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(54) Title: HEAT EXCHANGERS FOR MICROSTRUCTURES

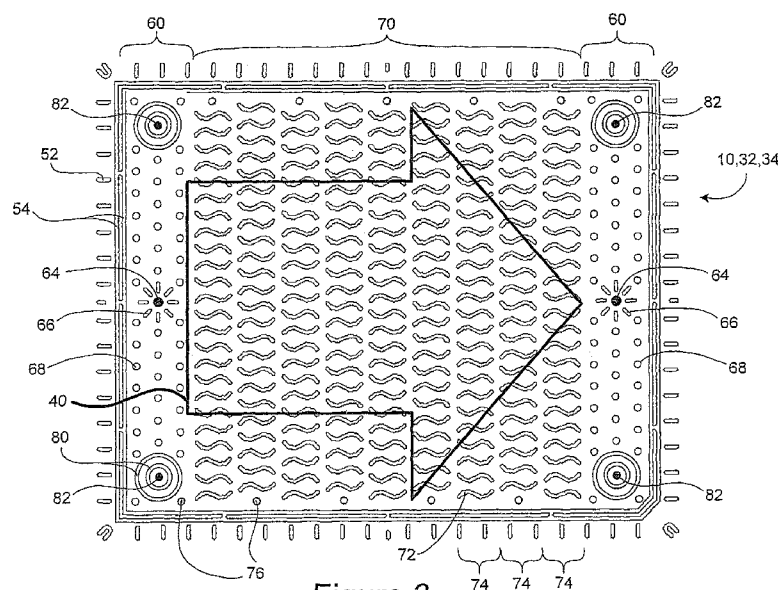


Figure 3

(57) Abstract: A multiple-layered microfluidic device [10] comprises at least a first fluid path [40, 40A, 40B] and at least a second fluid path [50], wherein the first fluid path [40, 40A, 40B] comprises a layer [32] or portion of a layer [32A, 32B] of the microfluidic device [10]. The first path [40, 40A, 40B] has multiple rows [74] of serpentine wall segments [72] positioned there along. The wall segments [72] extend in a direction along the first path [40]. The rows [74] extend along a direction cross-ways to the first path [40, 40A, 40B]. Adjacent ones of wall segments [72] within a row [74] are arranged such that concave portions [73] face concave portions [73] of adjacent ones of segments [72], while convex portions [75] face convex portions [75] of adjacent ones of segments [72].

HEAT EXCHANGERS FOR MICROSTRUCTURES

PRIORITY

[0001] This application claims priority to European Patent Application number 08305844.6, filed November 26, 2008, titled "Heat Exchangers For Microstructures".

BACKGROUND

[0002] This application relates to devices and methods for heat exchangers useful in microfluidic devices.

SUMMARY

[0003] According to one aspect of the invention, a multiple-layered microfluidic device comprises at least a first fluid path and at least a second fluid path, wherein the first fluid path comprises a layer or portion of a layer of the microfluidic device. The first path has multiple rows of serpentine wall segments positioned there along. The wall segments extend in a direction along the first path. The rows extend along a direction cross-ways to the first path. Adjacent ones of wall segments within a row are arranged such that concave portions face concave portions of adjacent ones of segments, while convex portions face convex portions of adjacent ones of segments.

[0004] Additional variations and features of the present invention are described below in connection with the figures, of which:

BRIEF DESCRIPTION OF THE DRAWINGS

[0005] Figure 1 is cross section of a microstructure in the form of a microfluidic device 10;

[0006] Figure 2 is another cross section of the microstructure or microfluidic device 10 of Figure 1;

[0007] Figure 3 is a cross-sectional plan view showing an arrangement of walls 34 of a

layer 32 of a microstructure such as the microfluidic device 10 of Figures 1 and 2;

[0008] Figure 4 is an enlarged partial plan view of the features shown in Figure 3;

[0009] Figure 5 is a plan view showing features of a comparative thermal control layer 33;

[0010] Figures 6-8 are graphs comparing the performance of an embodiment of a microstructure having the structure of Figure 3 to an embodiment of a microstructure having the comparative structure of Figure 5;

[0011] Figure 9 is a plan view showing the layout of an alternative embodiment of a layer 32 of a device 10 like that of Figure 1, but with the layer 32 divided into to sections 32A and 32B.

DETAILED DESCRIPTION

[0012] Whenever possible, the same reference numerals will be used throughout the drawings to refer to the same or like parts. Features described as desirable are preferred but optional, representative of variations of the invention.

[0013] Within this document, “microstructure” refers to single-piece or otherwise generally permanently assembled microfluidic devices having internal passages with at least one characteristic cross-sectional dimension in the few-millimeter to sub-millimeter range; “microreactor” refers to a device for performing chemical or physical processes or combinations thereof, the device including one or more microstructures.

[0014] The methods and/or devices disclosed herein are generally useful in performing any process that involves mixing, separation, extraction, crystallization, precipitation, or otherwise processing fluids or mixtures of fluids, including multiphase mixtures of fluids—and including fluids or mixtures of fluids including multiphase mixtures of fluids that also contain solids—within a microstructure. The processing may include a physical process, a chemical reaction defined as a process that results in the interconversion of organic, inorganic, or both organic and inorganic species, a biochemical process, or any other form of processing. The following non-limiting list of reactions may be performed with the disclosed methods and/or devices: oxidation; reduction; substitution; elimination; addition; ligand exchange; metal exchange; and ion exchange. More specifically, reactions of any of the following non-limiting list may be performed with the

disclosed methods and/or devices: polymerisation; alkylation; dealkylation; nitration; peroxidation; sulfoxidation; epoxidation; ammoxidation; hydrogenation; dehydrogenation; organometallic reactions; precious metal chemistry/ homogeneous catalyst reactions; carbonylation; thiocarbonylation; alkoxylation; halogenation; dehydrohalogenation; dehalogenation; hydroformylation; carboxylation; decarboxylation; amination; arylation; peptide coupling; aldol condensation; cyclocondensation; dehydrocyclization; esterification; amidation; heterocyclic synthesis; dehydration; alcoholysis; hydrolysis; ammonolysis; etherification; enzymatic synthesis; ketalization; saponification; isomerisation; quaternization; formylation; phase transfer reactions; silylations; nitrile synthesis; phosphorylation; ozonolysis; azide chemistry; metathesis; hydrosilylation; coupling reactions; and enzymatic reactions.

[0015] Figure 1 is cross section of a microstructure in the form of a microfluidic device 10. The microfluidic device 10 may be comprised of multiple substrates 20, generally at least four as shown at the top of the figure, but more may be included if desired, up to “n” total, as labeled at the left edge of Figure 1. Between each adjacent pair of the multiple substrates 20, a layer 30 of the device 10 is defined, such that multiple layers are present, generally at least three, and optionally more, with up to n-1 total layers, as labeled at the right edge of Figure 1. The substrates 20 are joined to each other and supported relative to one another by walls 34 (for ease of viewing, not all labeled), some of which are cut by the cross section of the figure, as indicated by the cross-hatching. Holes 64, which may extend through one or more of the substrates 20, provide external access to a first fluidic path 40 defined through one or more of the layers 30, in this case through the two layers 32 of the layers 30. Alternative access methods, such as passages through walls 34, may also be used if desired.

[0016] In the use of the device, the first fluidic path 40 is desirably used for flowing a thermal control fluid, but may also, in some devices or for performing some operations, be used to flow a process fluid such as a mixture in a reaction process or the like. Figure 2 is another cross section of the microstructure or microfluidic device 10 of Figure 1 taken in a different plane parallel to that of Figure 1. In the cross section of Figure 2 may be seen holes 82 which provide access to a second fluidic path 50 defined through one or more layers 30 of the device, in this case through the one layer 31. Layer 31, bordered on

both major sides thereof by layers 32, is typically but not necessarily used for reaction fluids or other process fluids for which heat exchange is desirable.

[0017] Various materials methods may be used to form microfluidic devices 10 of the type shown in Figures 1 and 2, including molding or otherwise forming a glass-containing frit to form walls 34 between substrates 20 of glass, ceramic, metal, or other materials, then heating or “firing” the frit walls to adhere the walls 34 and substrates 20 together, forming a microfluidic device. Such processes are disclosed and described, for example, in US Patent No. 7007709, A Microfluidic Device and Manufacture Thereof, assigned to the assignee of the present invention.

[0018] The results of some alternate forming methods are represented in the cross section of Figure 2. In one alternate method, a glass or glass-ceramic sheet or a glass-containing frit composition may be formed by heating together with a mold as disclosed and described for example in EP Application No. 07300835, Method for Making Microfluidic Devices and Devices Produced Thereof, filed 28 February 2007 and assigned to the present assignee, or by injection molding as disclosed and described for example in US Patent Publication No. 2007-0154666, Powder Injection Molding of Glass and Glass-Ceramics, filed 31 March 2006 and assigned to the present assignee. If walls 34 are formed onto a substrate 20 from a frit or from a flat glass precursor, a thin layer 22 of the consolidated frit material or a thin layer 22 of the molded glass material may remain on the substrate surfaces, such that said thin layer 22 lines the respective layer 30 of the device 10, as shown at the top of Figure 2. As a further alternative, rather than molding onto a substrate, walls 34 and an adjacent substrate may be molded as a single piece, as represented in Figure 2 by the fourth from the top of the substrates 20 and the associated walls 34, which are all part of one consolidated structure 24, such that a separate substrate, in the sense of a pre-formed generally flat object on which other structures are placed or formed, is not required. Another single piece molded layer may have cooperating walls 34a that meet and seal to walls 34. Any other appropriate forming methods may also be applied to form the structures of the present invention.

[0019] Figure 3 is a cross-sectional plan view of a device 10 such as that in Figures 1 and 2, showing the structures of walls 34 of a layer 32 including a fluid path 40 extending from one hole or entrance port 64 to another. The cross section is taken through the walls

34, and only walls 34 are shown, with the exception of ports or holes 64, 82.

[0020] The layer 32 shown in Figure 3 includes various types of walls 34. A doubled boundary wall 54 laterally encloses the fluidic path 40 of the layer 32. Short oblong buttress walls 52 surround the boundary wall 54. In the corners of the layer, double circular walls 80 surround holes 82 that provide access to other layers not in the plane of the figure. Short oblong walls 66 in a radiating pattern around each hole 64 give added support to the substrate material in the region of the input and output ports or holes 64. A second fluidic path 50 such as in Figure 2, in a layer not in the plane of Figure 3, may be accessed via one or more of the holes 82.

[0021] The first fluidic path 40 comprises a layer 32 having multiple rows 74 of serpentine wall segments 72 positioned therein. The wall segments are walls 34 that connect the two substrates 20 bounding the layer 32, as shown in Figure 1. As shown in Figure 3, the serpentine wall segments 72 extend in a direction along the direction of the first path 40. As seen in Figure 3 and in Figure 4, which is an enlarged partial cross-sectional plan view of the features shown in Figure 3, the rows 74 of wall segments 72 extend along a direction cross-ways to the first path 40, with adjacent ones of the wall segments 72 within a row 74 arranged such that concave portions 73 face concave portions 73 of adjacent ones of segments 72, while convex portions 75 face convex portions 75 of adjacent ones of segments 72.

[0022] As seen in Figures 3 and 4, the segments 72 are desirably S-shaped, although longer serpentine forms may also be used. The segments 72 of each row 74 are desirably at offset positions relative to the segments 72 of an adjacent row 74. The amount of offset is desirably equal to one-half of the center-to-center distance within the row 74 from one segment 72 to the next segment 72. Referring to Figure 4, the distance 96 between facing concave portions 73 of adjacent ones of the segments 72 is desirably at least twice the distance 94 between facing convex portions 75 of adjacent ones of segments 72 within a row 74.

[0023] As shown in Figure 3, layer 32 desirably includes a relatively more open region 60 adjacent to the first row of the rows 74, and adjacent to the last row of the rows 74. The openness may be achieved, while still providing adequate pressure resistance for the enclosed layer 32, by using walls 34 that take the form of small round pillars 68 in the

more open region 60. Pillars 68 provide structural strength without providing as much flow resistance as in the region 70 comprised of the rows 74 of serpentine wall segments 72. Thus regions 60 can act as low-flow-resistance manifolds and collectors for the region 70 of rows 40 of segments 72.

[0024] The staggered rows 74 of serpentine wall segments 72 of the present invention act to minimize the thickness of boundary layers by producing Dean vortices and/or other secondary flows in a fluid flowing along the first path 40. The secondary flows thus generated significantly improve heat exchange performance of the microfluidic device 10 with only modest increase in pressure drop relative to comparative designs. It is also desirable to minimize the boundary layer by limiting the height 33 (see Figure 2) of the layer(s) 32 in which the fluid path 40 is contained. The height 33 of the layer 32 in which the path 40 is contained desirably in the range of 0.2 to 3 millimeters, more desirably in the range of 0.2 to 1.5 millimeters, most desirably in the range of 0.2 to 0.75 millimeters. The thickness 98 of the segments 72 is desirably in the range of 0.5 to 1.5 millimeters (see Figure 4). The distance 94 between facing convex portions 75 of adjacent ones of segments 72 is desirably in the range of 2-4 millimeters, and the distance 96 between facing concave portions 73 of adjacent ones of segments 72 is desirably in the range of 4-8 millimeters, and is desirably at least twice the distanced 94. The distance 92 between adjacent rows 74 is desirably in the range of 1-3 millimeters.

[0025] A previous design of a device 10 having a layer 33 as shown in Figure 5 was tested for comparative purposes. In the device of Figure 5, in the layer 33 of Figure 5, segmented walls 102 are not serpentine and are not arranged in staggered or offset rows, but are arranged instead to guide a fluid relatively smoothly from one access hole or port 104 to the other.

Experimental

[0026] Comparative tests were performed for a device 10 having a layer 32 as shown in Figure 3 and for a comparative device 10 having a layer 33 as shown in Figure 5. The second fluid path, path 50 of Figure 2, was structured essentially identically in both devices, and the heat exchanged between a fluid in the respective layer 32 or 33 and a fluid in the second fluid path. The results are reflected in the Table below.

[0027] The table shows the relative characteristics and performance of a device 10 of the present invention having one or more layers 32 as shown in Figures 3 and 4, relative to a comparative device similar to the inventive device but having one or more layers 33 as shown in Figure 5, for three different fluids flowed in the respective layers 32, 33. As may be seen from the table, for only an 18% relative increase in pressure drop (measured for water at 1 liter/minute), a heat transfer increase of up to 40% was realized.

[0028] More detailed test results are shown in the graphs of Figures 6-8, each of which shows the achieved volumetric heat transfer coefficient in W/m^3K , as a function of flow rate of fluid in the first path 40 in milliliters per minute. Figure 6 shows results for ethanol as the fluid in path 40, with trace 120 indicating the performance of the inventive device including the structure of Figure 3, while trace 110 indicates the performance of the comparative device including the structure of Figure 5. Figure 7 shows results for silicone oil as the fluid, with trace 140 indicating the performance of the inventive device including the structure of Figure 3, while trace 130 indicates the performance of the comparative device including the structure of Figure 5. Figure 8 shows the results for water with trace 160 indicating the performance of the inventive device including the structure of Figure 3, while trace 150 indicates the performance of the comparative device including the structure of Figure 5. As may be seen from Figures 6-8, significant improvement was found for all three fluids, even with the height corresponding to height 33 of Figure 2 being slightly greater in the inventive device than in the comparative device (see the Table below).

[0029] Figure 9 is a plan view showing the layout of an alternative embodiment of a thermal control layer 32 in a device 10 according to the present invention. In devices of the type shown in Figure 9, an individual layer 32 may be divided into two or more sections, section 32A and section 32B in this case, with each section 32A, 32B having its own respective path 40A, 40B. Further, as in section 32A, the path 40A may lie in different directions in different portions of the layer 32. Separate sections 32A and 32B allow different temperatures to be maintained in different parts of the thermal control layer 32 and hence in different parts of the device 10. Separate sections also allow the generation of different thermal profiles in different parts of the device 10, or thermal profiles lying in different directions, or both.

<u>TABLE</u>		
	Inventive Device	Comparative Device
Internal volume, ml	8.2 (height = 630 μm)	8.5 (610 μm)
Pressure drop, mbar (water, liter/min)	118	100
Heat transfer improvement (over comparative device for the listed fluid)		
- ethanol	30 %	NA
- silicone oil	40 %	NA
- water	40 %	NA
Pressure resistance, bar	17.1	15.7

What is claimed is:

1. A multiple-layered microfluidic device [10] comprising at least a first fluid path [40, 40A, 40B] and at least a second fluid path [50], wherein the first fluid path [40, 40A, 40B] comprises a layer [32] or portion of a layer [32A, 32B] of the microfluidic device [10], the first path [40, 40A, 40B] having multiple rows [74] of serpentine wall segments [72] positioned there along, said wall segments [72] extending in a direction along the first path [40, 40A, 40B], said rows [74] extending along a direction cross-ways to the first path [40, 40A, 40B], with adjacent ones of wall segments [72] within a row [74] arranged such that concave portions [73] face concave portions [73] of adjacent ones of segments [72], while convex portions [75] face convex portions [75] of adjacent ones of segments [72].
2. The device according to claim 1 wherein the segments [72] are S-shaped.
3. The device according to either claim 1 or claim 2 wherein the segments [72] of each row [74] are at offset positions relative to the segments [72] of an adjacent row [74].
4. The device according to claim 3 wherein the segments [72] of each row [74] are at offset positions relative to the segments [72] of an adjacent row [74], the offset amount being one half of the distance within the row [74] from one segment [72] to the next segment [72].
5. The device according to any of claims 1-4 further comprising a first relatively

more open region [60] adjacent to a first row of the rows [74].

6. The device according to any of claims 1-5 comprising a second relatively more open region [60] adjacent to a last row of the rows [74].

7. The device according to any of claims 1-6 wherein the height [33] of the layer [32] or portion of layer [32A, 32B] of the path [40, 40A, 40B] is in the range of 0.2 to 1.5 millimeters.

8. The device according to claim 7 wherein the height [33] of the layer [32] or portion of layer [32A, 32B] of the path [40, 40A, 40B] is in the range of 0.2 to 0.75 millimeters.

9. The device according to any of claims 1-8 wherein the thickness [98] of the segments [72] is in the range of 0.5 to 1.5 millimeters.

10. The device according to any of claims 1-8 wherein the distance [96] between facing concave portions [73] of adjacent ones of segments [72] is at least twice the distance [94] between facing convex portions [75] of adjacent ones of segments [72] within the rows [74].

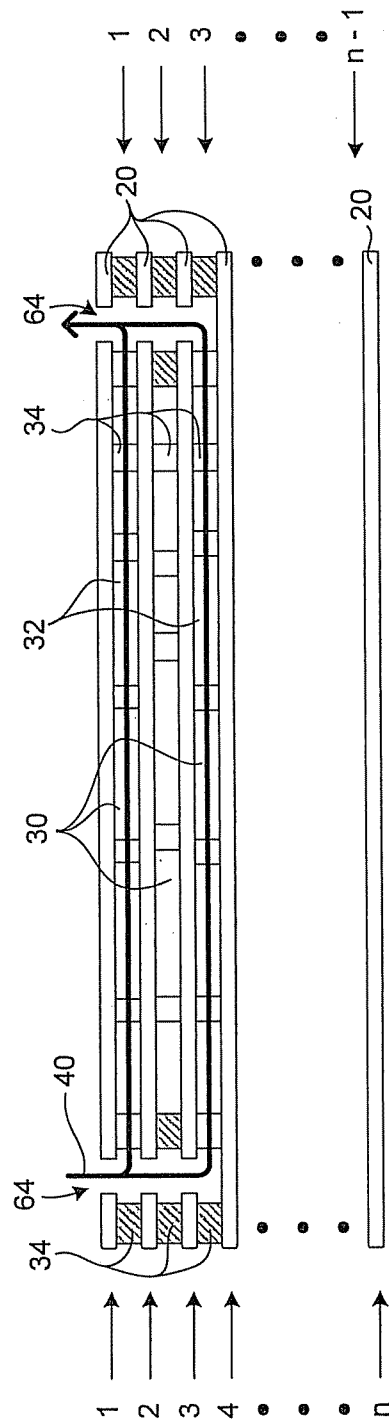


Figure 1

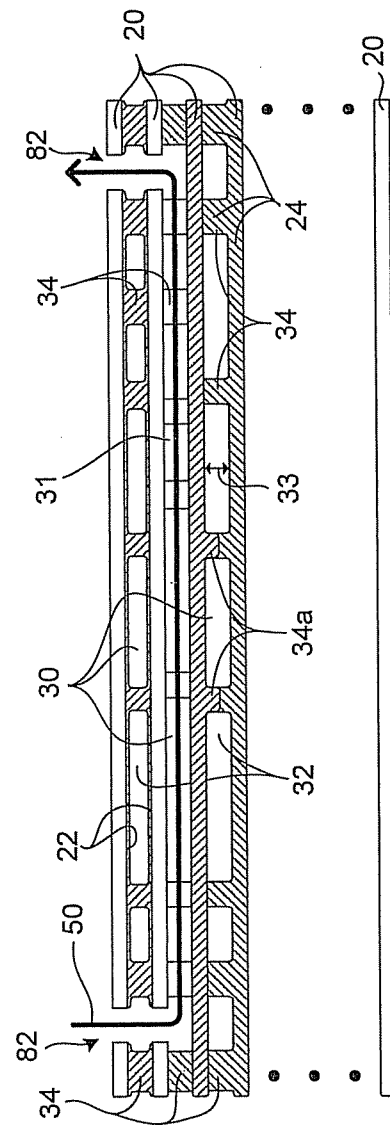


Figure 2

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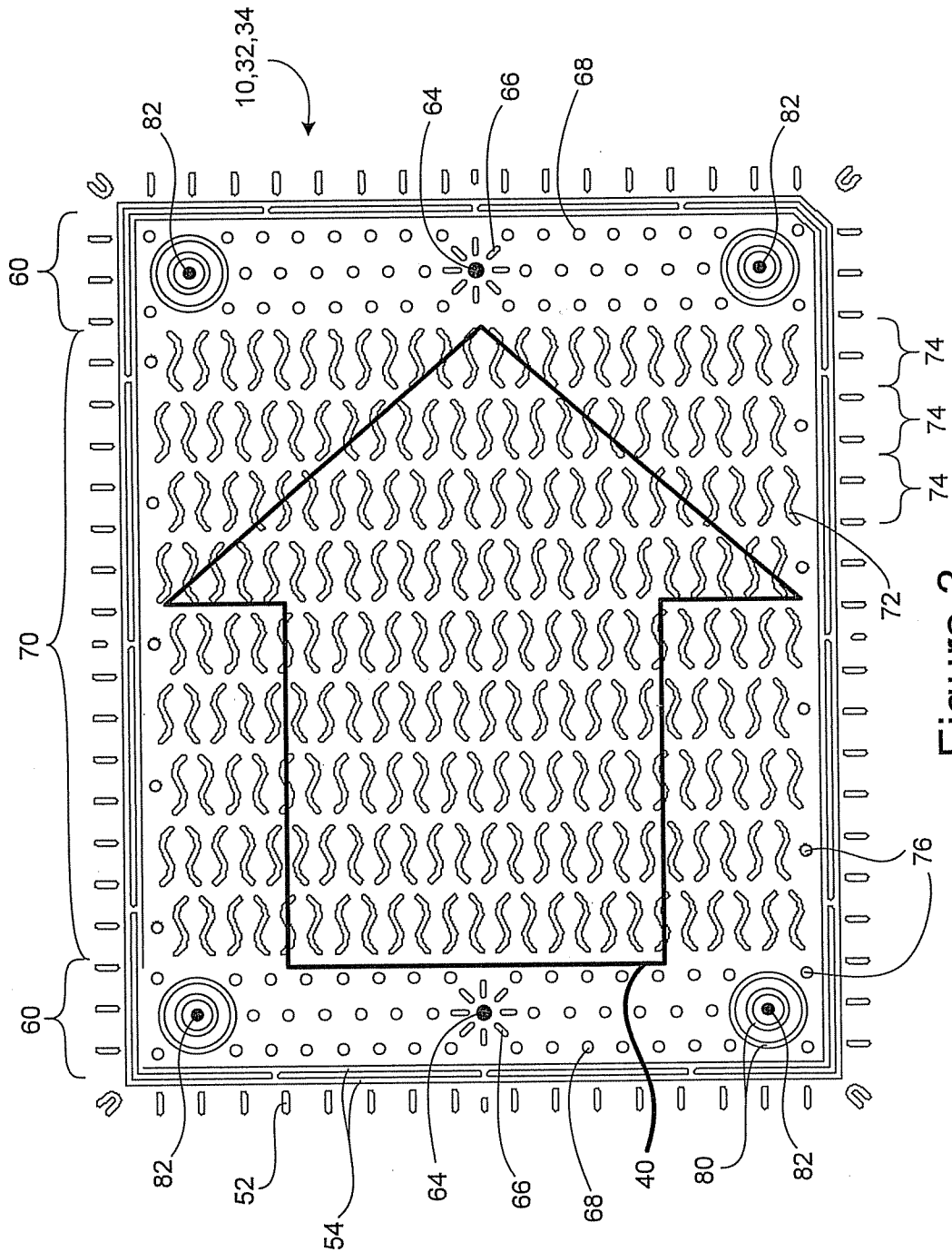


Figure 3

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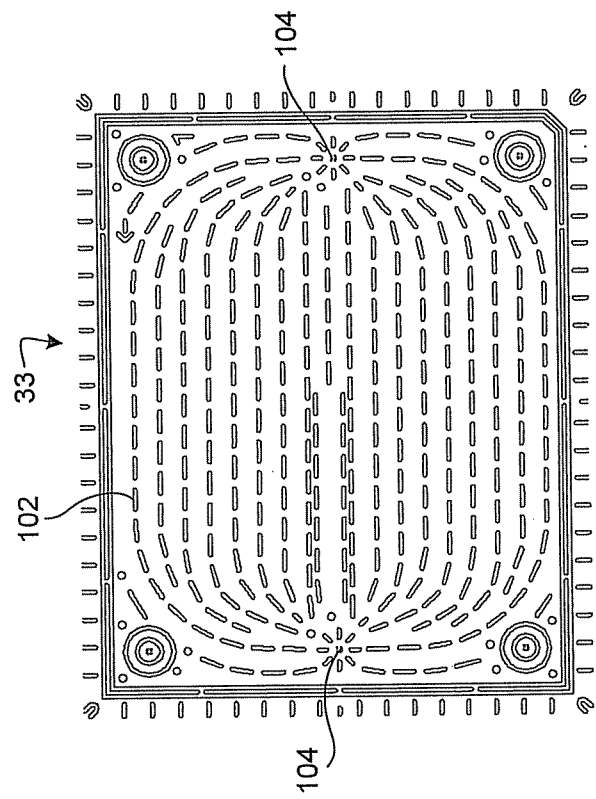


Figure 5

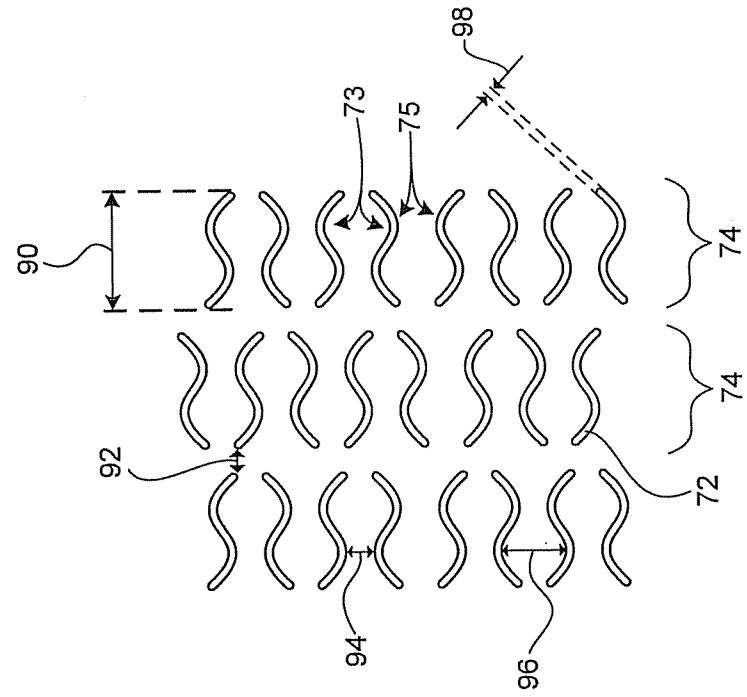


Figure 4

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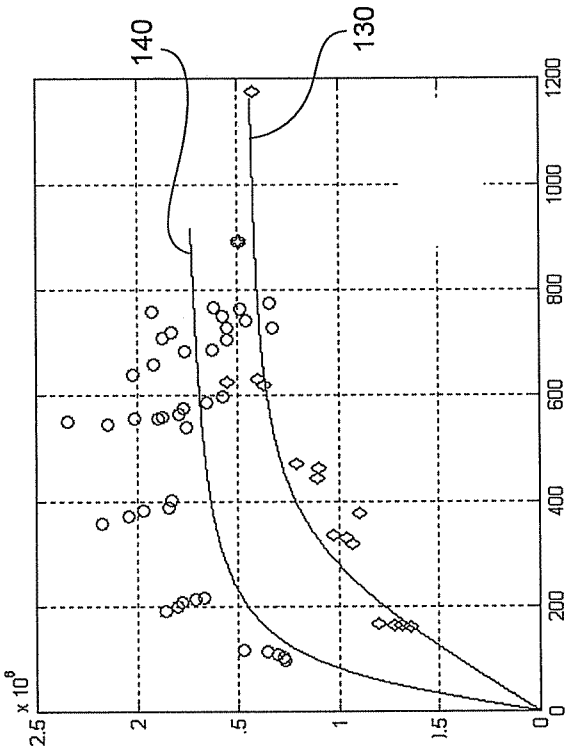


Figure 7

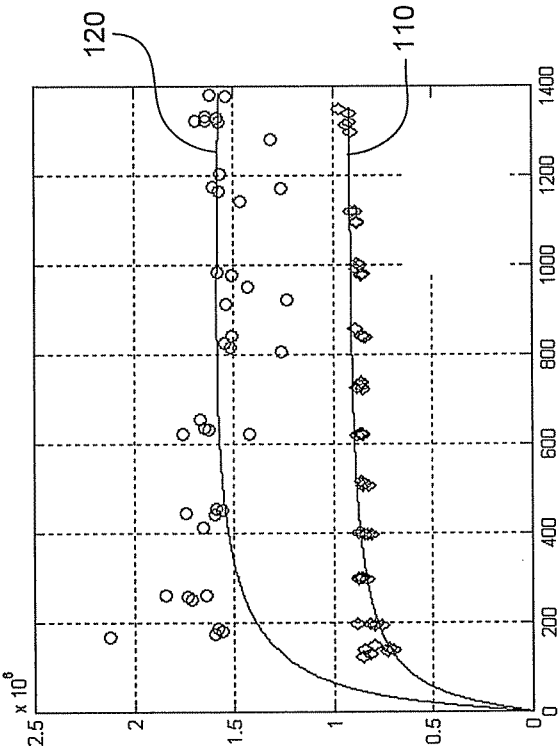


Figure 6

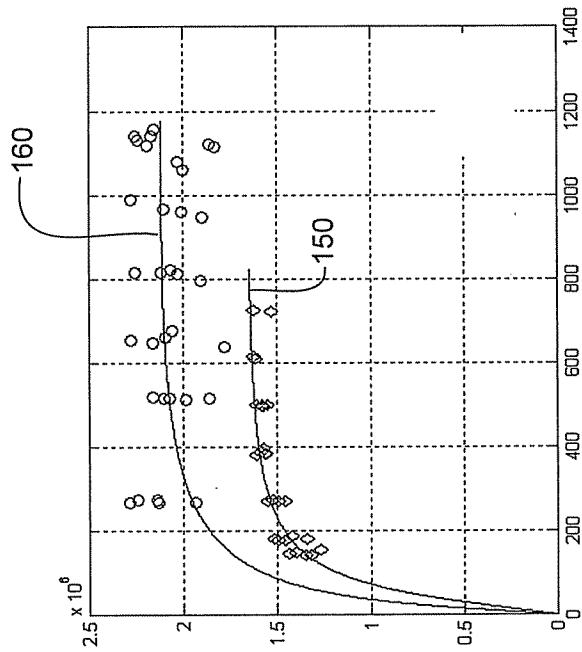


Figure 8

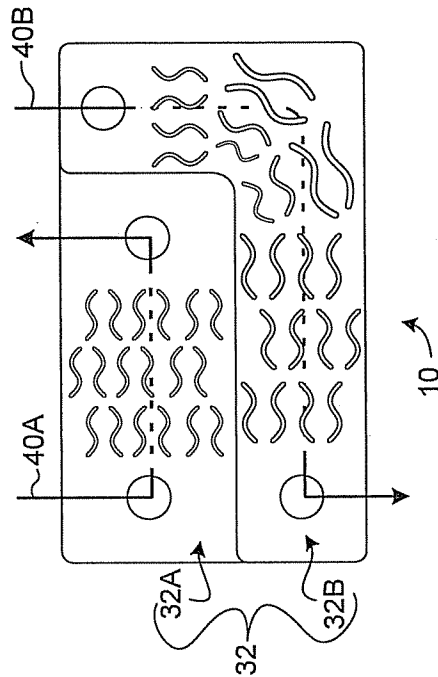


Figure 9

INTERNATIONAL SEARCH REPORT

International application No.

PCT/US 09/65630

A. CLASSIFICATION OF SUBJECT MATTER

IPC(8) - H05K 7/20 (2010.01)

USPC - 361/699; 165/104.26; 165/80.4; 174/15.1

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)

USPC - 361/699; 165/104.26; 165/80.4; 174/15.1

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

USPC - 361/699; 165/104.26; 165/80.4; 174/15.1 (text search)

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

PubWest: PGPB,USPT,USOC,EPAB,JPAB; Google

search terms used: microfluidic, s-shaped, heat exchanger, microreactor, wall segment, micro, small, concave, faces, serpentine, fins, offset, minimize, flow resistance plates curved

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
Y	EP 1 854 536 A1 (Barthe et al.) 14 November 2007 (14.11.2007), fig 9; para [0043], [0044], [0066]	1-4
Y	US 7,334,631 B2 (Yasuyoshi et al.) 26 February 2008 (26.02.2008), col 5, ln 9-47	1-4
Y	US 6,180,846 B1 (Dandekar et al.) 30 January 2001 (30.01.2001), Fig. 7	1-4

☐ Further documents are listed in the continuation of Box C.


* Special categories of cited documents:

"A" document defining the general state of the art which is not considered to be of particular relevance

"E" earlier application or patent but published on or after the international filing date

"L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)

"O" document referring to an oral disclosure, use, exhibition or other means

"P" document published prior to the international filing date but later than the priority date claimed

"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention

"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone

"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art

"&" document member of the same patent family

Date of the actual completion of the international search

08 January 2010 (08.01.2010)

Date of mailing of the international search report

12 FEB 2010

Name and mailing address of the ISA/US

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Lee W. Young

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INTERNATIONAL SEARCH REPORT

International application No.

PCT/US 09/65630

Box No. II Observations where certain claims were found unsearchable (Continuation of item 2 of first sheet)

This international search report has not been established in respect of certain claims under Article 17(2)(a) for the following reasons:

1. ☐ Claims Nos.:
because they relate to subject matter not required to be searched by this Authority, namely:
2. ☐ Claims Nos.:
because they relate to parts of the international application that do not comply with the prescribed requirements to such an extent that no meaningful international search can be carried out, specifically:
3. ☒ Claims Nos.: 5-10
because they are dependent claims and are not drafted in accordance with the second and third sentences of Rule 6.4(a).

Box No. III Observations where unity of invention is lacking (Continuation of item 3 of first sheet)

This International Searching Authority found multiple inventions in this international application, as follows:

1. ☐ As all required additional search fees were timely paid by the applicant, this international search report covers all searchable claims.
2. ☐ As all searchable claims could be searched without effort justifying additional fees, this Authority did not invite payment of additional fees.
3. ☐ As only some of the required additional search fees were timely paid by the applicant, this international search report covers only those claims for which fees were paid, specifically claims Nos.:
4. ☐ No required additional search fees were timely paid by the applicant. Consequently, this international search report is restricted to the invention first mentioned in the claims; it is covered by claims Nos.:

Remark on Protest

- ☐ The additional search fees were accompanied by the applicant's protest and, where applicable, the payment of a protest fee.
- ☐ The additional search fees were accompanied by the applicant's protest but the applicable protest fee was not paid within the time limit specified in the invitation.
- ☐ No protest accompanied the payment of additional search fees.