

(19) World Intellectual Property Organization
International Bureau



(43) International Publication Date
25 May 2001 (25.05.2001)

PCT

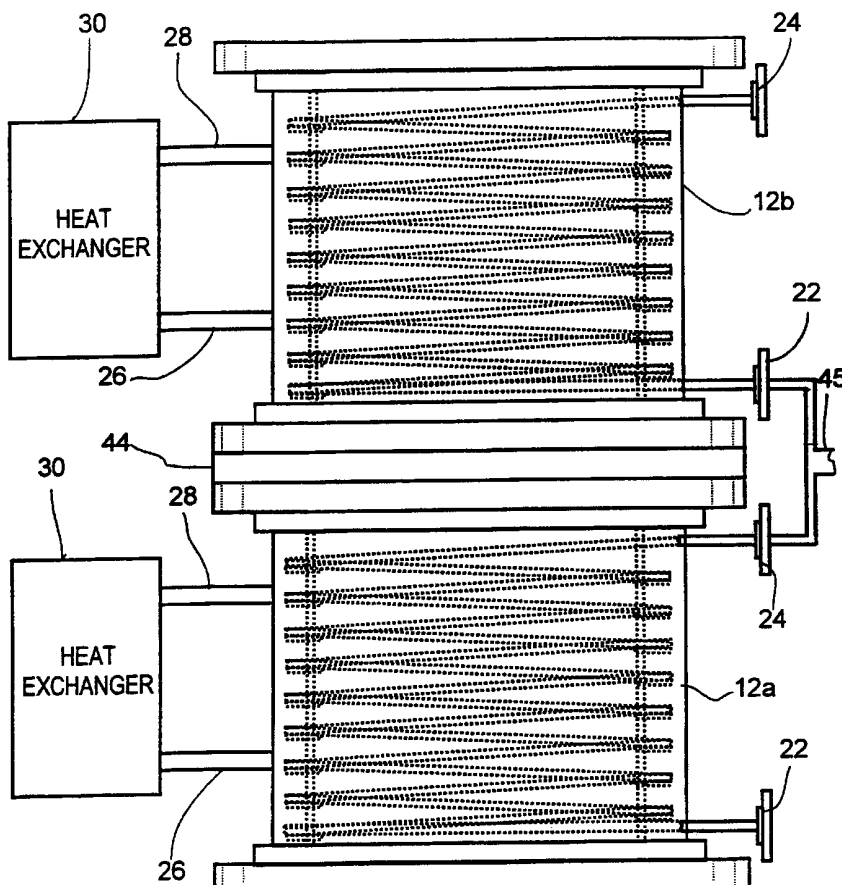
(10) International Publication Number
WO 01/36088 A1

- (51) International Patent Classification⁷: **B01J 19/24**, (72) Inventors; and
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(22) International Filing Date: 3 August 2000 (03.08.2000) **HINZ, Werner** [DE/US]; 25145 Old Depot Court, Grosse
(25) Filing Language: English (74) Agents: **MEYER, Udo** et al.; BASF Aktiengesellschaft,
D-67056 Ludwigshafen (DE).
(26) Publication Language: English (81) Designated States (national): AE, AG, AL, AM, AT, AU,
AZ, BA, BB, BG, BR, BY, BZ, CA, CH, CN, CR, CU, CZ,
DE, DK, DM, DZ, EE, ES, FI, GB, GD, GE, GH, GM, HR,
HU, ID, IL, IN, IS, JP, KE, KG, KP, KR, KZ, LC, LK, LR,
LS, LT, LU, LV, MA, MD, MG, MK, MN, MW, MX, MZ,
NO, NZ, PL, PT, RO, RU, SD, SE, SG, SI, SK, SL, TJ, TM,
TR, TT, TZ, UA, UG, US, UZ, VN, YU, ZA, ZW.
(30) Priority Data: 09/442,881 18 November 1999 (18.11.1999) US (84) Designated States (regional): ARIPO patent (GH, GM,
KE, LS, MW, MZ, SD, SL, SZ, TZ, UG, ZW), Eurasian

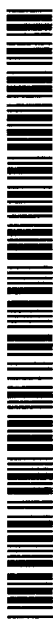
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[Continued on next page]

(54) Title: MODULAR REACTOR FOR CONTINUOUS POLYMERIZATION PROCESSES



(57) Abstract: A reactor assembly (10) for a continuous reaction process includes a module (12) having an outer tubular wall (16) defining an annular chamber (18). A spiral reaction tube (20) having an inlet end (22) and an outlet end (24), each of the ends extending out of the chamber, is spirally wound in the annular chamber (18) for transferring a reaction mixture through the chamber. In one embodiment, each module (12) further includes a heat exchanger inlet (26) and a heat exchanger outlet (28) that are operably connected to a heat exchanger that continuously flows a heat exchanger media through the annular chamber (18). A plurality of the modules (12) including a first module and a second module are connectable in series to thereby form a continuous reactor.



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patent (AM, AZ, BY, KG, KZ, MD, RU, TJ, TM), European patent (AT, BE, CH, CY, DE, DK, ES, FI, FR, GB, GR, IE, IT, LU, MC, NL, PT, SE), OAPI patent (BF, BJ, CF, CG, CI, CM, GA, GN, GW, ML, MR, NE, SN, TD, TG).

For two-letter codes and other abbreviations, refer to the "Guidance Notes on Codes and Abbreviations" appearing at the beginning of each regular issue of the PCT Gazette.

Published:

— *With international search report.*

MODULAR REACTOR FOR CONTINUOUS POLYMERIZATION PROCESSES

Description

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The subject invention relates to a continuous reactor for manufacturing a polymer. More specifically, the subject invention relates to a reactor assembly for creating a continuous reactor.

- 10 One common example of a polymer is a polyol, which is a building component of urethane polymers. Formation of a polyol typically requires that an alkoxylation reaction be performed. This type of reaction typically begins by mixing three reactants such as, for example, an alkylene oxide, an initiator having a reactive
- 15 hydrogen reactive with the alkylene oxide, and a catalyst such as potassium hydroxide. The most common type of reactor used for carrying out the alkoxylation reaction is a batch reactor into which the reactants are added in bulk to fulfill the stoichiometric requirements for manufacturing a desired polyol. Heating or
- 20 cooling the walls of the batch reactor with a large capacity heat exchanger fulfills the thermal requirements of the alkoxylation reaction. Additional reactants may be added during the reaction.

While the batch reactor has been the industrial standard for producing polyols, it has not proven to be cost efficient. In addition, the structure of the batch reactor does not facilitate the precise process control required to meet modern industrial quality standards for polymers.

- 30 Operating a batch reactor requires labor intensive attention to meet the stoichiometric requirements for producing an acceptable final product. The reactants are often manually added to the batch reactor to start the reactions therein. Subsequent reactant additions need to be made as the initiating reactants are
- 35 depleted. This requires closely monitoring the reaction time in the manufacturing environment. Should an operator misjudge a reaction time and not add a reactant as needed, the polymer batch is in jeopardy of being damaged.

- 40 An additional disadvantage of a batch reactor is the inability to precisely control the manufacturing process due to the large volumes of reactants in the reactor. Adjusting the chemical balance and temperatures in the reactor is difficult to perform rapidly due to the volume size of an industrial batch reactor.
- 45 Thus, it is often not feasible to run a first portion of the

reaction at a first reaction temperature and a second portion of the reaction at a second reaction temperature.

After a batch reaction has been completed, the reactor is shut
5 down for removal of the final product and for cleaning. Removing the final product from the batch reactor is labor intensive. In addition, while the product is being removed the reactor is not in use, which reduces manufacturing efficiency.

10 A need exists for a continuous reactor that provides the ability to continuously produce a polymer, such as a polyol. A continuous reactor that provides the ability to efficiently and precisely adjust the stoichiometric balance during production would improve the quality of the end product. Also, a continuous reac-
15 tor that allows for precise thermal control of the process would improve the quality of the end product.

In one embodiment, a continuous reactor for a continuous reaction process, comprises a plurality of modules, including at least a
20 first module and a second module, operably connected in series forming said continuous reactor. Each of the modules has an outer tubular wall defining an annular chamber, and a spiral reaction tube having an inlet end and an outlet end with each of the ends extending out of the chamber. The spiral reaction tube
25 is spirally wound in the chamber for transferring a reaction mixture through the chamber and the outlet end of the first module is operably connected to the inlet end of the second module.

Segregating the reactor into separate modules that can be connec-
30 ted in series provides manufacturing flexibility. The reaction tube has a smaller volume than a batch reactor for producing equivalent volumes of polymers; therefore, smaller volumes of reactants are disposed within the manufacturing process when using a continuous reactor. This reduces the potential for large
35 chemical spills. In addition, the operating conditions are easier to control for smaller volumes of reactants than for larger volumes because adjustments can be made more rapidly.

In the modular continuous reactor additional reactants can be ad-
40 ded in precise quantities at locations between each module. Separate modules that are operably connected in series also provide the ability to differentially control the reaction temperature at different stages of a reaction. The chambers of each module can communicate with separate heat exchangers, each heat exchanger
45 separately controlling the temperature of each module. Therefore, the first module can be heated to initiate a reaction, and

subsequent modules can be chilled for absorbing exothermic heat from the reaction.

Other advantages of the present invention will be readily appreciated as the same becomes better understood by reference to the following detailed description when considered in connection with the accompanying drawings wherein:

Figure 1a is a sectional view of a module of the subject invention;

Figure 1b is a top view of the module shown in Figure 1a;

Figure 2a is a schematic diagram of a plurality of modules aligned as a continuous reactor;

Figure 2b is a schematic diagram of an alternative embodiment of a plurality of modules aligned as a continuous reactor;

Figure 3 is a perspective view of a seal of the subject invention;

Figure 4 is an expanded view of a reactant feed line between adjacent modules;

Figure 5 is a perspective view of an inner tubular wall of the subject invention;

Figure 6 is a sectional view of a first and a second module connected in series; and

Figure 7 is a sectional view of a module having a plurality of spiral reaction tubes connected in series.

Referring to the Figures, wherein like numerals indicate like or corresponding parts throughout the several views, a reactor assembly for a continuous reaction process is generally shown at 10 in Figure 1a. The reactor assembly 10 includes a module 12 having an outer tubular wall 16 defining an annular chamber 18 within the outer tubular wall 16. The outer tubular wall 16 is contemplated to be between two feet and ten feet in diameter. Optionally, module 12 may include an inner tubular wall 14 that is inside the outer tubular wall 16 as shown in Figure 1a. It will be understood by one of ordinary skill in the art that inner tubular wall 14 is not necessary. When the inner tubular wall 14 is included, the annular chamber 18 is also defined by it and located between the inner tubular wall 14 and the outer tubular

wall 16. The inner tubular wall 14 also functions as a baffle as discussed below.

The module 12 further includes a reaction tube 20. The reaction tube 20 has an inlet end 22 and an outlet end 24, each of which extends out of the chamber 18. The reaction tube 20 is spirally wound, with a spiral diameter d_1 , in the chamber 18. The reaction tube 20 carries the reactants and transfers a reaction mixture through the module 12 and the chamber 18. The reaction tube 20 is preferably made of stainless steel; however, other materials may be selected that are compatible with the desired reactants.

For a given diameter outer tubular wall 16, the diameter d_1 of the spiral is selected to be about 1 to 2 inches less in diameter. Thus if the inner diameter of the outer tubular wall 16 were 48 inches, the spiral diameter d_1 would be between 47 and 46 inches. The spiral diameter of the reaction tube 20 and its internal diameter are specifically chosen to induce turbulent or pseudo-turbulent flow, defined as a flow with eddy current mixing off a continuously curved wall, within the reaction tube 20, which is beneficial to the polymerization reactions. The length of the reaction tube 20 can be adjusted for providing a desired reaction time within a module 12. The length can be varied between about thirty feet to several hundred feet depending upon the type of reaction desired in the reaction tube 20. The internal diameter of the reaction tube 20 can be varied between $\frac{1}{4}$ to 3.0 inches depending on the design of the reactor assembly 10. Generally there are about 15 to 30 complete spirals per module 12, but this number can vary depending on the design characteristics. The dimensions given for the reaction tube 20 are for example purposes only and can be modified to adjust the reaction time within each module 12.

In a preferred embodiment the reaction tube 20 is supported within annular chamber 18 by a plurality of rods 21 that extend from the inner tubular wall 14, as shown in Figure 1a. Preferably the inner tubular wall 14 includes internally threaded apertures (not shown) and the rods 21 include an enlarged portion having external threads (not shown). To assemble this embodiment, the rods 21 are slid from inside the inner tubular wall 14 and then threaded into the aperture to extend through the inner tubular wall 14 as shown.

In an alternative embodiment, shown in Figures 6 and 7, the reaction tube 20 is supported within the annular chamber 18 by a plurality of support rods 21 that extend from an inner surface 23

of the outer tubular wall 16. The support rods 21 preferably include external threads (not shown) that permit the support rods to be threaded into threaded apertures (not shown) in the inner surface 23, thus secured to the inner wall 23.

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Each annular chamber 18 includes a fluid inlet 26 and a fluid outlet 28 for continuously flowing a heat exchange fluid through the annular chamber 18 for controlling the reaction temperature within the reaction tube 20 disposed within the annular chamber 18. The heat exchange fluid preferably comprises a liquid, but it may also comprise a gas such as, for example, air. In one embodiment, each module 12 includes a heat exchanger 30 (see Figure 2a) operably connected to the fluid inlet 26 and the fluid outlet 28 for transferring a heat exchange fluid therethrough. In such an embodiment, the thermal environment of the reaction tube 20 of each module 12 can be tailored to a specific reaction.

The inner tubular wall 14 functions as a baffle and preferably creates a turbulent flow of the heat exchange fluid around the reactor tube 20. As is known in the art of heat transfer, turbulent flow transfers heat more efficiently than laminar flow does and is therefore, preferred for efficient heat exchange. In one embodiment, the inner tubular wall 14 includes a plurality of apertures 31 (see Figure 5) disposed therein for allowing the heat exchange fluid to flow into a space 32 defined by the inner tubular wall 14. Inside space 32 the heat exchange fluid flows in a predominantly laminar manner and this functions primarily as a heat sink for stabilizing the reaction temperature within the module 12. Outside the inner tubular wall 14, the heat exchange fluid flows in a turbulent manner for providing efficient heat exchange between the reaction tube 20 and the fluid. In an alternative embodiment the inner tubular wall 14 is a cylinder and it does not include any perforations.

As shown in Figures 1a and 1b, the module 12 includes an upper rim 40 opposite a lower rim 42. In a preferred embodiment, a seal 44 (shown in Figure 3) is affixed to each of the rims 40,42 with a fastening device (not shown) for retaining the heat exchange fluid within the annular chamber 18 and space 32. Alternatively, a plurality of modules 12 can be connected in series via their upper rim 40 and lower rim 42 without the use of the seal 44. In this embodiment the annular chambers 18 and inner spaces 32, if the inner tubular wall 14 is included, are all in communication with each other, thus each module 12 will be held at the same reaction temperature. The seal 44 can take the form of a blind flange as is known to those of skill in the art. In one embodiment, the rims 40,42 each include a plurality of rim

apertures 46, and the seal 44 includes a plurality of seal apertures 47 that can be aligned with the rim apertures 46. The apertures 46,47 receive a bolt (not shown) for affixing the seal 44 to each of the rims 40,42. However, other equivalent methods of attachment are known in the art and will suffice, such as, for example, a clamp.

A plurality of the modules 12, including a first module 12a and a second module 12b, are operably connected in series to form a continuous reactor shown schematically at 11 in Figures 2a and 2b. In forming the continuous reactor 11, the outlet end 24 of the reaction tube 20 from the first module 12a is operably connected to the inlet end 22 of the reaction tube 20 from the second module 12b.

By connecting several modules 12 in series, a chemical reaction that has typically required a batch process, can now be performed as a continuous process. For example, an alkoxylation reaction for producing a polyol is conventionally conducted in a batch reactor. Depending upon operating parameters and a desired final molecular weight of the polyol, any number of modules 12 can be operably connected in series. It is contemplated that only one module 12 may be required to achieve a chemical reaction desired to obtain a specific final polymer. Alternatively, as many as fourteen or more modules 12 might be required to manufacture the desired polymer. An additional benefit of the modules 12 is that the modules 12 can be individually drained for minimizing the amount of lost reactant during product changes.

Figures 2a and 2b show two alternative embodiments of the continuous reactor 11. In Figure 2a, the inlet end 22 of the reaction tube 20 in the first module 12a of the continuous reactor 11 is operably connected to a first feed line 34, a second feed line 36, and a third feed line 38. The first feed line 34 is connected to a first reactant supply tank 35, the second feed line 36 is connected to a second reactant supply tank 37, and the third feed line 38 is connected to a third reactant supply tank 39. In Figure 2b, the inlet end 22 of the reaction tube 20 in the first module 12a of the continuous reactor 11 is operably connected to the first reactant feed line 34 and the second reactant feed line 36. In both embodiments the subsequent inlet ends 22 of each reaction tube 20 in each of the modules 12b - 12x is operably connected to both the second and third feed lines 36 and 38. The outlet end 24 of each reaction tube 20 is operably connected to the inlet end 22 of the reaction tube 20 in the next module 12. A static mixer 48 ensures complete mixing of the reactants.

As would be understood by one of ordinary skill in the art, the present design would permit any number of feed lines and reactant supply tanks to feed into the inlet end 22 of any reaction tube 20. In addition, each supply tank can supply either a single reactant or a mixture of reactants. In addition, at any point between two reaction tubes 20, product could be removed or further manipulated prior to entry through the inlet end 22 of the next reactor tube 20.

Each module 12 further includes a heat exchanger 30 for maintaining the reaction temperature. Each feed line 34, 36, and 38 further includes a pump 48 for transferring reactants from the first 35, second 37, and third reactant supply tanks 39, respectively, through the associated feed line 34, 36, 38. In a preferred embodiment the reactants are fed at a pressure greater than the vapor pressures of the reactants. In such an embodiment, the pumps 46 maintain the pressure in the feed lines 34, 36, and 38, and reaction tubes 20 at a pressure between 200 to 1500 pounds per square inch. The upper limit of the pressure is determined by the structural stability of the reaction tube 20, other materials may allow even higher pressures, thus 1500 pounds per square inch is for example purposes only. It is possible that one or all of the reactants have a low vapor pressure. As is known in the art of manufacturing polyols, the alkoxylation reaction is most efficient when all the reactants are in the liquid phase. Thus, it is desired to deliver the reactants into the reaction tube 20 in the liquid phase. These high pressures ensure that reactants with low vapor pressure, for example, propylene oxide or ethylene oxide, are maintained in the liquid state. In an alternative embodiment, the pressure produced by the pumps 46 is lower at a level sufficient to exceed the pressure in reaction tube 20 so that the reactants will continue to enter the modules 12. Reaction product leaving the last module 12x is routed to either storage tanks or is further processed to purify the reaction product as is known in the art. For example, following a continuous alkoxylation reaction the product would be stripped of residual unreacted alkylene oxide and catalyst and then further purified.

The continuous reactor 11 shown in Figures 2a and 2b can be used for the continuous production of various polyols. Figure 2a permits the three major reactants in an alkoxylation reaction an alkylene oxide, initiator, and catalyst to be added to the first module 12a. Subsequent modules receive additional alkylene oxide and catalyst, if necessary. In Figure 2b only two reactants are delivered to the first module 12a, this design is useful when the alkylene oxide can self catalyze a reaction with an initiator in

the absence of additional catalyst. In Figure 2b subsequent modules receive the alkylene oxide and, optionally, catalyst through the second and third feed lines 36 and 38 respectively. Typical alkylene oxides are ethylene oxide, propylene oxide, and butylene oxide. Many other alkylene oxides are known in the art. The initiator can comprise any polyol or a reactive amine such as toluene diamine. Catalysts are well known and include potassium hydroxide, sodium hydroxide, cesium hydroxide, Lewis acids and double metal complex catalysts. Ethylene oxide can self catalyze with toluene diamine as the initiator. As will be understood by one of ordinary skill in the art the continuous reactor 11 discloses allows for great flexibility in the reaction parameters. One can create a polyol that has a block, random or heteric configuration by altering what is fed in at each module 12. The design also provides the ability to maintain optimum stoichiometric ratios for the desired reaction rate throughout the continuous reactor 11. As a reactant is depleted within the reaction tube 20, the reactant is replaced at the inlet end 22 of each module 12. Some or all of the reactants can be added at the various modules 12 depending upon the stoichiometric ratios desired. Figure 4 shows an enlarged view of the third reactant operably connected to the inlet end 22 of the second module 12b.

As a polymerization reaction, such as the alkoxylation reaction, proceeds through the continuous reactor 11 towards the termination stage of the reaction, the reaction mixture generally becomes more viscous thus reducing the volumetric flow rate. In addition, introducing additional volumes of reactants to various modules 12 of the continuous reactor reduces the effective volumetric flow rate. To maintain a constant volumetric flow rate throughout the reactor, the reaction tube 20 disposed within the later modules 12 is greater in diameter than the reaction tube 20 disposed within the first modules 12. For example, the inside diameter of the reaction tube 20 disposed within the first five modules 12 can be $\frac{3}{4}$ inch, and the inside diameter of the reaction tube 20 disposed within the remaining modules 12 can be $1\frac{1}{4}$ inches. If needed, each subsequent module 12 includes a reaction tube 20 having an internal diameter greater than the diameter of the reaction tube 20 in the prior module 12 for maintaining constant reactant flow rate throughout the continuous reactor 11.

The plurality of modules 12 can be configured in any formation desired for meeting processing and facility requirements. The modules 12 can be stacked upon each other, as shown in Figure 6, or they may be separated but operably connected. Figure 6 shows a first module 12a and a second module 12b operably connected to each other in series with seal 44 between them. Additionally,

the outlet end 24 of the first module 12a is operably connected to the inlet end 22 of the second module 12b. A feed line 45 permits addition of additional reactants to the product exiting the first module 12a for reaction in the second module 12b. A
5 separate heat exchanger 30 can be operably connected to each module 12a, 12b. This allows different temperatures to be achieved within each module 12, and therefore, at different stages of the reaction. For example, the first module 12a can be heated to initiate the reaction and subsequent modules 12 can be cooled for
10 absorbing exothermic heat from the reaction. Alternatively, the annular chambers 18 and space 32 of each module 12 can be operably connected to each other by eliminating seal 44, plugging a fluid inlet 26 in one module 12 and a fluid outlet 38 in another module 12 and connecting the remaining fluid inlet 26 and fluid
15 outlet 28 to a single heat exchanger 30 effectively forming a single large thermal chamber 18 for the continuous reactor 11. If the modules 12 are connected in this fashion, a uniform reaction temperature within both of the reaction tubes 20 throughout the continuous reactor 11.

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In Figure 7 an alternative embodiment of a module 12 is shown. In this embodiment the module 12 is elongated and includes a plurality of reaction tubes 20, in the figure two are shown but, the module could include over seven reaction tubes 20. In this embodi-
25 ment, the outlet 24 of each reaction tube 20 exits the outer tubular wall 16 and is operably connected to the inlet 22 of a subsequent reaction tube 20. The module 12 is connected to a single heat exchanger 30 and thus each reaction tube 20 is maintained at the same reaction temperature. Feed line 45 is operably
30 connected between subsequent reaction tubes 20 to allow for introduction of additional reactants.

The invention has been described in an illustrative manner, and it is to be understood that the terminology, which has been used,
35 is intended to be in the nature of words of description rather than of limitation.

Obviously, many modifications and variations of the present invention are possible in light of the above teachings. It is,
40 therefore, to be understood that within the scope of the appended claims, wherein reference numerals are merely for convenience and are not to be in any way limiting, the invention may be practiced otherwise than as specifically described.

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What is claimed is:

1. A continuous reactor for a continuous reaction process, said
5 continuous reactor comprising:

a plurality of modules, including at least a first module and
a second module, operably connected in series forming said
continuous reactor;
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each of said modules having an outer tubular wall defining an
annular chamber, and a spiral reaction tube having an inlet
end and an outlet end with each of said ends extending out of
said chamber, said spiral reaction tube being spirally wound
15 in said chamber for transferring a reaction mixture through
said chamber; and said outlet end of said first module being
operably connected to said inlet end of said second module.
2. A continuous reactor as set forth in claim 1 wherein, said
20 annular chamber of each of said modules includes a fluid in-
let and a fluid outlet, said fluid inlet and said fluid out-
let connected to a heat exchanger, said heat exchanger conti-
nuously flowing a heat exchange fluid through said chamber
for establishing and maintaining a reaction temperature in
25 said annular chamber and said reaction tube of each of said
modules.
3. A continuous reactor as set forth in claim 2 wherein said re-
actor includes a first heat exchanger operably connected to
30 said annular chamber of said first module, and a second heat
exchanger operably connected to said annular chamber of said
second module, and said first heat exchanger maintains a
reaction temperature within said first module that is diffe-
rent from the reaction temperature maintained by said second
35 heat exchanger within said second module.
4. A continuous reactor as set forth in claim 1 wherein said in-
let end of each of said reaction tubes of each of said modu-
les is operably connected to at least one reactant supply
40 tank through a feed line.
5. A continuous reactor as set forth in claim 1 wherein said in-
let end of plurality of said reaction tubes is operably con-
nected to a plurality of reactant supply tanks through a plu-
45 rality of feed lines.

6. A continuous reactor as set forth in claim 5 wherein each of said feed lines includes a pump, said pump transferring a reactant or a mixture of reactants through said feed line at a pressure greater than the vapor pressure of said reactant or mixture of reactants.
- 5
7. A continuous reactor as set forth in claim 6 wherein each of said pumps pressurizes an associated reactant or mixture of reactants to a pressure between 200 to 1000 pounds per square inch.
- 10
8. A continuous reactor as set forth in claim 6 wherein each of said pumps pressurizes an associated reactant or mixture of reactants to a pressure greater than a pressure in said plurality of reaction tubes.
- 15
9. A continuous reactor as set forth in claim 1 wherein said reaction tube disposed within each of a portion of said plurality of modules has an internal diameter that is greater than an internal diameter of said reaction tube disposed within said first module.
- 20
10. A continuous reactor as set forth in claim 1 wherein said first and said second modules each include an upper rim and a lower rim, and a fastening device secures said upper rim of said first module to said lower rim of said second module.
- 25
11. A continuous reactor as set forth in claim 10 further including a seal disposed between said upper rim of said first module and said lower rim of said second module.
- 30
12. A continuous reactor as set forth in claim 1, wherein each of said modules further includes an inner tubular wall spaced from said outer tubular wall with said annular chamber defined between said inner tubular wall and said outer tubular wall, and said spiral reaction tube is spirally wound in said annular chamber.
- 35
13. A continuous reactor for a continuous reaction process, said continuous reactor comprising:
- 40
- a plurality of modules, including at least a first module and a second module, operably connected in series forming a continuous reactor;
- 45

each of said modules having a spiral reaction tube and an outer tubular wall, said outer tubular wall defining an annular chamber, said annular chamber including a fluid inlet and a fluid outlet;

5

said spiral reaction tube spirally wound and supported in said annular chamber, said reaction tube having an inlet end and an outlet end, each of said ends extending through said outer tubular wall for transferring a reaction mixture through said annular chamber, said outlet end of said first module being operably connected to said inlet end of said second module; and

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a heat exchanger connected to at least one of said modules through said fluid inlet and said fluid outlet, said heat exchanger continuously flowing a heat exchange fluid through said annular chamber for one of cooling and heating.

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14. A continuous reactor as set forth in claim 13, wherein each of said modules further includes an inner tubular wall spaced from said outer tubular wall with said annular chamber defined between said inner tubular wall and said outer tubular wall, and said spiral reaction tube is spirally wound in said annular chamber between said inner tubular wall and said outer tubular wall.

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15. A continuous reactor as set forth in claim 13, wherein: said annular chamber of said first module is in fluid communication with said annular chamber of said second module; said fluid inlet of said first module and said fluid outlet of said second module are operably connected to said heat exchanger; and said reaction tubes of said first and said second module are maintained at the same reaction temperature by said heat exchanger.

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16. A continuous reactor as set forth in claim 13, wherein said fluid inlet and said fluid outlet of said first module are connected to a first heat exchanger, said first heat exchanger maintaining said first module at a first reaction temperature and said fluid inlet and said fluid outlet of another of said plurality of modules are connected to a second heat exchanger said second heat exchanger maintaining said another of said plurality of modules at a second reaction temperature, said second reaction temperature other than said first reaction temperature.

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17. A continuous reactor as set forth in claim 13, wherein said inlet end of each of said reaction tubes is operably connected to at least one reactant supply tank through a feed line.

5 18. A continuous reactor for a continuous reaction process, said continuous reactor comprising:

10 a plurality of modules, including at least a first module and a second module, operably connected in series forming a continuous reactor;

15 each of said modules having an inner tubular wall and an outer tubular wall with an annular chamber defined between said inner and said outer tubular walls, said annular chamber including a fluid inlet and a fluid outlet;

20 a spiral reaction tube spirally wound and supported in said annular chamber, said reaction tube having an inlet end and an outlet end, each of said ends extending through said outer tubular wall for transferring a reaction mixture through said annular chamber, said outlet end of said first module being operably connected to said inlet end of said second module; and

25 a heat exchanger connected to at least one of said modules through said fluid inlet and said fluid outlet, said heat exchanger continuously flowing a heat exchange fluid through said annular chamber for one of cooling and heating.

30 19. A continuous reactor as set forth in claim 18, wherein said fluid inlet and said fluid outlet of said first module are connected to a first heat exchanger, said first heat exchanger maintaining said first module at a first reaction temperature and said fluid inlet and said fluid outlet of another of said plurality of modules are connected to a second heat exchanger said second heat exchanger maintaining said another of said plurality of modules at a second reaction temperature, said second reaction temperature other than said first reaction temperature.

40

20. A continuous reactor as set forth in claim 18, wherein said inlet end of each of said reaction tubes is operably connected to at least one reactant supply tank through a feed line.

45 21. A reactor assembly for a continuous reaction process, said assembly comprising:

- 5 a module having an outer tubular wall defining an annular chamber; and a spiral reaction tube having an inlet end and an outlet end each of said ends extending out of said chamber, said spiral reaction tube spirally wound in said annular chamber for transferring a reaction mixture through said chamber.
- 10 22. A reactor assembly as set forth in claim 21 wherein said module further includes support rods disposed therein, said support rods supporting said spiral reaction tube.
- 15 23. A reactor assembly as set forth in claim 21 wherein said reaction tube has an internal diameter from about 0.25 inches to about 3 inches.
- 20 24. A reactor assembly as set forth in claim 21 wherein said outer tubular wall has an internal diameter of from about two feet to about ten feet.
- 25 25. A reactor assembly as set forth in claim 21 wherein said reaction tube has a spiral diameter of from about 20 inches to about 10 feet.
- 30 26. A reactor assembly as set forth in claim 21, wherein said module further includes an inner tubular wall spaced from said outer tubular wall with said annular chamber defined between said outer tubular wall and said inner tubular wall, and said spiral reaction tube is spirally wound in said annular chamber.
- 35 27. A reactor assembly as set forth in claim 26 wherein said inner tubular wall includes a plurality of apertures, said apertures permitting fluid communication between said annular chamber and a space defined by said inner tubular wall.
- 40 28. A reactor assembly as set forth in claim 21, wherein said module further includes an fluid inlet and a fluid outlet in communication with said annular chamber, said fluid inlet and said fluid outlet for providing a continuous flow a of heat exchange fluid through said annular chamber.
- 45 29. A continuous reactor assembly as set forth in claim 21, wherein said module further includes an upper rim opposite an lower rim, each of said rims including a seal secured to said rim, said annular chamber defined by said outer tubular wall and said seals.

30. A continuous reactor assembly as set forth in claim 21, wherein said module further includes a plurality of spiral reaction tubes operably connected in series, including a first spiral reaction tube and a second spiral reaction tube; each of said spiral reaction tubes having a an inlet end and an outlet end extending out of said chamber, and said outlet end of said first spiral reaction tube operably connected to said inlet end of said second spiral reaction tube.
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- 10 31. A reactor assembly for a continuous reaction process, said assembly comprising:
- a module having an outer tubular wall defining an annular chamber, a fluid inlet and a fluid outlet in communication with said annular chamber for providing a continuous flow of a heat exchange fluid through said annular chamber; and
- 15
- a spiral reaction tube having an inlet end and an outlet end each of said ends extending out of said chamber, said spiral reaction tube spirally wound in said annular chamber for transferring a reaction mixture through said chamber.
- 20
32. A continuous reactor assembly as set forth in claim 31, wherein said module further includes a plurality of spiral reaction tubes operably connected in series, including a first spiral reaction tube and a second spiral reaction tube; each of said spiral reaction tubes having a an inlet end and an outlet end extending out of said chamber, and said outlet end of said first spiral reaction tube operably connected to said inlet end of said second spiral reaction tube.
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33. A continuous reactor assembly as set forth in claim 31, further including a heat exchanger in communication with said fluid inlet and said fluid outlet, said heat exchanger for continuously flowing a heat exchange fluid through said annular chamber.
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34. A reactor assembly for a continuous reaction process, said assembly comprising:
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- a module having an inner tubular wall and an outer tubular wall defining an annular chamber between them, a fluid inlet and a fluid outlet in communication with said annular chamber;
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a spiral reaction tube having an inlet end and an outlet end each of said ends extending out of said chamber, said spiral reaction tube spirally wound in said annular chamber for transferring a reaction mixture through said chamber; and

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a heat exchanger in communication with said fluid inlet and said fluid outlet, said heat exchanger continuously flowing a heat exchange fluid through said annular chamber.

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FIG.1A

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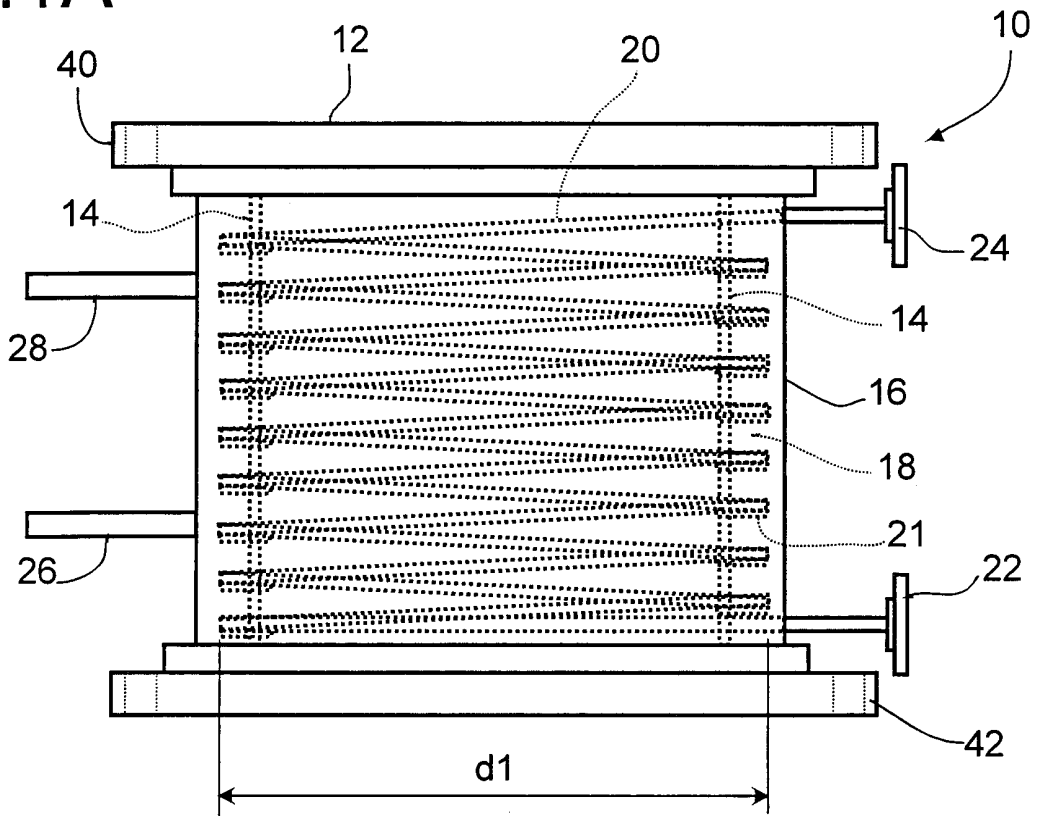


FIG.1B

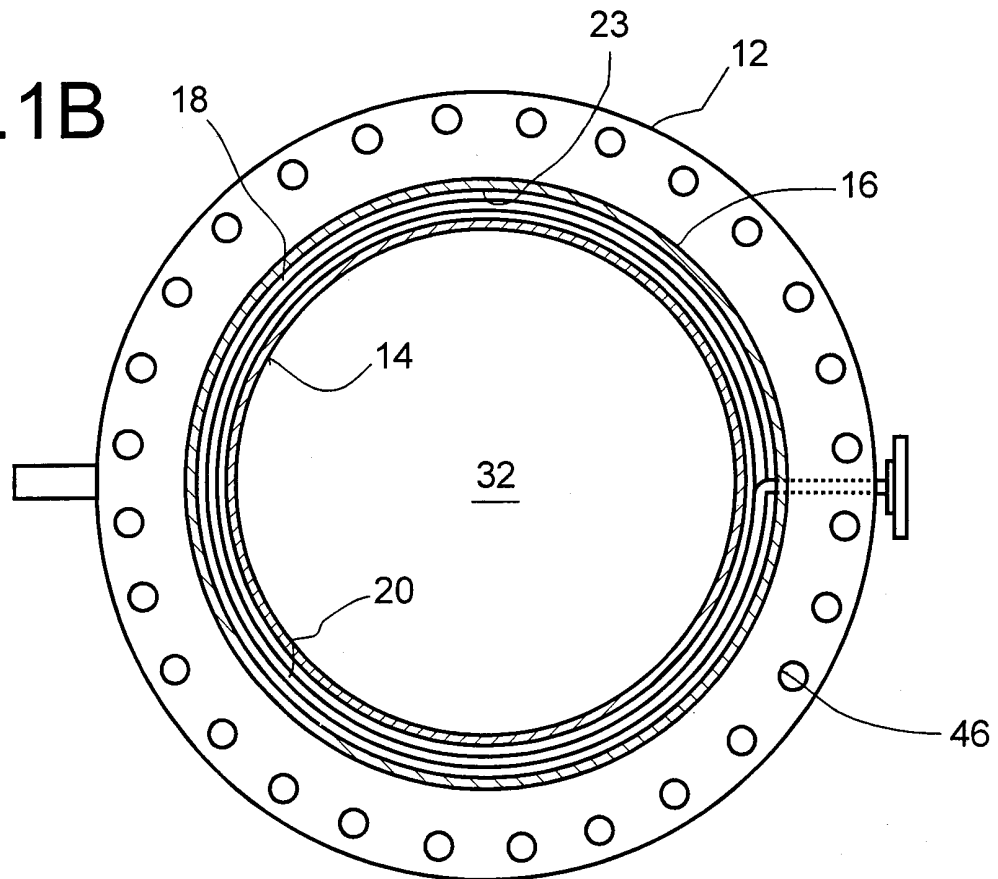


FIG.2A

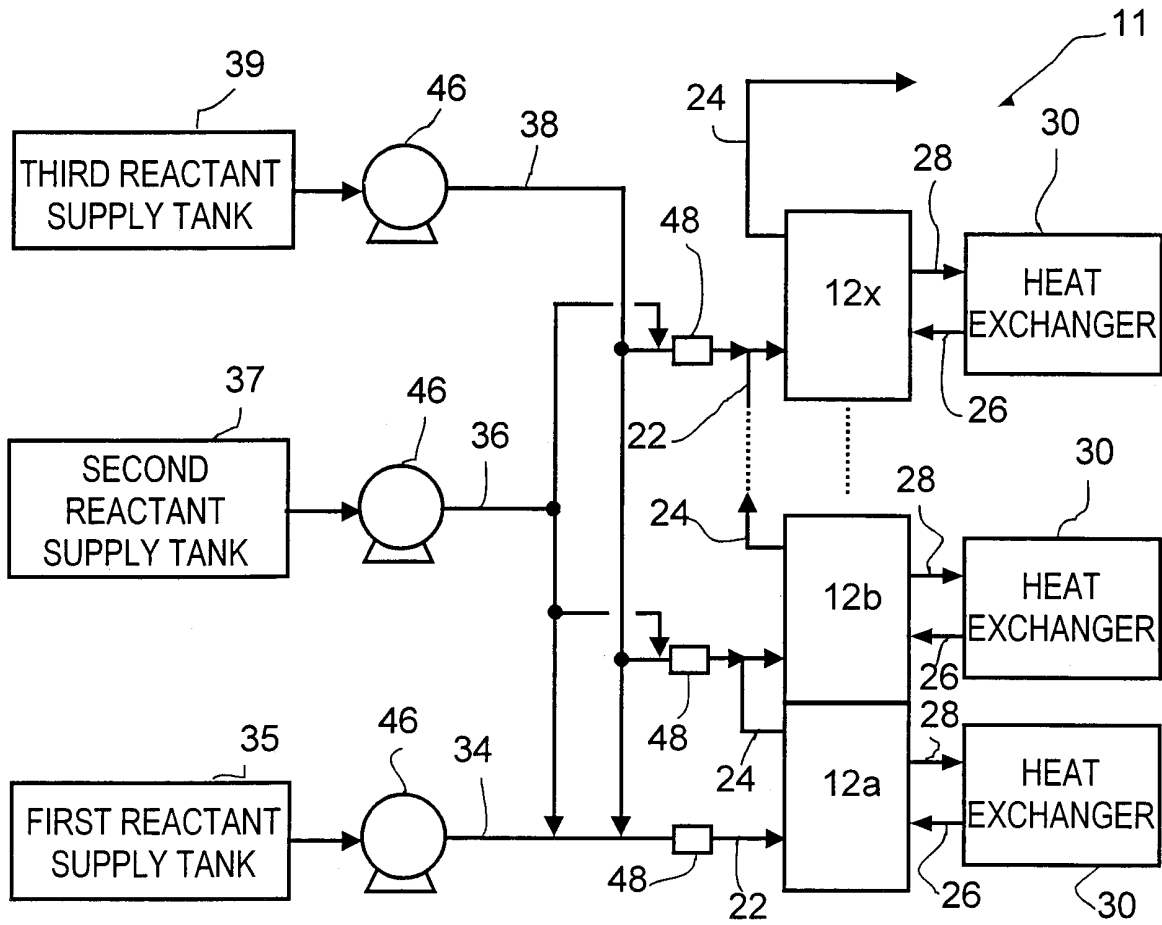


FIG.2B

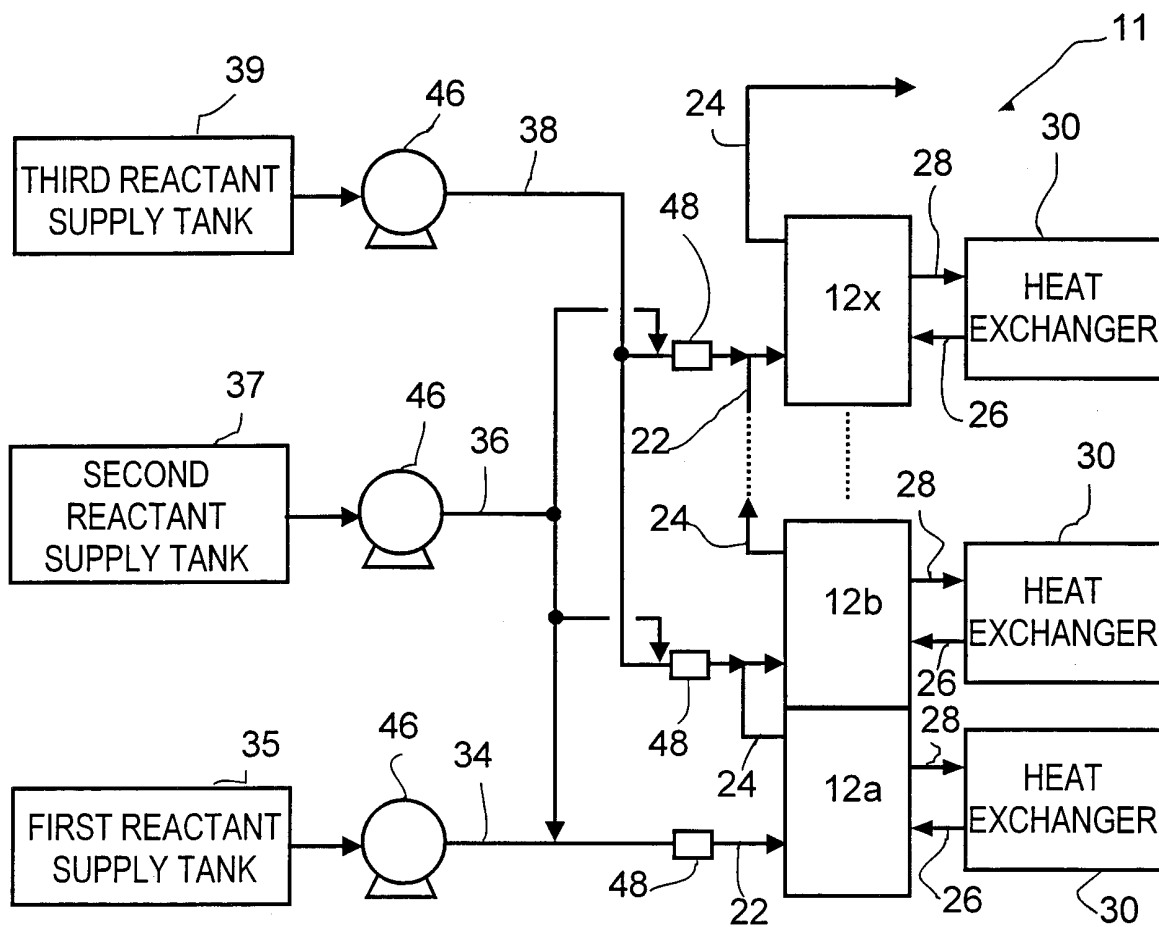


FIG.3

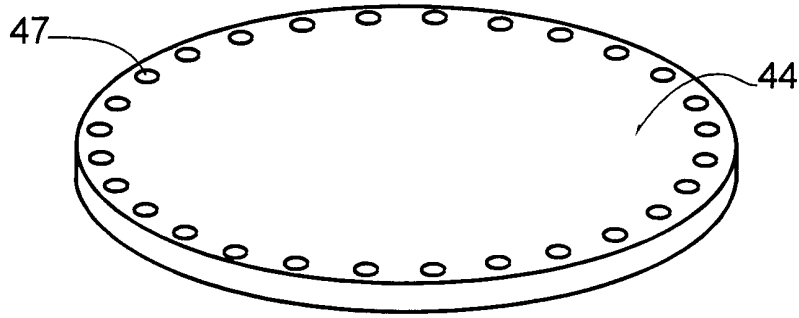


FIG.4

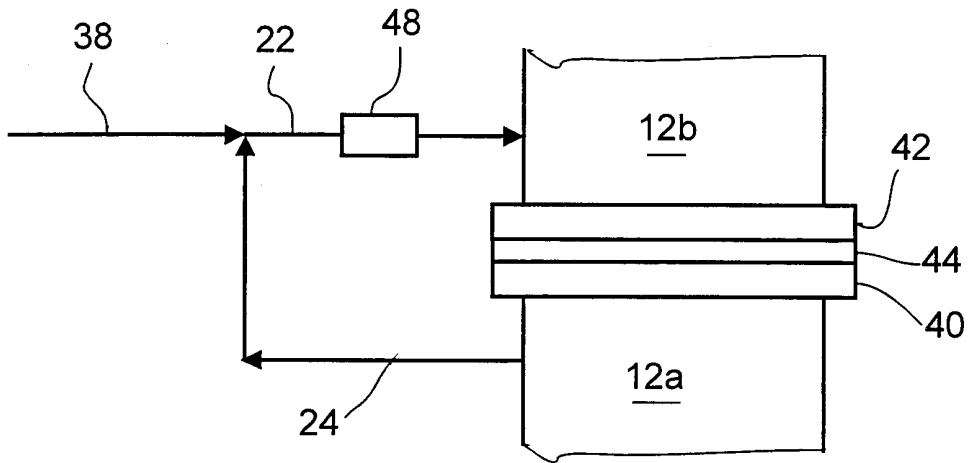


FIG.5

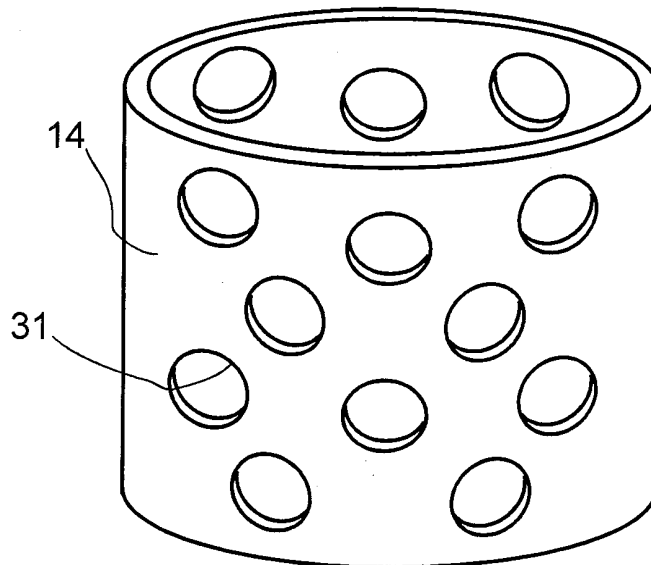


FIG.6

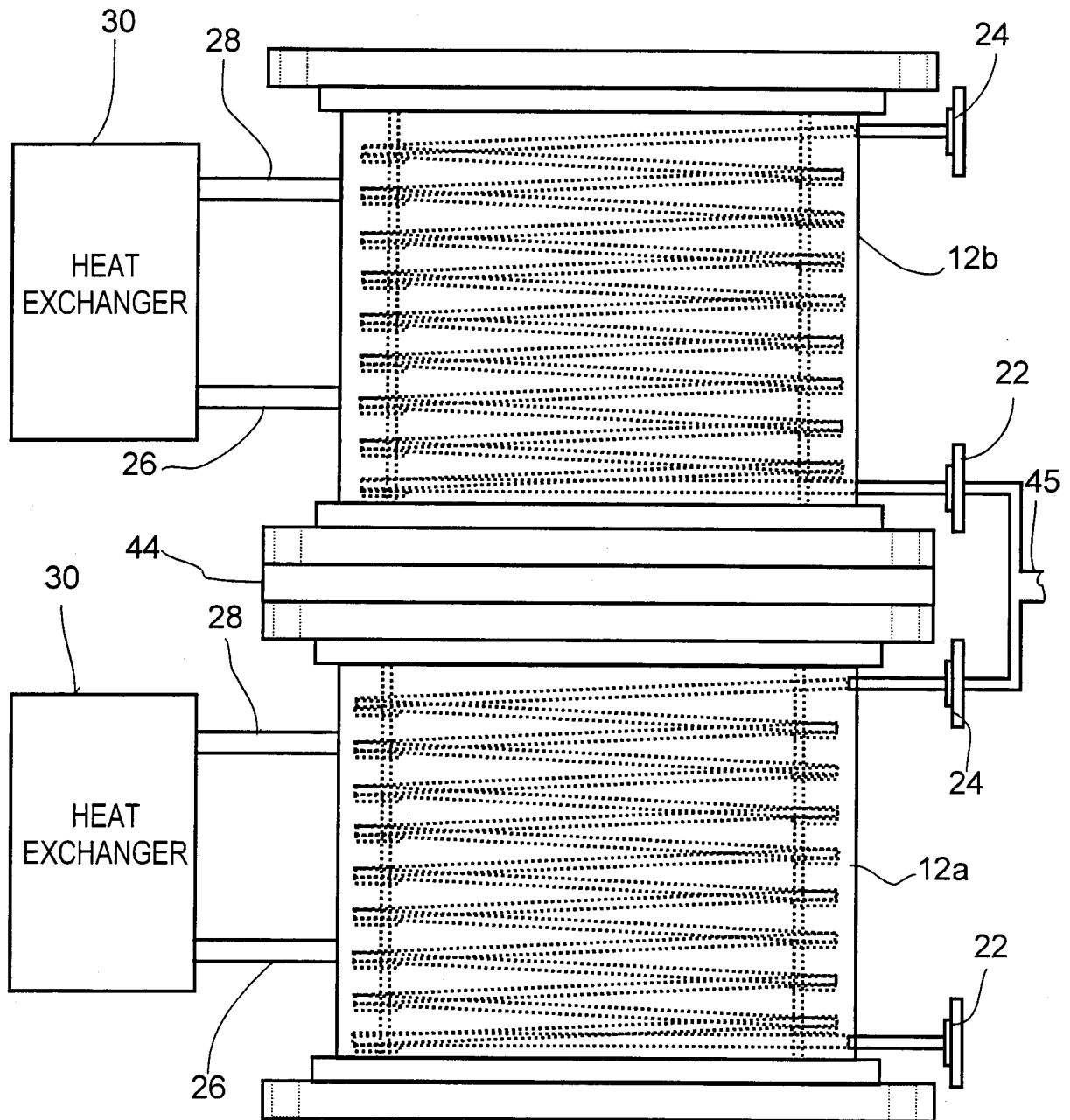
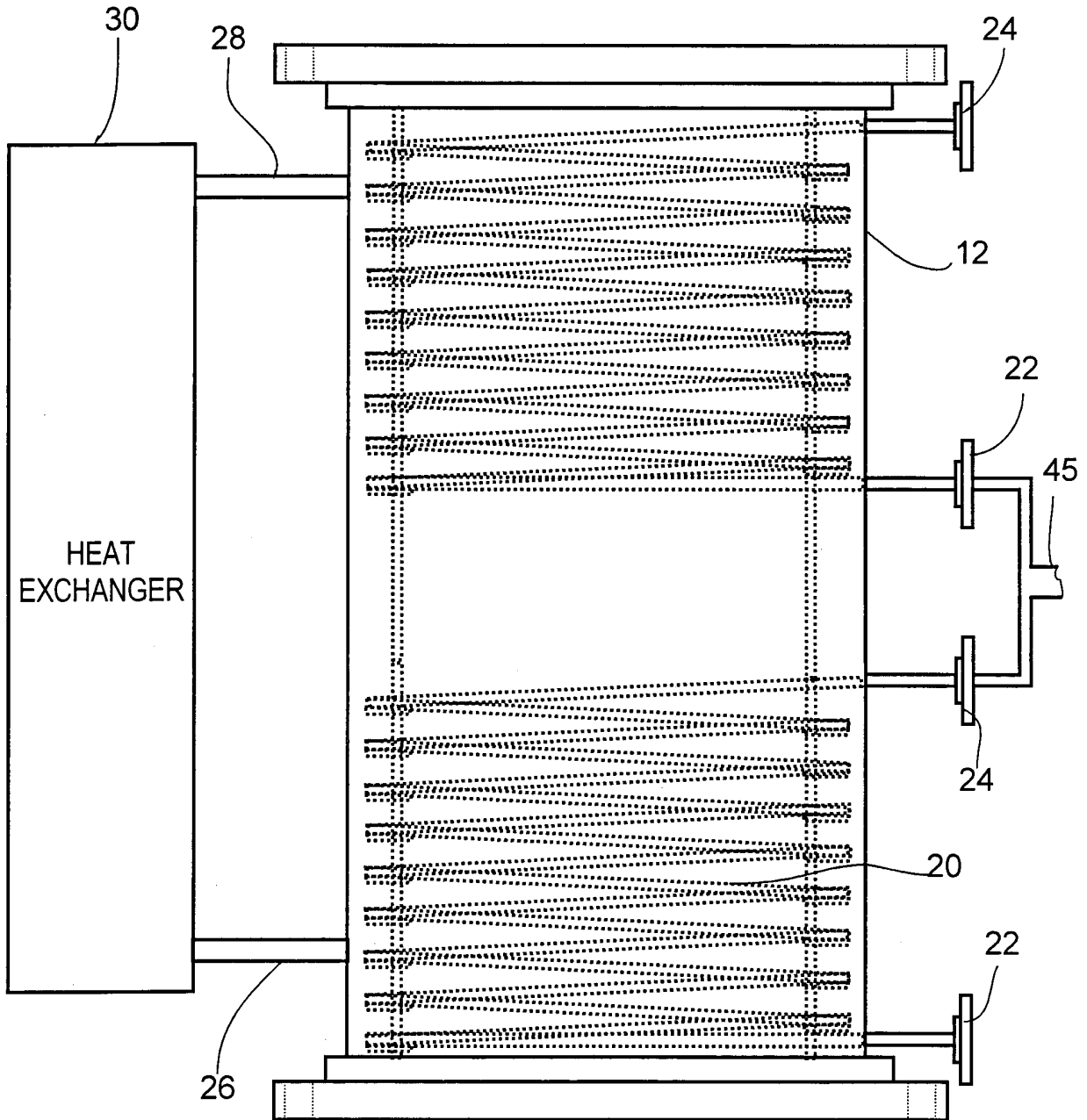


FIG. 7



INTERNATIONAL SEARCH REPORT

International Application No

PCT/EP 00/07501

A. CLASSIFICATION OF SUBJECT MATTER
 IPC 7 B01J19/24 C08G65/26

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)
 IPC 7 B01J C08G

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

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

EPO-Internal, WPI Data, PAJ

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category °	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X Y	US 4 241 043 A (HETZEL HARTMUT) 23 December 1980 (1980-12-23) the whole document ---	21, 26, 28, 31, 34 1-9, 12-20, 23, 32, 33
Y	DE 196 15 974 A (TOHOKU ELECTRIC POWER CO ;OHEI DEVELOPMENTAL IND CO (JP)) 24 October 1996 (1996-10-24) page 3, line 32 -page 4, line 19 page 4, line 62 -page 5, line 14 page 5, line 63 -page 7, line 20 figures 3-12 --- -/--	1-9, 12-20, 23, 32, 33

Further documents are listed in the continuation of box C. Patent family members are listed in annex.

° Special categories of cited documents :

<p>"A" document defining the general state of the art which is not considered to be of particular relevance</p> <p>"E" earlier document but published on or after the international filing date</p> <p>"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)</p> <p>"O" document referring to an oral disclosure, use, exhibition or other means</p> <p>"P" document published prior to the international filing date but later than the priority date claimed</p>	<p>"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</p> <p>"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</p> <p>"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.</p> <p>"&" document member of the same patent family</p>
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Date of the actual completion of the international search 23 October 2000	Date of mailing of the international search report 30/10/2000
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Name and mailing address of the ISA European Patent Office, P.B. 5818 Patentlaan 2 NL - 2280 HV Rijswijk Tel. (+31-70) 340-2040, Tx. 31 651 epo nl, Fax: (+31-70) 340-3016	Authorized officer Vlassis, M
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INTERNATIONAL SEARCH REPORT

Interr. Application No

PCT/EP 00/07501

C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT

Category °	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
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INTERNATIONAL SEARCH REPORT

Information on patent family members

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PCT/EP 00/07501

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