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(54) Title: DISTRIBUTIVELY COOLED, INTEGRATED WATER-GAS SHIFT REACTOR AND VAPORIZER

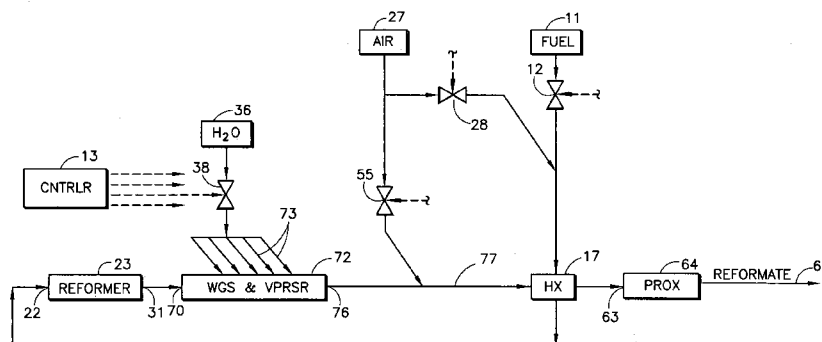


FIG.2

(57) Abstract: A hydrocarbon fuel processor includes a reformer (23) providing hydrogen-rich reformat gas to an integrated water-gas shift reactor (WGS) and vaporizer (72), the outlet of which is provided to a preferential oxidizer (64) so as to provide hydrogen-rich reformat gas in a conduit (67) which is low in carbon dioxide. The integrated WGS/vaporizer (72) may have a plurality of liquid water inlets (73) dispersed along substantially the entire length of the catalyst bed (83); or liquid water may be provided in a porous tube (86, 94) or perforated tube (95) separated from catalyst by a screen (89), along the length of the reactor. The temperature can be controlled to an optimum profile, high at the inlet and decreasing along the length of the reactor as steam is generated for the water-gas shift reaction. The catalyst may be separated from inlets (73) by inert material (95). The tube may have a trefoil configuration (94).

Distributively Cooled, Integrated Water-Gas Shift Reactor and Vaporizer

Technical Field

[0001] Liquid water is introduced in a controlled manner along the entire length of a water-gas shift reactor catalyst bed, simultaneously cooling the reactor and generating the steam necessary for the shift reaction to occur.

Background Art

[0002] The production of hydrogen-rich reformat gas from a variety of hydrocarbon-containing feedstocks typically involves reformation of the feedstock to provide hydrogen-rich reformat gas, which also includes an objectionable amount of carbon monoxide, some carbon dioxide (which is benign in most hydrogen utilizations), some unreformed hydrocarbon and small amounts of other gases. The reformation is usually followed by water-gas shift in which some of the steam and carbon monoxide are converted to hydrogen and carbon dioxide, thereby increasing the good (H₂) and decreasing the bad (CO). There may be two or more shift reactors in series. The shift reactions may be followed by catalytic preferential oxidization reactions (PROX) in which carbon monoxide is preferentially oxidized to carbon dioxide, while at the same time only a small amount of hydrogen may be converted to water.

[0003] One exemplary form of a fuel processor described above is illustrated in Fig. 1. Fuel, which is a hydrocarbon-containing feedstock, is fed from a source 11 at a rate regulated by a valve 12 in response to a controller 13. The fuel then is heated in a first heat exchanger 17, where the temperature of the fuel cell is increased to a first temperature of between about 100°C (210°F) and about 150°C (300°F) and then through another heat exchanger 18 to warm the fuel to a second temperature which may be between about 250°C (480°F) and about 300°C (570°F). Fuel then passes through a conduit 21 to the inlet 22 of a reformer 23. Air from a source 27 (which may be ambient) is mixed at the inlet 22 at a rate regulated by a valve 28 in response to the controller 13. At the same time, water may also be injected at the reformer inlet. The reformer provides

the reformat gas in a conduit 31 to the inlet 32 of a high temperature water-gas shift reactor 33 (WGS). Water from a source 36 is mixed with the reformat at the inlet 32 as regulated by a valve 38 in response to the controller 13. The output of the high temperature water-gas shift reactor in a conduit 41 is passed through the heat exchanger 18 to an inlet 43 of a low temperature water-gas shift reactor 44, and then passed in a conduit 47 through the heat exchanger 17.

[0004] The reformat, having been cooled sufficiently by the incoming fuel in the heat exchangers 17, 18, is passed over a conduit 50 to the inlet 51 of a first PROX 52. Air is introduced at the inlet 51 in a manner regulated by a valve 53 in response to the controller 13.

[0005] From the first PROX 52, the reformat is passed through a conduit 55 to the inlet 56 of a heat exchanger 57. Air is mixed with the reformat at the inlet 56 as regulated by a valve 60 in response to the controller 13. From the heat exchanger 57, the reformat is passed through a conduit 62 to the inlet 63 of a second PROX 64, the output of which comprises the useful reformat in a conduit 67, rich in hydrogen and relatively low in carbon monoxide.

[0006] In the water-gas shift reactors 33, 44, the amount of carbon monoxide in the product decreases as the temperature decreases, but the activity of the water-gas shift catalysts at lower temperatures (such as below 250°C (480°F) is relatively low. This equilibrium limitation requires very careful control over the operating temperature of the water-gas shift reactors, which increases the cost and volume of the fuel processor system. Typical carbon monoxide mitigation, involving two shift reactors 33, 44 and two preferential oxidizers 52, 64, can leave as much as 0.5% carbon monoxide in the reformat. Since current PROX catalysts are not sufficiently selective toward oxidizing CO, hydrogen is also oxidized in an exothermic process which creates temperature problems. The control over the air to carbon monoxide ratio in the PROX is therefore absolutely critical. If the WGS temperature and the PROX air/CO ratios are carefully controlled, a carbon monoxide level between about 0.002% and about 0.0005% could be achieved. However, lack of control in any of the shift reactors or PROXs could result in over cooling and insufficient CO removal,

or undercooling with thermal runaway and the back conversion of carbon dioxide and hydrogen into carbon monoxide and water. Thermal runaway also causes deactivation of high temperature, non-oxidizable water-gas shift catalysts (at temperatures above about 350°C (660°F)).

[0007] By decreasing the carbon monoxide concentration exiting the second water-gas shift reactor 44 from about 0.5% CO to about 0.3% CO, the first stage PROX reactor 52 could be eliminated, thereby decreasing cost, volume and weight, while increasing efficiency. However, the equilibrium limitation for conventional water-gas shift reactions prevent reaching such a low level of carbon monoxide. To achieve that, the WGS would have to be operated with a temperature much lower than the optimum temperature. This significantly reduces the CO removal rate because the rate of reaction decreases exponentially with temperature reduction, thereby requiring a much larger WGS. A larger WGS will dramatically increase, particularly if the WGS catalyst contains noble metal.

Summary

[0008] To mitigate the foregoing problems, liquid water is introduced in a controlled manner along substantially the entire length of the water-gas shift reactor catalyst, thereby cooling the reactor as well as increasing the rate of reaction, which is nearly proportional to the concentration of water. Either water or steam will shift the equilibrium favorably, thereby permitting a reduction in reactor volume. Through proper injection of water an ideal temperature profile in the bed (which is a function of the specific catalyst kinetics) can be achieved, thus maximizing CO conversion for a given amount of catalyst.

[0009] Because the actively cooled WGS operates at a significantly lower temperature, rapid deactivation of the WGS catalyst is avoided. Since the catalyst, operating at lower temperatures, does not rapidly deactivate, extra catalyst required in order to have enough catalyst after a life cycle (which could be on the order of 40,000 hours) is no longer necessary. This in turn allows an additional reduction in size, reaching sizes of about half of conventional water-gas shift reactors. This

significant reduction in size results in significant savings in the cost of the catalyst, as well as reduction in volume and weight.

[0010] Variations will be apparent in the light of the following detailed description of exemplary embodiments, as illustrated in the accompanying drawings.

Brief Description of the Drawings

[0011] Fig. 1 is a simplified, schematic block diagram of an exemplary form of a reformat generating, hydrocarbon fuel processor known to the art.

[0012] Fig. 2 is a simplified, schematic block diagram of a fuel processor employing a distributively cooled, water-gas shift reactor.

[0013] Fig. 3 is a simplified, stylized illustration of an exemplary structure for distributing liquid water along the length of the catalyst bed in a WGS.

[0014] Fig. 4 is a sectioned, front elevation view of an alternative embodiment of a distributively cooled, integrated water-gas shift reactor and vaporizer.

[0015] Fig. 5 is a front elevation section of a trefoil porous tube for use in the embodiment of Fig. 4.

[0016] Fig. 6 is a simplified perspective view of a plurality of casings 90 containing apparatus of the type illustrated in Figs. 4 and 5.

[0017] Fig. 7 is a front elevation section of another embodiment.

[0018] Fig. 8 is a fragmentary, front elevation section of still another embodiment.

Mode(s) of Implementation

[0019] Referring now to Fig. 2, reformat at the outlet 31 of the reformer 23 is fed to an inlet 70 of a water-gas shift reactor and vaporizer (WGS/vaporizer) 72 which has a plurality of liquid water inlets 73 receiving liquid water through the valve 38 from the source of water 36. The liquid water is introduced throughout substantially the entire length of the water-gas shift catalyst bed which cools the catalyst, vaporizing the liquid water and providing additional steam for the reaction. The temperature of the

process is controlled to achieve an optimum temperature profile, which starts as a high temperature at the inlet 70 and decreases from the beginning to the end of the catalyst bed, rather than increasing along the length of the catalyst bed, as in the prior art. The optimum temperature profile is provided within a narrow range, simply by controlling the amount of water provided to the WGS/vaporizer 72. From an outlet 76, reformat is passed through a conduit 77 to the heat exchanger 17 and thence to the second PROX 64 for reduction of carbon monoxide and increase of hydrogen, as described hereinbefore.

[0020] One simple form of implementing a WGS/vaporizer 72 is illustrated in Fig. 3. Therein, the water inlets 73 are distributed along casing 74 of the WGS/vaporizer 72 so as to introduce liquid water directly to the catalyst 83. The inlets may be on two or more sides of the reactor, or may be in a spiral pattern around the reactor. The catalyst 83 is thereby cooled mostly by evaporation, the resulting steam being taken up in the water-gas shift process. Sensible heat cooling also occurs throughout the length of the WGS/vaporizer 72. Thus, the water-gas shift reaction is distributively cooled and the temperature is controlled to an optimum profile throughout the length of the bed of catalyst 83.

[0021] In Fig. 4, the water inlet 73 feeds water into a porous tube 86 that is suspended by separators 87, such as ribs or a spiral outer wall configuration, so as to be separated from a screen 89 which could be wire mesh or a perforated plate. The porous tube 86 introduces water into the reaction along the whole length of the reactor 72a. The screen prevents the catalyst 83 from contacting the surface of the porous tube 87, which may have water on it. Although a single inlet 70 is shown for the untreated reformat, a plurality of inlets may be used, or a manifold may be fashioned to suit any implementation of shift reactor and vaporizer.

[0022] This arrangement precludes catalyst spalling and loss which may occur as a result of direct contact between catalyst and liquid water (rather than steam). In the embodiment of Fig. 4, the porous tube 86 will become very hot due to the exothermic reaction between steam and the untreated reformat at the catalyst. This will vaporize the outermost film of water, the pressure of water and pressure of vaporization contributing to

cause the steam to pass through the porous tube plate 86 into the space 92 between the tube 86 and the screen 89. Any water which may pass through the porous tube 86 will vaporize in the space 92, prior to passing through the screen to mix with the untreated reformat at the catalyst.

[0023] It has been determined that for a 150kW fuel cell power plant, about one square meter of external tube surface would be required. In order to reduce the necessary length of the perforated tube and screen arrangement as in Fig. 4, the shape of the tube could be altered in a variety of fashions in order to present more surface area per linear foot. One possibility is illustrated by a tube 94 in Fig. 5 which has a trefoil configuration.

[0024] In order to reduce length, a plurality of arrangements such as that illustrated in Fig. 4, with or without an altered configuration of the tube 86, 94, has a plurality of tube and screen combinations within a plurality of casings 74 configured together, as shown in Fig. 6, in order to achieve the required amount of external tube surface for the desired flow of reformat, at the temperature of the tube maintained by the heat of the exothermic reaction.

[0025] Another embodiment of reactor 72b, shown in Fig. 7, utilizes a tube 95 with small perforations therein which introduces liquid water along the length of the WGS/vaporizer 72, causing atomized mists 96 of water to enter the space 92 between the tube 95 and the screen 89; the mist quickly vaporizes due to the temperature of the space 92.

[0026] A simpler embodiment of reactor 72c, shown in Fig. 8, avoids direct contact of liquid water on the catalyst with a small amount of temperature-resistant inert material 98 such as porous ceramic, adjacent each water inlet 73. The water is therefore vaporized by the heat of the inert material and reaches the catalyst 83 as steam.

Claims

1. A method comprising:

(a) providing a water-gas shift reactor (72) of a given length containing a bed of water-gas shift reaction catalyst (83) extending along substantially the length of the reactor; and

5 (b) flowing reformat gas containing at least hydrogen and carbon monoxide through the catalyst;

characterized by:

(c) introducing liquid water along substantially the length of the reactor.

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2. A method according to claim 1 further characterized in that:

said step (c) includes vaporizing said water with the heat of the steam/reformat reaction prior to contact with the catalyst.

3. A method according to claim 2 further characterized in that:

said step (c) includes vaporizing said water within a porous tube which extends along the length of the reactor.

4. A method according to claim 2 further characterized in that:

said step (c) includes misting said water through apertures in a tube which extends along the length of the reactor.

5. A method according to claim 1 further characterized in that:

said step (c) comprises injecting water into said catalyst bed along substantially the length of the reactor.

6. Apparatus comprising:

a water-gas shift reactor (72) of a given length containing a bed of water-gas shift reaction catalyst (83) extending along substantially the length of the reactor, said reactor configured (23, 70) to flow hydrogen-rich reformat gas containing at least hydrogen and carbon monoxide through the catalyst;

5

characterized by:

said water-gas shift reactor configured to receive liquid water (36) and introduce (73) liquid water along substantially the length of the water-gas shift reactor.

10

7. Apparatus according to claim 6 further characterized by: the reactor (72) having a permeable tube (86, 94, 95) extending substantially the length of the reactor and surrounded by the catalyst (83).

8. Apparatus according to claim 7 further characterized by: the catalyst (83) being held within a screen (89) separated a small distance from the tube (86, 94, 95).

9. Apparatus according to claim 8 further characterized in that: said screen (89) is wire mesh.

10. Apparatus according to claim 7 further characterized in that: said tube (86, 94) is porous to allow vaporized water to pass therethrough and reach the catalyst (83) as steam.

11. Apparatus according to claim 7 further characterized in that: said tube (96) has small perforations which atomize the water into a mist before reaching the catalyst (83).

12. Apparatus according to claim 7 further characterized in that: a plurality of liquid water inlets (73) disposed along substantially the length of the reactor (72) introduce liquid water along substantially the length of the reactor.

13. Apparatus according to claim 12 further characterized in that: the catalyst (83) is separated from liquid water at the water inlets (73) by inert material (98).

14. Apparatus according to claim 13 further characterized in that:
the inert material (98) is ceramic.

AMENDED CLAIMS

received by the International Bureau on 23 October 2009 (23.10.09)

Claims

1. A method of cooling and increasing the rate of reaction in a water-gas shift reactor, comprising:
 - (a) providing a water-gas shift reactor (72) of a given length containing a bed of water-gas shift reaction catalyst (83) extending along substantially the length of the reactor; and
 - (b) flowing reformat gas containing at least hydrogen and carbon monoxide through the catalyst;
characterized by:
 - (c) introducing liquid water along substantially the length of the reactor.
2. A method according to claim 1 further characterized in that:
said step (c) includes vaporizing said water with the heat of the steam/reformat reaction prior to contact with the catalyst.
3. A method according to claim 2 further characterized in that:
said step (c) includes vaporizing said water within a porous tube which extends along the length of the reactor.
4. A method according to claim 2 further characterized in that:
said step (c) includes misting said water through apertures in a tube which extends along the length of the reactor.
5. A method according to claim 1 further characterized in that:
said step (c) comprises injecting water into said catalyst bed along substantially the length of the reactor.
6. Apparatus comprising:
a water-gas shift reactor (72) of a given length containing a bed of water-gas shift reaction catalyst (83) extending along substantially the length of the reactor, said reactor configured (23, 70) to flow hydrogen-rich reformat gas containing at least hydrogen and carbon monoxide through the catalyst;

characterized by:

said water-gas shift reactor configured to receive liquid water (36) and introduce (73) liquid water along substantially the length of the water-gas shift reactor.

7. Apparatus according to claim 6 further characterized by:
the reactor (72) having a permeable tube (86, 94, 95) extending substantially the length of the reactor and surrounded by the catalyst (83).

8. Apparatus according to claim 7 further characterized by:
the catalyst (83) being held within a screen (89) separated a small distance from the tube (86, 94, 95).

9. Apparatus according to claim 8 further characterized in that:
said screen (89) is wire mesh.

10. Apparatus according to claim 7 further characterized in that:
said tube (86, 94) is porous to allow vaporized water to pass therethrough and reach the catalyst (83) as steam.

11. Apparatus according to claim 7 further characterized in that:
said tube (96) has small perforations which atomize the water into a mist before reaching the catalyst (83).

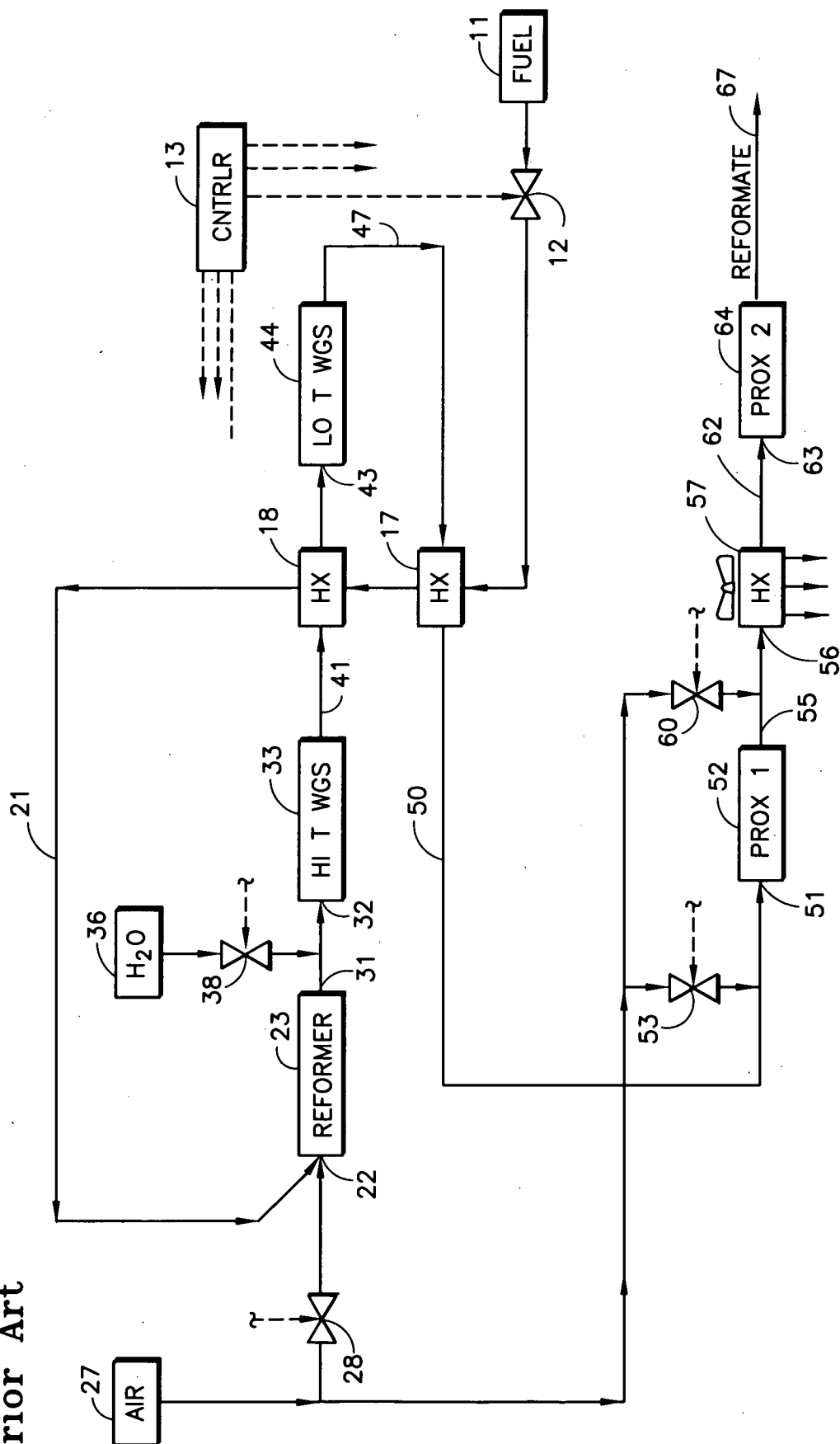
12. Apparatus according to claim 7 further characterized in that:
a plurality of liquid water inlets (73) disposed along substantially the length of the reactor (72) introduce liquid water along substantially the length of the reactor.

13. Apparatus according to claim 12 further characterized in that:
the catalyst (83) is separated from liquid water at the water inlets (73) by inert material (98).

14. Apparatus according to claim 13 further characterized in that:
the inert material (98) is ceramic.

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FIG. 1
Prior Art



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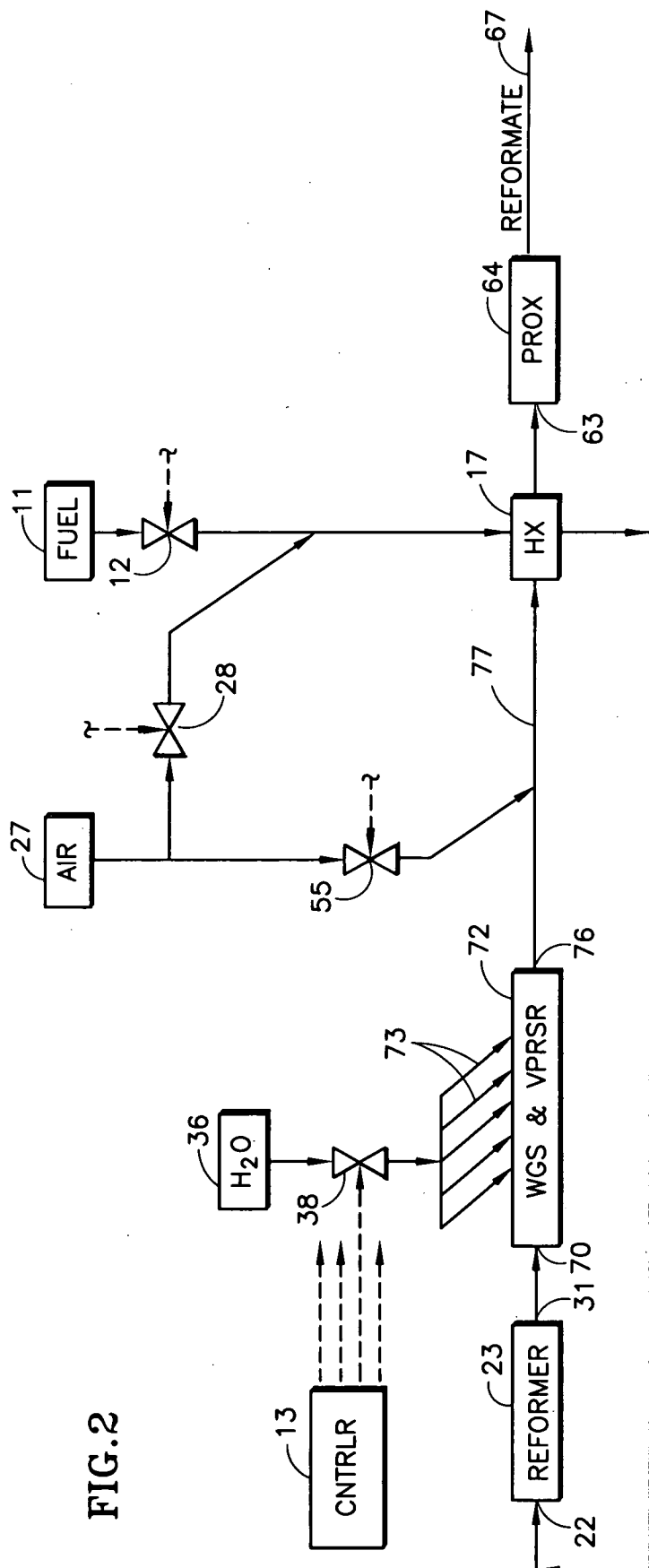


FIG.2

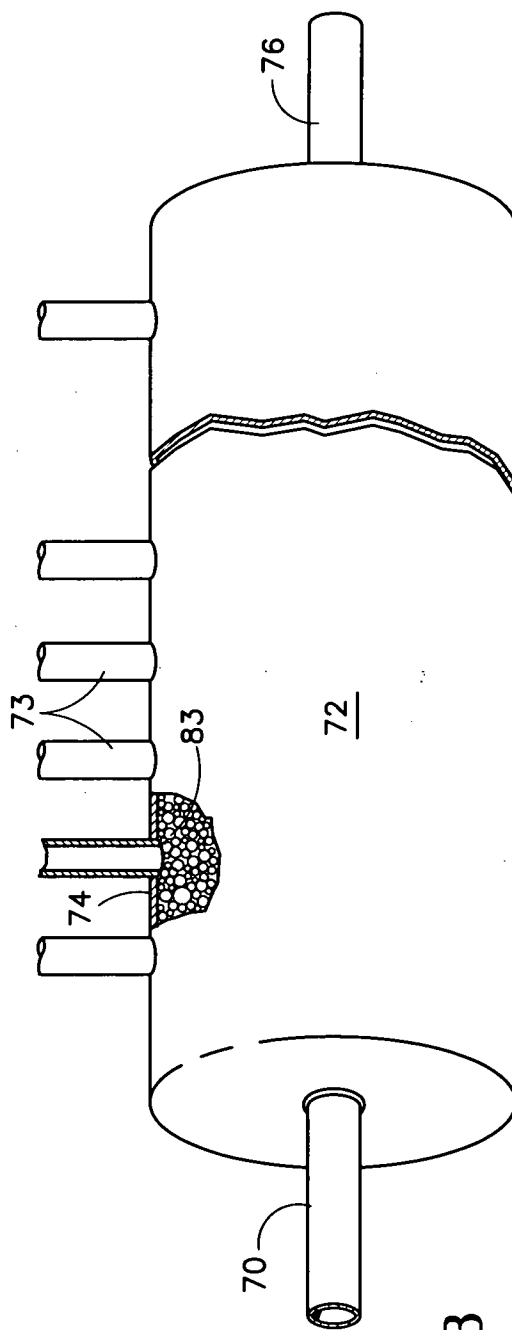


FIG.3

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FIG. 4

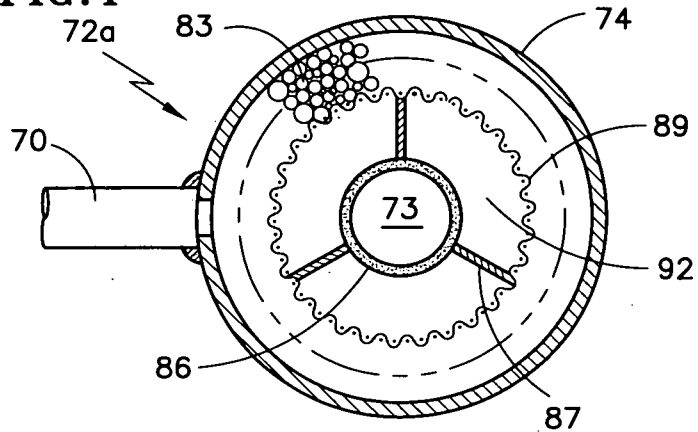
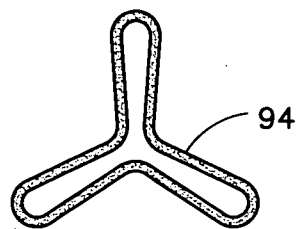


FIG. 5

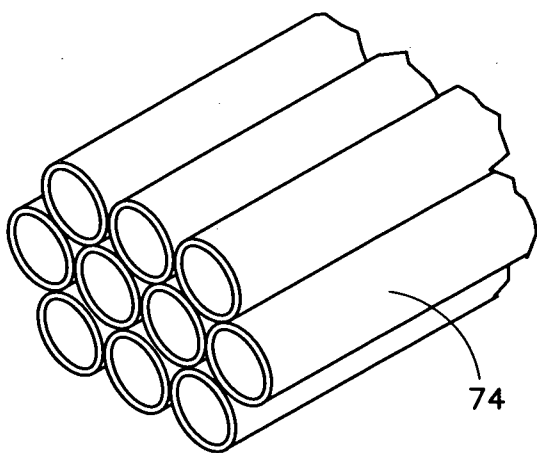


FIG. 6

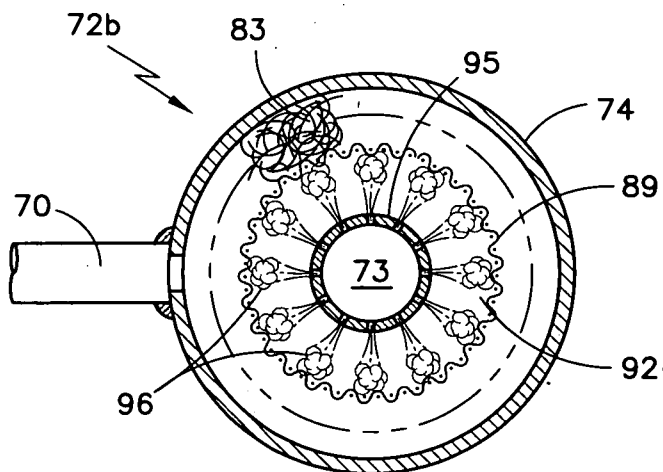


FIG. 7

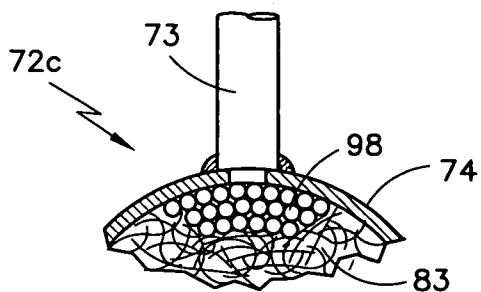


FIG. 8

A. CLASSIFICATION OF SUBJECT MATTER

B01J 8/08(2006.01)i, B01J 8/04(2006.01)i, B01J 12/00(2006.01)i, C01B 3/02(2006.01)i, C01B 3/38(2006.01)i, C01B 3/32(2006.01)i, C01B 3/48(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)

IPC B01J 8/08

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)

eKOMPASS (KIPO internal) water-gas, shift, reactor, cooling, catalyst, hydrocarbon, fuel and similar terms

C. DOCUMENTS CONSIDERED TO BE RELEVANT

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A	US 07220392 B2 (Xiaoyang Rong and Brian E. Cheadle) 22 May 2007 See the whole document	1 - 14
A	US 2006-0045828 A1 (Timothy Mark Aaron et al.) 02 March 2006 See the whole document	1 - 14
A	WO 2004-022480 A2 (Shell Oil Company and Miglin, Maria, Therese) 18 March 2004 See the whole document	1 - 14
A	WO 2002-0045836 A1 (Texaco Development Corporation) 13 June 2002 See the whole document	1 - 14



Further documents are listed in the continuation of Box C.



See patent family annex.

* Special categories of cited documents:

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"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

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"&" document member of the same patent family

Date of the actual completion of the international search

30 SEPTEMBER 2009 (30.09.2009)

Date of mailing of the international search report

30 SEPTEMBER 2009 (30.09.2009)

Name and mailing address of the ISA/KR



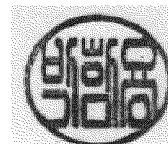
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INTERNATIONAL SEARCH REPORT

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