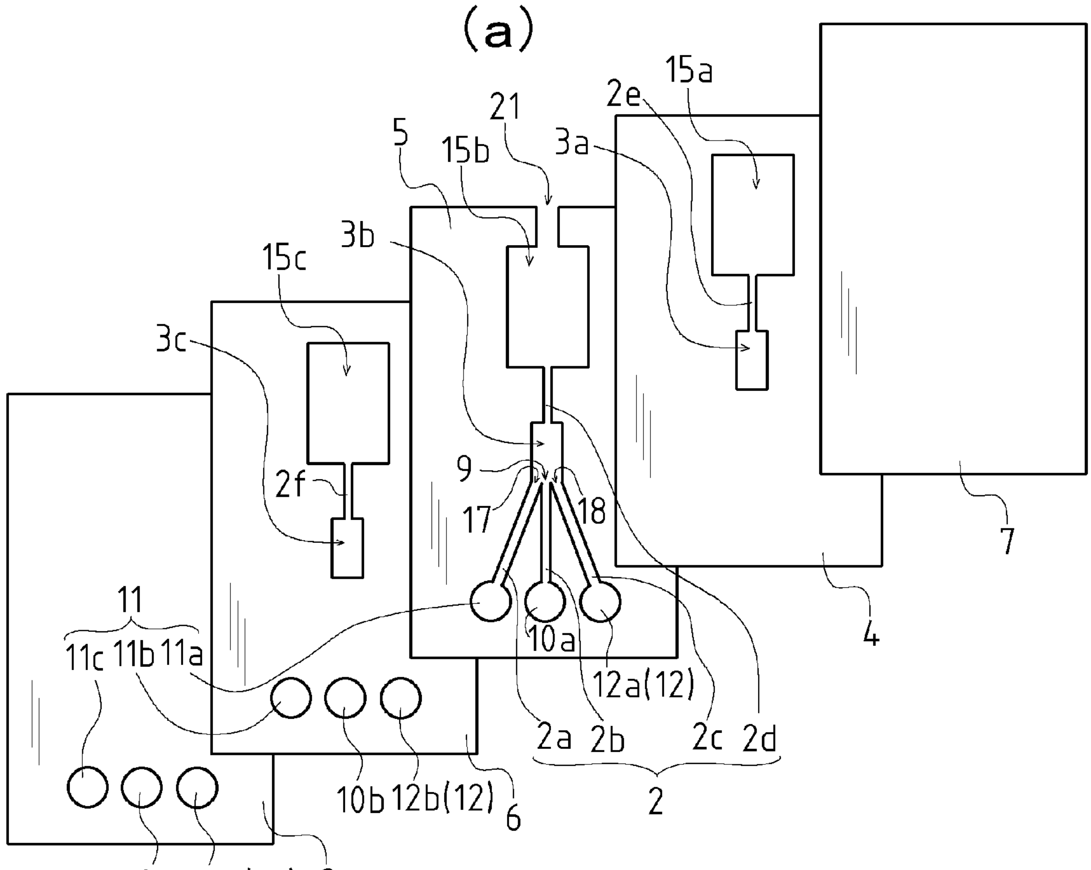
7. The microdroplet preparation device according to any one of claims 1 to 6, wherein the space portions formed in the spacer substrates joined to the respective sides of the flow-channel substrate have the same shape.

8. The microdroplet preparation device according to any one of claims 1 to 7, wherein the spacer substrates joined to the respective sides of the flow-channel substrate have the same thickness.

9. The microdroplet preparation device according to any one of claims 1 to 5, wherein
each of the spacer substrates joined to the respective sides of the flow-channel substrate and including the respective space portions each formed so as to have a predetermined space in the corresponding one of the vertical directions with respect to the merge section of the flow-channel substrate is integrated with the corresponding one of the cover substrates each joined to the corresponding outer side of the corresponding spacer substrate so as to close both outer sides, and
the space portions are provided so as to correspond in position to the respective outer sides of the merge section of the flow-channel substrate, and an integrated lamination is formed so as to sandwich the flow-channel substrate, between both the integrated substrates, from the respective sides of the flow-channel substrate such that positions of the space portions match a position of the merge section of the flow-channel substrate.

Fig.1

(a)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3a
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15b
3b
15c
3c
2f
9
17
18
7
11
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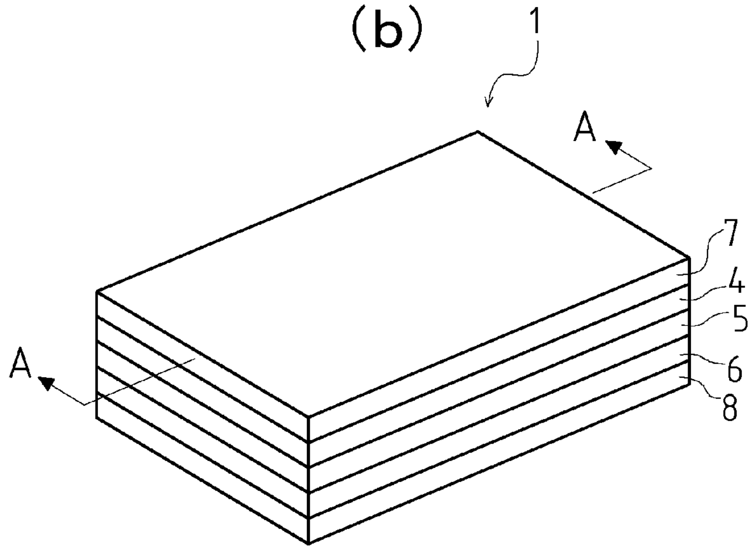
10a
12a(12)
4
2a 2b
2c 2d
10b 12b(12)
6
2
10c 12c(12) 8

(b)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8
A

Fig.2

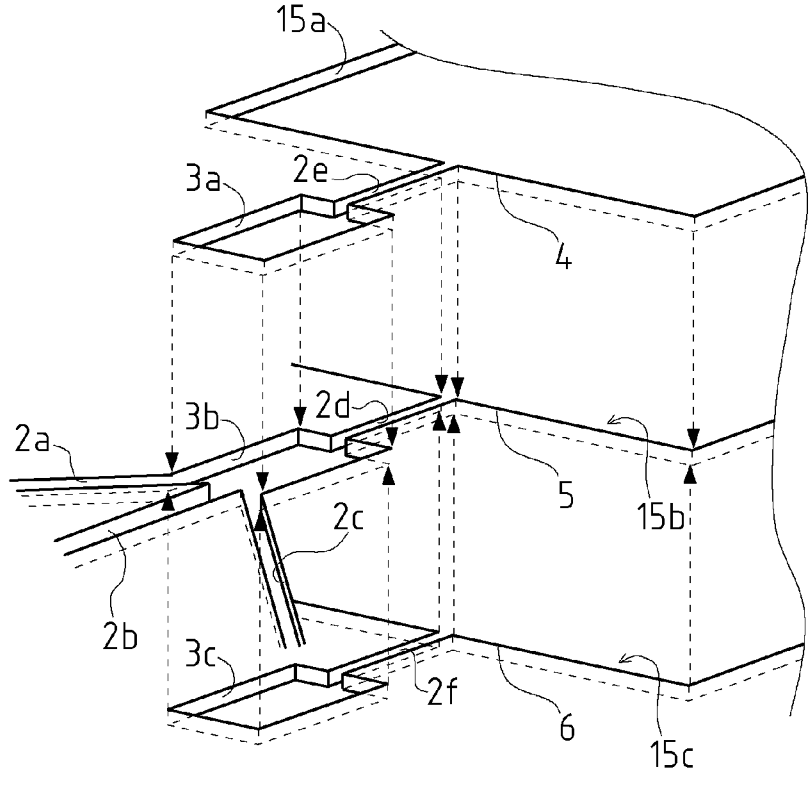
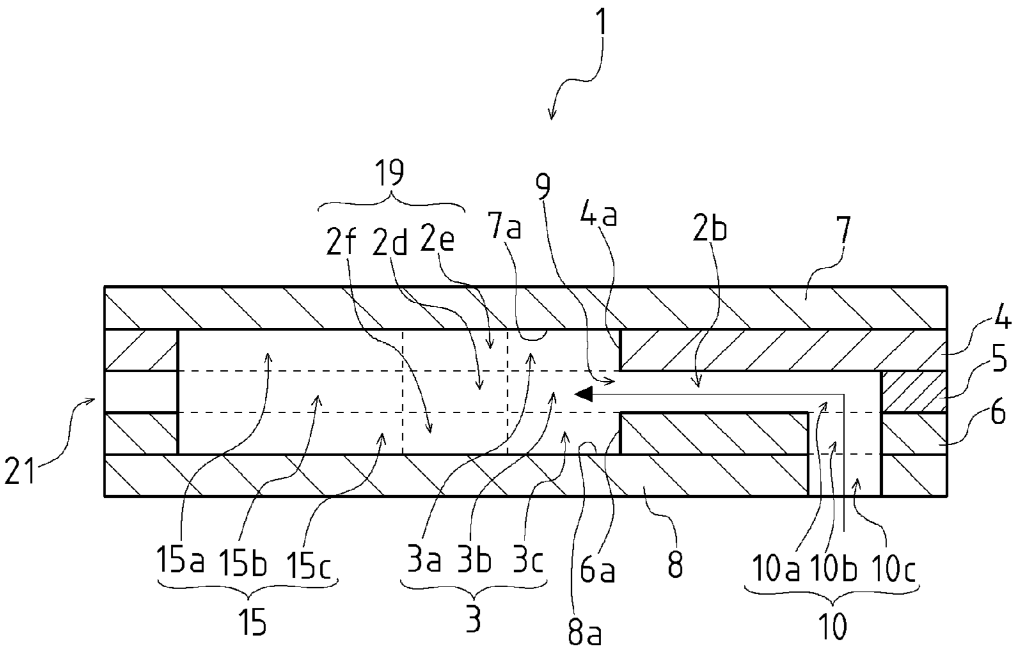


Fig.3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2b
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2f 2d 2e
7a
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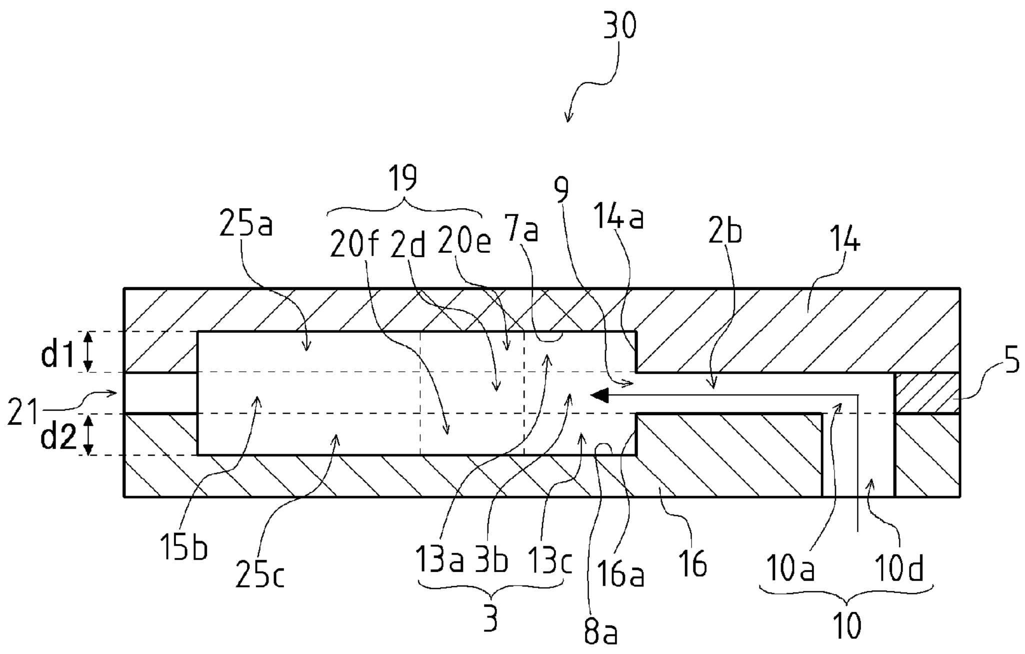
6a
8
8a
10a 10b 10c
10

Fig.4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9
25a
20f 2d 20e
7a
14a
2b
14
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d2
15b
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13a 3b 13c
3
16a
8a
16
10a
10d
10

Fig.5

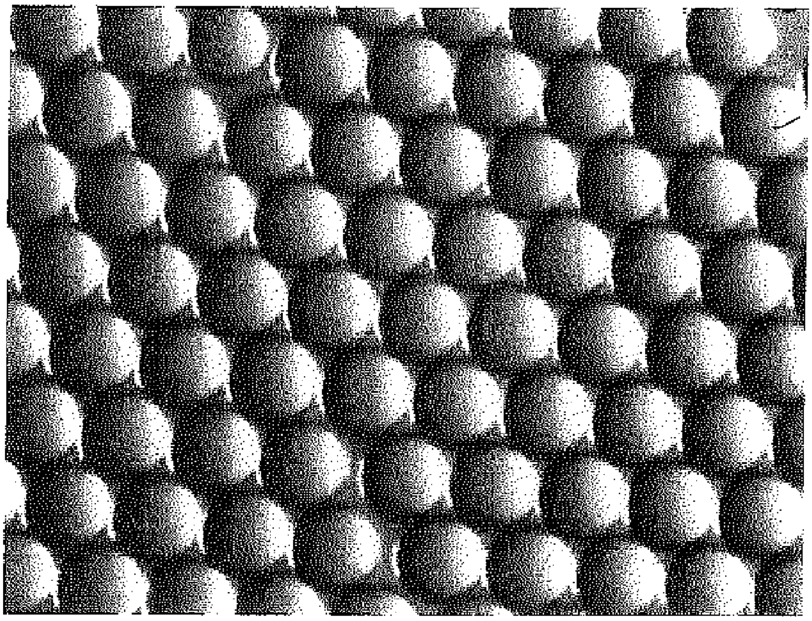


Fig.6

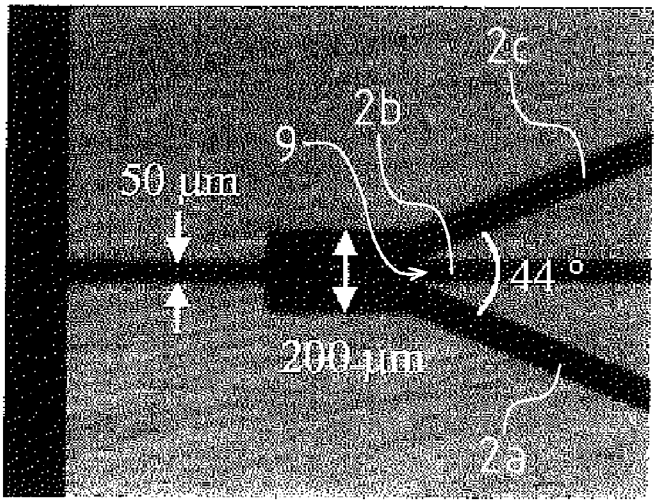
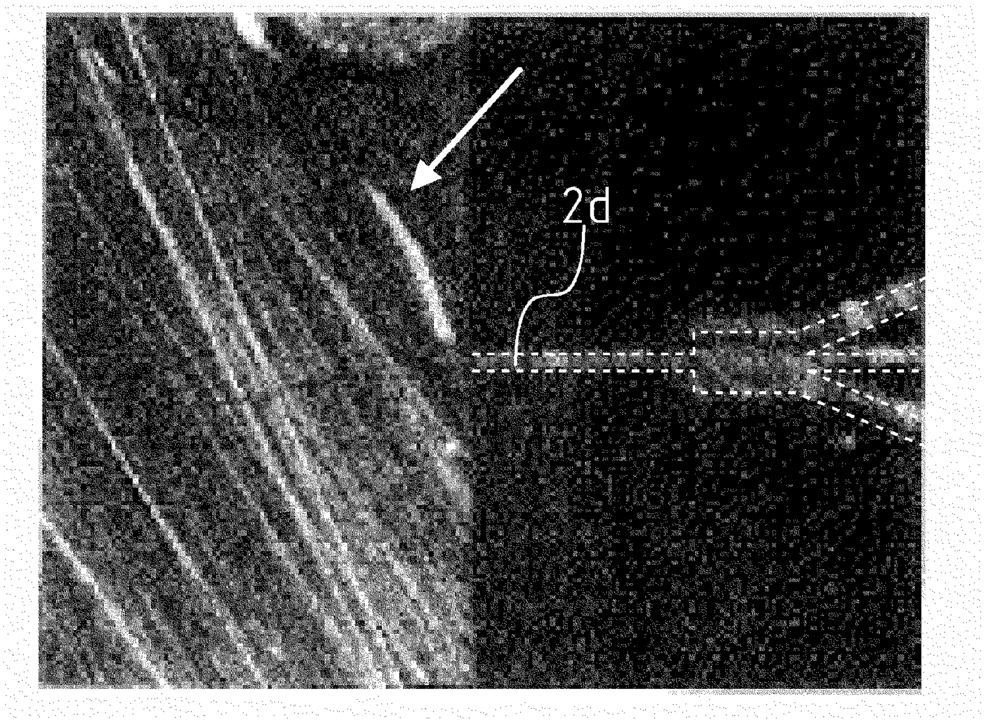


Fig.7



INTERNATIONAL SEARCH REPORT

International application No.
PCT/JP2009/055816

A. CLASSIFICATION OF SUBJECT MATTER
B01J19/00(2006.01)i, *B01F3/08*(2006.01)i, *B01F5/00*(2006.01)i, *B01J13/00*(2006.01)i, *B81B1/00*(2006.01)i

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)
B01J19/00-19/32, B01F1/00-B01F5/26, B01J13/00-B01J13/22, B81B1/00-7/04, B81C1/00-5/00

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

Jitsuyo Shinan Koho	1922-1996	Jitsuyo Shinan Toroku Koho	1996-2009
Kokai Jitsuyo Shinan Koho	1971-2009	Toroku Jitsuyo Shinan Koho	1994-2009

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

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X A	JP 2006-43617 A (Hitachi Industries Co., Ltd.), 16 February, 2006 (16.02.06), Par. Nos. [0001], [0038], [0046]; Figs. 1, 2, 5 & US 2006/0029528 A1 & EP 1623760 A2 & DE 602005005574 T2 & TW 247626 B & CN 1730142 A	1,2,5 3,4,6-9
A	JP 2007-521944 A (Velocys, Inc.), 09 August, 2007 (09.08.07), Par. Nos. [0001], [0046]; Fig. 9 & US 2005/0133457 A1 & EP 1700075 A1 & WO 2005/060658 A1 & CA 2550079 A1 & CN 1993174 A	1-9

☒ Further documents are listed in the continuation of Box C. ☐ See patent family annex.

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"&" document member of the same patent family

Date of the actual completion of the international search	Date of mailing of the international search report
27 May, 2009 (27.05.09)	09 June, 2009 (09.06.09)

Name and mailing address of the ISA/	Authorized officer
Japanese Patent Office Facsimile No.	Telephone No.

Form PCT/ISA/210 (second sheet) (April 2007)

INTERNATIONAL SEARCH REPORT

International application No.
PCT/JP2009/055816

C (Continuation). DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	JP 2006-272268 A (Fuji Photo Film Co., Ltd.), 12 October, 2006 (12.10.06), Par. Nos. [0064], [0070]; Fig. 8 (Family: none)	1-9
A	JP 2004-122107 A (Tosoh Corp.), 22 April, 2004 (22.04.04), Par. Nos. [0001] to [0004] & US 2003/0201022 A1 & US 2006/0231136 A1 & US 2008/0246172 A1 & EP 1358931 A2	1-9

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REFERENCES CITED IN THE DESCRIPTION

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