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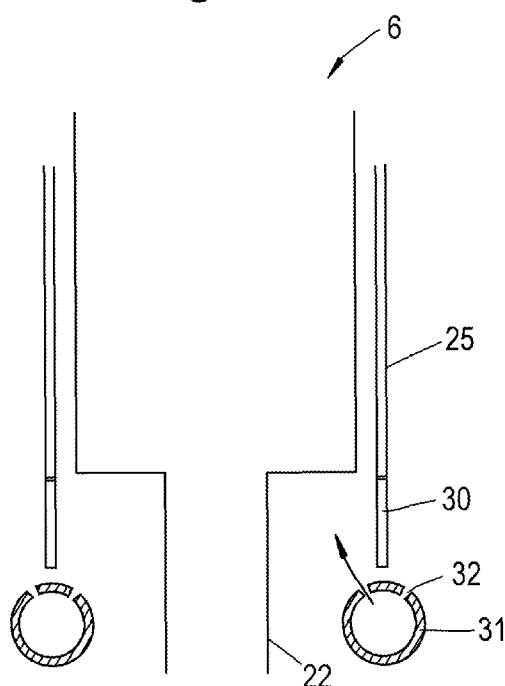
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(54) Title: REACTOR COMPRISING COOLING MODULES

Fig.7



(57) Abstract: The invention relates to a reactor (1) for carrying out an exothermic process comprising a reactor shell (2), inlets (3, 7) for introducing reactants and coolant into the reactor shell (2), outlets (8) for removing product and coolant from the reactor shell (2), and a plurality of cooling modules (6), the reactor comprising for at least some of the modules (6) a skirt (30) for guiding gas underneath the modules (6).



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REACTOR COMPRISING COOLING MODULES

The present invention relates to a reactor for carrying out an exothermic process, such as a Fischer-Tropsch process, comprising cooling modules. The reactor is compartmentalized. The invention further relates to the use of such a reactor for carrying out an exothermic process.

As is explained in WO 2005/075065, Fischer-Tropsch processes are often used for the conversion of gaseous hydrocarbon feed stocks into liquid and/or solid hydrocarbons. The feed stock, e.g. natural gas, associated gas, coal-bed methane, residual (crude) oil fractions, coal and/or biomass is converted in a first step to a mixture of hydrogen and carbon monoxide, also known as synthesis gas or syngas. The synthesis gas is then converted in a second step over a suitable catalyst at elevated temperature and pressure into paraffinic compounds ranging from methane to high molecular weight molecules comprising up to 200 carbon atoms, or, under particular circumstances, more.

Numerous types of reactor systems have been developed for carrying out the Fischer-Tropsch reaction. Fischer-Tropsch reactor systems include fixed bed reactors, in particular multi-tubular fixed bed reactors, fluidised bed reactors, such as entrained fluidised bed reactors and fixed fluidised bed reactors, and slurry bed reactors, such as three-phase slurry bubble columns and ebullated bed reactors.

The Fischer-Tropsch reaction is highly exothermic and temperature sensitive and thus requires careful

temperature control to maintain optimum operating conditions and hydrocarbon product selectivity.

Commercial fixed-bed and three-phase slurry reactors typically utilise boiling water to remove reaction heat.

5 In fixed-bed reactors, individual reactor tubes are located within a shell containing water/steam typically fed to the reactor via flanges in the shell wall. The reaction heat raises the temperature of the catalyst bed within each tube. This thermal energy is transferred to
10 the tube wall forcing the surrounding water to boil. In the slurry design, cooling tubes are placed within the slurry volume and heat is transferred from the liquid continuous matrix to the tube walls. The production of steam within the tubes provides cooling.

15 US 2,853,369 describes a compartmentalized slurry reactor. The reactor comprises heat exchange pipes and vertical shafts. The shafts are placed at a relatively high level in the reactor, and form walls along the mid part of the heat exchange pipes. Gas may enter the
20 reactor via a single inlet in the bottom of the reactor, or via a gas distribution means across the bottom which directs the gas towards the bottom of the reactor. US 2,853,369 mentions that in the sump underneath the shafts the gas distribution is large and uneven.

25 It is an object of the present invention to provide a reactor which allows relatively simple yet robust construction and operation.

To this end, the reactor according to the present invention is characterised in that the reactor comprises
30 a plurality of cooling modules and for at least some of the modules a skirt for guiding gas underneath the modules.

The reactor is suitable for carrying out an exothermic process, such as a Fischer-Tropsch process.
35 The reactor comprises a reactor shell, inlets for

introducing reactants and coolant into the reactor shell, outlets for removing product and coolant from the reactor shell.

5 A reactor according to the present invention is a slurry reactor. The cooling modules in the reactor according to the present invention are suitable for use in a slurry reactor. The cooling tubes of the cooling modules are normally placed within the volume in which the reaction takes place and heat is transferred from the
10 liquid continuous matrix to the tube walls.

The catalyst in the reaction volume may be a particulate catalyst. Additionally or alternatively, the catalyst in the reaction volume may be a structured catalyst, for example a shaped porous structure. A
15 structured catalyst may form an ebullated bed. A structured catalyst may be fixed in the reaction volume. When the catalyst is fixed in the slurry reactor, the reactor may be referred to as an "immobilized slurry reactor".

20 The cooling modules in the reactor of the current invention may be any type of cooling module. Examples of suitable cooling modules are described in WO 2005/035108, WO 2005/065813, WO 2005/075065, WO 2006/097906, and US 6,060,524.

25 Preferably the cooling module comprises a coolant inlet, a coolant distribution chamber, a plurality of cooling tubes, a coolant collection chamber, and a coolant discharge.

The cooling tubes are preferably arranged as
30 separate cooling tubes. When the cooling module is in use, coolant may pass from the coolant distribution chamber through the cooling tubes to the coolant collection chamber. Preferably at least 80%, more preferably at least 90%, of the cooling tubes are
35 arranged separately with a distance to the nearest

cooling tube of at least 1 cm, preferably at least 2 cm. Preferably at least 80%, more preferably at least 90%, of the cooling tubes have a distance of at least 1 cm, preferably at least 2 cm, to its nearest cooling tube
5 along the length of the cooling tubes. The distance between two adjacent cooling tubes in the cooling module of the present invention preferably is less than 50 cm, more preferably less than 20 cm, along the length of the cooling tubes.

10 In one embodiment of the reactor of the present invention, one or more of the cooling modules comprises one or more passages extending through the distribution chamber to enable fluid communication between the space
15 underneath the distribution chamber, and the space between the cooling tubes. The cooling module(s) may additionally comprise one or more passages extending through the collection chamber to enable fluid communication between the space between the cooling tubes
20 and the space above the collection chamber. The passages, preferably a plurality of tubes, extending through the distribution chamber on the one hand provide an effective (upward) passage for the (gaseous) reactants and, in some embodiments, passage of the (liquid) product and on the
25 other hand enable a relatively straightforward construction of the bottom header and, if desired, the top header of the cooling module. Further, if the passages are evenly distributed, e.g. in rows or in a pattern having a square, rectangular or triangular pitch,
30 over the cross-section of the distribution chamber, the bottom header contributes to an even distribution of gaseous reactants entering the module. In another aspect, at least one of the distribution chamber and the collection chamber comprises two at least substantially
35 parallel plates interconnected by means of the passage

tubes. As a result of this structural connection, the passage tubes add to the mechanical strength of the header and bear part of the internal and external pressure load, exerted by the (evaporating) cooling medium and reactants and product respectively, as well as the structural load exerted on the bottom header by the mass of the module itself.

In one embodiment of the reactor of the present invention, a structured catalyst is placed between the cooling tubes of the cooling modules, such as shaped porous structures e.g. woven or non-woven and optionally compressed metal fabrics, e.g. in the form of sheets or contained in a cage. This configuration combines the advantage of a fixed bed reactor in that substantially no filtering of catalyst particles is required and the advantage of a slurry reactor, i.e. relatively high transfer of heat from the product to the coolant.

In another aspect, the cooling tubes are enveloped by one or more walls to contain reactants and product within the module, thus compartmentalizing the reactor in the radial direction and preferably at least up to the level of the structured catalyst (catalyst bed) in the reactor. Compartmentalizing the reactor facilitates scaling up in that a larger reactor can be obtained by using more of the same compartments (multiplication) having predictable hydrodynamic behavior. Thus, large scale hydrodynamics can be avoided and the risks of scaling up are reduced.

In one aspect, the reactor comprises several cooling modules, at least one cooling module being enveloped by one or more walls. Two walls may be connected to each other. Alternatively, there may be a space between two adjacent walls along the side of the walls which is substantially parallel to the length of the cooling tubes. The length of the walls may, for example, extend

along the cooling tubes from the distribution chamber up to the collection chamber of the cooling module.

Alternatively, the walls may, for example, extend along the cooling tubes from the top of the distribution chamber up to about 50 to 70% of the length of the cooling tubes. The distance between two opposite substantially parallel walls preferably is in the range of from 0.5 m to 10 m, more preferably in the range of from 0.5 m to 6 m, even more preferably in the range of from 0.5 m to 3 m. A wall preferably has a thickness in the range of from 0.5 mm to 12 mm, more preferably in the range of from 2 to 10 mm. The width of a wall preferably is in the range of from 5 cm to 15 m, more preferably in the range of from 1 m to 9 m.

In yet another aspect, the reactor comprises one or more perforated baffles, preferably at regular intervals along the length of the cooling tubes. The flow of gas and liquid can be influenced by selecting a suitable pattern for and dimensions of the perforations. I.e., the baffles can be used as redistributors for the gas and liquid inside the modules. Further, the baffles can provide support for any catalyst system that might be installed between cooling tubes and add mechanical strength to the module, e.g. by preventing tube buckling and module twisting. Baffles are preferably placed substantially horizontal.

The shape, size and configuration of the cooling modules and their arrangement within a reactor are governed primarily by factors such as the capacity, operating conditions and cooling requirements of the reactor. The cooling modules may have any cross-section which provides for efficient packing of cooling modules within a reactor, for example, the cooling module may be of square, triangular, rectangular, trapezoidal (especially covering three equilateral triangles) or

hexagonal cross-section. A cooling module having a square cross-section is advantageous in terms of lateral movement of the modules within the reactor during installation and removal and in providing uniform cooling throughout the reactor volume.

The cross-sectional area of the cooling modules may typically be about 0.1 to 5.00 m², preferably about 0.16 to 2.00 m², depending on the number and configuration of cooling tubes employed and the cooling capacity required.

The cooling tubes preferably have a length of about 4 to about 40 metres, more preferably a length of about 10 to about 25 metres. A cooling tube may have any cross section, for example, square or circular, preferably circular. Further, the outer diameter of each of the cooling tubes is preferably in a range from about 1 to about 10 cm, more preferably in a range from about 2 to about 5 cm.

A reactor according to the invention comprises a plurality of cooling modules which are typically placed in parallel.

In one aspect, the reactor comprises a grid or set of beams for supporting the modules near the bottom of the reactor and optionally one or more further grids or sets of beams for guiding the modules during installation in and removal from the reactor.

At least some of the cooling modules in a reactor according to the invention comprise a skirt, e.g. attached to or as an integral part of the beams or grid or directly to the corresponding modules or attached to the walls, for guiding feed gas underneath the modules. Reactant gas is guided underneath the modules, flows upwards, and enters the space between the cooling tubes. In some embodiments the gas may pass through passages in the distribution chamber of a cooling module before it enters the space between the cooling tubes. In some

embodiments the gas passes alongside the distribution chamber of a cooling module before it enters the space between the cooling tubes. Reactant gas might follow a preferred path (bypass) instead of being evenly distributed over the cooling modules. By guiding reactant gas underneath the modules, bypass of gas can be reduced or avoided.

A skirt preferably has a thickness in the range of from 0.5 mm to 12 mm, more preferably in the range of from 2 to 10 mm. A skirt preferably extends downwards from the module with a length in the range of from 10 cm to 5 m, more preferably 10 cm to 2 m, even more preferably in the range of from 50 cm to 1 m. The width of the skirt, horizontally along a side of the cooling module, preferably is in the range of from 5 cm to 15 m, more preferably in the range of from 1 m to 9 m.

The reactants inlet of the reactor may be connected to a gas distribution system with several gas outlets. A gas distribution system may, for example, consist of pipes with orifices, nozzles and/or spargers.

The gas outlets of a gas distribution system are preferably directed towards the cooling modules, so that the gas flow is guided and is evenly distributed over the cooling modules. The gas outlets of a gas distribution system are preferably directed to the cavities under the cooling modules that are defined by the skirts.

The cooling modules, the skirts, and the optional walls, baffles, and gas distribution system in a reactor according to the invention preferably are able to withstand the conditions of an exothermic reaction. More preferably, they are able to withstand Fischer Tropsch reaction conditions. A cooling module, wall, baffle, and/or skirt can be made of any material, and preferably is made of sheet metal, titanium, carbon steel, graphite, stainless steel, alumina, and/or carbon fibre reinforce

steel. A cooling module, wall, baffle, and/or skirt is most preferably steel, especially carbon steel or stainless steel.

The reactor preferably comprises between 1 and 100 cooling modules, more preferably between 2 and 100 cooling modules, even more preferably between 12 and 65, most preferably between 24 and 50.

The invention will now be explained in more detail with reference to the drawings, which show an example of a reactor according to the invention.

Figure 1 is a vertical cross-section of a reactor according to the present invention.

Figures 2 and 3 are lateral cross-sections, at II and at III respectively, of the reactor shown in Figure 1. Figure 2 shows a gas distribution system. Figure 3 shows coolant inlet piping.

Figures 4A and 4B are perspective views of a cooling module used in the reactor shown in Figure 1.

Figure 5 is a top view of the distribution chamber shown in Figures 4A and 4B.

Figure 6 is a top view of a perforated baffle.

Figures 7 and 8 show two different embodiments of a gas supply for the cooling modules in the reactor.

Figures 1 to 3 show a reactor 1 for carrying out an exothermic process, such as a Fischer-Tropsch process, comprising a reactor shell 2, at least one reactant inlet 3, at least one product outlet (not shown), at least one top outlet and liquid-gas separator (not shown), a cooling system 5 comprising a plurality of cooling modules 6, and inlets 7 and outlets 8 for a coolant. The reactor 1 further comprises skirts 30 for guiding reactant gas from the reactant inlet and gas distribution system to the cooling modules 6 inside the reactor 1.

The upper part of the reactor 1 comprises a flanged dome 10 having an inner diameter equal to that of the main cylindrical section of the reactor 1, which dome 10 provides access to the interior of the reactor 1 and enables the cooling modules 6 to be installed in and removed from the reactor 1.

Figures 4A to 8 show a cooling module 6 having a square cross-section and comprising, from bottom to top, a coolant distribution chamber 15, an array of cooling tubes 16, and a coolant collection chamber 17.

As shown in Figures 5 and 6, the cooling tubes 16 are arranged in rows separated by a distance sufficient to accommodate a structured catalyst, in particular shaped porous structures such as woven or non-woven and optionally compressed metal fabrics, e.g. in the form of blankets 24 (only three shown), between the rows of cooling tubes 16. Fischer-Tropsch catalysts are known in the art and typically include a Group VIII metal component, preferably cobalt, iron and/or ruthenium, more preferably cobalt. Suitable catalyst structures are disclosed in, e.g., WO 2006/037776 and WO 2007/068732.

In the embodiment shown in the Figures, the collection chamber 17 is identical to the distribution chamber 15. However, typically, the collection chamber will be different, e.g. may comprise an outlet having a larger diameter to take account of the increased volume of evaporated coolant.

The cooling tubes 16 are enveloped by walls 25 (omitted in Figures 4A to 6) extending from the level of the distribution chamber 15 to level of the collection chamber 17 to contain reactants and product within the module 6. In an alternative embodiment, the wall(s) terminate at a distance below the collection chamber, e.g. extend just up to the top level of the structured catalyst (catalyst bed) in the reactor.

Baffles 26 comprising, as shown in Figure 6, rows of relatively small perforations 27 are provided at regular intervals along the length of the cooling tubes 16 to redistribute the gas and product inside the modules 6 and to provide support for the structured catalyst 24.

The cooling modules 6A adjacent the reactor wall 2 have a different cross-section to maximize reactor volume utilization.

As shown in Figures 7 and 8, skirts 30 are present below each of the modules 6 for guiding gas. In the embodiment shown in Figure 7, pipes 31 run below and parallel to the skirts 30 and are provided with orifices 32 or nozzles directed towards the cavities defined by the skirts 30. In the alternative embodiment shown in Figure 8, an annular pipe 33 is provided around the inlet 22 of each of the modules 6.

During operation, coolant, typically water and/or steam, is fed through the inlet 7 to the distribution chamber of each of the modules 6. There, the coolant is distributed over the cooling tubes 16 and flows through the tubes 16 to the collection chamber 17 where it is collected and discharged via the outlet 8. Heat is transferred from the structured catalyst and the liquid surrounding the cooling tubes 16 to the coolant as it passes through the modules 6 and in particular as the coolant flows through the cooling tubes 16.

Syngas is fed through the inlet 3 to the pipes 31, and into the cavities defined by the skirts 30. Reactant gas is guided underneath the modules by skirts 30.

The modules can be installed by removing the dome and subsequently lowering the cooling modules into position in the reactor shell without the need for any personnel to be inside at the bottom of the reactor.

The invention is not limited to the embodiment described above, which can be varied in several ways

within the scope of the claims. For instance, the reactor can be provided with a sub-dome or manhole, having a diameter significantly smaller than that of the cylindrical section of the reactor. In that case,
5 internal lifting means (not shown) such as a temporary internal hoist fixed in the space above the cooling modules and below the ceiling of the reactor shell can be provided to facilitate lateral movement of the modules within the reactor shell, e.g. from the central-most
10 position to the designated positions and vice versa.

In a further example, the reactor according to the present invention can be used for other exothermic processes including hydrogenation, hydroformylation, alkanol synthesis, the preparation of aromatic urethanes
15 using carbon monoxide, Kölbel-Engelhard synthesis, and polyolefin synthesis.

C L A I M S

1. Reactor (1) for carrying out an exothermic process comprising a reactor shell (2), inlets (3, 7) for introducing reactants and coolant into the reactor shell (2), outlets (8) for removing product and coolant from the reactor shell (2), and a plurality of cooling modules (6), the reactor comprising for at least some of the modules (6) a skirt (30) for guiding gas underneath the modules (6).

2. Reactor (1) according to claim 1, wherein at least some of the skirts (30) are provided with an individual gas supply (31; 33).

3. Reactor (1) according to claim 2, wherein the gas supplies comprise pipes (31) running below and parallel to the skirts (30) and are provided with orifices (32) or nozzles directed towards the cavities defined by the skirts (30).

4. Reactor (1) according to any one of the preceding claims, wherein one or more of the cooling modules (6) comprises a coolant inlet (22), a coolant distribution chamber (15), a plurality of cooling tubes (16), a coolant collection chamber (17), and a coolant discharge.

5. Reactor (1) according to claim 4, wherein at least 80% of the cooling tubes (16) of the cooling modules (6) are arranged separately with a distance to the nearest cooling tube of at least 1 cm.

6. Reactor (1) according to claim 4 or 5, wherein one or more of the cooling modules (6) comprises one or more passages extending through the distribution chamber (15) to enable fluid communication between the space on one side of the distribution chamber (15) and the space between the cooling tubes (16), and wherein the cooling module comprises one or more passages extending through

the collection chamber (17) to enable fluid communication between the space between the cooling tubes (16) and the space above the collection chamber (17).

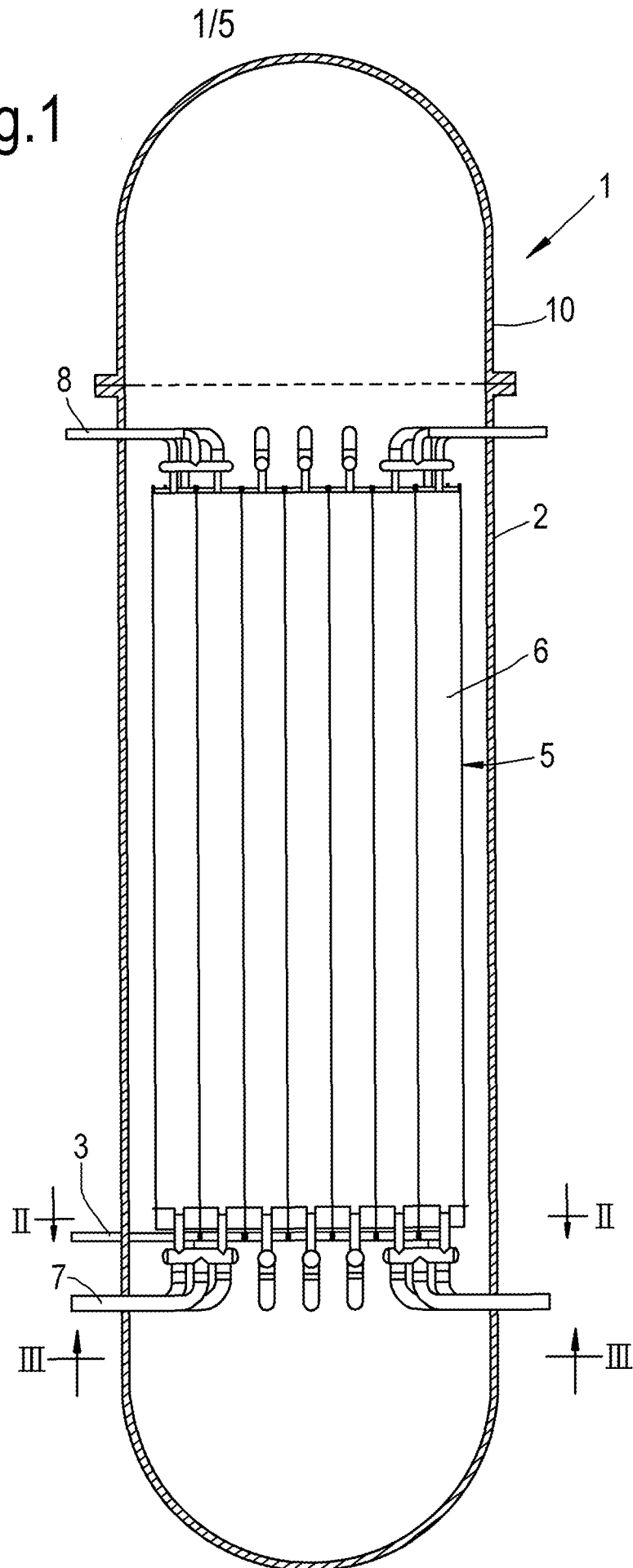
7. A reactor (1) according to any one of claims 4 to 6, wherein a structured catalyst (24) is placed between the cooling tubes (16).

8. A reactor (1) according to any one of claims 4 to 7, wherein the cooling tubes (16) are enveloped by one or more walls (25) to contain reactants and product within the module (6).

9. A reactor (1) according to any one of the preceding claims, wherein the cooling module (6) comprises one or more baffles along the height of the module (6), the baffles preferably comprising perforations to redistribute the reactants over the cross-section of the module (6).

10. Use of a reactor (1) according to any one of the preceding claims, for carrying out an exothermic process.

Fig.1



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Fig.2

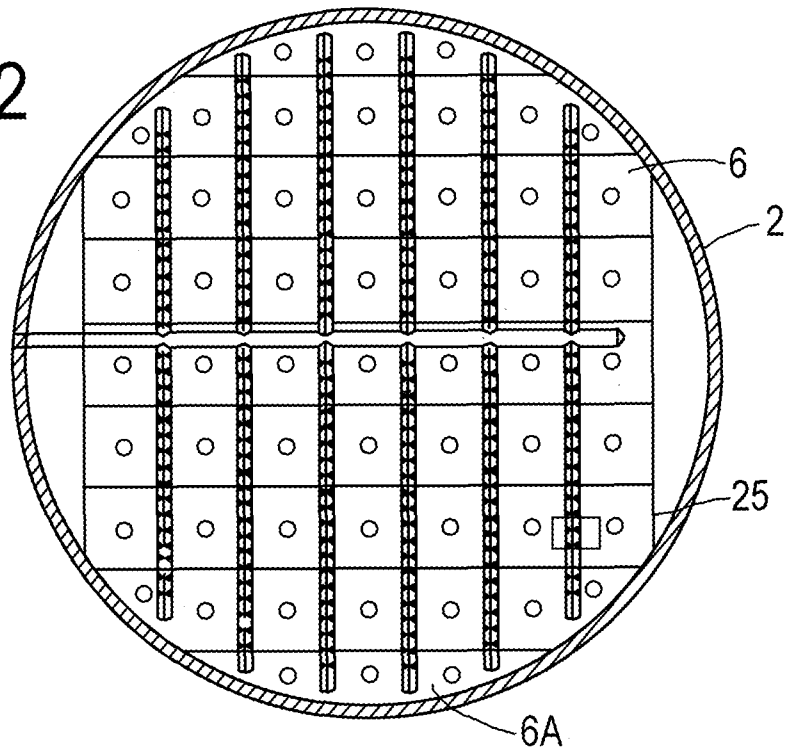
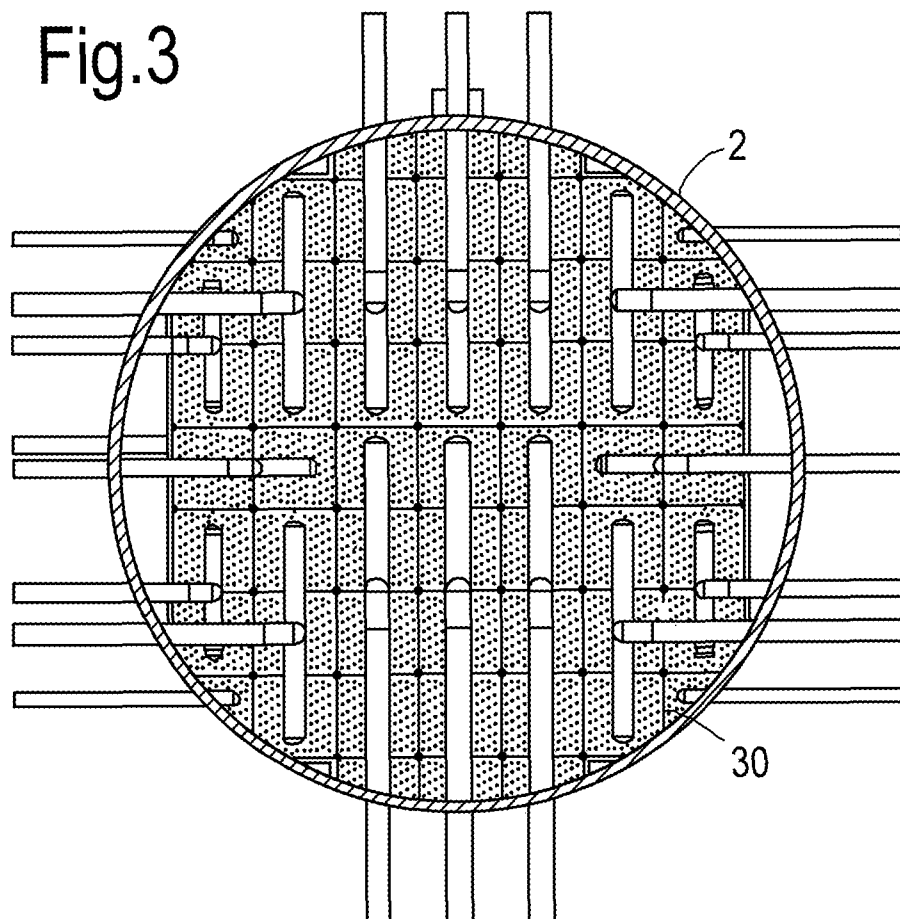


Fig.3



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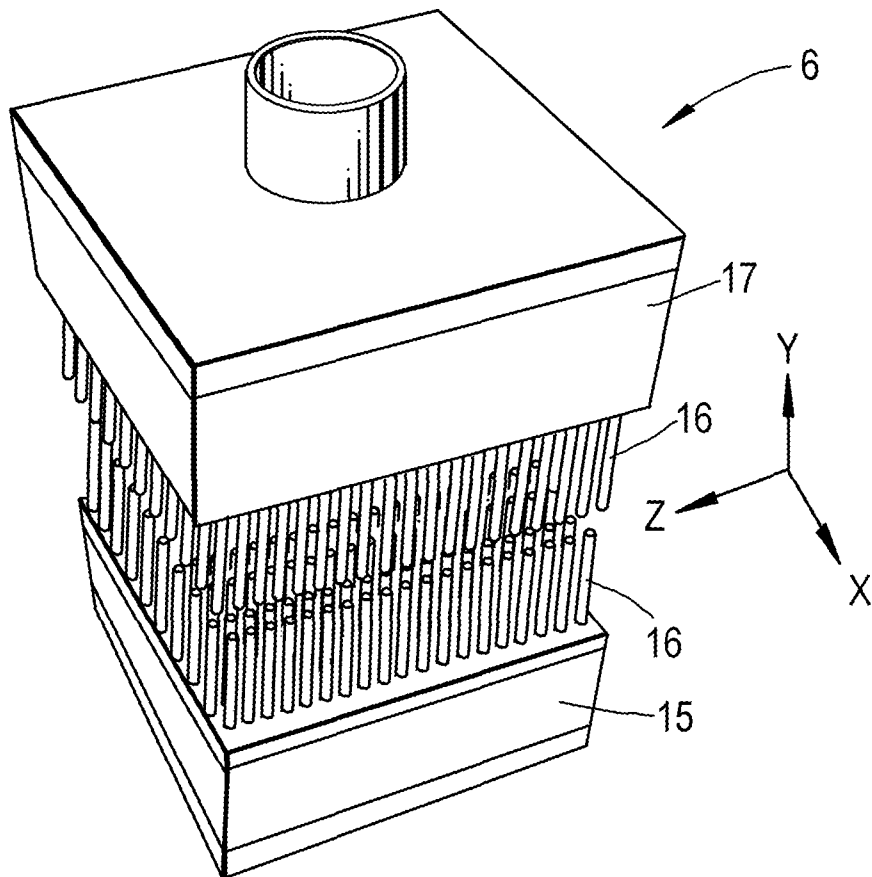


Fig.4A

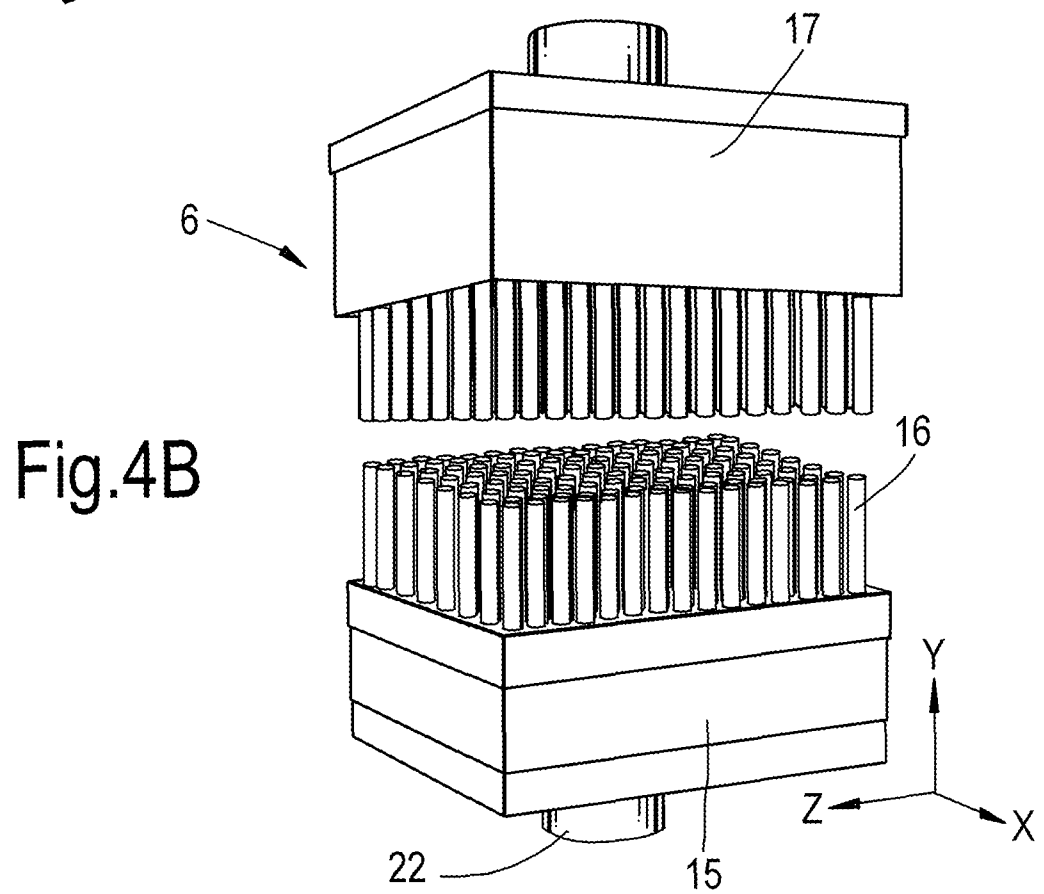


Fig.4B

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Fig.5

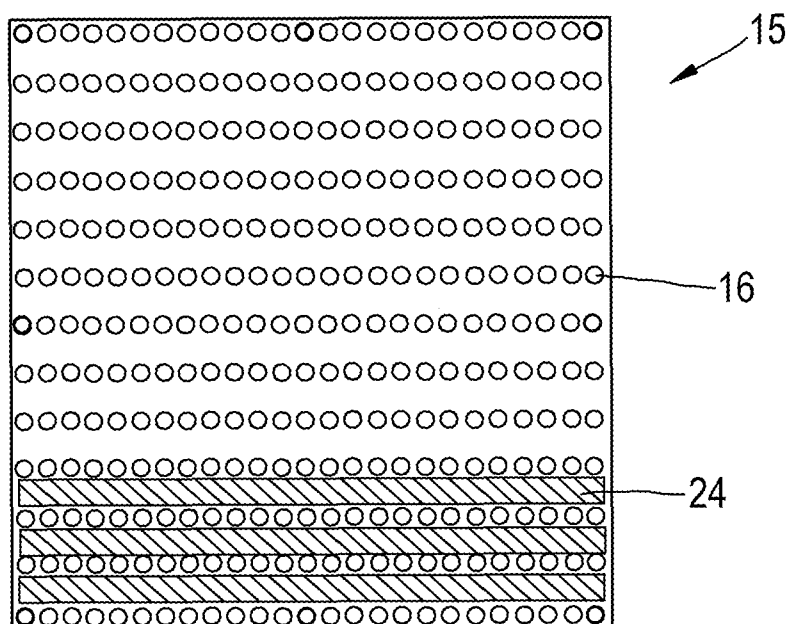


Fig.6

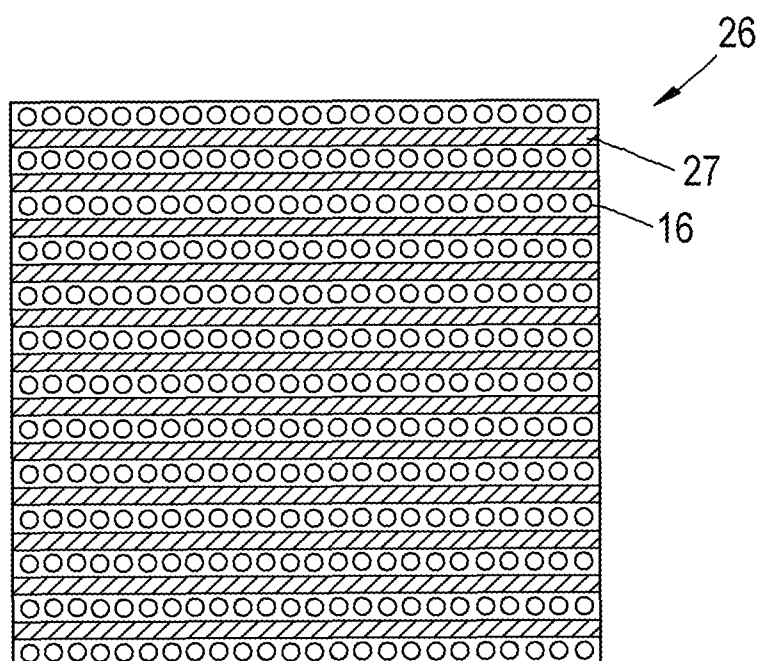


Fig.8

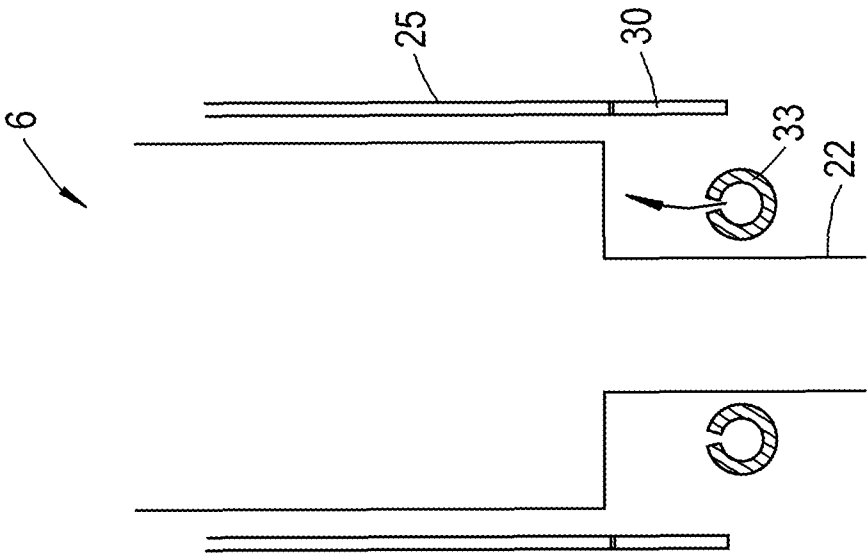
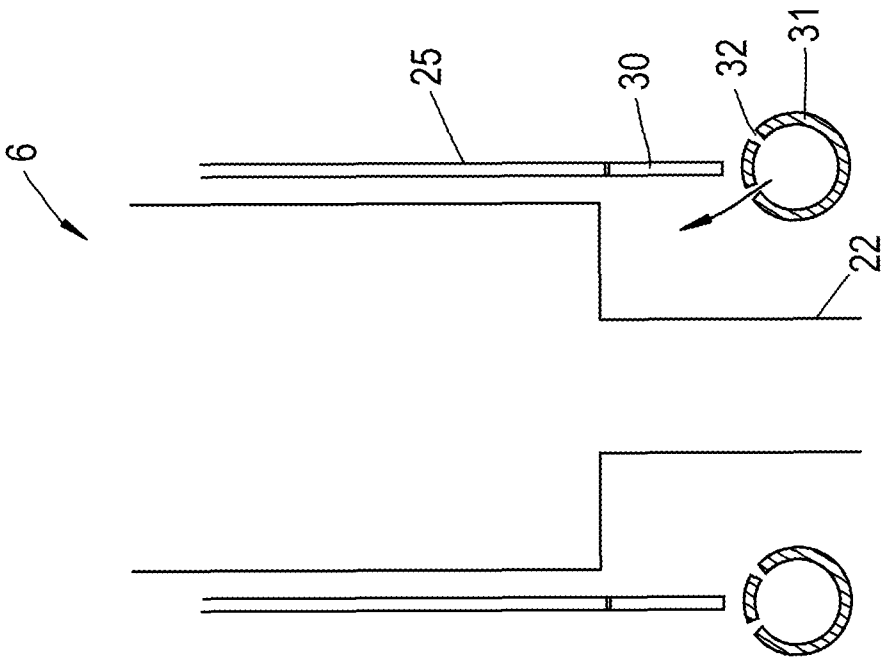


Fig.7



INTERNATIONAL SEARCH REPORT

International application No
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A. CLASSIFICATION OF SUBJECT MATTER INV. B01J8/02 B01J15/00 C10G2/00 F28D7/00 C07C1/04 ADD. B01J8/22 B01J12/00		
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) B01J C10G F28D C07C		
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		
C. 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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A	WO 2005/084790 A1 (SHELL INT RESEARCH [NL]; BOER ANNE [NL]; SCHRAUWEN FRANCISCUS JOHANNES) 15 September 2005 (2005-09-15) page 1, lines 1-8 page 11, line 31 - page 13	1, 4, 5, 10
A	FR 2 900 065 A1 (INST FRANCAIS DU PETROLE [FR]; ENI SPA [IT]) 26 October 2007 (2007-10-26) the whole document	1, 10
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☒ Further documents are listed in the continuation of Box C. ☒ See patent family annex.		
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Date of the actual completion of the international search 22 July 2010		Date of mailing of the international search report 30/07/2010
Name and mailing address of the ISA/ European Patent Office, P.B. 5818 Patentlaan 2 NL - 2280 HV Rijswijk Tel. (+31-70) 340-2040, Fax: (+31-70) 340-3016		Authorized officer Veronesi, Sergio

INTERNATIONAL SEARCH REPORT

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