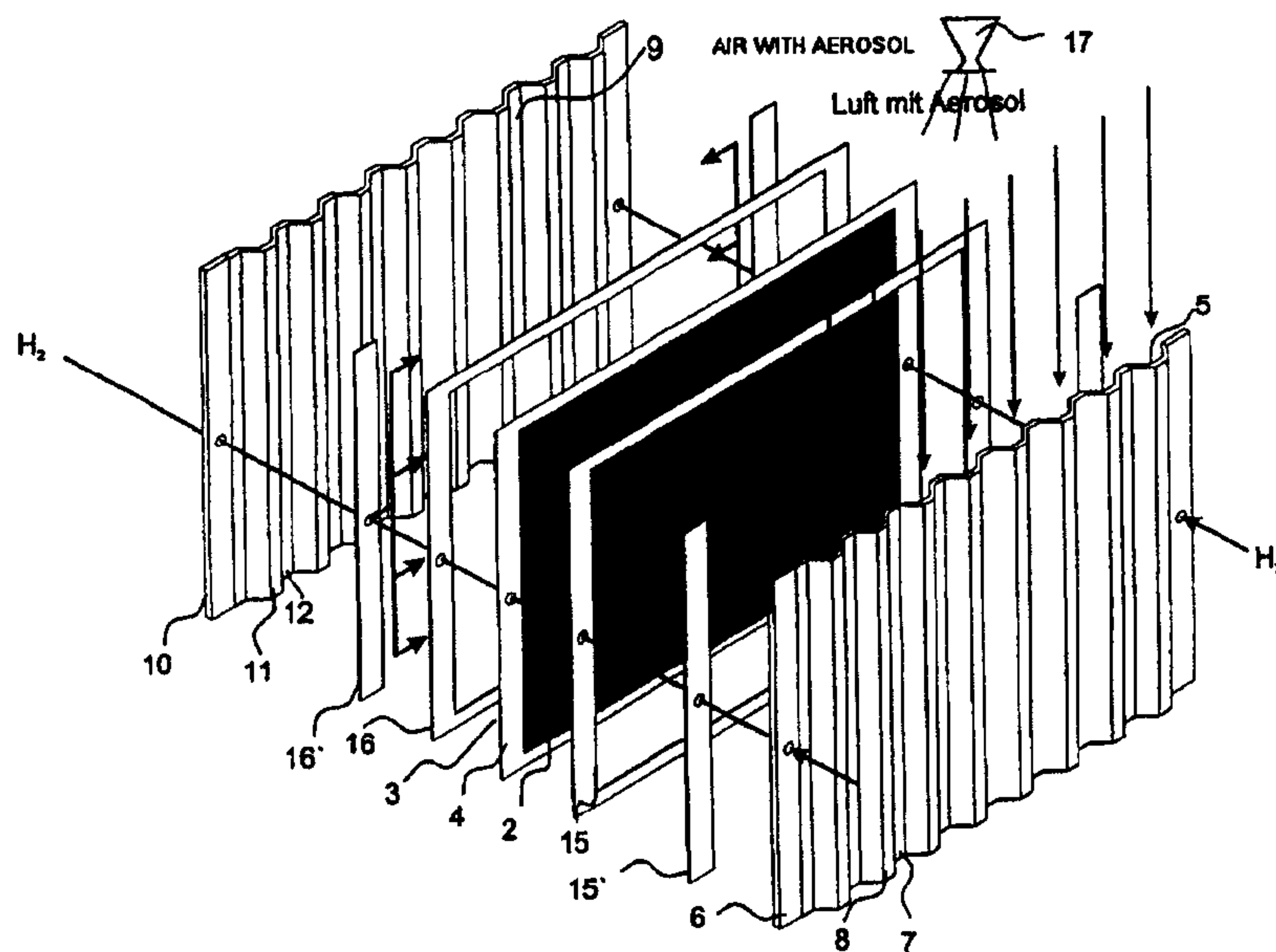




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(54) **REFROIDISSEMENT ET HUMIDIFICATION DE PILES A
COMBUSTIBLES A POLYELECTROLYTES**
(54) **COOLING AND WETTING POLYMER-ELECTROLYTE FUEL
CELLS**



(57) Une pile à combustible à polyélectrolyte (1) utilisant l'air comme agent d'oxydation, sous faible surpression, et différents gaz de combustion, en particulier l'hydrogène, est refroidie par alimentation d'eau liquide directement dans les arrivées de gaz (5, 9) de l'air de combustion et, éventuellement, du gaz de combustion. L'eau introduite dans les arrivées de gaz sert à même temps à l'humidification du polyélectrolyte solide (4).

(57) A polymer-electrolyte fuel cell (1) using air as oxidizing agent, under low overpressure, and different combustion gases, in particular hydrogen, is cooled by feeding liquid water directly into the gas passages (5, 9) of the combustion air and possibly of the combustible gas. The water introduced into the gas passages also serves to wet the solid polymer electrolyte (4).

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Abstract

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10 A polymer electrolyte fuel cell (1) for the oxidizing
agent air at slight overpressure and for various
burnable gases, in particular hydrogen, is cooled by
supply of liquid water directly to the gas channels (5,
9) of the combustion air and possibly of the burnable
gas. The water introduced into the gas channels simul-
taneously serves for humidifying the solid polymer elec-
15 trolyte (4).

(Fig. 1)

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**Cooling and Humidifying of
Polymer Electrolyte Fuel Cells**

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The invention relates to fuel cells which contain solid polymer membranes as electrolytes, preferably use hydrogen as burnable gas and use air or oxygen under low pressure as oxidizing agent. The invention relates furthermore to a method of simultaneously cooling the fuel cells and humidifying the polymer electrolyte membranes.

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Polymer electrolyte membrane fuel cells, as they are commonly employed for producing electric current, contain an anode, a cathode and an ion exchange membrane disposed therebetween. A plurality of fuel cells constitutes a fuel cell stack, with the individual fuel cells being separated from each other by bipolar plates acting as current collectors. For generating electricity, a burnable gas, e.g. hydrogen, is introduced into the anode region, and an oxidizing agent, e.g. air or oxygen, is introduced into the cathode region. Anode and cathode, in the regions in contact with the polymer electrolyte membrane, each contain a catalyst layer. In the anode catalyst layer, the fuel is oxidized thereby forming cations and free electrons, and in the cathode catalyst layer, the oxidizing agent is reduced by taking up electrons. The cations migrate through the ion exchange membrane to the cathode and react with the reduced oxidizing agent, thereby forming water when hydrogen is used as burnable gas and oxygen is used as oxidizing agent. In the reaction of burnable gas and oxi-

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dizing agent, there are released considerable amounts of heat that must be dissipated by cooling. Cooling so far has been achieved by cooling channels in the bipolar plates through which deionized water is flown.

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With this kind of cooling, tremendous material problems result since there are typically about 50 to 300 bipolar plates connected in series, with the cooling water thus electrically joining together different potentials. The result thereof are material decompositions. In accordance therewith, solely graphite or gold-plated metal are feasible as material for the bipolar plates.

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Furthermore, it is necessary to keep the polymer membrane moist, since the conductivity value of the membrane is greatly dependent on its water content. To prevent drying up of the membrane, there was thus required a complex system for humidifying the reaction gases.

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It is the object of the invention to make available a polymer electrolyte fuel cell and a polymer electrolyte fuel cell stack, respectively, in which the polymer electrolyte membrane of a fuel cell has the optimum moisture content at all times during operation and in which at the same time sufficient cooling is ensured.

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An additional object of the invention consists in making available a method which renders possible to keep the polymer electrolyte membrane of a polymer electrolyte fuel cell at an optimum moisture content during operation of the fuel cell and at the same time to sufficiently cool the fuel cell.

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These objects are met by the polymer electrolyte fuel cell according to claim 1, the polymer electrolyte fuel

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cell stack according to claim 11, the method of cooling and humidifying a polymer electrolyte fuel cell according to claim 12 and the method according to claim 22.

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Polymer electrolyte membranes require a high water content to ensure optimum conductivity for H^+ ions. The water content must be maintained as a rule by supply of water, as otherwise the burnable gas flows and oxidizing agent gas flows flowing through the cell dry up the membrane. However, to counteract possible drying up by the addition of an excess of water, is not sensible since water in too large quantities results in flooding of the electrodes, i.e. the pores of the electrodes are clogged. Simple ascertaining and regulating the particular amount of water required has not been possible so far.

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Preferred embodiments are indicated in the respective dependent claims.

In the drawings:

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Fig. 1 shows a preferred embodiment of a fuel cell according to the invention,

Fig. 2 shows a circuit for measuring the impedance of a fuel cell.

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Fig. 3 shows the dependency of the conductivity a Nafion® membrane on the water content of the membrane.

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A polymer electrolyte fuel cell according to the invention uses air or oxygen at slight overpressure as oxidizing agent. Preferred is an overpressure of less

- 4 -

than 2 bar, with an overpressure of less than 0.5 bar being particularly preferred. The necessary pressure difference can also be obtained by suction. As burnable gas, preferably hydrogen is used, but the use of other
5 burnable gases is in principle possible as well. As polymer electrolyte membrane, preferably Nafion® is employed. Hydrogen is supplied to the individual fuel cells of a stack and distributed via gas channels in the anode region. Air is supplied at the same time and distributed via gas channels in the cathode region. The
10 hydrogen migrates to the anode catalyst layer and forms cations there which migrate through the electrolyte, a proton exchange membrane, to the cathode. At the cathode, oxygen migrates to the cathode catalyst layer and
15 is reduced there. During the reaction with the cations, water is created as reaction product. Due to the reaction heat, the water formed evaporates, which results in a certain cooling effect. This cooling effect, however, is not sufficient on the one hand, and on the
20 other hand the membrane in the course of operation of the fuel cell is increasingly depleted of humidity.

As can be seen from Fig. 3 for Nafion® NE 105 (30 °C), the conductivity of ion-conducting membranes increases
25 with the H₂O content. $N(H_2O)/N(SO_3H)$ designates the number of water molecules per sulphonic acid remainder of the membrane.

A reduction of the moisture content of the solid polymer electrolyte membrane of a fuel cell thus has the consequence that its internal resistance increases, i.e. that
30 its conductivity value decreases. The conductivity value of the membrane is dependent in extreme manner on its water content. What is essential for efficient operation of a polymer electrolyte fuel cell is thus that the
35 polymer electrolyte membrane at all times have the optimum

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humidity corresponding to the particular operating conditions (temperature, load, air ratio).

5 For maintaining optimum humidity, it is possible according to the invention to determine during operation of the fuel cell, preferably regularly or continuously, whether the membrane is moist in optimum manner or whether the addition of water is necessary and which quantity of water needs to be added, respectively.

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The amount of water added basically can vary very much. It is dependent on the particular operating conditions of the fuel cell, and it is dependent in particular also on the type of cooling of the fuel cell. Frequently,
15 fuel cells are fed with water for cooling which, depending on the construction of the fuel cells, humidifies to a certain extent also the membrane. As a rule, less additional water has to be supplied than in case of cells employing exclusively air cooling, for
20 example.

The conductivity value of the membrane depends on the water content thereof. During operation of a fuel cell, however, the conductivity value of the membrane cannot
25 be measured directly. According to the invention, preferably the impedance of the fuel cell (value of the impedance or particularly preferred, the real part of the impedance) is ascertained. Since the conductivity value of the membrane is a continual, monotonic function of these quantities, the necessary amount of water
30 can also be regulated on the basis of the impedance.

A possible circuit for measuring the impedance of a fuel cell is shown in Fig. 2.

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Direct measurement of the conductivity value and thus of the moisture content of a polymer electrolyte membrane of a fuel cell by determination of the impedance is carried out by modulation of the cell voltage with an alternating signal having a frequency of 1 to 20 kHz. In case of a fuel cell stack, suitably the average moisture content of several membranes is measured. The quotient of alternating voltage and the resulting current response is a measure for the moisture. In Fig. 2, BZ represents the fuel cell and R_L represents the load resistor. Connected in parallel to the load resistor is an assembly of capacitor C, resistor R and alternating voltage source U, which is suitable to produce small alternating voltages (in the order of magnitude of about 10 mV) and large currents (in the order of magnitude of about 10 A). The voltage of the fuel cell is modulated by the alternating signal (about 1 to 20 kHz). The alternating voltage component U effects an alternating current I to be superimposed on the fuel cell current. The quotient of alternating voltage and alternating current is a measure for the impedance of the fuel cell and thus a measure for the moisture of the polymer electrolyte membrane and for the necessary amount of water to be added, respectively.

However, the amount or value of the impedance is dependent, in addition to the conductivity of the membrane, on further determinative quantities, namely on the size of the catalyst surface in contact with the membrane, the ohmic resistance of the electrodes and the poisoning of the membrane by foreign ions. These quantities are subject to a certain amount of change in the course of the service life of a fuel cell, with the deviations due to change of the ohmic resistance of the electrodes and due to poisoning of the membrane by foreign ions being as a rule negligible. In the course of the life of a

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fuel cell, the value of the impedance which corresponds to the optimum membrane moisture under the given operating conditions (desired or set value of the amount of the impedance), can thus vary. Thus, the desired value to be observed of the amount of the impedance should be set each time anew in the course of arising maintenance work. In doing so, the new desired value is determined by maximizing the performance of the fuel cell. During operation of the fuel cell, the optimum desired value can be matched anew in alternative manner by Fuzzy logic or other methods familiar to the expert, in accordance with the changed conditions.

A measure for the conductivity of the membrane that is largely independent of the catalyst surface (whose change in essence is responsible for the change of the desired value of the impedance) is obtained if, in addition to the amount of the impedance, its phase angle is considered as well. If the real part of the impedance determined electronically therefrom is regarded as regulating variable, one sole desired value can be employed even over the entire service life of the fuel cell.

During operation of the fuel cells, the impedance (amount or real part) can be measured continuously or at regular intervals. In case a too low conductivity value of the membrane or membranes is calculated on the basis of the measurement, water is supplied to the system, for example by electronically controlled opening of water inlet valves, as is usual, until the desired value of the impedance is reached again.

In case of fuel cell stacks with a plurality of fuel cells, it is favorable not to determine the amount or the real part of the impedance for each membrane indivi-

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dually, but to determine average values for a plurality of cells of the stack or even for all cells of the stack jointly and to arrange the necessary addition of water in accordance therewith.

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Irrespective of the manner of determination of the optimum water content of the membrane and the regulation of the water introduction, it is possible according to the invention to use membrane humidifying water simultaneously for cooling the fuel cell and for thus ensuring sufficient cooling. This is achieved according to the invention in that in case of a fuel cell designed as outlined hereinbefore, ion-free water in liquid form is introduced directly into the gas channels for the combustion air. As an alternative, the water can also be introduced directly into the gas channels for the burnable gas.

A proven solution is the introduction of water both in the cathode region and in the anode region, particularly with operating conditions causing severe drying up of the membrane.

The liquid water evaporates in the hot fuel cell and effects efficient cooling of the cell due to the phase conversion taking place. Furthermore, it penetrates into the polymer electrolyte membrane and keeps it moist.

The easiest possibility of adding the necessary amount of water to the air flow and to the air and/or hydrogen flow, respectively, consists in introducing the water into the gas channels by means of a metering pump, in numerous thin lines, e.g. capillaries. In doing so, no substantial mixing of the water with the air and the burnable gas, respectively, takes place, so that the

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free water surface available for evaporation is relatively small.

5 A considerably larger free water surface and thus faster humidifying of the membrane and more efficient cooling is obtained if the required amount of water is added to the reaction gas flows in mixed form, i.e. as aerosol. The water-in-air aerosol and, if applicable, the water-in-burnable gas aerosol contain water in the form of
10 droplets with a size of 2 to 20 μm , which ensure rapid vaporization or evaporation. The aerosol can be produced for instance with the aid of ultrasonic atomizers or nozzles. The simplest production of the aerosol, which at the same time requires the least amount of energy,
15 takes place by means of ultrasonic atomizers at frequencies of at least 100 kHz.

A particularly advantageous embodiment of the invention consists in designing the passages or channels for receiving the water-in-air aerosol and the water-in-burnable gas aerosol, respectively, as shown in Fig. 1. In a fuel cell stack, each fuel cell is confined on the anode side and on the cathode side by a bipolar plate
20 10, 6 each. The anode-side bipolar plate simultaneously is the cathode-side bipolar plate of a neighboring cell and the cathode-side bipolar plate at the same time is the anode-side bipolar plate of the other neighboring cell.

30 The bipolar plate, at least in a partial region, is of corrugated sheet structure, i.e. it has alternating elevations and depressions. A surface of the bipolar plate 6 contacts, with its elevations 7, the cathode region 2 of the fuel cell, whereby the depressions 8 located
35 between two adjacent elevations each together with the cathode region form channels 5 for receiving water-in-

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air aerosol. In corresponding manner, the bipolar plate 10 contacts with one surface the anode region 3 of the cell, so that the depressions 12 located between two adjacent anode-side elevations 11 each also form channels 9 together with the anode region 3. These can serve for taking up water-in-burnable gas aerosol.

In the embodiment shown in Fig. 1, hydrogen as burnable gas is introduced perpendicularly to the plate surface through bores. The hydrogen first enters channel 9 in communication with the supply opening and from there diffuses or flows into the adjacent porous anode region. From there, the hydrogen diffuses in part to the anode catalyst layer and in part into additional gas channels 9 in the plane of the anode region. Because of the outstanding diffusion properties of hydrogen, the entire anode region thus is uniformly supplied with hydrogen without a problem.

If cooling water is to be supplied as well along with the burnable gas, it is as a rule more advantageous to choose the same type of feeding as in the cathode region, i.e. to feed fuel and water into each individual channel 9. Because of the poor diffusion properties of water in comparison with hydrogen, only little water would penetrate the anode otherwise, and the cooling effect would thus be low.

The construction has no separate cooling channels whatsoever. A specific advantage consists in particular in that the path of the aerosol through the channels 5 of the cell constitutes a straight line. The corrugated sheet structure of the bipolar plate with straight gas paths permits to minimize depositions of the aerosol and to conduct the necessary volume flows with low pressure drop.

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Flooding and clogging of the water-conducting paths by water droplets, as is frequently the case with porous plates, does not take place. Besides, the "corrugated sheet plate" can be manufactured very inexpensively and simply in terms of manufacturing technology.

The anode and cathode regions are each designed as diffusion layers carrying a suitable catalyst and disposed on the opposite sides of the polymer electrolyte membrane 4.

Air gaskets 15, 15' and hydrogen gaskets 16, 16' seal the cell in gastight manner.

To increase the dwell period of the water in the cell and to thus enable complete evaporation, the walls of the gas channels 5 and/or the gas channels 9 can be coated with a hydrophilic absorbent layer, for instance with felt. The hydrophilic, absorbent layer distributes the introduced amount of water in particularly even manner and retains the same up to evaporation.

The amount of water required for obtaining optimum membrane humidification, as outlined hereinbefore, can be determined and regulated electronically. The amount of water introduced into the fuel cell has to fulfil two tasks: cooling the cell and humidifying the membrane. For regulation of the required amount of water, however, only the setting of the suitable membrane moisture is taken into consideration. Depending on the parameters temperature, load, air ratio and the like, the optimum membrane moisture and thus the optimum conductivity value of the membrane is determined experimentally. The addition of water varies depending on the conductivity value to be reached. The cell temperature varies within

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wide limits depending on the operating conditions. As long as sufficient water is introduced to ensure optimum membrane moisture, a sufficient cooling effect, however, is ensured as well.

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For keeping the moisture content of the reaction gases and the temperature thereof along the direction of flow as constant as possible in a fuel cell or fuel cell stack, the reaction gas, in particular the air, may be caused to pass the cell stack several times. This takes place by recirculation of the air/water mixture leaving the fuel cells and the burnable gas/water mixture leaving the fuel cells, respectively, to the respective suction or intake flow.

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Thus, it is possible according to the invention in a polymer electrolyte fuel cell, by introducing ion-free water in liquid form directly into the gas channels of the combustion air and/or the burnable gas, to ensure at the same time keeping of an optimum membrane moisture and, thus, an optimum conductivity value of the membrane as well as sufficient cooling of the fuel cell.

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Translation of the Annex to the IPER

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Claims

1. A polymer electrolyte fuel cell (1) comprising an
10 anode region (3), a cathode region (2), a polymer
electrolyte membrane (4) disposed therebetween, a
means for supplying air as oxidizing agent to the
cathode region, gas channels (5) for distributing
15 the air in the cathode region, a means for supplying
burnable gas to the anode region, and gas channels
(9) for distributing the burnable gas in the anode
region, as well as bipolar plates (10, 6) confining
the cell on the anode side or on the anode side and
20 on the cathode side and being corrugated at least in
a partial region and having elevations (11, 7) and
depressions (12, 8), with the depressions (8) on the
cathode side constituting the gas channels (5) for
distributing the air in the cathode region and the
25 depressions (12) on the anode side constituting the
gas channels (9) for distributing the burnable gas
in the anode region,
characterized by
- a means for introducing water in liquid form
30 directly into the gas channels (9) of the
burnable gas in the anode region in an amount
necessary to keep the membrane moist and to cool
the fuel cell, and
 - by a means for returning burnable gas/water
mixture leaving the fuel cell, or air/water
35 mixture and burnable gas/water mixture, to the

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means for supplying burnable gas or air and burnable gas.

2. The polymer electrolyte fuel cell (1) of claim 1,
5 characterized in that there is provided a means for introducing water in liquid form directly into the gas channels (5) of the air in the cathode region.
3. The polymer electrolyte fuel cell (1) of claim 1 or
10 2,
characterized in that the means for introducing water in liquid form is designed such that, during introduction, no substantial mixing of water and burnable gas or of air and burnable gas and water
15 and burnable gas takes place.
4. The polymer electrolyte fuel cell (1) of any of
claims 1 to 3,
characterized in that the means for introducing
20 water comprises a plurality of thin lines opening into the gas channels (9) of burnable gas or into the gas channels (5; 9) of air and burnable gas.
5. The polymer electrolyte fuel cell (1) of claim 1 or
25 2,
characterized in that the means for introducing water contains means for producing an aerosol of water in burnable gas or of water in air and water in burnable gas.
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6. The polymer electrolyte fuel cell (1) of claim 5,
characterized in that the means for producing the aerosol are nozzles.
- 35 7. The polymer electrolyte fuel cell (1) of claim 5,

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characterized in that the means for producing the aerosol are ultrasonic atomizers (17).

- 5 8. The polymer electrolyte fuel cell (1) of any of claims 1 to 7, characterized in that the gas channels (9) or the gas channels (5) and the gas channels (9) have walls coated with a hydrophilic absorbent layer.
- 10 9. The polymer electrolyte fuel cell (1) of any of claims 1 to 8, characterized in that a means is provided for electronically determining the amount of water required for setting the optimum conductivity value of the
15 membrane (4).
- 20 10. The polymer electrolyte fuel cell (1) of claim 9, characterized in that a means is provided for measuring the moisture content of the membrane (4) by modulating the cell voltage with an alternating signal.
- 25 11. A fuel cell stack comprising a plurality of fuel cells (1) according to any of claims 1 to 10, characterized by a means for measuring the average moisture content of a plurality of membranes.
- 30 12. A method of cooling and humidifying a polymer electrolyte fuel cell (1) comprising an anode region (3), a cathode region (2), a polymer electrolyte membrane (4) disposed therebetween, a means for supplying air as oxidizing agent to the cathode region, gas channels (5) for distributing the air in
35 the cathode region, a means for supplying burnable gas to the anode region, and gas channels (9) for

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distributing the burnable gas in the anode region, as well as bipolar plates (10; 6) confining the cell on the anode side and/or on the cathode side and being corrugated at least in a partial region and having elevations (11; 7) and depressions (12, 8), with the depressions (8) on the cathode side constituting the gas channels (5) for distributing the air in the cathode region and/or the depressions (12) on the anode side constituting the gas channels (9) for distributing the burnable gas in the anode region, characterized in that, for simultaneously cooling the fuel cell and humidifying the polymer electrolyte membrane, a required amount of water in liquid form is introduced directly into the gas channels of the burnable gas, and in that the burnable gas or the oxidizing agent and the burnable gas are re-circulated.

13. The method of claim 12, characterized in that for simultaneously cooling the fuel cell and humidifying the polymer electrolyte membrane, a required amount of water is introduced directly into the gas channels (5) of the air and the gas channels of the burnable gas (9).

14. The method of claim 12, characterized in that the amount of water required for cooling and humidifying is added to the burnable gas or to the air and the burnable gas without substantial mixing therewith.

15. The method of any of claims 12 to 14, characterized in that the required amount of water is introduced through a plurality of thin lines opening into the gas channels (9) of the burnable

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gas or into the gas channels (5, 9) of air and burnable gas.

16. The method of claim 12,
5 characterized in that the required amount of water is added to the burnable gas or to the air and the burnable gas in mixed form using an aerosol.
17. The method of claim 16,
10 characterized in that the aerosol is produced with the aid of nozzles.
18. The method of claim 16,
15 characterized in that the aerosol is produced with the aid of ultrasonic atomizers (17).
19. The method of any of claims 12 to 18,
20 characterized in that the dwell period of the cooling water in the cell is increased by coating the walls of the gas channels (9) or of the gas channels (5) and the gas channels (9) with a hydrophilic absorbent layer.
20. The method of any of claims 12 to 19,
25 characterized in that the required amount of water is determined electronically by experimentally determining the optimum membrane moisture and regulation of the water addition as a function of the membrane moisture.
21. The method of claim 20,
30 characterized in that measuring of the moisture content of the membrane (4) is carried out by modulating the cell voltage with an alternating
35 signal.

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22. A method of cooling and humidifying a fuel cell stack comprising a plurality of fuel cells (1) according to any of claims 12 to 21, characterized in that the average moisture content of several membranes is measured for determining the amount of water required.

Structure of a fuel cell stack

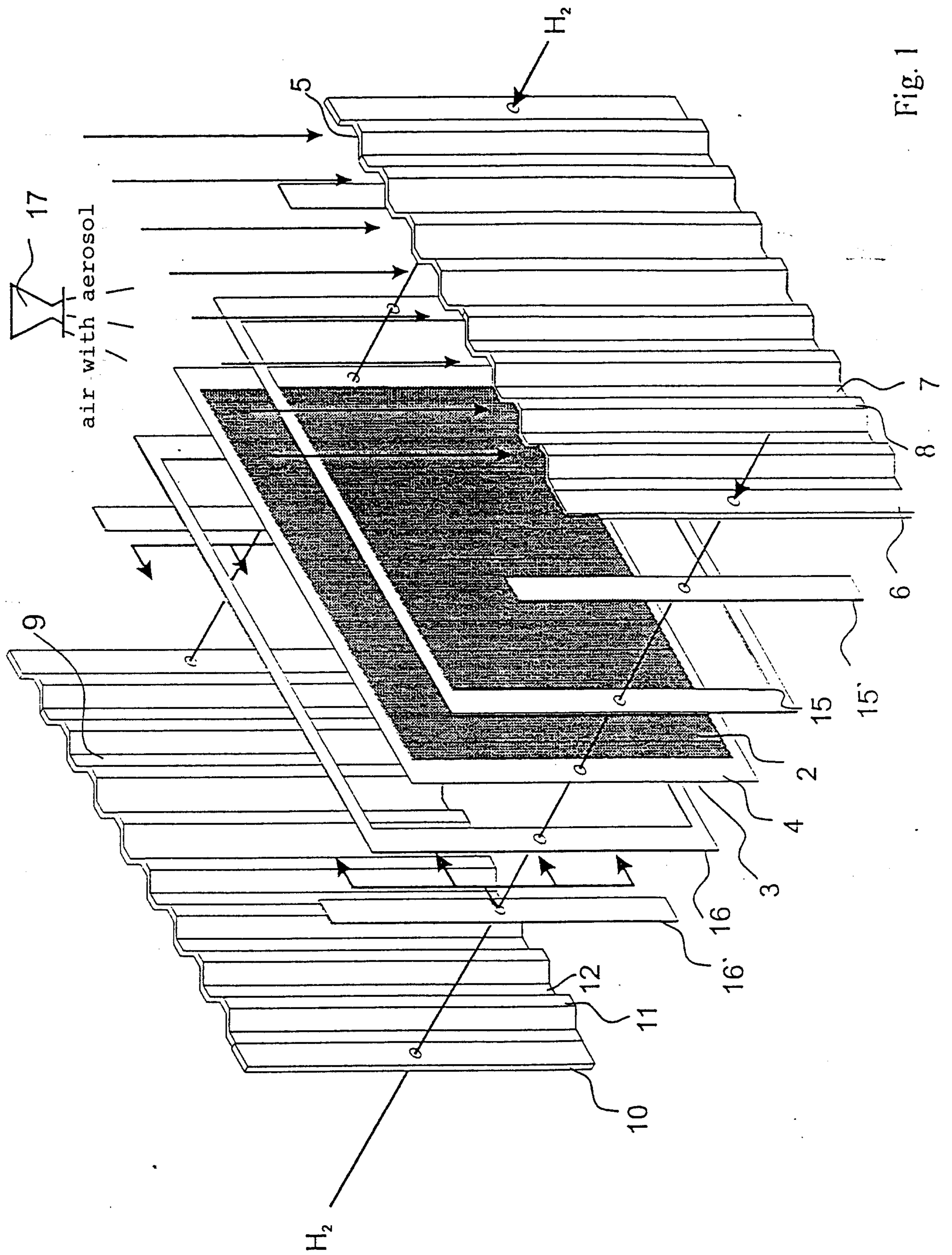


Fig. 1

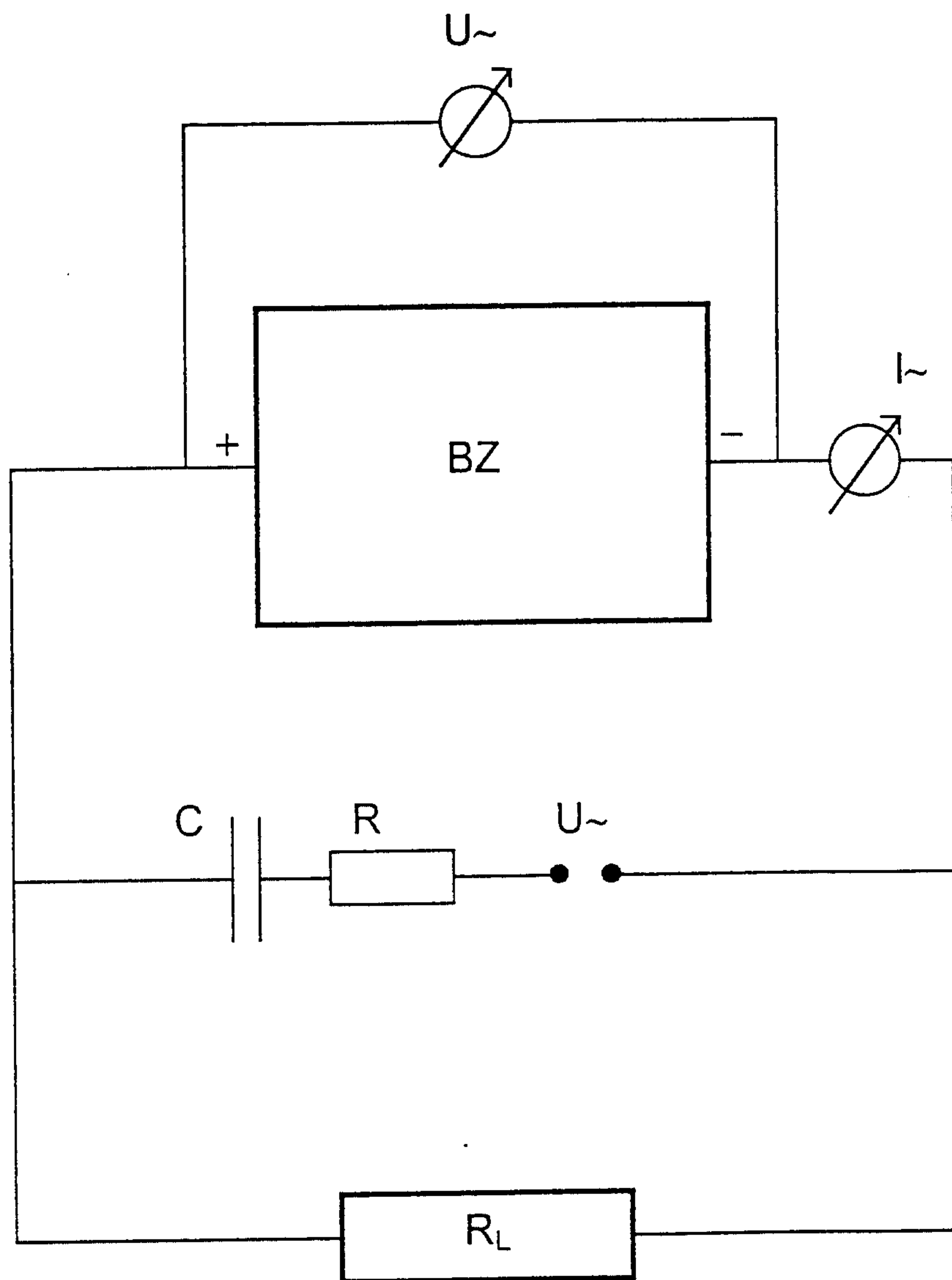


Fig. 2

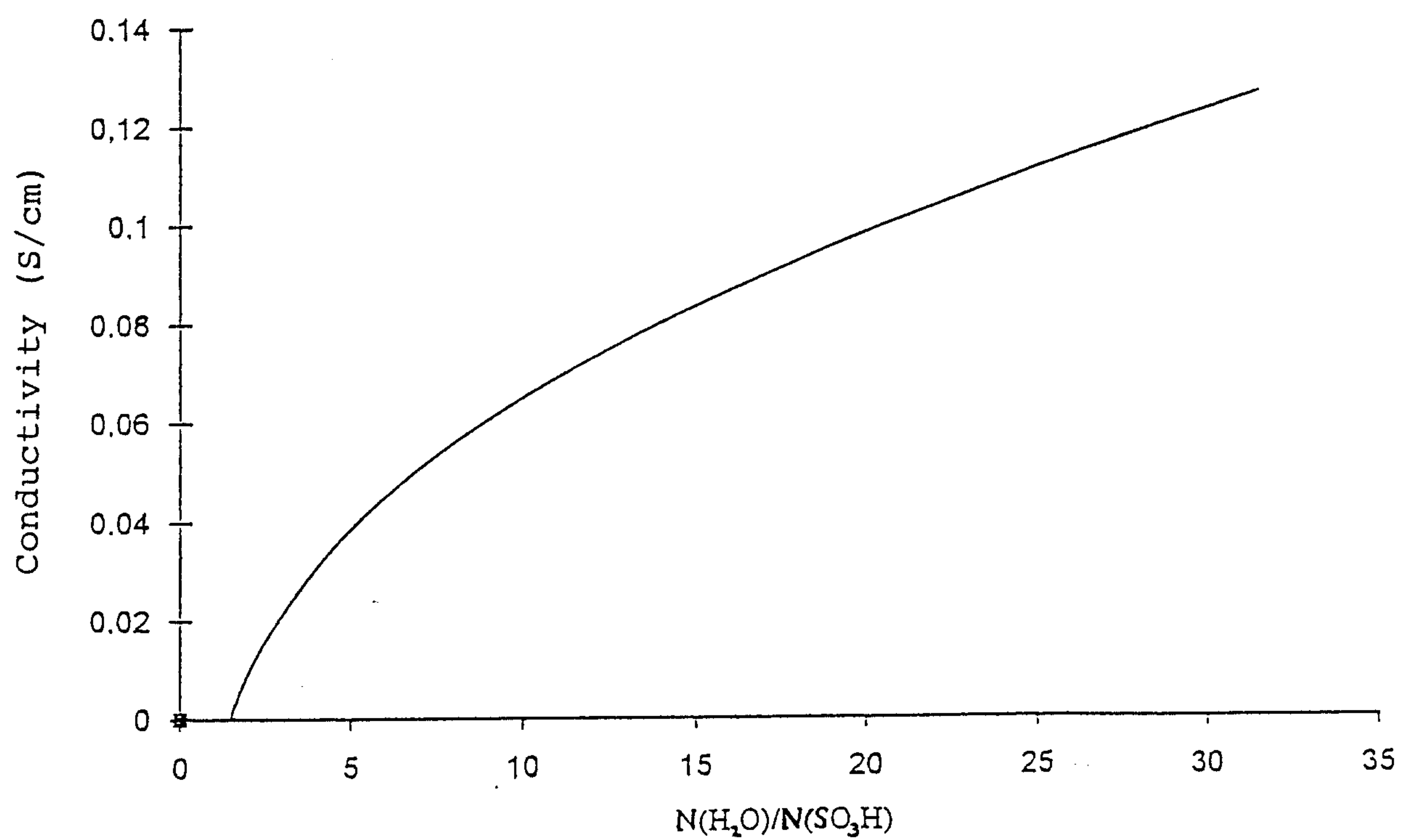


Fig. 3