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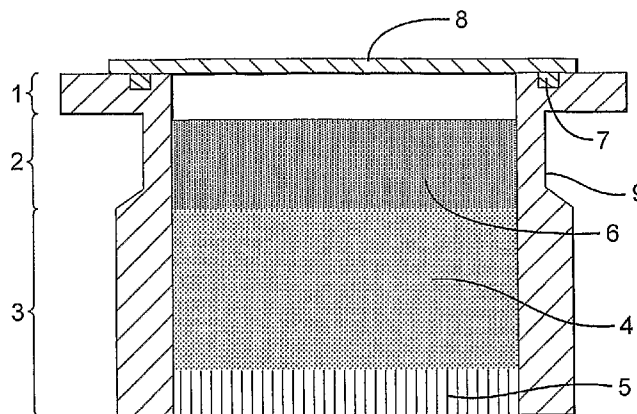
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(54) Title: CATALYST HOLDER, APPARATUS CONTAINING IT AND PROCESS FOR THE REACTION OF AN HYDRO-CARBON WITH OXYGEN



(57) Abstract: The present invention relates to a catalyst holder, and in particular to a catalyst holder for a catalyst for the reaction of a hydrocarbon with oxygen, wherein the catalyst holder comprises: (i) a first part which is adapted to provide a sealing engagement between the catalyst holder and a source of reactants, (ii) a second part, downstream of the first part as defined with respect to a flow of reactants, and (iii) a third part, downstream of the first and second parts as defined with respect to a flow of reactants and which is adapted to house the catalyst bed, the catalyst holder having a wall which defines the internal cross-sections of the first, second and third parts and extends from the first part to the third part, and wherein the wall of the second part has an average thickness narrower than the average wall thickness of the third part.

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CATALYST HOLDER, APPARATUS CONTAINING IT AND PROCESS FOR THE REACTION OF AN HYDROCARBON WITH OXYGEN

The present invention relates to a catalyst holder, and, in particular to a catalyst holder for a catalyst for the reaction of a hydrocarbon with oxygen.

Numerous processes are known in which a hydrocarbon is reacted with oxygen over a catalyst. One example of such a process is the catalytic partial oxidation of methane. Typical catalytic partial oxidation processes are described, for example, in WO 01/46068, WO 01/46069 and WO 02/88021.

A further example is the autothermal cracking of hydrocarbons, such as ethane, to produce olefins. Autothermal cracking is a route to olefins in which a hydrocarbon feed is mixed with oxygen and passed over an autothermal cracking catalyst. The autothermal cracking catalyst is capable of supporting combustion beyond the fuel rich limit of flammability. Combustion is initiated on the catalyst surface and the heat required to raise the reactants to the process temperature and to carry out the endothermic cracking process is generated in situ. The autothermal cracking of paraffinic hydrocarbons is described in, for example, EP 332289B; EP-529793B; EP-A-0709446 and WO 00/14035.

During the reactions of hydrocarbons with oxygen the hydrocarbons and oxygen need to be mixed then contacted with the catalyst. The mixed hydrocarbon and oxygen (reactants) stream is typically pre-heated prior to contacting the catalyst, either before or after mixing, or both, and is often at high pressures. The mixed reactants stream may be within flammability limits and hence it is desired to pass the mixed reactants into contact with the catalyst as quickly as possible. Thus, it is desirable to minimise the distance between the reactant mixing/pre-heat section and the catalyst.

On contact with the catalyst highly exothermic combustion occurs, causing the front face of the catalyst bed to reach temperatures significantly in excess of the pre-heated mixed reactants stream. This heat is conducted by the catalyst holder, and hence the catalyst holder and other parts of the reactor upstream of the catalyst bed can reach temperatures significantly higher than that of the upstream mixing/pre-heat section.

This can cause problems with the mixed reactants stream coming into contact with very high temperature surfaces upstream of the actual catalyst bed. Rapid changes in temperature between the catalyst holder and the rest of the reactor during start-up and shut-down of the reaction can also cause problems with differential expansion, causing potential reactor stress and sealing problems.

A catalyst holder has now been developed to overcome the above problems.

Thus, in a first aspect, the present invention provides a catalyst holder for a catalyst bed, wherein the catalyst holder comprises:

- 5 i) a first part which is adapted to provide a sealing engagement between the catalyst holder and a source of reactants,
 - ii) a second part, downstream of the first part as defined with respect to a flow of reactants, and
 - iii) a third part, downstream of the first and second parts as defined with respect to a flow of reactants and which is adapted to house a catalyst bed,
- 10 the catalyst holder having a wall which defines the internal cross-sections of the first, second and third parts and extends from the first part to the third part, wherein the average thickness of the wall in the second part is narrower than the average thickness of the wall in the third part.

In the process of the present invention, to mitigate the conduction of heat from the wall adjacent to the catalyst bed in the third part to the first part, a section of the wall of the catalyst holder above the third part and below the first part is provided which has an average thickness narrower than the average wall thickness of the third part.

As well as providing a generally cooler upstream part of the catalyst holder, by mitigating the conduction of heat to the first part of the catalyst holder, changes in temperature between the first part of the catalyst holder and the rest of the upstream reactor on reaction start-up and reactor shut-down are minimised. In the absence of the thinned second part according to the present invention, the whole of the catalyst holder will rapidly heat up on start-up, whereas the rest of the upstream reactor would heat up more slowly (due to its distance from the catalyst and its larger thermal mass) leading to differential expansion and potential stresses where the catalyst holder is connected to the reactor.

As a further advantage, as well as mitigating heat conduction, the thinner second part, due to it being less rigid than the third part, also provides flexibility that can accommodate differential expansion of the third part of the catalyst holder compared to the first part during start-up and shut-down of reaction.

30 The preferred average thickness of the second part will depend on the specific reaction for which the catalyst is being used, including such factors as the size of the catalyst holder (which itself depends on the size of the catalyst bed) and the catalyst

temperature during reaction. Generally, however, the average thickness of the wall in the second part will be less than 8mm, especially less than 5mm. Although thinner walls will mitigate heat conduction further still and also provide more flexibility, usually the minimum thickness of the wall in the second part is more than 1mm for structural
5 (integrity) reasons.

In comparison to the average thickness in the third part, the average thickness in the second part is usually less than 75% of this average, such as less than 50% of the average in the third part. The actual average wall thickness in the third part is not critical, but, generally, will be at least 10mm.

10 The source of reactants is typically an upstream reactor section in which the reactants are provided, and preferably mixed and pre-heated (hereinafter defined as a reactant supply section). The catalyst holder according to the process of the present invention is suitable for any reaction in which a large temperature differential exists between the catalyst bed and the reactant supply during reaction i.e. for highly exothermic
15 reactions. The reactants preferably comprise a hydrocarbon and oxygen.

By "sealing engagement" is meant that the first part is connected to the source of the reactants with a gas tight seal such that the reactants flow from the source into the catalyst holder.

20 The sealing engagement of the first part to the source of reactants may be by any suitable means, but is preferably provided by a suitable flange which may be bolted to a matching flange or flanges on the source of reactants.

The third part is adapted to house the catalyst bed. Typically, the third part defines a suitable volume into which the catalyst bed is placed, the catalyst bed being held within the third part by the presence of a support grid at its base (which generally also coincides
25 with the base of the catalyst holder). The catalyst bed may be in any suitable form. In one example, the catalyst bed may be in the form of a single catalyst structure, such as a foam or monolith. Alternatively, the catalyst bed may be in the form of a bed of catalyst particles.

In use, it is generally desired to ensure that gas flows through the catalyst bed,
30 entering only at the front (upstream) face and exiting only at the back (downstream) face. This may be a feature of the catalyst itself, for example straight channel monoliths. Where this is not the case, or even where it is but to prevent reactants bypassing the catalyst bed,

the edges of the catalyst bed may be wrapped in a suitable sealing material, such as binderless paper.

It is also preferred that the wall of the catalyst holder through the first, second and third parts is solid to ensure that all the gas entering the first part (upstream end) of the catalyst holder passes through the catalyst bed in the third part.

Preferably, the first, second and third parts of the catalyst holder together provide an internal volume of constant internal cross-section.

The catalyst holder may be made of any material capable of handling the temperatures of the hydrocarbon oxidation reaction. Preferred materials of construction are high temperature alloys, especially nickel containing heat resistant alloys. Particularly preferred materials are inconel, paralloy, e.g. H39WM, and Incoloy, e.g. 800HT.

Surfaces of the catalyst holder may be aluminized.

In a preferred embodiment of the present invention the first part of the catalyst holder is providing with cooling means such that the first part of the catalyst holder is maintained at a temperature at least 100°C below the front face temperature of the catalyst bed during reaction. Preferably the cooling means is such that the first part of the catalyst holder is maintained at a temperature at least 200°C, such as at least 300°C below the front face temperature of the catalyst bed (during reaction).

The temperature of the first part of the catalyst holder upstream of the catalyst is preferably maintained at a temperature independent of the temperature of the front face of the catalyst bed (rather than relative to it), for example is maintained at a temperature of below 300°C, preferably below 200°C. Most preferably, the first part of the catalyst holder is maintained at a temperature at or less than the temperature of the (pre-heated) reactants. This temperature is generally limited by flammability constraints, which will be composition dependent, and, thus, the most preferred maximum temperature will be dependent on the process in which the catalyst holder is to be used.

Preferably, the cooling means comprises channels through which a cooling fluid can flow during reaction to maintain the first part of the catalyst holder at the required temperature.

In one embodiment, the cooling channels through which the cooling fluid can flow may be located within the first part of the catalyst holder itself.

Alternatively, the cooling channels may be provided in a separate cooling plate, which plate is held in (thermal) contact with the first part of the catalyst holder to maintain this part of the catalyst holder at the required temperature. The use of a separate cooling plate is preferred due to the generally simpler engineering required. The (thermal) contact may be obtained simply by holding the cooling plate and the catalyst holder tightly together. Alternatively, a heat transfer compound, for example a silicone heat transfer compound, may be used to provide conductivity between the cooling plate and the catalyst holder.

Typically, the cooling in said channels is achieved by flowing a cooling fluid, such as water or oil, through the channels at a rate sufficient to remove the required amount of heat. A preferred cooling medium is water at a temperature in the range 70°C to 140°C.

By the use of a catalyst holder which is cooled, problems with the mixed reactants stream coming into contact with very high temperature surfaces of the catalyst holder upstream of the actual catalyst are further minimised.

In addition, by maintaining the first part of the catalyst holder upstream of the catalyst at lower temperature, changes in temperature between the first part of the catalyst holder and the rest of the reactor on reaction start-up and reactor shut-down are also further minimised.

In a further embodiment, the present invention provides an apparatus comprising, a reactant supply section, a reaction section, which is positioned downstream of the reactant supply section with respect to the flow of the reactants, and which comprises the catalyst holder as defined herein, and a product removal section, downstream of the catalyst holder with respect to the flow of the reactants.

More preferably, the present invention provides an apparatus for the reaction of a hydrocarbon with oxygen, which comprises:

- a) a reactant supply section, which comprises a hydrocarbon supply, an oxygen supply and a mixing section for mixing the hydrocarbon and oxygen,
- b) a reaction section, downstream of the reactant supply section with respect to the flow of the reactants, which comprises a catalyst holder as described herein, and
- c) a product removal section, downstream of the catalyst holder with respect to the flow of the reactants,

characterised in that the reactant supply section is connected in sealed engagement with the first part of the catalyst holder, and such that the third part of the catalyst holder resides within the upper section of the product removal section with a gap between the outer surface of the third part of the catalyst holder and the inner surface of the upper section of the product removal section, said gap being purged with a suitable purge gas.

The gap provides space for differential expansion of the third part of the catalyst holder compared to the product removal section.

Preferably the purge gas is an inert gas, such as nitrogen. The purge prevents products exiting the catalyst holder from flowing into and/or stagnating in the gap (where they would be exposed to the high temperature surfaces of the second part of the catalyst holder).

More preferably, to reduce the volume of purge gas required, the gap is filled with a flexible material, and at least the lower part of this material is purged to prevent products exiting the catalyst holder from flowing into or through the material.

The flexible material may, for example, be a suitable sponge or mesh material which can withstand the required temperatures and can be compressed as the catalyst holder expands on reaction start-up. A suitable material is "Interam" (trade mark) produced by 3M.

The reactant supply section may be configured in any suitable configuration in which the hydrocarbon and oxygen are supplied and mixed to obtain a mixed reactants stream which can be passed to the catalyst bed (catalyst holder). Preferably a pre-heated, mixed reactants stream is obtained, either by heating the hydrocarbon and oxygen prior to mixing or after mixing, or a combination of both.

One example of a suitable mixing system is described in WO 01/18451, which describes a tangential mixing device for mixing a gaseous stream comprising a fuel and a gaseous oxidant and to a process for the catalytic partial oxidation of a hydrocarbon fuel using the mixing device. Most preferably, the mixing and pre-heating section utilises first and second supply means for the respective reactants each comprising a plurality of outlets, as described in WO 2004/074222.

"Product removal section" as used herein refers to the part of the reactor downstream of the catalyst into which the product stream from the reaction on the catalyst subsequently flows. Various product treatments may be required in the product removal

section depending on the reaction being performed, such techniques generally being those known to the person skilled in the art for said processes.

For example, in the autothermal cracking of hydrocarbons to produce olefins, the product stream typically exits the reaction zone as a gaseous product stream at a
5 temperature greater than 800°C e.g. greater than 900°C and, especially when also at pressure, it is preferred that the product stream is rapidly cooled. This ensures a high olefinic yield because the product cooling step slows down the rate of reaction in the gaseous product stream thus preventing further reactions taking place.

Preferably the temperature of the product stream is reduced to 800°C preferably to
10 600°C within 60mS preferably 40mS and advantageously 20mS from exiting the reaction zone.

Advantageously the rapid cooling may be achieved by injecting a condensate into the gaseous product stream, preferably at multiple points, such that the vaporisation of the condensate cools the gaseous product stream.

15 The condensate may be a gas or a liquid. When the condensate is gas it is preferably an inert gas. Preferably the condensate is a liquid e.g. water.

Injecting the condensate at high pressure and high temperature ensures that a large proportion of the condensate instantaneously vaporizes at the reactor pressure and therefore provides a very rapid temperature drop in the gaseous product stream.
20 Consequently the condensate, such as water, is usually injected at a pressure higher than the pressure of the gaseous product stream, such as 100 barg and is usually injected at a temperature of between 100-400°C and preferably between 200-350°C e.g. 300°C.

In a further aspect the present invention also provides a process for the reaction of a hydrocarbon with oxygen, which process comprises passing a mixed gaseous reactant
25 stream comprising said hydrocarbon and oxygen into contact with a catalyst bed in a catalyst holder as described herein, for example in the apparatus as described.

Suitably a resistance zone is provided upstream of and in contact with the front face of the catalyst, as described in WO 2004/074222. The resistance zone is porous and ensures dispersion of the reactants as they pass through the zone, such that they leave the
30 resistance zone substantially uniformly distributed over the cross-sectional area of the resistance zone.

The resistance zone may be formed of a porous metal structure, but preferably the porous material is a non metal e.g. a ceramic material. Suitable ceramic materials include lithium aluminium silicate (LAS), alumina (α -Al₂O₃), yttria stabilised zirconia, alumina titanate, niascon, and calcium zirconyl phosphate. A preferred porous material is gamma
5 alumina. The porous material may be in the form of spheres, other granular shapes or ceramic foams.

The oxygen may be provided as any suitable molecular oxygen containing gas, such as molecular oxygen itself or air.

The hydrocarbon may be any suitable hydrocarbon depending on the process to be
10 operated.

In one embodiment, the process is a process for the production of synthesis gas by the catalytic partial oxidation of methane.

In a second embodiment, the process is a process for the production of olefins by the autothermal cracking of a hydrocarbon.

The autothermal cracking (ATC) process will now be described in more detail.
15

For use in an autothermal cracking process, when a cooling fluid is used the preferred cooling fluid is water at a temperature in the range 70°C to 140°C. At least the first part of the catalyst holder is thus maintained at a maximum of 140°C.

Suitable hydrocarbons for autothermal cracking typically have at least 2 carbon
20 atoms. For example, the hydrocarbon may be a gaseous hydrocarbon, such as ethane, propane or butane or a liquid hydrocarbon, such as a naphtha or an FT liquid.

Preferably, hydrogen is co-fed. Hydrogen co-feeds are advantageous because, in the presence of the catalyst, the hydrogen combusts preferentially relative to hydrocarbon, thereby increasing the olefin selectivity of the overall process. The amount of hydrogen
25 combusted may be used to control the amount of heat generated and hence the severity of cracking. Thus, the molar ratio of hydrogen to oxygen can vary over any operable range provided that the ATC product stream comprising olefins is produced. Suitably, the molar ratio of hydrogen to oxygen is in the range 0.2 to 4, preferably, in the range 0.2 to 3.

The hydrocarbon to be cracked and oxygen may be contacted with the catalyst
30 capable of supporting combustion in any suitable molar ratio, provided that the ATC product stream comprising olefins is produced. The preferred stoichiometric ratio of hydrocarbon to oxygen is 5 to 16, preferably, 5 to 13.5 times, preferably, 6 to 10 times the

stoichiometric ratio of hydrocarbon to oxygen required for complete combustion of the hydrocarbon to carbon dioxide and water.

The mixed feedstream is passed over the catalyst at a gas hourly space velocity which is pressure dependent and typically greater than $10,000 \text{ h}^{-1} \text{ barg}^{-1}$, preferably greater than $20,000 \text{ h}^{-1} \text{ barg}^{-1}$ and, most preferably, greater than $100,000 \text{ h}^{-1} \text{ barg}^{-1}$. For example, at 20 barg pressure, the gas hourly space velocity is most preferably, greater than $2,000,000 \text{ h}^{-1}$. It will be understood, however, that the optimum gas hourly space velocity will depend upon the nature of the feed composition.

The autothermal cracking step may suitably be carried out at a catalyst exit temperature in the range 600°C to 1200°C . Suitably the catalyst exit temperature is at least 720°C such as at least 750°C . Preferably, the autothermal cracking step is carried out at a catalyst exit temperature in the range 850°C to 1050°C and, most preferably, in the range 850°C to 1000°C .

The autothermal cracking step is usually operated at a pressure of greater than 0.5 barg, preferably at a pressure of least 10 barg, and more preferably at a pressure of at least 20 barg. The pressure is preferably less than 50 barg, and more preferably less than 35 barg, for example in the range 20 to 30 barg.

The catalyst may be any suitable catalyst for autothermal cracking. The catalyst is capable of supporting combustion beyond the fuel rich limit of flammability. The catalyst usually comprises a Group VIII metal as its catalytic component. Suitable Group VIII metals include platinum, palladium, ruthenium, rhodium, osmium and iridium. Rhodium, and more particularly, platinum and palladium are preferred. Typical Group VIII metal loadings range from 0.01 to 100 wt %, preferably, between 0.01 to 20 wt %, and more preferably, from 0.01 to 10 wt % based on the total dry weight of the catalyst.

Where a Group VIII catalyst is employed, it is preferably employed in combination with a catalyst promoter. The promoter may be a Group IIIA, IVA, and/or VA metal. Alternatively, the promoter may be a transition metal; the transition metal promoter being a different metal to that which may be employed as the Group VIII transition metal catalytic component. Preferred promoters are selected from the group consisting of Ga, In, Sn, Ge, Ag, Au or Cu. The atomic ratio of Group VIII B metal to the catalyst promoter may be 1 : 0.1 - 50.0, preferably, 1: 0.1 - 12.0.

Preferred examples of promoted catalysts include Pt/Ga, Pt/In, Pt/Sn, Pt/Ge, Pt/Cu, Pd/Sn, Pd/Ge, Pd/Cu, Rh/Sn, Pt/Pd/Cu and Pt/Pd/Sn catalysts.

For the avoidance of doubt, the Group VIII metal and promoter in the catalyst may be present in any form, for example, as a metal, or in the form of a metal compound, such as an oxide.

The catalyst may be unsupported, such as in the form of a metal gauze, but is preferably supported. Any suitable support may be used such as ceramic or metal supports, but ceramic supports are generally preferred. Where ceramic supports are used, the composition of the ceramic support may be any oxide or combination of oxides that is stable at high temperatures of, for example, between 600°C and 1200°C. The support material preferably has a low thermal expansion co-efficient, and is resistant to phase separation at high temperatures.

Suitable ceramic supports include cordierite, lithium aluminium silicate (LAS), alumina (α -Al₂O₃), yttria stabilised zirconia, alumina titanate, niascon, and calcium zirconyl phosphate. The ceramic supports may be wash-coated, for example, with γ -Al₂O₃.

The catalyst capable of supporting combustion beyond the fuel rich limit of flammability may be prepared by any method known in the art. For example, gel methods and wet-impregnation techniques may be employed. Typically, the support is impregnated with one or more solutions comprising the metals, dried and then calcined in air. The support may be impregnated in one or more steps. Preferably, multiple impregnation steps are employed. The support is preferably dried and calcined between each impregnation, and then subjected to a final calcination, preferably, in air. The calcined support may then be reduced, for example, by heat treatment in a hydrogen atmosphere.

Although the catalyst bed has been described above in terms of a single catalyst bed, the catalyst bed may alternatively be present as a sequential catalyst bed, as described, for example, in WO 02/04389.

The invention will now be illustrated by way of Figures 1 to 5 and the following example, wherein:

Figure 1 shows in schematic form a catalyst holder according to the present invention;

Figure 2 shows in schematic form the catalyst holder in situ in a reactor;

Figures 3 to 5 show temperature profiles with and without cooling of the top surface of the catalyst holder.

In particular, Figure 1 shows a catalyst holder comprising a first part (1), a second part (2) and a third part (3). A catalyst (4) is held within the third part (3) supported on a support grid (5). A heat shield (6) is provided on the front face of the catalyst (4).

The first part comprises a flange which has a gasket (7) to provide a sealing arrangement with a reactant supply (not shown).

The first part (1) is in contact with a cooling plate (8) comprising cooling channels (not shown) through which a cooling fluid can be flowed to provide cooling of the first part. As shown, the cooling plate extends across the top of the catalyst holder, and hence need be provided with holes or other gaps through which the reactant stream may flow. As an alternative, the cooling plate may be in the form of a band around the top of the first part of the catalyst holder.

The wall of the catalyst holder in the second part (2) has a narrower thickness than that of the third part (shown as (9)).

Figure 2 shows a catalyst holder similar to Figure 1 located within a reactor. The catalyst holder is held in sealing engagement with an upstream section of the reactor (10, only the bottom part of which is shown) using a gasket (7) and flange bolts (11).

In this Figure, although shown by separate shading to distinguish it, the cooling plate is provided as an integral part of the upstream reaction section to which the catalyst holder is connected. Thus, the connection between the catalyst holder and the upstream reaction section may be the same as the connection of the catalyst holder to the cooling plate, as shown. However, it will also be readily apparent to the person skilled in the art that alternative connection configurations may equally be suitable as long as the cooling of the first part and sealed engagement are obtained. For example, the cooling plate may be provided as a separate plate, and may instead be connected to the catalyst holder independently of the connection of the catalyst holder to the upstream reactor section.

Figure 2 also shows part of the downstream section of the reactor (12, only the top part of which is shown). The gap between the third part of the catalyst holder and the reactor wall (12) is filled with a mesh (13) and in use may be purged with an inert gas.

Example 1 (Comparative)

This comparative example uses a catalyst holder wherein the wall of the catalyst holder (below the first part) has the same thickness in the second and third parts (and no specific cooling is provided).

5 The catalyst holder is shown in schematic form in Figure 3. Under the conditions of reaction, the catalyst is at 900°C and the incoming gaseous stream is at approximately 250°C.

As shown in Figure 3 the entire catalyst holder reaches a temperature approaching 900°C, the top of the catalyst holder reaching a temperature between 800°C and 900°C.

10 This will also result in an increase in the temperature of the upstream reactor.

Example 2

This example shows in schematic form the temperature profile of a catalyst holder with a thinned second part according to the present invention, as simulated by finite element analysis. The second section has thickness of 3mm.

15 As with the Comparative Example, the catalyst is at 900°C and the incoming gaseous stream is at approximately 250°C.

The results are represented schematically in Figure 4, which shows a portion of the catalyst holder (comprising a first part (1), a second part (2), a third part (3), catalyst (4) and heat shield (6)). (Note that the "gap" between the catalyst/heat shield and the catalyst holder is present only to aid clarity.)

20 The temperature at the top of the catalyst holder is significantly reduced compared to Comparative Example 1. In the actual analysis, the top of the catalyst holder is 366°C.

Example 3

25 The example shows the temperature profile of a catalyst holder with a thinned second part according to the present invention and with the provision of an additional cooling plate attached to its top surface, as simulated by computational fluid dynamics (CFD).

30 As with Example 2, the catalyst is at 900°C, the incoming gaseous stream is at approximately 250°C and the second section has thickness of 3mm. The cooling plate was cooled to 100°C using water flowing through channels therein.

The results are represented schematically in Figure 5, which shows a portion of the catalyst holder (comprising a first part (1), a second part (2), a third part (3), catalyst (4),

heat shield (6) and cooling plate (8)). The top of the catalyst holder is maintained at approximately 100°C. The majority of the first part is also maintained at a relatively low temperature (100-200°C), and only the third part of the catalyst holder reaches anywhere near the 900°C temperature of the catalyst.

CLAIMS

1. A catalyst holder for a catalyst bed, wherein the catalyst holder comprises:
- 5 (i) a first part which is adapted to provide a sealing engagement between the catalyst holder and a source of reactants,
- (ii) a second part, downstream of the first part as defined with respect to a flow of reactants, and
- (iii) a third part, downstream of the first and second parts as defined with respect to a flow of reactants and which is adapted to house a catalyst bed,
- 10 the catalyst holder having a wall which defines the internal cross-sections of the first, second and third parts and extends from the first part to the third part, wherein the average thickness of the wall in the second part is narrower than the average thickness of the wall in the third part.
2. A catalyst holder according to claim 1, wherein the first part of the catalyst holder
- 15 is provided with cooling means such that the first part of the catalyst holder is maintained at a temperature at least 100°C below the front face temperature of the catalyst bed during reaction.
3. A catalyst holder according to claim 1, wherein the first part of the catalyst holder is provided with cooling means such that the first part of the catalyst holder is maintained
- 20 at a temperature below 300°C, preferably below 200°C.
4. A catalyst holder according to claim 2 or claim 3 wherein the cooling means comprises channels through which a cooling fluid can flow during reaction to maintain the
- at first part of the catalyst holder at the required temperature
5. An apparatus for the reaction of a hydrocarbon with oxygen, which comprises:
- 25 a) a reactant supply section, which comprises a hydrocarbon supply, an oxygen supply and a mixing section for mixing the hydrocarbon and oxygen,
- b) a reaction section, downstream of the reactant supply section with respect to the flow of the reactants, which comprises a catalyst holder as described in
- 30 any one of claims 1 to 4, and
- c) a product removal section, downstream of the catalyst holder with respect to the flow of the reactants,

wherein the reactant supply section is connected in sealed engagement with the first part of the catalyst holder, and such that the third part of the catalyst holder resides within the upper section of the product removal section with a gap between the outer surface of the third part of the catalyst holder and the inner surface of the upper section of the product
5 removal section, said gap being purged with a suitable purge gas.

6. A process for the reaction of a hydrocarbon with oxygen, which process comprises passing a mixed gaseous reactant stream comprising said hydrocarbon and oxygen into contact with a catalyst bed in a catalyst holder as claimed in any one of claims 1 to 4 or in an apparatus as claimed in claim 5.

Fig.1

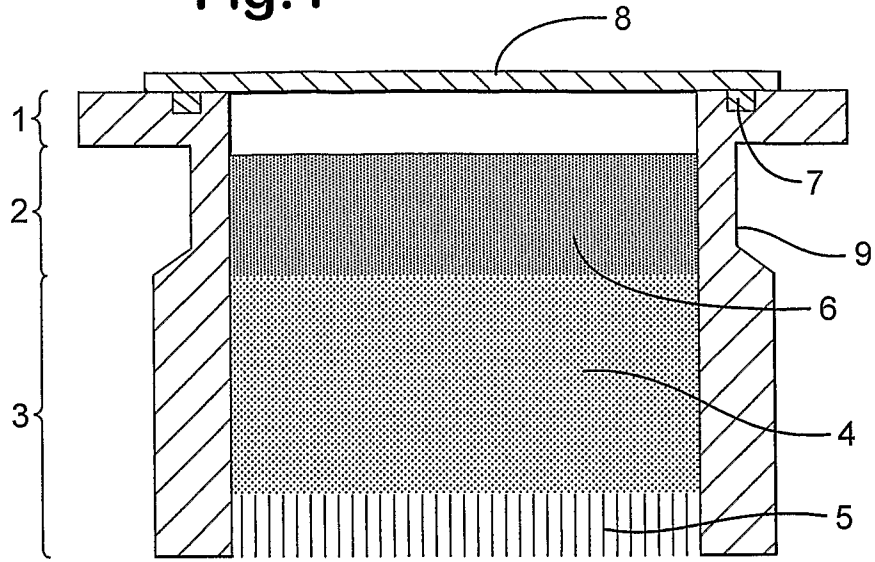


Fig.2

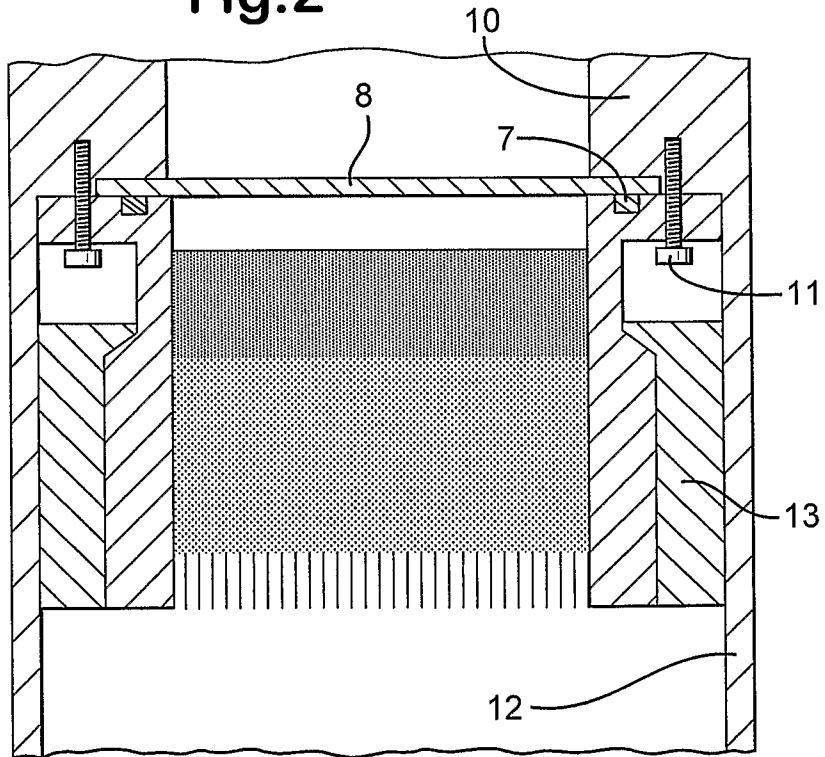


Fig.3

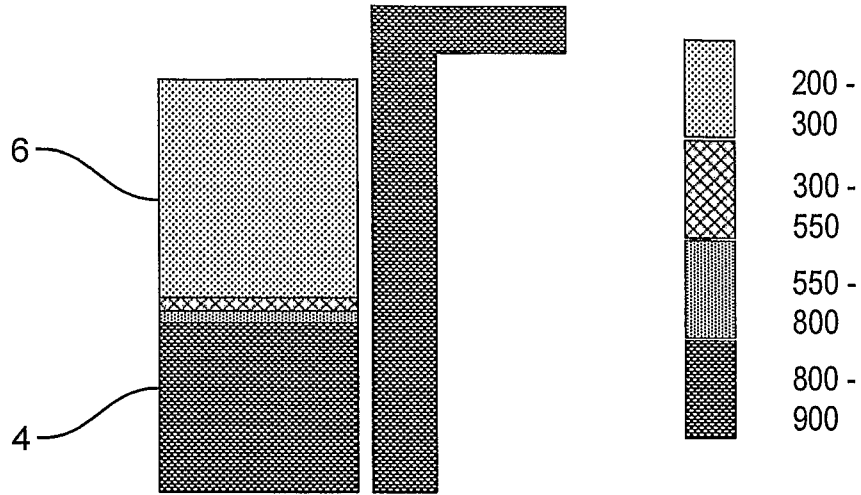


Fig.4

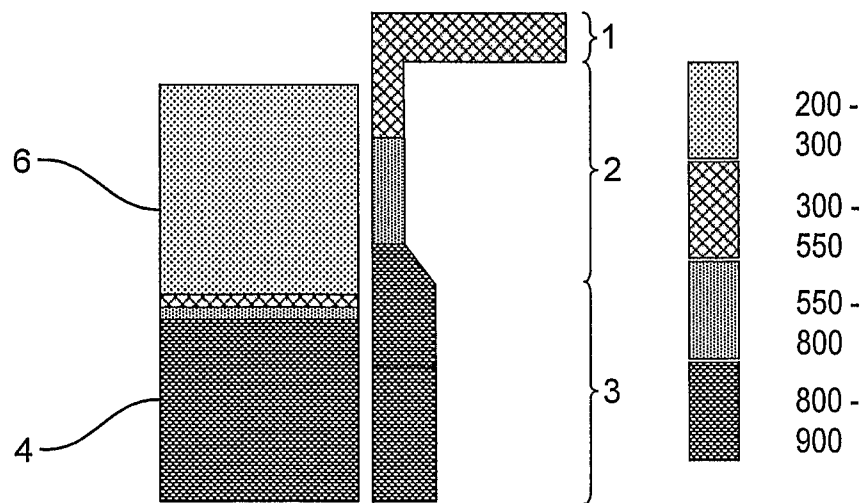
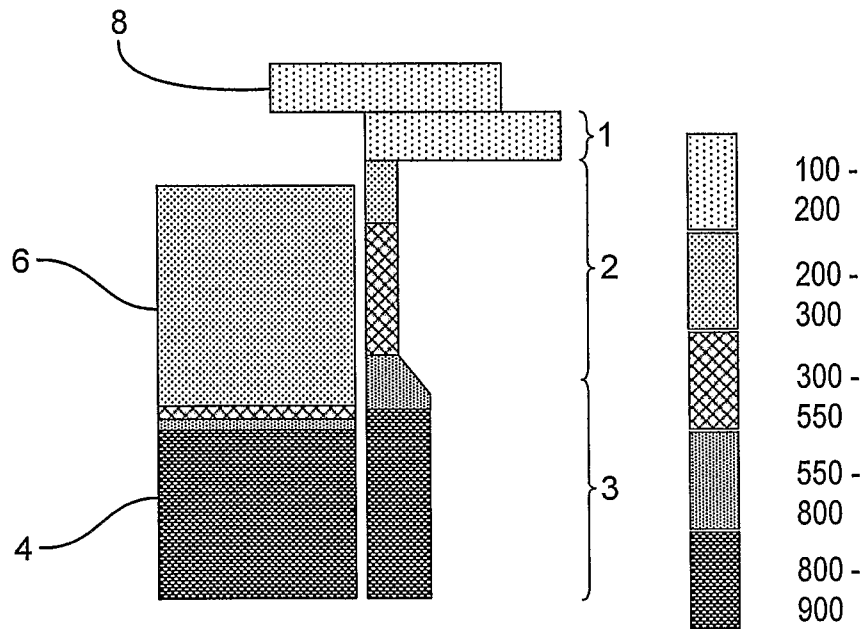


Fig.5



INTERNATIONAL SEARCH REPORT

International application No
PCT/GB2006/004642

A. CLASSIFICATION OF SUBJECT MATTER
INV. B01J8/02

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 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.
X	US 3 195 988 A (ROBERTS EDWARD S ET AL) 20 July 1965 (1965-07-20) column 2, lines 61-69; figure 1	1,5
A	GB 1 334 577 A (UNIVERSAL OIL PROD CO) 24 October 1973 (1973-10-24) claim 1; figure 1	1,5,6
A	WO 2004/074222 A (BP CHEM INT LTD [GB]; COLMAN DEREK ALAN [GB]; MATTHEWMAN MICHAEL JOHN) 2 September 2004 (2004-09-02) cited in the application page 16, lines 21-33; figure 1	2-4
A	US 2004/171894 A1 (COLMAN DEREK ALAN ET AL) 2 September 2004 (2004-09-02) paragraphs [0018], [0073], [0075], [0076], [0078]; figure 1	2-4

Further documents are listed in the continuation of Box C.

See patent family annex.

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- *Z* document member of the same patent family

Date of the actual completion of the international search

15 February 2007

Date of mailing of the international search report

05/03/2007

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INTERNATIONAL SEARCH REPORT

Information on patent family members

International application No PCT/GB2006/004642
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