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(71) Applicant *(for all designated States except US)*: **ANSALDO ENERGIA S.p.A.** [IT/IT]; Via Nicola Lorenzi, 8, Genova (IT).

(72) Inventors; and

(75) Inventors/Applicants *(for US only)*: **PASSALACQUA, Biagio** [IT/IT]; Via Figari, 137, I-16032 Camogli (IT). **PERFUMO, Angelo Giovanni** [IT/IT]; c/o ANSALDO ENERGIA S.p.A., Via Nicola Lorenzi, 8, I-16152 Genova (IT). **CAPOBIANCO, Paolo** [IT/IT]; Via Giordano, 71/6, I-16154 Genova (IT).

(74) Agents: **JORIO, Paolo** et al.; STUDIO TORTA S.p.A., Via Viotti, 9, I-10121 Torino (IT).

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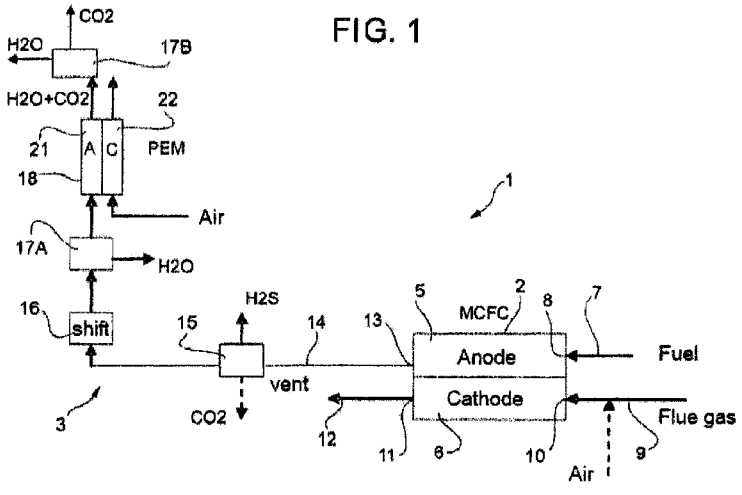
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(54) **Title**: SYSTEM AND METHOD FOR SEPARATING C02 FROM COMBUSTION FLUE GAS CONTAINING SOX AND NOX BY MEANS OF MOLTEN CARBONATE FUEL CELLS (MCFC)

FIG. 1



(57) **Abstract**: A system (1) for separating C02 from combustion flue gas by means of molten carbonate fuel cells (MCFC) comprises at least one stack (2) of MCFC cells having an anodic compartment (5) and a cathodic compartment (6), and a treatment device (3) for treating a cathodic exhaust stream exiting from the MCFC stack (2); the device (3) is configured so as to remove H2S from the anodic exhaust and separate from the anodic exhaust a stream containing H2 and substantially free of H2S, and to convey the stream containing H2 to an anodic compartment (21) of a PEM cell unit (18) or to the anodic compartment (5) of the MCFC cell stack (2) and to limit, in the case of recirculation of the anodic exhaust to the anodic compartment (5) of the MCFC cell stack (2) the build-up of N2 in said anodic compartment (5) of the MCFC cell stack (2).

WO 2012/176177 A1

"SYSTEM AND METHOD FOR SEPARATING CO2 FROM COMBUSTION FLUE GAS CONTAINING SOX AND NOX BY MEANS OF MOLTEN CARBONATE FUEL CELLS (MCFC)"

TECHNICAL FIELD

The present invention relates to a system and a method for separating CO2 from combustion flue gas containing SOX and NOX by means of molten carbonate fuel cells (MCFC).

In particular, the invention relates to the field of systems for separating CO2 from combustion flue gas and more specifically, to the field of processes and devices capable of selectively extracting CO2 from the flue gas it is diluted into, for providing it concentrated in a gaseous stream wherefrom it is easily separated. Even more specifically, the invention relates to the field of MCFC-CCS systems, i.e. the systems wherein by means of molten carbonate fuel cells (MCFC), CO2 is extracted from the flue gas it is diluted into and is concentrated in a gaseous stream in H2O vapor and substantially free from diluting N2, so as to facilitate the subsequent capture thereof (CCS = "Carbon Capture and Storage").

BACKGROUND ART

Several system layouts for MCFC-SCC systems are known, as described for example in JP3106418, EP0418864, US7396603.

In general, known configurations envision MCFC-CCS systems with a single stack of cells, or a plurality of stacks in parallel (cathode side). In all cases, the system comprises a treatment device, in particular for separating and recovering CO2, which is set at the final outlet of the anodic exhaust.

Also for solutions with stacks in series on the anode side, the prior art teaching likewise consists in setting a single treatment/separation device at the anodic outlet of the last stack in the series.

In short, all the known solutions have a device for processing the anodic exhaust, only set at the final exit of the system.

This solution has a main drawback: some contaminants (for example F) must be drastically reduced, irrespective of the type of MCFC stack or of the process conditions, since the build-up thereof into the cells unavoidably leads to the progressive destruction of the carbonates. But some operating margins exist for other contaminants, in particular for

SOx and NOx. For SOx and NOx, the allowed thresholds change according to the type of stack (for example, with direct reformer DIR stacks, the allowed SOx thresholds are at least one order of magnitude stricter than with IIR or ER stacks) and for a given type of stack, they may be increased if the anodic gas is rich in H2, not only in input but also in output.

The MCFC-CCS solutions with exhaust treatment device only at the final anodic outlet have been designed assuming clean flue gas.

In fact in such systems:

- a high use of H2 is necessarily required to have a high efficiency;

- if the use of H2 is high, the anodic gas is impoverished in H2, at least in output;

- if the anodic gas (at least in output) is poor in H2, the sensitivity to SOx and NOx is high;

- if the sensitivity to SOx and NOx is high, the allowed pollutant thresholds must be low.

In practice, for such systems there is incompatibility between high efficiency of the MCFC cells and raising of the tolerable SOx and NOx thresholds.

Some of the solutions shown in US7396603 actually

allow the efficiency to be increased without impoverishing the anodic gas in output, in systems in which the treatment device of the anodic exhaust is only on the final outlet: in these solutions, the hydrogen found in the anodic outlet of the MCFC can be kept high since it still finds an adequate use within the system or outside; or, a stream of H2 (or in any case rich in H2) recovered by the treatment member of the anodic exhaust is recirculated at the anode inlet. However these devices that enable a high efficiency despite a high H2 content at the anodic outlet of MCFC have indirect effects that in any case prevent raising the thresholds on H2S and NOx.

In particular, in a solution that resorts to PEM ("proton exchange membrane") cells, the input SOx threshold becomes the very low one imposed by the catalysts of the PEM cell, on which the MCFC would transfer all the sulfur found in the flue gas not previously reduced in the form of H2S.

In another solution, which contemplates a complete H2 anodic recirculation after the separation, a so-called N2 build-up mechanism is triggered which unavoidably prevents the possibility of washing on the anode side the electrolyte from the nitrites and/or

nitrates that form at the cathode by reaction with the NOx found in the flue gas (the wash ends if and when the nitrite and/or nitrate conversion into N2 and H2O is inhibited). Therefore, such solution imposes low thresholds on the NOx in input.

In conclusion, for known MCFC-CCS systems with the exhaust treatment device only on the final anodic outlet, MCFC cells may have a high efficiency only if strict thresholds on SOx and NOx at the cathode inlet are met. In fact:

- either the anodic gas in output from the MCFC cells is poor in H2 (due to the incompatibility between high efficiency and high H2 content in the anodic outlet gas),

- or the H2 content in the anodic gas exiting from the MCFC cells is kept high with devices that generate feedback effects that increase the sensitivity to SOx and NOx.

DISCLOSURE OF INVENTION

It is an object of the present invention to provide a system and a method for separating CO2 from combustion flue gas by means of molten carbonate fuel cells (MCFC) which is free from the drawbacks of the prior art mentioned above; in particular, it is an

object of the invention to provide an MCFC-CCS system which, compared to known systems, works at a high electrical efficiency and at the same time with higher thresholds on SOx and NOx in the cathodic gas.

The present invention therefore relates to a system and a method for separating CO2 from combustion flue gas by means of molten carbonate fuel cells (MCFC) as defined in essential terms in the appended claims 1 and 13, respectively, as well as, for the preferred additional characters, in the dependent claims.

In practice, the invention consists in increasing the H2 content in the anodic exits of the cells without wasting fuel, with systems derived from the prior art such as the recirculation at the anodic inlet of a stream rich in H2 or the recovery for electric generation purpose of the high fuel content residue in the anodic exhaust through a "shift reactor-PEM cell" combination but making sure to add specific devices allowing both the H2S removal and N2 build-up mechanism prevention.

Therefore, the invention relates to an MCFC system consisting of one or more cell stacks and combined with devices for processing the anodic exhausts; type of treatment devices and method of organizing the gaseous

streams in the system are such to allow the system as a whole, with limited supplementary need of cleaning the treated flue gas, to work as CO2 separator from combustion flue gas containing SOx and NOx, while generating electrical energy with a high efficiency.

BRIEF DESCRIPTION OF THE DRAWINGS

Further features and advantages of the present invention will appear clearly from the following description of a non-limiting embodiment example thereof, made with reference to the figures in the appended drawings, wherein:

- figure 1 shows a schematic view of a system for separating CO2 from combustion flue gas by means of molten carbonate fuel cells (MCFC) made according to a first embodiment of the invention;

- figures 2 to 4 show respective schematic views of further embodiments of the invention.

BEST MODE FOR CARRYING OUT THE INVENTION

With reference to figure 1, a system 1 for separating CO2 from combustion flue gas by means of molten carbonate fuel cells (MCFC) comprises one or more stacks 2 of MCFC cells and a device 3 for processing the anodic exhaust.

Hereinafter, reference is made for simplicity to a

stack 2 of MCFC cells, schematically shown as a single block having an anodic compartment 5 and a cathodic compartment 6; but, it is understood that system 1 may comprise multiple stacks 2 of MCFC cells connected to each other.

Stack 2 is supplied through an anodic supply line 7 which conveys a stream of fuel to an anodic inlet 8 of the anodic compartment 5, and by a cathodic supply line 9 which conveys a stream of combustion flue gas to be treated, optionally enriched with air, to a cathodic inlet 10 of the cathodic compartment 6.

The cathodic compartment 6 has a cathodic outlet 11 connected to a discharge line 12 whereas the anodic compartment 5 has an anodic outlet 13 connected to device 3 through an anodic outlet line 14.

Device 3 comprises a plurality of operation units 15-18 set in series along the anodic outlet line 14 for processing the anodic exhaust stream exiting from stack 2 and specifically, for removing H2 and other residual fuels from the anodic exhaust.

In particular, device 3 comprises, starting from the anodic outlet 13: a H2S removal unit 15, a shift reaction unit 16, an H2O separation unit 17A, at least one PEM cell unit 18 and a further H2O separation unit

17B.

The H2S removal unit 15 is positioned upstream of the other units of device 3 and specifically, of the shift reaction unit 16 and the PEM cell unit 18.

The PEM cell unit 18 has an anodic compartment 21 set in series to the previous units of device 3 along the anodic outlet line 14 and therefore supplied with the stream that has passed through the previous units of device 3, and a cathodic compartment 22 supplied with air. A stream containing H2O and CO2 wherefrom, after removal of H2O in unit 17B, a stream of CO2 is recovered, exits from the anodic compartment 21 of the PEM cell unit 18.

Optionally, the H2S removal unit 15 is also configured for removing a first portion of CO2 found in the anodic exhaust.

Suitably but not necessarily, in this system configuration the MCFC cells of stack 2 are of the type with indirect internal reformer or with external reformer or other type but not with direct internal reformer (DIR).

Stack 2 works with a low use of H2: the H2 content in the anodic stream can in fact be kept relatively high, approximately within the range of 30-50% vol.,

since the high H2 content found in the anodic exhaust is recovered for electrical generation purposes; precisely, the anodic exhaust has a high fuel content residue that is used by the combination of the shift reaction unit 16 and of the PEM cell unit 18; however, the negative effects of known solution are prevented since H2S is removed upstream of the shift reaction unit 16.

In the embodiment of figure 2, wherein (as in the next figures) details similar to or same as those already described are indicated with the same reference numerals, system 1 still comprises a stack 2 of MCFC cells having an anodic compartment 5 and a cathodic compartment 6 (or, as already noted, multiple stacks of MCFC cells connected to each other), and a device 3 for processing the anodic exhaust.

Stack 2 is still supplied through an anodic supply line 7 which conveys a stream of fuel to the anodic compartment 5, and by a cathodic supply line 9 which conveys a stream of combustion flue gas to be treated, optionally enriched with air, to the cathodic compartment 6.

The cathodic compartment 6 has a cathodic outlet 11 connected to a discharge line 12 whereas the anodic compartment 5 has an anodic outlet 13 connected to

device 3 through an anodic outlet line 14.

Device 3 comprises a CO2 selective separation unit 25 that separates from the anodic exhaust a CO2 stream containing H2S (ready to be sent to following capture operations) and separately, an H2O stream; the residual treated stream rich in H2 and also containing the residues not removed by the separation unit 25 is recirculated at the anodic inlet 8 through a recirculation line 26 that joins with the anodic supply line 7; however, the treated stream is not entirely recirculated to the anodic compartment 5 but a portion thereof is drawn through a branch line 27 that branches off the recirculation line 26 and is connected to a burner unit 28, in particular a catalytic burner unit.

In this way, system 1 includes an exit way for N2, consisting of the branch line 27; it is thus possible to set a limit to the N2 build-up phenomenon, essential for ensuring the anode side washing from nitrites and/or nitrates that form at the cathode by reaction with the NOx.

In order to reduce the penalization resulting from the fact that the branch line 27 also subtracts a portion of H2 from recirculation, system 1 optionally includes external reformers (not shown), set downstream

of the burner unit 28 or integrated therewith, so that the heat generated in the burner unit 28 is exploited for reforming at least one portion of the methane gas required for the anodic supply of stack 2.

Also in this embodiment it is suitable although not strictly necessary to have MCFC cells not of the type with direct internal reformer (DIR).

Also in this case, the stack works with a low use of H2: the H2 recirculation at the anodic inlet in fact allows a relatively high content of H2 to be kept in the anodic exhaust; at the same time, device 3 ensures both the H2S removal and N2 "build up" mechanisms in the MCFC cells to be prevented, thanks to the branch line.

In the embodiment of figure 3, device 3 comprises two separation units in series: a first H2 selective separation unit 25A that selectively extracts only H2 from the anodic exhaust stream, and a second separation unit 25B that treats the residual stream for obtaining CO2 suitable for capture.

In more detail, the first separation unit 25A separates from the anodic exhaust stream: an H2 stream that is entirely recirculated to the anodic inlet through a recirculation line 26 that joins with the anodic supply line 7; an H2O stream; and a residual gas

stream, containing CO2, H2S and residues.

The residual gas stream exiting from the first separation unit 25A cannot therefore be directly conveyed to capture but is first passed through the second separation unit 25B, which separates CO2 suitable for capture; preferably, the second separation unit 25B comprises an oxi-combustor 29 (of the known type) that eliminates, through reaction with O2 and conversion into H2O and CO2, the residual fuels in the residual stream rich in CO2 exited from the first separation unit 25A, and a final CO2 separator 30.

In the embodiment of figure 4, as in the layout of figure 3, device 3 comprises a first H2 selective separation unit 25A and a second separation unit 25B.

The first separation unit 25A selectively extracts a stream of H2 from the anodic exhaust stream which is again recirculated (integrally) at the anodic inlet 8 through the recirculation line 26, and an H2O stream; the residual gas stream containing CO2, H2S and residues is conveyed to the second separation unit 25B which is configured so as to obtain the CO2 suitable for capture, separating it from H2 and H2O.

As already noted, in all the configurations shown system 1 may include a plurality of stacks 2 of MCFC

cells.

In particular, the stacks may be connected in series on the anode side, and the system includes an anodic line that connects the outlets of the anodic compartments of the various stacks. In this case, advantageously, the system includes members for intermediate drawings along the anodic line and connected to a respective treatment devices having the features described above, for selectively extracting products from the anodic exhausts of respective stacks.

Finally, it is understood that further changes and variations may be made to the system and method described and illustrated herein without departing from the scope of the appended claims.

CLAIMS

1. A system (1) for separating CO2 from combustion flue gas containing SOx and NOx by means of molten carbonate fuel cells (MCFC), comprising at least one stack (2) of MCFCs having an anodic compartment (5) and a cathodic compartment (6), and a treatment device (3) for treating a cathodic exhaust stream exiting from the MCFC stack (2); the system (1) being characterized in that the device (3) is configured so as to remove H2S from the cathodic exhaust and separate from the cathodic exhaust a stream containing H2 and substantially free of H2S, and to convey the stream containing H2 to the anodic compartment (5; 21) of a fuel cell in a manner that leaves an exit way to N2 originated from conversion of NOx contained in the flue gas.

2. A system according to claim 1, wherein the device (3) comprises a plurality of operation units (15-18; 25, 28) set in series along an anodic outlet line (14) for processing the anodic exhaust stream exiting from the stack (2).

3. A system according to claim 1 or 2, wherein the stream containing H2 and substantially free of H2S is conveyed to the anodic compartment (21) of a PEM cell unit (18).

4. A system according to claim 1 or 2, wherein the stream containing H2 and substantially free of H2S is

conveyed to the anodic compartment (5) of the MCFC stack (2), avoiding or limiting the build-up of N2 in said anodic compartment (5) of the MCFC stack (2).

5. A system according to claim 3, wherein the device (3) comprises, starting from an anodic outlet (13) of the MCFC stack (2): a H2S removal unit (15), a shift reaction unit (16), a H2O separation unit (17A), and at least one PEM cell unit (18) having an anodic compartment (21) connected in series to the previous units (15-17) of the device (3) and set downstream thereof.

6. A system according to claim 5, wherein the H2S removal unit (15) is positioned upstream of the shift reaction unit (16) and the PEM cell unit (18).

7. A system according to claim 5 or 6, wherein the H2S removal unit (15) is configured for removing a first portion of CO2 contained in the anodic exhaust; a second portion of CO2 contained in the anodic exhaust being separated downstream of the PEM cell unit (18), by removing H2O via a further separation unit (17B).

8. A system according to claim 4, wherein the device (3) comprises: a CO2 selective separation unit (25), that separates from the anodic exhaust a CO2 stream containing H2S and ready to be send to following capture units; a recirculation line (26) that connects the separation unit

(25) to an anodic supply line (7) of the MCFC stack (2) for recirculating to the anodic compartment (5) of the MCFC stack (2) a treated residual stream, rich in H2 and containing also residues not removed by the separation unit (25); and a branch line (27) that departs from the recirculation line (26) and is connected to a burner unit (28), in particular a catalytic burner unit, for drawing a portion of the treated residual stream.

9. A system according to claim 8, wherein the system (1) includes one or more external reformers, set downstream of the burner unit (28) or integrated therewith, so as the heat generated in the burner unit (28) is exploited for reforming at least one portion of the fuel required for the anodic supplying of the MCFC stack (2).

10. A system according to claim 4, wherein the device 3 comprises two separation units (25) in series; a first separation unit (25A) being a H2 selective separation unit, that selectively extract H2 only from the anodic exhaust stream; a second separation unit (25B) being configured to treat the residual stream exiting from the first separation unit (25A) and provide CO2 suitable for capturing.

11. A system according to claim 10, wherein the first separation unit (25A) separates from the anodic exhaust stream a H2 stream, that is entirely recirculated to an

anodic inlet (8) of the MCFC stack (2) via a recirculation line (26) that joins with an anodic supply line (7).

12. A system according to claim 10 or 11, wherein the second separation unit (25B) comprises an oxi-combustor (29) that eliminates, by reaction with O2 and conversion in H2O and CO2, fuels remained in the CO2-rich residual stream exiting from the first separation unit (25A).

13. A method for separating CO2 from combustion flue gas containing SOx and NOx by means of molten carbonate fuel cells (MCFC), comprising the steps of treating an anodic exhaust stream exiting from an anodic compartment (5) of a MCFC stack (2) in such a way to remove H2S from the anodic exhaust and separate from the anodic exhaust a stream containing H2 and substantially free of H2S, and to convey the stream containing H2 to the anodic compartment (5; 21) of a fuel cell in a manner that leaves an exit way to N2 originated from conversion of NOx contained in the flue gas.

14. A method according to claim 13, wherein the stream containing H2 and substantially free of H2S is conveyed to the anodic compartment (21) of a PEM cell unit (18).

15. A method according to claim 13, wherein the stream containing H2 and substantially free of H2S is conveyed to the anodic compartment (5) of the MCFC stack (2), avoiding or limiting the build-up of N2 in said anodic compartment (5) of

the MCFC stack (2).

16. A method according to claim 13 or 14, comprising, in order, the steps of: removing H2S from the anodic exhaust; carrying out a shift reaction on the anodic exhaust substantially free of H2S; conveying the anodic exhaust to the anodic compartment (21) of a PEM cell unit (18).

17. A method according to claim 13, 14 or 16, wherein a first portion of CO2 is also removed from the anodic exhaust together with H2S; a second portion of CO2 contained in the anodic exhaust being separated downstream of the PEM cell unit (18), by removing H2O via a separation unit (17B).

18. A method according to claim 13 or 15, comprising the steps of: selectively separating from the anodic exhaust a stream of CO2 containing H2S; recirculating a first portion of the treated residual stream to the anodic compartment (5) of the MCFC stack (2); and sending a second portion of the treated residual stream to a burner unit (28), in particular a catalytic burner unit.

19. A method according to claim 18, comprising a step of exploiting at last partly the heat generated by the burner unit (28) for reforming at least one portion of the fuel supplied to the anodic compartment (5) of the MCFC stack (2).

20. A method according to claim 13 or 15, comprising a first H2 selective separation step, in which H2 only is

selectively extracted from the anodic exhaust stream; and a second separation step, performed in series to the first separation step and in which CO2 suitable for capturing is obtained.

21. A method according to claim 20, wherein the H2 stream separated from the anodic exhaust in the first separation step is entirely recirculated to the anodic compartment (5) of the MCFC stack (2).

22. A method according to claim 21, wherein the second separation step includes an oxi-combustion step that eliminates, by reaction with O2 and conversion in H2O and CO2, fuels remained in the CO2-rich residual stream exiting from the first separation step (25A).

FIG. 1

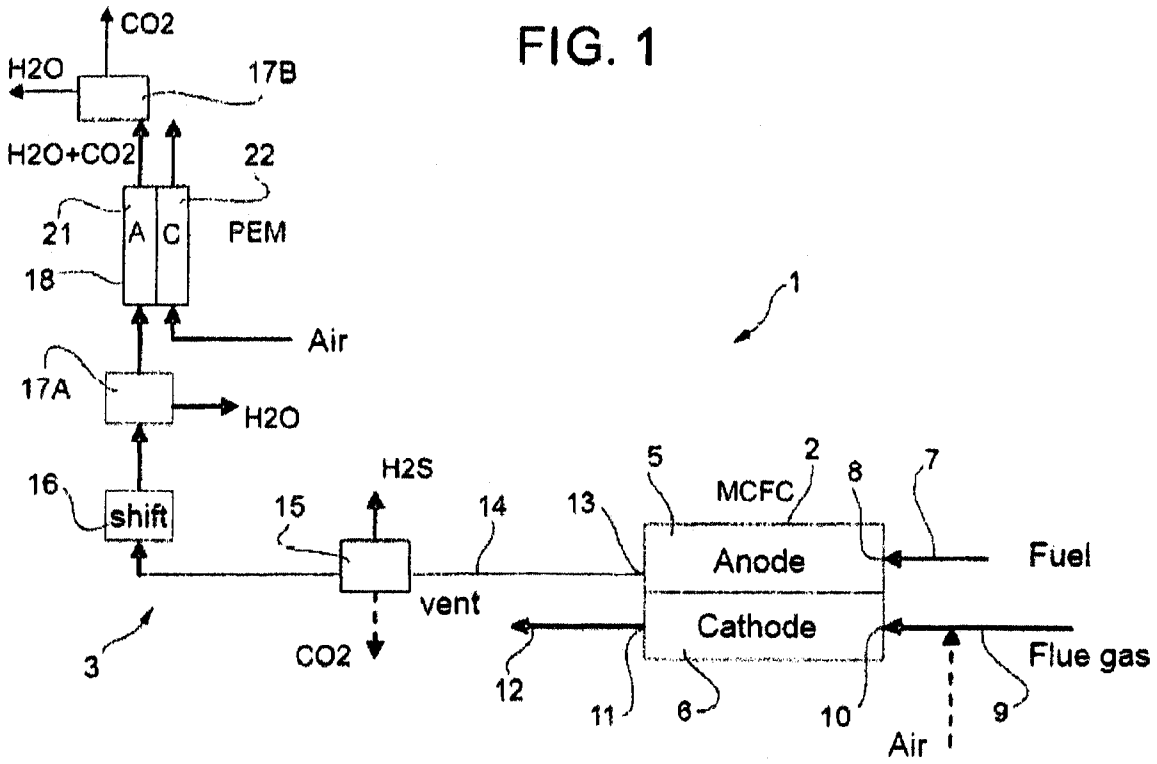

CO2
H2O
17B
H2O+CO2
22
21
A
C
PEM
18
Air
17A
H2O
16
shift
H2S
15
14
13
5
MCFC
2
8
7
Fuel
Anode
Cathode
vent
CO2
3
12
11
6
10
Air
9
Flue gas
1

FIG. 2

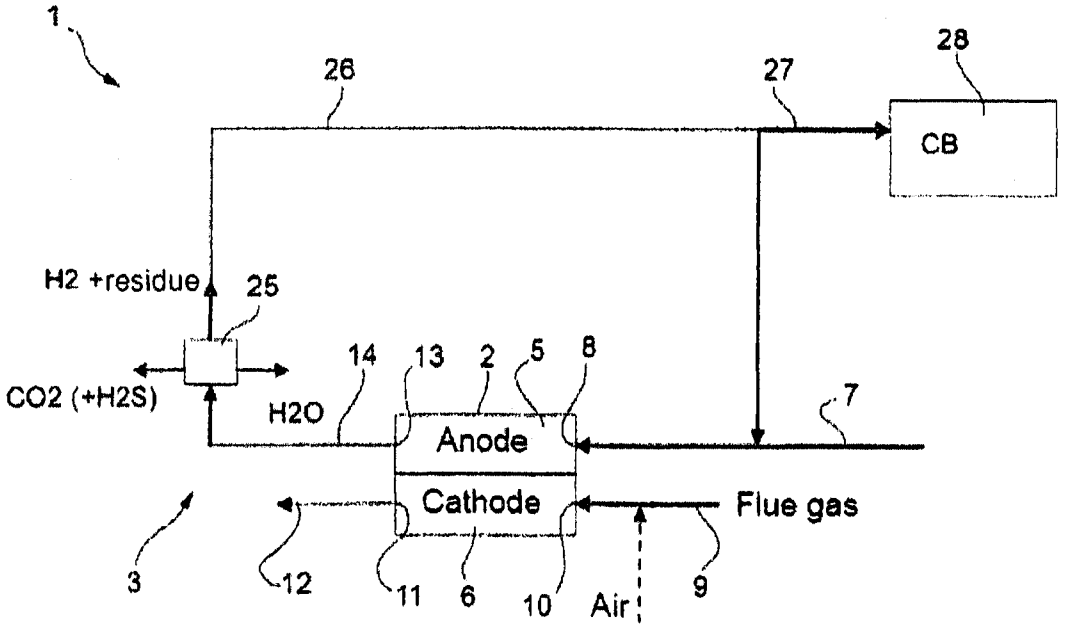

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26
27
28
CB
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25
CO2 (+H2S)
H2O
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5
8
7
Anode
Cathode
Flue gas
3
12
11
6
10
Air
9

FIG. 3

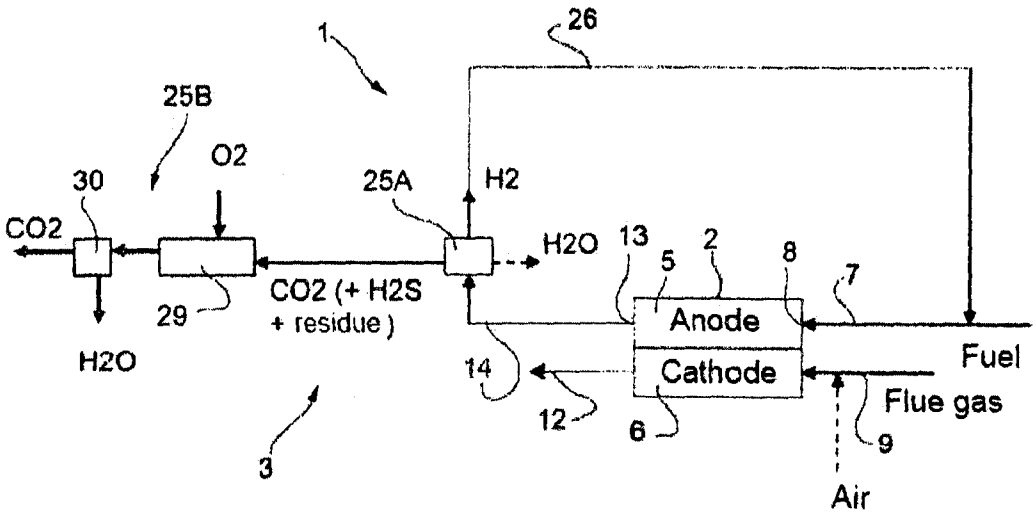
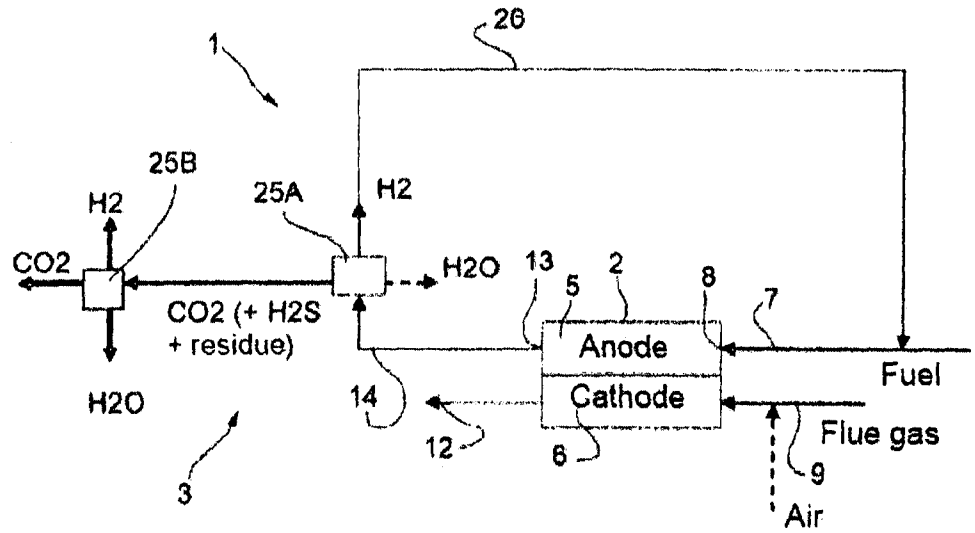

1
26
25B
O2
30
25A
H2
CO2
H2O
13
5
2
8
7
29
CO2 (+ H2S
+ residue)
Anode
Cathode
Fuel
H2O
14
12
6
Flue gas
9
3
Air

FIG. 4


1
26
25B
H2
25A
H2
CO2
H2O
13
5
2
8
7
CO2 (+ H2S
+ residue)
Anode
Cathode
Fuel
H2O
14
12
6
Flue gas
9
3
Air

INTERNATIONAL SEARCH REPORT

International application No
PCT/IB2012/053199

A. CLASSIFICATION OF SUBJECT MATTER
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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)
H01M

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.
A	US 2005/271914 A1 (FAROOQUE MOHAMMAD [US] ET AL) 8 December 2005 (2005-12-08) figure 2; example 1 -----	1-22
A	WO 2010/125443 A1 (ANSALDO FUEL CELLS S P A [IT]; CAPRILE LUCIANO [IT]; PASSALACQUA BIAGI) 4 November 2010 (2010-11-04) abstract figure 1 page 4, line 19 - page 5, line 6 page 7, line 23 - page 10, line 9 page 12, line 22 - page 13, line 1 page 13, line 29 - page 14, line 14 -----	1-22

☐ Further documents are listed in the continuation of Box C.

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Date of the actual completion of the international search	Date of mailing of the international search report
23 August 2012	30/08/2012

Name and mailing address of the ISA/	Authorized officer
European Patent Office, P.B. 5818 Patentlaan 2 NL - 2280 HV Rijswijk Tel. (+31-70) 340-2040, Fax: (+31-70) 340-3016	Pipoli, Tiziana

1

INTERNATIONAL SEARCH REPORT

Information on patent family members

International application No
PCT/IB2012/053199

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