

(54) Title: METHOD FOR THE INTEGRATED OPERATION OF A FUEL CELL AND AN AIR SEPARATOR

(57) Abstract: The invention relates to a method for the integrated operation of a fuel cell and an air separator, comprising the steps of supplying fuel to an anode of the fuel cell and supplying an oxidiser gas to the cathode of the fuel cell. The invention is characterised in that the electricity formed in the fuel cell is at least partly used in an air separator separating nitrogen and oxygen from air, and wherein the oxygen formed is supplied to the cathode of the fuel cell.
Method for the integrated operation of a fuel cell and an air separator

The present invention relates to a method for the integrated operation of a fuel cell and an air separator, comprising the steps of supplying fuel to an anode of the fuel cell and supplying an oxidiser-comprising gas to the cathode of the fuel cell.

Such a method is generally known in the art. For example, when a fuel such as natural gas (methane CH₄) is supplied to an anode of a fuel cell and an oxidiser such as oxygen is supplied to a cathode of that fuel cell, oxidation of the fuel will take place in the fuel cell with the aid of the supplied oxygen. In the case of natural gas a complete conversion may take place, producing CO₂ and H₂O. These gases are released as gas stream from the anode. An optional possibility is an incomplete conversion of, for example, the natural gas, whereby carbon monoxide (CO) and hydrogen (H₂) are produced. These gases also will be released as gas stream from the anode.

The object of the invention is to provide an improved method of the kind mentioned in the preamble.

It is also an object of the invention to, at a natural-gas mixing station, use a fuel cell in such a manner that by means of the fuel cell the costs for bringing the natural gas to a suitable calorific value and Wobbe Index, are greatly reduced.

It is also an object of the invention to greatly reduce the use of natural gas in general.

Another object of the invention is to increase the output of an air separator.

An additional object of the invention is to use a fuel cell in an efficient manner when extracting natural gas.

In order to achieve at least one of the above-mentioned objectives, the invention provides a method as mentioned in the preamble, characterised in that the electricity formed in the fuel cell is at least partly used in an air separator separating nitrogen and oxygen from air, and wherein
oxidiser. This affords the advantage that when separating nitrogen from air, no or less gas needs to be ventilated or stored. According to the prior art, the oxygen produced in the air separator was ventilated to the outside air. In some cases, where the produced oxygen was stored, costs were incurred for its storage and its transport, causing the profit to dwindle.

The invention provides the advantage that all the gasses that are produced are utilised while no transport costs are incurred.

It was further shown that much more electricity was produced in the fuel cell when using oxygen formed in the air separator than was required for the separation of that amount of oxygen. The surplus electricity may be supplied to the electricity grid.

According to a preferred embodiment, the nitrogen formed is supplied to a natural-gas stream whose calorific value is higher than a standard calorific value, so as to reduce that calorific value to the standard calorific value. The nitrogen stream obtained with an air separator, for example, a cryogenic air separator as known in the prior art, is suitable for being supplied to a natural-gas stream to be used by end-users.

Since the fuel cell will be installed at a natural-gas mixing station where nitrogen is usually supplied to the extracted natural gas, the fuel to be supplied to the anode of the fuel cell is preferably natural gas.

It is further preferred that the reaction in the fuel cell be chosen from complete conversion of the fuel, incomplete conversion of the fuel so as to form at least H₂ (hydrogen), and a combination of these. When natural gas is supplied as fuel, CO₂ and H₂O will be formed in a complete conversion. In a partial conversion of the natural gas carbon monoxide (CO) and H₂ will be formed. A possible option is a combination of these two types, allowing all the above-mentioned reactions to take place. This will also provide a so-called shift reaction between carbon monoxide and water, whereby carbon dioxide and hydrogen will be formed.
first fuel cell at least hydrogen is formed, said formed hydrogen is supplied to the anode of a second fuel cell. This second fuel cell may, for example, be a low-temperature fuel cell.

According to another embodiment, it is possible to supply the hydrogen produced in the fuel cell to the natural-gas stream. An ensuing reduction of the calorific value may optionally be augmented by also admixing nitrogen produced in the air separator if the original calorific value and/or Wobbe Index of the natural gas is higher than a desired value. Apart from the production of electricity and heat in the fuel cell, there is in that case also an increase in the volume stream of the natural gas. The heat produced in the fuel cell may suitably be used for heating the produced and still to be expanded natural-gas stream.

If the first fuel cell is used for the conversion of natural gas, said first fuel cell is preferably of the internal reforming type. A fuel cell of this type is a high-temperature fuel cell, for example, of the MCFC or the SOFC type. Such fuel cells as well as their use are generally known in the art. A further explanation concerning these fuel cells will therefore not be given.

Both the first fuel cell, which may for example be of the high-temperature internal reforming type, and the second fuel cell, which may for example be of the low-temperature type such as a polymer fuel cell, or of the PAFC type, may at the cathode be supplied with oxygen obtained from the air separator. Since this is pure O₂ (without CO₂) it is also possible to use an AFC. When the oxidiser gas being supplied to the cathode of the fuel cell is such pure oxygen, there is a voltage rise in the fuel cell compared with when the oxidiser is air. By Nernst's Law it is simple to work out that the voltage rise will be approximately 4-7\% depending on the operating temperature of the fuel cell.

It is also preferred that at least a part of the heat formed in the fuel cell or fuel cells be used for heating the nitrogen and oxygen formed in the air separator. Since such nitrogen and oxygen are produced cryogenically, they must be heated to approximately 10-20°C or higher before they can be
is suitable for the fuel cell before they are supplied to the fuel cell. When hydrogen or nitrogen produced in the fuel cell is supplied to a natural-gas stream, the heat it carries from the fuel cell may be used to prevent condensable compounds present in the natural gas from condensing during the expansion of the natural gas at the gas extraction station. This reduces the consumption of natural gas, which so far in the prior art is combusted to provide heat for the natural gas to be expanded.

It is also preferred that the cathodic gas stream be subjected to a further treatment in order to remove the residual oxygen therefrom, for example, by supplying it to a catalytic oxidiser and subsequently supplying at least part of the treated gas stream to the natural-gas stream.

It is also preferred that the cathodic gas stream be supplied to the cathode of a low-temperature fuel cell.

It is further preferred that hydrogen from an anodic gas stream from the first fuel cell be supplied to the anode of a second fuel cell, and a cathodic gas stream from the first fuel cell, which compared with air has a reduced oxygen content, be supplied to the cathode of the second fuel cell.

It is also preferred that the fuel cell to which oxygen or air is supplied be a high-temperature fuel cell, preferably of the internal reforming type, for example an SOFC type or an MCFC type.

In addition it is preferred that at least a portion of the produced hydrogen be admixed with a natural-gas stream, and at least a portion of the produced electricity be used in the air separator.

Finally, it is preferred that at least a portion of the electricity formed in the fuel cell be supplied to the electricity grid.

The invention is not limited to the above description of a preferred embodiment, wherein the fuel used is natural gas. Other fuels may also be used in the method according to the present invention. The invention is limited by the appended claims only.
CLAIMS

1. A method for the integrated operation of a fuel cell and an air separator, comprising the steps of supplying fuel to an anode of the fuel cell and supplying an oxidiser-comprising gas to the cathode of the fuel cell, characterised in that the oxygen formed in an air separator is supplied to the cathode of a fuel cell as oxidiser.

2. A method according to claim 1, characterised in that the electricity formed in the fuel cell is at least partly used in an air separator separating nitrogen and oxygen from air.

3. A method according to claim 1 or 2, characterised in that the nitrogen formed is supplied to a natural-gas stream whose calorific value is higher than a standard calorific value, so as to reduce that calorific value to the standard calorific value.

4. A method according to claim 1 or 2, characterised in that the fuel supplied to the anode of the fuel cell is natural gas.

5. A method according to one or several of the preceding claims, characterised in that the reaction in the fuel cell is chosen from complete conversion forming CO₂ and H₂O, incomplete conversion forming CO and H₂ (hydrogen), and a combination of these.

6. A method according to claim 5, wherein in a first fuel cell at least H₂ is formed, characterised in that at least part of the formed H₂ is supplied to the anode of a second fuel cell.

7. A method according to claim 6, characterised in that the oxygen formed in an air separator is supplied to the cathode of the second fuel cell.

8. A method according to one or several of the claims 1 to 7, characterised in that at least a part of the heat formed in the fuel cell is used for heating the nitrogen and oxygen formed in the air separator.

9. A method according to claim 1, characterised in that the cathodic gas stream is subjected to a further treatment in order to remove the residual oxygen therefrom, for ex-
sequently supplying at least part of the treated gas stream to the natural-gas stream.

10. A method according to one or several of the claims 1 to 4, characterised in that the cathodic gas stream is supplied to the cathode of a low-temperature fuel cell.

11. A method according to claim 5, wherein in a first fuel cell at least hydrogen is formed, characterised in that hydrogen from an anodic gas stream from the first fuel cell is supplied to the anode of a second fuel cell, and a cathodic gas stream from the first fuel cell, which compared with air has a reduced oxygen content, is supplied to the cathode of the second fuel cell.

12. A method according to one of the preceding claims, characterised in that the fuel cell to which oxygen or air is supplied is a high-temperature fuel cell, preferably of the internal reforming type, for example an SOFC type or an MCFC type.

13. A method according to claim 5, wherein at least hydrogen is formed, characterised in that at least a portion of the produced hydrogen is admixed with a natural-gas stream.

14. A method according to one or several of the preceding claims, characterised in that at least a part of the electricity formed in the fuel cell is supplied to the electricity grid.
### INTERNATIONAL SEARCH REPORT

**International application No:** PCT/NL2006/000400

**A. CLASSIFICATION OF SUBJECT MATTER**

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<th>H01M8/04</th>
<th>H01M8/06</th>
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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

**C. DOCUMENTS CONSIDERED TO BE RELEVANT**

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Further documents are listed in the continuation of Box C.

**D. DOCUMENTS MEETING THE CONSIDERATION CRITERIA**

* Special categories of cited documents:
  *A* document defining the general state of the art which is not considered to be of particular relevance
  *E* earlier document but published on or after the international filing date
  *L* document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)
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*X* document of particular relevance: the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone

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*A* document member of the same patent family

**Date of the actual completion of the International search**

4 January 2007

**Date of mailing of the International search report**

12/01/2007

**Name and mailing address of the ISA**

European Patent Office, P.B. 5818 Patentlaan 2 NL-2280 HV Rijswijk Tel. (+31-70) 340-2040, Tx. 31 651 epos nl, Fax (+31-70) 340-3516

Authorized officer

Kiliaan, Sven
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