

(12) **UK Patent**

(19) **GB**

(11) **2497787**

(45) Date of B Publication

(13) **B**

26.06.2019

(54) Title of the Invention: **Hydrogen quality monitor**

(51) INT CL: **H01M 8/04089** (2016.01) **H01M 8/04537** (2016.01) **H01M 8/0662** (2016.01)

(21) Application No: **1122035.7**

(22) Date of Filing: **21.12.2011**

(43) Date of A Publication **26.06.2013**

(56) Documents Cited:

GB 2478829 A JP 2006059745 A
US 20070244601 A US 20060057034 A
US 20040247952 A US 20030057109 A

(58) Field of Search:

As for published application 2497787 A viz:

INT CL **H01M**

Other: **On line databases WPI,EPODOC**
updated as appropriate

Additional Fields

Other: **None**

(72) Inventor(s):

**Christopher James Kirk
Simon Edward Foster**

(73) Proprietor(s):

**Intelligent Energy Limited
Charnwood Building, Holywell Park, Ashby Road,
Loughborough, Leicestershire, LE11 3GB,
United Kingdom**

(74) Agent and/or Address for Service:

**Creation IP Limited
Hillington Park Innovation Centre, 1 Ainslie Road,
GLASGOW, G52 4RU, United Kingdom**

GB 2497787 B

Figure 1

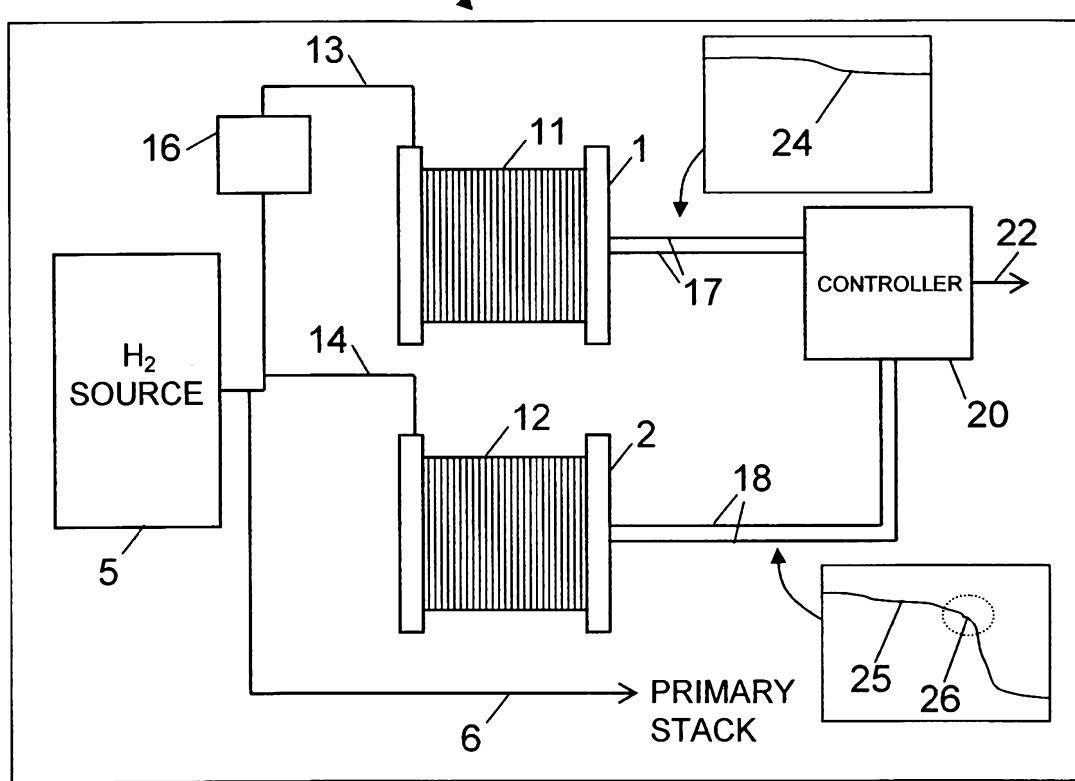
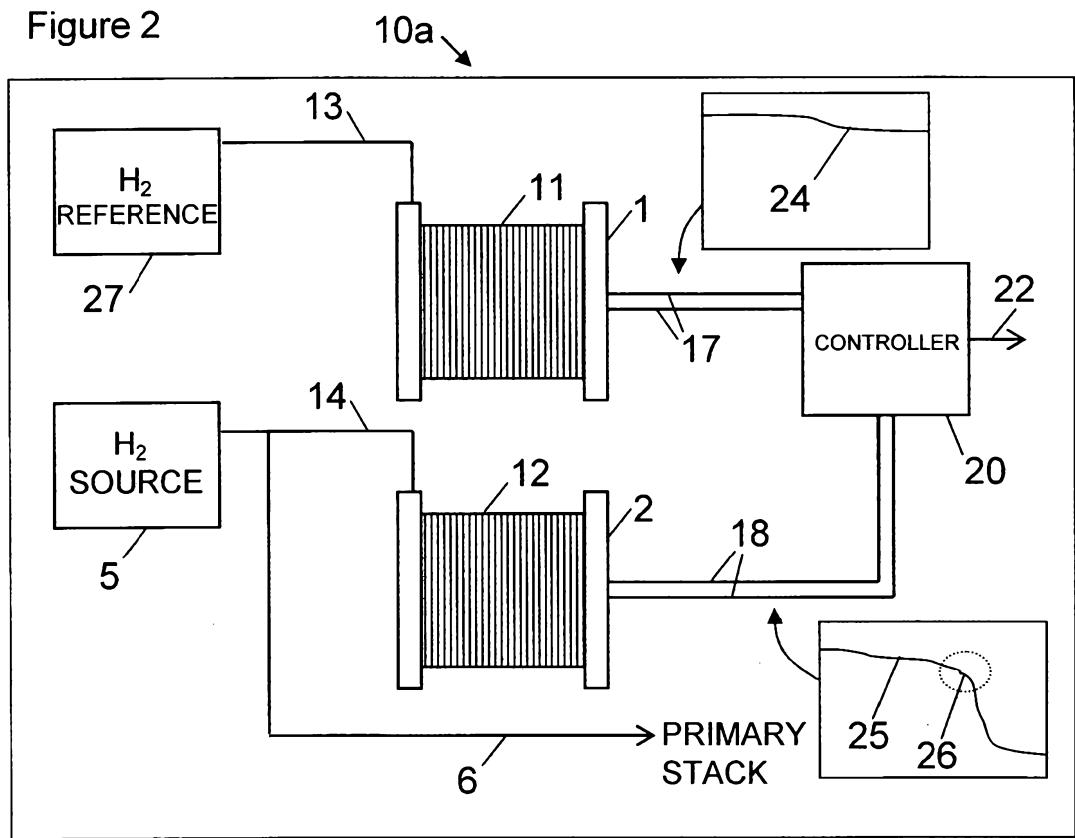


Figure 2



28 03 19

HYDROGEN QUALITY MONITOR

The present invention relates to apparatus and methods for monitoring the quality of a hydrogen supply.

5

The use of hydrogen as a fuel for the generation of electrical power in fuel cells is becoming of increasing importance. Purity of the hydrogen supply is important for optimal electrical power generation and for maintaining fuel cells using that hydrogen in optimal condition.

10

Currently, hydrogen used in fuel cell systems is often synthesized through the steam reforming of natural methane gas. Even where best quality practices are used, a number of contaminants may be present in the hydrogen fuel which are harmful to fuel cell operation. Although the harm is usually reversible, in the worst cases a high degree of contamination may be present including some compounds which may cause irreversible harm to the fuel cell.

15

It is an objective to provide a convenient hydrogen quality monitor that is particularly, though not exclusively, suited to monitoring fuel supplies to fuel cells.

20

The present invention provides a method of monitoring hydrogen purity, the method comprising:

25

supplying hydrogen fuel from a first, purified, hydrogen source to a first fuel inlet of a first fuel cell configured to generate electrical current from the electrochemical reaction of hydrogen and oxidant;

supplying hydrogen fuel from a second hydrogen source to a second fuel inlet of a second fuel cell configured to generate electrical current from the electrochemical reaction of hydrogen and oxidant;

30

applying an electrical load to each fuel cell and determining an electrical output of each fuel cell;

comparing the electrical outputs of the first and second fuel cells with a comparator; and

providing an indication of hydrogen purity of the second hydrogen source based on an output of the comparator;

wherein a first hydrogen purification device is provided, coupled between the first fuel inlet and the second fuel inlet, to receive hydrogen from the second hydrogen source, and to provide purified hydrogen from the second hydrogen source to the first fuel inlet via the purification device, as the first, purified, hydrogen source; and

5 wherein discrimination between different contaminants in the hydrogen of the second hydrogen source is determined by at least one of:

- a) cleaning the second fuel cell with purified hydrogen following a contamination event and then observing change of electrical output over time; and
- 10 b) providing a third fuel cell; and either
 - (i) providing purified hydrogen from the first hydrogen purification device to the third fuel cell, the third fuel cell being configured to have a different sensitivity to specific contaminants from that of the first fuel cell; or
 - 15 (ii) providing a second hydrogen purification device receiving hydrogen from the second hydrogen source and configured to provide differently purified hydrogen from that of the first purification device to the third fuel cell; and
- 20 obtaining an electrical output of the third fuel cell in the same way as for the first and second fuel cells, and comparing electrical outputs of the first and third fuel cells to obtain an indication of contaminant type.

The first hydrogen purification device employed in the method of the invention may include a catalytic purifier. The first hydrogen purification device may comprise a palladium membrane. The comparator may be configured to determine a rate of change of voltage and / or current for each of the first and second fuel cells over a period of time. The second hydrogen source may be a steam reformer. The first fuel cell may comprise a plurality of series-connected fuel cells in a stack and / or the second fuel cell may comprise a plurality of series-connected fuel cells in a stack. The first fuel cell and the second fuel cell may form part of a single fuel cell stack. The first fuel cell and the second fuel cell may each be of the proton exchange membrane type. The first fuel cell and the second fuel cell may be integrated into a larger primary fuel cell stack.

28 03 19

Also described herein, but not in accordance with the claimed invention, is a hydrogen purity monitor comprising:

a first fuel cell configured to generate electrical current from the electrochemical reaction of hydrogen and oxidant, having a first fuel inlet configured to receive hydrogen from a first hydrogen source;

a second fuel cell configured to generate electrical current from the electrochemical reaction of hydrogen and oxidant, having a second fuel inlet configured to receive hydrogen from a second hydrogen source;

10 a control system configured to apply an electrical load to each fuel cell and determine an electrical output of each fuel cell, the control system including a comparator for comparing the electrical outputs of the first and second fuel cells; and

a purity monitor output configured to give an indication of hydrogen purity based on an output of the comparator.

15 The hydrogen purity monitor may include a first hydrogen source comprising a hydrogen source of known purity. The known purity hydrogen source may be a hydrogen tank containing a reference gas. The hydrogen purity monitor may include a hydrogen purification device coupled between the first fuel inlet and the second fuel inlet to receive hydrogen from the second hydrogen source, and to provide hydrogen from the second hydrogen source to the first fuel inlet via the purification device as the first hydrogen source.

20 The present invention will now be described by way of example and with reference to the accompanying drawings in which:

25 Figure 1 shows a schematic diagram of a fuel cell based hydrogen quality monitor; and

Figure 2 shows a schematic diagram of an alternative fuel cell based hydrogen quality monitor, not in accordance with the claimed invention.

30 A convenient hydrogen purity monitoring system described here uses fuel cell technology in order to monitor hydrogen quality. The purity monitoring system provides contamination detection and can be used at hydrogen fuelling stations in order to assess fuel purity before it is delivered to customers. The purity monitoring system can also be 35 used to monitor the hydrogen supply being fed to an operational fuel cell being used as

28 03 19

an electrical power supply for a building or vehicle, for example (referred to herein as a primary fuel cell). The purity monitoring system can be used as a periodic testing system or as an "in-line", continuously-operating fuel monitor.

5 The purity monitoring system uses a configuration of at least two fuel cells in order to monitor hydrogen purity. An advantage of using fuel cells to perform the hydrogen purity monitoring is that it is relatively inexpensive compared to existing elemental analysis apparatus and methods. Another advantage of a fuel cell based purity monitoring system is that, by their very nature, the fuel cells performing the purity monitoring can readily be
10 configured to be sensitive to exactly the same contaminants that are harmful to operation of a primary fuel cell stack with which the purity monitor can be associated.

Figure 1 shows a schematic diagram illustrating the principles of operation of a first configuration of hydrogen purity monitor 10. The purity monitor 10 includes a first fuel cell 11 and a second fuel cell 11. The first fuel cell 11 is a reference fuel cell and may further comprise a number of individual fuel cells disposed in series-connected configuration as a reference fuel cell stack 1. The second fuel cell 12 is a test fuel cell and may further comprise a number of individual fuel cells disposed in series connected configuration as a test fuel cell stack 2. The reference cell 11 has a fuel inlet 13 and the test cell 12 has a fuel inlet 14. In this arrangement, the fuel inlets 13, 14 are both supplied from a common hydrogen source 5. Hydrogen source 5 may be any form of hydrogen source including, but not limited to, any form of storage tank or vessel, a continuous piped supply, or a hydrogen generator such as a steam reforming system. The fuel inlet 13 is connected to the hydrogen source 5 by way of a purifier 16. The purifier 16 may be any form of filter capable of removing contaminants that would degrade the electrical performance of the reference fuel cell 11 and the test fuel cell 12. For example, any form of catalyst-activated purifier could be used. A preferred purifier is a palladium membrane. The purifier is preferably situated between the inlet 13 and the inlet 14 and the hydrogen source 5. Any suitable purifier or in-line gas purification method
25 may be used, such as those based on an adsorption method using porous media or pressure swing adsorption. A range of possible hydrogen purifiers are commercially available, such as the MicroTorr ® range from SAES Pure Gas Inc.

28 03 19

5

The hydrogen source 5 may also include an output 6 which is common to that supplied to fuel inlet 14, and which is coupled to a primary fuel cell stack power source for electrical power generation (not shown).

5 The reference fuel cell 11 has an electrical output 17 and the test fuel cell 12 has an electrical output 18. Both electrical outputs 17, 18 are connected to a controller 20. Controller 20 is configured to apply an electrical load (not shown) to each of the fuel cells 11, 12 and to monitor the electrical outputs 17, 18 of the fuel cells 11, 12. The controller 20 also includes a comparator (not shown) which compares the electrical outputs 17, 18 of the fuel cells 11, 12. The controller 20 also provides a purity monitor output 22 configured to give an indication of hydrogen purity of the hydrogen source 5 based on an output of the comparator.

15 In use, the hydrogen source 5 supplies hydrogen fuel to the reference fuel cell 11 via the purifier 16, but supplies hydrogen fuel directly to the test fuel cell 12 without purification. By comparing performance metrics from the reference fuel cell 11 and from the test fuel cell 12, it is possible to test for the presence of contaminants in the hydrogen fed to the test fuel cell that are specifically harmful to fuel cell operation and which degrade electrical performance of the test fuel cell.

20 The controller 20 may be configured to carry out performance metrics continuously, periodically or intermittently. The performance metrics may include measuring fuel cell voltage at a constant output current and I or output current at a constant voltage for each of the reference cell and the test cell. As shown in the inset output graphs 24, 25, the 25 rate of any voltage loss 26 in the test cell 12 compared to the reference cell 11 is related to the quantity and type of contamination in the hydrogen source fuel. The comparison with the reference cell 11 provides normalization of the measurements for environmental changes, such as temperature, humidity, air contamination and other factors that affect fuel cell performance.

30 Any suitable algorithm may be used for monitoring and comparing the relative performance of the reference and test cells 11, 12. An exemplary algorithm may determine a rate of change of voltage output for each of the reference and test cells and determine a purity level based on the difference in the respective rates of change. An 35 exemplary algorithm may determine a purity level based on an absolute difference in the

voltage outputs of the reference and test cells. The controller may be configured to trigger an alarm condition when the difference established exceeds a predetermined maximum, either transiently or over a defined period of time. A rate of change of voltage output may give an indication of the severity of contamination of the hydrogen supply.

5

Discrimination between different contaminants could be made by providing additional reference fuel cells that are each supplied with hydrogen from the hydrogen source 5 by way of different purifiers or contaminant filters, each filter configured to remove specific contaminants.

10

Alternatively, or in addition, discrimination between different contaminants could be made by providing further reference and test fuel cells with cells that have different catalysts, membranes or other features that are sensitive to different specific contaminants.

15

In an alternative arrangement, (not in accordance with the claimed invention) shown in figure 2, the hydrogen purity monitor 10a is provided with a separate high purity hydrogen source 27 instead of a purified supply from the main hydrogen source 5. In this arrangement, the high purity hydrogen source 27 can be a small storage vessel or tank

20

of known high purity hydrogen reference gas, e.g. hydrogen of at least a known level of purity in a high integrity vessel. In other respects, the purity monitor 10a operates in the same way as the purity monitor 10 of figure 1.

25

The fuel cells of the hydrogen purity monitor 10, 10a are preferably of the proton exchange membrane type although other fuel cell types capable of generating electrical current from the electrochemical reaction of hydrogen and oxygen can be used.

30

The reference and test fuel cells 11, 12 may form part of one or more fuel cell stacks. In one arrangement, one or more series-connected reference cells may be coupled to one or more series-connected test cells in a single fuel cell stack. Appropriate voltage monitoring terminals can be provided in known manner in the stack from the relevant cells or groups of cells to provide the requisite outputs 17, 18. The stack would be provided with the necessary separate reference fuel supply for the reference cell or cells and test fuel supply for the test cell or cells. Integrating reference and test cells into the same stack could provide an advantage in that the ambient conditions (e.g. temperature,

28 03 19

28 03 19

pressure, humidity etc) for operation of the reference and test fuel cells are more closely matched, thereby reducing any electrical output variation between the cells arising from a difference in ambient conditions.

- 5 The purifier 16 could also be integrated into the same fuel cell stack as the reference cells, e.g. by having a catalyst surface in a plate adjacent to the reference cell or cells and providing appropriate fluid flow ports for delivery of hydrogen so that the purifier and reference cell or cells are fluidically in series.
- 10 In another arrangement, the hydrogen purity monitor 10 or 10a can be integrated into a primary fuel cell stack that provides power to an external load, e.g. an automotive power unit. Appropriate voltage monitoring terminals can be provided in known manner in the primary stack from cells that are designated as the reference and test cells to provide the requisite outputs 17, 18. The primary stack would be provided with the necessary 15 separate reference fuel supply for the reference cell or cells. The rest of the stack that serves as a primary stack power supply and the test cells would be provided with fuel from the source 5.
- 20 In another arrangement, the hydrogen purity monitoring system could be modular such that reference and 1 or test fuel cells and palladium membranes could be replaced on a regular basis either after a certain time period or after a contamination event.

The purity monitor may be configured to run for a set time period after a fuel delivery to the main fuel storage tank 5. Alternatively, a sample volume of a fuel delivery could be 25 taken prior to filling the tank 5 to avoid dilution of contaminants in a fuel delivery. If the difference in voltage drop between the test and reference cells were to be above a preset value, the system may be configured to trigger a shut down in the delivery station and / or a primary fuel cell operating from a tank, or to trigger an alarm condition for a more detailed analysis of the fuel source.

30 After a contamination event, a test cell could be cleaned with purified hydrogen which could give some indication of the type of contamination. For example:
(i) an immediate improvement in test cell electrical output could indicate that the contamination event corresponded to concentration contamination (dilution) with a

contaminant having no direct effect on the fuel cell catalyst but causing a reduction in hydrogen concentration;

(ii) an improvement over time in test cell electrical output could indicate that the contamination event corresponded to a reversible catalyst contamination e.g. with CO;

5 (iii) no or little improvement over time in test cell electrical output could indicate that the contamination event corresponded to an irreversible catalyst contamination e.g. with sulphur compounds.

201218

CLAIMS

1. A method of monitoring hydrogen purity, the method comprising:
 - supplying hydrogen fuel from a first, purified, hydrogen source to a first fuel inlet
 - 5 of a first fuel cell configured to generate electrical current from the electrochemical reaction of hydrogen and oxidant;
 - supplying hydrogen fuel from a second hydrogen source to a second fuel inlet of a second fuel cell configured to generate electrical current from the electrochemical reaction of hydrogen and oxidant;
 - 10 applying an electrical load to each fuel cell and determining an electrical output of each fuel cell;
 - comparing the electrical outputs of the first and second fuel cells with a comparator; and
 - providing an indication of hydrogen purity of the second hydrogen source based
 - 15 on an output of the comparator;
 - wherein a first hydrogen purification device is provided, coupled between the first fuel inlet and the second fuel inlet, to receive hydrogen from the second hydrogen source, and to provide purified hydrogen from the second hydrogen source to the first fuel inlet via the purification device, as the first, purified, hydrogen source; and
 - 20 wherein discrimination between different contaminants in the hydrogen of the second hydrogen source is determined by at least one of:
 - a) cleaning the second fuel cell with purified hydrogen following a contamination event and then observing change of electrical output over time; and
 - 25 b) providing a third fuel cell; and either
 - (i) providing purified hydrogen from the first hydrogen purification device to the third fuel cell, the third fuel cell being configured to have a different sensitivity to specific contaminants from that of the first fuel cell; or
 - 30 (ii) providing a second hydrogen purification device receiving hydrogen from the second hydrogen source and configured to provide differently purified hydrogen from that of the first purification device to the third fuel cell; and
 - obtaining an electrical output of the third fuel cell in the same way as for the first
 - 35 and second fuel cells, and comparing electrical outputs of the first and third fuel cells to obtain an indication of contaminant type.

2. The method of monitoring according to claim 1 in which the first hydrogen purification device includes a catalytic purifier.

5 3. The method of monitoring according to claim 1 in which the first hydrogen purification device comprises a palladium membrane.

4. The method of monitoring according to claim 1 in which the comparator is configured to determine a rate of change of voltage and/or current for each of the first 10 and second fuel cells over a period of time.

5. The method of monitoring according to claim 1 in which the second hydrogen source is a steam reformer.

15 6. The method of monitoring according to claim 1 in which the first fuel cell comprises a plurality of series-connected fuel cells in a stack and/or in which the second fuel cell comprises a plurality of series-connected fuel cells in a stack.

20 7. The method of monitoring according to claim 1 in which the first fuel cell and the second fuel cell form part of a single fuel cell stack.

8. The method of monitoring according to claim 7 in which the single fuel cell stack is integrated into a larger, primary fuel cell stack, that provides power to an external load.

25 9. The method of monitoring according to claim 1 in which the first fuel cell and the second fuel cell are each of the proton exchange membrane type.

201218