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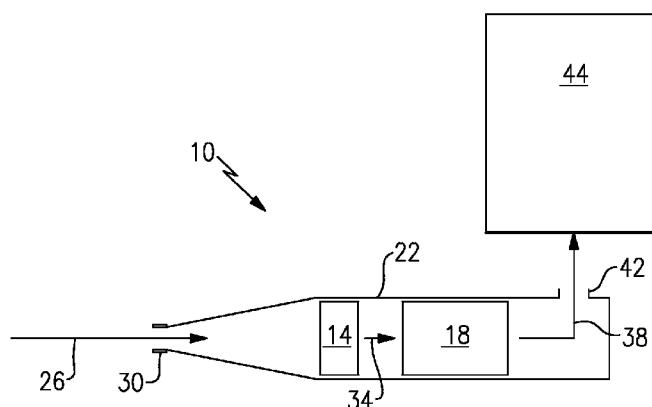


FIG.1

(57) Abstract: An example adsorber assembly includes an adsorber and a housing. The housing extends between an inlet and an outlet. The housing is configured to communicate a reformate gas through at least a portion of the adsorber. The adsorber includes a material coating selected from a group consisting of cerium oxide, a rare earth oxide, and combinations of these. The material coating having a thickness and being configured to react with hydrogen sulfide throughout the thickness.

ADSORBER ASSEMBLY

STATEMENT REGARDING FEDERALLY SPONSORED RESEARCH OR DEVELOPMENT

[0001] This invention was made with government support under Contract No. N00014-09-C-0430, awarded by the United States Navy. The Government may have certain rights in this invention.

BACKGROUND

[0002] This disclosure relates generally to treating reformat gas and, more particularly, to removing hydrogen sulfide from reformat gas supplied to a fuel cell stack.

[0003] Many fuel cell stacks require a supply of reformat gas. Autothermal reformer (ATR) reactors may supply the reformat gas. The ATR reactors use a supply of fuel and oxygen to generate the reformat gas, which may include sulfur in the form of hydrogen sulfide. As known, sulfur can poison electrodes within a fuel cell stack, such as the anode electrodes within individual solid oxide fuel cells. Accordingly, the reformat gas is typically treated to remove some of the sulfur before the reformat gas enters the fuel cell stack. In one example, the reformat gas must contain less than 100 parts per billion by volume of hydrogen sulfide to avoid poisoning the electrodes.

[0004] Zinc oxide is often used as an adsorbent to remove sulfur from the reformat gas. The effectiveness of the zinc oxide as an adsorbent varies with temperature. Zinc oxide is most effective at temperatures below about 400°C, which is lower than the temperature of the reformat gas exiting the autothermal reformer and higher than the desired temperature of the reformat gas entering the fuel cell stack. Using zinc oxide at lower temperatures may result in coke deposition that can decrease efficiency of the overall system. Thus, in arrangements using zinc oxide as an adsorbent, the reformat gas is cooled and then reheated before entering the fuel cell stack. Cooling and reheating the reformat gas requires multiple heat exchangers.

SUMMARY

[0005] An example adsorber assembly includes an adsorber and a housing. The housing extends between an inlet and an outlet. The housing is configured to communicate a

reformate gas through at least a portion of the adsorber. The adsorber includes a material coating selected from a group consisting of cerium oxide, a rare earth oxide, manganese oxide, and combinations of these. The material coating having a thickness and being configured to react with hydrogen sulfide throughout the thickness.

[0006] In another embodiment, the adsorber includes a material coating selected from a group consisting of cerium oxide, lanthanum oxide, and combinations of these in the form of (CeO₂, La₂O₃).

[0007] In another embodiment, the adsorber includes a material coating selected from a group consisting of cerium oxide, lanthanum oxide, manganese oxide and combinations of these in the form of (CeO₂, La₂O₃, MnO).

[0008] In yet another embodiment, the adsorber includes a material coating selected from a group consisting of cerium oxide, a rare earth oxide (REO) selected from the group of lanthanum, praseodymium, neodymium, samarium, and gadolinium and combinations of these. In this embodiment, the material coating is comprised of cerium oxide, a rare earth oxide selected from the group of lanthanum, praseodymium, neodymium, samarium, and gadolinium, manganese oxide and combinations of these in the form of (CeO₂, REO, MnO).

[0009] An example fuel cell adsorber arrangement includes a reformer and a fuel cell stack. An adsorber includes a material selected from a group consisting of cerium oxide, lanthanum oxide, and combinations of these. The reformate gas communicates between the reformer and the fuel cell stack through the adsorber. The reformate gas communicated from the trap has less hydrogen sulfide than the reformate gas communicated to the adsorber. The layer of material has a thickness and is configured to react with hydrogen sulfide throughout the thickness.

[0010] An example method of conditioning a reformate gas stream includes communicating a reformate gas stream through an adsorber and adsorbing material from the reformate gas stream using a material within the adsorber. The material is selected from a group consisting of cerium oxide, a rare earth oxide, and combinations of these. The adsorber is configured such that the entire thickness of the layer of material is able to absorb hydrogen sulfide. The method then communicates the reformate gas to a fuel cell stack. The rare earth oxide is lanthanum oxide in one example.

[0011] These and other features of the disclosed examples can be best understood from the following specification and drawings, the following of which is a brief description.

BRIEF DESCRIPTION OF THE FIGURES

[0012] Figure 1 shows a schematic view of an example fuel cell adsorber arrangement.

[0013] Figure 2 shows a partial cutaway view perspective view of the Figure 1 adsorber assembly.

[0014] Figure 2A shows an end view of a portion of the Figure 2 adsorber assembly.

[0015] Figure 2B shows a partial section view at line 2-2 in Figure 2.

[0016] Figure 3 shows a perspective view of a portion of the adsorber in the Figure 2 adsorber assembly.

DETAILED DESCRIPTION

[0017] As shown in Figure 1, an example adsorber assembly 10 includes a reformer 14 and an adsorber 18 housed within a housing 22. An inlet gas stream 26 enters the housing 22 at an inlet 30 and flows through the reformer 14. The inlet gas stream 26 is treated within the reformer 14. A reformat gas stream 34 or syngas flows from the reformer 14 to the adsorber 18. The adsorber 18 removes hydrogen sulfide from the reformat gas stream 34. An outlet gas stream 38 then flows from the adsorber 18 through an outlet 42 in the housing 22 to a fuel cell stack 44.

[0018] The example fuel cell stack 44 includes a plurality of individual solid oxide fuel cells that provide power in a known manner. The fuel cell stack 44 utilizes the outlet gas stream 38 to support the chemical reactions that provide electrical power.

[0019] In this example, the inlet gas steam 26 includes a hydrocarbon fuel, steam and air. The outlet gas stream 38 includes hydrogen, carbon monoxide, carbon dioxide, nitrogen, methane, water, and trace compounds. Notably, the outlet gas stream 38 includes less hydrogen sulfide than the reformat gas stream 34. In one example, the outlet gas stream 38 includes less than 100 parts per billion by volume of hydrogen sulfide, and the reformat gas stream 34 includes about 648 parts per billion by volume of hydrogen sulfide or higher concentrations of hydrogen sulfide.

[0020] Referring to Figures 2-2B with continuing reference to Figure 1, the example adsorber 18 (or trap) includes a base 46 that is washcoated with a layer 48 of adsorber material. The adsorber material is a single rare earth oxide, e.g., CeO_2 or Ce_2O_3 or La_2O_3 , a binary oxide, e.g., $(\text{CeO}_2, \text{La}_2\text{O}_3)$, a ternary oxide, e.g., $(\text{CeO}_2, \text{La}_2\text{O}_3, \text{MnO})$ or a multi-metal oxide, e.g., $(\text{CeO}_2, \text{REO}, \text{MnO})$ wherein the binary, ternary, or multi-metal oxide materials are formed as a compound oxide or a finely inter-dispersed mixture of the constituent oxides, in this example. These single, binary, ternary or multi-metal oxide materials are prepared by well established processes to prepare powders that have high surface area and high sintering reactivity.

[0021] The example layer 48 covers each surface of the base 46 with the adsorber material. Other examples cover less of the base 46 leaving some of the base 46 exposed.

[0022] The example base 46 is made of a cordierite material or another ceramic material. The base 46 includes multiple pores 50 each having a rectangular or near square cross-section. The pores are oftentimes referred to as channels or cells, and have macroscale dimensions, i.e., widths greater than about 50 micrometers. These pores or channels or cells are inherently different from the microscopic pores of the adsorber material layer 48 as will be discussed hereinafter. The pores 50 extend longitudinally through the adsorber 18 along an axis A aligned with the direction of flow through the adsorber 18. The base 46 includes a plurality of walls 52 that establish the pores 50. In another example, the pores 50 have a honeycomb or hexagonal cross-section. In another example, the pores 50 have a circular cross-section.

[0023] The example base 46 ceramic is known in the art as a honeycomb or ceramic monolith or ceramic monolith substrate. These monoliths are widely used in the automotive business as a support for the catalyst washcoatings of the catalytic converter used in exhaust gas clean-up. The high open flow area of these honeycomb substrates ensure that gas can flow through the ceramic with minimum resistance to flow, i.e., low pressure drop, at high gas velocities. The example base 46 with the washcoating adsorber material for the removal of hydrogen sulfide from reformat gas is projected to lead to pressure drops of less than 3 kPa, and most preferably less than 1 kPa.

[0024] The example pores 50 have a width X of about 1 millimeter and a height Y of about 1 millimeter. The thickness T_1 of cordierite in the walls 52 is about 100 micrometers. The layer 48 has a thickness T_2 that is less than 50 micrometers. The layer 48 is washcoated to each

side of the walls 52. Thus, the total thickness T_T of the example walls 52 is equal to $T_2 + T_1 + T_2$, which is less than 200 micrometers. The layer 48 thickness needs to be as large as possible in order to increase the sulfur capacity of the adsorber material. However, the layer 48 thickness is limited by spallation, which tends to occur with increasing layer 48 thickness. As a result, the optimum layer 48 thickness is a compromise, or tradeoff, between these two opposing effects.

[0025] The adsorber material layer 48 needs to be sufficiently porous so as to promote the diffusional transport of hydrogen sulfide in the pore gas space to the interior surfaces of the adsorber material and ensure as complete reaction as possible of the adsorber material. The porosity of the layer 48 should be as high as possible, preferably up to about 50%, but it should not compromise the structural stability of the coating. The pore size distribution should be tailored so as to promote the hydrogen sulfide diffusional transport rates, in other words, have large enough pores so that the transport of H_2S occurs in the pore gas space under bulk or molecular diffusion, yet provide high enough surface area to promote the reaction of H_2S with the adsorbent material. The latter implies the need to have some pores of small size, in which diffusional transport may be controlled by Knudsen diffusion. A combination of large and small pores is desirable, and this combination of pore sizes is usually referred to as a bimodal distribution of the pores in the adsorber material 48 in some examples. The large pores of the adsorber material may have a diameters in the range of 0.5 to 5 micrometers whereas the small pores may have a diameter of 0.05 to 0.5 micrometers. As will be discussed later, the layer 48 is applied to the honeycomb by wash-coating and sintering and the formation of the larger pores is achieved by incorporating pore former materials, i.e., organic or carbon based solids which burn off during the heat treatment and sintering process in air leaving behind larger voids and pores.

[0026] The layer 48 of the rare earth oxide adsorbs hydrogen sulfide from the reformat gas stream 34 as the reformat gas stream 34 flows through the adsorber 18. During the early stages of adsorption, the areas A_1 of the walls 52 closest to the inlet 30 have reacted with the hydrogen sulfide. The area of the walls 52 that have reacted with the hydrogen sulfide increases (e.g., to area A_2) as more hydrogen sulfide within the reformat gas stream 34 has moved through the adsorber 18 and more of the layer 48 reacts with the hydrogen sulfide.

[0027] In this example, the layer 48 is a cerium oxide that reduces to Ce_2O_3 in reformat gas. In another example, the layer 48 is a lanthanum oxide La_2O_3 . Other examples use binary or ternary alloys of cerium oxide or lanthanum oxide, multi-metal oxide adsorber

material referred to earlier. In another example, the layer 48 is a cerium-lanthanum-manganese oxide, this is another multi-metal oxide adsorber material in one other example. In yet another example, the layer 48 is comprised of a multi-metal oxide based on cerium oxide, a rare earth oxide (REO) selected from the group of lanthanum, praseodymium, neodymium, samarium, and gadolinium and combinations of these which may be represented by the simplified formula (CeO_2 , REO, MnO). In this example, the solid phase of the entire thickness T_2 of the layer is configured to react with hydrogen sulfide within the reformat gas stream 34.

[0028] The sulfur adsorption capacity of these example oxides ranges from a high of about 115 mg/g at 600 C with no steam present in the gas to about 1 mg/g in gas streams containing 10 mol% steam at residence times of about 2 milliseconds.

[0029] Referring to Figure 4, the example housing 22 is a cylindrical housing extending from the inlet 30 to the outlet 42. The housing 22 has a frustoconical shape near the inlet 30, to promote near uniform flow distribution across the reformer 14 and reduce pressure losses.

[0030] In this example, a radiation shield 72 is disposed against the reformer 14. The inlet gas stream 26 passes through the radiation shield 72 before entering the reformer 14. The radiation shield in this example is a perforated metal or ceramic disk. In other instances, the radiation shield is made of a high porosity metal or ceramic foam.

[0031] The example reformer 14 is a catalytic autothermal reformer. In another example, the reformer 14 is a catalytic partial oxidation reformer or a catalytic steam reformer. The example reformer 14 is a porous ceramic with a catalyst and has a diameter D of about 12.065 cm. The catalysts used in the autothermal reformer, partial oxidation reformer, and steam reformer are well known in the art and may be comprised of a combination of noble and base metals on inert ceramic supports, with or without promoters, that optimize selectivity and reforming efficiency, mitigate carbon formation and impart resistance to sulfur poisoning.

[0032] In this example, a spacer 76 is disposed between the adsorber 18 and the reformer 14. The reformat gas stream 34 flows through an aperture 78 in the spacer 76.

[0033] A seal 80 surrounds a radially outer surface of the adsorber 18 such that the adsorber 18 is radially spaced from the housing 22. The housing 22 is a metallic material in this example. The housing 22 expands and contracts with temperature changes at a different rate than the adsorber 18. The seal 80 ensures that no gaps form between the adsorber 18 and the

housing 22 so that the reformat gas stream 34 is forced to move through the adsorber 18 rather than moving through the gaps. In this example, the seal 80 is an interam intumescent mount seal.

[0034] A maintenance cap 84 connects to the adsorber 18 and the seal 80 with a plurality of pins 88. The maintenance cap 84 enables a user to remove the adsorber 18 from the housing 22 by pulling the maintenance cap 84 in a direction Z. Once removed, the user can replace the adsorber 18 with a different or fresh or unreacted or virgin adsorber. Replacing the adsorber 18 may be necessary when substantially all the material in the layer 48 has reacted with hydrogen sulfide and the adsorber 18 is no longer effectively removes hydrogen sulfide. The example adsorber 18 is thus disposable and replaceable.

[0035] Another example of the adsorber assembly 10 uses a rare earth element in a granular form to remove hydrogen sulfide from the reformat gas stream 34. The granular form is used instead of the layer 48 on the base 46. The granular form may be held within a container (not shown) in the housing 22. The granular form of the rare earth element may induce a significant pressure drop within an adsorber assembly due to the granular form blocking flow through the adsorber assembly 10.

[0036] Features of the disclosed examples include using a rare earth oxide to remove hydrogen sulfide from reformat gas. The example rare earth oxides are chemically stable for hydrogen sulfide adsorption at temperatures near 800°C and these adsorber materials can be used for reformat gas clean-up at temperatures higher than about 500°C. Thus, another feature of the disclosed examples includes removing hydrogen sulfide from reformat gas without requiring significant temperature changes to the reformat gas. Still other features of the disclosed examples include eliminating the need for heat exchangers or complicated piping and fittings when moving reformat gas to a fuel cell. Another feature is substantially eliminating the possibility of coke formation and deposition using a low weight and low volume assembly which minimizes heat losses and essentially maintains the gas stream temperature near constant at the desired temperature level to eliminate the possibility of coke formation.

[0037] The preceding description is exemplary rather than limiting in nature. Variations and modifications to the disclosed examples may become apparent to those skilled in the art that do not necessarily depart from the essence of this disclosure. Thus, the scope of legal protection given to this disclosure can only be determined by studying the following claims.

CLAIMS

We claim:

1. An adsorber assembly, comprising:

an adsorber; and

a housing extending between an inlet and an outlet, the housing configured to communicate a reformat gas through at least a portion of the adsorber, wherein the adsorber includes a material coating selected from a group consisting of cerium oxide, a rare earth oxide, manganese oxide and combinations thereof, the material coating having a thickness and being configured to react with hydrogen sulfide throughout the thickness.

2. The adsorber assembly of claim 1, wherein the rare earth oxide comprises lanthanum.

3. The adsorber assembly of claim 1, wherein the rare earth oxide is selected from the group of, praseodymium, neodymium, samarium, and gadolinium and a combination thereof.

4. The adsorber assembly of claim 1, wherein the adsorber comprises pores configured to convect gas flow.

5. The adsorber assembly of claim 4, wherein the pores have a rectangular cross-section.

6. The adsorber assembly of claim 4, wherein the pores extend along an axis aligned with a longitudinal axis of the housing.

7. The adsorber assembly of claim 1, wherein the adsorber is configured to operate in a temperature range of 500° C to 800° C.

8. The adsorber assembly of claim 1, wherein the adsorber comprise a cordierite base.

9. The adsorber assembly of claim 8, wherein the material is washcoated on the cordierite base.

10. The adsorber assembly of claim 1, wherein the material coating includes cerium oxide and a rare earth oxide.

11. The adsorber assembly of claim 1, wherein the material coating includes cerium oxide, the rare earth oxide, and manganese oxide.

12. A fuel cell adsorber arrangement comprising:

a reformer;

a fuel cell stack; and

an adsorber that includes a layer of material selected from a group consisting of cerium oxide, lanthanum oxide, manganese oxide and combinations of these, wherein a reformat gas communicates between the reformer and the fuel cell stack through the trap, the reformat gas communicated from the trap having less hydrogen sulfide than the reformat gas communicated to the trap due to reactions with the layer of material, wherein the layer of material has a thickness and is configured to react with hydrogen sulfide throughout the thickness.

13. The fuel cell adsorber arrangement of claim 12, wherein the reformer is a catalytic autothermal reformer.

14. The fuel cell adsorber arrangement of claim 12, wherein the fuel cell stack comprises a multiple of individual solid oxide fuel cell assemblies.

15. The fuel cell adsorber arrangement of claim 12, wherein the hydrogen sulfide concentration in the reformat gas communicated from the trap is less than 500 parts per billion by volume.

16. A method of conditioning a reformat gas stream comprising:

communicating a reformat gas stream through an adsorber;

adsorbing hydrogen sulfide from the reformat gas stream using a layer of material within the adsorber, the layer of material selected from a group consisting of cerium oxide, a rare earth oxide, manganese and combinations of these, the adsorber configured such that the entire thickness of the layer of material is able to absorb hydrogen sulfide; and

communicating the reformat gas stream to a fuel cell stack.

17. The method of claim 16, wherein the rare earth oxide comprises a lanthanum oxide.

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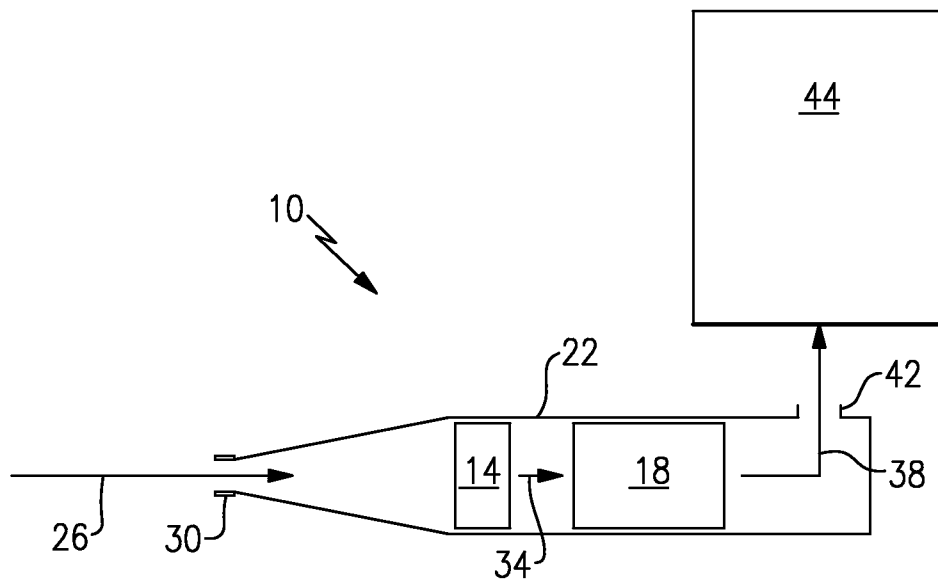


FIG. 1

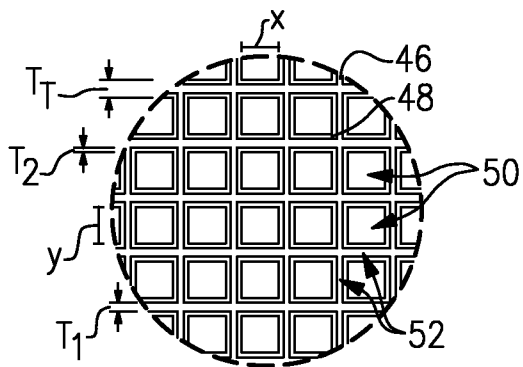


FIG. 2A

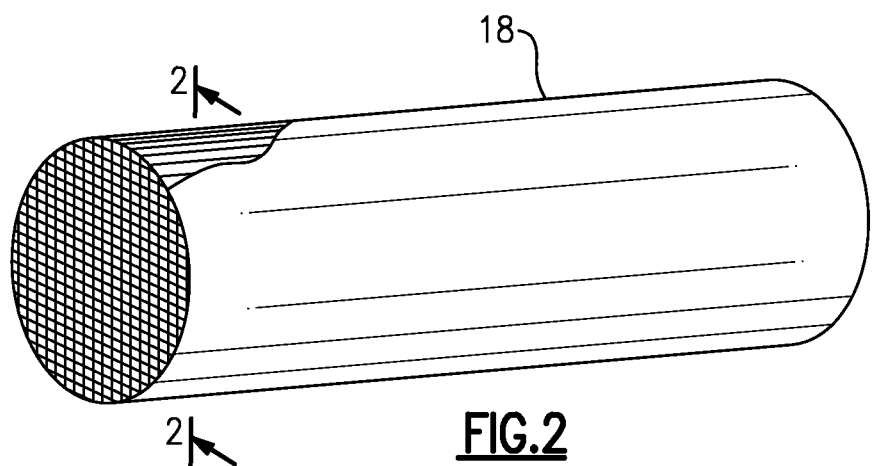


FIG. 2

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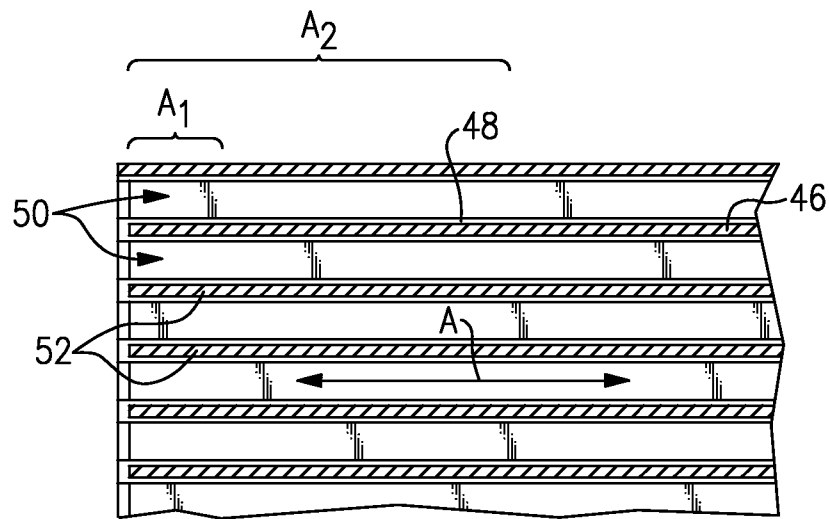


FIG. 2B

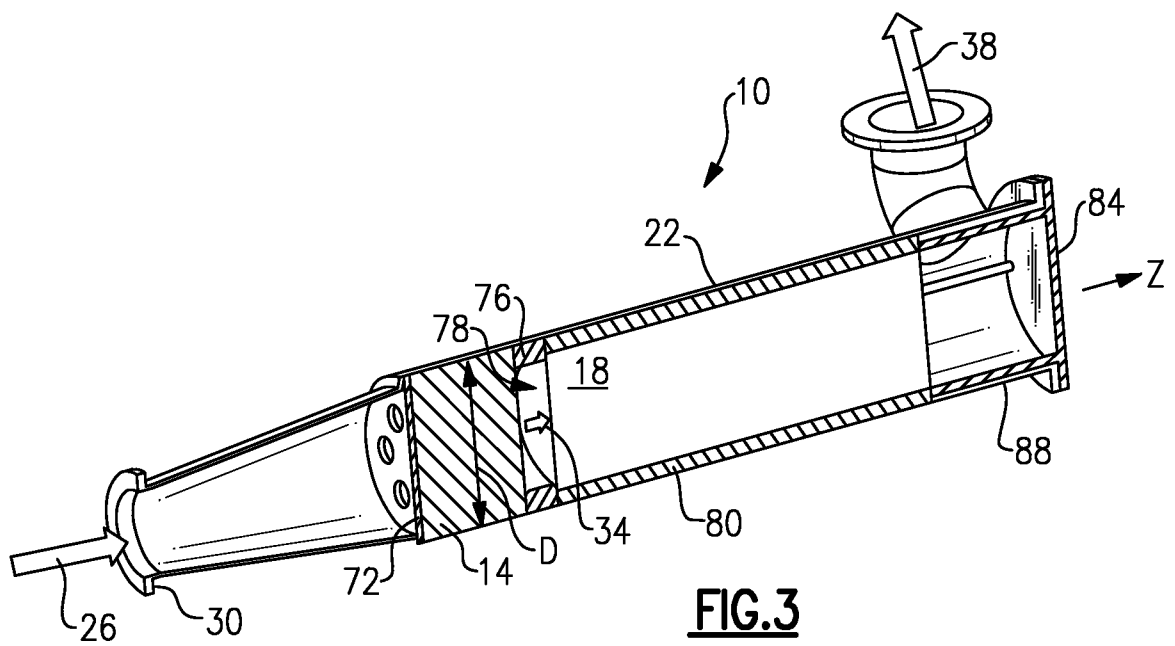


FIG. 3

A. CLASSIFICATION OF SUBJECT MATTER**H01M 8/06(2006.01)i, H01M 8/04(2006.01)i, B01D 53/04(2006.01)i, B01J 20/02(2006.01)i**

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)

H01M 8/06; B01D 53/02; B01D 24/00; H01M 8/04; C01F 17/00; F01N 3/00; B01D 53/48; B32B 3/12; B01J 8/04; B01J 35/02

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

Korean utility models and applications for utility models

Japanese utility models and applications for utility models

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

eKOMPASS(KIPO internal) & Keywords: hydrogen sulfide, adsorber, fuel cell, reformate.

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	US 2008-0210090 A1 (ZUBERI, BILAL) 04 September 2008	1-2, 4-7, 12, 14-17
Y	See abstract, figures 5-14, paragraphs [0047]-[0080], claims 1-16.	3, 8-11, 13
Y	US 2008-0267848 A1 (STEPHANOPOULOS, MARIA F. et al.) 30 October 2008 See abstract, figures 19-37, paragraphs [0095]-[0325], claims 74-75.	3, 10-11
Y	US 2005-0032640 A1 (HUANG, HE et al.) 10 February 2005 See abstract, figures 1-5, paragraphs [0003]-[0026], claims 9-16.	8-9
Y	US 2007-0012028 A1 (WEISSMAN, WALTER et al.) 18 January 2007 See abstract, figures 1-4, paragraphs [0063]-[0092], claims 1-4.	13
X	US 2002-0136936 A1 (GRIEVE, MALCOLM JAMES et al.) 26 September 2002 See abstract, figures 3-5, paragraphs [0036]-[0043], claims 1-20.	1, 12, 16



Further documents are listed in the continuation of Box C.



See patent family annex.

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Date of the actual completion of the international search

14 SEPTEMBER 2011 (14.09.2011)

Date of mailing of the international search report

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INTERNATIONAL SEARCH REPORT

Information on patent family members

International application No.

PCT/US2010/053921

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