

- (51) International Patent Classification:
B01J 19/08 (2006.01) *B01J 35/56* (2024.01)

(21) International Application Number:
PCT/EP2024/068225

(22) International Filing Date:
28 June 2024 (28.06.2024)

(25) Filing Language:
English

(26) Publication Language:
English

(30) Priority Data:
23182589.4 30 June 2023 (30.06.2023) EP

(71) Applicant: **TOPSOE A/S** [DK/DK]; Haldor Topsøes Allé 1, 2800 Kgs. Lyngby (DK).

(72) Inventors: **THOMSEN, Søren Gyde**; c/o Topsoe A/S, Haldor Topsøes Allé 1, 2800 Kgs. Lyngby (DK). **HANSEN, Anders Helbo**; c/o Topsoe A/S, Haldor Topsøes Allé 1, 2800 Kgs. Lyngby (DK). **BOE, Michael**; c/o Topsoe A/S, Haldor Topsøes Allé 1, 2800 Kgs. Lyngby (DK). **THOMSEN, Uffe Bach**; c/o Topsoe A/S, Haldor Topsøes Allé 1, 2800 Kgs. Lyngby (DK). **MORTENSEN, Peter Mølgaard**; c/o Topsoe A/S, Haldor Topsøes Allé 1, 2800 Kgs. Lyngby (DK).

(81) Designated States (unless otherwise indicated, for every kind of national protection available): AE, AG, AL, AM, AO, AT, AU, AZ, BA, BB, BG, BH, BN, BR, BW, BY, BZ, CA, CH, CL, CN, CO, CR, CU, CV, CZ, DE, DJ, DK, DM, DO, DZ, EC, EE, EG, ES, FI, GB, GD, GE, GH, GM, GT, HN, HR, HU, ID, IL, IN, IQ, IR, IS, IT, JM, JO, JP, KE, KG, KH, KN, KP, KR, KW, KZ, LA, LC, LK, LR, LS, LU, LY, MA, MD, MG, MK, MN, MU, MW, MX, MY, MZ, NA, NG, NI, NO, NZ, OM, PA, PE, PG, PH, PL, PT, QA, RO, RS, RU, RW, SA, SC, SD, SE, SG, SK, SL, ST, SV, SY, TH, TJ, TM, TN, TR, TT, TZ, UA, UG, US, UZ, VC, VN, WS, ZA, ZM, ZW.

(84) Designated States (unless otherwise indicated, for every kind of regional protection available): ARIPO (BW, CV, GH, GM, KE, LR, LS, MW, MZ, NA, RW, SC, SD, SL, ST, SZ, TZ, UG, ZM, ZW), Eurasian (AM, AZ, BY, KG, KZ, RU, TJ, TM), European (AL, AT, BE, BG, CH, CY, CZ, DE, DK, EE, ES, FI, FR, GB, GR, HR, HU, IE, IS, IT, LT, LU, LV, MC, ME, MK, MT, NL, NO, PL, PT, RO, RS, SE, SI, SK, SM, TR), OAPI (BF, BJ, CF, CG, CI, CM, GA, GN, GQ, GW, KM, ML, MR, NE, SN, TD, TG).

(54) Title: A CATALYST ASSEMBLY AND METHOD OF ASSEMBLING SAME

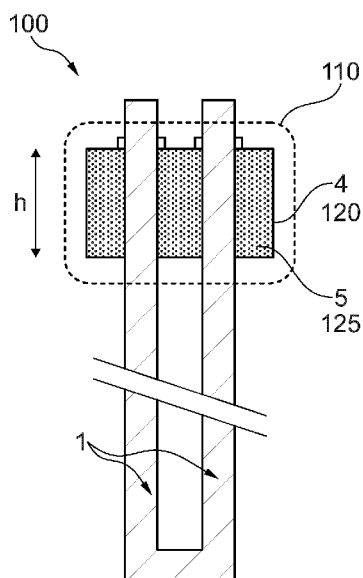


Fig. 2a

(57) Abstract: A catalyst assembly comprising two or more adjacent structured catalysts for catalysing an endothermic reaction of a feed gas being converted to a product gas, each structured catalyst comprising a macroscopic structure, the macroscopic structure comprising an electrically conductive material for resistance heating and supporting a catalytically active material, wherein: the structured catalysts each extend longitudinally from a first end forming an inlet for feed gas to a second end forming an outlet for product gas; and the assembly comprises a support structure that is electrically insulated from the electrically conductive material and provides a gaseous seal between the structured catalysts, to substantially prevent feed gas flow therebetween.

Declarations under Rule 4.17:

- *as to the identity of the inventor (Rule 4.17(i))*
- *as to applicant's entitlement to apply for and be granted a patent (Rule 4.17(ii))*
- *as to the applicant's entitlement to claim the priority of the earlier application (Rule 4.17(iii))*

Published:

- *with international search report (Art. 21(3))*
- *before the expiration of the time limit for amending the claims and to be republished in the event of receipt of amendments (Rule 48.2(h))*
- *in black and white; the international application as filed contained color or greyscale and is available for download from PATENTSCOPE*

A CATALYST ASSEMBLY AND METHOD OF ASSEMBLING SAMEField

5 The disclosure generally relates to a support structure for a catalyst assembly or a catalytic reactor, a method of assembling a catalyst comprising the support structure, and use of the catalyst assembly or catalytic reactor in an endothermic reaction, where heat for the endothermic reaction is provided by resistance heating.

10 Background

In electrically heated catalytic reactors for endothermic reactions such as those disclosed in WO2019228797, WO2021260108 and WO2023274939 (incorporated herein by reference), gas to be processed is fed to the structured catalysts, which provide heat to the endothermic chemical reaction (e.g. steam methane reforming or reverse water gas shift reactions) by resistance heating. Each structured catalyst comprises a macroscopic structure. The term "macroscopic structure" denotes a structure which is large enough to be visible with the naked eye, without magnification. The dimensions of the macroscopic structure are typically in the range of centimetres or meters. Each structured catalyst may comprise a single macroscopic structure or multiple macroscopic structures. Preferably, the macroscopic structure is extruded, fabricated from corrugated sheet metal or 3D printed, because the pressure drop from the inlet to the outlet may be reduced considerably compared to arrangements where the catalyst material is in the form of pellets or similar.

The macroscopic structure comprises an electrically conductive material for heating by resistance. The macroscopic structure also supports a catalytically active material, which may be on a ceramic coating such as in WO2023274939A1, and may be provided on at least part of the exposed surface area of the macroscopic structure. The surface area of the macroscopic structure itself, the fraction of the macroscopic structure coated with a ceramic coating, the type, features and structure (e.g. particle size, typically within the range of 2 nm – 1000 nm, thickness typically in the range of 10-500 µm) of the coating and the amount and composition of the catalytically active material may be suitably tailored to the reaction and operating conditions.

Synthesis gas production typically takes place in large chemical plants, due to the energy intensive reactions needed to facilitate the production. The toxicity of the synthesis gas (especially due to the content of carbon monoxide) poses a significant risk.

There is a need to improve the efficiency of such reactions, to minimise power consumption and losses of potentially toxic gases.

Brief summary of the invention

The present invention provides a catalyst assembly comprising two or more adjacent structured catalysts for catalysing an endothermic reaction of a feed gas being converted to a product gas, each structured catalyst comprising a macroscopic structure, the macroscopic structure comprising an electrically conductive material for resistance heating and supporting a catalytically active material, wherein: the structured catalysts each extend longitudinally from a first end forming an inlet for feed gas to a second end forming an outlet for product gas; and the assembly comprises a support structure that is electrically insulated from the electrically conductive material and provides a gaseous seal between the structured catalysts, to substantially prevent feed gas flow therebetween, and wherein the support structure is selected from the group consisting of

- a support structure comprising a container or enclosure containing a castable material between and/or surrounding the structured catalysts, and
- a support structure comprising a support member having one or more openings for receiving the structured catalysts, and a flexible sealing member disposed around and/or circumscribing the openings.

The present invention further provides a method of assembling a catalyst comprising two or more adjacent structured catalysts for catalysing an endothermic reaction of a feed gas being converted to a product gas, each structured catalyst comprising a macroscopic structure, the macroscopic structure comprising an electrically conductive material for heating by resistance and supporting a catalytically active material, the method comprising: positioning the structured catalysts in a support structure; and providing a gaseous seal between the structured catalysts, to substantially prevent feed gas flow therebetween.

The present invention further provides additional embodiments as claimed in the dependent claims.

The support structure provides a structural support for a catalyst assembly which may comprise several structured catalysts, which are preferably supported by the support structure substantially at their proximal ends, near their inlets. In some embodiments, two structured catalysts are placed vertically next to each other and are rigidly electrically connected at their distal ends to form a U-shaped pair. Several such U-shaped pairs may be connected in series. A number of support structures may be located in a vessel or reactor.

The support structure beneficially electrically insulates the (or sub-groups, e.g. U-shaped pairs of) structured catalysts from one another, allowing for high voltages such as up to 5000 V, 2000 V, 1000 V, or 500 V to be applied to the macroscopic structures, while maintaining a sufficiently high insulation resistance with respect to ground, such as above 0.3 Ohm/V, preferably above 0.5 Ohm/V. Said voltage can be supplied as either alternating current (AC) or direct current (DC).

Furthermore, the support structure provides a gaseous seal between the structured catalysts, at least partially filling at least some of any gaps therebetween, to minimise and substantially prevent feed gas (or more generally fluid) flow circumventing the macroscopic structures, so that none or only a minor fraction, preferably a very minor fraction (e.g. <10%, <5%, <1%, or <0.1%) of the feed fluid can bypass the structured catalysts.

Meanwhile, the support structure permits thermal expansion of the structured catalysts, permitting differences in thermal expansion along the length of the structured catalysts, which have a high operating temperature and have higher operating temperatures towards the outlets compared to the inlets. In particular, U-shaped pairs of structured catalysts may have different rates of expansion on their sides which can also be accommodated by the support structure.

Brief description of the figures

Embodiments of the disclosure will now be described, by way of example only, with reference to the accompanying drawings, in which:

Figure 1 is a schematic cross-section view of a first catalyst assembly;

Figure 2a is a schematic cross-section view of the catalyst assembly of figure 1 comprising a first support structure.

Figure 2b is a schematic cross-section view of a vessel comprising multiple catalyst assemblies according to figure 2a.

Figure 3a is a schematic cross-section view of the catalyst assembly of figure 1 comprising a second support structure.

Figure 3b is an enlarged schematic cross-section view of the second support structure of figure 3a.

Figure 4a is a schematic cross-section view of the catalyst assembly of figure 1 comprising a third support structure.

Figure 4b is an enlarged schematic cross-section view of the third support structure of figure 4a.

Figure 5 is a schematic perspective view of the catalyst assembly of figure 1 comprising a fourth support structure.

Figure 6 is a schematic perspective view of a filter arrangement for a catalyst assembly.

Detailed description of the disclosure

Aspects and features of certain examples and embodiments are described herein. Some aspects and features of certain examples and embodiments may be implemented conventionally and these are not described in detail in the interest of brevity. It will thus be appreciated that aspects and features of apparatuses and methods discussed herein which are not described in detail may be implemented in accordance with any suitable conventional techniques. Whilst various different examples are disclosed and discussed herein, any and all combinations of features in the different examples are explicitly contemplated within the scope of this disclosure.

Figure 1 is a schematic cross-section view of a first catalyst assembly 100 for catalysing an endothermic reaction of a feed gas being converted to a product gas.

The assembly 100 of figure 1 comprises a pair of vertically adjacent structured catalysts 1, each of the structured catalysts 1 extending longitudinally from a first (proximal) end forming an inlet for feed gas to a second (distal) end forming an outlet for product gas. Each structured catalyst 1 comprises a macroscopic structure, the macroscopic structure comprising an electrically conductive material (e.g. comprising Fe, Ni, Cu, Co, Cr, Al, Si or an alloy thereof, where such alloys may comprise other elements such as Mn, Y, Zr, C, Co, Mo or combinations thereof), for heating by resistance and supporting a catalytically active material. The macroscopic structures may be extruded, 3D printed and/or be monolithic structures. The macroscopic structures provide multiple channels for feed gas passing through the structured catalyst 1 from the inlet.

The macroscopic structures may generally, in some examples of the disclosure, comprise a circumferential wall encircling an internal space and/or may comprise one or more internal walls forming a plurality of flow channels from the first (inlet) end to the second (outlet) end. The macroscopic structures may be any shape, e.g. substantially circular or rectangular in a cross-section, perpendicular to the longitudinal direction.

The distal ends of the structured catalysts 1 are rigidly connected by a connector 2 to form a U-shaped arrangement. The structured catalysts 1 are also electrically connected together at their distal ends via the connector 2, forming a U-shaped arrangement providing an electrical current flow path through both the first and the second structured catalysts. A conductor or electrical circuit 3 connects the catalyst assembly to an external power supply, shown here at the proximal ends of the structured catalysts, but may be located elsewhere. In the U-shaped arrangement of figure 1, the electrical current flow path extends through the U-shaped pair of catalysts 1 in a U-shaped loop, from the first end of the first (left-hand) structured catalyst 1 through the first and the second structured catalysts, back to the first end of the second (right-hand) structured catalyst 1.

In large assemblies, multiple pairs of structured catalysts 1 may be connected together in series or in parallel. For example, the assembly comprises at least two pairs of structured catalysts comprising a first pair, a second pair and optionally one or more intermediate pairs, wherein:

the first structured catalyst of the first pair is electrically connected substantially at its first end to a conductor, and

the second structured catalyst of the first pair is electrically connected substantially at its first end to the first structured catalyst of the second pair or the intermediate pair,

the second structured catalyst of the intermediate pair, if present, is electrically connected substantially at its first end to the first structured catalyst of the second pair, and

the second structured catalyst of the second pair is electrically connected substantially at its first end to a conductor

Figures 2-5 illustrate support structures 110 in accordance with the disclosure, which are generally configured to prevent feed gas flow circumventing the inlets and may at least partially fill at least some of any (preferably substantially all) space(s) between adjacent structured catalysts 1 and/or any spaces between the structured catalysts 1 and walls of the vessel containing the structured catalysts 1.

Figure 2a is a schematic cross-section view of the catalyst assembly 100 of figure 1, comprising a first support structure 110 for a pair of structured catalysts 1.

As shown in figure 2a, the structured catalysts 1 are adjacent, but with some distal separation (gap) therebetween. The support structure 110 is electrically insulated from the electrically conductive material of the macroscopic structures (e.g. by part of the assembly 100 such as the structure 110 comprising an insulating material and/or part of the assembly 100, e.g. the structured catalysts 1 comprising an insulating layer) and the structure 110 provides a gaseous seal between the structured catalysts 1, to substantially prevent feed gas flow therebetween.

In figure 2a, the support structure 110 comprises a container in the form of an enclosure 120 containing a castable material 125. In figure 2a, the enclosure 120 fully encapsulates the space both between and around the structured catalysts 1, thus the enclosure 120 surrounds the structured catalysts 1. The enclosure 120 may be of any suitable cross-sectional shape, e.g. circular, rectangular (including square) or triangular.

The enclosure 120 itself may comprise a metallic or ceramic outer wall containing the castable material 125 and/or a metallic or ceramic inner wall around each structured catalyst 1. In some embodiments, the structured catalysts 1 may be wrapped with a metallic or ceramic lining (e.g. ceramic paper) during assembly, forming the inner wall, and the castable material is poured into the container 120 to set around the structured catalysts 1.

The castable material 125 itself may comprise a flowable material (e.g. comprising a ceramic, such as cement) and may be configured to flow during assembly (e.g. when heated to a suitable temperature to be self-flowing, to fill voids) and then subsequently dry/harden, forming a rigid structure which retains the structured catalysts 1, which thus then suspends them from the enclosure 120.

5

In figure 2a, a top surface of the enclosure 120 is located proximal (near) to, but downstream from, the inlets at the first (proximal) ends of the structured catalysts 1 and thus the enclosure 120 encapsulates the structured catalysts 1 proximal (near) to their respective inlets, extending downstream away from the inlets. This is beneficial because it minimises voids/gaps for inlet gas to bypass the structured catalysts 1. In some examples (not shown in figure 2a), the enclosure 120 is substantially flush with the inlets to minimise gaps and/or may substantially surround the structured catalysts 1 at their first ends / inlets to minimise gaps. In other examples, the support structure 110 is located elsewhere along the structured catalysts 1, e.g. distal from the inlets, including upstream or downstream of the inlets, more generally between the inlets and outlets, or proximal to the outlets at the second (distal) ends of the structured catalysts 1.

10

15

More generally, in some examples, the support structure 110 preferably provides the gaseous seal substantially at or proximal to the first ends / inlets of the structured catalysts 1, e.g. flush with or downstream from the first ends / inlets, substantially sealing within the upper 30%, 25%, 20%, 15%, 10% or 5% of the total length of the structured catalysts 1, since the first ends operate at lower temperatures than the second ends and hence the support structure 110 is then not exposed to such high temperatures, nor does it constrain thermal expansion of the catalysts 1 at such high temperatures, i.e. the structured catalysts 1 can expand freely axially (downwards) as they are heated. Moreover, such an arrangement located substantially at or proximal to the first (proximal) ends beneficially reduces possible void space for feed gas to circumvent the inlets. Furthermore, such an arrangement located substantially at or proximal to the first (proximal) ends prevents gas backflow from the spaces between the structured catalysts 1 as well as turbulence in the feed gas flow created by such backflow. Furthermore, minimising the height *h* of the enclosure 120 is beneficial to minimise weight of the enclosure 120 and to minimise restriction of axial expansion of the structured catalysts 1.

20

25

30

Figure 2b is a schematic cross-section view of a vessel 200 comprising an assembly 100 comprising multiple pairs of structured catalysts 1. A vessel or reactor may generally comprise one or more assemblies 100, which may each comprise e.g. 2-500, 10-100 or 20-50 structured catalysts 1. As shown in figure 2b, preferably, the support structure 110 extends and substantially seals between the structured catalysts 1 and one or more walls 205 of the vessel, so as to minimize feed gas circumventing the inlets, i.e. by travelling through any spaces between the catalysts 1 and the side walls 205 of the vessel 200. The support structure 110 may comprise a single container 120 having multiple openings for receiving the catalysts 1 (as shown), or multiple containers 120 between and/or surrounding the structured catalysts 1.

35

40

Figures 3a and 3b are schematic cross-sections view of the catalyst assembly 100 of figure 1 comprising a second support structure 110. Key differences to the arrangement of figures 2a and 2b are highlighted here.

5

In figures 3a and 3b, the support structure 110 comprises a horizontal support member 130 having an opening through which the structured catalysts 1 extend, and flexible sealing members 135 for sealing the opening.

- 10 The opening(s) may take any suitable form, such as comprising through-apertures and/or slots/recesses (e.g. recesses circumscribing through-apertures) and may generally, in some examples, be sized to provide an interference fit with the structured catalysts 1 in use, particularly both when not operating and when operating at the designed operating temperature(s) (e.g. up to 600, 800 or 1200 °C), so that the structure 110 supports the weight of the catalysts 1 and they are
- 15 securely suspended/hanging from the structure 110 and are able to undergo axial thermal expansion. The materials for the support structure 110, including any flexible sealing member(s) 135, can be chosen accordingly, noting that the flexible sealing member(s) 135 are electrically non-conducting (insulating), preferably with a resistivity higher than 0.001 (10e-4) Ωm , more preferably higher than 0.01, 0.1 or 1 (10e-3 to 10e-1) Ωm . Minimising the thickness t of the member 130 (e.g. $t \leq 20$ mm or $\leq$
- 20 10 mm) is beneficial to minimise weight and restriction of thermal expansion. Preferable materials include steels, including stainless steels with chemical resistance according to the intended use.

The flexible sealing member 135 may comprise any suitable material or combination of materials for assisting with sealing. In particular, the sealing member 135 may be compressible and/or be

25 ceramic- and/or silicone- based, which are particularly suited to the operating temperatures for this application. Particular examples include the sealing member 135 comprising a gasket, an o-ring, putty, rope and/or gland packing. In figure 3b, the sealing member 135 comprises a layered sandwich comprising inner (lower) and outer (upper) layers of gland packing 135a with ceramic sealing putty 135b in-between.

30

In figures 3a and 3b, the opening receives the U-shaped pair of structured catalysts 1 and the flexible sealing members 135a, 135b circumscribe the opening (i.e. are disposed around the inner periphery of the opening, lining the opening) to seal around the structured catalysts 1, thus the support member 130 does not have direct contact with the structured catalysts 1. The flexible sealing members 135a, 135b are also disposed between the structured catalysts 1. More generally, individual openings for each structured catalyst 1 may be provided (as shown in figures 4a and 4b), and/or the flexible sealing members 135 may be disposed around and/or circumscribing the or each opening (i.e. layered above/below the opening, also shown in figures 4a and 4b, and/or disposed around the inner periphery of the opening, lining the opening), to seal around and/or between the structured catalysts

40 1.

As shown in figure 3b, the support structure 110 may also comprise a fitting 133, such as a sleeve or lining, on/around the structured catalysts 1, where the flexible sealing member 135 seals between the support member 130 and the fitting 133 (rather than directly between the support member 130 and the structured catalysts 1). The fitting 133 may be thermally and/or electrically insulating, e.g. comprising a ceramic, preferably having a thermal conductivity of $< 20 \text{ W/(mK)}$, more preferably $< 1 \text{ W/(mK)}$ and/or preferably an electrical resistivity higher than $0.001 \text{ (} 10\text{e-}4 \text{)} \Omega\text{m}$, more preferably higher than 0.01 , 0.1 or $1 \text{ (} 10\text{e-}3 \text{ to } 10\text{e-}1 \text{)} \Omega\text{m}$. Accordingly, the fitting 133 may comprise an electrically insulating layer, electrically insulating the support structure 110 from the electrically conductive material, and/or a thermally insulating layer, thermally insulating the support structure 110 from the electrically conductive material which is heated by resistance. The fitting 133 may be secured to the structured catalyst 1 by any suitable means, such as an interference fit around the structured catalyst 1 or complementary threads on the fitting 133 and the structured catalyst 1.

As shown in figure 3b, the support member 130 may comprise a protrusion 131 forming a recess or slot 132 circumscribing one or more of the openings in the support member 130 for one or more of the structured catalysts 1, the recess or slot 132 being suitable for at least partially receiving and/or engaging one or more protrusions 134 of the fitting 133 and/or for at least partially receiving the flexible sealing member 135 (i.e. in combination with the fitting 133 or without the fitting 133).

In figure 3b, the fitting 133 comprises 3 protrusions 134, with two protrusions 134 on the left-hand side forming a (backwards) c-shaped recess for receiving the flexible member 135a on the left-hand side, and a single protrusion 134 on the right-hand side forming a lip for receiving the flexible member 135a on the right-hand side. In other examples, any number, any shape and any combination of recesses and protrusions may be provided on the support member 130, the catalyst 1 and the fitting 133.

Optionally, the support structure 110 may further comprise one or more fasteners 140 (not shown) configured to compress the sealing member 135 in use, to enhance the seal. Any suitable fastener 140 may be used, including clamps and threaded fasteners such as bolts and tightening rings. Additionally, the support structure 110 may further comprise bellows configured to permit thermal expansion of the structured catalysts 1 in one or more axes. An example illustrating fasteners and a bellows arrangement is discussed with reference to figures 4a and 4b.

Figures 4a and 4b are schematic cross-sections view of the catalyst assembly 100 of figure 1 comprising a third support structure 110. Key differences to the earlier arrangements are highlighted here. In particular, compared to the arrangement of figures 3a and 3b, the arrangement of figures 4a and 4b additionally comprises a fastener 140 for compressing the sealing members 135, a bellows arrangement for permitting axial expansion of the structured catalysts 1, and a feed gas funnel 152.

In figures 4a and 4b, the support structure 110 comprises a first, horizontal support member 130 having a pair of openings, each opening for receiving a second, vertical extension support member 150. The extension support members 150 of the support structure 110 each comprise an opening for receiving an individual structured catalyst 1. For the inlet end of the left structured catalyst 1, the extension support member 150 comprises bellows, which are shown in more detail in figure 4b, whilst for the inlet end of the right structured catalyst 1, the extension support member 150 does not comprise bellows.

The bellows permit axial thermal expansion of the structured catalysts 1. More generally, in some examples, each end of the U-shaped pair may be received with or without bellows - although only one extension support member 150 comprises bellows in figure 4b, other examples include the other or both extension support members 150 comprising the bellows, arrangements comprising multiple bellows and arrangements where the bellows receive multiple catalysts 1 e.g. wherein the extension support member 150 comprises multiple openings for receiving the structured catalysts 1.

In figure 4b, the support structure 110 further comprises a pair of flexible sealing members 135a, 135b layered above and below the opening respectively, sandwiched between an insulating fitting 133 which circumscribes the opening. The flexible sealing members 135a, 135b and the fitting 133 may have the same functionality as described above with reference to figures 3a and 3b.

In the arrangement of figure 4b, the first support member 130 secures around the second extension support members 150 which receive the structured catalysts 1. In figure 4b, the support structure 110 also comprises an optional fastener 140, configured to compress the sealing members 135, to enhance the seal.

More generally, in some examples, the bellows arrangement may comprise any suitable combination of the functionality of the fitting 133, the sealing members 135a, 135b and/or the fastener 140. For example, a combined sleeve fitting 133 and sealing member 135 may be provided in the form of a gasket or similar sealing member, e.g. having an I-shaped cross-section. In another example, the bellows arrangement may be in the form of a flanged connector having apertures for the fastener 140 and comprise a non-electrically conducting and/or thermally insulating flexible body (e.g. comprising nitrile rubber or mica), combining the sealing and insulating functionality of the fitting 133 and the sealing members 135a, 135b.

In embodiments, the support structure 110 may generally, in some examples, be shaped to direct feed gas flow to the inlets of the structured catalysts 1, e.g. by comprising one or more flow guide members such as deflectors or funnels. Optionally, as shown in figure 4b, the assembly may comprise an inlet tube 151 and/or a funnel 152 directing feed gas to the structured catalysts 1.

Figure 5 is a schematic perspective view of the catalyst assembly 100 comprising a fourth support structure 110, supporting a quadrant comprising two pairs of structured catalysts 1. The support structure of figure 5 is similar to that of figures 2a and 2b, comprising a cylindrical open container 120 containing a castable material which has solidified around the quadrant of catalysts 1, encapsulating them so that the container 120 completely surrounds the structured catalysts 1 proximal to their first ends / inlets. The catalyst assembly 100 of figure 5 is modular and thus removable/replaceable as a single unit - the container 120 of figure 5 comprises a flange 122 for removably securing the assembly 100 within a vessel (not shown), the vessel having walls with one or more apertures for receiving one or more assemblies 100, which can be secured by fasteners through the fastener openings in the flange 122.

Figure 5 also illustrates two conductors 3a for supplying power to the catalyst assembly 100, which may be a DC or AC supply. There are two U-shaped pairs of structured catalysts 1 shown in the assembly 100 figure 5, with the catalysts 1 in each pair being electrically connected together in the U-shaped arrangement by a connector 2. A first conductor 3a connects to a first catalyst 1 of the first pair; and a second conductor 3a connects to a catalyst 1 of the second pair, as shown. An n-shaped conductor 3b is provided for transferring the power supply between the two U-shaped pairs of catalysts 1, completing the circuit.

Figure 6 is a schematic perspective view of an optional filter arrangement for the inlets to the structured catalysts 1 in a catalyst assembly 100. The filter arrangement of figure 6 comprises a filter 160 and a distributor 170 (see inset) for each inlet. In the example of figure 6, the filter 160 comprises a fine mesh, e.g. of mesh size substantially in the range of 50-1000 μm , to remove contaminants such as dust from the feed gas or vessel. In figure 6, the filter 160 extends across the full cross-sectional area of each inlet and comprises a bulbous or hemispherical central portion, but any other shape and configuration may be used in other examples. The filter 160 is preferably removable, e.g. secured by fasteners.

The optional filter arrangement of figure 6 also comprises a distributor 170 across each inlet (see inset - shown separately in figure 6, since the distributor 170 lies under the filter 160), for distributing feed gas flow across the cross-section of the inlet. The distributor 170 beneficially assists in distributing feed gas flow over the full cross-section of the macrostructures. In figure 6, the distributor 170 comprises a plate with apertures uniformly spaced across its surface, to distribute feed gas accordingly. In other examples, the aperture distribution is configured to modify the flow distribution, e.g. direct flow towards and/or away from particular regions, e.g. by varying the aperture sizes and/or distribution across the plate. The aperture size for the distributor 170 is larger than the aperture size of the mesh filter 160.

Figure 6 also illustrates an optional connection sub-assembly which comprises a copper connector 32 having a fastener 165 in the form of bolts for a busbar connection, to provide power to the assembly 100.

5 Further preferred aspects of support structures 110 in accordance with the disclosure are now described.

In some embodiments, the support structure 110 has an electrical resistance of $> 1000 \text{ Ohm}$, $> 10 \text{ kOhm}$, $> 100 \text{ kOhm}$, or preferably $> 1 \text{ MOhm}$, e.g. as measured between one of the structured
10 catalysts and an external grounded periphery (such as an outer metal wall) of the catalyst assembly.

The support structure 110 is also preferably highly heat resistant, e.g. operable in temperatures of up to 600 , 800 or 1200°C , preferably operable at up to 800°C at or proximal to the first end (e.g. within the upper 30%, 25%, 20%, 15%, 10% or 5% of the total length) and up to 1200°C at or proximal to
15 the second end (e.g. within the lower 30%, 25%, 20%, 15%, 10% or 5% of the total length), since these are typical operating temperatures for catalysing an endothermic reaction of a feed gas being converted to a product gas. Particularly suitable materials for the support structure 110 include ceramics. Particular examples include castable refractory materials such as high alumina castables (comprising calcium aluminates), more specifically a self-flowing high alumina castables.

20 The support structure 110 may also have a coefficient of thermal expansion substantially equal to or smaller than a coefficient of thermal expansion of a circumferential (e.g. outer) wall of the structured catalysts 1, so that the contact surfaces of the support structure 110 and the catalysts 1 expand at similar rates, to minimize the risk of expansion fracture.

25 The catalyst assembly 100 may be implemented in a catalytic reactor for catalysing an endothermic reaction of a feed gas, comprising one or more such assemblies 100. The reactor may further comprise a shell housing the one or more assemblies 100, the shell comprising an inlet for receiving feed gas and an outlet for product gas. The reactor may also comprise a thermal insulation layer
30 between the structured catalysts 1 and the shell (the thermal insulation layer preferably having a thermal conductivity of $< 20 \text{ W/(mK)}$, more preferably $< 1 \text{ W/(mK)}$) and one (or more) connectors for electrically connecting the one or more assemblies 100 to an external power supply, which may be AC or DC, particularly a three-phase AC supply. The reactor may be used for any suitable reaction, but particularly may be used for one or more endothermic reactions such as steam methane
35 reforming, hydrogen cyanide formation, methanol cracking, ammonia cracking, reverse water gas shift and dehydrogenation.

A method of assembling a catalyst comprising two or more adjacent structured catalysts 1 for catalysing an endothermic reaction of a feed gas being converted to a product gas is also envisaged,
40 comprising positioning the structured catalysts 1 in a support structure 110 (in accordance with any

combination of the above disclosed features) and providing a gaseous seal between the structured catalysts 1, to substantially prevent feed gas flow between the structured catalysts 1. The assembly 100 may be heat-treated in a gaseous atmosphere comprising one or more of nitrogen, oxygen, hydrogen, argon and helium at a temperature of up to 600°C to cure any ceramic or castable material, e.g. when using an enclosure 120 comprising a castable material 125.

In the context of the disclosure, an interference fit should be understood as a fastening between two tight-fitting parts, providing a connection which is held together by friction.

The various embodiments described herein are presented only to assist in understanding and teaching the claimed features. These embodiments are provided as a representative sample of embodiments only, and are not exhaustive and/or exclusive. It is to be understood that advantages, embodiments, examples, functions, features, structures, and/or other aspects described herein are not to be considered limitations on the scope of the invention as defined by the claims or limitations on equivalents to the claims, and that other embodiments may be utilised and modifications may be made without departing from the scope of the claimed invention.

Various embodiments of the invention may suitably comprise, consist of, or consist essentially of, appropriate combinations of the disclosed elements, components, features, parts, steps, means, etc., other than those specifically described herein. In addition, this disclosure may include other inventions not presently claimed, but which may be claimed in future.

Protection may also be sought for any features disclosed in any one or more published documents referenced herein in combination with the present disclosure.

Index to reference numerals

1	structured catalyst
2	connector
3	conductor / electrical circuit
12	additional connector
100	catalyst assembly
110	support structure
120	enclosure
122	flange
125	castable material
130	first support member
131	support member protrusion
132	support member recess
133	fitting

134	fitting protrusion
135	sealing member
140	fastener
150	second (extension) support member
5 151	inlet tube
152	funnel
160	filter
165	busbar fastener
170	distributor

10

Particular features

1. A catalyst assembly comprising two or more adjacent structured catalysts for catalysing an endothermic reaction of a feed gas being converted to a product gas, each structured catalyst comprising a macroscopic structure, the macroscopic structure comprising an electrically conductive material for resistance heating and supporting a catalytically active material, wherein:

15

the structured catalysts each extend longitudinally from a first end forming an inlet for feed gas to a second end forming an outlet for product gas; and

the assembly comprises a support structure that is electrically insulated from the electrically conductive material and provides a gaseous seal between the structured catalysts, to substantially prevent feed gas flow therebetween, and wherein the support structure is selected from the group consisting of

20

- a support structure comprising a container or enclosure containing a castable material between and/or surrounding the structured catalysts, and
- a support structure comprising a support member having one or more openings for receiving the structured catalysts, and a flexible sealing member disposed around and/or circumscribing the openings.

25

2. The assembly of clause 1, wherein the support structure encapsulates the structured catalysts, preferably around their respective inlets.

30

3. The assembly of clause 1 or 2, wherein:

the container or enclosure comprises a metallic or ceramic outer wall containing the castable material; and/or

35

the container or enclosure comprises a metallic or ceramic inner wall around each structured catalyst; and/or

the castable material comprises a flowable material comprising a ceramic and is configured to subsequently harden.

40

4. The assembly of clause 1 or 2, wherein:
the sealing member is compressible; and/or
the sealing member comprises a ceramic- and/or silicone- based sealing member; and/or
the sealing member comprises one or more of: a gasket, an o-ring, putty, rope and/or gland
5 packing.

5. The assembly of clause 1, 2 or 4, wherein:
the support member comprises recesses circumscribing the openings for the structured
catalysts, the recesses being configured to receive the sealing member; and/or
10 the support structure comprises a fastener configured to compress the sealing member;
and/or
the support structure comprises bellows configured to permit thermal expansion of the
structured catalysts in one or more axes.

15 6. The assembly of any preceding clause, wherein the support structure:
is shaped to direct feed gas flow to the inlets; and/or
is configured to at least partially fill at least some of any space(s) between the adjacent
structured catalysts; and/or
provides the gaseous seal substantially at or proximal to the first ends of the structured
20 catalysts; and/or
has an electrical resistance of $> 1000 \text{ Ohm}$, $> 10 \text{ kOhm}$, $> 100 \text{ kOhm}$, or preferably > 1
 MOhm ; and/or
comprises an electrically insulating layer, electrically insulating the support structure from the
electrically conductive material; and/or
25 is heat resistant, operable in temperatures of up to 600 , 800 or 1200°C , preferably operable
at up to 800°C at or proximal to the first end and/or up to 1200°C at or proximal to the second end;
and/or
has a coefficient of thermal expansion substantially equal to or smaller than a coefficient of
thermal expansion of a circumferential wall of the structured catalysts; and/or
30 provides an interference fit, securing the structured catalysts.

7. The assembly of any preceding clause, wherein the macroscopic structures:
comprise a circumferential wall encircling an internal space; and/or
comprise one or more internal walls forming a plurality of flow channels from the first end to
35 the second end; and/or
are substantially circular or rectangular in a cross-section, perpendicular to the longitudinal
direction.

8. The assembly of any preceding clause, comprising one or more pairs of first and second
40 structured catalysts wherein the first and second structured catalysts in each pair are electrically

connected together substantially at or proximal to their second ends, to provide an electrical current flow path from the first end of the first structured catalyst through the first and the second structured catalysts, to the first end of the second structured catalyst.

5 9. The assembly of clause 8 comprising at least two pairs of structured catalysts, wherein:
the first structured catalyst of a first pair is electrically connected substantially at its first end to
a second structured catalyst of a second pair or to a conductor, and
the second structured catalyst of the first pair is electrically connected substantially at its first
end to a first structured catalyst of the second pair or to a conductor.

10 10. The assembly of any preceding clause, further comprising:
a filter across one or more of the inlets, for filtering feed gas; and/or
a distributor across one or more of the inlets, for distributing feed gas flow across the cross-
section of the inlets.

15 11. A vessel comprising one or more of the assemblies according to any preceding clause,
wherein the support structure extends and substantially seals between the structured catalysts and
one or more walls of the vessel.

20 12. A catalytic reactor for catalysing an endothermic reaction of a feed gas, comprising:
one or more assemblies or the vessel according to any preceding clause;
a shell housing the one or more assemblies, the shell comprising an inlet for receiving feed
gas and an outlet for product gas;
a thermal insulation layer between the structured catalysts and the shell; and
25 one or more connectors for electrically connecting the one or more assemblies to an external
power supply.

13. Use of the catalyst assembly, the vessel or the catalytic reactor of any preceding clause in an
endothermic reaction, the reaction optionally comprising one or more of: steam methane reforming,
30 hydrogen cyanide formation, methanol cracking, ammonia cracking, reverse water gas shift and
dehydrogenation.

14. A method of assembling a catalyst comprising two or more adjacent structured catalysts for
catalysing an endothermic reaction of a feed gas being converted to a product gas, each structured
35 catalyst comprising a macroscopic structure, the macroscopic structure comprising an electrically
conductive material for heating by resistance and supporting a catalytically active material, the
method comprising:

positioning the structured catalysts in a support structure; and
providing a gaseous seal between the structured catalysts, to substantially prevent feed gas
40 flow therebetween.

15. The method according to clause 14, where the assembly is heat treated in a gaseous atmosphere comprising one or more of nitrogen, oxygen, hydrogen, argon and helium at a temperature of up to 600°C.

5

CLAIMS

1. A catalyst assembly comprising two or more adjacent structured catalysts for catalysing an endothermic reaction of a feed gas being converted to a product gas, each structured catalyst
5 comprising a macroscopic structure, the macroscopic structure comprising an electrically conductive material for resistance heating and supporting a catalytically active material, wherein:

the structured catalysts each extend longitudinally from a first end forming an inlet for feed gas to a second end forming an outlet for product gas; and

the assembly comprises a support structure that is electrically insulated from the electrically
10 conductive material and provides a gaseous seal between the structured catalysts, to substantially prevent feed gas flow therebetween, and wherein the support structure is selected from the group consisting of

- a support structure comprising a container or enclosure containing a castable material between and/or surrounding the structured catalysts, and
- 15 • a support structure comprising a support member having one or more openings for receiving the structured catalysts, and a flexible sealing member disposed around and/or circumscribing the openings.

2. The assembly of claim 1, wherein the support structure encapsulates the structured catalysts,
20 preferably around their respective inlets.

3. The assembly of claim 1 or 2, wherein:

the container or enclosure comprises a metallic or ceramic outer wall containing the castable material; and/or

25 the container or enclosure comprises a metallic or ceramic inner wall around each structured catalyst; and/or

the castable material comprises a flowable material comprising a ceramic and is configured to subsequently harden.

30 4. The assembly of claim 1 or 2, wherein:

the sealing member is compressible; and/or

the sealing member comprises a ceramic- and/or silicone- based sealing member; and/or

the sealing member comprises one or more of: a gasket, an o-ring, putty, rope and/or gland
35 packing.

5. The assembly of claim 1, 2 or 4, wherein:

the support member comprises recesses circumscribing the openings for the structured catalysts, the recesses being configured to receive the sealing member; and/or

the support structure comprises a fastener configured to compress the sealing member;
40 and/or

the support structure comprises bellows configured to permit thermal expansion of the structured catalysts in one or more axes.

6. The assembly of any preceding claim, wherein the support structure:

is shaped to direct feed gas flow to the inlets; and/or

is configured to at least partially fill at least some of any space(s) between the adjacent structured catalysts; and/or

provides the gaseous seal substantially at or proximal to the first ends of the structured catalysts; and/or

has an electrical resistance of $> 1000 \text{ Ohm}$, $> 10 \text{ kOhm}$, $> 100 \text{ kOhm}$, or preferably $> 1 \text{ MOhm}$; and/or

comprises an electrically insulating layer, electrically insulating the support structure from the electrically conductive material; and/or

is heat resistant, operable in temperatures of up to 600 , 800 or $1200 \text{ }^{\circ}\text{C}$, preferably operable at up to 800°C at or proximal to the first end and/or up to 1200°C at or proximal to the second end; and/or

has a coefficient of thermal expansion substantially equal to or smaller than a coefficient of thermal expansion of a circumferential wall of the structured catalysts; and/or

provides an interference fit, securing the structured catalysts.

7. The assembly of any preceding claim, wherein the macroscopic structures:

comprise a circumferential wall encircling an internal space; and/or

comprise one or more internal walls forming a plurality of flow channels from the first end to the second end; and/or

are substantially circular or rectangular in a cross-section, perpendicular to the longitudinal direction.

8. The assembly of any preceding claim, comprising one or more pairs of first and second structured catalysts wherein the first and second structured catalysts in each pair are electrically

connected together substantially at or proximal to their second ends, to provide an electrical current flow path from the first end of the first structured catalyst through the first and the second structured catalysts, to the first end of the second structured catalyst.

9. The assembly of claim 8 comprising at least two pairs of structured catalysts comprising a first pair, a second pair and optionally one or more intermediate pairs, wherein:

the first structured catalyst of the first pair is electrically connected substantially at its first end to a conductor, and

the second structured catalyst of the first pair is electrically connected substantially at its first end to the first structured catalyst of the second pair or the intermediate pair,

the second structured catalyst of the intermediate pair, if present, is electrically connected substantially at its first end to the first structured catalyst of the second pair, and

the second structured catalyst of the second pair is electrically connected substantially at its first end to a conductor.

5

10. The assembly of any preceding claim, further comprising:

a filter across one or more of the inlets, for filtering feed gas; and/or

a distributor across one or more of the inlets, for distributing feed gas flow across the cross-section of the inlets.

10

11. A vessel comprising one or more of the assemblies according to any preceding claim, wherein the support structure extends and substantially seals between the structured catalysts and one or more walls of the vessel.

15

12. A catalytic reactor for catalysing an endothermic reaction of a feed gas, comprising:

one or more assemblies or the vessel according to any preceding claim;

a shell housing the one or more assemblies, the shell comprising an inlet for receiving feed gas and an outlet for product gas;

a thermal insulation layer between the structured catalysts and the shell; and

20

one or more connectors for electrically connecting the one or more assemblies to an external power supply.

13. Use of the catalyst assembly, the vessel or the catalytic reactor of any preceding claim in an endothermic reaction, the reaction optionally comprising one or more of: steam methane reforming, hydrogen cyanide formation, methanol cracking, ammonia cracking, reverse water gas shift and dehydrogenation.

25

14. A method of assembling a catalyst comprising two or more adjacent structured catalysts for catalysing an endothermic reaction of a feed gas being converted to a product gas, each structured catalyst comprising a macroscopic structure, the macroscopic structure comprising an electrically conductive material for heating by resistance and supporting a catalytically active material, the method comprising:

30

positioning the structured catalysts in a support structure; and

providing a gaseous seal between the structured catalysts, to substantially prevent feed gas flow therebetween,

35

optionally, heat treating the assembly in a gaseous atmosphere comprising one or more of nitrogen, oxygen, hydrogen, argon and helium at a temperature of up to 600°C.

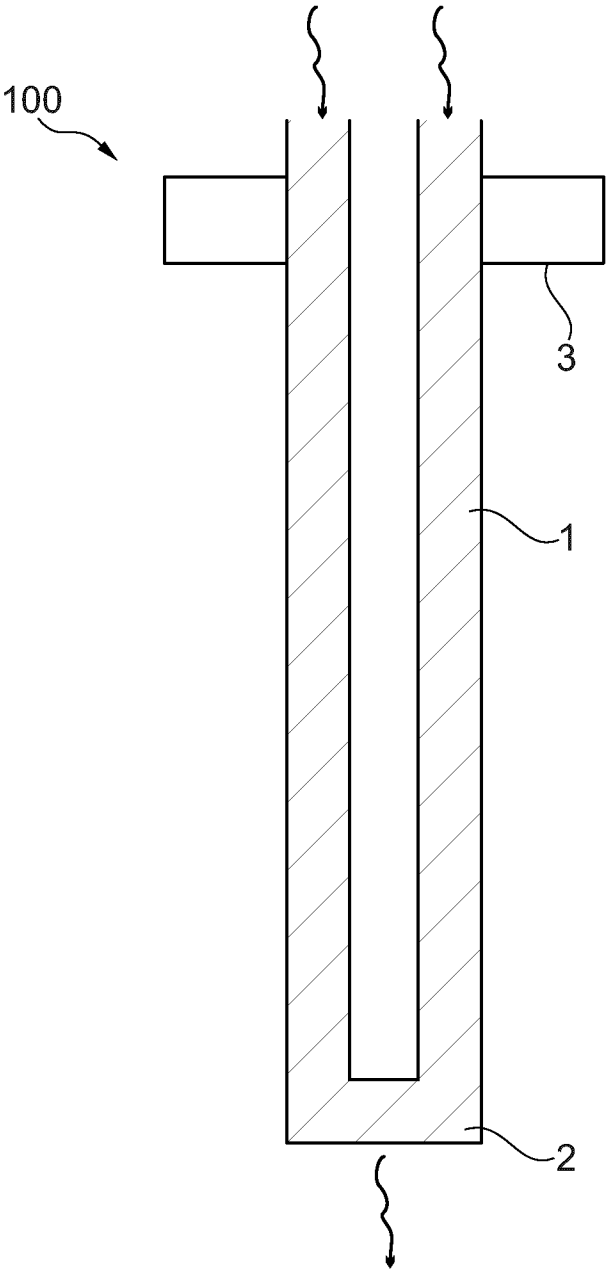


Fig. 1

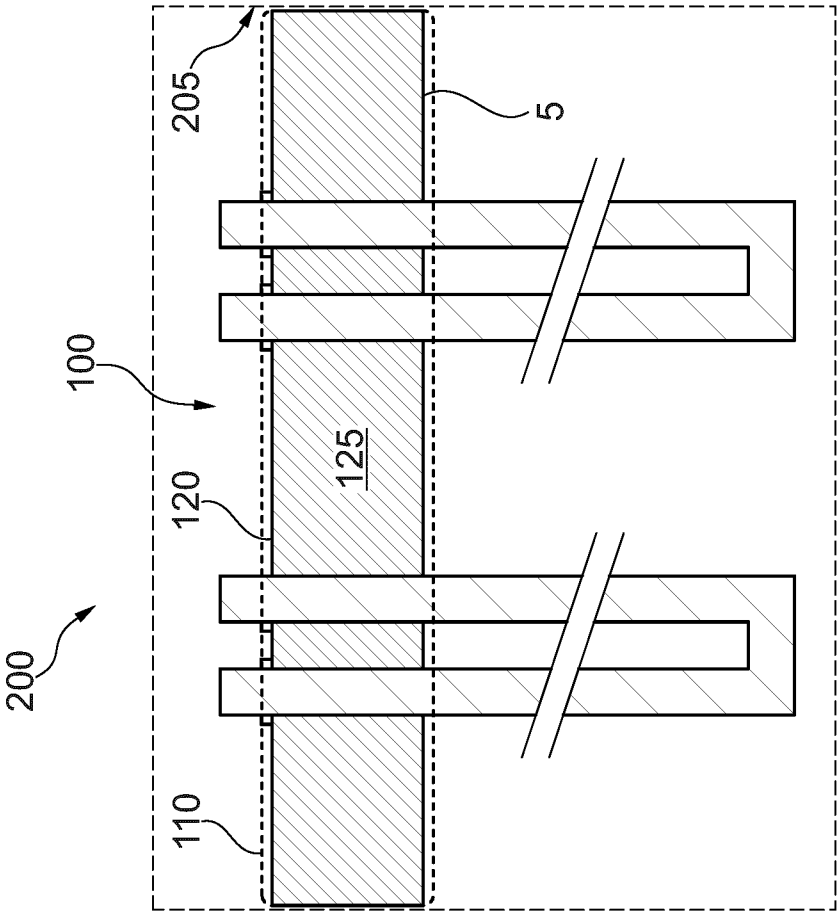


Fig. 2a

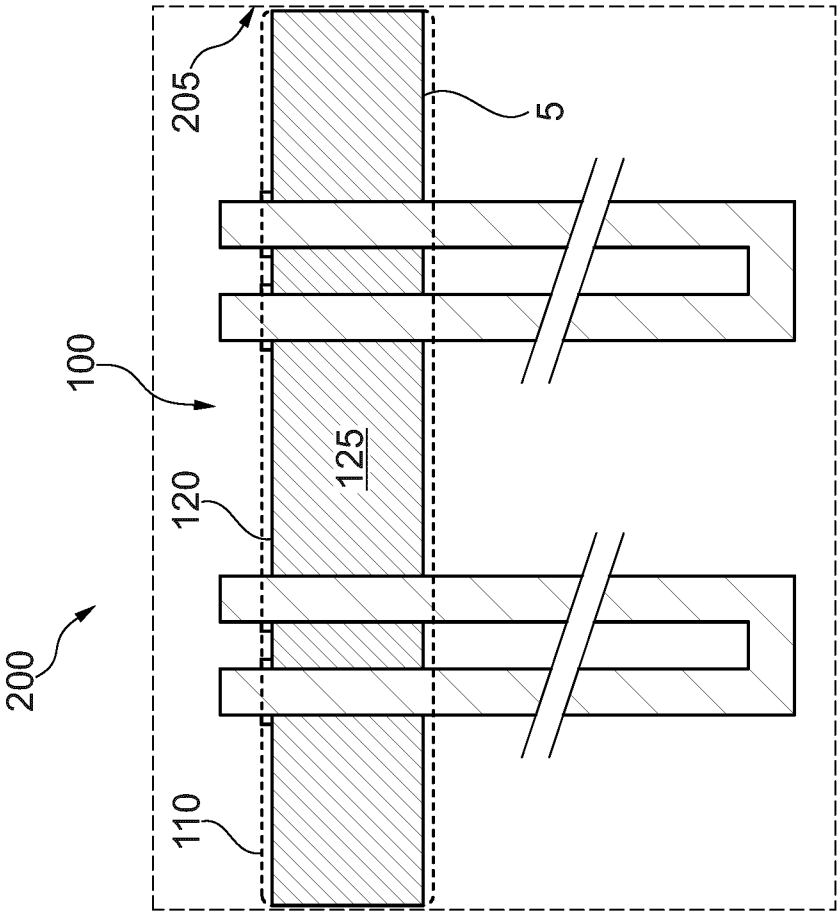


Fig. 2b

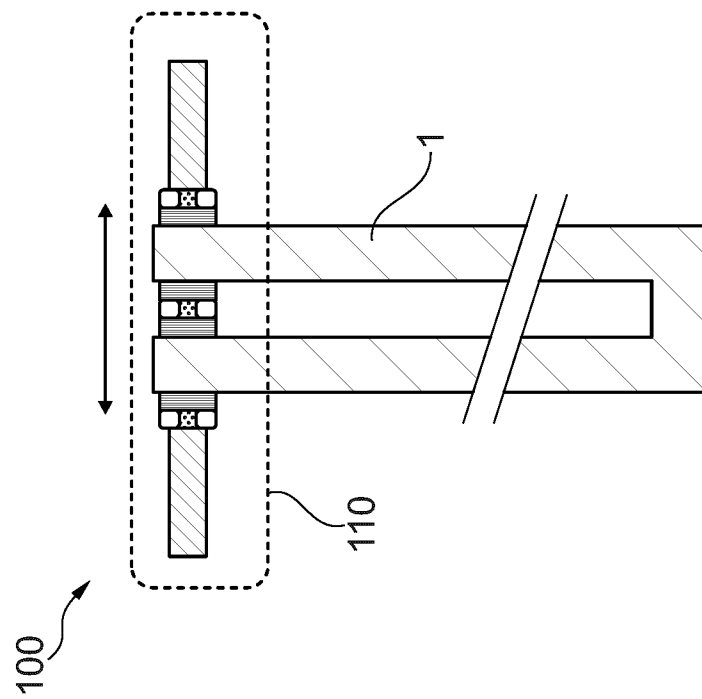


Fig. 3a

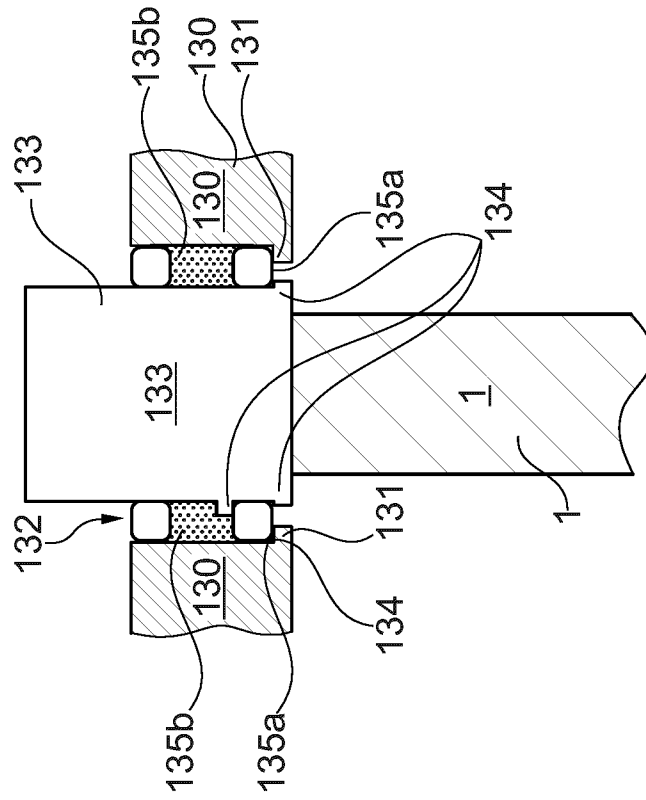


Fig. 3b

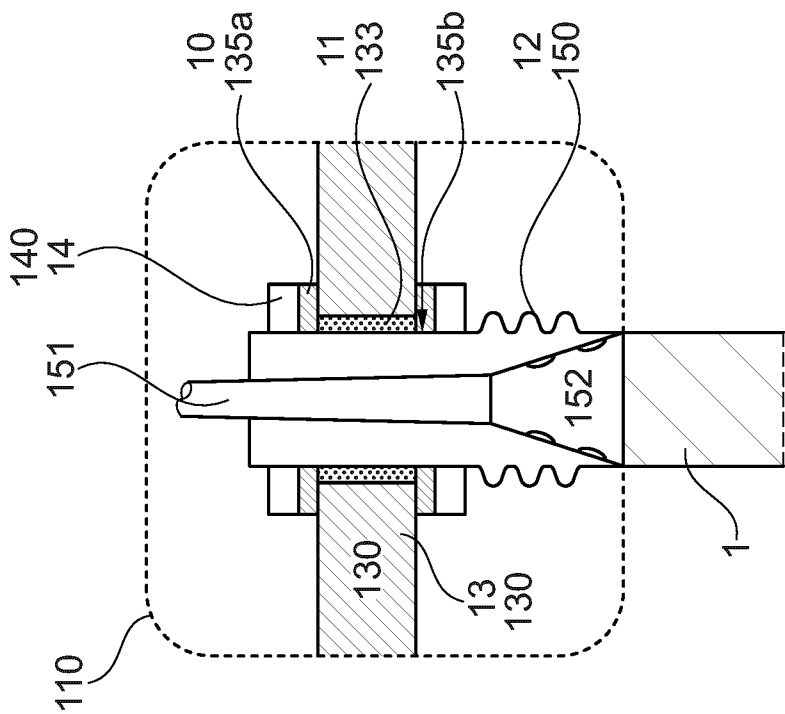


Fig. 4b

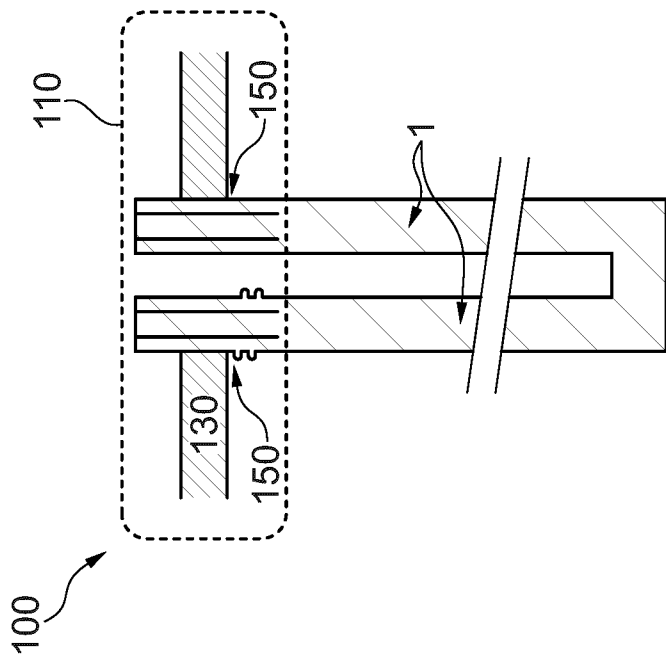


Fig. 4a

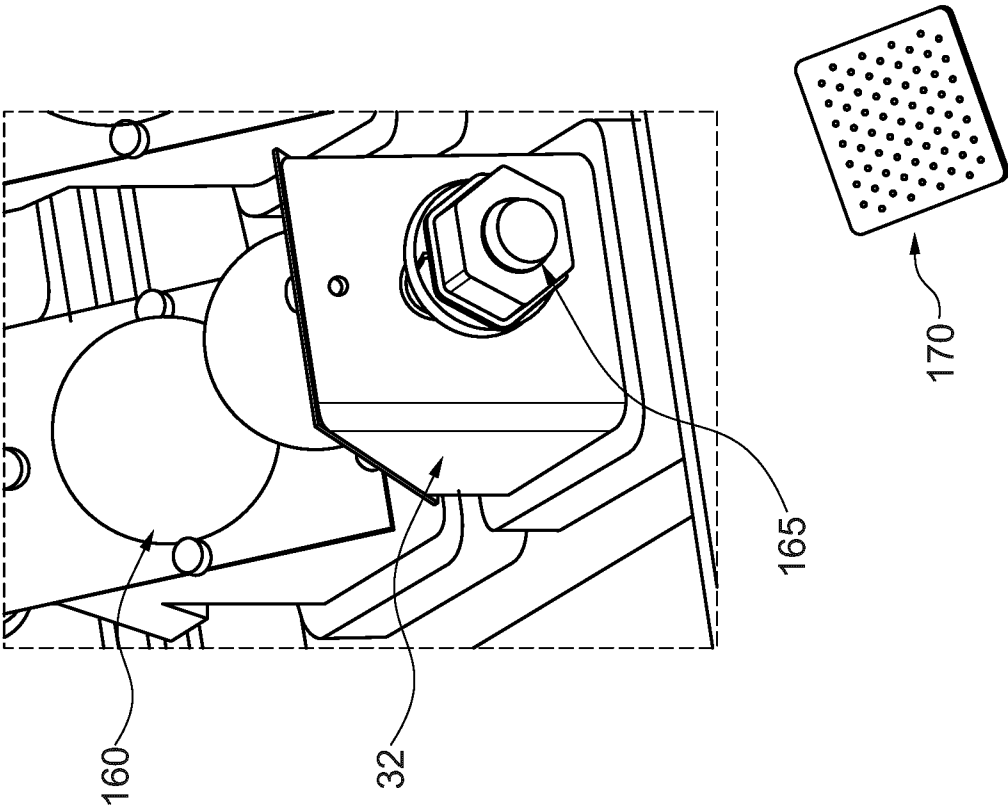


Fig. 5

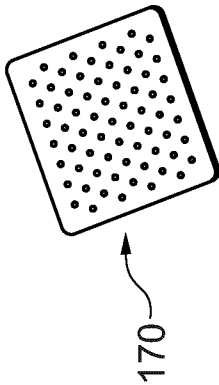


Fig. 6

INTERNATIONAL SEARCH REPORT

International application No

PCT/EP2024/068225

A. CLASSIFICATION OF SUBJECT MATTER

INV. B01J19/08 B01J35/56
ADD.

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

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 practicable, search terms used)

EPO-Internal**C. DOCUMENTS CONSIDERED TO BE RELEVANT**

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	US 2022/242727 A1 (MORTENSEN PETER MØLGAARD [DK] ET AL) 4 August 2022 (2022-08-04)	1 - 7, 11 - 14
Y	figures 2, 3a, 3b -----	9
X	US 2022/362736 A1 (MORTENSEN PETER MØLGAARD [DK] ET AL) 17 November 2022 (2022-11-17)	1 - 8, 10 - 14
Y	figures 2, 3a, 3b -----	9
X	US 2023/166247 A1 (KLEIN ROBERT [DK] ET AL) 1 June 2023 (2023-06-01)	1 - 7, 11 - 14
Y	figures 2, 3a, 3b -----	9
Y	US 2022/363537 A1 (MORTENSEN PETER MØLGAARD [DK] ET AL) 17 November 2022 (2022-11-17) paragraph [0023] -----	9



Further documents are listed in the continuation of Box C.



See patent family annex.

* 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

23 September 2024

Date of mailing of the international search report

25/10/2024

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

Pasanisi, Andrea

INTERNATIONAL SEARCH REPORT

Information on patent family members

International application No

PCT/EP2024/068225

Patent document cited in search report	Publication date	Patent family member(s)	Publication date
US 2022242727 A1	04-08-2022	AU 2020360477 A1	03-03-2022
		CA 3148729 A1	08-04-2021
		CN 114450083 A	06-05-2022
		EP 4038013 A1	10-08-2022
		JP 2022550813 A	05-12-2022
		KR 20220075347 A	08-06-2022
		US 2022242727 A1	04-08-2022
		WO 2021063795 A1	08-04-2021

US 2022362736 A1	17-11-2022	CN 114555219 A	27-05-2022
		DK 4037821 T3	04-09-2023
		EP 4037821 A1	10-08-2022
		ES 2955244 T3	29-11-2023
		KR 20220069071 A	26-05-2022
		US 2022362736 A1	17-11-2022
		WO 2021063793 A1	08-04-2021

US 2023166247 A1	01-06-2023	AU 2021295431 A1	24-11-2022
		CA 3185989 A1	30-12-2021
		CL 2022003533 A1	11-08-2023
		CN 115720533 A	28-02-2023
		EP 4171796 A1	03-05-2023
		FI 4171796 T3	20-06-2024
		KR 20230028255 A	28-02-2023
		PL 4171796 T3	05-08-2024
		US 2023166247 A1	01-06-2023
		WO 2021260108 A1	30-12-2021

US 2022363537 A1	17-11-2022	CA 3153229 A1	08-04-2021
		CN 114466694 A	10-05-2022
		EP 4038016 A1	10-08-2022
		KR 20220077135 A	08-06-2022
		US 2022363537 A1	17-11-2022
		WO 2021063792 A1	08-04-2021
