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(54) Titre : PROCEDE D'ELECTROLYSE ET CELLULES D'ELECTROLYSE
(54) Title: ELECTROLYSIS METHOD AND ELECTROLYTIC CELLS

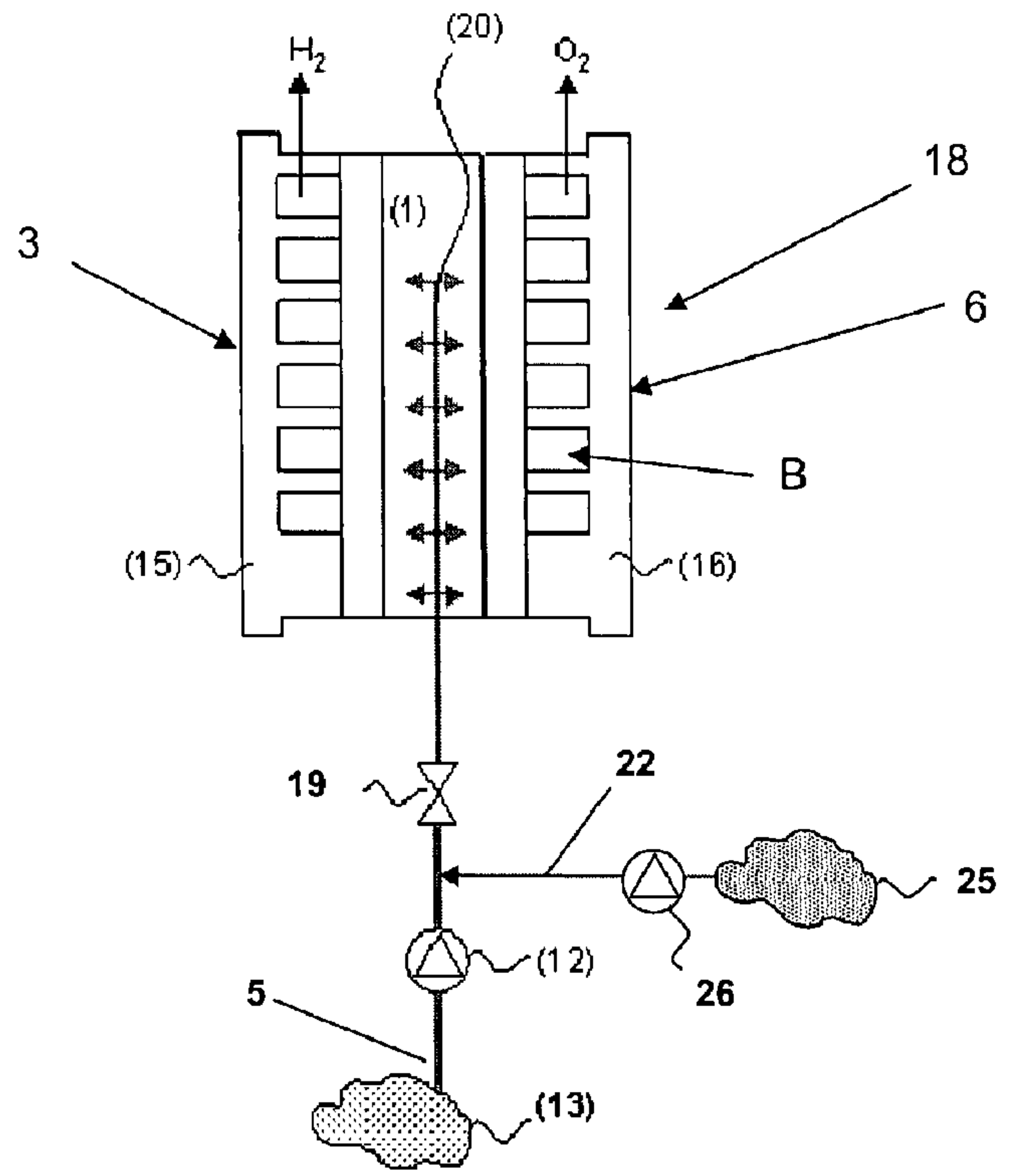

Fig. 4

(57) Abrégé/Abstract:
The invention relates to an electrolysis method for electrolytic cells having an electrode-membrane-electrode assembly comprising two porous electrodes (3, 6) having a porous membrane (1) located therebetween and filled with electrolyte (22) or having an ion

(57) Abrégé(suite)/Abstract(continued):
exchange membrane located therebetween, one or more liquids (22, 5) being led directly into the membrane (1) of the electrode-membrane-electrode assembly (3, 1, 6), wherein the one or more liquids (22, 5) are guided in a channel structure (20) arranged in the membrane (1). The invention further relates to an electrolytic cell (18) comprising porous electrodes (3, 6), between which a porous membrane (1) is arranged, wherein a liquid electrolyte (22) is fixed in the pores of electrodes (3, 6) and membrane (1), a product gas chamber (2, 15) adjoining the cathode (3), a further product gas chamber (7, 16) adjoining the anode (6), and an arrangement for feeding a liquid (22, 5) to the electrodes (3, 6), wherein a channel structure (20), in which distribution of the liquid (22, 5) is provided, is arranged in the membrane (1).

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[Fortsetzung auf der nächsten Seite]

(54) Title: ELECTROLYSIS METHOD AND ELECTROLYTIC CELLS

(54) Bezeichnung : ELEKTROLYSEVERFAHREN UND ELEKTROLYSEZELLEN

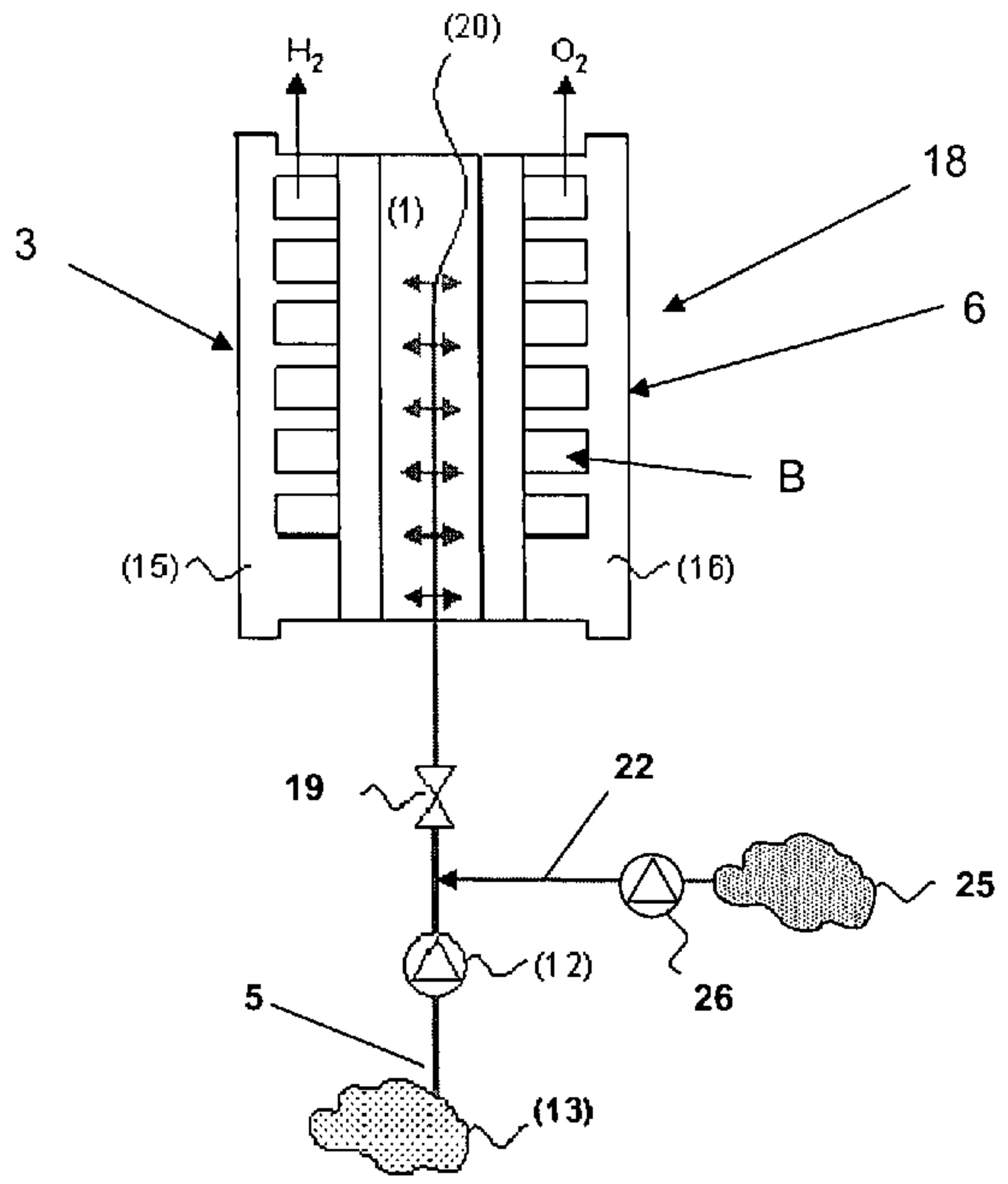

Fig. 4

(57) Abstract: The invention relates to an electrolysis method for electrolytic cells having an electrode-membrane-electrode assembly comprising two porous electrodes (3, 6) having a porous membrane (1) located therebetween and filled with electrolyte (22) or having an ion exchange membrane located therebetween, one or more liquids (22, 5) being led directly into the membrane (1) of the electrode-membrane-electrode assembly (3, 1, 6), wherein the one or more liquids (22, 5) are guided in a channel structure (20) arranged in the membrane (1). The invention further relates to an electrolytic cell (18) comprising porous electrodes (3, 6), between which a porous membrane (1) is arranged, wherein a liquid electrolyte (22) is fixed in the pores of electrodes (3, 6) and membrane (1), a product gas chamber (2, 15) adjoining the cathode (3), a further product gas chamber (7, 16) adjoining the anode (6), and an arrangement for feeding a liquid (22, 5) to the electrodes (3, 6), wherein a channel structure (20), in which distribution of the liquid (22, 5) is provided, is arranged in the membrane (1).

(57) Zusammenfassung:

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Veröffentlicht:

— *mit internationalem Recherchenbericht (Artikel 21 Absatz 3)*

— *vor Ablauf der für Änderungen der Ansprüche geltenden Frist; Veröffentlichung wird wiederholt, falls Änderungen eingehen (Regel 48 Absatz 2 Buchstabe h)*

Die Erfindung betrifft ein Elektrolyseverfahren für Elektrolysezellen mit einer Elektroden-Membran-Elektroden-Anordnung umfassend zwei poröse Elektroden (3, 6) mit dazwischen liegender poröser Membran (1) gefüllt mit Elektrolyt (22) oder mit dazwischen liegender lonenaustauschermembran, wobei ein oder mehrere Flüssigkeiten (22, 5) direkt in die Membran (1) der Elektroden-Membran-Elektroden- Anordnung (3, 1, 6) geführt werden, wobei die ein oder mehreren Flüssigkeiten (22, 5) in einer in der Membran (1) ausgeführten Kanalstruktur (20) geführt werden. Die Erfindung betrifft ferner eine Elektrolysezelle (18) umfassend poröse Elektroden(3, 6), zwischen denen eine poröse Membran (1) angeordnet ist, wobei in den Poren von Elektroden (3, 6) und Membran (1) ein flüssiger Elektrolyt (22) fixiert ist, eine an die Kathode (3) angrenzende Produktgaskammer (2, 15), eine an die Anode (6) angrenzende weitere Produktgaskammer (7, 16) und eine Anordnung zur Zuführung einer Flüssigkeit (22, 5) zu den Elektroden (3, 6), wobei in der Membran (1) eine Kanalstruktur (20) ausgeführt ist, in welcher eine Verteilung der Flüssigkeit (22, 5) vorgesehen ist.

Electrolysis method and electrolytic cells

The invention relates to an electrolysis process according to the features of the preamble of Patent Claim 1 and to an electrolytic cell according to the features of the preamble of Patent Claim 2.

US 5,916,505 A has disclosed a fuel cell arrangement in which water or electrolyte is pumped through the cell. US 4,330,378 A has disclosed an electrolytic cell in which water or electrolyte is pumped through the electrode.

A known device for the electrolysis of water with a fixed, alkaline electrolyte is known, for example, from US 2009/008261 A1 or from R. J. Davenport et al., Space water electrolysis: space station through advanced missions, Journal of Power Sources, 36 1991, 235-250. The key part of this device is the electrolytic cell. It comprises the following components:

- electrodes between which a porous membrane (diaphragm) is arranged, an electrolyte solution being fixed in the pores of the electrodes and the membrane by capillary forces;
- a hydrogen product gas chamber H_2 adjacent to the cathode;
- an oxygen product gas chamber O_2 adjacent to the anode;
- a chamber for the feedstock, namely water H_2O, which is separated from the H_2 product gas chamber by a membrane.

For the further construction and function of an electrolytic cell, reference is made to the description of DE 195 35 212 C2.

DE 195 35 212 C2 has also disclosed an alkaline electrolysis process using an immobilized potassium hydroxide solution, in which the water is transferred into an electrode-membrane-electrode system by diffusion via a hydrophobic porous membrane adjacent to the cathode gas chamber. This process has the disadvantage that, when the electrolytic cell is switched off, the diffusion process can only be stopped by flushing out the entire water reservoir in the stack with an inert gas. Since the diffusion process is controlled by way of the temperature of the water, there is quite a time delay in the control.

The invention is based on the objective of providing an alkaline electrolysis process for hydrogen electrolysers by which the disadvantages of the prior art are overcome. A further objective is to implement an electrolytic cell of a simple construction, with which it is possible to switch the electrolysis process on and off quickly without diffusion processes leading to a dilution of the immobile electrolyte solution.

These objectives are achieved by an electrolysis process according to the features of the current Claim 1, and an electrolytic cell according to Claim 2. Advantageous refinements are the subject of subclaims.

In accordance with the process according to the invention for alkaline electrolytic cells with an electrode-membrane-electrode arrangement comprising two porous electrodes with a membrane lying in between, one or several liquids is/are introduced directly into the electrode-membrane-electrode arrangement. According to the invention, the one or several liquids is/are conveyed directly in a structure of channels implemented in the membrane.

Hereinafter, an electrode-membrane-electrode arrangement or EME system is understood as meaning an arrangement of two electrodes and a membrane arranged between the electrodes, a liquid electrolyte being fixed in the membrane. The membrane, in this case, can be a porous membrane or an ion-exchange membrane.

The one or several liquids is/are introduced directly into the structure of channels of the membrane. This ensures that the one or several liquids is/are distributed uniformly in the membrane or electrode.

The electrolysis causes liquid, for example water, in the structure of channels of the membrane to be used up. The respective liquid, for example water, is then replenished accordingly in the structure of channels. It is to be mentioned that, according to the invention, the liquid is not passed through the structure of channels but is introduced or conducted into the structure of channels, where it may be used up.

The liquid is consequently an immobilized liquid, which when used up accordingly, for example when water is converted into hydrogen and oxygen in the electrolysis, is topped up by replenishing the corresponding liquid in the structure of channels.

According to the invention, the electrolytic cell comprises porous electrodes between which a porous membrane is arranged, a liquid electrolyte being fixed in the pores of the electrodes and of the membrane, a product gas chamber adjacent to the cathode, a further product gas chamber adjacent to the anode and an arrangement for feeding water to the electrodes. According to the invention, a structure of channels in which a distribution of the liquid, expediently water or electrolyte, is provided is implemented in the membrane.

The structure of channels may expediently be a structure of microchannels and/or nanochannels. The membrane is expediently a proton-conducting membrane or an ion-exchange membrane. In one particular embodiment, the membrane may also be implemented in a multi-layered form.

A concentrated, aqueous solution of high electrical conductivity is expediently used as an electrolyte. Preferred exemplary embodiments for this are: acids, bases and inorganic salt solutions of high electrical conductivity, such as for example: potassium hydroxide or other alkaline and alkaline-earth hydroxides in concentrations of approximately 5 to 12 mol/litre; sulphuric acid of approximately 2 to 5 mol/litre; phosphoric acid, etc.

One advantage of the invention is that, in the case of alkaline electrolytic cells with an immobilized electrolyte, water and/or electrolyte is fed to the EME system directly, so that no inert gas flushing is necessary when it is switched off and, in addition, the delayed response of the system to changes in load is reduced. Furthermore, in the case of polymer electrolyte cells, water is likewise fed directly to the EME system, so that the hydrogen and oxygen gas emerging leaves the cell as gas and no further gas/liquid separation is needed.

The invention and further advantages of the invention are explained in more detail by means of the drawings, in which:

Fig. 1 schematically shows an electrolytic cell according to the invention for the alkaline electrolysis,

Fig. 2 schematically shows an exemplaryarrangement of microchannels in a diaphragm for the alkaline electrolysis,

Fig. 3 schematically shows a polymer-membrane electrolytic cell according to the prior art,

Fig. 4 schematically shows a polymer-membrane electrolytic cell according to the invention,

Fig. 5 schematically shows an electrolytic cell arrangement according to the invention with three electrolytic cells by way of example.

Alkaline electrolysis

Fig. 1 schematically shows an alkaline electrolytic cell according to the invention, in which the electrolyte 22, for example a potassium hydroxide solution, is immobilized by the porosity of the electrodes 3, 6 and of the membrane 1 (diaphragm).

If a voltage is then applied to the current lines 8 by means of a power source 9, water molecules from the aqueous electrolyte 22 are split into their components hydrogen, H_2, and oxygen, O_2. At the cathode 3 H_2 is produced, which flows as a gas into the H_2-gas chamber 2. At the anode 6 gaseous oxygen is produced, which flows into the O_2-gas chamber 7. As this process continues, the electrolyte 22 becomes more and more concentrated unless water is replenished correspondingly. In the electrolytic cell 18 according to the invention, it is provided that water to be split is then fed to the immobilized electrolyte 22.

According to the invention, the feeding of the water to be split takes place by way of a structure of channels 20, which is implemented in the membrane 1 and/or in at least one electrode 3, 6. Through this structure of channels 20, the water to be split is introduced into the EME system 3, 1, 6 over a large surface area and is distributed in the EME system 3, 1, 6 over a large surface area. Numerous further possibilities for feeding water directly into the EME system 3, 1, 6 are conceivable. Figure 2 shows by way of example a membrane 1 with parallel channels 20 and a surrounding annular channel.

The electrolytic cell, namely the structure of channels 20 of the membrane 1 of the electrolytic cell, is connected to a water reservoir 13 by way of a control valve 19 and a first pump 12. For specific applications, the capillary forces in the EME system 3, 1, 6 may also be sufficient to suck the water to be split from a water reservoir 13 without a pump 12.

In the case of prolonged operation of the alkaline electrolytic cell or of instances of improper operation, liquid electrolyte may be discharged from the EME system 3, 1, 6 by way of the product gas chambers, as a result of which the electrolyte concentration in the electrolytic cell is diluted and the performance of the cell drops. By way of the electrolyte reservoir 25 and the pump 26, electrolyte can expediently also be fed to a cell by way of the structure of channels.

By way of the control valve 19 and a second pump 26, the electrolytic cell 18, namely the structure of channels 20 of the membrane 1 of the electrolytic cell, is connected to an electrolyte reservoir 25. It is consequently possible for the electrolyte 22 to be introduced into the membrane 1 and distributed on the membrane surface directly by way of the structure of channels 20.

The structure of channels 20 is constructed in such a way that the water is thus uniformly distributed over the surface area of the membrane 1 of the EME system 3, 1, 6 and mixed with the electrolyte 22, and negligible concentration gradients form over the cross section of the surface area.

If the electrolysis process is then stopped and the power supply to the electrolytic cell is interrupted, no water is replenished either if the control valve 17 is closed or if the pump 12 is switched off. In this state, an alkaline electrolytic cell can go for any length of time without flushing of the electrolytic cell being necessary.

Direct supply of water to be split or of the electrolyte to the membrane 1 of the EME system 3, 1, 6 offers the following advantages here:

- the electrolytic cell involves simple equipment, because the water diffusion layer is omitted and there is no need for flushing with inert gas after switching off,
- higher current densities,
- faster control response,
- possible replenishing discharged electrolyte.

Polymer-membrane electrolysis

The same arrangement with a separate supply of water to be split to the membrane 1 of the EME system 3, 1, 6 can also be used for an electrolytic cell with a polymer electrolyte. Figure 3 shows the schematic construction of a membrane-electrolyte electrolytic cell according to the prior art. In this example, a proton-conducting membrane is used as a membrane 1. However, an ion-exchange membrane or anion-exchange membrane in which hydroxyl ions undertake the conduction of the current may also be used as a membrane 1.

According to the generally known prior art, on the anode side 6 water is pumped through a structure of channels (not shown) of the bipolar plate B. A portion of the water is split. The oxygen will herein bubble into the water flow and be carried thereby out of the electrolytic cell 18. On the cathode side 3, the hydrogen enters the inter-mediate spaces of the bipolar plate B. For better removal of the hydrogen, water circulating in a cycle on the cathode side 3 is also often used to discharge the hydrogen. To obtain gaseous H_2 and O_2, this necessitates a phase separation, which requires additional equipment.

Fig. 4 shows a membrane-electrolyte electrolytic cell according to the invention, in which the water to be split is provided to be fed to the anode 6 over a large surface area by way of a system of microchannels into the membrane 1. The water is introduced into the membrane 1 by way of a system of microchannels 20. Gaseous H_2 and O_2 are produced at the cathode 3 and at the anode 6 and escape by way of channels (not shown) in the bipolar plates 15 and 16. Direct water supply offers the following advantages here:

- Omitting the gas/liquid separators, including pipework and pumps. In particular in the case of pressure electrolytic cells, this leads to a considerable reduction in weight. Furthermore, no phase separation is required, which is desirable in particular for applications under weightlessness.
- Lower expenditure of energy as a result of omitting the pumps.

Fig. 5 schematically shows according to the invention an exemplary electrolytic cell arrangement with three electrolytic cells 18 by way of example. In the case of immobilized alkaline electrolytic cells or polymer-membrane electrolytic cells, each electrolytic cell 18 consists of the EME system 3, 1, 6 and the H_2 und O_2 gas chambers with the respective framework structure for removing the H_2 and O_2.

The water from the water reservoir 13 is pressurized centrally by a pump 12 and then fed individually, by way of control valves 19, to each electrolytic cell 18, namely to the structure of microchannels in the membrane (not shown). However, it is also possible for each electrolytic cell 18 to be connected to a respective pump, which in turn is connected to the water reservoir 13. In this implementation, the control valves 19 for each electrolytic cell 18 could be omitted.

In the electrolytic cell arrangement that is shown in Fig. 5, each electrolytic cell is also supplied with an electrolyte from an electrolyte reservoir 26. The electrolyte from the electrolyte reservoir 25 is then pressurized centrally by a pump 26 and fed individually, by way of control valves 19, to each electrolytic cell 18, namely to the structure of microchannels in the membrane (not shown). However, it is also possible for each electrolytic cell 18 to be connected to a respective pump, which in turn is connected to the electrolyte reservoir 25.

The cooling of an electrolytic cell 18 is effected by way of cooling cells 23, which are arranged between the individual electrolytic cells 18 and through which cooling water flows in parallel. The cooling water cycle 23a, which is isolated from the electrochemical process, consequently serves exclusively for cooling. In the case of alkaline electrolytic cells, the diffusion membrane, as described in DE 195 35 212 C2, is omitted. In the case of the polymer-membrane electrolytic cell, the O_2-and/or H_2-water cycle, and consequently the phase separation, is omitted.

Material tolerances and different thermal behaviour of individual cells may cause differences in the operating behaviour and the ageing of individual cells. In an extreme case, it was possible in the prior art for a single cell to fail and thereby render the entire stack of cells inoperative.

The following further advantages are provided by the water being supplied directly:

a) By way of an individual pump 12 with individual activation for each cell or by means of a central pump 12 for all the electrolytic cells 18 and individually activatable control valves 19 for each electrolytic cell 18, the water supply and any necessary electrolyte refill for an electrolytic cell 18 can take place separately from the other cells 18. The possibility of an individual water supply is the prerequisite for running each electrolytic cell 18 at its optimum operating point.

b) By stopping the water supply and bridging the power supply, together with closing the H_2 and O_2 channels of an electrolytic cell 18, it would also be possible to isolate defective cells from the stack of several electrolytic cells 18.

Patent claims

1. An electrolysis process for electrolytic cells with an electrode-membrane-electrode arrangement comprising two porous electrodes (3, 6) with a porous membrane (1) filled with electrolyte (22) lying in between or with an ion-exchange membrane lying in between,
   characterized in that
   one or several liquids (22, 5) is/are introduced directly into the membrane (1) of the electrode-membrane-electrode arrangement (3, 1, 6), the one or several liquids (22, 5) being introduced into a structure of channels (20) implemented in the membrane (1).

2. The electrolytic cell (18) comprising
   porous electrodes (3, 6) between which a porous membrane (1) is arranged, a liquid electrolyte (22) being fixed in the pores of the electrodes (3, 6) and of the membrane (1),
   a product gas chamber (2, 15) adjacent to the cathode (3),
   a further product gas chamber (7, 16) adjacent to the anode (6) and
   an arrangement for feeding a liquid (22, 5) to the electrodes (3, 6),
   characterized in that
   a structure of channels (20) in which a distribution of the liquid (22, 5) is provided is implemented in the membrane (1).

3. The electrolytic cell according to Claim 2,
   characterized in that
   the liquid is water (5) to be split or an electrolyte (22).

4. The electrolytic cell according to either of preceding Claims 2 and 3,
   characterized in that
   the membrane (1) is a proton-conducting membrane or an anion-exchange membrane.

5. The electrolytic cell according to Claim 4,
characterized in that
the membrane (1) is implemented in one or several layers.

6. The electrolytic cell according to one of preceding Claims 2 to 5,
characterized in that
the structure of channels (20) is connected to a water reservoir (13) by way of a first pump (12).

7. The electrolytic cell according to Claim 6,
characterized in that
the structure of channels (20) is connected to an electrolyte reservoir (25) by way of a second pump (26).

8. The electrolytic cell according to one of preceding Claims 2 to 7,
characterized in that
the structure of channels (20) is a structure of microchannels and/or nanochannels.

9. An arrangement comprising one or several electrolytic cells connected in series or parallel according to one of the preceding Claims 2 to 8.

10. The arrangement according to Claim 9,
characterized in that
the structure of channels (20) of each electrolytic cell (18) is connected to a first pump (12) by way of a respective control valve (19) and the first pump (12) is connected to a water reservoir (13).

11. The arrangement according to Claim 9,
characterized in that
the structure of channels (20) of each electrolytic cell (18) is connected to a water reservoir (13) by way of a respective control valve (19) and by a respective pump (12).

12. The arrangement according to one of Claims 10 and 11,
characterized in that
each control valve (19) is connected to an electrolyte reservoir (25) by way of a further pump (26).

13. The arrangement according to one of preceding Claims 8 to 12,
characterized in that
cooling cells (23) through which a cooling medium flows are arranged between the electrolytic cells (18).

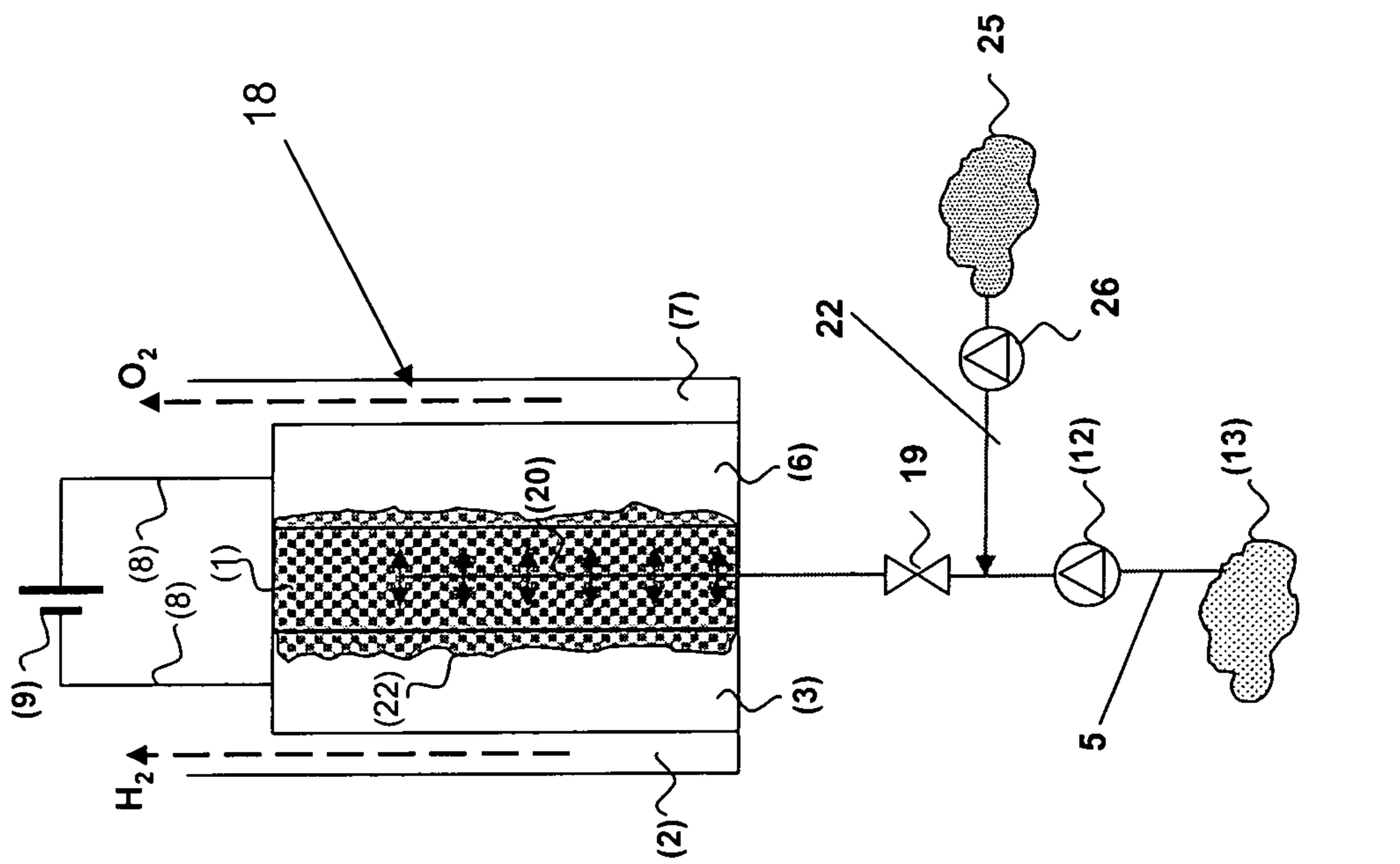

Fig. 1

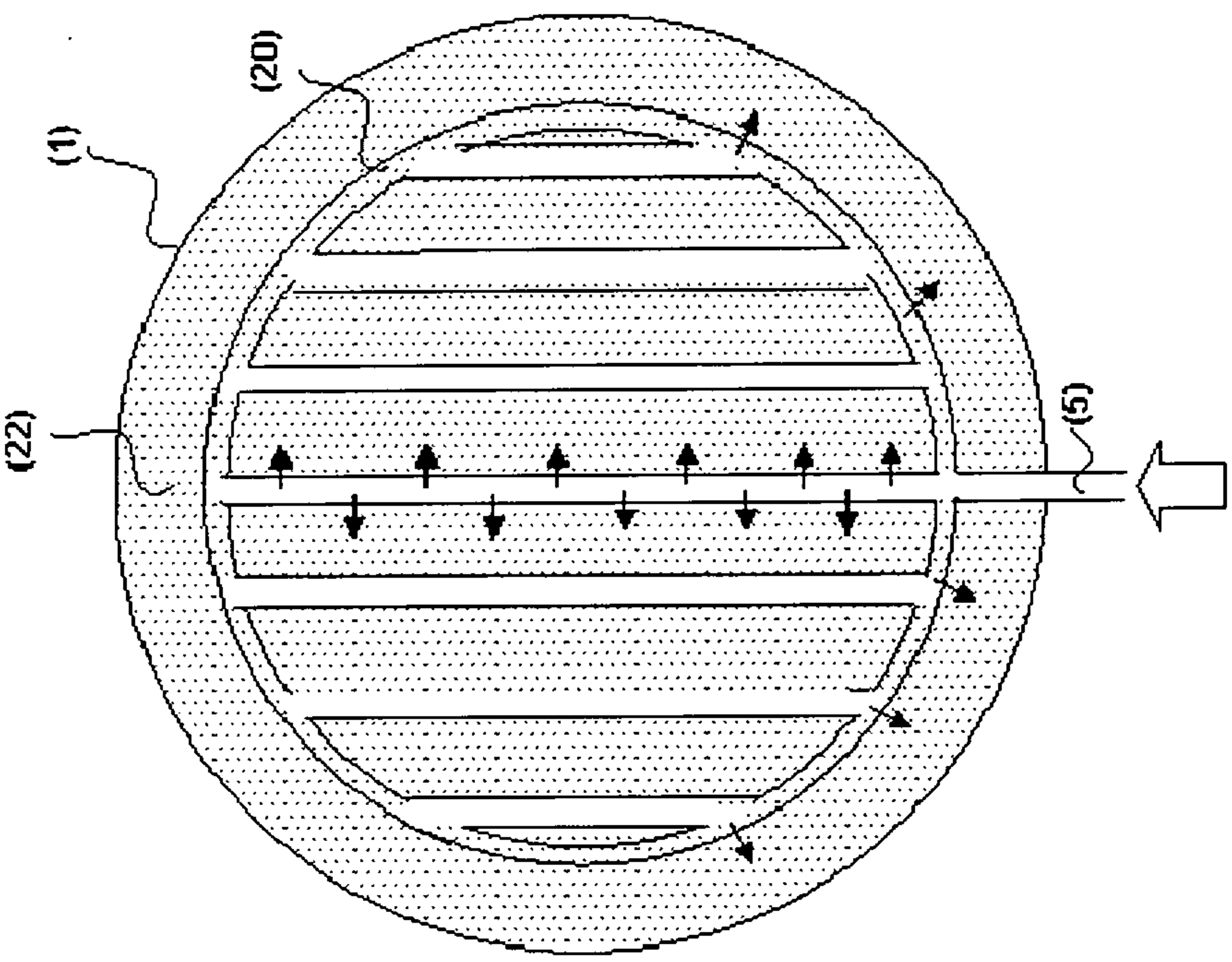

Fig. 2

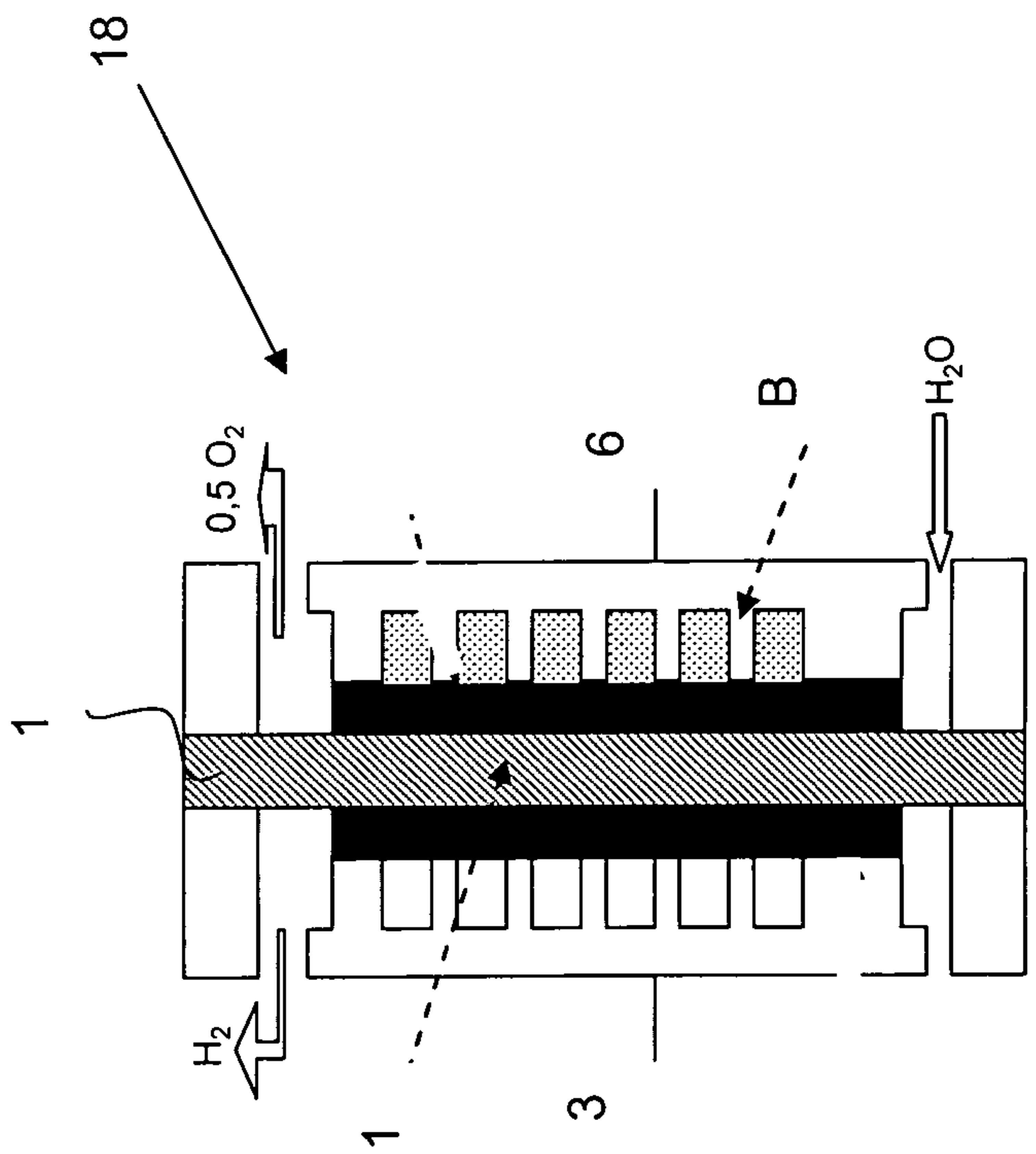

Fig. 3

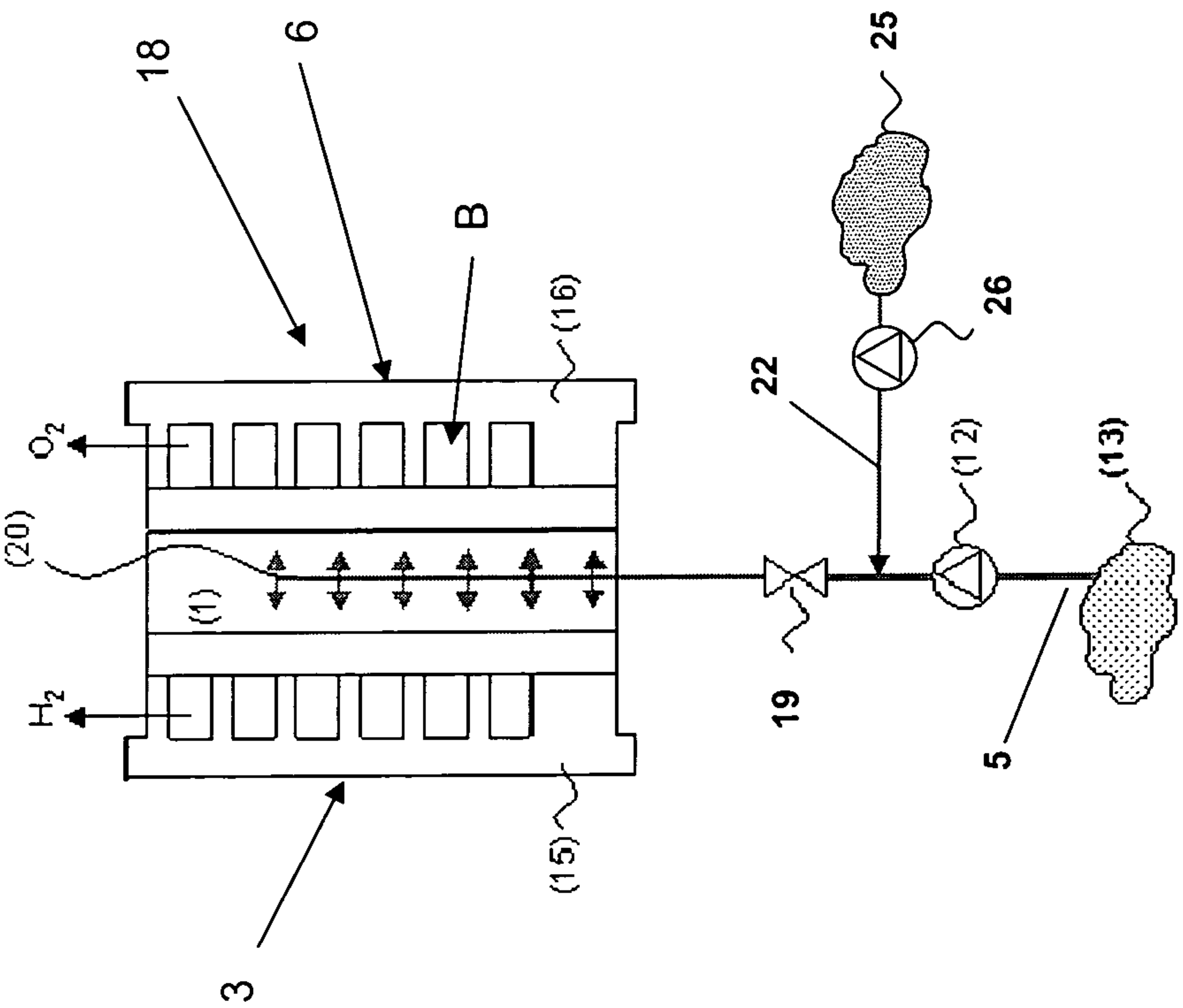

Fig. 4

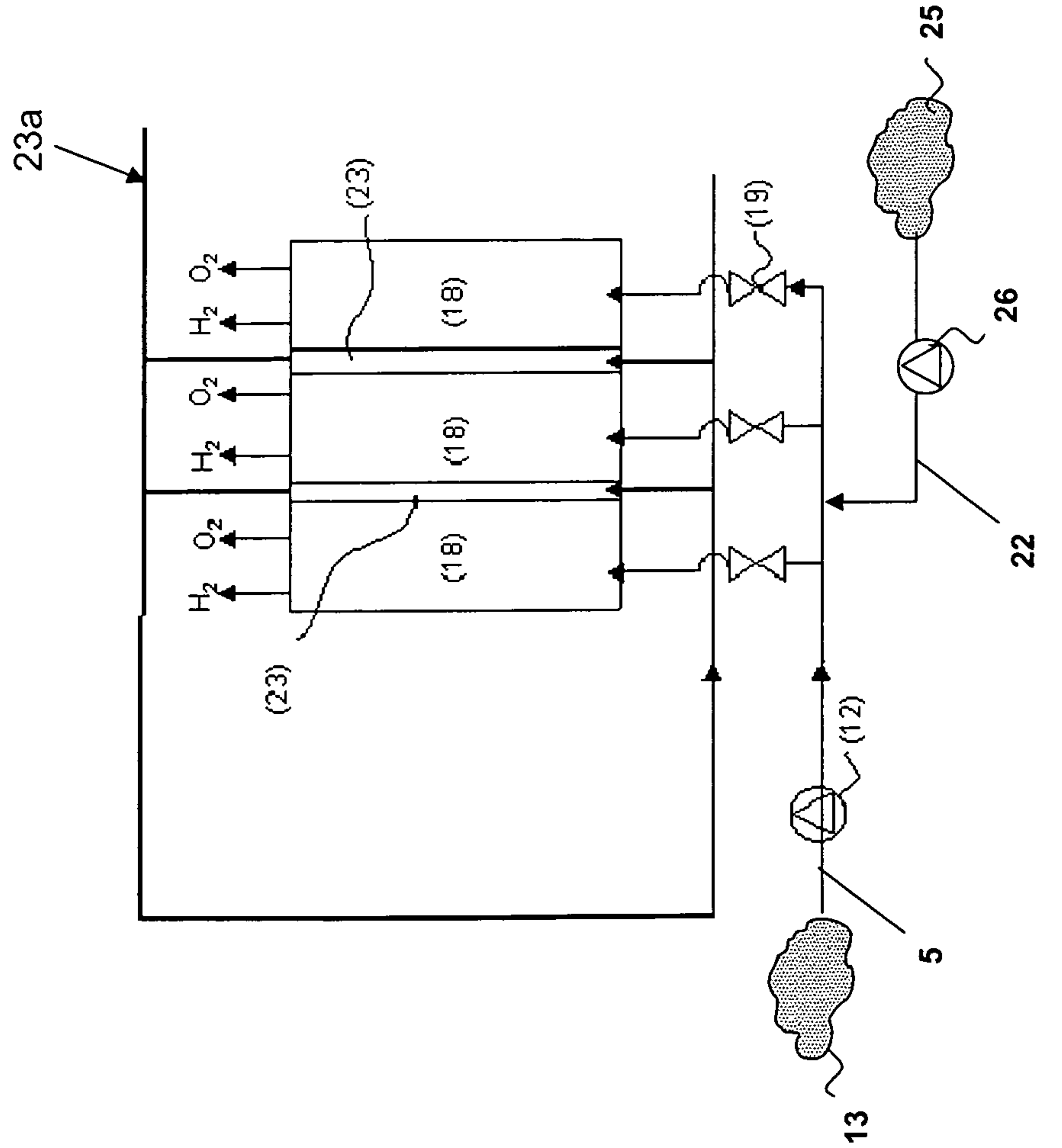

Fig. 5

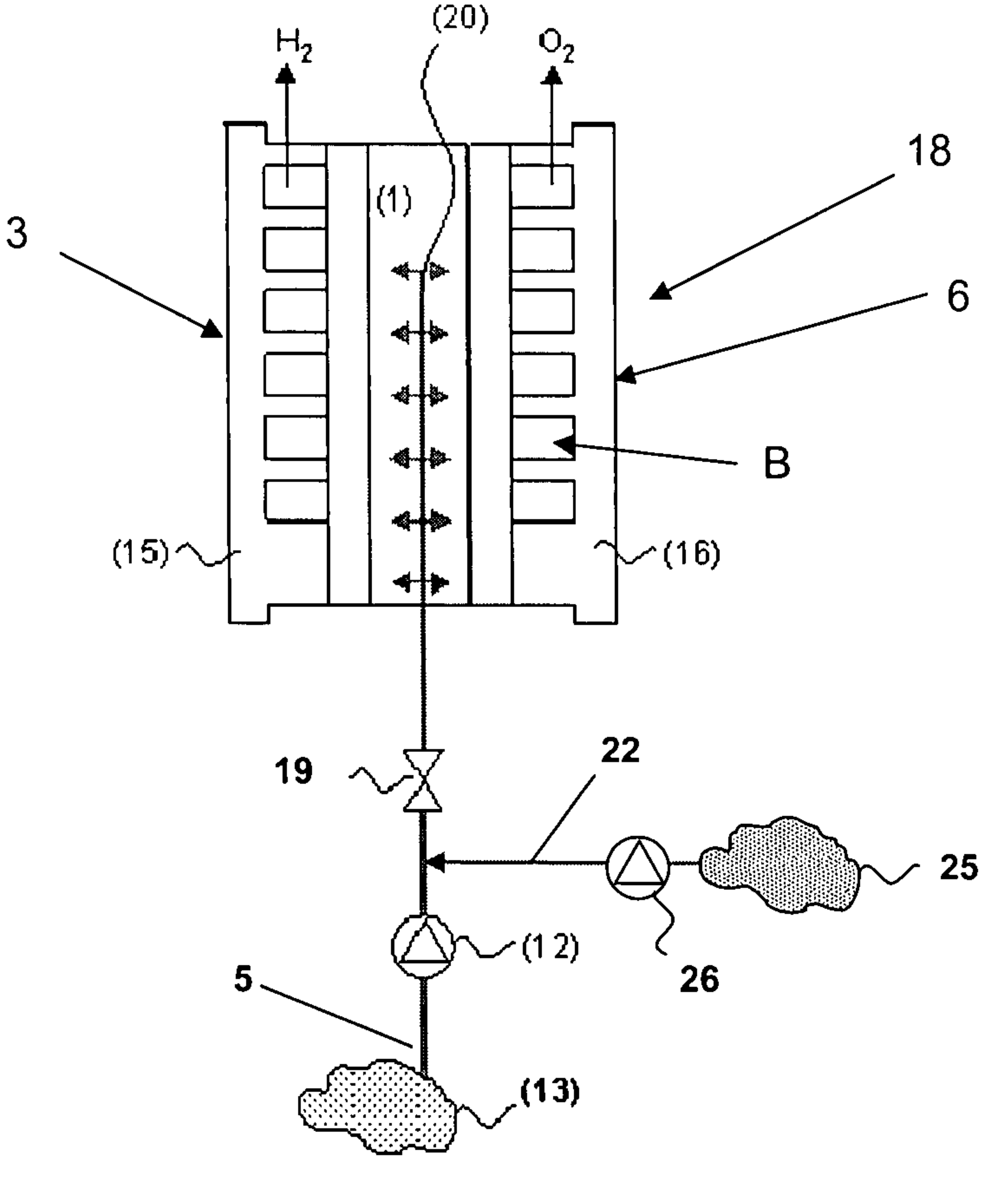

Fig. 4