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CA, CH, CL, CN, CO, CR, CU, CZ, DE, DJ, DK, DM, DO,
DZ, EC, EE, EG, ES, FI, GB, GD, GE, GH, GM, GT, HN,
HR, HU, ID, IL, IN, IR, IS, JO, JP, KE, KG, KH, KN, KP,
KR, KW, KZ, LA, LC, LK, LR, LS, LU, LY, MA, MD, ME,
MG, MK, MN, MW, MX, MY, MZ, NA, NG, NI, NO, NZ,
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UG, ZM, ZW), Eurasian (AM, AZ, BY, KG, KZ, RU, TJ,
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EE, ES, FI, FR, GB, GR, HR, HU, IE, IS, IT, LT, LU, LV,

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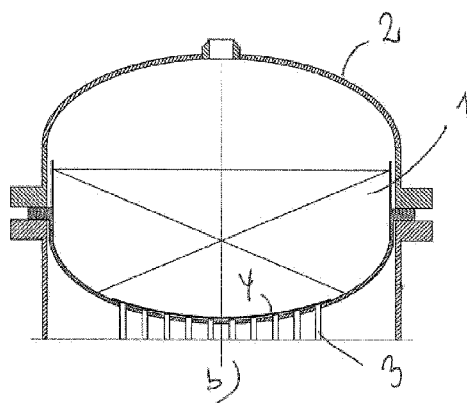
Declarations under Rule 4.17:

- 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))
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(54) Title: MULTI BED REACTOR COMPRISING AN ADIABATIC AND A HEATED CATALYST BED



(57) Abstract: The present application relates to a reactor comprising a heated catalyst bed and a guard layer and/or adiabatic catalyst, as well as a method for revamping an existing reactor having at least a heated catalyst bed.



Title: Multi bed reactor comprising an adiabatic and a heated catalyst bed

During the last 25 years use of separate prereforming reactors has become common part of the steam reforming section in synthesis gas preparation plants. In the beginning this was driven by the need to reform feed stocks with content of higher hydrocarbons. Today prereforming is also used to reduce the size of the downstream primary reformer and to protect the catalyst in the primary reformer even in case of feedstocks with low content of hydrocarbon. A prereforming step is an excellent catalyst guard removing for example all Sulphur components quantitatively. The prereforming – primary reforming concept is a well-established concept in the industry

There is still a reluctance to use prereforming in the ammonia industry. Partly because of conservatism, partly because of process integration problems as a prereforming step may reduce required steam production.

Prereforming protects the primary reforming catalyst typically increasing the expected lifetime from 3-5 to 10 years. Over a 10 years' period 1 to 2 primary reformer catalyst exchanges are thus avoided. As it is time consuming to change primary reformer catalyst this is a huge economic benefit, more than covering the cost of having a prereforming step.

Heat exchange reformers, where a hot gas stream is used as heat source for the reforming process through heat exchange, are typical very compact designs where catalyst is even more difficult to change than in a primary reformer. It is therefore very relevant to have a prereforming step before a heat exchange reformer. Heat exchange reformers are often used for revamps to increase the reforming capacity. This is also feasible for ammonia plants, which typically have no prereforming step and where it typically will not be economically feasible to install a separate prereforming step.

In a first aspect of the present invention is provided a reactor which provides reduced equipment/piping cost.

In a first aspect of the present invention is provided a reactor which provides reduced plot space requirement thereby enabling cost savings and increased versatility of the

reactor.

These and other advantages are achieved by a reactor comprising a guard layer and/or adiabatic catalyst layer and a heated catalyst bed. I.e. by the present invention a single reactor comprising a heated catalyst bed as well as a guard layer and/or adiabatic catalyst layer is provided whereby significant CAPEX savings may be achieved compared to known multi reactor setups.

The present invention thus combines prereforming and heat exchange reforming in a single reactor with no or minimal impact on the reactor dimensions. The added cost of having a prereforming step protecting the heat exchange reforming step is therefore merely the catalyst cost making the combined solution economically feasible against a solution with no prereforming step, considering the heat exchange reformer catalyst exchange cost.

Combining a guard/catalyst layer and a heat exchange reactor step in one reactor provides:

- Reduced equipment/piping cost
- Reduced plot space requirement saving cost and increased possibility to install the solution as part of a revamp of an existing plant.
- Possibility for easy exchange of guard/catalyst layer without affecting the heat exchange catalyst.

Preferably the heated catalyst bed is downstream the guard layer and/or adiabatic catalyst layer. I.e. preferably the outlet from the guard layer and/or adiabatic catalyst layer is the inlet to the heated catalyst bed. This way the reaction gas which enters the reactor first passes the guard layer and/or adiabatic catalyst layer and thereafter the heated catalyst bed.

When the reaction gas first passes the guard layer and/or adiabatic catalyst layer i.e. when the adiabatic catalyst layer is arranged upstream the heated catalyst bed the lifetime of the catalyst in the heated catalyst bed may be significantly increased.

Having the guard layer and/or adiabatic catalyst layer separate from the heated catalyst bed it is possible to change the guard layer and/or adiabatic catalyst more frequently than the catalyst in the heated catalyst layer. If for example the heated catalyst bed is an exchange reformer, the process of changing the catalyst in the heated catalyst bed is relatively demanding in which case it may be highly desirable to be able to enhance the life time of the heated catalyst by adding the guard layer and/or adiabatic catalyst layer which furthermore may be simpler/faster/easier to change.

The guard layer and/or adiabatic catalyst layer may e.g. be a Sulphur guard, a hydrocarbon desulphurisation catalyst and/or a hydrocarbon steam reforming catalyst.

The catalyst in the heated catalyst bed may preferably be a hydrocarbon steam reforming catalyst.

The present reactor may be specifically designed and provided from new to comprise the heated catalyst bed as well as the guard layer and/or adiabatic catalyst layer.

However, the present reactor may alternatively be provided as a revamp solution to existing reactors comprising one or more heated catalyst beds. For example the present reactor may be provided as a revamp to existing exchange reformers.

In existing reactors, such as exchange reformers, a heated catalyst bed is present in the reactor. Above the heated catalyst bed is a hollow in known setups due to gas distribution requirements. The applicant has shown that this free space above the heated catalyst bed in a revamp solution advantageously may be used for a guard layer and/or adiabatic catalyst layer.

The invention is not limited to the prereforming – primary reformer concept but can also be used for other processes where a combination of an adiabatic/guard reactor and a heat exchange reactor is used. An example of this could be a steam fired reverse shift reactor in combination with a chlorine/Sulphur guard layer. Similarly, the invention is valid for process combinations where the first step is the heat exchange reactor and the second step a guard/catalyst layer. The heat exchange reactor has inherently also an empty room in the outlet head providing room for a guard/catalyst layer.

The guard/catalyst layer can for example be used to remove/convert bi-products formed in the heat exchange reactor step

5 Table 1 shows a typical set of process conditions for a heat exchange reformer. The REACTOR INLET stream is send through a number of reforming catalyst containing tubes. These tubes are heated from outside by cooling the HEAT SOURCE gas. Before leaving the reactor the outlet gas from the catalyst containing tubes is mixed with the cooled heat source gas to form the final PRODUCT gas.

10 Table 1

STREAM DESCRIPTION	INLET REACTOR	HEAT SOURCE	PRODUCT
FLOW (nm ³ /h)	31631,40	263331,60	308659,20
TEMP (°C)	359,88	1010,52	798,95
PRESSURE (bar g)	30,94	30,94	30,32
COMPOSITION (dry mole%)			
<i>Hydrogen</i>	<i>3,75</i>	<i>51,31</i>	<i>54,67</i>
<i>Nitrogen</i>	<i>1,51</i>	<i>27,45</i>	<i>23,56</i>
<i>Carbon Monoxide</i>	<i>0,00</i>	<i>13,31</i>	<i>13,73</i>
<i>Carbon Dioxide</i>	<i>2,12</i>	<i>7,45</i>	<i>7,48</i>
<i>Argon</i>	<i>0,02</i>	<i>0,33</i>	<i>0,28</i>
<i>Methane</i>	<i>84,23</i>	<i>0,14</i>	<i>0,29</i>
<i>Ethane</i>	<i>4,47</i>	<i>0,00</i>	<i>0,00</i>
<i>Propane</i>	<i>2,53</i>	<i>0,00</i>	<i>0,00</i>
<i>n-Butane</i>	<i>0,48</i>	<i>0,00</i>	<i>0,00</i>
<i>n-Pentane</i>	<i>0,09</i>	<i>0,00</i>	<i>0,00</i>
<i>n-Hexane</i>	<i>0,08</i>	<i>0,00</i>	<i>0,00</i>
<i>Isobutane</i>	<i>0,55</i>	<i>0,00</i>	<i>0,00</i>
<i>2-Methylbutane</i>	<i>0,16</i>	<i>0,00</i>	<i>0,00</i>

Table 1: Heat exchange reformer without adiabatic prereformer bed

Table 2

STREAM DESCRIPTION	INLET REACTOR	INTERMEDIATE	HEAT SOURCE	PRODUCT
FLOW (nm ³ /h)	31631,40	32387,80	263331,60	308638,00
TEMP (°C)	359,88	350,72	1010,52	798,78
PRESSURE (bar g)	31,44	30,95	30,94	30,32
COMPOSITION (dry mole%)				
<i>Hydrogen</i>	3,75	9,93	51,31	54,66
<i>Nitrogen</i>	1,51	1,23	27,45	23,56
<i>Carbon Monoxide</i>	0,00	0,01	13,31	13,72
<i>Carbon Dioxide</i>	2,12	6,31	7,45	7,48
<i>Argon</i>	0,02	0,02	0,33	0,28
<i>Methane</i>	84,23	82,50	0,14	0,29
<i>Ethane</i>	4,47	0,00	0,00	0,00
<i>Propane</i>	2,53	0,00	0,00	0,00
<i>n-Butane</i>	0,48	0,00	0,00	0,00
<i>n-Pentane</i>	0,09	0,00	0,00	0,00
<i>n-Hexane</i>	0,08	0,00	0,00	0,00
<i>Isobutane</i>	0,55	0,00	0,00	0,00
<i>2-Methylbutane</i>	0,16	0,00	0,00	0,00
Table 2: Heat exchange reformer with adiabatic prereformer bed				

Table 2 shows the effect of sending the INLET REACTOR gas through an adiabatic pre-reforming catalyst layer before it is send through the heated reforming catalyst containing tubes. The INTERMEDIATE shows the outlet stream from the adiabatic pre-reforming catalyst layer. It is seen that no higher hydrocarbons are present in this stream. Furthermore, Sulphur components in the INLET REACTOR stream will be absorbed by the pre reforming catalyst. These two effects increase the lifetime of the reforming catalyst in the tubes from approximate 3 years to 10 years. As a secondary effect the capacity can be increased.

Figure 1 b) – f) show various examples of how a guard/adiabatic catalyst layer (1) can be integrated in a heat exchange reformer (2) with no or minimum change of the reactor dimensions. The adiabatic catalyst layer (1) is placed in the entrance head of the heat exchange reformer (2), where there typically is an empty room, such that the incoming gas is send through this adiabatic catalyst layer 1 before entering the catalyst tubes (3). Figure 1 b) provides example of a simple solution placing the guard/catalyst

layer directly on the top of the tube sheet only separated by a grid (4) preventing catalyst from falling into the tubes. Figure 1 c) – f) provides various examples of one or more cassette's (5) which easily can be taken in and out of the reactor for quick catalyst exchange. Solutions are not limited to what is shown in Figure 1. The principle can also be adopted in heat exchange reformer designs with different entrance head design and dimensions. Typically, there will inherently be an empty entrance room used for gas distribution which is large enough to install a feasible guard/catalyst layer. Figure 1 a) shows a standard heat exchange reformer without an adiabatic catalyst layer.

Claims

1. A reactor comprising an adiabatic catalyst bed and a heated catalyst bed.
- 5 2. A reactor according to claim 1 wherein the product gas leaving the adiabatic catalyst bed is inlet gas to the heated catalyst bed.
3. A reactor according to claim 1 and 2 wherein the adiabatic catalyst bed comprises a hydrocarbon steam reforming catalyst.
- 10 4. A reactor according to claim 1 and 2 wherein the adiabatic catalyst bed comprises a hydrocarbon desulphurisation catalyst.
5. A reactor according to claim 1 and 2 wherein the heated catalyst bed comprises a hydrocarbon steam reforming catalyst.
- 15 6. A reactor according to any of the preceding claims wherein said reactor is a heat exchange reformer.
- 20 7. A hydrocarbon steam reforming process using the reactor according to any of the claims 1 - 6.
8. A method for revamping an existing reactor having at least a heated catalyst bed, said method comprising
- 25 - providing a guard layer and/or adiabatic catalyst layer in said existing reactor.
9. A method according to claim 8 wherein the product gas wherein the heated catalyst bed is downstream the adiabatic catalyst bed.
- 30 10. A method according to claim 8 or 9 wherein the adiabatic catalyst bed is filled with hydrocarbon steam reforming catalyst.

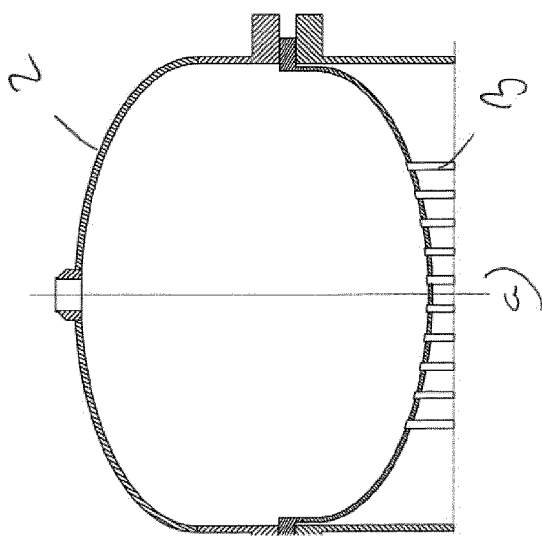
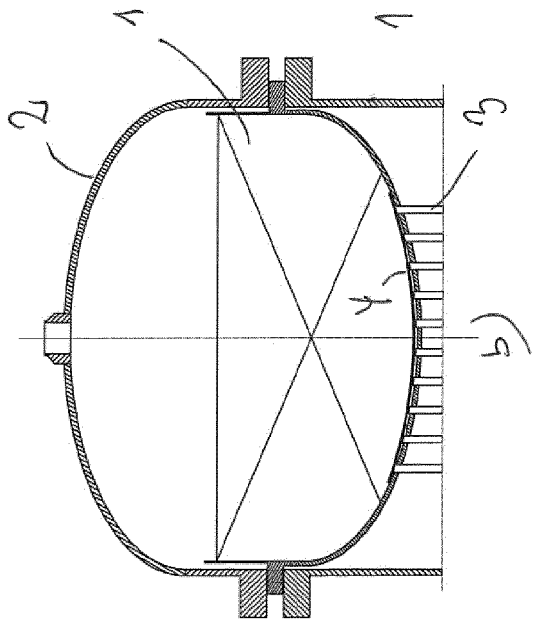
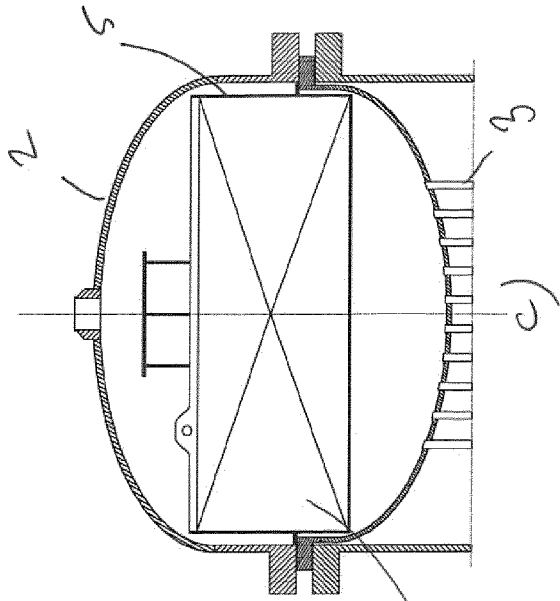
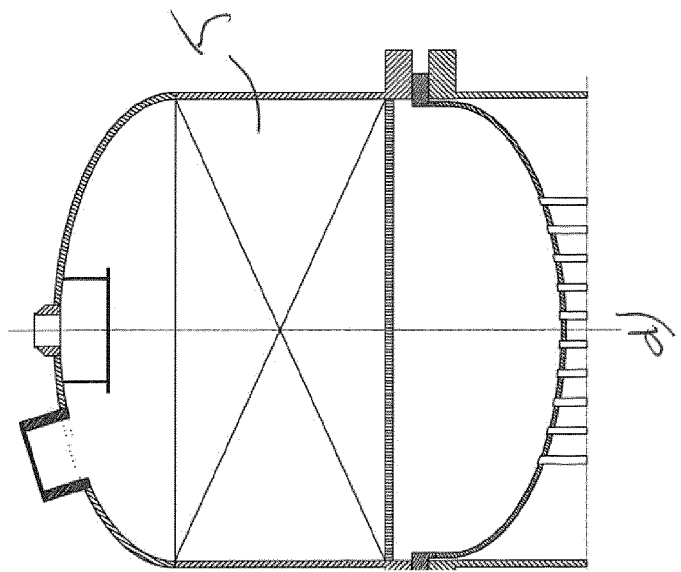
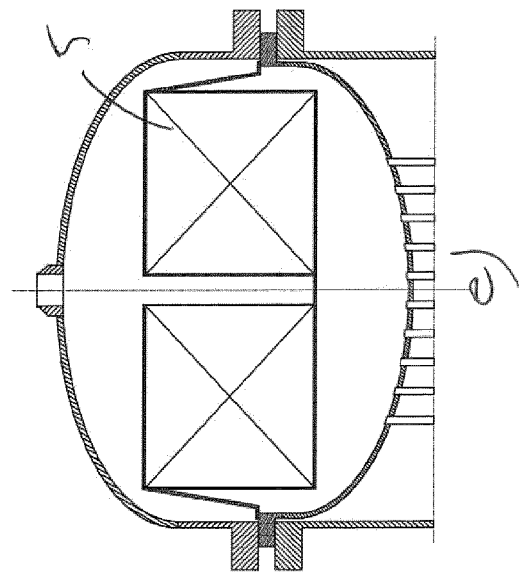
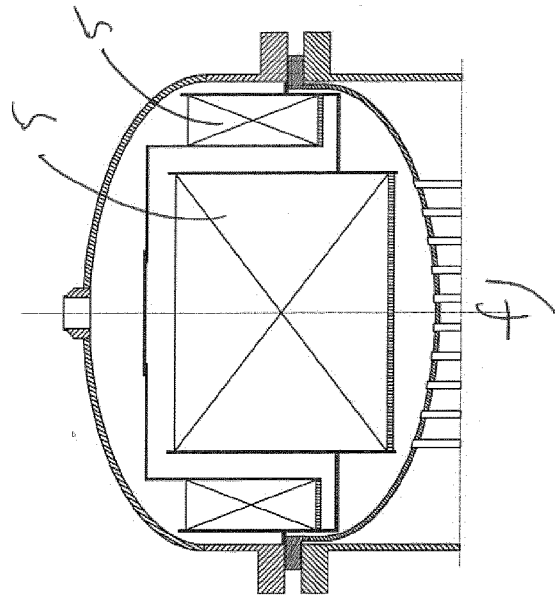


Fig 1



INTERNATIONAL SEARCH REPORT

International application No
PCT/EP2017/080157

A. CLASSIFICATION OF SUBJECT MATTER
INV. B01J8/04
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, 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	CN 2 621 805 Y (HUADONG TECH UNIV) 30 June 2004 (2004-06-30) abstract figure 1 claims 1, 2 page 5, lines 28-29	1,2,8-10
X	WO 2007/000244 A1 (HALDORE TOPSOE AS [DK]; BOE MICHAEL [DK]; HANSEN JOHN BOGILD [DK]) 4 January 2007 (2007-01-04) abstract figure 1 claims 1, 9 page 24, lines 16-17	1-7

Further documents are listed in the continuation of Box C.

See patent family annex.

* Special categories of cited documents :

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- "E" earlier application or patent but published on or after the international filing date
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Date of the actual completion of the international search 1 February 2018	Date of mailing of the international search report 20/02/2018
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Name and mailing address of the ISA/ European Patent Office, P.B. 5818 Patentlaan 2 NL - 2280 HV Rijswijk Tel. (+31-70) 340-2040, Fax: (+31-70) 340-3016	Authorized officer Biscarat, Jennifer
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INTERNATIONAL SEARCH REPORT

International application No
PCT/EP2017/080157

C(Continuation). DOCUMENTS CONSIDERED TO BE RELEVANT		
Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
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X	----- US 5 464 606 A (BUSWELL RICHARD F [US] ET AL) 7 November 1995 (1995-11-07) abstract figure 2	1,2
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INTERNATIONAL SEARCH REPORT

Information on patent family members

International application No PCT/EP2017/080157

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