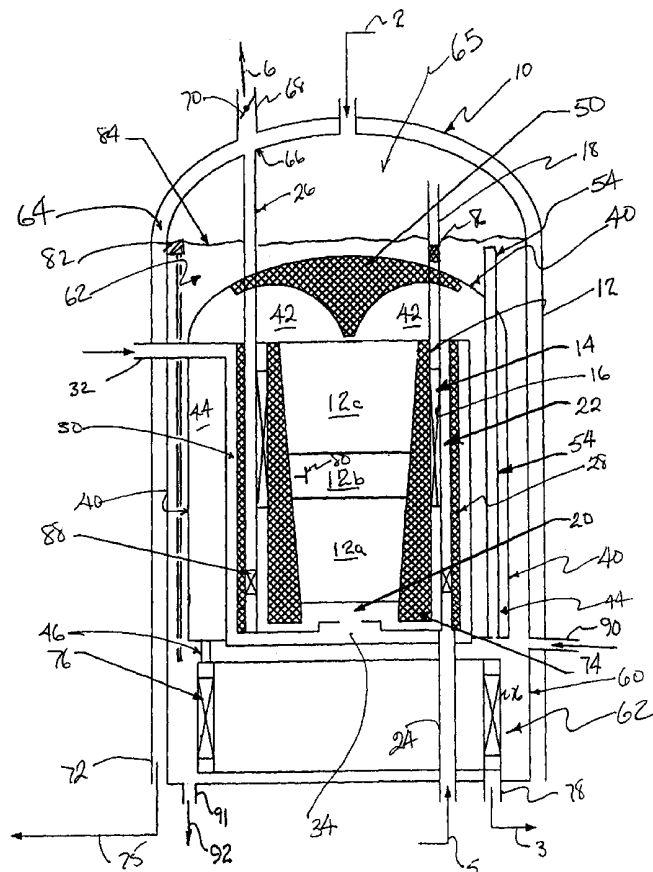




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(54) Title: APPARATUS FOR PRODUCING HYDROGEN



(57) Abstract: A fuel processor apparatus for converting a hydrocarbon or an oxygenate feed stream to produce a fuel stream for a fuel cell is provided wherein the heat required to sustain the reforming reaction and the cooling required to moderate and direct high and low-temperature shift reactions occur in a thermosiphon boiler vessel (60). The thermosiphon boiler vessel (60) provides an essentially isothermal steam generator and heat removal system which improves the stability of the integrated reaction zone by providing a uniform temperature profile and strategic temperature control.



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APPARATUS FOR PRODUCING HYDROGEN

BACKGROUND OF THE INVENTION

This invention relates to reactors for the production of hydrogen used in conjunction with a fuel cell for the production of electricity. More particularly, the invention relates to an integrated reactor for converting a hydrocarbon or alcohol to produce a fuel stream for an electrochemical reaction zone.

Systems for contacting solids and particulate material are well known and routinely employed in the processing of gases, the production of chemicals, and the refining of petroleum. The particulate materials in most cases comprise catalysts or adsorbents and the process streams are gaseous or liquid mixtures of reactants, product or streams undergoing separation.

One particularly well-known method of contacting particulate material with a fluid stream retains the particulate solid material as a bed of particulate material through which the fluid stream passes. A multitude of arrangements with various bed geometries is known for contacting the particulate material with the fluid streams. Such arrangements include radial flow beds where particulate solids are retained in an annular ring or downflow or upflow beds where fluid streams pass through a cylindrical bed or laminar bed of particulate solids.

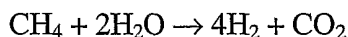
The use of fuel cells to generate electrical power for electricity or to drive a transportation vehicle relies upon the generation of hydrogen. Because hydrogen is difficult to store and distribute, and because hydrogen has a low volumetric energy density compared to fuels such as gasoline, hydrogen for use in fuel cells will have to be produced at a point near the fuel cell, rather than be produced in a centralized refining facility and distributed like gasoline. To be effective, hydrogen generation for fuel cells must be smaller, simpler, and less costly than hydrogen plants for the generation of industrial gasses. Furthermore, hydrogen generators for use with fuel cells will have to be integrated with the operation of the fuel cell and be sufficiently flexible to efficiently provide a varying amount of hydrogen as demand for electric power from the fuel cell varies.

Hydrogen is widely produced for chemical and industrial purposes by converting materials such as hydrocarbons and methanol in a reforming process to produce a synthesis gas. Such production usually takes place in large facilities which are rarely turned down in production for even a few days per year. In addition, the operation of the industrial hydrogen production facilities is often integrated with associated facilities to improve the use of energy for the overall complex. Synthesis gas is the name generally given to a gaseous mixture principally comprising carbon monoxide and hydrogen, but also possibly containing carbon dioxide and minor amounts of methane and nitrogen. It is used, or is potentially useful, as feedstock in a variety of large-scale chemical processes, for example: the production of methanol, the production of gasoline boiling range hydrocarbons by the Fischer-Tropsch process and the production of ammonia.

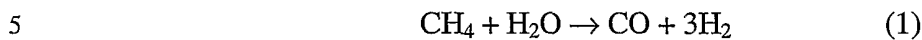
Processes for the production of synthesis gas are well known and generally comprise steam reforming, autothermal reforming, non-catalytic partial oxidation of light hydrocarbons or non-catalytic partial oxidation of any hydrocarbons. Of these methods, steam reforming is generally used to produce synthesis gas for conversion into ammonia or methanol. In such a process, molecules of hydrocarbons are broken down to produce a hydrogen-rich gas stream. A paper titled "Will Developing Countries Spur Fuel Cell Surge?" by Rajindar Singh, which appeared in the March 1999 issue of *Chemical Engineering Progress*, page 59-66, presents a discussion of the developments of the fuel cell and methods for producing hydrogen for use with fuel cells. The article particularly points out that the partial oxidation process is a fast process permitting small reactors, fast startup, and rapid response to changes in the load, while steam reforming is a slow process requiring a large reactor and long response times, but operates at a high thermal efficiency. The article highlights one hybrid process which combines partial oxidation and steam reforming in a single reaction zone as disclosed in US-A-4,522,894.

Modifications of the simple steam reforming processes have been proposed to improve the operation of the steam reforming process. In particular, there have been suggestions for improving the energy efficiency of such processes in which the heat available from the products of a secondary reforming step is utilized for other purposes within the synthesis gas production process. For example, processes are described in US-A-4,479,925 in which heat from the products of a secondary reformer is used to provide heat to a primary reformer.

The reforming reaction is expressed by the following formula:



where the reaction in the reformer and the reaction in the shift converter are respectively expressed by the following simplified formulae (1) and (2):



In the water gas shift converter which typically follows a reforming step, formula (2) is representative of the major reaction.

10 US-A-4,925,456 discloses a process and an apparatus for the production of synthesis gas which employs a plurality of double pipe heat exchangers for primary reforming in a combined primary and secondary reforming process. The primary reforming zone comprises at least one double-pipe heat exchanger-reactor and the primary reforming catalyst is positioned either in the central core or in the annulus thereof. The invention is further characterized in that the secondary reformer effluent is
15 passed through which ever of the central core or the annulus is not containing the primary reforming catalyst counter-currently to the hydrocarbon-containing gas stream.

US-A-5,181,937 discloses a system for steam reforming of hydrocarbons into a hydrogen rich gas which comprises a convective reformer device. The convective reformer device comprises an outer shell enclosure for conveying a heating fluid
20 uniformly to and from a core assembly within the outer shell. The core assembly consists of a multiplicity of tubular conduits containing a solid catalyst for contacting a feed mixture open to the path of the feed mixture flow such that the path of the feed mixture flow is separated from the heating fluid flow in the outer shell. In the process, an autothermal reformer fully reforms the partially reformed (primary reformer) effluent
25 from the core assembly and supplies heat to the core assembly by passing the fully reformed effluent through the outer shell of the convective reforming device.

WO 99/36351 discloses a reformer reactor for producing a hydrogen-rich gas which includes four sequentially adjacent reaction zones and a product gas collection space. The reformer reactor comprises a reactor geometry such that the flow path is
30 directed in diverging radial directions away from the first reaction zone and through the subsequent zones. In addition the reactor is provided with a heat exchange means

disposed in one of the downstream reaction zones to use heat developed in the reaction zones to preheat the feed stream. This heat exchange means is employed to regulate the heat exchange in the reactor to achieve a desired temperature in the catalyst zone. The heat exchange means disclosed in WO 99/36351 comprises helical tube sections
5 disposed in the catalyst zones to the feed stream.

WO 99/36351 and similar axial flow arrangements based on locating adjacent reaction zones in a manner to facilitate heat exchange and then disposing within those reaction zones coils of heat exchanger tubes suffer from the practical problem of regulating and controlling the heat exchange in the coils and the mechanical stresses
10 generated in the heat exchange tubes. Simple axial flow systems employing double pipe heat exchange zones which rely on simple convection between adjacent zones are not able to remove a sufficient amount of heat because the heat exchange medium is typically limited to the gaseous reaction mixture which has a low thermal conductivity. In addition, the placement of coils within relatively narrow catalytic zones of these close-
15 coupled reaction systems can result in poor flow distribution of reactants and an unfavorable temperature profile in the direction perpendicular to the flow of the reactants. The equilibrium reactions presented herein above are sensitive to temperature and may exhibit a reversal when the temperature of the particular reaction zone falls outside an effective operating range. Furthermore, two phase flow conditions may arise
20 within the coils resulting in vibration and bumping which can result in mechanical failure of the coil and can raise safety issues related to hydrogen leaks and fire. Integrated reaction systems are sought which avoid the above-mentioned problems.

It is an objective of the present invention to provide compact fuel processor for use with fuel cell systems.

25 It is an objective of the present invention to provide a fuel processor reactor which provides a uniform temperature profile to avoid wide temperature swings and minimize mechanical and thermal stress.

It is an objective of the present invention to provide an isothermal heat medium having a heat capacity greater than the feed or the reactor products to strategically
30 control the temperature throughout the fuel processor zone.

SUMMARY OF THE INVENTION

The present invention is directed to a fuel processor to be used in conjunction with a fuel cell for the generation of electric power. The reactor portion of the fuel

processor comprises an arrangement wherein particulate solids are disposed in concentric annular catalyst zones and wherein the catalyst is retained in relatively narrow vertically extended annular flow channels within the concentric annular catalyst zones. The particulate solids remain in a fixed position and the fluids move through catalyst and heat transfer zones which are strategically arranged to avoid the use of heating coils and other heat transfer surfaces which might interrupt flow or create hot spots and dead zones in a fuel processor while maintaining the overall apparatus in a stable thermal condition. Strategically positioned thermosiphon tubes are used to define vertical flow channels for heat transfer fluids. This is particularly important in applications that require or benefit from heating or cooling of the particulate solids and fluids within the reaction zones to control the temperature of a reaction or other processing. In such arrangements, the vertical tubes provide a large area of heat transfer surface by which a heat transfer fluid may indirectly contact one surface of the tube while the other surface contacts the particulate solids and fluids. For example, indirect heat transfer in which a heat exchange fluid contacts a catalyst and a reaction fluid or a reaction fluid in combination with another fluid can be used to supply or withdraw the heat of reaction in an endothermic or exothermic process to establish isothermal conditions in the reaction zone.

In one embodiment, the present invention comprises an apparatus for generating hydrogen from a fuel stream for use in conjunction with a fuel cell. The apparatus comprises a first catalyst chamber which has an upper end, a lower end, a hollow interior containing an upper zone, a center zone, and a lower zone. A second catalyst chamber annularly surrounds the first catalyst chamber. At least a portion of the second catalyst chamber is in intimate thermal contact with the first catalyst chamber. The second catalyst chamber has an annular catalyst zone, a vertical feed conduit at a top end and a fluid distributor/mixer at a lower end. The fluid distributor/mixer is in fluid communication with the lower zone of the first catalyst chamber. A burner zone annularly surrounds the second catalyst chamber. The burner zone has a lower burner inlet and an upper flue gas outlet. The burner zone is in intimate thermal contact with the second catalyst chamber. An insulation zone is disposed annularly about the burner zone. An air pre-heater zone is disposed annularly about the insulation zone. The air pre-heater zone comprises an air inlet conduit and a pre-heated air outlet which is in fluid communication with the fluid distributor/mixer. A third catalyst chamber is annularly disposed on the air pre-heater zone and is disposed over the first catalyst chamber. The

third catalyst chamber has a top catalyst zone in fluid communication with the upper catalyst zone of the first catalyst chamber; a second annular catalyst zone in fluid communication with the top catalyst zone; a shift effluent conduit in fluid communication with the second annular catalyst zone; and an upper insulation zone
5 extending over the top catalyst zone. A plurality of steam conduits is disposed in the second annular catalyst zone. The plurality of steam conduits extends vertically through the second annular catalyst zone, the top catalyst zone, and the upper insulation zone. The third catalyst chamber is vertically disposed in a boiler vessel having a boiler interior. The plurality of steam conduits is in fluid communication with the boiler
10 interior. A flue gas exchange zone is disposed essentially fully enclosing the boiler vessel and in intimate thermal contact with the boiler vessel. The flue gas exchange zone has a hot gas inlet in fluid communication with the upper flue gas outlet of the burner zone, a flue gas vent, and a cooled gas outlet.

BRIEF SUMMARY OF THE DRAWINGS

15 FIG. 1 shows a vertical cross-sectional view of the present invention.

FIG. 2 shows a cross sectional view of the present invention from an overhead perspective.

FIG. 3 shows a burner zone detail.

DETAILED DESCRIPTION OF THE DRAWINGS

20 This invention applies to arrangements for the integration of highly exothermic and endothermic reaction zones into a single reaction zone with the ability to manage the heating and cooling requirements in a manner to provide a stable operation of the combination. The key to achieving a stable operation is the attainment of an essentially isothermal heat sink which provides a continuous source of cooling to a highly
25 exothermic reaction system while providing a continuous source of steam at a critical point in the process.

Referring to FIG. 1, the apparatus 10 for the generation of a hydrogen fuel stream from a feed stream comprising a hydrocarbon or an oxygenate is shown in cross section. The apparatus of the present invention comprises a first catalyst chamber 12. The first
30 catalyst chamber 12 has a hollow interior which has an upper zone 12c at an upper end, a lower zone 12a at a lower end and a center zone 12b between the upper zone 12c and the

lower zone 12a. The lower catalyst zone 12a contains a reforming catalyst or a mixture of a reforming catalyst and an inert catalyst layer. The center zone 12b contains a partial oxidation catalyst, and the upper zone 12c contains a reforming catalyst. An igniter 80 is disposed within the center zone 12b to provide initial heat to begin the partial oxidation reaction during start up. The inner wall of the first catalyst chamber is insulated with a first insulation zone, or a layer of insulation 74. It is preferred that the insulation layer 74 be tapered along the length of the first catalyst chamber such that the diameter of the upper catalyst zone 12c is greater than the diameter of the lower catalyst zone 12a. Thus, the first catalyst chamber comprises a progressively increasing layer of insulation 24 from the upper end to the lower end of the first catalyst chamber 12. A second catalyst chamber 14 annularly surrounds the first catalyst chamber 12 and at least a portion of the second catalyst chamber 14 is in intimate thermal contact with the first catalyst zone offset by the layer of insulation 74. It is important to maintain some degree of thermal separation between the first catalyst chamber and the second catalyst chamber to permit the operation of the first catalyst chamber to take place at very high effective partial oxidation conditions, while the second catalyst chamber is maintained at effective pre-reforming conditions which are more moderate than the partial oxidation conditions. The second catalyst chamber 14 has an annular catalyst zone 16. The annular catalyst zone 16 contains a pre-reforming catalyst for the partial conversion of the feed stream in the presence of steam at effective pre-reforming conditions to at least partially convert the feed stream into a pre-reforming effluent stream comprising hydrogen, carbon monoxide, carbon dioxide, and water. The second catalyst chamber 14 has a vertical feed conduit 18 at a top end and a fluid distributor/mixer 20 at a lower end. The fluid distributor/mixer 20 is in fluid communication with the lower zone 12a of the first catalyst chamber 12. The feed stream and steam enter the second catalyst chamber via the vertical feed conduit 18 and contact the pre-reforming catalyst in the second catalyst chamber 14 and the pre-reforming effluent stream from the annular catalyst zone 16 is passed to the fluid distributor/mixer 20. A burner zone 22 is disposed annularly surrounding the second catalyst chamber 14. The burner zone 22 has a lower burner inlet 24 through which a fuel/air mixture in line 5 is introduced. The burner zone 22 has an upper flue gas outlet 26 and the burner zone 22 is in intimate thermal contact with the second catalyst chamber 14. Heat produced from the combustion of fuel/air mixture in line 5 provides heat to the pre-reforming or annular catalyst zone 16. The burner zone 22 may contain

any suitable burner catalyst 88 or combustion promoter containing a sufficient amount of noble metal such as platinum or palladium to carry out the combustion of the fuel/air mixture. In the operation of the fuel cell, anode waste gases will be generated which may be incorporated into the fuel/air mixture in line 5. A second insulation zone 28 is disposed annularly about or surrounding the burner zone 22 to provide a thermal separation between the burner zone 22 and an air pre-heater zone 30 which is disposed annularly about or surrounding the burner zone 22. The air pre-heater zone 30 comprises an air inlet conduit 32 and an air pre-heater outlet 34. The pre-heated air outlet is in fluid communication with the fluid distributor/mixer 20. In the fluid distributor/mixer 20, the pre-heated air stream is mixed with the pre-reforming effluent stream and passed as a reforming feed admixture to the lower catalyst zone 12a containing a reforming catalyst to continue the reforming conversion to produce a reforming effluent stream. The reforming effluent stream is passed counter-current to the direction of the feed stream in the pre-reforming annular catalyst zone 16 to the center zone containing a partial oxidation catalyst. In the center zone 12b, the reforming effluent stream in the presence of the pre-heated air stream withdrawn from the air pre-heater outlet 34 undergoes a further conversion to hydrogen and carbon oxides which continue to flow counter-currently to the direction in which the feed stream was introduced to the feed conduit 18 through the upper zone 12c containing a reforming catalyst. Heat produced in the center zone 12b during partial oxidation provides heat to the adjacent reforming zones 12a and 12c and to the annularly surrounding pre-reforming catalyst zone 16 to the extent permitted by insulation layer 74. A net reforming effluent stream is withdrawn from the upper zone 12c and passed to a third catalyst chamber 40 which is annularly disposed on the air pre-heater zone 30 and disposed over the first catalyst chamber 12. The third catalyst chamber 40 comprises a top catalyst zone 42 containing a high-temperature water gas shift catalyst and a second annular catalyst zone 44 which surrounds and is in intimate thermal contact with the air pre-heater zone 30. The top catalyst zone 42 is in fluid communication with the upper zone 12c to permit the flow of the net reforming effluent to contact the high-temperature water gas shift catalyst in the top catalyst zone 42 wherein at least a portion of the carbon monoxide undergoes a shift reaction to produce additional hydrogen and produce a high-temperature shift effluent stream. The top catalyst zone 42 is in fluid communication with the second annular catalyst zone 44 and permits the high-temperature shift effluent stream to be passed co-currently to the

direction in which the feed stream was introduced to the feed conduit through the second annular catalyst zone 44 which contains a low-temperature water gas shift catalyst. An upper insulation zone 50 is disposed on and extends over the top catalyst zone 42. The high-temperature water gas shift reaction is an exothermic reaction. The upper zone 12c is operated in a manner which will provide some heat to the adjacent second annular catalyst zone 44, but it is desired to establish a falling temperature profile in the top catalyst zone 42 relative to the temperature of the upper zone 12c. The upper insulation zone 50 prevents excess heat transfer away from the top catalyst zone 42. The second annular catalyst zone contains a low-temperature shift catalyst and must be maintained at effective low-temperature shift conditions to prevent the loss of hydrogen by a reversal of the equilibrium reaction whereby hydrogen would combine with the carbon monoxide to produce water and carbon dioxide. To moderate the temperature of the second annular catalyst zone a plurality of steam conduits 54 is disposed therein. The plurality of steam conduits 54 extends vertically through the second annular catalyst zone 44, the top catalyst zone 42, and the upper insulation zone 50. The third catalyst chamber 40 is vertically disposed in a boiler interior 62 of a boiler vessel 60, and the plurality of steam conduits 54 are in fluid communication with the boiler interior 62 which is filled with fluid such as water or a mixture of water and a high boiling additive such as a glycol and/or a glycol ether to a fluid level 84 and controlled with a fluid level control means 82 such as a standpipe to maintain the fluid level to at least partially submerge the upper insulation zone 50. The plurality of steam conduits 54 act as thermosiphon tubes to heat the water in the boiler interior to generate steam and create a vertical thermosiphon flow which is counter-current to the both the direction of the feed stream flow and the direction of the flow of the high-temperature water gas shift effluent through the second annular catalyst zone, or low-temperature water gas shift zone 44. The plurality of steam conduits 54 herein act as thermosiphon tubes to generate steam from water in the boiler interior 62 and pass steam or a two-phase mixture of hot water and steam vertically upwards to a termination point above the upper insulation zone 50 and below the fluid level. The fluid level in the boiler interior 62 is maintained at an effective height to permit the thermosiphon flow in the steam conduits 54 by the introduction of fluid via fluid inlet 90 at a point below the third catalyst chamber 40, or by the removal of a boiler fluid stream 92 via fluid outlet 91. The effective height of the fluid level will depend upon the nature of the feed stream. For example, a fuel processor of the present design

for a feed comprising methane will require a relatively low fluid level control point consistent with the steam-to-carbon ratio required in the hydrogen production process. A higher water level control point is required for an LPG stream such as propane, which requires a relatively higher steam-to-carbon ratio in the hydrogen production process. In this manner, as the number of carbon atoms per molecule in the feed increases, the fluid level in the boiler interior 62 increases. If a standpipe is employed to establish the fluid level in the boiler interior when propane is used, the standpipe will extend to a greater height above the upper insulation zone 50 than when methane is used as fuel. When the fluid level is low, only those portions of the process providing the direct process heat which is sufficient for a feed such as methane. When the higher fluid level is required to produce more steam, additional heat from the flue gas is obtained via a submerged exchanger (not shown) or by passing the flue gas through the flue gas exchange zone 64 shown external to the boiler interior 62. This additional heat is required to maintain the boiler interior at effective thermosiphon conditions and to provide additional steam to achieve the desired steam-to-carbon ratio in the feed stream to the fuel processor. Steam generated in this manner is combined with the feed stream introduced in line 2 to a vapor space 65 above the fluid level 84 and in fluid communication with the vertical feed conduit 18.

The boiler vessel 60 is completely enclosed in a flue gas exchange zone 64 and is in intimate thermal contact therewith. The flue gas exchange zone 64 is in fluid communication with the upper flue gas outlet 26 and has a flue gas vent 68 and a cooled gas outlet 72. Flue gas generated in the burner zone 22 is conducted through the upper flue gas outlet 26 to a hot gas inlet 66 of the flue gas exchange zone 64. A damper 70 in the flue gas vent 68 is employed to control the circulation of flue gas in the flue gas exchange zone 64. Cooled flue gas is withdrawn in line 75 and hot flue gas exits the flue gas exchange zone 64 in line 6.

Returning to the second annular catalyst zone 44, this zone 44 has a shift effluent conduit 46 for withdrawing a low-temperature shift effluent stream from the second annular catalyst zone 44. The shift effluent conduit is in fluid communication with the second annular catalyst zone 44 and a fourth catalyst zone 76 which may be disposed in the interior of the boiler vessel 60 at a point below the fluid level and preferably below the second annular catalyst zone 44. The fourth catalyst zone 76 contains a preferential oxidation catalyst for the selective oxidation of carbon monoxide to carbon dioxide in the

presence of an oxygen-containing stream to produce a hydrogen fuel stream comprising less than about 50 ppm-vol carbon monoxide. More preferably, the hydrogen fuel stream comprises less than about 10 ppm-vol carbon monoxide. The fourth catalyst has a second effluent conduit 78 in fluid communication with the fourth catalyst zone and extending
5 beyond the flue gas exchange zone 64 for withdrawing the hydrogen fuel stream in line 3. In the operation of the process for the generation of hydrogen the fourth catalyst zone is maintained at essentially isothermal conditions, including a preferential oxidation temperature of about 105° to about 125°C. The hydrogen fuel stream in line 3 is passed to a fuel cell zone (not shown) for the generation of electric power which is withdrawn
10 from the fuel cell zone. Unconsumed hydrogen in the fuel cell is returned to the apparatus of the present invention as anode waste gas which is admixed with air or air in combination with cathode waste gas and returned to be burned in the burner zone 22 via line 5. For safety considerations and to contain the flame fronts within the appropriate zones, a first flame arrestor means 86 comprising a ring or a plug of sintered metal or
15 ceramic is disposed in the vertical feed conduit 18. The first flame arrestor means 86 may also be disposed at any point downstream of the pre-reforming annular catalyst zone 16 and the fluid distributor/mixer 20. A second flame arrestor means 88 is disposed at any point in the air pre-heater zone 30. Flame arrestor means are well known to those skilled in the art and need not be further discussed herein.

FIG. 2 shows a vertical cross section of the apparatus 10 of the present invention
20 to illustrate the relative relationship of the concentric catalyst and heat transfer zones. Referring to FIG. 2, the apparatus 10 comprises a flue gas exchange zone 64 in intimate thermal contact with and fully enclosing a boiler vessel 60. The boiler vessel 60 has a boiler interior 62 which contains a fluid comprising water and a third catalyst chamber
25 40 disposed therein. The third catalyst chamber 40 comprises a second annular catalyst zone 44 through which a plurality of steam conduits 54 extends vertically. The second annular catalyst zone is disposed on and is in intimate thermal contact with an air pre-heater zone 30 into which extends an air inlet conduit 32. An insulation zone 28 is disposed between the air pre-heater zone 30 and a burner zone 22. The burner zone 22
30 annularly surrounds and is in intimate thermal contact with a first annular catalyst zone 16 in a second catalyst chamber 14. The second catalyst chamber 14 is disposed annularly on and in intimate thermal contact with a first catalyst chamber 12. The first catalyst chamber is lined with a layer of insulation 74.

FIG. 3 shows a detail A-A of FIG. 2 with reference to the internal arrangements of the burner zone 22 and the second catalyst chamber 14. The burner zone can comprise an extended heat transfer surface 23 such as fins for exchanging heat between the burner zone and the flue gas passing therethrough. The second catalyst chamber can contain a
5 fixed bed of catalyst particles, a fixed bed of catalyst and uncoated vertically aligned fins, or a plurality of vertically aligned fins coated with catalyst 15.

In all cases, it is understood that the above-identified arrangements are merely illustrative of the many possible specific embodiments which represent application of the present invention. Numerous and varied other arrangements can readily be devised in
10 accordance with the principles of the present invention without departing from the spirit and scope of the invention.

CLAIMS:

1. An apparatus for generating a hydrogen product gas stream from a feed stream, said apparatus comprising:

a first catalyst chamber (12) having a upper end, a lower end, a hollow interior containing an upper zone (12c), a center zone (12b), and a lower zone (12a);

a second catalyst chamber (14) annularly surrounding the first catalyst chamber (12), at least a portion of said second catalyst chamber (14) being in intimate thermal contact with the first catalyst chamber (12), said second catalyst chamber (14) a having an annular catalyst zone (16), a vertical feed conduit (18) at a top end and a fluid distributor/mixer (20) at a lower end, said fluid distributor/mixer (20) being in fluid communication with the lower zone (12a) of the first catalyst chamber (12);

a burner zone (22) annularly surrounding the second catalyst chamber (14), said burner zone (22) having a lower burner inlet (24) and an upper flue gas outlet (26), said burner zone (22) being in intimate thermal contact with the second catalyst chamber (14);

an insulation zone (28) disposed annularly about the burner zone;

an air pre-heater zone (30) disposed annularly about the insulation zone (28), said air pre-heater zone (30) comprising an air inlet conduit (32) and a pre-heated air outlet (34) being in fluid communication with the fluid distributor/mixer (20);

a third catalyst chamber (40) annularly disposed on the air pre-heater zone (30) and disposed over the first catalyst chamber (12), said third catalyst chamber (40) having a top catalyst zone (42) in fluid communication with the upper zone (12c) of the first catalyst chamber (12), a second annular catalyst zone (44) in fluid communication with the top catalyst zone (42), a shift effluent conduit (46) in fluid communication with the second annular catalyst zone (44), and an upper insulation zone (50) extending over the top catalyst zone (42);

.0a plurality of steam conduits (54) disposed in the second annular catalyst zone (44) extending vertically through the second annular catalyst zone (44), the top catalyst zone (42), and the upper insulation zone (50);

a boiler vessel (60) having a boiler interior (62) has a fluid inlet (90) in fluid communication with the boiler interior (62) and a fluid outlet (91) from said boiler interior (62) wherein the third catalyst chamber (40) is vertically disposed in the boiler interior (62) and said plurality of steam conduits (54) are in fluid communication with the boiler interior (62); and

a flue gas exchange zone (64) disposed essentially fully enclosing the boiler vessel being in intimate thermal contact with the boiler vessel (60) and having a hot gas inlet (66) in fluid communication with the upper flue gas outlet (26) of the burner zone (22), a flue gas vent (68), and a cooled gas outlet (72).

2. The apparatus of claim 1 wherein the first catalyst chamber (12) comprises a vertical orientation and comprises a progressively increasing layer of insulation (74) from the upper end to the lower end.

3. The apparatus of claim 1 wherein the flue gas vent includes a flue gas damper (70).

4. The apparatus of claim 1 further comprising a fourth catalyst zone (76) disposed in the boiler vessel (60) said fourth catalyst zone being in fluid communication with the shift effluent conduit (46) and a second effluent conduit (78) for withdrawing the hydrogen product gas stream.

5. The apparatus of claim 1 further comprising an igniter means (80) in the center zone (12c) of the first catalyst chamber (12).

6. The apparatus of claim 1 further comprising a fluid level control means (82) disposed in the boiler vessel (60) to maintain a fluid level (84) at least partially submerging the third catalyst chamber (40).

7. The apparatus of claim 6 wherein the plurality of steam conduits (54) terminates at a point at or above the upper insulation zone (50) and below the fluid level (84).

8. The apparatus of claim 6 wherein the fluid level control means (82) comprises a standpipe means (82) in fluid communication with the fluid outlet (91) and at a height of the fluid level (84).

9. A process for controlling a steam-to-hydrocarbon ratio of a feed stream to an apparatus for generating a hydrogen product gas stream for a fuel cell, said processing comprising passing said feed stream and a steam stream to a fuel processor, said fuel processor being disposed in a boiler interior and submerged in a fluid below a fluid level

at an effective height to provide the steam-to-hydrocarbon ratio and said fuel processor being in intimate thermal contact with the fluid.

10. The process of claim 9 wherein the fuel processor comprises a steam reforming zone, a partial oxidation zone, a combustion zone, and a water gas shift
5 reaction zone.

11. The process of claims 9 or 10 wherein a fuel stream is passed to the combustion zone to produce a flue gas stream and the flue gas stream is passed to a flue gas exchange zone to provide additional steam.

Fig. 1

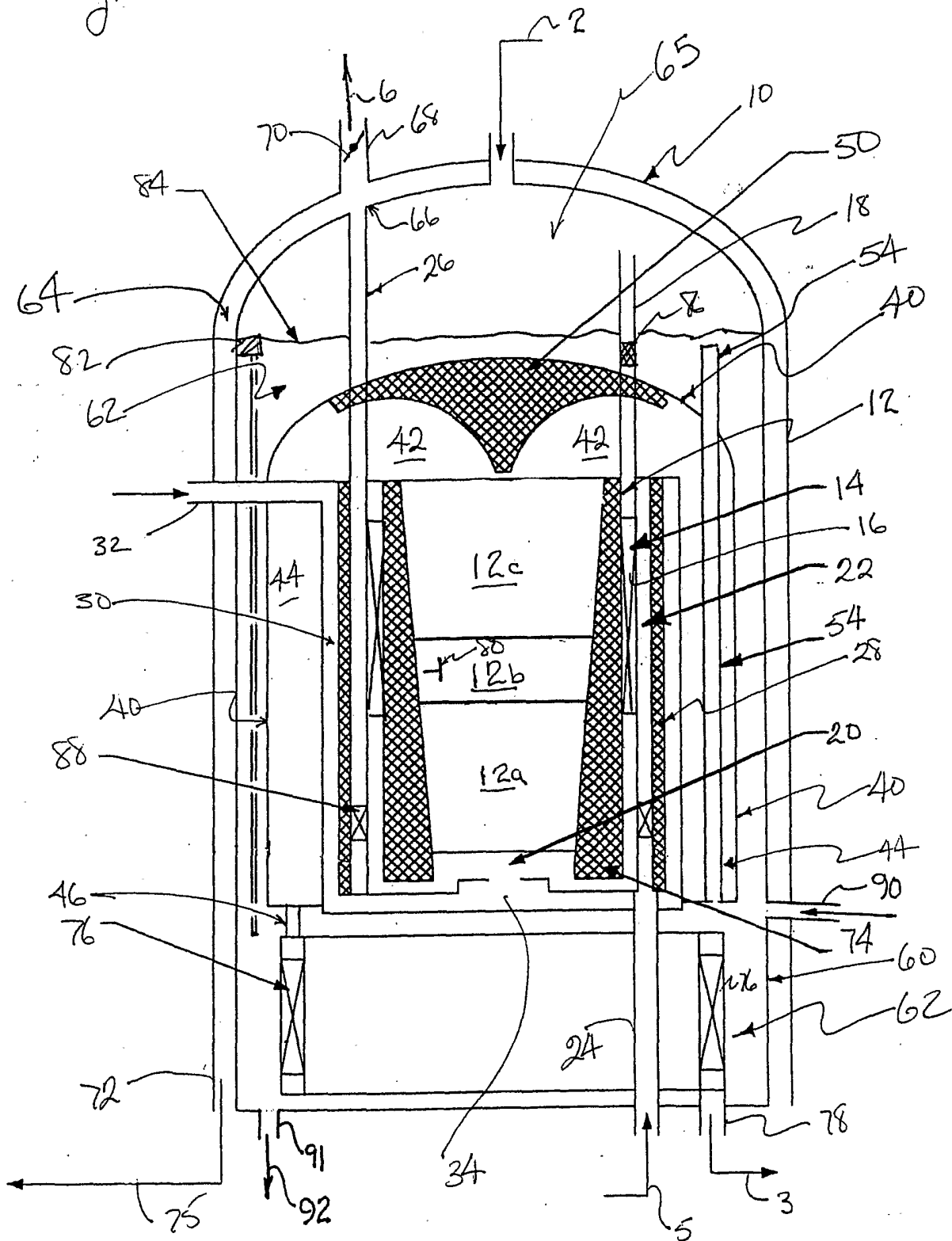


Fig. 3

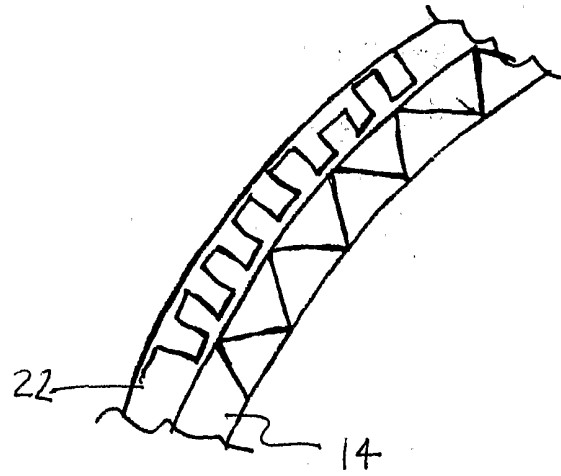
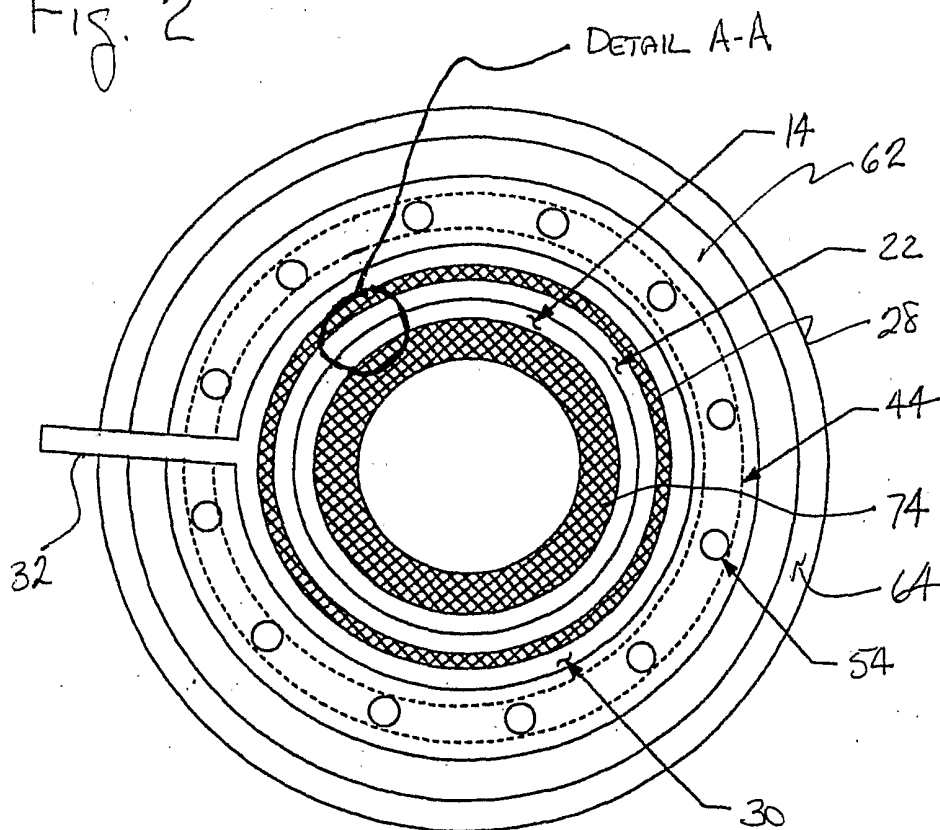


Fig. 2



INTERNATIONAL SEARCH REPORT

International Application No

PC1/US 01/19510

A. CLASSIFICATION OF SUBJECT MATTER

IPC 7 C01B3/38 C01B3/48 B01J8/04 C01B3/32

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 7 C01B 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, PAJ

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	US 3 541 729 A (DANTOWITZ PHILIP) 24 November 1970 (1970-11-24)	9,11
A	column 7, line 6-27; claim 1; figure 1 ---	1-8
A	US 4 861 348 A (KOYAMA KAZUHIITO ET AL) 29 August 1989 (1989-08-29) claim 1; figures 6-8 ---	1-8
A	US 5 006 316 A (ZARDI UMBERTO ET AL) 9 April 1991 (1991-04-09) the whole document ---	9-11
A	EP 0 600 621 A (ROLLS ROYCE & ASS) 8 June 1994 (1994-06-08) claim 1; figures 1-3 -----	1-8



Further documents are listed in the continuation of box C.



Patent family members are listed in annex.

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Date of the actual completion of the international search

18 October 2001

Date of mailing of the international search report

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INTERNATIONAL SEARCH REPORT

Information on patent family members

International Application No

PCT/US 01/19510

Patent document cited in search report		Publication date	Patent family member(s)	Publication date
US 3541729	A	24-11-1970	NONE	
US 4861348	A	29-08-1989	JP 2528836 B2	28-08-1996
			JP 63093343 A	23-04-1988
			JP 1951499 C	28-07-1995
			JP 6079664 B	12-10-1994
			JP 63126539 A	30-05-1988
			US 4935037 A	19-06-1990
US 5006316	A	09-04-1991	CA 1306848 A1	01-09-1992
			CN 88103614 A	14-12-1988
			IN 171435 A1	10-10-1992
EP 0600621	A	08-06-1994	CA 2109655 A1	03-06-1994
			DE 69309862 D1	22-05-1997
			DE 69309862 T2	24-07-1997
			EP 0600621 A1	08-06-1994
			JP 6239601 A	30-08-1994
			US 5458857 A	17-10-1995