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(54) **METHOD OF CONTROLLING AND MAXIMIZING THE ELECTRIC EFFICIENCY AND THE POWER OUTPUT OF A FUEL CELL**

VERFAHREN ZUR STEUERUNG UND ZUR MAXIMIERUNG DER ELEKTRISCHEN LEISTUNG UND DER AUSGANGSLEISTUNG EINER BRENNSTOFFZELLE

PROCÉDÉ DE RÉGLAGE ET DE MAXIMISATION DU RENDEMENT ÉLECTRIQUE ET DU RENDEMENT EN PUISSANCE D'UNE PILE À COMBUSTIBLE

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US-B1- 7 569 293

- **SCHIAVETTI PIERLUIGI ET AL: "Efficiency measurement and uncertainty discussion of an electric engine powered by a â self-breathingâ and â self-humidifiedâ proton exchange membrane fuel cell", REVIEW OF SCIENTIFIC INSTRUMENTS, AIP, MELVILLE, NY, US, vol. 78, no. 8, 20 August 2007 (2007-08-20), pages 85107-85107, XP012104176, ISSN: 0034-6748, DOI: 10.1063/1.2769576**

EP 2 710 658 B1

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Description**Field of the Invention**

5 **[0001]** The present invention relates to a novel method of controlling and maximizing the electric efficiency and the power output of a fuel cell.

Background of the invention

10 **[0002]** As it is known in the art, a fuel cell is an electrochemical device or apparatus suitable for producing an electromotive force by taking advantage of electron exchange occurring in Red-Ox reactions. The electric efficiency of an electrochemical device is measured by the ratio of chemical energy conversion - which is here defined as Gibbs free energy associated with an unitary molar amount of reagents involved in a Red-Ox reaction which takes place in the electrochemical device in question into electric power.

15 **[0003]** So far, it has been the general pursuit the reduction to a minimum of the production costs of a fuel cell, that are directly proportional to the involved electrodes surface. Such a need has directed the research efforts towards developing fuel cells with the highest possible ratio of electric power density supplied per unit of active surface area of the electrodes.

20 **[0004]** Moreover, the fuel cell development strategies of today, are based on the assumption, unrelated to the specific FC technology involved (PEM, ACFC, MCFC, SOFC, etc...), that the internal dissipative resistances of a fuel cell are set by the technological characteristics of the apparatus itself, such as type of materials, catalysts, cell architecture, and the electrolytes, while considering mostly marginal the dependence between the internal dissipative resistances and some macroscopic operating parameters such as pressure, flow, and average current density produced per unit of electrodes active surface, and some local operating parameters such as local current density and dilution gradient of the reagent mixtures along the overall feed channels. The same assumptions are considered, therefore, also valid with regard to the electric efficiency of fuel cells.

25 **[0005]** Up to date, in accordance with the theoretical model mentioned above, there are two equivalents methods used to control the output power and the electrical efficiency of a fuel cell. These methods comprise changing either the active surface area of the electrodes involved, or - as it is also shown in US-7,569,293 B1 and the article "Efficiency measurement and uncertainty discussion of an electric engine powered by a self-breathing and self-humidified proton exchange membrane fuel cell" of August 20, 2007 - Review of Scientific Instruments, AIP, Melville, NY, U.S. vol.78 and vol.8 - only the flow rate of the used reagents. The choice of either method depends entirely on how one chooses to define the nominal power (commercial power) of the used fuel cell.

30 **[0006]** The first method is applicable in the case where the nominal power of the used fuel cell is coincident with the power produced by it under the operating conditions of maximum efficiency (temperature, flow and pressure of the reactants, electric load, etc.). In such a case, is possible to define a power control method based on the simple variation of the overall electrode active surface area involved in the reaction while maintaining unchanged the remaining operating conditions.

35 **[0007]** The second method, however, as disclosed in US-7,569,293 B1, makes it possible to find out the relative maximum efficiency of the fuel cell when its nominal power is defined as the maximum output power (to draw a parallel with the automotive field where the power of an engine is expressed in HP at maximum torque).

40 **[0008]** As also mentioned in the two prior documents mentioned above, the consolidated operation model makes it possible to identify a "maximum relative efficiency" for each specific technology that is fixed by the above mentioned technological features of the cell.

45 **[0009]** This assumption together with the need of reducing the production costs of fuel cells has led to the pursuit of materials, catalysts, cell architectures, electrolytes and processes geared to obtaining the lowest possible internal resistance and the highest possible current density per unit of active surface area.

[0010] As a result of this, all fuel cells presently on the market reach a maximum relative efficiency between 48 and 55%.

50 **[0011]** With reference to a fuel cell, "main Red-Ox reactions" are defined as the Red-Ox reactions involved in the production of an electric current flowing in an external circuit connected in parallel to the fuel cell. Any chemicals not involved in the main Red-Ox reactions are defined as "dilutents", whereas any chemicals actually involved in the main Red-Ox reactions are defined as "reagents". Of course, dilutents, here includes all the range of possible chemicals.

Summary of the invention

55 **[0012]** The main object of the present invention is to provide a method of controlling the electric efficiency of a fuel cell in an easy and rapid way.

[0013] Another object of the present invention is to provide a method of controlling the electric efficiency of a fuel cell

before the operating of the fuel cell itself.

[0014] Another object of the present invention is to provide a method of controlling the electric power output of a fuel cell which is suitable for dynamically optimizing according to requirements the balance between output power and current efficiency, thereby increasing the energy conversion efficiency close to 100% with respect to the Gibbs free energy associated with the reactions taking place in the fuel cell.

[0015] Another object of the present invention is to provide a method of controlling the electric power output of a fuel cell independently from the technological characteristics of the fuel cell and the chemical-kinetic phenomena occurring in the fuel cell.

[0016] Another object of the present invention is to provide a method of controlling the electric power output of a fuel cell independently from the amount of the electrodes active surface area involved in the reactions.

[0017] Another object of the present invention is to provide a method of measuring the electric efficiency of a fuel cell.

[0018] Another object of the present invention is to provide a method of controlling and gauging the electric power output of a fuel cell suitable for controlling any type of fuel cell.

[0019] Another object of the present invention is to provide a method of controlling the ratio between thermal power and electric power produced by a fuel cell.

[0020] Another object of the present invention is to provide a fuel cell in which the electric efficiency can be rapidly controlled.

[0021] Another object of the present invention is to provide a fuel cell which is suitable for obtaining a high electric efficiency.

[0022] Another object of the present invention is to provide a fuel cell for maximizing the electric efficiency and the power output of a fuel cell.

[0023] According to a first aspect of the present invention a method of controlling a fuel cell is provided, which comprises a step of controlling the fuel cell electric efficiency :

- by checking and adjusting the dilution gradient of the reagents along the overall electrodes active surface, i. e. the variation of reagent dilutions among different parts of the electrodes active surface, and
- by checking and/or adjusting the local current density produced in the fuel cell per unit of electrodes active surface.

[0024] Advantageously the method of controlling a fuel cell according to the present invention comprises the following steps:

- determining, at a predetermined operation pressure, an electric efficiency curve per unit of active surface area as a function of the local current density produced in the fuel cell per unit of active surface area;
- establishing a desired electrical efficiency value, thereby determining a suitable current density in accordance to the electric efficiency curve; and
- producing in the fuel cell a local current density value equal to, or lower than, said suitable local current density.

[0025] More advantageously, the method comprises the following steps:

- determining, at a predetermined operation pressure, an electric power output curve per unit of active surface area as a function of the local current density produced in the fuel cell per unit of active surface area,
- determining the total amount of active surface;
- establishing a desired electric power output value, thereby determining a electrical efficiency and a local current density in accordance with the electric efficiency curve and to the electric power output curve.

[0026] Still more advantageously, provided that the dilution gradient along the overall feeding channel of at least one reagent used to fuel the reaction occurring in the fuel cell is checked or adjusted, the local current density is established on the base of the local dilution value(s) of the at least one reagent.

[0027] Preferably, the local current density is increased or decreased either by adding or subtracting reagents or diluents either already available in, or before or while being fed to, the cathodic and/or anodic area of said fuel cell, thereby decreasing or increasing the electric efficiency.

[0028] According to another aspect of the present invention, there is provided a method of evaluating the electric efficiency of a fuel cell comprising the following steps:

- arranging a circuit including a fuel cell and a resistor electrically connected with the fuel cell,
- supplying the circuit with reagents having a known dilution, and with determined dilution gradient along the overall electrodes active surface;
- evaluating a reference electric efficiency for the fuel cell supplied with the reagents having known dilution;

- evaluating the reference voltage drop across the resistor;
- changing the dilution of at least one reagent;
- changing the resistance of the resistor until a voltage drop across the resistor equal to the reference voltage drop is obtained; and
- evaluating the electric efficiency of the fuel cell on the base of the reference electric efficiency of the fuel cell, of the dilution of the reagents to be fed to the fuel cell before and after changing the dilution of the at least one reagent, and on the base of the ratio of resistor resistances before and after changing the resistance of the resistor.

[0029] According to further aspect of the present invention, there is provided a method of evaluating the electric efficiency of a fuel cell comprising the following steps:

- arranging a first fuel cell, whose electric efficiency is to be determined, in electrical connection with a first resistor of known resistance value, thereby obtaining a first circuit;
- arranging a second circuit including a second fuel cell identical to the first fuel cell, and a second resistor electrically connected with the second fuel cell,
- supplying the first and the second circuit with reagents having a known dilution(s), and with determined dilution gradient along the overall electrodes active surface;
- evaluating the electric efficiency of the second fuel cell on the base of its respective reagent dilution(s);
- evaluating the voltage drop across the resistor of the first and of the second circuit;
- keeping the second resistor (R^1) at a predetermined value and varying the first resistor (R^λ) until the same voltage drop across the resistor of the first and the second circuit is obtained, thereby determining a first resistor value; and
- evaluating the electric efficiency of the first fuel cell on the base of the electric efficiency of the second fuel cell, of the reagent dilution(s) to be fed to the fuel cell, and on the base of the ratio of first resistor value to second predetermined resistor value.

[0030] According to a further aspect of the present invention a fuel cell is provided, which is suitable for obtaining an electric power output comprising:

- a casing in which an intermediate space is delimited between a cathodic space and an anodic space;
- at least one electrolytic material located in the intermediate space;
- a positive electrode electrically connected to the end of said electrolytic material at the cathodic space;
- a negative electrode electrically connected to the end of the electrolytic material at the anodic space;
- an oxidant inlet mixture in fluid communication with the cathodic space; and
- a reducing inlet mixture in fluid communication with the anodic space,

the fuel cell comprising:

- means for checking and adjusting the dilution gradient of the reagents along the overall electrodes active surface, i. e. the variation of reagent dilutions among different parts of the electrodes active surface, and
- control means of the electric efficiency of the fuel cell including means suitable for checking and/or adjusting the current density produced in the fuel cell per unit of active surface area.

[0031] Advantageously, said means suitable for checking the current density comprises means suitable for checking the dilution value of the reagents of the fuel cell.

Brief Description of the Drawings

[0032] Further aspects and advantages of the present invention are detailed by the following description of specific embodiments of the present invention, referring to the accompanying drawings, in which:

- Figure 1 shows a diagram relating to an electric power output curve and an electric efficiency curve of a fuel cell as a function of the dilution parameter λ of the inlet oxidant mixture (ratio between the volume of pure oxygen and pure nitrogen used as diluent, at a temperature of 20°C and a pressure of 1 atm);
- Figure 2 shows a diagram relating to a polymer electrolyte fuel cell electric power output curve as a function of the dilution parameter λ of the inlet oxidant mixture (ratio between the volume of pure oxygen and pure nitrogen used as diluent, at a temperature of 20°C and a pressure of 1 atm) ;
- Figure 3 shows a diagram relating to an electric efficiency curve of a polymer electrolyte fuel cell as a function of the oxygen concentration in an inlet oxidant mixture with pure nitrogen at a temperature of 20°C ad a pressure of

1 atm; and

- Figures 4 to 7 show schematically a method and a respective fuel cell according to the present invention.

[0033] In the drawings, equivalent or similar parts or components were marked with the same reference numerals.

Detailed Description of the Preferred Embodiments

[0034] Experimental investigations carried out and reported in the present application show that another method of controlling the output power and the electrical efficiency of a fuel cell has been devised, which differs from the two previously described methods both in so far the methodology and the results are concerned. This method is based on the control of some local operation parameters such as local current density and local dilution of the reagents along the overall electrodes active surface, and the dilution gradient of the same along the overall reagent feed channels, i. e. the variation of reagent dilutions among different parts of the electrodes active surface.

[0035] A method in accordance with present invention is based on the finding that a strict correlation exists between the local current density produced on the electrodes of a fuel cell per unit of electrodes active surface (determined by the local dilution parameter of the reagents along the overall electrodes active surface, provided that the dilution gradient of the reagent mixtures along the overall feeding channel is controlled and adjusted) and the electric efficiency of a fuel cell. Such a correlation is prompted by the intrinsic characteristics of the fuel cell, such as fuel cell architecture, electrolyte(s), catalyst(s) and the thermodynamics of the reaction taking place in the fuel cell, i. e. independently of the used technology. More particularly, the same correlation is apparently due to an interaction between the intrinsic electronic feature of a fuel cell and an intrinsic characteristic of the reaction occurring in it.

[0036] The applicant's research results have shown that:

- unlike the methods described in US-7,569,293 B1 of August 4, 2009, and in the article "Efficiency measurement and uncertainty discussion of an electric engine powered by a self-breathing and self-humidified proton exchange membrane fuel cell" of August 20, 2007 Review of Scientific Instruments, AIP, Melville, NY, U.S. vol.78 and vol.8, according to which some general operating parameters, such as the average surface current density produced on the electrodes of a fuel cell by varying over time the amount of active electrode surface, or, equivalently, the flow rate of feeding flows of the reagents are dynamically controlled,
- by dynamically controlling local operating parameters such as the local current density generated along the overall active electrode surface of a fuel cell, provided that the inlet and outlet flow rate ratio of the reagent mixtures (lamination ratio), and then the dilution gradient of the reagent mixtures along the overall active electrode surface are controlled and adjusted, it is possible to control the electric efficiency of a fuel cell.

[0037] Preferably, local operating parameters are designed to include the dilution parameter of the reagent mixtures provided that the local dilution gradient of the reagent mixtures is controlled or adjusted along the overall active electrode surface of the fuel cell, and preferably kept as close as possible to zero.

[0038] From an electronic point of view, a fuel cell can be regarded as a source of current with a capacitor in parallel whose capacitance is determined by the technological characteristics of the specific fuel cell involved. Owing to the kinetics of the electrochemical reactions involved, an electrochemical noise is generated which is proportional to the local rate of electrochemical reactions, this resulting in an electrical noise being obtained at the plates of the capacitor that has a frequency and an amplitude proportional to the local density of the current produced per unit of active electrodes surface. The higher the local current density produced the higher the cathode overvoltage and the electrical power dissipated by Joule effect on the capacitor, which causes a temperature increase in the fuel cell and thus a reduction in the electricity efficiency.

[0039] The process described in U.S. Patent 7,569,293 B1 August 4, 2009, makes it possible to control through variation of the inlet reagents flow rate the average current density produced by the fuel cell (i.e., as defined by the ratio $\langle I \rangle = I / A$, where I is the electric current flowing in the resistance short-circuited to the cell, and A is the total electrode surface). As known in the art, the consumption of reagent along the active electrode surface is directly proportional to the local intensity of the electrochemical reactions which is proportional, in turn, to the local availability in the proximity of the electrodes active surface. This causes an exponential decay of the reagent concentration in the mixture along the electrodes surface, thus inducing a variation in the active electrode surface involved in the reaction and in the dilution gradient of the reagent along the overall electrode surface. In this sense, a variation of the reagent inlet flow rate, as described in US-7,569,293 B1, corresponds to a variation of the electrode active surface.

[0040] With respect to this procedure, the method described in the present application makes it possible to control and maintain constant along the overall electrode active surface the locally produced current density (defined as $I^* = dl/dA$

where dI is the electric current produced on the element of the electrode surface dA , such that $\langle I \rangle = \int_{\omega} I^* dA$,

[0041] This method therefore allows to control and maintain constant on the overall electrode active surface the local intensity of the electrochemical reaction (and thus the electric energy dispersion for capacitive effect of the fuel cell) and not (as taught in the above-mentioned prior art documents) its average value on the electrode active surface, with much less important results in terms of control of the dispersion, and thus of electric efficiency.

[0042] To this regards, the applicant of the present invention has discovered that each fuel cell may be properly described by two characteristic curves or diagrams, i. e. the electric power output curve (hereinafter $EPC(P_{OX}; P_{RED})$) defined as the local electric power density produced per unit of active electrode surface of a fuel cell as a function of the local intensity of electrochemical reaction (that is as a function of the local dilution parameter of the reagents in conditions of zero gradient of the same along the overall electrode surface), and the electric efficiency curve (hereinafter $EEC(P_{OX}; P_{RED})$) also parameterized as a function of the local current density produced per unit of electrode active surface. Such curves describe the variation of electric power output and cell electric efficiency, respectively, as a function of the local current density of the fuel cell, in the conditions and according to the method of the present invention. Such curves are shown in the diagram of Fig. 1, and generally depend upon the pressure of reagents in the feeding chamber and upon the technological characteristics of the fuel cell as both depend upon the value of the capacitance of the capacitor referred to above.

[0043] These curves radically differ from the curves described in "Efficiency measurement and uncertainty discussion of an electric engine powered by a self-breathing and self-humidified proton exchange membrane fuel cell" of August 20, 2007 Review of Scientific Instruments, AIP, Melville, NY, U.S. and vol.78 vol.8, and US-7,569,293 B1 of August 4, 2009, and indicated as the Polarization Curves (or curves VI) or Power Curves (or curve WI).

[0044] Such known (polarization and power) curves are obtained by short-circuiting a variable resistance to the fuel cell and measuring the potential drop and the electric current across the resistor upon varying the same.

[0045] Polarization Curves show the behavior of the potential available at the heads of a fuel cell by varying the total current produced by the same, or, equivalently, by varying the average surface current density per unit of electrode active surface as defined above. Similarly, the Power curves show the variation of the electric power generated by the fuel cell upon varying the average surface current density per unit of electrode active surface.

[0046] The diagram of Fig. 1 shows that in the tested fuel cell a considerably higher increase in electrical efficiency was obtained for oxygen dilutions (mean dilution of oxygen in air nitrogen) higher than a specific threshold value, i. e. 21%. It should be noted that according to the present invention, electrical efficiency is increased, provided that the dilution gradient of the oxygen along the overall electrodes active surface is checked and adjusted, if the oxygen concentrations in the oxidant inlet mixtures are reduced, but a remarkable increase of the electrical efficiency is obtainable for a concentration of oxygen lower in the oxidant inlet mixture than 21%.

[0047] These experimental results are of substantial interest in that they open up new opportunities for developing novel energy conversion systems, both primary, e. g. thermal power plants based on combustion of fossil fuels, and secondary ones, e. g. motors, energy storage systems, auxiliary equipment, and the like, and thus, no longer based on thermo-kinetic technologies that are quite limited in efficiency but rather on high-efficiency electro-chemical technologies. Thus, owing to the present invention both efficiency and costs can be improved.

[0048] More particularly, primary conversion systems can include, for example fuel cells supplied by methane, ethane, propane, gasified coal, any reformed hydrocarbon, whereas second conversion systems can include, for example fuel cells supplied by any reformed hydrocarbon or reforming-derived hydrogen. Referring to fuel cells supplied by reforming-derived hydrogen or gasified coal the supplied reducing mixture can include CO and CO₂, which act as performing diluents of the reducing reagent, e. g. hydrogen.

[0049] The same results also show that it is possible to design electric power control systems, even integrated in the fuel cell, suitable for dynamically optimizing, according to requirements, the balance between output power and efficiency, and to increase the efficiency of energy conversion of the fuel cell to values very close to 100% with respect to the Gibbs free energy associated to the amount of moles of reagents involved in the reaction occurring in the apparatus.

[0050] Such an increase in performance of a fuel cell is obtained by modifying the local electric current density produced on the electrode active surfaces by balancing the lamination ratio of the feeding fluxes, i. e. by checking and adjusting the dilution gradient of the reagents along the overall electrodes active surface, and by acting on the dilution of the oxidant and/or the reducing agent(s) used to fuel the reaction occurring in the apparatus.

[0051] Other methods may be used for setting the local current density produced on the electrode active surface, e. g. by changing the permeability of the gas diffusion layers, the conductivity of the electrodes or the electrolyte concentration. These techniques involve direct manipulation of the used fuel cell and do not allow a dynamic control of the obtained current density as it is obtainable by acting on the dilution of the oxidant and/or the reducing agent/s.

[0052] Moreover, the local current density can be controlled or adjusted owing to the combination of the above-mentioned techniques.

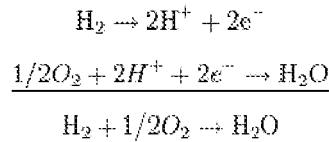
[0053] Thus, the control method according to the present invention is applicable independently from the technology of the adopted fuel cell. Moreover, the intrinsic chemical-kinetic phenomenon on which it is based makes it possible to apply the control method according to the invention to fuel cells of any type regardless of the geometry of the cell, the electrochemical process involved, or the type of fuel being used.

[0054] Once the characteristic curves $EPC(P_{OX};P_{RED})$ and $EEC(P_{OX};P_{RED})$ of an fuel cell are experimentally determined, one can increase (up to a maximum concentration value set by the reagent supply source), or decrease (with no limitation) the dilution or concentration of the reagent(s) taking part in the reactions occurring in the fuel cell. Of course, before such increasing-decreasing step the dilution gradient of the reagents along the overall electrodes active surface should be checked and adjusted, and preferably be kept as close as possible to zero.

[0055] An increase or decrease in reagent(s) concentration can be caused either by adding or subtracting reagent(s) or diluent(s) either already available in, or before or while being fed to the cathodic and/or anodic area of the fuel cell. The reagent(s) or diluent(s) can be added or subtracted either in a chemical or in a physical way.

Example: Application of the electric efficiency and power output control method to a PEM fuel cell:

[0056] A polymeric electrolyte fuel cell (PEM) was used to produce an electromotive force exploiting the following Red-Ox reaction between oxygen and hydrogen:



Main Red-Ox reaction

[0057] The fuel cell was fed with hydrogen obtained by electrolysis and with variable stoichiometric mixtures of oxygen-nitrogen (having a different value of the mean stoichiometric ratio λ).

[0058] Function λ , when parameterized in one dimension on the z coordinate axis along the channel or chamber of the cathodic space, is generally defined as:

$$\lambda := \frac{V_{O_2} [l]}{V_{mix} [l]} = \lambda(z, t) \rightarrow (0,1) \quad (1)$$

where V_{O_2} is the volume of pure oxygen at ambient pressure and at a temperature of 4°C, V_{mix} is the volume of oxidant inlet mixture (in the same standard conditions), and t is the time. In the case of an ideal transport of oxygen through the porous layers (which are usually located on the cathodic electrode and are arranged to increase the oxygen distribution thereon), and assuming the condition of minimum feeding flux of oxidant mixture:

$$\phi_{feed}^{O_2} \approx \phi_{consumption}^{O_2} \quad (2)$$

where $\phi_{feed}^{O_2}$ is the inlet flux and $\phi_{consumption}^{O_2}$ is the consumption flux, the local oxygen concentration decreases exponentially with z according to the following algorithm:

$$\lambda(z) = \lambda_{bulk} e^{\frac{-z}{\alpha}} \quad (3)$$

where λ_{bulk} is the stoichiometric ratio between oxygen and oxidant mixture (i. e. oxygen plus diluent) in the oxidant inlet mixture (see below).

[0059] It was chosen to work with a cell having a square active surface (2 cm side) with a central opening through which oxygen was supplied into the cathodic channel or chamber. In order to satisfy the invariance condition for the gradient of the oxidant mixture dilution parameter along the overall active electrode surface, a lamination ratio equal to 0,9867 \approx 1 was set, i.e. such that

$$\phi_{feed}^{O_2} \geq 75 \cdot \phi_{consumption}^{O_2} \quad (4)$$

[0060] In order to satisfy the time invariance condition

$$\frac{\partial \lambda}{\partial t} = 0 \quad (5)$$

the cell was fed at constant pressure (a slight atmospheric overpressure) and constant stoichiometric gas ratio λ_{bulk} .

[0061] Measures of the open circuit cell potential and the current flowing in the circuit were made by measuring the voltage drop across a known resistance electrically connected to the cell, in parallel therewith. The measures performed made it also possible to determine the cathode overvoltage as a function of current density ($\eta = E - E_{eq}$).

Table 1

%O ₂ λ	(mA)	H (1 Ω) (mV)
100,0%	400	-634
50,0%	396	-638
33,3%	392	-642
20,0%	381	-653
16,7%	372	-669
13,3%	330	-704
10,0%	351	-683
6,7%	328	-706
5,0%	265	-769

[0062] The current measurements show, at constant cathode active surface area Θ^c , a decrease in the mean surface current density, while λ lowers towards 0.

[0063] The electrical efficiency μ^e of the cell can be defined as:

$$\mu^e = \frac{W^e}{K_{O_2} V^c} \in [0,1] \quad (6)$$

where $K_{O_2} 285.83 \left[\frac{KJ}{mol} \right]$ is the molar density of the free Gibbs energy associated with the oxygen with respect to the reaction:



$V^c \left[\frac{mol}{s} \right]$ is the oxygen consumption in the unity of time t and per unit of electrode active surface and $W^e \left[\frac{KJ}{s} \right]$

is the electric power generated by the fuel cell. More particularly, one can observe that with $\mu^e=1$, all free Gibbs energy in the reaction (6) is transformed into electric energy.

[0064] Now one can observe that:

$$V^c = \frac{\lambda\phi}{\Theta^c} \quad (8)$$

5 where Θ^c is the cathode active surface area and ϕ the flux of the inlet oxidant or oxidizing mixture:

$$\mu^e = \frac{W^e}{K_{O_2}} \frac{\Theta^c}{\lambda\phi} \quad (9)$$

10 **[0065]** We observe first that in general it is quite reasonable to suppose, as long as it can be considered negligible edge effects, the following condition of invariance.

$$\frac{\partial\mu^e}{\partial\Theta^c} = 0 \quad (10)$$

15 **[0066]** Suppose now to consider also a condition of invariance of the electric efficiency from the dilution parameter:

$$\frac{\partial\mu^e}{\partial\lambda} = 0 \quad (11)$$

20 **[0067]** By definition it is known that:

$$\mu^e = \frac{W^e}{K_{O_2}} \frac{\Theta^c}{\lambda\phi} \quad (12)$$

25 **[0068]** Now if the conditions (10) and (11) must be respected, the quantity W^e should be given by the following formula:

$$W^e = A \cdot \frac{\lambda}{\Theta^c} \quad (13)$$

30 where A is a suitable constant of proportionality.

[0069] Thus, if the assumption (11) is correct, a linear dependence between the cell electric power output and the dilution parameter λ should occur.

35 **[0070]** However, we carried out a number of tests, the results thereof are shown in the Graph of Figure 2, which clearly demonstrated that such a linear dependence is not obtained, thereby demonstrating false the hypothesis (11).

[0071] Such observations and tests make it possible to deduce the existence of a non-trivial functional relationship linking those quantities, and in particular allow observing a noticeable improvement in power efficiency by reducing the surface density of the current, which means in particular that in the range of measures considered in place of the report condition (11) is instead the condition:

$$\frac{\partial\mu^e}{\partial\lambda} \leq 0 \quad (14)$$

40 **[0072]** As it will be understood, from the formula (12) both an explicit relation between the electric efficiency and the dilution parameter and an implicit relation can be evaluated, the implicit relation occurring since, as a matter of fact, W^e must depend upon the dilution parameter. Moreover, tests carried out by the applicant of the present application have shown that W^e must depend upon the dilution parameter in a non-linear way, and thus such tests have confirmed that the electric efficiency μ^e depends upon the dilution parameter, and, in turn, upon the current density.

45 **[0073]** More particularly, it is of interest to note that the electrical efficiency of the fuel cells depends substantially on external factors such as the dilution parameters λ , i. e. intrinsic parameters to the red-ox process rather than structural

parameters or parameters concerning the geometry of the cell.

[0074] In view of the relation (9), the above described protocol makes it possible also to determine the relative electric efficiency $\mu^e(\lambda)/\mu^e(\lambda^*)$, that represents the gain on a known value of the electrical efficiency $\mu^e(\lambda^*)$.

5 **[0075]** Suppose to compare two circuits each comprising a fuel cell, and a resistance R^λ (the resistor of the first fuel cell) and R^1 (the resistor of the second fuel cell) electrically connected, connected in parallel with the fuel cell, the first cell being fed with an oxidant mixture with $0 < \lambda < 1$, and the second cell with an oxidant mixture having $\lambda = 1$.

[0076] The fuel cell of one circuit is identical to the fuel cell of the other circuit, i. e. the fuel cells have the same technology and the same amount of electrodes active surface area involved in the reactions.

10 **[0077]** The oxygen consumption occurs according to the relation $V^c = \frac{\lambda\phi}{\Theta^c}$ and linearly depends upon λ . The oxygen consumption of the two respective circuits will occur according to the following relation:

15
$$V^c_\lambda = \lambda V^c_1 \tag{15}$$

[0078] If one compares the electric power output dissipated by the two circuits with the same consumption, one has to consider λ circuits with $FC(0 < \lambda < 1)$ and 1 with $FC(\lambda = 1)$.

20 **[0079]** The only one equation describing the electrical behavior of the circuit is that concerning the mean surface current density:

$$I = \bar{J}\Theta^c = k^* F V^c \Theta^c \tag{16}$$

25 **[0080]** The variable to be set is the resistance R^λ and/or R^1 .

[0081] A choice would be to select R^λ and R^1 so that in both circuits the voltage drop across the resistor is the same.

[0082] The current density is proportional to the dilution parameter, so that through the circuit fed with diluted oxygen a lower current with respect to the circuit fed with pure oxygen will flow. In order to obtain the same ΔV , the resistors in the two circuits must satisfy the condition $R^\lambda > R^1$.

30 **[0083]** It is possible to proceed by keeping R^1 at a fixed value and varying the resistor R^λ in order to determine x (see following formula (18)) in such a way that:

35
$$\frac{\Delta V^\lambda}{R^\lambda} = I^\lambda = k^* F \frac{\lambda\phi}{\Theta^c} \Theta^c = k^* F \lambda \phi = \lambda I^1 = \lambda \frac{\Delta V^1}{R^1} \tag{17}$$

or in such a way as both the following conditions are satisfied:

40
$$\begin{cases} I^\lambda = \lambda I^1 \\ R^\lambda = x R^1 \end{cases} \tag{18}$$

45 **[0084]** Bearing in mind that by definition

$$\mu^e(\lambda)/\mu^e(1) = \frac{V^c(1) W^e(1)}{V^c(\lambda) W^e(\lambda)} \tag{19}$$

50 **[0085]** it is possible to obtain

$$\mu^e(\lambda)/\mu^e(1) = \frac{I(1) W^e(\lambda)}{I(\lambda) W^e(1)} = \frac{I(1)}{\lambda I(1)} \frac{\lambda^2 I^2(1) x R^1}{I^2(1) R^1} = \lambda x \tag{20}$$

55 that is

$$\mu^e(\lambda)/\mu^e(1) = \lambda x \quad (21)$$

[0086] Accordingly, the applicant of the present invention, on the base of his findings that the electric efficiency of a fuel cell depends upon the local current density produced over all the electrode active surface, and so can be controlled by checking and adjusting, and preferably fixing the lamination ratio of the feeding fluxes and by setting the dilution of the reagents to be fed to the fuel cell, has also developed a method of evaluating and controlling the electric efficiency of the fuel cell on the base of the electric efficiency of a reference fuel cell having a known electric efficiency.

[0087] Clearly, a method of evaluating and controlling the electric efficiency of a fuel cell can also be obtained by fixing the dilution parameter of the fuel cell, and by evaluating the electric efficiency thereof, thereby obtaining a reference electric efficiency.

[0088] After that, the electric efficiency during the operation of the same fuel cell can be evaluated at any time on the base of the above-mentioned formula (21).

[0089] Thus, owing to a method according to the present invention it is possible to evaluate an electric efficiency without using sensors or similar components, but referring to a reference electric efficiency of the same fuel cell or of a reference fuel cell.

Table 2

%O ₂ λ	x	x λ
100,0%	1	1,00
50,0%	2,828	1,41
33,3%	4,85	1,62
20,0%		
16,7%	11,16	1,86
13,3%	15,1	2,01
10,0%	22,9	2,29
6,7%	35,1	2,35
5,0%	48,2	2,41

[0090] The diagram of Fig. 3 shows that in the tested fuel cell a considerably higher increase in electrical efficiency was obtained for oxygen concentrations (mean concentration of oxygen in nitrogen) lower than a specific threshold value, i. e. 16%.

[0091] Thus, the present invention relates to a method of controlling a fuel cell, which comprises a step of controlling the fuel cell electric efficiency:

- by checking and adjusting the dilution gradient of the reagents along the overall electrodes active surface, and
- by checking and/or adjusting the local current density produced in the fuel cell per unit of electrode active surface .

[0092] As it will be understood, owing to the present invention, it is possible also to maximize the absolute electric efficiency (here to be intended not with respect to the specific technology but with respect the total Gibbs free energy produced by the involved reaction) and the power output of a fuel cell.

[0093] According to the present invention, there are two main patterns of operation of a fuel cell depending on whether the cathodic and/or anodic active surface area involved in the reactions is constant or variable.

[0094] In the case of a fuel cell having cathodic and anodic active surface area involved in the reaction constant over time, the whole output power is predetermined by the total amount of active surface area according to the EPC(P_{OX};P_{RED}) curve. Accordingly, once the characteristic curves are known, and, preferably, once the forecasted plan of the load curve, i. e. the curve describing the time-varying power supply required to the fuel cell throughout the operative life thereof (hereinafter LC), is also known, one can determine the total amount of active surface area that has to be involved in the reaction and the optimal dilution of the reagent(s) in order to maximize the average electric efficiency provided by the fuel cell over time.

[0095] The fuel cell can be so designed as to operate with a reagent dilution(s) constant or variable over time by equipping the apparatus with an (integrated) control system arranged to control the dilution of the used oxidants and/or reducing agents.

[0096] The expression "raw mixtures" in the present invention is designed to indicate any mixture of reagents and diluents having an arbitrary (not controlled) stoichiometric ratio between the chemicals, i. e. between reagents and diluents. Such raw mixtures can be supplied by the atmosphere (air used as oxidant mixture) or from a storage.

[0097] The expression "inlet mixtures", instead, indicates all the mixtures of reagents and diluents that are used to feed the electro chemical reactions in a fuel cell without undergoing any treatment process to modify the stoichiometric ratio of the chemicals composed. To obtain inlet mixtures, in general, it is possible to mix together an arbitrary number of raw mixtures containing reagents and diluents in different proportions. It should be understood, that when the raw mixtures have a correct (i. e. suitable for being used in the fuel cell) stoichiometric ratio, the inlet mixtures correspond to the raw mixtures.

[0098] When operating in a constant reagents dilution configuration, where the required inlet reagents concentrations are, respectively, $C_0^{(ox)}$ for the oxidizing mixture and $C_0^{(red)}$ for the reducing mixture, the concentration $C_0^{(ox)}$ and $C_0^{(red)}$ is obtained by addition to, or subtraction from, diluent or reagent substances, depending upon whether the raw mixtures are more diluted or less diluted than the inlet reagents mixture concentrations $C_0^{(ox)}$ and $C_0^{(red)}$.

[0099] Either an external addition or subtraction can be carried out.

[0100] A scheme of external addition or adjunction is shown in Figure 4, where the concentrations of reagents and diluents in the raw mixtures are deemed to be known, whereas the inlet mixtures are prepared by mixing available raw mixtures in proportions suitable for feeding inlet mixtures in which the reagents and the diluents are in a desired stoichiometric ratio.

[0101] The inlet mixtures can be obtained directly by mixing the feeding raw mixtures immediately upstream of the fuel cell or by pre-mixing the raw mixtures in storage devices, from which the inlet mixtures are obtainable. The inlet mixtures thus obtained are then caused to flow on the active surfaces of the fuel cell and discharged when the concentration of the reagents is lower than a predetermined limit value. In this case, it is possible to obtain changes over the time dilution of the reagents intervening directly in the pre-mixing phase by varying (increasing or decreasing) the flow rate of the raw mixtures or by changing the concentration of any species (i. e. reagents and diluents) in each raw reagent mixture.

[0102] More particularly, in Figure 4 a fuel cell 1 is shown, which is suitable for obtaining an electric power output, and comprises a casing (not shown) in which an intermediate space 2 is delimited between a cathodic space 3, and an anodic space 4, at least one electrolytic material being located in the intermediate space 4, whereas a positive electrode 5 is electrically connected to the end of said electrolytic material at the cathodic space 3, and a negative electrode 6 is electrically connected to the end of the electrolytic material at the anodic space 4.

[0103] According to the present invention, the electrolytic material can be, for example, selected from the group including salt water, aqueous alkaline (KOH) solution, polymer membrane, molten phosphoric acid (H_3PO_4), molten carbonate, solid oxide, such as O^{2-} -conducting ceramic oxide or H^+ -conducting ceramic oxide.

[0104] Furthermore, the fuel cell includes:

- means for checking and adjusting the dilution gradient of the reagents along the overall electrodes active surface;
- control means of the electric efficiency of said fuel cell comprising suitable means suitable for checking and/or adjusting the current density produced in the fuel cell per unit of active surface area.

[0105] Advantageously, the means for checking and adjusting the dilution gradient of the reagents along the overall electrodes active surface comprises means suitable for maintaining constant and null the dilution gradient of the reagents along the overall electrodes active surface, i. e. the variation of reagent dilutions among different parts of the electrodes active surface.

[0106] Moreover, the fuel cell includes an oxidant inlet mixture 7 in fluid communication with the cathodic space 3, and a reducing inlet mixture 8 in fluid communication with the anodic space 4.

[0107] Particularly referring to the fuel cell shown on Figure 4, oxidant inlet mixture is obtained by mixing a number (two shown on Figure) of raw mixtures 7a, whereas reducing inlet mixture 8 is obtained by mixing a number (two shown on Figure) of raw mixtures 8a.

[0108] Discharge mixtures 3a and 4a from the cathodic space 3, and anodic space 4, respectively, are also shown.

[0109] Optionally, the fuel cell can include control means of the electric efficiency including means suitable for checking the dilution value of the reagents of the fuel cell.

[0110] Preferably, the fuel cell comprises:

- means for checking the dilution of the oxidant and/or reducing agent(s) used to fuel the reaction that will occur out in the fuel cell;
- means for comparing the checked dilution(s) with desired maximum dilution value(s); and
- means for controllably increasing or decreasing the dilution of said reagents, thereby decreasing or increasing the electric efficiency of said fuel cell, such means for controllably increasing or decreasing can include a source of

diluent(s) in fluid communication with the cathodic space, the anodic space, the oxidant inlet mixture and/or the reducing inlet mixture.

5 **[0111]** Advantageously, the fuel cell can comprise means for varying the electric efficiency including means for increasing or decreasing the current density, which can include means for adding or subtracting reagents or diluents either already available in, or before or while being fed to, the cathodic and/or anodic area of said fuel cell.

10 **[0112]** A scheme of internal adjunction is shown in Figure 5, where the concentration of available raw mixtures is known, and it is possible to provide a diluent ambient directly in the reaction chamber, i. e. the cathodic and anodic spaces of the fuel cell. Then a previously known volume of diluents mixture is injected under pressure into the electro-chemical reaction chamber. The fuel cell is fed with the mixture containing the reagent with a feed flow adjusted with respect to the reagents consumption rate in order to maintain a constant dilution of the reagents in the reaction chambers or channels of the fuel cell. The means for adjusting the dilution gradient of the reagents along the overall electrodes active surface can include a mixer 9, located in the cathodic and/or anodic space, and suitable for maintaining the oxidant and/or reducing, respectively, mixture in a desired turbulent motion condition. The mixing speed is fixed or adjustable in order to obtain a desired lamination ratio parameter (thus maintaining in an established range the dilution gradient of the oxidant mixture and/or reducing mixture along the overall electrode active surface). The mixing speed allows to fix the ratio between the velocity of flow skimming the electrode active surface and the consumption flow rate of oxygen through the same. The exhaust flows in this case are proportional to the portion of diluents contained in the feeding mixtures. Thus, it is possible to make changes in the time dilution of the reagents by varying the pressure of the diluent mixtures in the reaction chamber.

20 **[0113]** A scheme of external subtraction is instead shown in Figure 6, where the concentration of available raw mixtures is known, and the inlet mixtures are prepared by removing reagents and/or diluents from the raw mixtures. Subtraction of one of the two species, or both, is carried out externally or outside the reaction chamber and can be obtained by applying any suitable physical or chemical method (e. g. adsorption membrane (PSA), fractional distillation or freezing, selective absorption membrane, and the like). To this end, subtraction streams 7b and 8b are shown.

25 **[0114]** The inlet mixtures thus obtained are then caused to flow on the active surfaces of the fuel cell and discharged when the concentrations of the reagents are lower than a predetermined limit value. In this case, the possibility of changing the dilution over time of reagents in the inlet mixture depends upon the adopted physical or chemical subtraction method.

30 **[0115]** In Figure 7 a scheme of internal subtraction is shown, where the concentrations of available raw mixtures are known, and the inlet mixtures are prepared by removing reagents and/or diluents from the raw mixtures. The subtraction of one of the two species, or both, is carried out inside the reaction chamber, i. e. the cathodic 3 and anodic 4 spaces, see subtraction streams 3b and 4b, and can be obtained by applying any suitable physical or chemical method (see also below). Also in this case the chamber is provided with a mixer 9 for the reasons above mentioned. The exhaust flows, generally consisting of mixtures containing reagents and/or diluents species and/or other chemicals, are then discharged from the reaction chamber. In this case, the possibility to change dilution over time of reagents in the inlet mixture depends upon the adopted physical or chemical subtraction method.

35 **[0116]** Working in reagents dilution configuration variable over time, the concentrations $C^{\text{ox}}(t)$ and $C^{\text{red}}(t)$ of, respectively, the oxidizers and reducing agents in the inlet mixture at time t , can be obtained by applying one of the above described adjunction or subtraction schemes or methods, or a combination thereof, depending whether, at time t , the reagent raw mixtures are over- or under diluted with respect to the concentrations $C^{\text{ox}}(t)$ and $C^{\text{red}}(t)$.

40 **[0117]** According to a variant, the amount of cathodic and/or anodic active surface area involved in the reactions changes, i. e. it/they is/are time depending, and in such case the fuel cell is equipped with a gauge system that makes it possible to actively control the size of active surface (or electrodes) involved in the reaction from time to time.

45 **[0118]** In this case the obtainable power output of the cell depend upon the total amount of active surface areas involved in the reaction at time t .

[0119] A fuel cell generally consists of a number of unitary fuel cells (the unit being arbitrary) combined with one another and connected to each other in series and/or in parallel, thereby forming a so-called fuel cell stack.

50 **[0120]** Generally, whatever is the specific gauging system used to vary over time the amount of active surface areas involved in the reaction, it is possible to schematically represent such system as a system that increases or decreases the number of working (fed with fuel) unitary fuel cells electrically connected in parallel and/or in series in a stack.

[0121] Also in this case the apparatus can be designed so as to work with reagent dilution(s) constant or variable over time.

55 **[0122]** The methods that can be applied to control the concentrations of reagents and diluents in the inlet mixtures, in both configurations with time constant or variable concentrations are those described above either taken alone or in combination with one another.

[0123] In contrast to the configuration described above in which the size of cathodic and/or anodic active surfaces is set and kept constant in time, in this configuration it is possible to change from time to time the electric power output of

the cell without changing the chosen electric efficiency of the fuel cell, achieved by setting the concentration of reagents in the inlet mixture, in line with the respective characteristic curve $EEC(P_{OX};P_{RED})$ of the fuel cell in question.

[0124] According to the state of the art, it is possible to increase or decrease the electric power output of a fuel cell system only by acting on the total amount of active surface area involved in the reactions, whereas the electric efficiency of the fuel cell is set by the technological characteristics of the cell itself.

[0125] On the contrary, according to the present invention, owing to the exploitation of the characteristic curves of electrochemical devices it is possible to obtain the following:

- by working with an active surface area and a reagents dilution configuration constant over time, it is possible to determine the total amount of active surface area that have to be involved in the reaction and the optimal dilution of the reagents in order to obtain the desired electric power output, and, differently from the state of the art, it is also possible, in accordance with the characteristic curves of the fuel cell and to the forecast plan of the load curve, to obtain an average electric efficiency higher than 90%;
- by working in an active surface area and a variable reagents dilution configuration constant over time, in accordance with the characteristic curves of the cell, $EPC(P_{OX};P_{RED})$ and $EEC(P_{OX};P_{RED})$, it is possible to increase or decrease from time to time the electric power output and the electric efficiency, i. e. to increase or decrease the consumption of reagents per unit of watt hour produced;
- by working in variable active surface area and reagents dilution configuration constant over time, it is possible to determine, in accordance with the $EEC(P_{OX};P_{RED})$ characteristic curve of the fuel cell being used, the optimal reagent dilution and the total amount of active surface area to be involved in order to obtain both maximum electric power output and the desired cell efficiency; a fuel cell according to the present invention in this case is suitable for producing a variable electric power output at constant electric efficiency; and
- by working in active surface area and reagents dilution configuration variable over time, it is possible to control the electric power output by acting on the total amount of active surface area involved in the reaction and/or by acting on the $EPC(P_{OX};P_{RED})$ characteristic curve of the apparatus to vary the concentration of reagents in the inlet reagent mixture; in this second case, depending upon the $EEC(P_{OX};P_{RED})$ characteristic curve of the cell, it is also possible to control the electric efficiency of the fuel cell.

Claims

1. A method of controlling a fuel cell, **characterized in that** it comprises a step of controlling the fuel cell electric efficiency,

- by checking and adjusting the dilution gradient of the reagents along the overall electrodes active surface, i. e. the variation of reagent dilutions among different parts of the electrodes active surface, and
- by checking and/or adjusting the local current density produced in the fuel cell per unit of electrodes active surface.

2. A method of controlling a fuel cell as claimed in claim 1, **characterized in that** during said step of checking and adjusting the dilution gradient of the reagents along the overall electrodes active surface, said dilution gradient is kept as close as possible to zero.

3. A method of controlling a fuel cell as claimed in claim 1 or 2, **characterized in that** it comprises the following steps:

- determining, at a predetermined operation pressure, an electric efficiency curve ($EEC(P_{OX};P_{RED})$) per unit of active surface area as a function of the local current density produced in the fuel cell per unit of electrodes active surface;
- establishing a desired electrical efficiency value, thereby determining a suitable local current density in accordance to the electric efficiency curve; and
- producing in the fuel cell a local current density value equal to, or lower than, said suitable local current density.

4. A method as claimed in claim 1, 2 or 3, **characterized in that** it comprises the following steps:

- determining, at a predetermined operation pressure, an electric power output curve ($EPC(P_{OX};P_{RED})$) per unit of active surface area as a function of the local current density produced in the fuel cell per unit of electrodes active surface,
- determining the total amount of active surface;

EP 2 710 658 B1

- establishing a desired electric power output value, thereby determining a electrical efficiency and a local current density in accordance with the electric efficiency curve ($EEC(P_{OX};P_{RED})$) and to the electric power output curve ($EPC(P_{OX};P_{RED})$).

- 5 **5.** A method as claimed in any previous claim, **characterized in that**:
- said local current density is established on the base of local dilution value(s) of at least one reagent used to fuel the reaction occurring in the fuel cell.
- 10 **6.** A method as claimed in claim 5, **characterized in that** it comprises the following steps:
- checking the dilution of the oxidant and/or reducing agent/s used to fuel a reaction that will occur in said fuel cell;
- comparing the checked dilution with said desired dilution value; and
- 15 - if the checked dilution/s is/are higher or lower than the desired maximum dilution value/s, controllably increasing or decreasing the dilution of the oxidant and/or reducing agent/s.
- 7.** A method as claimed in any previous claim, **characterized in that** said local current density is increased or decreased either by adding or subtracting reagents or diluents either already available in, or before or while being fed to, the cathodic and/or anodic area of said fuel cell, thereby decreasing or increasing the electric efficiency.
- 20 **8.** A method as claimed in any previous claim, **characterized in that** it comprises a step of evaluating a load curve (LC) describing the time-varying power supply required by the fuel cell throughout the operation life thereof.
- 9.** A method as claimed in any previous claim, **characterized in that** it comprises a step of varying the total amount of active surface area involved in the reaction of the fuel cell, whereby ensuring the required power supply for the fuel cell.
- 25 **10.** A method as claimed in any previous claim, **characterized in that** it comprises the following steps:
- 30 - feeding inlet mixtures, including reagents and diluents, onto the active surfaces of the fuel cell, and discharging them when the concentrations of the reagents are lower than predetermined limit values,
- said diluents being chemicals taking no part in the main Red-Ox reactions occurring in said fuel cell, whereas the reagents are involved in the main Red-Ox reactions occurring in said fuel cell, and the fuel cell includes at least one reaction chamber in which the main Red-Ox reaction occur.
- 35 **11.** A method as claimed in claim 10, **characterized in that** it comprises the step of prearranging and mixing a plurality of raw mixtures, thereby obtaining said inlet mixtures.
- 40 **12.** A method as claimed in claim 11, **characterized in that** the concentration of reagents and diluents of said raw mixtures is known, whereas the inlet mixtures are prepared by mixing available raw mixtures in suitable proportions for feeding inlet mixtures in which the reagents and the diluents are in a desired stoichiometric ratio.
- 13.** A method as claimed in claim 11, **characterized in that** the concentration of raw mixtures is known, and a dilution step is carried out directly in the reaction chamber of the fuel cell.
- 45 **14.** A method as claimed in claim 11, **characterized in that** a diluent mixture is injected under pressure into the reaction chamber of the fuel cell, and the fuel cell is fed with a mixture containing the reagent at a feed flow rate adjusted with respect to the reagents consumption rate, thereby maintaining a constant dilution of the reagents in the reaction chamber of the fuel cell.
- 50 **15.** A method as claimed in claim 11, **characterized in that** the concentration of raw mixtures is known, and the inlet mixtures having a dilution value equal to, or lower than, said maximum dilution value/s are prepared by removing reagents and/or diluents from the raw mixtures, subtraction of said reagents and/or diluents being carried out outside the reaction chamber of the fuel cell by a physical or chemical method.
- 55 **16.** A method as claimed in claim 11, **characterized in that** the concentration of said raw mixtures is known, and the inlet mixtures having a dilution value equal to, or lower than, said maximum dilution value/s are prepared by removing

reagent(s) and/or diluent(s) from the raw mixtures, the subtraction of said reagent(s) and/or diluent(s) being carried out inside the reaction chamber of the fuel cell by a physical or chemical method.

5 17. A method as claimed in any previous claim, **characterized in that**

- said local current density is established or adjusted on the base of the permeability of the gas diffusion layers, the conductivity of the electrodes or the electrolyte concentration.

10 18. A method of evaluating the electric efficiency of a fuel cell comprising the following steps:

- arranging a circuit including a fuel cell and a resistor electrically connected with said fuel cell,
 - supplying said circuit with reagents having known dilution, and with determined dilution gradient along the overall electrodes active surface;
 - evaluating a reference electric efficiency for the fuel cell supplied with said reagents having known dilution;
 15 - evaluating the reference voltage drop across the resistor;
 - changing the dilution of at least one reagent;
 - changing the resistance of the resistor until a voltage drop across the resistor equal to the reference voltage drop is obtained; and
 - evaluating the electric efficiency of the fuel cell on the base of the reference electric efficiency of the fuel cell,
 20 of the dilution of the reagents to be fed to the fuel cell before and after changing the dilution of the at least one reagent, and on the base of the ratio of resistor resistances before and after changing the resistance of the resistor.

25 19. A method of evaluating the electric efficiency of a fuel cell comprising the following steps:

- arranging a first fuel cell, whose electric efficiency is to be determined, in electrical connection with a first resistor of known resistance value, thereby obtaining a first circuit;
 - arranging a second circuit including a second fuel cell identical to the first fuel cell, and a second resistor electrically connected with the second fuel cell,
 30 - supplying the first and the second circuit with reagents having a known dilution(s), and with determined dilution gradient along the overall electrodes active surface;
 - evaluating the electric efficiency of the second fuel cell on the base of its respective reagent dilution(s);
 - evaluating the voltage drop across the resistor of the first and of the second circuit;
 - keeping the second resistor (R^1) at a predetermined value and varying the first resistor (R^λ) until the same voltage drop across the resistor of the first and the second circuit is obtained, thereby determining a first resistor value; and
 35 - evaluating the electric efficiency of the first fuel cell on the base of the electric efficiency of the second fuel cell, of the reagent dilution(s) to be fed to the fuel cell, and on the base of the ratio of first resistor value to second predetermined resistor value.

40 20. A fuel cell suitable for obtaining an electric power output comprising:

- a casing in which an intermediate space is delimited between a cathodic space and an anodic space;
 - at least one electrolytic material located in said intermediate space;
 45 - a positive electrode (5) electrically connected to the end of said electrolytic material at the cathodic space (3);
 - a negative electrode (6) electrically connected to the end of the electrolytic material at the anodic space (4);
 - an oxidant inlet mixture in fluid communication with said cathodic space (3); and
 - a reducing inlet mixture in fluid communication with said anodic space (4), **characterized in that** it comprises:
 - means for checking and adjusting the dilution gradient of the reagents along the overall electrodes active surface, i. e. the variation of reagent dilutions among different parts of the electrodes active surface, and
 50 - control means of the electric efficiency of said fuel cell including means suitable for checking and/or adjusting the current density produced in the fuel cell per unit of active surface area.

55 21. A fuel cell as claimed in claim 20, **characterized in that** said means for checking and adjusting the dilution gradient of the reagents along the overall electrodes active surface comprises means suitable for maintaining constant and null the dilution gradient of the reagents along the overall electrodes active surface, i. e. the variation of reagent dilutions among different parts of the electrodes active surface.

22. A fuel cell as claimed in claim 20 or 21, **characterized in that** said means suitable for checking the current density comprises means suitable for checking the dilution value of the reagents of the fuel cell.

23. A fuel cell as claimed in claim 20, 21 or 22, **characterized in that** it comprises means for checking the dilution of the oxidant and/or reducing agent(s) used to fuel the reaction that will occur out in said fuel cell; means for comparing the checked dilution(s) with desired maximum dilution value(s); and means for controllably increasing or decreasing the dilution of said reagents, thereby decreasing or increasing the electric efficiency of said fuel cell.

24. A fuel cell as claimed in any previous claim 20 to 23, **characterized in that** said it comprises means for varying the electric efficiency including means for increasing or decreasing the current density.

25. A fuel cell as claimed in claim 24, **characterized in that** said means for increasing or decreasing the current density comprises means for adding or subtracting reagents or diluents either already available in, or before or while being fed to, the cathodic and/or anodic area of said fuel cell.

26. A fuel cell as claimed in any previous claim 20 to 25, **characterized in that** said means for adjusting the dilution gradient of the reagents along the overall electrodes active surface, comprises a mixer (9) located in said cathodic and/or anodic space and suitable for maintaining the oxidant and/or reducing, respectively, mixture in a desired turbulent motion condition.

Patentansprüche

1. Verfahren zur Regelung einer Brennstoffzelle, **dadurch gekennzeichnet, dass** es einen Schritt des Regelns der elektrischen Effizienz der Brennstoffzelle umfasst

- durch Überprüfung und Einstellung des Verdünnungsgradienten der Reagenzien entlang der gesamten aktiven Oberfläche der Elektroden, d.h. der Variation der Reagensverdünnungen zwischen verschiedenen Teilen der aktiven Oberfläche der Elektroden; und

- durch Überprüfung und/oder Einstellung der lokalen Stromdichte, die in der Brennstoffzelle pro Einheit der aktiven Oberfläche der Elektroden erzeugt wird.

2. Verfahren zur Regelung einer Brennstoffzelle nach Anspruch 1, **dadurch gekennzeichnet, dass** während des Schritts des Überprüfens und Einstellens des Verdünnungsgradienten der Reagenzien entlang der gesamten aktiven Oberfläche der Elektroden der Verdünnungsgradient so nahe wie möglich bei null gehalten wird.

3. Verfahren zur Regelung einer Brennstoffzelle nach Anspruch 1 oder 2, **dadurch gekennzeichnet, dass** es die folgenden Schritte umfasst:

- Bestimmen, bei einem vorbestimmten Betriebsdruck, einer elektrischen Effizienzkurve ($EEC(P_{OX}; P_{RED})$) pro Einheit des aktiven Oberflächenbereichs als eine Funktion der lokalen Stromdichte, die in der Brennstoffzelle pro Einheit der aktiven Oberfläche der Elektroden erzeugt wird;

- Erstellen eines gewünschten elektrischen Effizienzwerts, dabei Bestimmen einer geeigneten lokalen Stromdichte gemäß der elektrischen Effizienzkurve; und

- Erzeugen, in der Brennstoffzelle, eines lokalen Stromdichtewerts, der gleich wie oder geringer als die geeignete lokale Stromdichte ist.

4. Verfahren nach Anspruch 1, 2 oder 3, **dadurch gekennzeichnet, dass** es die folgenden Schritte umfasst:

- Bestimmen, bei einem vorbestimmten Betriebsdruck, einer elektrischen Leistungsausgabekurve ($EPC(P_{OX}; P_{RED})$) pro Einheit des aktiven Oberflächenbereichs als eine Funktion der lokalen Stromdichte, die in der Brennstoffzelle pro Einheit der aktiven Oberfläche der Elektroden erzeugt wird,

- Bestimmen des Gesamtumfangs der aktiven Oberfläche;

- Erstellen eines gewünschten elektrischen Leistungsausgabewerts, dabei Bestimmen einer elektrischen Effizienz und einer lokalen Stromdichte gemäß der elektrischen Effizienzkurve ($EPC(P_{OX}; P_{RED})$) und der elektrischen Leistungsausgabekurve ($EPC(P_{OX}; P_{RED})$)

5. Verfahren nach einem der vorhergehenden Ansprüche, **dadurch gekennzeichnet, dass**

- die lokale Stromdichte auf der Grundlage eines/von lokalen Verdünnungswerts/Verdünnungswerten von mindestens einem Reagens erstellt ist, das verwendet wird, um die Reaktion, die in der Brennstoffzelle vorgeht, in Gang zu setzen.

6. Verfahren Anspruch 5, **dadurch gekennzeichnet, dass** es die folgenden Schritte umfasst:

- Überprüfen der Verdünnung des Oxidationsmittels und/oder des/der Reduktionsmittel, das/die verwendet wird/werden, um eine Reaktion in Gang zu setzen, die in der Brennstoffzelle stattfinden wird;
 - Vergleichen der überprüften Verdünnung mit dem gewünschten Verdünnungswert; und
 - wenn die überprüfte(n) Verdünnung(en) höher oder niedriger als der/die maximale(n) Verdünnungswert(e) ist/sind, kontrollierte Erhöhung oder Verringerung der Verdünnung des Oxidationsmittels und/oder des/der Reduktionsmittel/s.

7. Verfahren nach einem der vorhergehenden Ansprüche, **dadurch gekennzeichnet, dass** die lokale Stromdichte entweder durch die Zugabe oder die Entnahme von Reagenzien oder Verdünnungsmitteln erhöht oder verringert wird, die entweder in, vor oder während sie dazugegeben werden, dem kathodischen und/oder dem anodischen Bereich der Brennstoffzelle bereits zur Verfügung stehen, wodurch die elektrische Effizienz verringert oder erhöht wird.

8. Verfahren nach einem der vorhergehenden Ansprüche, **dadurch gekennzeichnet, dass** es einen Schritt des Bewertens einer Lastkurve (LC) umfasst, die die zeitabhängige Stromversorgung beschreibt, die von der Brennstoffzelle während der gesamten Lebensdauer davon erforderlich ist.

9. Verfahren nach einem der vorhergehenden Ansprüche, **dadurch gekennzeichnet, dass** es einen Schritt des Variierens des gesamten Umfangs des aktiven Oberflächenbereichs umfasst, der an der Reaktion der Brennstoffzelle beteiligt ist, wodurch die erforderliche Stromversorgung für die Brennstoffzelle sichergestellt wird.

10. Verfahren nach einem der vorhergehenden Ansprüche, **dadurch gekennzeichnet, dass** es die folgenden Schritte umfasst:

- Zuführen von Eintrittsmischungen, darin eingeschlossen Reagenzien und Verdünnungsmittel, auf die aktiven Oberflächen der Brennstoffzelle und deren Entladung, wenn die Konzentrationen der Reagenzien geringer als die vorbestimmten Grenzwerte sind,

wobei die Verdünnungsmittel Chemikalien sind, die nicht an den hauptsächlichen Red-Ox-Reaktionen beteiligt sind, die in der Brennstoffzelle stattfinden, während die Reagenzien an den hauptsächlichen Red-Ox-Reaktionen beteiligt sind, die in der Brennstoffzelle stattfinden, und die Brennstoffzelle mindestens eine Reaktionskammer umfasst, in der die hauptsächliche Red-Ox-Reaktion stattfindet.

11. Verfahren nach Anspruch 10, **dadurch gekennzeichnet, dass** es den Schritt des vorherigen Anordnens und Mischens einer Vielzahl von rohen Mischungen umfasst, wodurch die Eintrittsmischungen erhalten werden.

12. Verfahren nach Anspruch 11, **dadurch gekennzeichnet, dass** die Konzentration der Reagenzien und Verdünnungsmittel der rohen Mischungen bekannt ist, während die Eintrittsmischungen durch die Mischung von verfügbaren rohen Mischungen in geeigneten Proportionen hergestellt werden, um Eintrittsmischungen zuzuführen, in denen die Reagenzien und die Verdünnungsmittel in einem gewünschten stöchiometrischen Verhältnis vorliegen

13. Verfahren nach Anspruch 11, **dadurch gekennzeichnet, dass** die Konzentration der rohen Mischungen bekannt ist und ein Schritt des Verdünnens direkt in der Reaktionskammer der Brennstoffzelle durchgeführt wird.

14. Verfahren nach Anspruch 11, **dadurch gekennzeichnet, dass** eine Verdünnungsmittelmischung unter Druck in die Reaktionskammer der Brennstoffzelle injiziert wird, und die Brennstoffzelle mit einer Mischung, die das Reagens enthält, mit einer Zufuhrflussrate versorgt wird, die mit Bezug auf die Reagensverbrauchsrate eingestellt ist, wobei eine konstante Verdünnung der Reagenzien in der Reaktionskammer der Brennstoffzelle aufrechterhalten wird.

15. Verfahren nach Anspruch 11, **dadurch gekennzeichnet, dass** die Konzentration der rohen Mischungen bekannt

ist und die Eintrittsmischungen einen Verdünnungswert aufweisen, der gleich oder geringer als der/die maximalen Verdünnungswert(e) ist/sind, die durch die Entfernung der Reagenzien und/oder Verdünnungsmittel von den rohen Mischungen hergestellt werden, wobei die Entnahme der Reagenzien und/oder Verdünnungsmittel außerhalb der Reaktionskammer der Brennstoffzelle durch ein physisches oder chemisches Verfahren durchgeführt wird.

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16. Verfahren nach Anspruch 11, **dadurch gekennzeichnet, dass** die Konzentration der rohen Mischungen bekannt ist und die Eintrittsmischungen einen Verdünnungswert aufweisen, der gleich oder geringer als der/die maximalen Verdünnungswert(e) ist/sind, die durch die Entfernung des Reagens/der Reagenzien und/oder des/der Verdünnungsmittel(s) von den rohen Mischungen hergestellt werden, wobei die Entnahme des Reagens/der Reagenzien und/oder des/der Verdünnungsmittel(s) außerhalb der Reaktionskammer der Brennstoffzelle durch ein physisches oder chemisches Verfahren durchgeführt wird.

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17. Verfahren nach einem der vorhergehenden Ansprüche, **dadurch gekennzeichnet, dass** die lokale Stromdichte auf der Grundlage der Permeabilität der Gasdiffusionslagen, der Leitfähigkeit der Elektroden oder der Elektrolytkonzentration erstellt oder eingestellt wird,

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18. Verfahren zur Bewertung der elektrischen Effizienz einer Brennstoffzelle, umfassend die folgenden Schritte:

- Anordnen einer Schaltung, darin eingeschlossen eine Brennstoffzelle und ein Widerstandsgerät, die elektrisch mit der Brennstoffzelle verbunden sind,
- Versorgen der Schaltung mit Reagenzien, die eine bekannte Verdünnung aufweisen, und mit einem bestimmten Verdünnungsgradienten entlang der gesamten aktiven Oberfläche der Elektroden,
- Bewerten einer elektrischen Referenzeffizienz für die Brennstoffzelle, die mit den Reagenzien mit einer bekannten Verdünnung versorgt wird;
- Bewertung des Referenz-Spannungsabfalls auf dem Widerstandsgerät;
- Ändern der Verdünnung von mindestens einem Reagens;
- Ändern des Widerstands eines Widerstandsgeräts, bis ein Spannungsabfall auf dem Widerstandsgerät gleich dem Referenz-Spannungsabfall erhalten wird; und
- Bewerten der elektrischen Effizienz der Brennstoffzelle auf der Grundlage der elektrischen Referenzeffizienz der Brennstoffzelle, der Verdünnung der Reagenzien, die der Brennstoffzelle zugeführt werden sollen, vor und nachdem die Verdünnung von mindestens einem Reagens geändert wird; und auf der Grundlage des Verhältnisses Widerstandsgerät Widerstände vor und nachdem der Widerstand des Widerstandsgeräts geändert wird.

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19. Verfahren zur Bewertung der elektrischen Effizienz einer Brennstoffzelle, umfassend die folgenden Schritte:

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- Anordnen einer ersten Brennstoffzelle, deren elektrische Effizienz bestimmt werden soll, in elektrischer Verbindung mit einem ersten Widerstandsgerät mit einem bekannten Widerstandswert, wodurch eine erste Schaltung erhalten wird;
- Anordnen einer zweiten Schaltung mit einer zweiten Brennstoffzelle, die identisch mit der ersten Brennstoffzelle ist, und eines zweiten Widerstandsgeräts, das elektrisch mit der zweiten Brennstoffzelle verbunden ist,
- Versorgen der ersten und der zweiten Schaltung mit Reagenzien, die (eine) bekannte Verdünnung(en) aufweisen, und mit einem bestimmten Verdünnungsgradienten entlang der gesamten aktiven Oberfläche der Elektroden,
- Bewerten der elektrischen Effizienz der zweiten Brennstoffzelle auf der Grundlage ihrer jeweiligen Reagensverdünnung(en);
- Bewerten des Referenz-Spannungsabfalls auf dem Widerstandsgerät der ersten und der zweiten Schaltung;
- Beibehalten des zweiten Widerstandsgeräts (R^1) auf einem vorbestimmten Wert und Variieren des ersten Widerstandsgeräts (R^λ), bis der gleiche Spannungsabfall auf dem Widerstandsgeräts der ersten und der zweiten Schaltung erhalten wird, wodurch ein erster Widerstandsgerätwert bestimmt wird; und
- Bewerten der elektrischen Effizienz der ersten Brennstoffzelle auf der Grundlage der elektrischen Effizienz der zweiten Brennstoffzelle der Verdünnung(en) der Reagenzien, die der Brennstoffzelle zugeführt werden sollen, und auf der Grundlage des Verhältnisses des ersten Widerstandsgerätwerts zum zweiten vorbestimmten Widerstandsgerätwert.

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20. Brennstoffzelle, die geeignet ist, um einen elektrischen Leistungsausgang zu erhalten, umfassend:

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- ein Gehäuse, in dem ein Zwischenraum zwischen einem kathodischen Raum und einem anodischen Raum begrenzt ist;

EP 2 710 658 B1

- mindestens ein elektrolytisches Material, das sich in dem Zwischenraum befindet;
- eine positive Elektrode (5), die elektrisch mit dem Ende des elektrolytischen Materials am kathodischen Raum (3) verbunden ist;
- eine negative Elektrode (6), die elektrisch mit dem Ende des elektrolytischen Materials am anodischen Raum (4) verbunden ist;
- eine Oxidationsmittel-Eintrittsmischung in fluidischer Kommunikation mit dem kathodischen Raum (3); und
- eine reduzierende Eintrittsmischung in fluidischer Kommunikation mit dem anodischen Raum (4), **dadurch gekennzeichnet, dass** sie Folgendes umfasst:
 - Mittel zur Überprüfung und Einstellung des Verdünnungsgradienten der Reagenzien entlang der gesamten aktiven Oberfläche der Elektroden, d.h. der Variation der Reagensverdünnungen zwischen verschiedenen Teilen der aktiven Oberfläche der Elektroden; und
 - Mittel zur Kontrolle der elektrischen Effizienz der Brennstoffzelle, darin eingeschlossen Mittel, die geeignet sind, um die Stromdichte zu überprüfen und/oder einzustellen, die in der Brennstoffzelle pro Einheit des aktiven Oberflächenbereichs erzeugt wird,

21. Brennstoffzelle nach Anspruch 20, **dadurch gekennzeichnet, dass** das Mittel zur Überprüfung und Einstellung des Verdünnungsgradienten der Reagenzien entlang der gesamten aktiven Oberfläche der Elektroden Mittel umfasst, die geeignet sind, um den Verdünnungsgradienten der Reagenzien entlang der gesamten aktiven Oberfläche der Elektroden konstant und null zu halten, d.h. die Variation von Reagensverdünnungen entlang verschiedener Teile der aktiven Oberfläche der Elektroden.

22. Brennstoffzelle nach Anspruch 20 oder 21, **dadurch gekennzeichnet, dass** das Mittel, das geeignet ist, um die Stromdichte zu überprüfen, geeignete Mittel umfasst, um den Verdünnungswert der Reagenzien der Brennstoffzelle zu überprüfen.

23. Brennstoffzelle nach Anspruch 20, 21 oder 22, **dadurch gekennzeichnet, dass** sie Mittel umfasst, um die Verdünnung des Oxidationsmittels und/oder des/der Reduktionsmittel zu überprüfen, das/die verwendet wird/werden, um die Reaktion in Gang zu setzen, die in der Brennstoffzelle stattfinden wird; Mittel, um die überprüfte(n) Verdünnung(en) mit der/den gewünschten maximalen Verdünnungswert(en) zu vergleichen, und Mittel, um kontrolliert die Verdünnung der Reagenzien zu erhöhen oder zu verringern, wodurch die elektrische Effizienz der Brennstoffzelle verringert oder erhöht wird.

24. Brennstoffzelle nach einem der vorhergehenden Ansprüche 20 bis 23, **dadurch gekennzeichnet, dass** sie Mittel umfasst, um die elektrische Effizienz zu variieren, darin eingeschlossen Mittel, um die Stromdichte zu erhöhen oder zu verringern.

25. Brennstoffzelle nach Anspruch 24, **dadurch gekennzeichnet, dass** das Mittel zur Verringerung oder Erhöhung der Stromdichte Mittel umfasst, um Reagenzien oder Verdünnungsmittel zuzugeben oder zu entnehmen, die entweder in, vor oder während sie dazugegeben werden, dem kathodischen und/oder dem anodischen Bereich der Brennstoffzelle bereits zur Verfügung stehen.

26. Brennstoffzelle nach einem der vorhergehenden Ansprüche 20 bis 25, **dadurch gekennzeichnet, dass** das Mittel, um den Verdünnungsgradienten der Reagenzien entlang der gesamten aktiven Oberfläche der Elektroden einzustellen, einen Mischer umfasst (9), der sich in dem kathodischen und/oder anodischen Raum befindet und geeignet ist, um jeweils das oxidierende- und/oder reduzierende Mischung in einem gewünschten Zustand der turbulenten Bewegung beizubehalten.

Revendications

1. Méthode de contrôle d'une pile à combustible, caractérisé en ceci qu'il comprend une étape consistant à contrôler l'efficacité électrique de la pile à combustible,

- en contrôlant et en ajustant le gradient de dilution des réactifs le long de la surface active des électrodes d'ensemble, c'est à dire la variation de dilution des réactifs entre les différentes parties des surfaces actives des électrodes, et
- en contrôlant et/ou ajustant la densité de courant locale produite dans la pile à combustible par unité de surface active d'électrodes.

EP 2 710 658 B1

2. Une méthode de contrôle de pile à combustible selon la revendication 1, caractérisée en ceci que pendant ladite étape de contrôle et d'ajustement du gradient de dilution des réactifs le long des surfaces actives globales des électrodes, ledit gradient de dilution est maintenu aussi proche que possible de zéro.
- 5 3. Une méthode de contrôle de la pile à combustible selon la revendication 1 ou 2, caractérisée en ceci qu'elle comprend les étapes suivantes:
- 10 - déterminer, à une pression de fonctionnement prédéterminée, une courbe d'efficacité électrique ($ECC(P_{OX};P_{RED})$) par unité de surface active en fonction de la densité de courant locale produite dans la pile à combustible par unité de surface active d'électrodes;
 - établir une valeur de rendement électrique souhaité, de manière à déterminer une densité de courant local approprié conformément à la courbe de rendement électrique; et
 - produire dans la pile à combustible une valeur de densité de courant local égale ou inférieure à ladite densité de courant locale appropriée.
- 15 4. Une méthode selon la revendication 1, 2 ou 3, caractérisée en ceci qu'elle comprend les étapes suivantes:
- 20 - déterminer, à une pression de fonctionnement prédéterminée, une courbe d'efficacité électrique ($EPC(P_{OX};P_{RED})$) par unité de surface active en fonction de la densité de courant locale produite dans la pile à combustible par unité de surface active d'électrodes;
 - déterminer la quantité totale de surface active;
 - établir une valeur de consigne de sortie de l'énergie électrique, en déterminant ainsi un rendement électrique et une densité de courant local conformément à la courbe de rendement électrique ($EEC(P_{OX};P_{RED})$) et de la courbe de puissance électrique de sortie ($EPC(P_{OX};P_{RED})$).
- 25 5. Une méthode selon l'une quelconque des revendications précédentes, caractérisée en ceci que:
- 30 - ladite densité de courant local est établie sur la base de la valeur ou des valeurs de dilution locale d'au moins un réactif utilisé pour alimenter la réaction ayant lieu dans la pile à combustible.
- 35 6. Une méthode selon la revendication 5, caractérisée en ceci qu'elle comprend les étapes suivantes:
- la vérification de la dilution de l'oxydant et/ou de(s) l'agent(s) réducteur(s) utilisés pour alimenter une réaction qui se produira dans ladite pile à combustible;
 - 40 - la comparaison de la dilution vérifiée avec ladite valeur de dilution voulue; et
 - si la(les) dilution(s) vérifiée(s) est(sont) supérieure(s) ou inférieure(s) à la(les) valeur(s) maximale(s) de dilution désirée, augmenter ou diminuer de manière contrôlable la dilution de(s) l'oxydant(s) et/ou de(s) l'agent(s) réducteur(s).
- 45 7. Une méthode selon l'une quelconque des revendications précédentes, caractérisée en ceci que ladite densité de courant locale est augmentée ou diminuée, soit en ajoutant ou en soustrayant des réactifs ou des diluants soit déjà disponibles dans, ou avant ou pendant l'introduction dans la cathode et/ou une zone anodique de ladite pile à combustible, diminuant ou augmentant ainsi l'efficacité électrique.
- 50 8. Une méthode selon l'une quelconque des revendications précédentes, caractérisée en ceci qu'elle comprend une étape consistant à évaluer une courbe de charge (LC) décrivant l'alimentation électrique variant dans le temps requise par la pile à combustible tout au long de la durée de vie de fonctionnement de celle-ci.
- 55 9. Une méthode selon l'une quelconque des revendications précédentes, caractérisée en ceci qu'elle comprend une étape consistant à faire varier la quantité totale de surface active intervenant dans la réaction de la pile à combustible, de sorte à assurer l'alimentation électrique requise pour la pile à combustible.
10. Une méthode selon l'une quelconque des revendications précédentes, caractérisée en ceci qu'elle comprend les étapes suivantes:
- alimenter les mélanges en entrée, y compris les réactifs et les diluants, sur les surfaces actives de la pile à combustible, et les décharger lorsque les concentrations des réactifs sont inférieures à des valeurs limites prédéterminées,

EP 2 710 658 B1

lesdits diluants étant des produits chimiques qui ne participent pas dans les principales réactions Red-Ox se produisant dans ladite pile à combustible, alors que les réactifs sont impliqués dans les principales réactions Red-Ox se produisant dans ladite pile à combustible, et la pile à combustible comprend au moins une chambre de réaction dans laquelle la réaction principale Red-Ox se produit.

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11. Une méthode selon la revendication 10, caractérisée en ceci qu'elle comprend l'étape de préarrangement et de mélange d'une pluralité de mélanges bruts, obtenant ainsi lesdits mélanges d'entrée.

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12. Une méthode selon la revendication 11, caractérisée en ceci que la concentration des réactifs et diluants desdits mélanges bruts est connue, alors que les mélanges d'entrée sont préparés en mélangeant les mélanges bruts disponibles dans des proportions appropriées pour l'alimentation des mélanges d'entrée dans lesquels les réactifs et les diluants sont dans un rapport stoechiométrique souhaité.

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13. Une méthode selon la revendication 11, caractérisée en ceci que la concentration de mélanges bruts est connue, et une étape de dilution est effectuée directement dans la chambre de réaction de la pile à combustible.

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14. Une méthode selon la revendication 11, caractérisée en ceci qu'un mélange de diluant est injecté sous pression dans la chambre de réaction de la pile à combustible, et la pile à combustible est alimentée avec un mélange contenant le réactif à un débit d'alimentation ajusté par rapport au taux de consommation des réactifs, ce qui maintient une dilution constante des réactifs dans la chambre de réaction de la pile à combustible.

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15. Une méthode selon la revendication 11, caractérisée en ceci que la concentration de mélanges bruts est connue, et les mélanges d'entrée ayant une valeur de dilution égale à, ou inférieure à ladite valeur maximale de dilution sont préparés par élimination des réactifs et/ou diluants des mélanges bruts, la soustraction desdits réactifs et/ou diluants étant effectuée à l'extérieur de la chambre de réaction de la pile à combustible par un procédé physique ou chimique.

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16. Une méthode selon la revendication 11, caractérisée en ceci que la concentration desdits mélanges bruts est connue, et les mélanges d'entrée ayant une valeur de dilution égale à, ou inférieure à ladite valeur maximale de dilution sont préparés en éliminant le(s) réactif(s) et/ou diluant(s) à partir des mélanges bruts, la soustraction dudit(desdits) réactif(s) et/ou diluant(s) étant réalisée à l'intérieur de la chambre de réaction de la pile à combustible par un procédé physique ou chimique.

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17. Une méthode selon l'une quelconque des revendications précédentes, caractérisée en ceci que:

- ladite densité de courant local est établie ou ajustée sur la base de la perméabilité des couches de diffusion de gaz, la conductivité des électrodes ou de la concentration d'électrolyte.

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18. Une méthode d'évaluation de l'efficacité électrique d'une pile à combustible comprenant les étapes suivantes:

- agencer un circuit comprenant une pile à combustible et une résistance connectée électriquement à ladite pile à combustible,

- fournir ledit circuit avec des réactifs ayant une dilution connue, et avec un gradient de dilution déterminé sur l'ensemble de la surface active des électrodes;

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- évaluer un rendement électrique de référence pour la pile à combustible fourni avec lesdits réactifs ayant une dilution connue;

- évaluer la chute de la tension de référence à travers la résistance;

- modifier la dilution d'au moins un réactif;

- changer la valeur de la résistance jusqu'à ce qu'une chute de tension aux bornes de la résistance égale à la chute de tension de référence soit obtenue; et

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- évaluer l'efficacité électrique de la pile à combustible sur la base de la référence d'efficacité électrique de la pile à combustible, de la dilution des réactifs à introduire dans la pile à combustible avant et après le changement de la dilution d'au moins un réactif, et sur la base du rapport des valeurs de la résistance avant et après un changement de valeurs de la résistance.

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19. Une méthode d'évaluation de l'efficacité électrique d'une pile à combustible comprenant les étapes suivantes:

- agencer une première pile à combustible, dont l'efficacité électrique est à déterminer, en liaison électrique avec une première résistance de valeur connue, de manière à obtenir un premier circuit;

EP 2 710 658 B1

- disposer un deuxième circuit comprenant une deuxième pile à combustible identique à la première, et une deuxième résistance connectée électriquement à la seconde pile à combustible,
 - fournir le premier et le deuxième circuit avec des réactifs ayant une(des) dilution(s) connue(s), et avec un gradient de dilution déterminé le long de l'ensemble de surface active des électrodes;
 - évaluer l'efficacité électrique de la seconde pile à combustible sur la base de sa(ses) dilution(s) de réactif respectif;
 - évaluer la chute de tension aux bornes de la résistance du premier et du deuxième circuit;
 - garder la seconde résistance (R^1) à une valeur prédéterminée et faire varier la première résistance (R^λ) jusqu'à obtenir la même baisse de tension dans la résistance du premier et du second circuit, déterminant ainsi une première valeur de la résistance; et
 - évaluer l'efficacité électrique de la première pile à combustible sur la base de l'efficacité électrique de la seconde pile à combustible, de la (des) dilution(s) de réactif à introduire dans la pile à combustible, et sur la base du rapport de la première valeur de résistance et de la seconde valeur de résistance prédéterminée.
- 15 **20.** Une pile à combustible appropriée pour obtenir une puissance électrique comprenant:
- un boîtier dans lequel un espace intermédiaire est délimité entre un espace cathodique et un espace anodique;
 - au moins un matériau électrolytique situé dans ledit espace intermédiaire;
 - une électrode positive (5) connectée électriquement à l'extrémité dudit matériau électrolytique à l'espace cathodique (3);
 - une électrode négative (6) connectée électriquement à la fin du matériau électrolytique à l'espace anodique (4);
 - un mélange d'entrée d'oxydant en communication fluide avec ledit espace cathodique (3); et
 - un mélange réducteur d'entrée en communication fluide avec ledit espace anodique (4), caractérisé en ceci qu'il comprend:
- les moyens de contrôle et d'ajustement du gradient de dilution des réactifs le long de la surface active d'ensemble des électrodes, c'est à dire la variation de dilution des réactifs entre les différentes parties des surfaces actives des électrodes, et
 - des moyens de commande de l'efficacité électrique de ladite pile à combustible comprenant des moyens aptes à vérifier et/ou ajuster la densité de courant produite dans la pile à combustible par unité de surface active.
- 30 **21.** Une pile à combustible selon la revendication 20, caractérisée en ceci que lesdits moyens de contrôle et de réglage du gradient de dilution des réactifs le long des surfaces actives d'ensemble des électrodes comprennent des moyens aptes à maintenir constant et nul le gradient de dilution des réactifs le long de la surface active d'ensemble des électrodes, c'est à dire la variation de la dilution des réactifs entre les différentes parties des surfaces actives des électrodes.
- 35 **22.** Une pile à combustible selon la revendication 20 ou 21, caractérisée en ceci que ledit moyen adapté pour vérifier la densité de courant comprend des moyens aptes à vérifier la valeur de dilution des réactifs de la pile à combustible.
- 40 **23.** Une pile à combustible selon la revendication 20, 21 ou 22, caractérisée en ceci qu'elle comprend des moyens de contrôle de la dilution de(s) agent(s) oxydant(s) et/ou réducteur(s) utilisé pour alimenter la réaction qui se produira dans ladite pile à combustible; un moyen pour comparer la(es) dilution(s) contrôlée(s) par la(es) valeur(s) de dilution maximum désirée(s); et des moyens pour augmenter ou diminuer de manière contrôlable la dilution desdits réactifs, diminuant ou augmentant ainsi le rendement électrique de ladite pile à combustible.
- 45 **24.** Une pile à combustible selon l'une quelconque des revendications précédentes 20 à 23, caractérisée en ceci qu'elle comprend des moyens pour faire varier l'efficacité électrique comprenant des moyens pour augmenter ou diminuer la densité de courant.
- 50 **25.** Une pile à combustible selon la revendication 24, caractérisée en ceci que lesdits moyens pour augmenter ou diminuer la densité de courant comprennent des moyens pour additionner ou soustraire des réactifs ou des diluants soit déjà disponibles dans, ou avant ou pendant l'introduction dans la zone cathodique et/ou anodique de ladite pile à combustible.
- 55 **26.** Une pile à combustible selon l'une quelconque des revendications précédentes 20 à 25, caractérisée en ceci que lesdits moyens pour ajuster le gradient de dilution des réactifs le long de la surface active d'ensemble des électrodes, comprennent un mélangeur (9) situé dans ledit espace cathodique et/ou anodique et adapté pour le maintien de l'oxydant et/ou réducteur, respectivement, en mélange turbulent dans un état de mouvement souhaité.

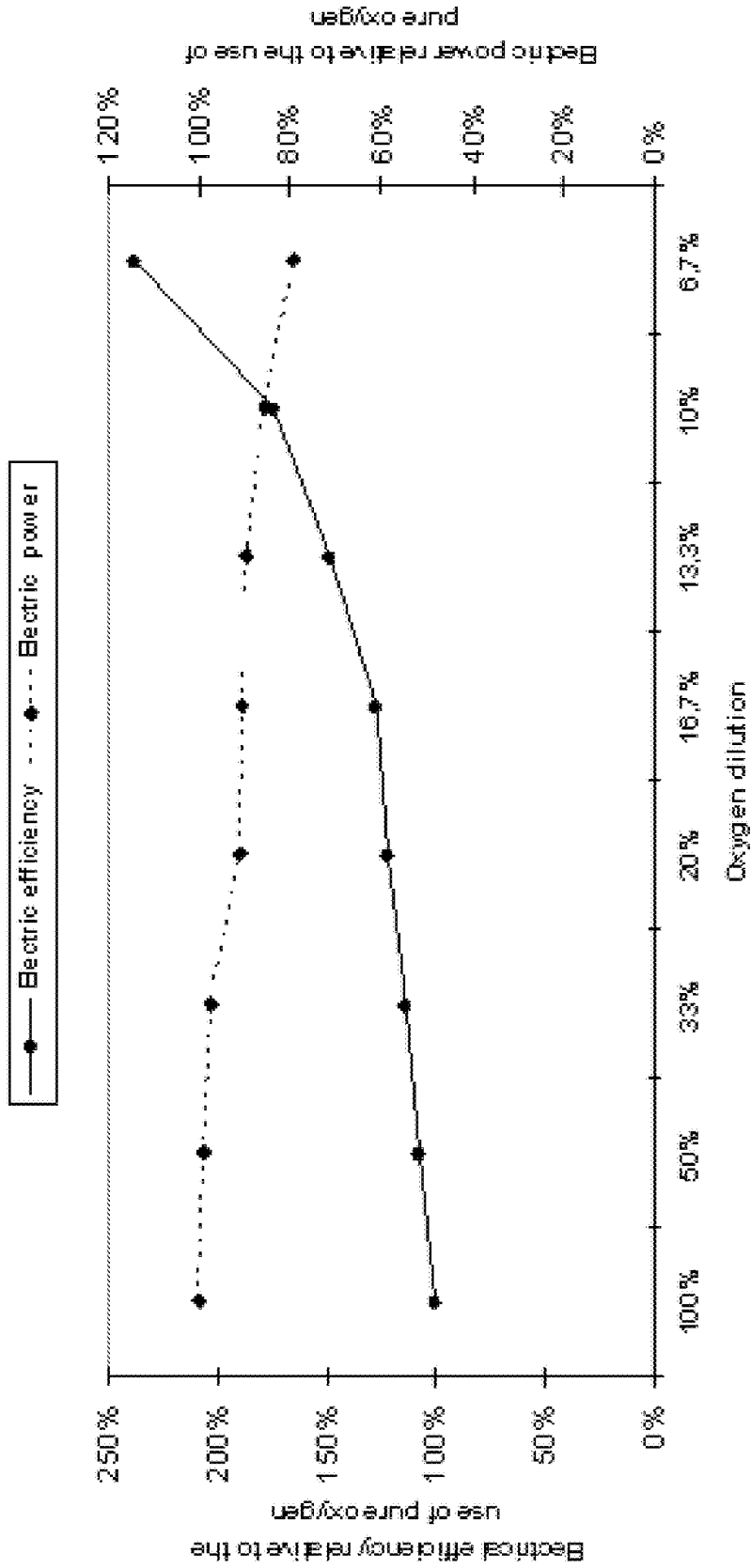


Fig. 1

Inlet Oxidant mixture prepared by using pure (99,995%) oxygen and pure nitrogen (99,98%) at a temperature of 20°C and a pressure of 1 atm.

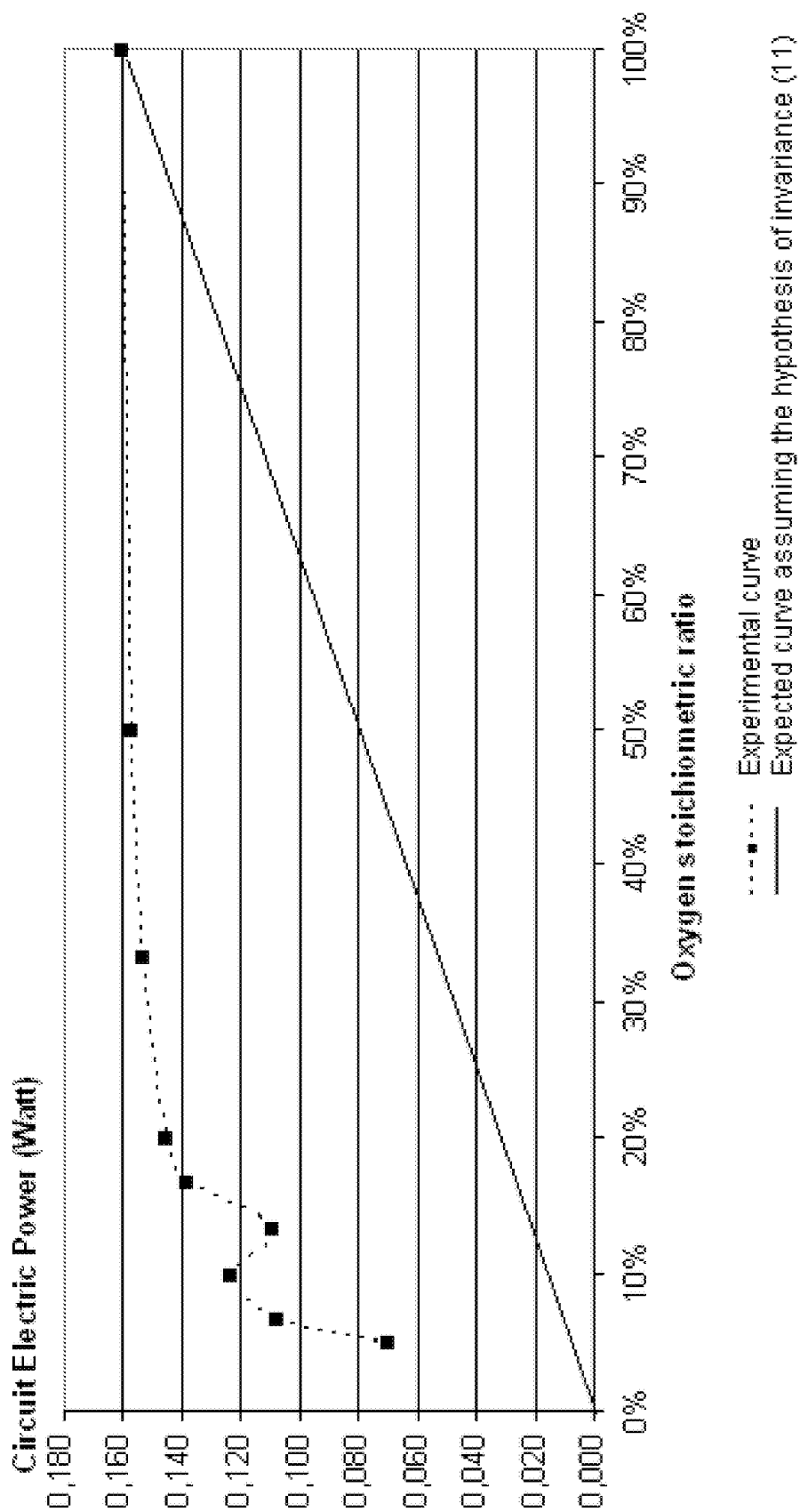
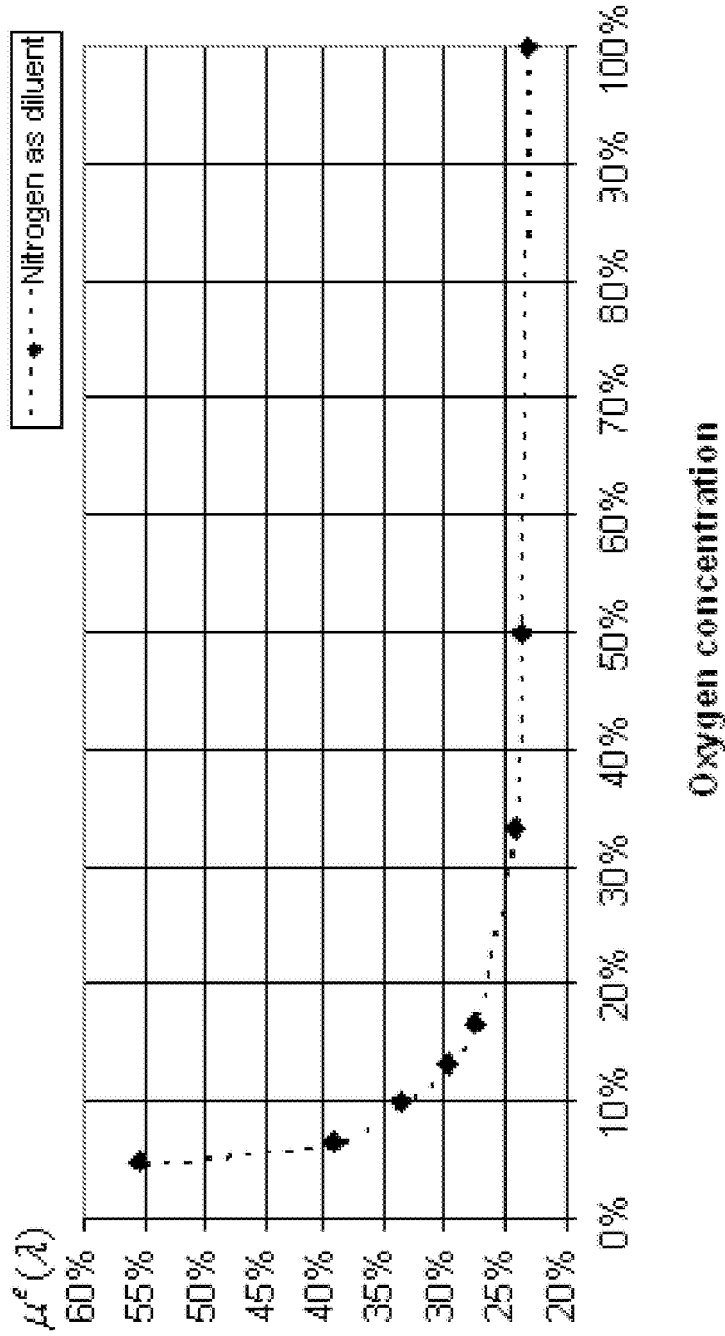


Fig. 2

Inlet Oxidant mixture prepared by using pure (99,995%) oxygen and pure nitrogen (99,998%) at a temperature of 20°C and a pressure of 1 atm.



Inlet Oxidant mixture prepared by using pure (99.995%) oxygen and pure nitrogen (99.98%) at a temperature of 20°C and a pressure of 1 atm.

Fig. 3

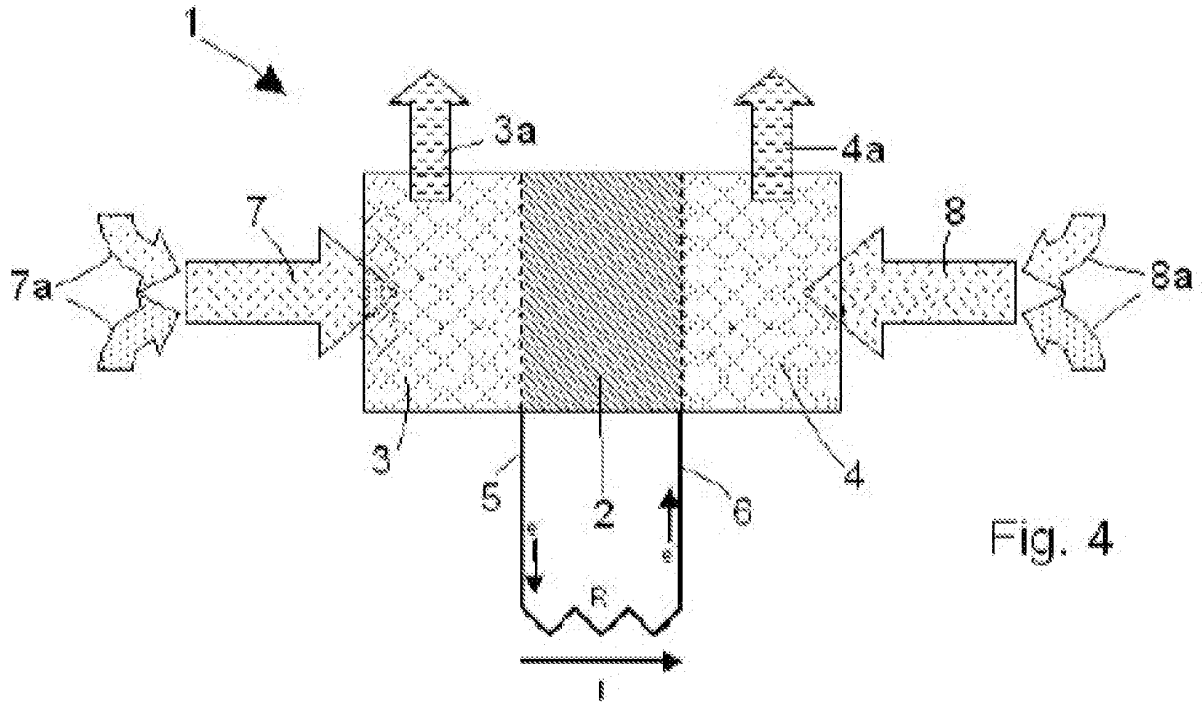


Fig. 4

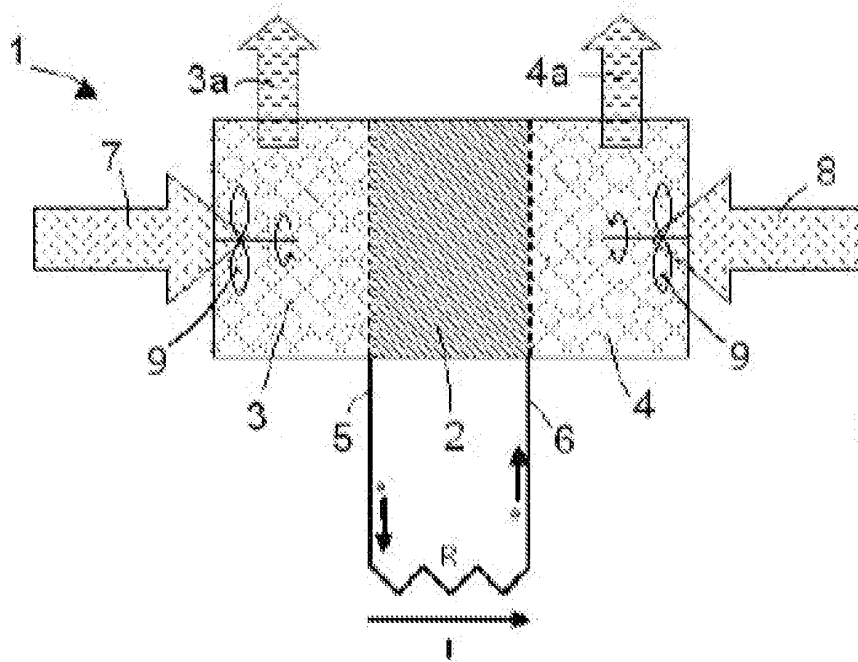
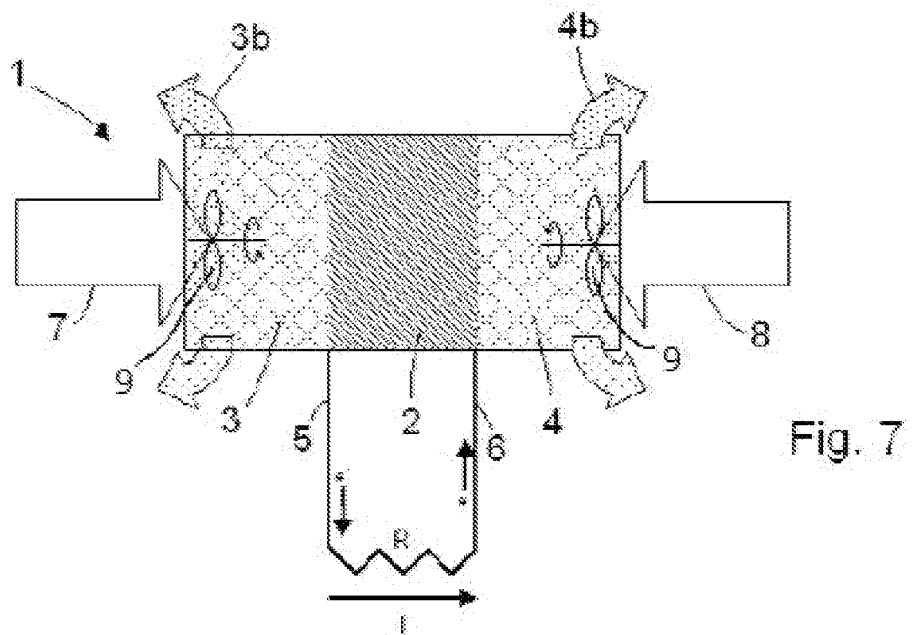
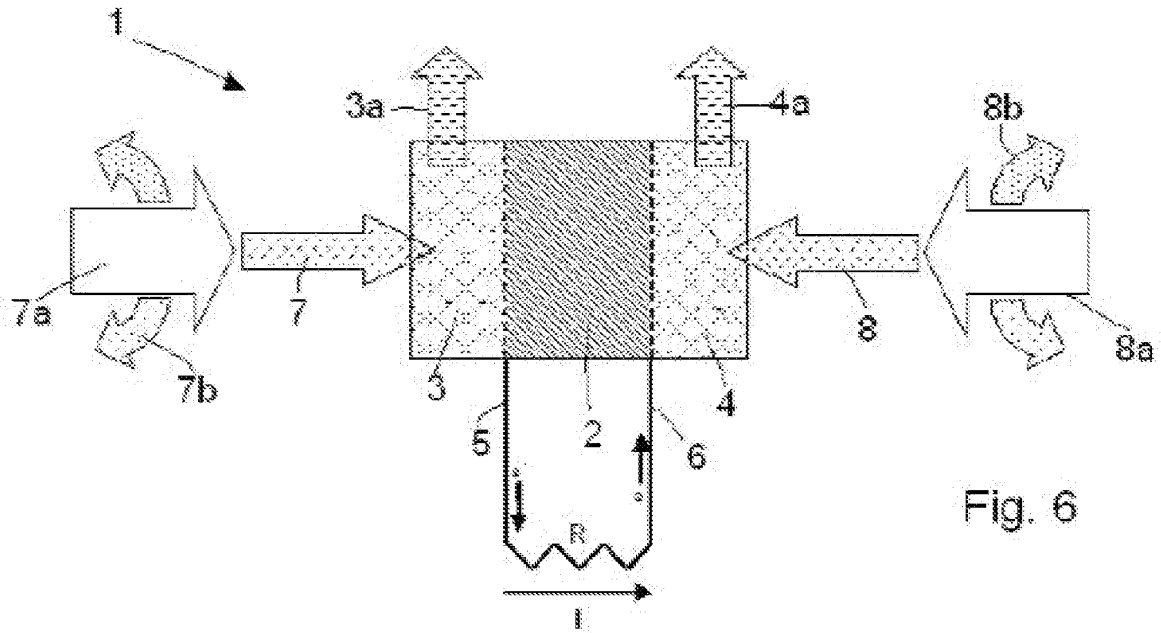


Fig. 5



REFERENCES CITED IN THE DESCRIPTION

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