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(71) Applicant: ZEG POWER AS [NO/NO]; Fantoftvegen 38, N-5072 Bergen (NO).

(72) Inventors: VIK, Arild; Slettenveien 76, N-5258 Blomsterdalen (NO). RAAHEIM, Arne; Rypelia 16, N-2932 Maura (NO).

(74) Agent: CURO AS; Industriveien 53, N-7080 Heimdal (NO).

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(54) Title: METHOD FOR SUSTAINABLE ENERGY PRODUCTION IN A POWER PLANT COMPRISING A SOLID OXIDE FUEL CELL

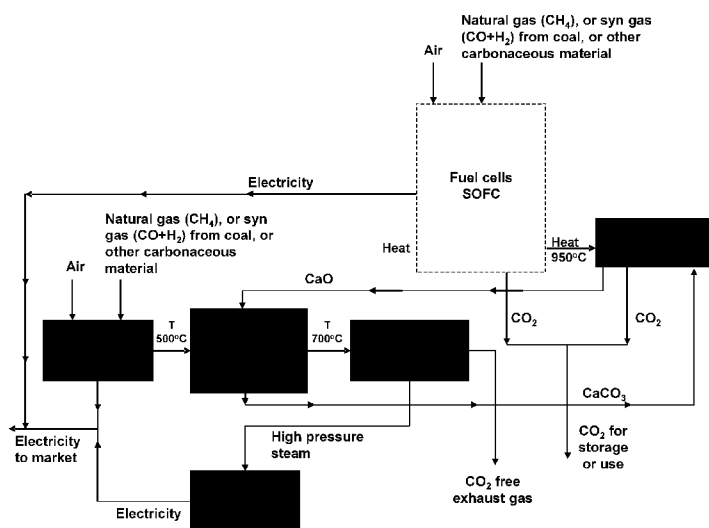


Fig. 2

(57) Abstract: Method for sustainable energy production in a plant comprising a solid oxide fuel cell (SOFC) in which method calcium oxide is being used as a sorbent in an exothermic carbon capture step. Calcium oxide is converted to calcium carbonate in the carbon capture step and subsequently converted to calcium oxide in an endothermic regeneration step, heated by the SOFC. The carbon capture step is performed in the presence of hot gas from another process, thus increasing the temperature of the hot gas thereby improving the cost-efficiency of said other process.

WO 2014/200357 A1

## Method for sustainable energy production in a power plant comprising a solid oxide fuel cell

The present invention relates to a method for sustainable energy production as indicated by the preamble of claim 1.

### 5 Background

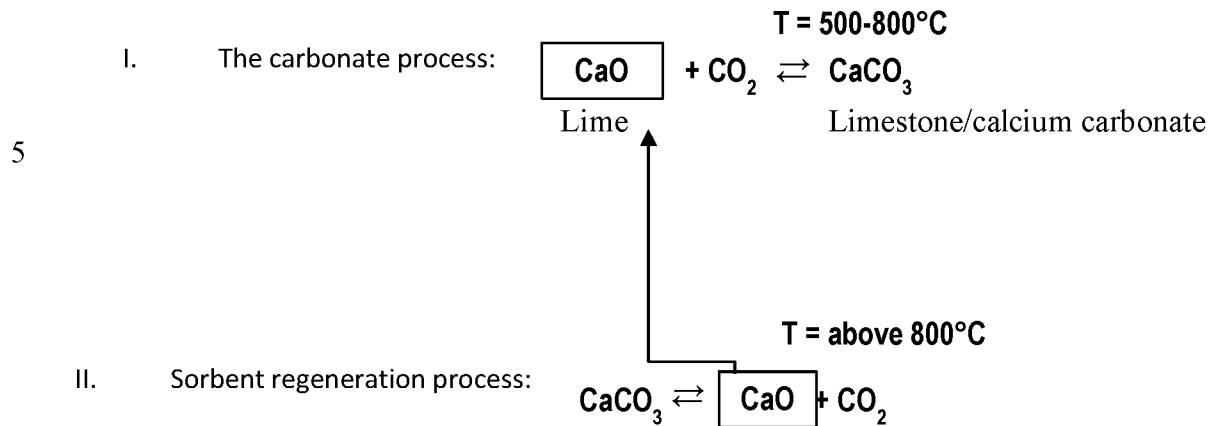
International Energy Agency (IEA) has defined energy efficiency as one of the most important steps for reduction of greenhouse gases (GHG) (see IEA's Blue Map Scenario 2010). In addition, successful development and application of CCS (carbon capture and storage) in the power sector and a number of energy-intensive industrial sectors (e.g. iron and steel, aluminium, silicon, ferrosilicon, cement, etc.) potentially represent the most important option for direct large scale  
10 reduction of GHG emissions in power production and industry.

The publication WO 2004/025767 (Vik et al.) discloses a plant for production of electricity from a hydrocarbon-containing flow. According to one embodiment, a solid oxide fuel cell (SOFC) is used for producing the electricity. The process involves reforming of the fuel in order to produce  
15 hydrogen before separating it from the other components to use pure hydrogen as the feed to the fuel cell. CO<sub>2</sub> produced during reforming may be captured directly in the process by the lime (CaO) sorbent in a hydrogen production reactor. The solid calcium carbonate (CaCO<sub>3</sub>) formed in this process is transferred to the sorbent regeneration reactor. The CO<sub>2</sub> formed in this process (see reaction II) may be stored or used in processes where CO<sub>2</sub> is needed. The process of Vik et al. is  
20 targeted towards applications where excess heat is not needed, and where high efficiency for the co-production of electricity and hydrogen is the primary object.

WO 2011/078681 A1 teaches a method that allows cost and energy-efficient sustainable recovery and energy production from any carbon-containing fuel (solid or gas), whether fossil or of bio origin. This method provides efficient CO<sub>2</sub> capture, and allows a highly versatile production of  
25 energy in the forms electricity, hydrogen and heat. By "versatility" in this respect it is understood that the ratio of the amounts of these energy forms may be varied within wide limits by simple change of parameters in the process. This publication also makes use of solid oxide fuel cell for energy production, in a process in which CaO is used to capture CO<sub>2</sub>. CaO is converted to CaCO<sub>3</sub> in this process and subsequently regenerated in a secondary closed loop process also called calcium-  
30 looping, further described below.

Lime (CaO) is used to capture CO<sub>2</sub>. Calcium carbonate (CaCO<sub>3</sub>)/limestone is then formed at a temperature of about 650°C (650°C ± 150°C), the carbonation process (see reaction below). The calcium carbonate (CaCO<sub>3</sub>) is transferred to another reactor for sorbent regeneration, the calcination process. In this process step, the calcium carbonate is heated to a temperature in

excess of 800°C (800°C-950°C) releasing CO<sub>2</sub> at the same time as the lime sorbent (CaO) is regenerated. The sorbent (CaO) is then transferred to the CO<sub>2</sub> capture reactor.



- 10 Calcium-looping has a number of advantages compared to closer-to-market CO<sub>2</sub> capture schemes (see Chemical Engineering Research and Design 89 (2011),: Review: *The calcium looping cycle for CO<sub>2</sub> capture from power generation, cement manufacture and hydrogen production*, Dean et al.). Post-combustion applications of Ca-looping are applicable in power production from natural gas (CH<sub>4</sub>), gasified coal (syn-gas) and in a range of different high temperature industry processes.
- 15 However, new technology focused on energy optimization (production of more el-power, with the same energy input), energy efficient CO<sub>2</sub> capture (by the Ca-looping method) and utilization of waste heat from high temperature industries, such as iron/steel, aluminium, silicon, ferrosilicon, cement, manufacturing and others, is needed.

### Objectives

- 20 It is the object of the present invention to further improve the methods of the inventions according to WO 2004/025767 and WO 2011/078681 A1, providing a method even more cost and energy efficient.

### The present invention

The object mentioned above is fulfilled by the present invention as defined by claim 1.

- 25 Preferred embodiments of the invention are disclosed by the dependent claims.

As will be seen by a person skilled in the art, the present method maintains the beneficial features of the prior inventions commented above, but involves a combination with other high temperature producing flue gas processes, making the overall process even more cost and energy efficient.

Thus, the exothermic nature of the carbon capture process in which CaO is converted to CaCO<sub>3</sub>, is utilized to further increase the temperature of the high temperature flue gas, making this flue gas even more useful for subsequent energy recovery utilizations.

Sustainability is still a keyword and common denominator for the overall process.

- 5 By "another process" as used herein is understood a process that runs principally independent of the energy production in the SOFC, and typically are conducted for the productions of mechanical goods like iron, steel, cement or the like. As such, the "another process" will typically be a process known per se, but not in the context here presented.

#### Detailed description of the invention

- 10 Different embodiments of the invention are illustrated below with reference to the to the enclosed drawings, where;

Fig. 1 gives a schematic view of a combined-cycle, gas-fired power plant without a CO<sub>2</sub>-capture facility.

Fig. 2 gives a schematic view of the present invention related to power production.

- 15 Fig. 3 and 3a, gives a schematic view of the present invention related to exhaust gases from high temperature industries (such as iron/steel, aluminium, silicon, ferrosilicon, other metals, cement, etc.), with total CO<sub>2</sub> capture

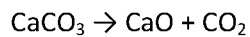
Fig. 4 gives a schematic view of the present invention related to power production combined with production of hydrogen.

- 20 Fig. 5 and 5a, gives a schematic view of the present invention related to exhaust gases from high temperature industries (such as iron/steel, aluminium, silicon, ferrosilicon, other metals, cement, etc.), with total integrated CO<sub>2</sub>-capture, combined with production of hydrogen.

Fig. 6 shows a variant of the embodiment shown in Figure 3, the exhaust gas being provided at a lower temperature.

- 25 Compared to the simple (general) outline of a combined cycle, gas-fired power plant (fig. 1), figure 2 illustrates a cost- and energy-efficient method for CO<sub>2</sub> capture, combined with production of more power (electricity) from the same fuel input to the combined gas-fired power facility. Figure 2 shows that natural gas (CH<sub>4</sub>) or syn-gas and air is fed to the gas turbine, producing power/electricity to the market. The hot exhaust (approx. 500°C) with CO<sub>2</sub> (3-5%) is transferred to a reactor (fluidized bed) where fine powdered hot lime (CaO) reacts with the CO<sub>2</sub> to form calcium carbonate. This reaction (CaO + CO<sub>2</sub> → CaCO<sub>3</sub>) is exothermic (with delta H° = +178 kJ/mol, Dean, C.
- 30

et al., 2011) and results in a temperature increase (500°C → 700°C or more), of the remaining gas (mainly N<sub>2</sub>, O<sub>2</sub>), and are thus providing additional energy to drive the steam cycle (production of more high pressure steam) by well-known technology. The CO<sub>2</sub> concentration in the flue gas from a NG fired power plant is typically 3-5%, giving a temperature increase of approximately 160°C-  
 5 260°C. The CO<sub>2</sub> concentration in the flue gas from a pulverized coal fired power plant is typically 10-15%, giving a temperature increase of approximately 480°C-660°C. The CO<sub>2</sub> concentration in the flue gas from an Integrated Gasification Combined Cycle fired power plant is typically 30-32%, giving a temperature increase of more than 1000°C. In the last two cases the CO<sub>2</sub> capture will have to be performed with integrated cooling/ heat exchange or in two or more steps to keep the  
 10 temperature sufficiently low to enable absorption. The absorption will stop when the temperature increase to the equilibrium temperature for the given CO<sub>2</sub> concentration. The equilibrium for the reaction



is given in Table 1.

15	550 °C	0.055	kPa
	587 °C	0.13	kPa
	605 °C	0.31	kPa
	680 °C	1.80	kPa
20	727 °C	5.9	kPa
	748 °C	9.3	kPa
	777 °C	14	kPa
	800 °C	24	kPa
	830 °C	34	kPa
25	852 °C	51	kPa
	871 °C	72	kPa
	881 °C	80	kPa
	891 °C	91	kPa
	898 °C	101	kPa
30	937 °C	179	kPa
	1082 °C	901	kPa
	1241 °C	3961	kPa

Table 1. Absorption equilibrium temperature and pressure

The calcium carbonate formed in the CO<sub>2</sub> capture reactor (carbonator) is transferred to the  
 35 calciner (see fig. 2) for regeneration of the CaO sorbent and release of CO<sub>2</sub> for storage or use, according to the equation;



(The heat exchange to cool hot CaO (to 650 ±150° C) from the Calciner, and to preheat the CaCO<sub>3</sub> before the calcination process is, for simplicity not shown in fig. 2.)

The heat necessary for the calcination is according to the present invention provided by Solid Oxide Fuel cells (SOFC, see fig. 2) instead of for example oxy-combustion of coal (Dean et al., 2011). Natural gas may be used as the primary energy source for the SOFC (see fig. 2). However, other gases, such as syngas or hydrogen (used in different embodiments) as suggested in (WO 2011/078681 Al, Raaheim and Vik) are alternative possibilities.

The CO<sub>2</sub> capture from the SOFC is arranged to be performed by combustion of the remaining fraction of fuel in the anode exhaust gas from the fuel cell in pure oxygen. The oxygen can be obtained by the use of an oxygen pump. It is worth noticing that this CO<sub>2</sub> capture method reduces the efficiency by 2-3% compared to a reduction between 5 and 10% by more conventional methods (WO 2011/078681 Al, Raaheim and Vik).

The performance of the SOFC is guided, or arranged, towards maximum production of heat (50-70%) and minimum production of electricity (30-50%). The production of heat from the SOFC would be in accordance with the heat energy required for the Ca-looping process, or the amount of CO<sub>2</sub> in the hot exhaust gas from the gas turbine. The co-produced electricity is delivered to the market, together with electric power produced by the gas and steam turbines (see fig. 2).

There are, however, other high temperature industrial situations (or industries), where it is a major challenge to obtain cost-efficient CO<sub>2</sub> capture.

These challenges can be met by the present invention.

Important stationary industrial CO<sub>2</sub> point source emitters, with hot exhaust or waste gases, are industries where metal oxides are reduced to metals by carbon as the reducing agent. GHG's are thus not only a penalty for the energy consumption (from fossil fuel) in high temperature industries, such gases are also present in the hot exhaust gases from the oxide to metal reduction itself. It should be noted that these emissions are always present regardless of the type of energy (fossil or renewable).

Important industries with hot exhaust gases where the cement industry represent a special case are as follows;

- 1 Iron (and steel) production normally has hot waste gases at 400-500°C. The CO<sub>2</sub> is derived both from the reduction of the oxide ore (Fe<sub>2</sub>O<sub>3</sub>, Fe<sub>3</sub>O<sub>4</sub>) and from limestone often used for slag generation ( $\text{CaCO}_3 \rightarrow \text{CaO} + \text{CO}_2$  and  $\text{CaO} + \text{SiO}_2 \rightarrow \text{CaSiO}_3$  (slag)).
- 2 Aluminium is formed at about 900°C. Exhaust carbon anode gases have a high temperature (about 600°C) and accounts for about 50% of total direct CO<sub>2</sub> equivalent emissions from aluminium production, with the remaining GHG's emitted being PFCs, including tetrafluoromethane (CF<sub>4</sub>) and hexafluorethane (C<sub>2</sub>F<sub>6</sub>). These pollutants (PFCs)

occur during "anode effects" caused by the depletion of the alumina content of the cell and have in recent years been dramatically reduced by the introduction of automated point feeders to ensure regular and sufficient feed of alumina to each cell in the pot line.

3 Silicon and ferrosilicon have hot waste gases at about 650°C. The exhaust gas also contains  
5 some NO<sub>x</sub> and SO<sub>2</sub>, with minor CO, CH<sub>4</sub>, VOC, N<sub>2</sub>O.

4 The hot exhaust gas(es) from the energy-demanding cement industry is mainly (80%)  
caused by decarbonation and decarbonation-fuel to release the CO<sub>2</sub> from limestone  
(CaCO<sub>3</sub> → CaO + CO<sub>2</sub>, occurring at about 900°C), Dean et al. (2011).

Similarly to the gas turbine case, the hot exhaust gases results in a temperature increase when  
10 CaO reacts with CO<sub>2</sub>. This, in turn, gives more heat recovery, steam generation and finally power  
production, as schematically illustrated in figure 3. Note that the CO<sub>2</sub> from the power production  
(electricity from SOFC and steam turbine) and the CO<sub>2</sub> from the metal reduction can both be  
captured by the same Ca-looping process (see fig. 3) and contributes strongly to a total integrated  
energy-efficient CO<sub>2</sub>-capture.

15 For some industrial processes the CO<sub>2</sub>-concentration in the hot exhaust or flue gas, may- as  
stated earlier - be too high for efficient CO<sub>2</sub>-capture and heat recovery in one step. The CO<sub>2</sub>-  
capture by the Ca-looping method and heat recovery for steam generation may thus be performed  
in two or more stages, depending on the CO<sub>2</sub>-capture efficiency at each stage, as shown in figure  
3a.

20 In order to obtain the highest efficiency for the thermal power generation cycle the carbonation  
reaction should preferably take place at the highest possible temperature according to table 1. For  
industrial processes with lower than optimal temperature the temperature can be increased by a  
heat exchanger.

Figure 6 shows how low temperature CO<sub>2</sub> containing exhaust gases can be used in the present  
25 invention. The exhaust gas is first heated in a heat exchanger to a temperature at which the CO<sub>2</sub>  
capture process proceeds at an acceptable reaction rate (approximately 500°C) and the resulting  
temperature increase (500°C → 700°C or more) can be utilized more efficient in a steam cycle for  
electricity production. After heat recovery the exhaust gas is fed to the heat exchanger to heat the  
incoming exhaust gas.

30 The captured CO<sub>2</sub> may be stored in for instance suitable geological formations, which, however,  
always result in an additional cost penalty. The CO<sub>2</sub> should thus preferably be used for industrial  
purposes.

If hydrogen is needed as a reducing agent, for example in the production of special metals (tungsten), or for nearby industries such as, refineries or chemical production plants, it is an option to combine the Ca-looping with hydrogen production, by adding a hydrocarbon gas (natural gas) or syn-gas formed from any carbonaceous material (together with a catalyst) and water to the carbonate forming process as illustrated in fig. 4, 5 and 5a. The hydrogen is in this case produced by the so-called sorption enhanced steam methane reforming process (SE-SMR), where the Ca-looping also represents an important part of an energy-efficient hydrogen production (Industrial and Engineering Chemistry Research 40 (23) 2001, 5102-5109, Hydrogen production using sorption-enhanced reaction, Ortiz and Harrison; Dean et al. (2011), WO 2004/025767 (Vik et al, (2004) and WO 2011/078681 A1 (Raaheim and Vik, 2011).)

The catalyst could be prevented from contamination/degradation by the hot industry flue gases, or power production exhaust gases (f. ex.  $\text{SO}_2$  or  $\text{NO}_x$ ), by size or magnetic separation between reactor 1 and 2 (see fig. 4, 5 and 5a). The total amount of natural gas (or syn-gas) may be reformed to hydrogen and used to feed the SOFC. If hydrogen is needed for other purposes it would be taken out from the SOFC as shown in fig. 5a

In figures 4, 5 and 5a, it is seen that the Ca-looping process is split in two, where one part of the Ca-looping provides CaO to the hydrogen production and the other part provides CaO for efficient  $\text{CO}_2$  capture from the gas turbine power production. It is also seen that the heat for the energy-demanding calcination process (Reactor 2, in figs. 4, 5 and 5a) for both splits are provided by the SOFC.

### Preferred embodiments

The high temperature industrial process combined with the SOFC based energy production can be any process generating hot  $\text{CO}_2$  containing flue gas and is preferably one chosen among processes for manufacturing of iron, steel, aluminium, silicon, ferrosilicon and cement. Be conducted in a number of ways, one of which is in a steam generator, said steam being used for subsequent production of electricity.

The temperature of the hot gas from another process may be raised by heat exchange when feasible, thus increasing the temperature from a lower temperature to a temperature above 500 °C.

The SOFC may be powered by a number of energy rich gases, among which methane containing gases, syn-gas and hydrogen are preferred, resulting in the production of heat, electricity and optionally hydrogen.



### Claims

1. Method for sustainable energy production in a plant comprising a solid oxide fuel cell (SOFC) in which method calcium oxide is being used as a sorbent in an exothermic carbon capture step by which calcium oxide is converted to calcium carbonate, the calcium carbonate subsequently being  
5 converted to calcium oxide in an endothermic regeneration step (heated by the SOFC),  
**characterized in** that the carbon capture step is performed in the presence of hot gas from another process, thus increasing the temperature of the hot gas thereby improving the cost-efficiency of said other process.
2. Method as claimed in claim 1 wherein said hot gas from another process is exhaust gas from a  
10 gas turbine.
3. Method as claimed in claim 2 wherein the temperature increased exhaust gas is used for heat recovery.
4. Method as claimed in claim 3 wherein the heat recovery takes place in a steam generator, said steam being used for subsequent production of electricity.
- 15 5. Method as claimed in claim 4 wherein said steam is high pressure steam.
6. Method as claimed in claim 1, wherein said hot gas from another process is exhaust gas from a high temperature industrial process.
7. Method as claimed in claim 6 wherein said high temperature industrial process is a process chosen among manufacturing of iron, steel, aluminium, silicon, ferrosilicon and cement.
- 20 8. Method as claimed in claim 6 or 7 wherein the temperature increased exhaust gas is used for heat recovery.
9. Method as claimed in claim 8 wherein the heat recovery takes place in a steam generator, said steam being used for subsequent production of electricity.
10. Method as claimed in claim 1, wherein said hot gas from another process is obtained by heat  
25 exchanging low temperature exhaust gas from an industrial process.
11. Method as claimed in claim 10 wherein said industrial process is a process chosen among manufacturing of iron, steel, aluminium, silicon, ferrosilicon and cement.
12. Method as claimed in claim 10 or 11 wherein the temperature increased exhaust gas is used for heat recovery.

13. Method as claimed in claim 12 wherein the heat recovery takes place in a steam generator, said steam being used for subsequent production of electricity.
14. Method as claimed in claim 13 wherein said steam is high pressure steam.
15. Method as claimed in any one of the preceding claims, wherein the SOFC is powered by a gas  
5 chosen among a methane containing gas, hydrogen, and syn-gas.
16. Method as claimed in any one of the preceding claims, wherein the energy produced by the SOFC is heat, electricity and optionally hydrogen.

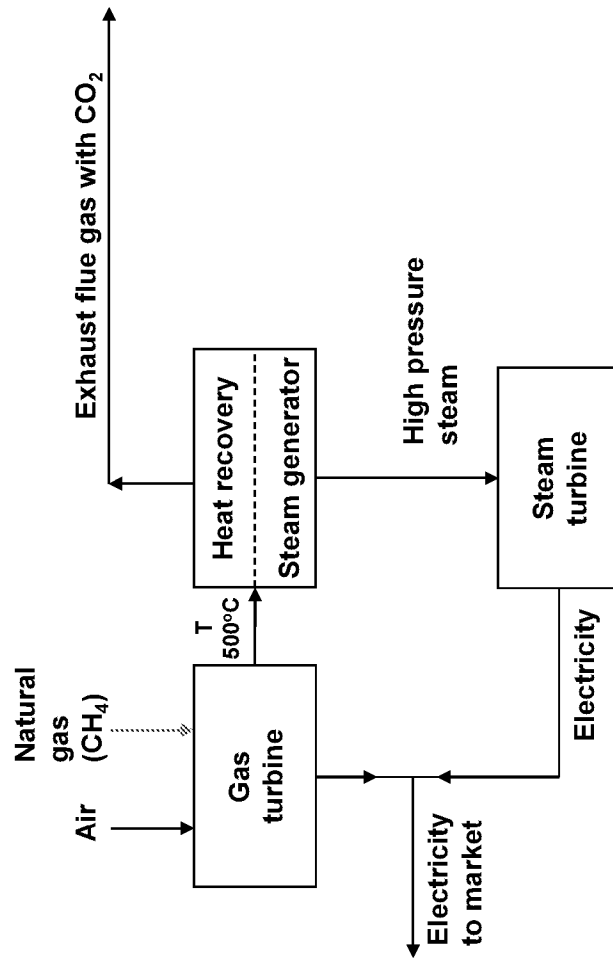


Fig. 1

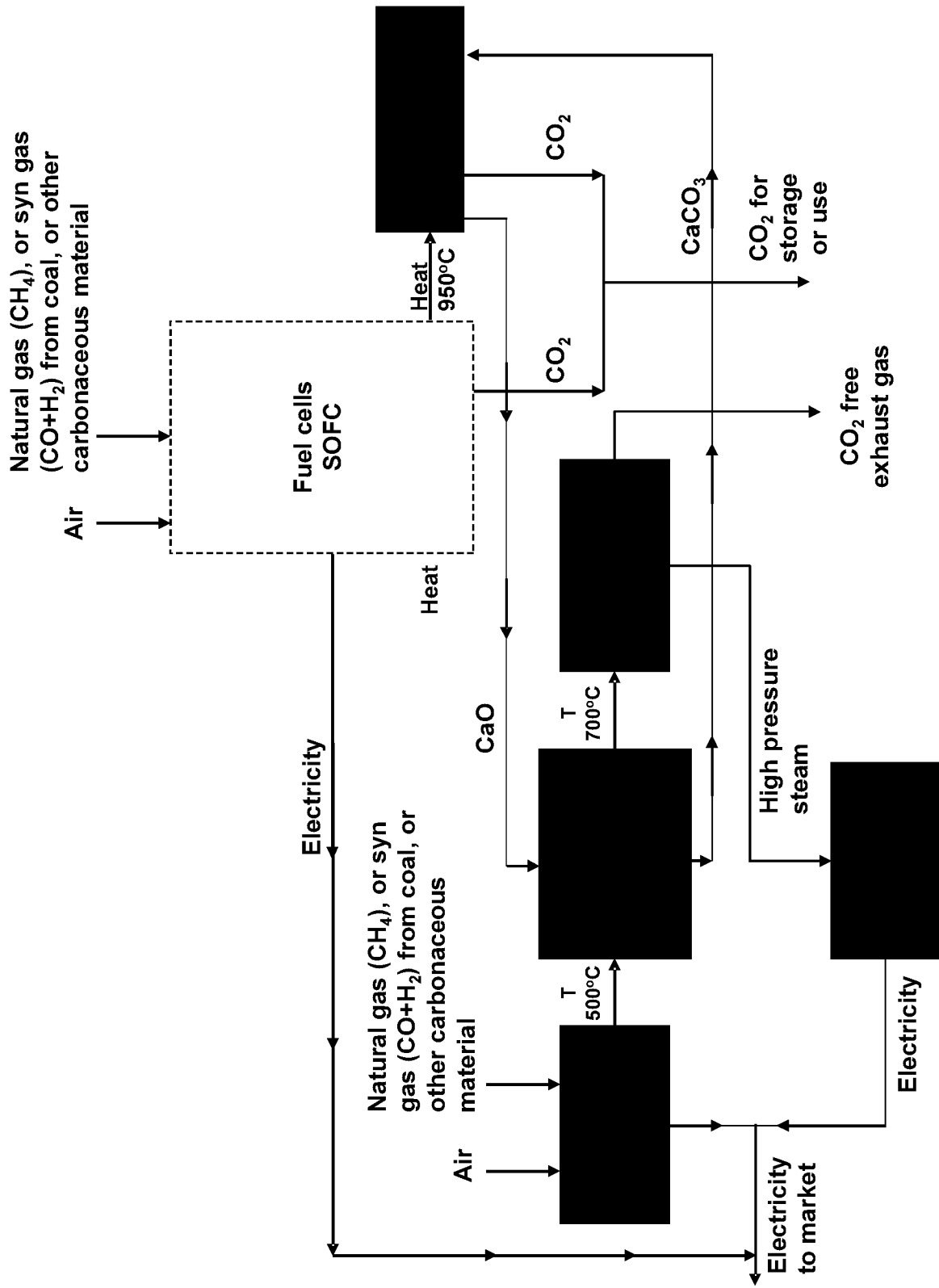


Fig. 2

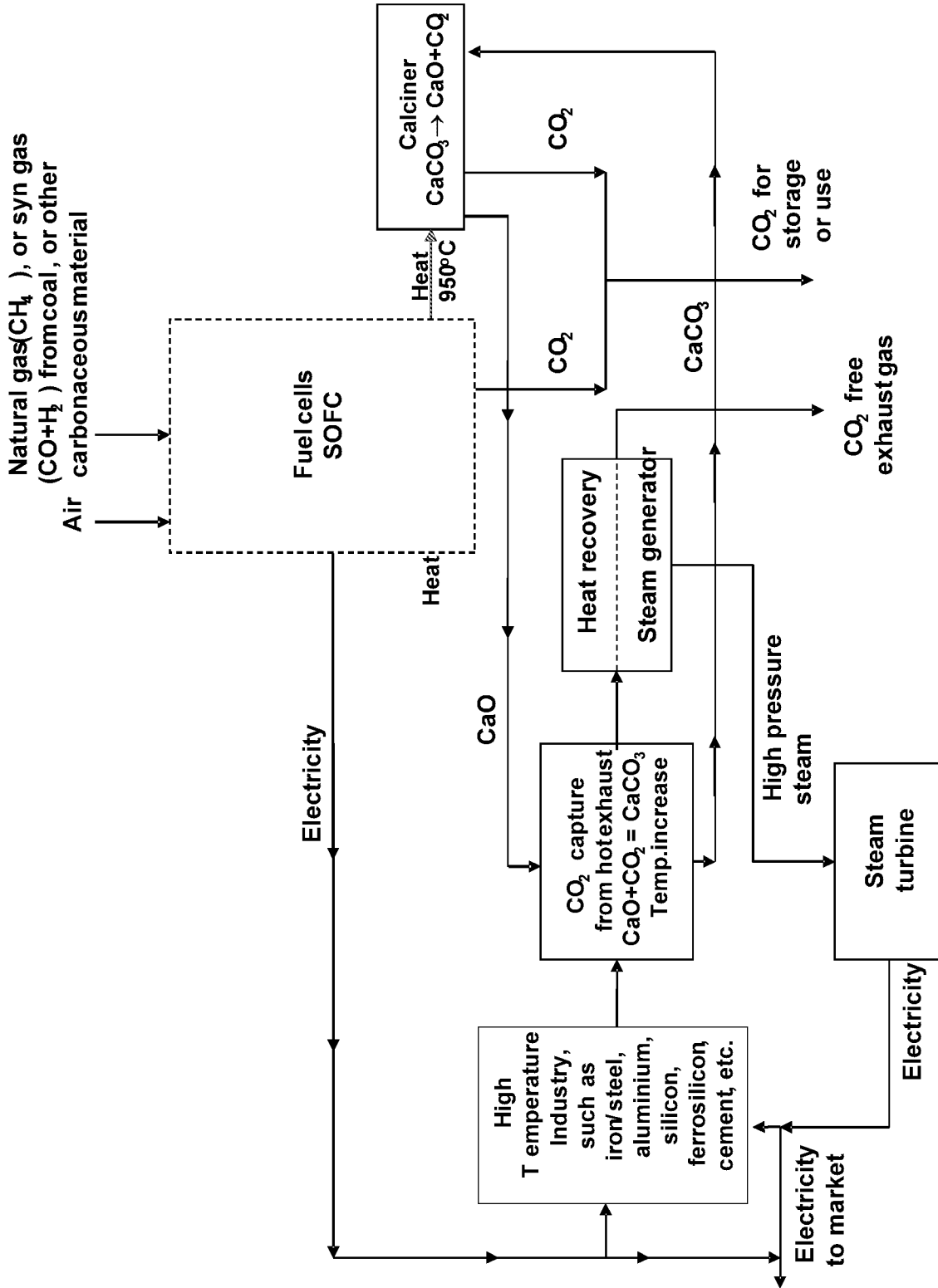


Fig. 3

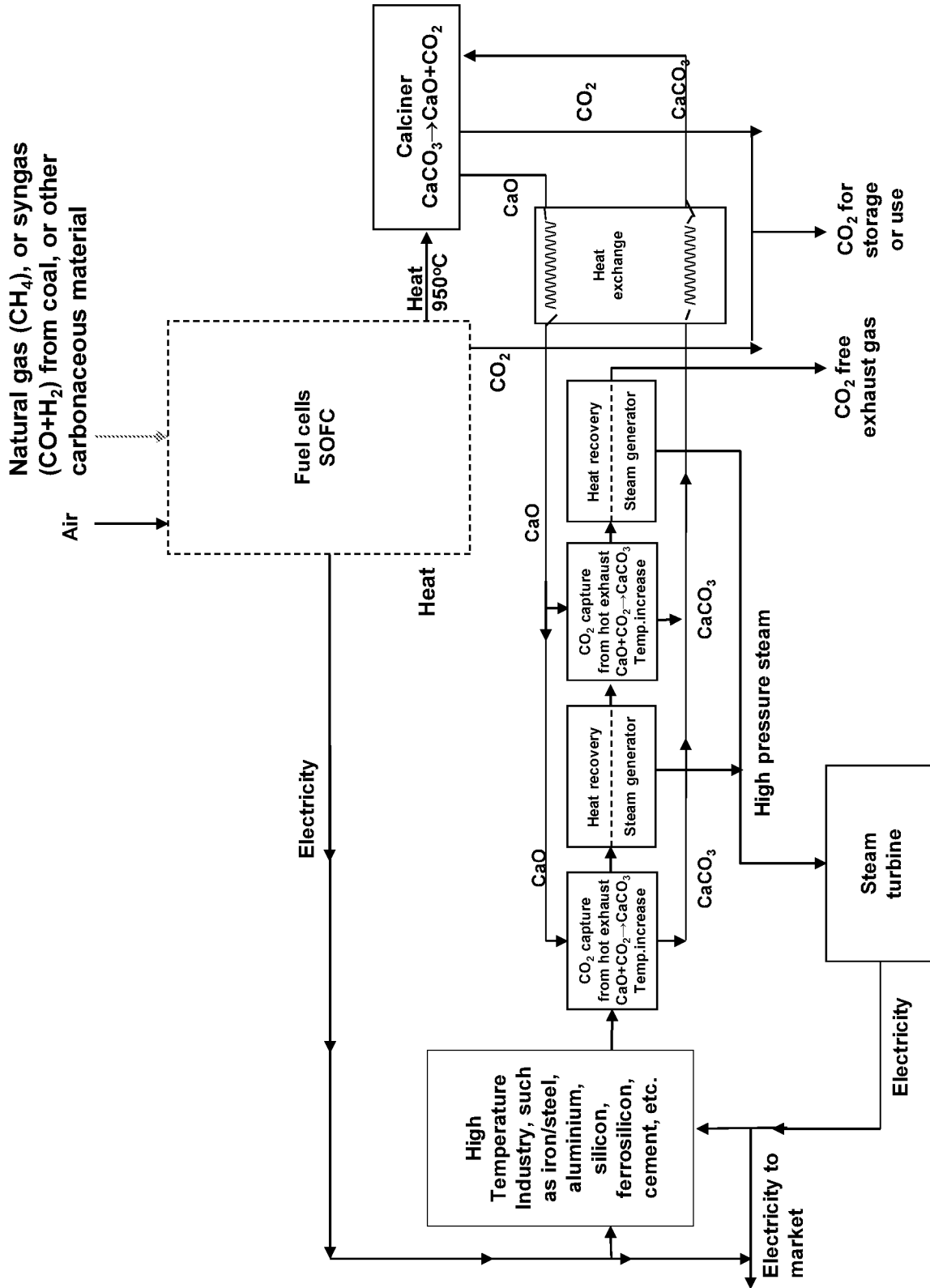


Fig. 3a

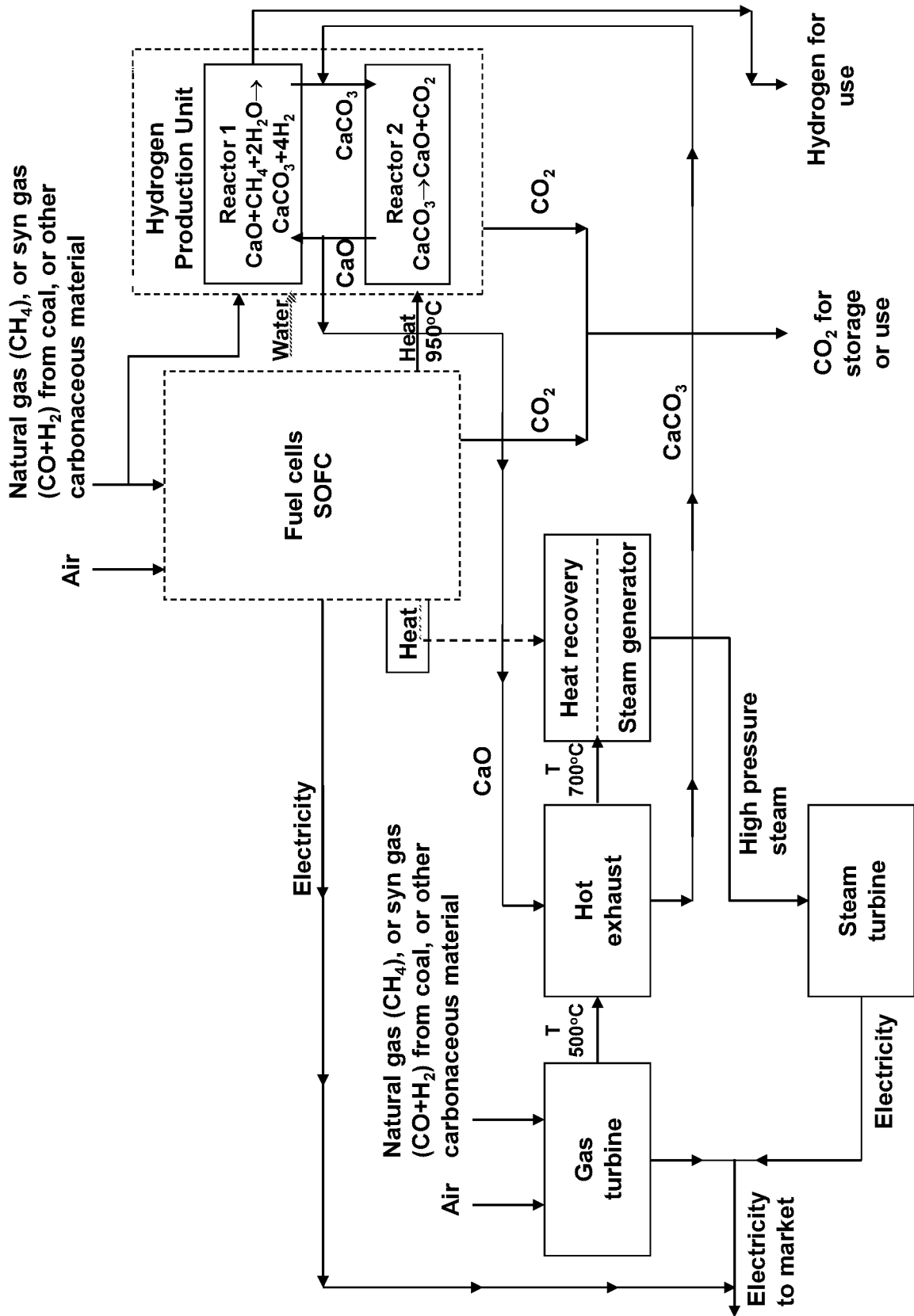


Fig. 4

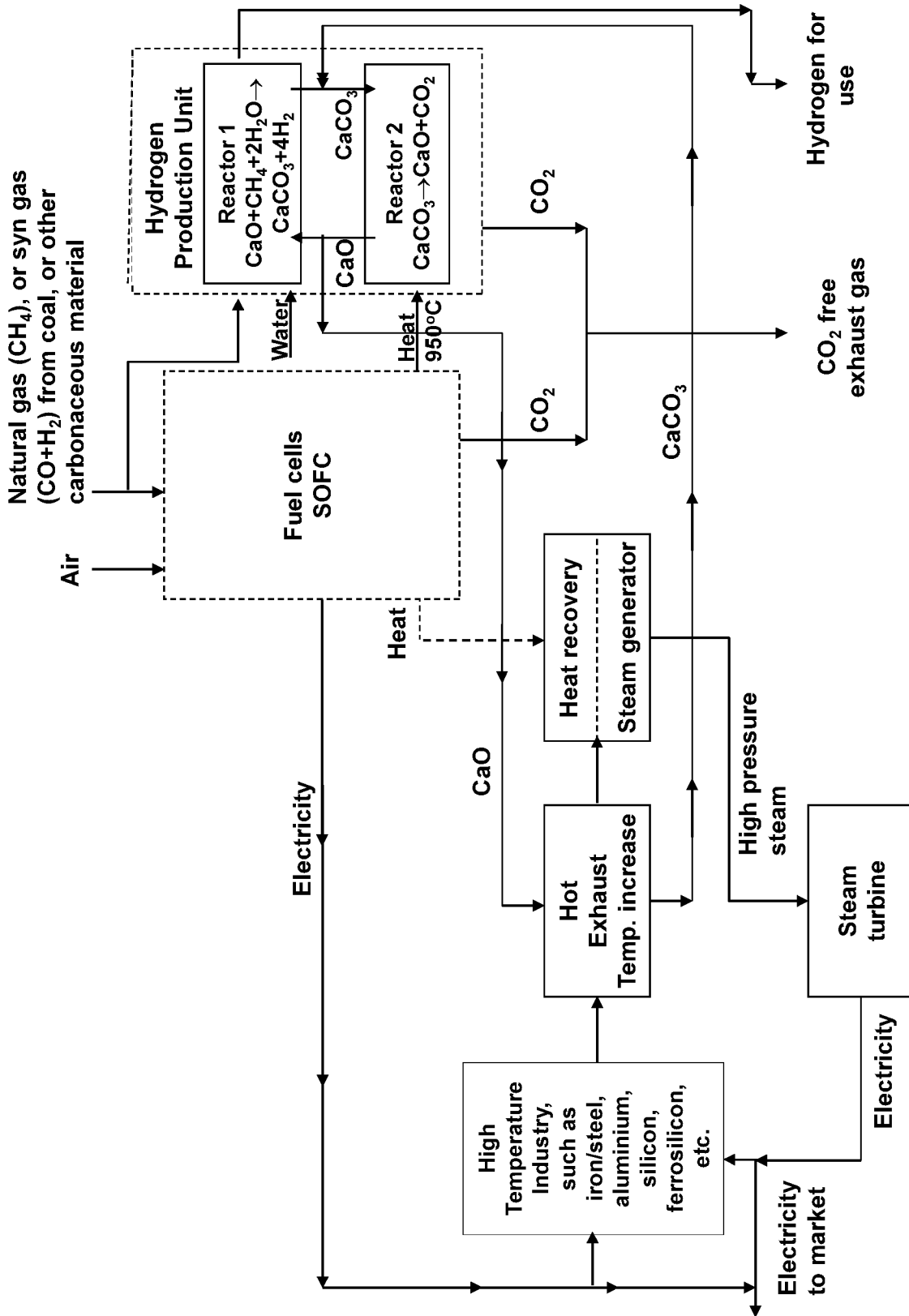


Fig. 5



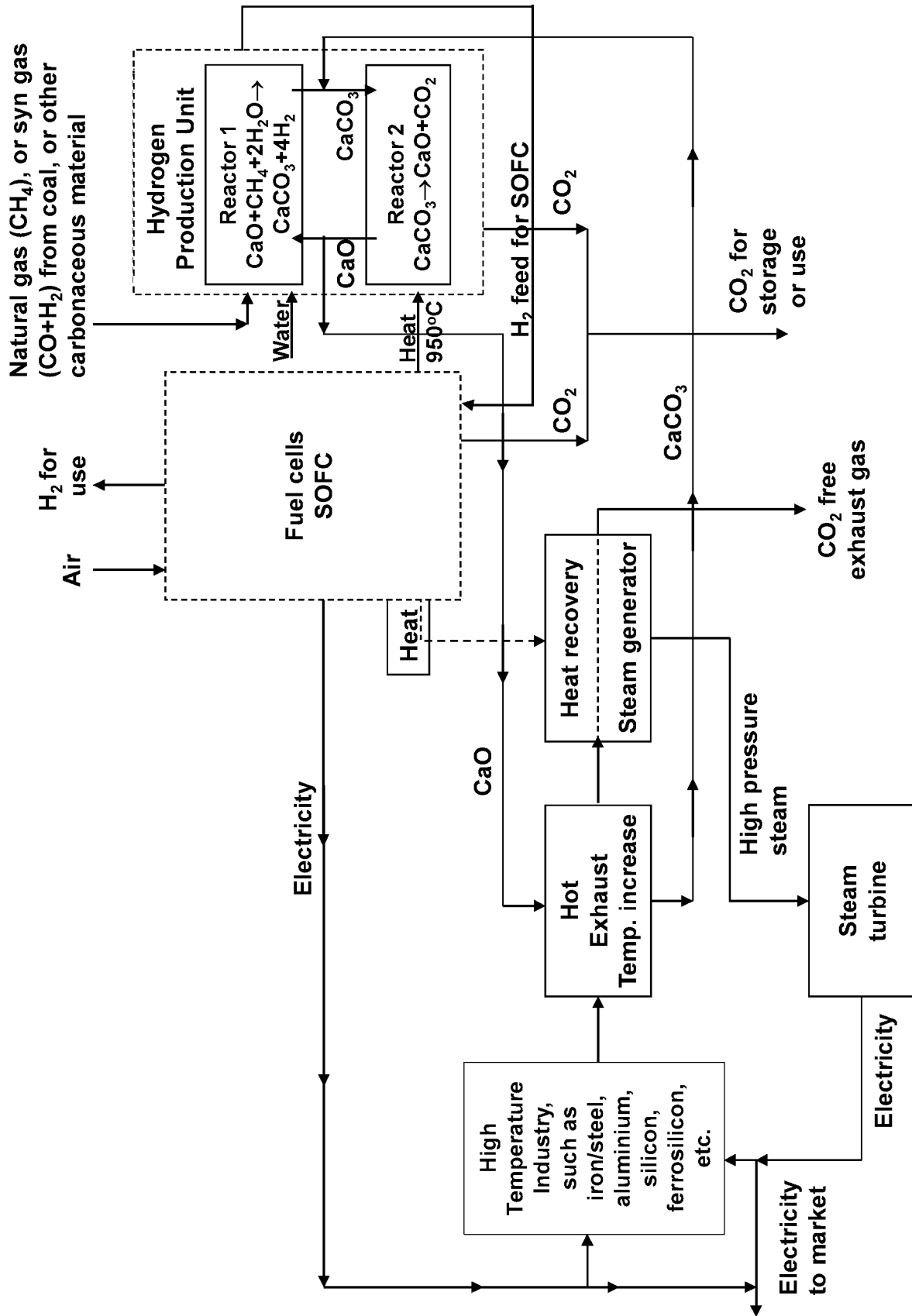


Fig. 5a

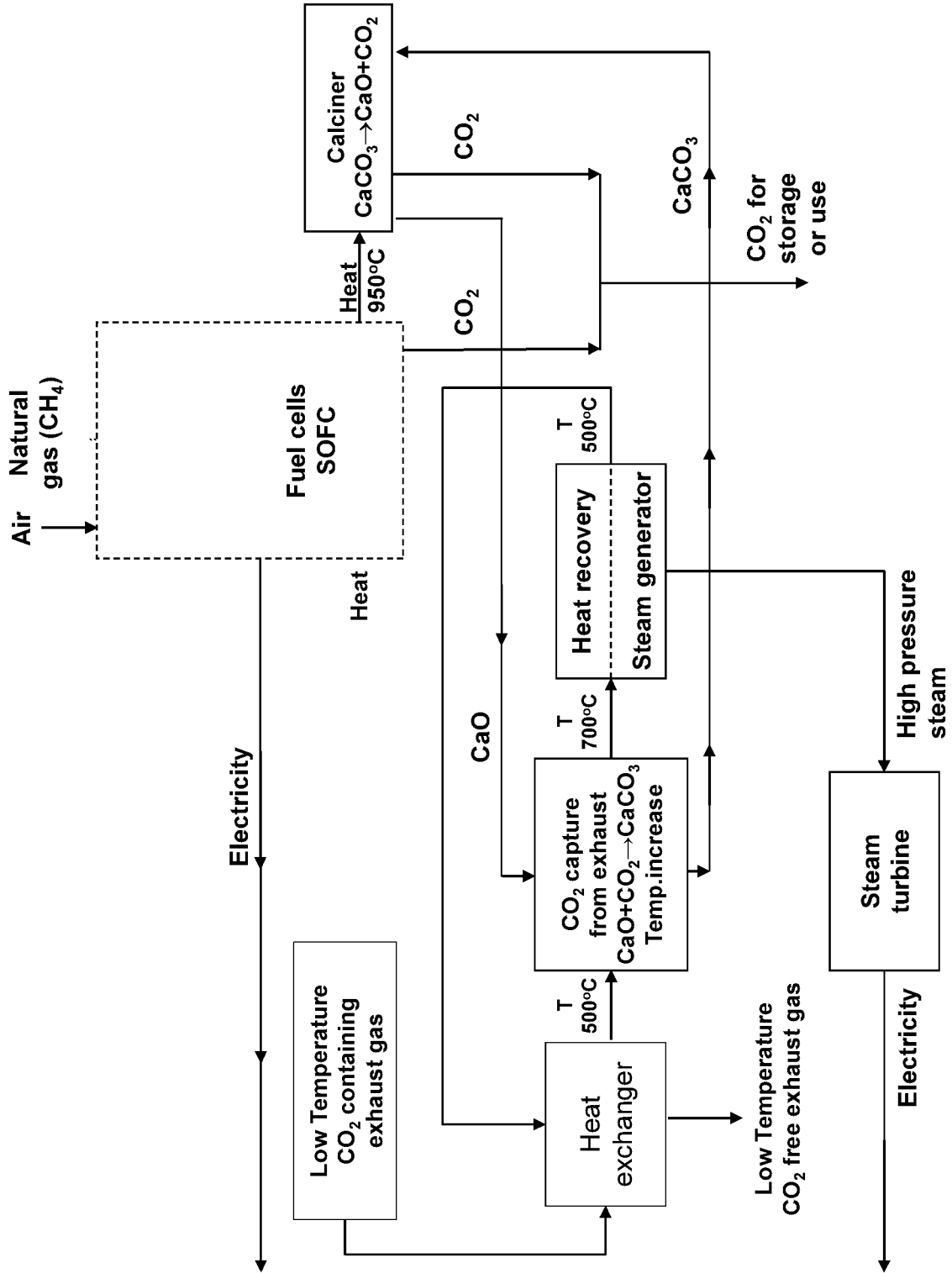


Fig. 6

## INTERNATIONAL SEARCH REPORT

International application No.

PCT/NO2014/050100

A. CLASSIFICATION OF SUBJECT MATTER B01D 53/14 (2006.01), B01D 53/34 (2006.01), B01D 53/62 (2006.01), B01D 53/74 (2006.01), B01D 53/96 (2006.01), B01J 20/04 (2006.01), B01J 20/34 (2006.01), C22B 21/00 (2006.01), C22C 33/00 (2006.01) 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) B01D, B01J, C22B, C22C		
Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched DK, NO, SE, FI: Classes as above.		
Electronic data base consulted during the international search (name of data base and, where practicable, search terms used) EPODOC, WPI, FULL TEXT: ENGLISH		
C. DOCUMENTS CONSIDERED TO BE RELEVANT		
Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	WO 01/42132 A1 (2001.06.14) (The Regents of the University of California), see page 1 line 6-7, page 6 line 1- page 7 line 14, page 8 line 8- page 10 line 2, page 13 line 8-10, page 19 line 4- page 21 line 18.	1-16
A	WO 2012/070954 A1 (2012.05.31) (Universitetet for miljø- og biovitenskap), see the whole document.	1-16
AD	WO 2011/078681 A1(2011.06.30) (Institutt for Energiteknikk), see the whole document.	1-16
<input type="checkbox"/> Further documents are listed in the continuation of Box C. <input type="checkbox"/> See patent family annex.		
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**INTERNATIONAL SEARCH REPORT**

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