



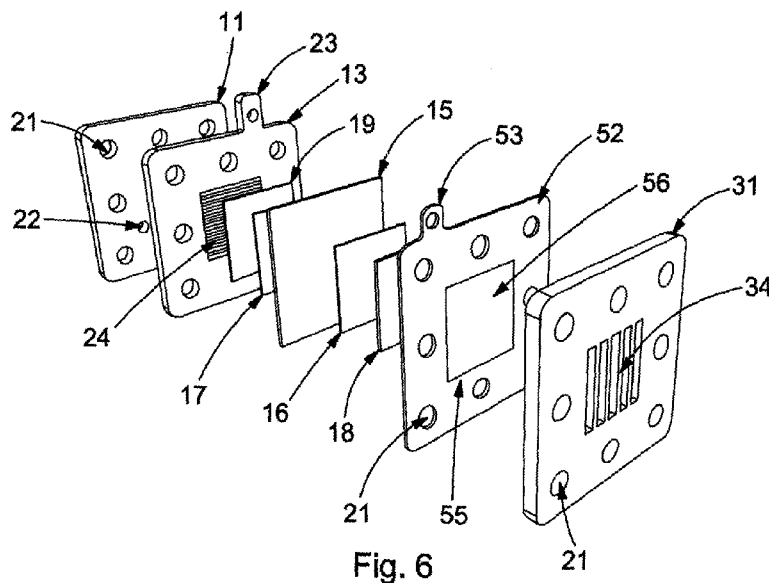
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(54) Title: PROTON EXCHANGE MEMBRANE FUEL CELL



(57) Abstract: A proton exchange membrane fuel cell comprising an electrically conductive anodic current collector plate, separated from an electrically conductive cathodic current collector plate by a proton exchange membrane, wherein at least a portion of one or both of the anodic and cathodic current collector plates is provided with a plurality of molecular pores.

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Proton Exchange Membrane Fuel Cell

The present invention relates to a proton exchange membrane (PEM) fuel cell, and to improvements made in the performance of such fuel cells.

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Fuel cells generally are well known in the field of power-generation technology. Their general principle of operation involves supplying a fuel gas to an anode and an oxidant to a cathode. The fuel and oxidant are separated by an "electrolyte", which prevents direct mixing of the two and, importantly, also forces electrons produced at the anode to travel *via* an external circuit to the cathode, whilst allowing the movement of certain ions between the anode and cathode to facilitate the required electrochemical reactions. Fuels suitable for use in such fuel cells include small, organic molecules such as hydrogen, methane and methanol, with oxygen being used as the oxidant. In particular, for an environmental perspective, the combination of hydrogen and oxygen is favoured, since the only resulting waste product is water.

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Of particular interest with regards to the present invention is a PEM fuel cell, in which an electrolytic PEM is used to separate a planar anode and a planar cathode. A fuel gas, such as hydrogen, is oxidised at the anode to release protons (H^+) and electrons (e^-), typically through the use of a catalyst in relation to the anode. Released protons travel through the membrane to the cathode, again provided with a catalyst in relation thereto, where they meet conducted electrons and react with the oxidant, typically oxygen, to produce water (H_2O) as a by-product.

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Where the PEM fuel cell is sealed to its external environment and both the fuel and the oxidant are supplied from suitable storage devices, it is known as a "conventional" non-air-breathing PEM fuel cell. Where the cathode compartment of the PEM fuel cell is open to the ambient environment, such that the oxidant is supplied by natural convection, the cell is known as an "air-breathing" PEM fuel cell. Such air-breathing PEM fuel cells are of increasing interest in the field of portable electronic devices, where they are in competition with conventional power generation means, such as lithium-ion batteries, primarily because of their simplicity; the only ancillary component needed to successfully operate such a fuel cell is a hydrogen storage device.

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In a typical PEM fuel cell, the combination of the electrolytic membrane interleaved between the electrodes is known as a membrane electrode assembly (MEA). Surrounding, on either side of, the MEA is a pair of gas diffusion layers and outwardly of the gas diffusion layers, a pair of current collector plates (also known as “flow field plates”) is provided. Further outwardly of the current collector plates is a pair of end plates, which define the outermost major surfaces of the fuel cell, although one or both of these may be omitted if the corresponding current collector plate is sufficiently rigid.

The gas diffusion layers are typically provided as bi-layers of a carbon-fibre-based material (having an overall thickness in the region of 200-400 μm), having a macroporous substrate (with pores sizes of the order of tens of microns) and a microporous overlayer (with pore sizes of the order of one micron). The current collector plates are typically provided as graphite-based plates (in the region of 3 mm thick) to ensure adequate electrical conductivity of electrons around the cell and thermal conductivity of heat generated away from the reaction sites in the cell, although other conductive materials, such as stainless steel or aluminium, alternatively may be used.

Typically, the areas of each of the planar anode, the planar cathode and the gas diffusion layers are reduced as compared to the overall surface area of each of the current collector plates. Channels (known as “flow field channels”), which may be of a serpentine, parallel or interdigitated flow configuration, are typically provided in a centralised region of the current collector plates, so as to be in register with the MEA and pair of gas diffusion layers once all these components are appropriately layered and formed into a cell, to allow the supply of fuel gas to the surface of the anode, *via* the intermediate gas diffusion layer, and to allow the supply of oxidant to the surface of the cathode, *via* the other intermediate gas diffusion layer and to allow water by-product to drain away from the cathode.

Once constructed, the electrochemical integrity of the fuel cell is typically provided by incorporating a sealing, fluid-impervious, peripheral gasket material between the end plates, which are held together, for example by bolts or other such suitable fastening means, using typically at least 2 Nm of torque, thereby ensuring intimate contact between the layers within. As such, the end plates must be sufficiently rigid to

withstand such compression, and thus made from, for example aluminium or stainless steel.

5 Figures 1 and 2 of the accompanying drawings show a prior art, non-air-breathing
“conventional” PEM fuel cell 10 comprising two end plates 11, between which is
provided an electrically conductive cathodic current collector plate 12 and an
electrically conductive anodic current collector plate 13, separated by a sealing gasket
material 14 provided between and around the peripheries of the collector plates.
10 Located inwardly of the periphery of each end plate 11 is a series of bolt holes 21, of
which eight are provided per plate, through which bolts (not shown) are locatable to
secure together the components of the cell and to provide the necessary degree of
compression. Also provided in each end plate 11 are with two inlet/outlet (I/O) holes
22, which facilitate transport of fuel and oxidant into, and water and heat out of, cell 10.
Plates 11 are identical such that when stacked in register, bolt holes 21 and I/O holes
15 22 are in alignment.

Figure 2 provides more detail regarding the interior of fuel cell 10, particularly of the
additional components of the cell found between each of the cathodic and anodic
current collector plates 12, 13. These additional components include an electrolytic
20 PEM 15 interleaved between a cathodic catalyst layer 16 and an anodic catalyst layer
17 (references to “cathodic” and “anodic” through this specification indicating the
relevant compartment of the cell to which like charged components reside), all of which
are interleaved between a cathodic gas diffusion layer 18 and an anodic gas diffusion
layer 19.

25 Each of cathodic current collector plate 12 and anodic collector plate 13 is also
provided with a series of bolt holes 21, of which eight are again provided, along with
two I/O holes 22, and furthermore each has a current collection tab 23. In a central
region on the innermost surface (as shown in Figure 2) of each plate 12, 13, flow field
30 channels 24 are provided. The surface areas of each of the PEM 15, catalyst layers
16, 17 and gas diffusion layers 18, 19 are reduced as compared to the surface areas of
each of the current collector plates 12, 13, so as to substantially overlap only with flow
field channels 24 therein.

In use, fuel such as hydrogen, methane and methanol from a dischargeable source and an oxidant such as oxygen from a different dischargeable source are provided to cell 10 *via* I/O holes 22 to the anode compartment and cathode compartment respectively to enable electrochemical operation of the cell.

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Figures 3 and 4 of the accompanying drawings show a prior art, air-breathing PEM fuel cell 30 which is very similar in its construction to fuel cell 10, apart from the features to be described below. For the sake of clarity, components in cell 30 which are the same as the components in cell 10 have been attributed the same reference numerals.

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Fuel cell 30 again comprises an electrolytic PEM 15 interleaved between a cathodic catalyst layer 16 and an anodic catalyst layer 17, all of which are interleaved between a cathodic gas diffusion layer 18 and an anodic gas diffusion layer 19. All of these layers are interleaved between anodic current collector plate 13, which is of the same design as that shown in Figure 2, and a cathodic current collector plate 32, which is different to cathodic current collector plate 12 shown in Figure 2. Finally, cell 30 is contained between anodic end plate 11, which is of the same design as that shown in Figure 2, and a cathodic end plate 31, which is different to the cathodic end plate 11 shown in Figure 2.

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Because cell 30 is air-breathing, both cathodic end plate 31 and current collector plate 32 are provided with a series of through-channels 34. In collector plate 32, these channels 34 are provided instead of the surface-deep flow field channels 24 in plate 12 of cell 10, whilst in end plate 31, channels 34 are provided instead of I/O holes 22 in plate 11.

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In use, hydrogen as fuel from a dischargeable source is provided to cell 30 *via* I/O holes 22 in the anodic end plate 11, whilst oxygen from the natural convection of air is able to pass *via* through-channels 34 to the cathode compartment of the cell 30 to enable electrochemical operation of the cell.

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As mentioned above, the attractiveness of PEM fuel cells, especially air-breathing PEM fuel cells, is ever increasing, however the power output obtained from such cells is relatively low compared to conventional PEM fuel cells, due to the dependence of the former on the natural convection of air for their oxidant supply. Attempts have

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previously been made to improve the performance of all PEM fuel cells, with emphasis having been placed on optimisation of the catalyst(s) used to effect the electrochemical reactions underpinning the operation of the cells.

5 Despite such research, there are still many opportunities for improving the performance of PEM fuel cells, especially air-breathing PEM fuel cells; the present invention has been conceived with the goal of improved performance in mind.

10 According to a first aspect of the invention there is provided a PEM fuel cell comprising: an electrically conductive anodic current collector plate, separated from an electrically conductive cathodic current collector plate by a PEM, wherein at least a portion of one or both of the anodic and cathodic current collector plates is provided with a plurality of molecular pores.

15 By the term "molecular pores" is meant apertures, of any configuration, which extend through the collector plate(s) from one major surface to the other major surface thereof, and which are of a size suitable for molecules to pass through substantially unhindered. Thus, the molecular pores may be nanometre sized, or preferably may be micron-sized, to enable passage of at least fuel and oxidant molecules therethrough.
20 The molecular pores comprised in the plurality may be identical, or substantially identical, or may be purposefully provided in different sizes in either a regular or irregular array.

25 Provision of such a PEM fuel cell provides performance advantages over known PEM fuel cells by enabling maximisation of one or more of mass, charge and heat transport through the cell by increasing thermal and/or electrical conductivity of the collector plate(s), by providing significantly shorter diffusion pathways for the fuel and oxidant reactants and/or products and/or by enabling better contact between the collector plate(s) and surrounding structure of the cell.

30 Said anodic and/or cathodic current collector plates may be made from a graphite-based material or a conductive material other than a graphite-based material.

35 In the latter case, the conductive material may be chosen from a metal, a metal alloy, a hybridised metal, a semimetal, a conductive ceramic and a metal-plated silicone.

Advantageously, said conductive material may be chosen from silver, copper, gold, aluminium, zinc, nickel, iron, platinum, titanium, chromium, lead, brass, bronze, carbonised steel, stainless steel, a stainless steel-polycarbonate-graphite hybrid and silicone. Such materials are believed to exhibit the minimum thermal and electrical
5 conductivities required, namely at least 20 W/(mK) and at least 100 S/m respectively, for the operation of the fuel cell to achieve the desired improvement in performance. Preferably, said conductive material may be chosen from aluminium, copper and stainless steel. Further preferably said conductive material is copper.

10 Although the material from which the anodic and/or cathodic current collector plates may be made may have the desired electrical and thermal conductivities, such materials are often susceptible to corrosion, and often on a relatively short timescale as compared to the theoretical operational lifetime of the fuel cell. Consequently, at least a portion of one surface of the anodic and/or cathodic current collector plates may be
15 coated with a corrosion-resistant material. Preferably, the active region of the plate, i.e. the region in which contact with fuel, oxidant (and water) occurs, is entirely provided with the coating of corrosion-resistant material. Further preferably, and for ease of manufacture, an entire plate may be so coated.

20 The corrosion-resistant material may be a precious metal, chosen from gold, silver, palladium, platinum, rhodium, iridium, ruthenium, osmium and rhenium. Further preferably the corrosion-resistant material may be chosen from gold, silver and platinum. Alternatively, the corrosion-resistant material may be a metal nitride, e.g. titanium nitride, a metal carbide, e.g. tungsten carbide, or a nickel-containing material.

25 For the avoidance of doubt, the corrosion-resistant material, having a protective function, would thus be different to the conductive material from which the collector plate(s) may be made.

Advantageously, the conductive material may be electroplated with the corrosion-
30 resistant material, preferably a precious metal.

When made from a conductive material (other than a graphite-based material, as discussed above), the anodic and/or cathodic current collector plate(s) preferably have a thickness in the range of from 50-200 μm , preferably in the range of from 60-175 μm ,
35 more preferably in the range of from 70-150 μm , and most preferably in the range of

from 80-120 μm . These thicknesses are clearly reduced as compared to the thickness of a conventional metallic or graphite-based current collector plate (of 2-10 mm), thereby facilitating increased electrical and thermal conductivities in a fuel cell according to the invention. Furthermore, by relative reduction of the diffusion path through a collector plate for oxidant supply and water removal, increased mass transport is achieved, which improves the rate of electrochemical reaction in the cell, thus improving its power output. Moreover, a reduction in thickness leads to an overall reduction in both the weight and the volume of the overall fuel cell, which is further advantageous.

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Regarding the plurality of molecular pores, said portion in which the plurality is provided may itself be provided as an inherent part of said plate, i.e. the plate may be a one-piece item having the plurality of molecular pores formed therein. Alternatively, said portion may be a separable, exchangeable part of said plate.

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Regardless of the manner in which the portion containing the plurality of molecular pores is provided, advantageously, said portion may be provided as a conductive molecular mesh. The molecular mesh may be made from a metal, a metal alloy, a hybridised metal, a semimetal, a conductive ceramic or a metal-plated silicone. Advantageously, the material from which the molecular mesh is made may be chosen from silver, copper, gold, aluminium, zinc, nickel, iron, platinum, titanium, chromium, lead, brass, bronze, carbonised steel, stainless steel, a stainless steel-polycarbonate-graphite hybrid and silicone. Preferably, said conductive material may be chosen from aluminium, copper and stainless steel. Further preferably said conductive material is copper.

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In the same manner as when the anodic and/or cathodic current collector plate(s) is made from a conductive material (other than a graphite-based material), the conductive molecular mesh may be coated with a corrosion-resistant material.

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The corrosion-resistant material may be a precious metal, chosen from gold, silver, palladium, platinum, rhodium, iridium, ruthenium, osmium and rhenium. Further preferably the corrosion-resistant material may be chosen from gold, silver and platinum. Alternatively, the corrosion-resistant material may be a metal nitride, e.g. titanium nitride, a metal carbide, e.g. tungsten carbide, or a nickel-containing material.

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For the avoidance of doubt, the corrosion-resistant material, having a protective function, would thus be different to the conductive material from which the molecular mesh may be made.

- 5 Advantageously, the conductive material may be electroplated with the corrosion-resistant material, preferably a precious metal.

The portion of the anodic and/or cathodic current collector plate(s) that may be provided with a plurality of molecular pores preferably has a thickness in the range of
10 from 50-200 μm , preferably in the range of from 60-175 μm , more preferably in the range of from 70-150 μm , and most preferably in the range of from 80-120 μm . These thicknesses are clearly reduced as compared to the overall, generally uniform thickness of a conventional metallic or graphite-based current collector plate, thereby facilitating increased electrical and thermal conductivities *via* said portion in a fuel cell
15 according to the invention. Furthermore, by relative reduction of the diffusion path through a collector plate for oxidant supply and water removal, increased mass transport is achieved, which improves the rate of electrochemical reaction in the cell, thus improving its power output.

20 Advantageously, the number of inherent micron-sized molecular pores per unit area provided in the plurality thereof in the portion of the anodic and/or cathodic current collector plate(s) may be at least 100 pores/cm², preferably between 200 and 1,500 pores/cm², more preferably between 250 and 1,400 pores/cm², and most preferably between 300 and 1,300 pores/cm². Assuming said portion has an area of, for example,
25 3.4 cm x 3.4 cm, for pores having a diameter of 150 μm and a centre-to-centre distance of 300 μm , the number of micropores provided in said portion would be 12,800. For a corresponding portion in which the pore diameter is 200 μm and the centre-to-centre distance is 400 μm , 7,200 micropores would be provided, whilst a pore diameter of 300 μm and a centre-to-centre distance of 500 μm would provide 3,200
30 such micropores.

A reduction in thickness of the plate(s) in the region provided with the plurality of molecular pores, in addition to the provision of molecular pores *per se* leads to an overall reduction in both the weight and the volume of the overall fuel cell, which is
35 further advantageous.

In a preferred embodiment of the invention, one or both of the anodic and cathodic current collector plates in the PEM fuel cell, further preferably only the cathode collector plate, is made from a conductive material other than a graphite-based material and at least a portion of said plate(s) is provided with a plurality of molecular pores.

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Alternatively, both the anodic and the cathodic current collector plates are so provided, and thus are identical.

In a yet further preferred embodiment according to the invention one or both the anodic and cathodic current collector plates are made from gold-plated, 100 μm thick copper having between 3,000 and 13,000 inherent micron-sized molecular pores provided therein for an active area of about ten square centimetres.

In a further development to the PEM fuel cell of the invention, when configured as a non-air-breathing cell, at least one further PEM may be interleaved between one or the other of the anodic current collector plate and the cathodic current collector plate, said plate thus becoming a bipolar plate, and a further anodic or cathodic current collector plate as appropriate. Indeed, several PEMs may be interleaved between sequential bipolar plates, which form the required electrodes. In such circumstances, one surface of a bipolar plate acts as an anode in relation to one adjacent PEM, whilst the other surface of the bipolar plate acts as a cathode in relation to the other (or next) adjacent PEM. Correspondingly for an air-breathing cell, since the cathodic side of the cell is open to the atmosphere, the same kind of interleaving is not possible; instead, multiple cells may be electrically connected adjacent to one another along their edges forming a monolayer.

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It is within the scope of the present invention that a PEM fuel cell may further comprise an anodic catalyst layer and a cathodic catalyst layer, one positioned each side of the PEM, inwardly of the anodic current collector plate and the cathodic current collector plate respectively.

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It is furthermore within the scope of the present invention that a PEM fuel cell may yet further comprise an anodic gas diffusion layer and a cathodic gas diffusion layer, each positioned outwardly of the anodic catalyst layer and the cathodic catalyst layer, but

inwardly of the anodic current collector plate and the cathodic current collector plate, respectively.

5 It is yet furthermore within the scope of the present invention that a PEM fuel cell may even further comprise one or a pair of end plates positioned outwardly of the (outermost) anodic and/or cathodic current collector plates.

10 Assuming sealing of the fuel cell by provision of a suitable gasket material or the like between the pair of end plates, in order for fuel and oxidant to be introduced into the cell, and for water to be able to escape, at least one molecular slot or channel may be provided in each of the end plates, preferably in a centralised region thereof, for allowing hydrogen or a hydrogen source into the cell at one end, for allowing oxygen or air into the cell at the other end, and for removal of excessive liquid water and non-reacted oxygen and hydrogen from the cell. The terms "molecular slot" and "molecular channel" should be construed in the same manner as "molecular pore" discussed earlier. Typically, these slots or channels may have micron-sized dimensions.

20 For the avoidance of any doubt, the PEM fuel cell described herein may be based on a conventional fuel cell, however the invention finds particular and preferred application when the fuel cell is an air-breathing fuel cell.

25 In a particularly preferred application of the present invention, a PEM fuel cell as described herein may be used to provide at least some, if not all, of the power required by an electronic device, preferably a portable electronic device, such as a laptop computer either directly or by means of charging/recharging a lithium-ion battery. The device may also be hand-held, and may be a telecommunications device, such as a mobile telephone or a tablet device. In all cases, the PEM fuel cell preferably weighs less than 500 g, further preferably less than 350 g and most preferably less than 200 g.

30 According to a second aspect of the present invention there is provided a method of improving the power output of a PEM fuel cell comprising the steps of:
providing at least a portion of one or both of an anodic current collector plate and a cathodic current collector plate, located either side of a PEM, with a plurality of molecular pores,

whereby any one or more of mass transport, charge transport and heat transfer through the cell is improved as compared to known PEM fuel cells.

5 Of course, in such a fuel cell, the improvement may only be observed once fuel (especially hydrogen) and an oxidant (especially oxygen from air) is supplied to the cell in order for the power output to be determined. The improvement may be achieved when the cell is operated continuously and/or when operated over a finite time period.

10 The improvement may be achieved by making a new PEM fuel cell or by retro-fitting the extant anodic current collector plate and/or the cathodic current collector plate in an existing PEM fuel cell with such plates as described above, whether by repair or upgrade.

15 Thus, the method of the invention may further comprise the step of forming, or having pre-formed, the plurality of molecular pores in a portion of one or both plates by any of the following techniques: laser-cutting, laser-drilling, acid-etching (also known as chemical milling), mechanical cutting and plasma cutting.

20 Preferably the number of inherent micron-sized molecular pores per unit area provided in the plurality thereof in the portion of the anodic and/or cathodic current collector plate(s) may be at least 100 pores/cm², preferably between 200 and 1,500 pores/cm², more preferably between 250 and 1,400 pores/cm², and most preferably between 300 and 1,300 pores/cm²; which may be so formed or pre-formed.

25 In a preferred embodiment of the invention, one or both of the anodic current collector plate and the cathodic current collector plate, further preferably only the cathode collector plate, may be provided in a conductive material other than a graphite-based material and at least a portion of one or both of said plates may be provided with the plurality of molecular pores.

30 Alternatively, both the anodic current collector plate and the cathodic current collector plate may be so provided.

Most preferably, one or both the anodic and cathodic current collector plates may be provided in gold-plated, 100 μm thick copper having between 3,000 and 13,000 inherent micron-sized molecular pores formed or pre-formed therein.

5 A further optional step according to the second aspect of the invention comprises, in the case of a non-air-breathing fuel cell, interleaving at least one further PEM between one or the other of the anodic current collector plate and the cathodic current collector plate, such that said plate is functional as a bipolar plate, and a further anodic or
10 provided and the overall power output of the fuel cell further increased. Alternatively, for an air-breathing cell, since the cathodic side of the cell is open to the atmosphere and the same kind of interleaving is not possible, the further optional step comprises electrically connecting two or more air-breathing cells along their adjacent edges to form a monolayer. Of course, any such stacking or provision of a monolayer will likely
15 be limited by the physical dimensions of the space available for the cell in a device, which must be borne in mind.

The peak power density (mW/cm^2) achievable by implementation of the improvement of the invention may be at least 20 %, preferably at least 25 %, and further preferably
20 at least 30 %, as compared to the peak power density achievable with an otherwise conventional PEM fuel cell operated without said improvement. This improvement is observed with both air-breathing and non-air-breathing PEM fuel cells configured according to the invention.

25 For a better understanding, the present invention will now be more particularly described, by way of non-limiting example only, with reference to certain of the accompanying schematic drawings (not to scale), in which:

Figure 1 is a perspective view of a prior art, non-air-breathing PEM fuel cell;

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Figure 2 is an exploded, perspective view of the fuel cell shown in Figure 1;

Figure 3 is a perspective view of a prior art, air-breathing PEM fuel cell;

35 Figure 4 is an exploded, perspective view of the fuel cell shown in Figure 3;

Figure 5 is a perspective view of an air-breathing PEM fuel cell according to the invention;

Figure 6 is an exploded, perspective view of the fuel cell shown in Figure 5;

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Figure 7 is a perspective view of a component of the fuel cell shown in Figures 5 and 6;

Figure 8 shows polarisation curves for an air-breathing PEM fuel cell according to the invention and for a prior art air-breathing PEM fuel cell (the plot showing cell potential, measured in volts (V), on the left-hand y-axis, power density, measured in milliwatts per square centimetre (mW/cm^2), on the right hand y-axis and current density, measured in milliamps per square centimetre (mA/cm^2), on the x-axis;

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Figure 9 is a Nyquist plot showing impedance data (the real part, Z_{Re} (Ohms), being plotted on the x-axis and the imaginary part, $-Z_{Im}$ (Ohms), being plotted on the y-axis) for the PEM fuel cell and the prior art PEM fuel cell of Figure 8;

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Figure 10 shows further polarisation curves for another air-breathing PEM fuel cell according to the invention and for the prior art air-breathing PEM fuel cell of Figure 8 (the plot showing cell potential, measured in volts (V), on the left-hand y-axis, power density, measured in milliwatts per square centimetre (mW/cm^2), on the right hand y-axis and current density, measured in milliamps per square centimetre (mA/cm^2), on the x-axis;

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Figure 11 shows repeated power density curves for the air-breathing PEM fuel cell of Figure 8 (the plot showing power density, measured in milliwatts per square centimetre (mW/cm^2), on the y-axis and current density, measured in milliamps per square centimetre (mA/cm^2), on the x-axis;

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Figure 12 is a plot of the time dependence (in hours) of the cell potential, measured in volts (V), for an air-breathing fuel cell according to the invention;

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Figure 13 shows polarisation curves for two different air-breathing PEM fuel cells according to the invention and for a prior art air-breathing PEM fuel cell (the plot showing cell potential, measured in volts (V), on the left-hand y-axis, power density,

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measured in milliwatts per square centimetre (mW/cm^2), on the right hand y-axis and current density, measured in milliamps per square centimetre (mA/cm^2), on the x-axis; and

5 Figure 14 shows polarisation curves for two different non-air-breathing PEM fuel cells according to the invention and for a prior art non-air-breathing PEM fuel cell (the plot showing cell potential, measured in volts (V), on the left-hand y-axis, power density, measured in milliwatts per square centimetre (mW/cm^2), on the right hand y-axis and current density, measured in milliamps per square centimetre (mA/cm^2), on the x-axis.

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Figures 5 and 6 show an air-breathing PEM fuel cell 50 which is similar in its construction to prior art, air-breathing fuel cell 30 shown in Figure 3 and 4, with notable differences however. For the sake of clarity, components in cell 50 which are the same as the components in cell 30 have been attributed the same reference numerals.

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Fuel cell 50 again comprises an electrolytic PEM 15 interleaved between a cathodic catalyst layer 16 and an anodic catalyst layer 17, all of which are interleaved between a cathodic gas diffusion layer 18 and an anodic gas diffusion layer 19. All of these layers are interleaved between anodic current collector plate 13, which is of the same design as that shown in Figure 4, and a cathodic current collector plate 52, which is different to cathodic current collector plate 32 shown in Figure 4. Finally, cell 50 is contained between anodic end plate 11 and a cathodic end plate 31, both of which are of the same design as those shown in Figure 4.

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25 As with the prior art PEM fuel cell 30 shown in Figures 3 and 4, cell 50 shown in Figures 5 and 6 is air-breathing. Consequently, cathodic end plate 31 is provided with a series of through-channels 34.

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Unlike fuel cell 30 shown in Figure 3 and 4 however, cell 50 comprises novel and inventive cathodic current collector plate 52, in which a central portion 55 is provided with a plurality 56 of molecular pores.

The PEM 15 may be any such suitable, semi-permeable ionomer membrane known in the art, including both pure polymer membranes and composite membranes having

other species embedded in the polymer matrix. However, preferably, the PEM is the fluoropolymer available under the trade name Nafion™, from Du Pont.

5 The cathodic and anodic catalyst layers 16, 17 may be provided as platinum-based catalysts (with platinum loadings of 0.4 and 0.2 mg/cm² respectively). Each of these catalyst layers may be obtained from Alfa Aesar, and manually sprayed as a solution onto the respective cathodic and anodic gas diffusion layers 18, 19. Thus, although Figure 6 (and indeed prior art Figures 2 and 4) shows the catalyst layers as being separate entities from the gas diffusion layers, in practice, the catalyst layers may be
10 formed on, and thus inseparable from, the gas diffusion layers.

The cathodic and anodic gas diffusion layers 18, 19 may be provided as Teflon™-treated carbon-fibre-based layers, e.g. SGL 10BC gas diffusion layers available from SGL Group, which have enhanced hydrophobicity thereby increasing their ability to
15 expel water.

The gasket material 14 used to seal the interior of fuel cell 50 between the end plate 11 and end plate 31 may be a silicone rubber, e.g. of 0.5 mm thickness, available from Samco Silicone Products.
20

In use, a fuel such as hydrogen or a source of hydrogen is supplied to fuel cell 50 *via* end plate 11 to the anodic compartment, whilst an oxidant such as pure oxygen or oxygen from air, is supplied *via* natural air convection to the other end plate 31 to the cathodic compartment. Each of the fuel and oxidant travels through the respective end
25 plate 11, 31 to meet anodic and cathodic current collector plates 13, 52 respectively, whereupon each is able to pass therethrough (as will be described in more detail below), *via* respective anodic and cathodic gas diffusion layers 19, 18 to respective anodic and cathodic catalyst layers 17, 16.

30 Upon reaching anodic catalyst layer 17, hydrogen is ionised to form hydrogen ions and free electrons. The liberated hydrogen ions diffuse through PEM 15 to the cathodic compartment, whilst the electrons are conducted by anodic gas diffusion layer 19 to anodic current collector plate 13, from where they are onwardly conducted *via* an external circuit (not shown), outside of fuel cell 50, to cathodic current collector plate
35 52. At cathodic catalyst layer 16, oxygen electrochemically reacts with hydrogen ions

that have diffused through PEM 15 and electrons that have been conducted from cathodic current collector plate 52 to form water vapour and to release heat.

5 The improvements observed with the present invention, as discussed in further detail in relation to Figures 8 to 14, is achieved because one (the cathodic current collector plate 52 as shown in Figures 5 and 6) or both (not shown) of anodic current collector plate 13 and cathodic current collector plate 52 has at least a portion thereof provided with a plurality of molecular pores.

10 In a preferred embodiment of the invention, as shown in Figure 7, cathodic current collector plate 52 is made from a conductive material other than a graphite-containing material, and so is able to be made much thinner than a conventional graphite-containing plate (the thickness being indicated by "t" in the figure), and is designed so that central portion 55 of said plate is provided with a plurality 56 of molecular pores, in
15 the form of a microporous molecular mesh. Again, a current collection tab 53 is provided. In one example, plate 52 is formed by acid-etching as a gold-plated, 100 μm thick copper plate having 12,800 inherent micron-sized molecular pores formed in its central region.

20 It would also be possible, although not explicitly illustrated in the accompanying figures, for a conventional graphite-containing current collector plate (or indeed any other known collector plate) to be adapted so as to be provided with a portion containing a plurality of molecular pores, for example in the form of a microporous molecular mesh, in a similar manner to portion 55 of plate 52 shown in Figure 7. In such a case, said
25 alternative portion would be of the same thickness, t, as the whole of plate 52 in Figure 7, whilst the remainder of the conventional plate would be much thicker. The alternative portion may be provided in a conductive material other than a graphite-containing material, such as gold-plated copper, and may also be provided as a separate part, to be fixed to/in an appropriate aperture in the otherwise conventional
30 plate.

Notwithstanding the aforementioned alternatives, with cathodic current collector plate 52 provided as shown in Figure 7 (provided as a gold-plated, 100 μm thick copper plate having 12,800 inherent micron-sized molecular pores formed in its central region) in
35 cell 50, the performance of air-breathing cell 50 was measured and compared to an

otherwise identical air-breathing cell however provided with a conventional graphite-containing cathodic current collector plate (such as is shown in Figure 3 and 4). Such a conventional plate is made from graphite (70-90 %) and a phenolic polymer (10-30%), and is available from Bac2 Limited, Basepoint Romsey, Romsey, SO51 9AQ, UK.

Figure 8 illustrates the polarization curves for a prior art air-breathing fuel cell (having square-shaped data points) and an air-breathing fuel cell operating in accordance with the present invention (having triangular-shaped data points). The solid data points relate to cell potential whilst the hollow data points relate to power density. From Figure 8 it is clear that the fuel cell of the invention performs significantly better than the prior art fuel cell; the peak power has been improved by about 31 % (or 36 % if the contact resistances associated with the collection terminals are not corrected for).

The curves shown in the Nyquist plot of Figure 9 were obtained by taking electrochemical impedance spectroscopy (EIS) measurements at 0.75 V cell potential. These show that the ohmic and charge resistances are significantly reduced, by about 41 % and 28 % respectively, for the fuel cell according to the invention (having solid square data points) compared to the prior art fuel cell (having solid triangular data points). It is thought that the reduced ohmic resistance may be mainly attributed to: (i) the establishment of better electrical contact between the metallic, cathodic current collector and the adjacent gas diffusion layer, and (ii) the increased electrical conductivity of the metallic collector as compared to the conventional graphite-based current collector (the former being several orders of magnitudes larger than the latter). Similarly, it is thought that the reduction in the charge transfer resistance, or kinetic resistance, in the case of the metallic, cathodic current collector may indicate that the activity of the catalyst has been increased, which is thought most likely to be due to enhanced reaction rate, and subsequently increased water generation.

Figure 10 illustrates the polarization curves for the same prior art fuel cell (having square-shaped data points) as tested for Figure 8 and another fuel cell operating in accordance with the present invention (having triangular-shaped data points), to test the reproducibility of the data obtained for Figure 8. The solid data points again relate to cell potential whilst the hollow data points again relate to power density. From Figure 10 the superiority of the metallic porous current collector over the conventional

collector is confirmed. Interestingly, the size of the improvement observed in Figure 10 is greater than that shown in Figure 9 (namely a 40 / 44 % difference, rather than a 31 / 36 % difference) in terms of the peak power.

- 5 Figure 11 illustrates the repeatability of the polarization measurements taken for the fuel cell of the invention; three separate measurements were taken, and as shown in Figure 11, the performance of the fuel cell of the invention is highly repeatable.

10 Figure 12 plots the time dependence (in hours) of the cell potential for the fuel cell to the invention when operated at 0.6A. Clearly, the performance of the cell is reasonably stable over the five hours tested, with particular stability having been observed after the first two hours of testing. For this particular fuel cell, a commercial water-repellent coating (RepelSmart™ available from Chamelic Limited, Leeds, UK) was applied in a thickness of the order of 1 micron or less to the surface of the cathodic current collector
15 which is directed towards the ambient external environment in order to prevent water accumulation and to ensure a continuous supply of oxygen to the cathodic electrode.

Notwithstanding the aforementioned alternatives, with a cathodic current collector plate provided as shown in Figure 7 (provided as a gold-plated, 100 µm thick copper plate
20 having 12,800 inherent micron-sized molecular pores formed in its central region) an air-breathing fuel cell that is otherwise identical to that shown in Figure 5 and 6, with the exception that an identical plate is also provided as the anodic current collector, the performance of said twice-modified cell was measured and compared to an otherwise identical cell however provided with conventional graphite-containing cathodic and
25 anodic current collector plates (of the type as described earlier).

Figure 13 illustrates the polarization curves for a prior art air-breathing fuel cell (having square-shaped data points) and the twice-modified air-breathing fuel cell described in the preceding paragraph operating in accordance with the present invention (having
30 circular-shaped data points). Also shown are the data points for the fuel cell of the invention shown in Figure 10 (having triangular-shaped data points). The solid data points relate to cell potential whilst the hollow data points relate to power density. From Figure 13 it is clear that the twice-modified fuel cell of the invention performs significantly better than the prior art fuel cell; the peak power has been improved by

about 60 %, and better than the fuel cell having only its cathodic current collector plate of the type shown in Figure 7 by about 25 %.

5 Finally, to illustrate the applicability of the present invention to non-air-breathing fuel cells, as well as to air-breathing fuel cells, polarization curves were generated, as shown in Figure 14, for a fuel cell as otherwise shown in Figures 1 and 2 but, firstly with only the cathodic current collector plate, and secondly with both the anodic and cathodic current collector plates, having been replaced by a current collector of the type shown in Figure 7, (provided as a gold-plated, 100 μm thick copper plate having
10 12,800 inherent micron-sized molecular pores formed in its central region).

Figure 14 illustrates the polarization curves for a prior art non-air-breathing fuel cell (having square-shaped data points), a non-air-breathing fuel cell having its cathodic current collector plate replaced operating in accordance with the present invention
15 (having triangular-shaped data points) and a non-air-breathing fuel cell having both its cathodic and anodic current collector plates replaced operating in accordance with the present invention (having circular-shaped data points). The solid data points relate to cell potential whilst the hollow data points relate to power density. From Figure 14 it is clear that the non-air-breathing fuel cell of the invention having only its cathodic current
20 collector plate replaced performs significantly better than the prior art fuel cell; the peak power has been improved by about 75 %, whilst the non-air-breathing fuel cell of the invention having both of its current collector plates replaced exhibits an improvement in peak power of about 135 % as compared to the prior art fuel cell.

25 Clearly, provision in at least a portion of one or both of an anodic current collector plate and a cathodic current collector plate of a plurality of molecular pores improves, and typically significantly improves, the performance of a PEM fuel cell, whether it is of the air-breathing type, reliant upon the natural convection of air for its oxidant supply or of the non-air-breathing type, reliant upon dischargeable sources of both fuel and oxidant.

CLAIMS:

1. A proton exchange membrane fuel cell comprising:
an electrically conductive anodic current collector plate, separated from an
5 electrically conductive cathodic current collector plate by a proton exchange
membrane,
wherein at least a portion of one or both of the anodic and cathodic current
collector plates is provided with a plurality of molecular pores.
- 10 2. A fuel cell as claimed in claim 1 wherein one or both of the anodic and cathodic
current collector plates is made from a graphite-based material or a conductive
material other than a graphite-based material.
- 15 3. A fuel cell as claimed in claim 2 wherein the conductive material is chosen from
a metal, a metal alloy, a hybridised metal, a semimetal, a conductive ceramic
and a metal-plated silicone.
- 20 4. A fuel cell as claimed in claim 3 wherein the conductive material is chosen from
silver, copper, gold, aluminium, zinc, nickel, iron, platinum, titanium, chromium,
lead, brass, bronze, carbonised steel, stainless steel, a stainless steel-
polycarbonate-graphite hybrid and silicone.
- 25 5. A fuel cell as claimed in claim 2 wherein the anodic and/or cathodic current
collector plates, when made from the conductive material, has at least a portion
of one surface thereof coated with a corrosion-resistant material.
- 30 6. A fuel cell as claimed in claim 5 wherein the corrosion-resistant material is a
precious metal and is different to the conductive material from which the
collector plate(s) is made.
- 35 7. A fuel cell as claimed in claim 4 wherein the conductive material is plated with
the precious metal.
8. A fuel cell as claimed in any of claims 2-7 wherein the anodic and/or cathodic
current collector plate(s), when made from the conductive material, has a

thickness in the range of from 50-200 μm , preferably in the range of from 60-175 μm , more preferably in the range of from 70-150 μm , and most preferably in the range of from 80-120 μm .

- 5 9. A fuel cell as claimed in any of claims 1-8 wherein the portion of the anodic and/or cathodic current collector plate(s) that is provided with a plurality of molecular pores is an inherent part of said plate.
- 10 10. A fuel cell as claimed in any of claims 1-9 wherein the portion of the anodic and/or cathodic current collector plate(s) that is provided with a plurality of molecular pores is a separable, exchangeable part of said plate.
- 15 11. A fuel cell as claimed in claim 9 or claim 10 wherein the portion of the plate(s) is provided as a conductive molecular mesh.
- 20 12. A fuel cell as claimed in claim 11 wherein the molecular mesh is made from a metal, a metal alloy, a hybridised metal, a semimetal, a conductive ceramic or a metal-plated silicone.
- 25 13. A fuel cell as claimed in claim 12 wherein the material from which the molecular mesh is made is chosen from silver, copper, gold, aluminium, zinc, nickel, iron, platinum, titanium, chromium, lead, brass, bronze, carbonised steel, stainless steel, a stainless steel-polycarbonate-graphite hybrid and silicone.
- 30 14. A fuel cell as claimed in claim 12 or claim 13 wherein the molecular mesh is coated with a corrosion-resistant material.
15. A fuel cell as claimed in claim 14 wherein the corrosion-resistant material is a precious metal and is different to the conductive material from which the molecular mesh is made.
16. A fuel cell as claimed in claim 15 wherein the conductive material is plated with the precious metal.

17. A fuel cell as claimed in any of claims 1-16 wherein the portion of the anodic and/or cathodic current collector plate(s) that is provided with a plurality of molecular pores has a thickness in the range of from 50-200 μm , preferably in the range of from 60-175 μm , more preferably in the range of from 70-150 μm , and most preferably in the range of from 80-120 μm .
18. A fuel cell as claimed in any of claims 1-17 wherein the number of inherent micron-sized molecular pores per unit area provided in the plurality thereof in the portion of the anodic and/or cathodic current collector plate(s) is at least 100 pores/ cm^2 , preferably between 200 and 1,500 pores/ cm^2 , more preferably between 250 and 1,400 pores/ cm^2 , and most preferably between 300 and 1,300 pores/ cm^2 .
19. A fuel cell as claimed in claim 1 wherein the anodic and the cathodic current collector plates are identical.
20. A fuel cell as claimed in any preceding claim wherein one or both of the anodic and cathodic current collector plates is made from gold-plated, 100 μm thick copper having between 300 and 1,300 inherent micron-sized molecular pores per square centimetre provided therein.
21. A fuel cell as claimed in any preceding claim further comprising an anodic catalyst layer and a cathodic catalyst layer, one positioned each side of the proton exchange member, inwardly of the anodic current collector plate and the cathodic current collector plate respectively.
22. A fuel cell as claimed in claim 21 further comprising an anodic gas diffusion layer and a cathodic gas diffusion layer, each positioned outwardly of the anodic catalyst layer and the cathodic catalyst layer, but inwardly of the anodic current collector plate and the cathodic current collector plate, respectively.
23. A fuel cell as claimed in claim 22 further comprising one or a pair of end plates positioned outwardly of the (outermost) anodic and/or cathodic current collector plates.

24. A fuel cell as claimed in claim 23 wherein the or each of the end plates is provided with at least one slot for allowing hydrogen or a hydrogen source into the cell at one end, and for allowing water out of the cell at the other end.
- 5 25. A fuel cell as claimed in any preceding claim being an air-breathing fuel cell.
26. Use of a fuel cell as claimed in claim 1 to provide at least some of the power required by an electronic device.
- 10 27. The use as claimed in claim 26 wherein the device is a telecommunications device, such as a mobile telephone or a tablet device.
28. A method of improving the power output of a proton exchange membrane fuel cell comprising the steps of:
- 15 providing at least a portion of one or both of an anodic current collector plate and a cathodic current collector plate with a plurality of molecular pores, whereby any one or more of mass transport, charge transport and heat transfer through the cell is improved as compared to known proton exchange membrane fuel cells.
- 20 29. A method as claimed in claim 28 comprising forming, or having pre-formed, the plurality of molecular pores in a portion of one or both plates by any of the following techniques: laser-cutting, laser-drilling, acid-etching, mechanical cutting, plasma cutting.
- 25 30. A method as claimed in claim 28 or claim 29 comprising forming, or having pre-formed, a number of inherent micron-sized molecular pores per unit area in the plurality thereof in the portion of the anodic and/or cathodic current collector plate(s) of at least 100 pores/cm², preferably between 200 and 1,500 pores/cm², more preferably between 250 and 1,400 pores/cm², and most preferably between 300 and 1,300 pores/cm².
- 30 31. A method as claimed in any one of claims 28-30 comprising providing one or both of the anodic current collector plate and the cathodic current collector plate
- 35 in a conductive material other than a graphite-containing material and providing

at least a portion of one or both of said plates with the plurality of molecular pores.

- 5 32. A method as claimed in any one of claims 28-31 wherein both the anodic current collector plate and the cathodic current collector plate are so provided.
- 10 33. A method as claimed in any one of claims 28-32 comprising providing one or both of the anodic and/or cathodic current collector plates in gold-plated, 100 μm thick copper having between 300 and 1,300 inherent micron-sized molecular pores per square centimetre formed therein.
- 15 34. A method as claimed in any one of claims 28-33 wherein the peak power density (mW/cm^2) achievable by implementation of the improvement of the invention is at least 20 %, preferably at least 25 %, and further preferably at least 30 %, as compared to an otherwise conventional proton exchange membrane fuel cell operated without said improvement.
- 20 35. A proton exchange membrane fuel cell substantially as hereinbefore described with reference to Figures 5 to 7 of the accompanying drawings.
36. Use of a proton exchange membrane fuel cell substantially as hereinbefore described.
- 25 37. A method of improving the power output of a proton exchange membrane fuel cell substantially as hereinbefore described.

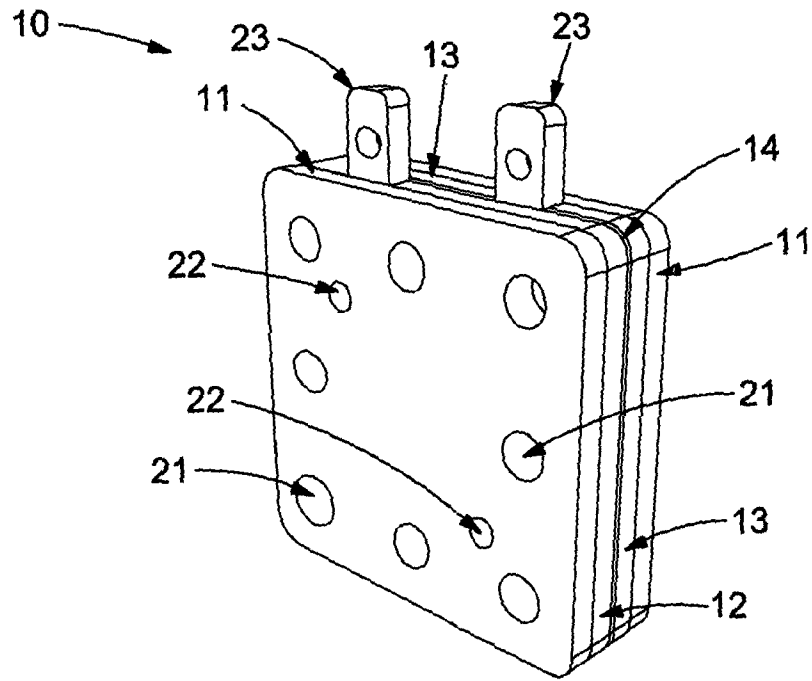


Fig. 1

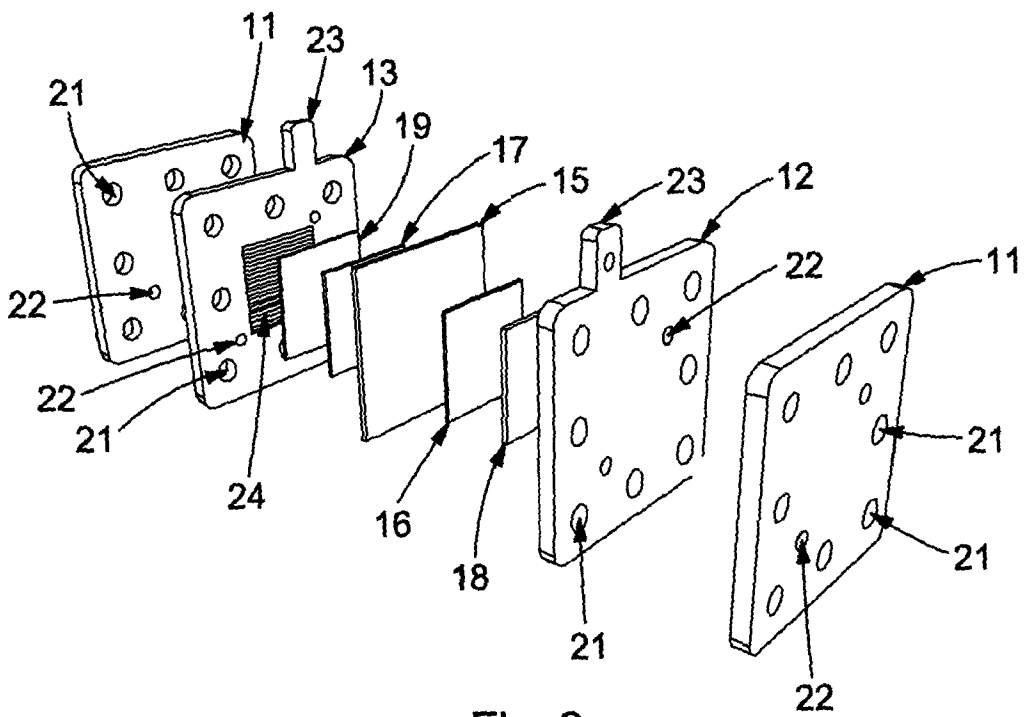


Fig. 2

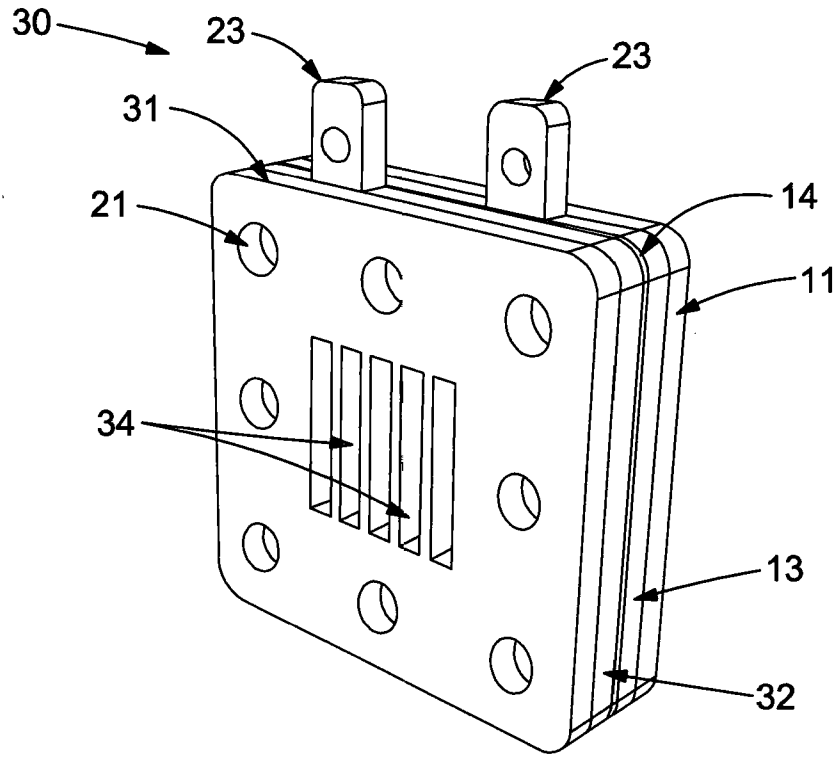


Fig. 3

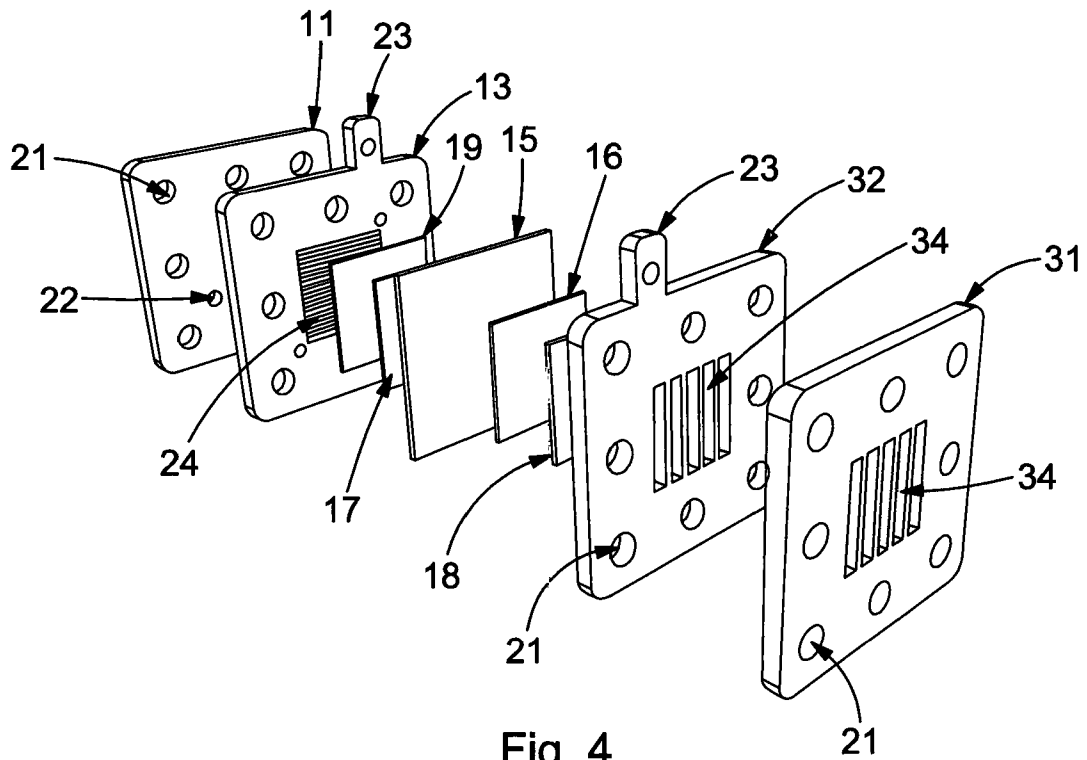


Fig. 4

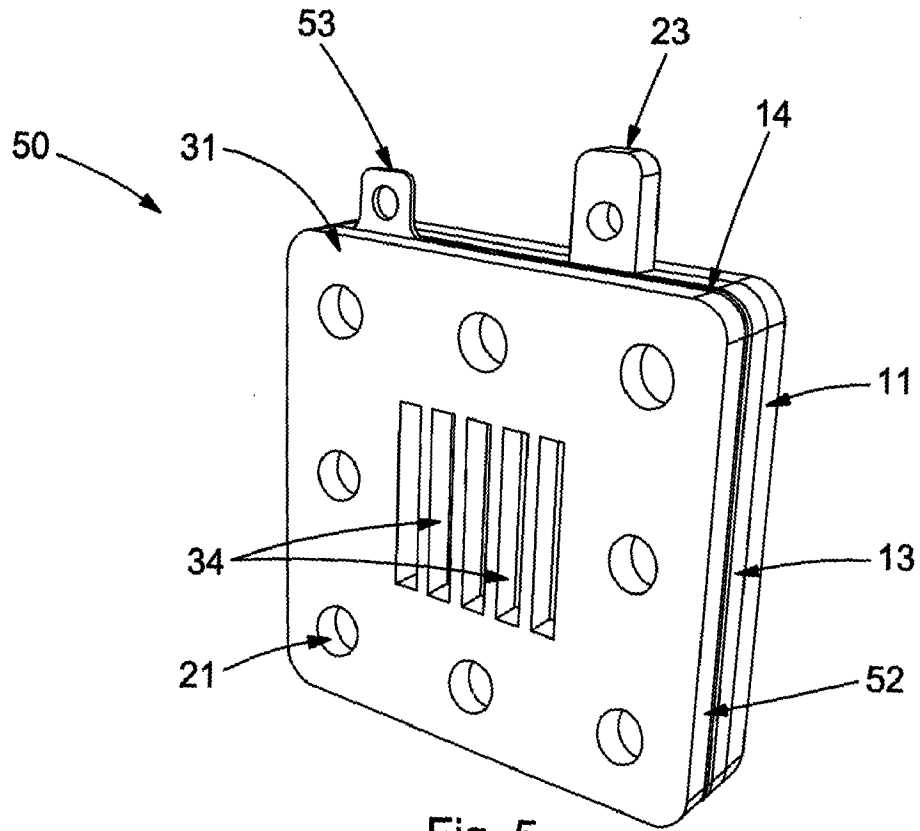


Fig. 5

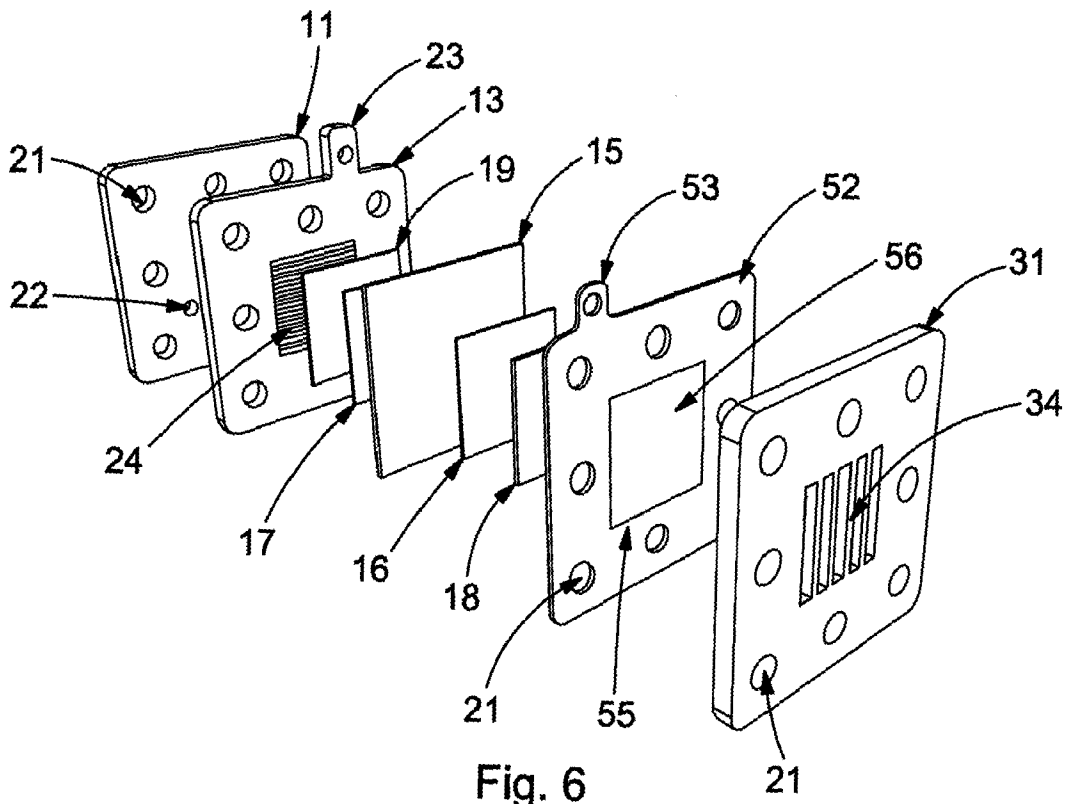


Fig. 6

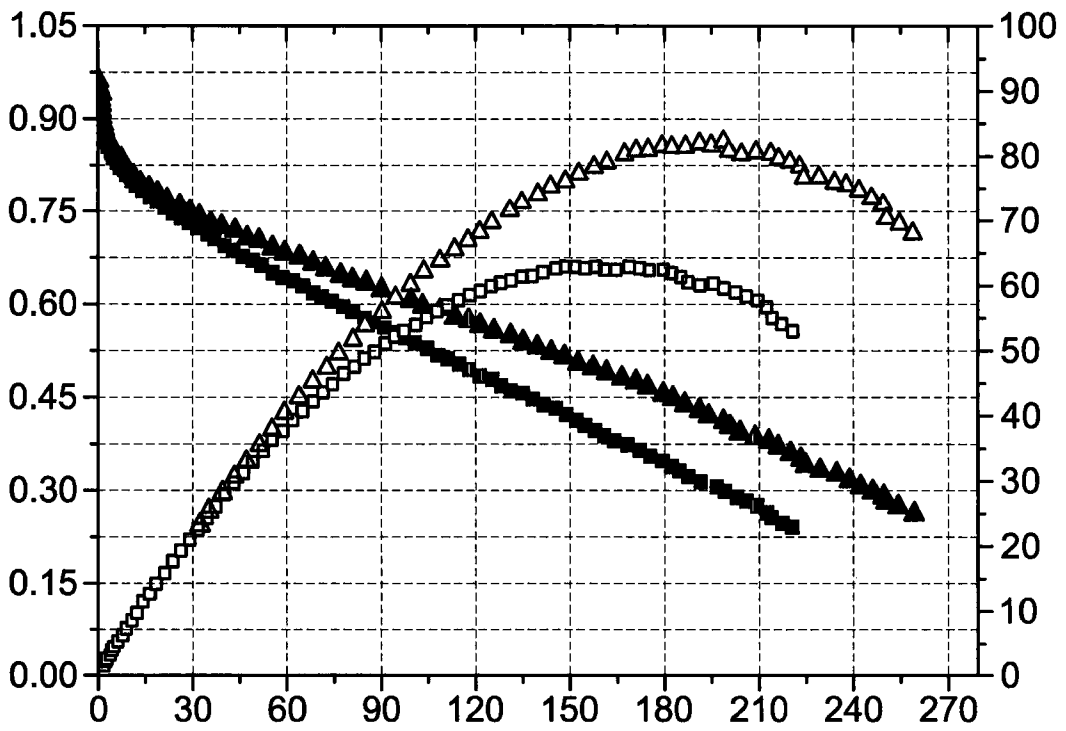
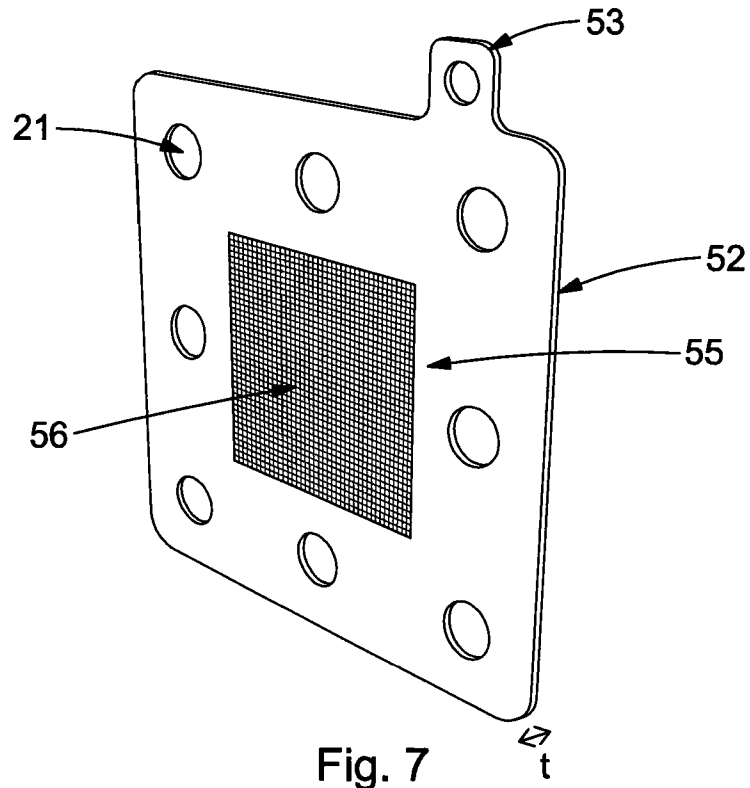


Fig. 8

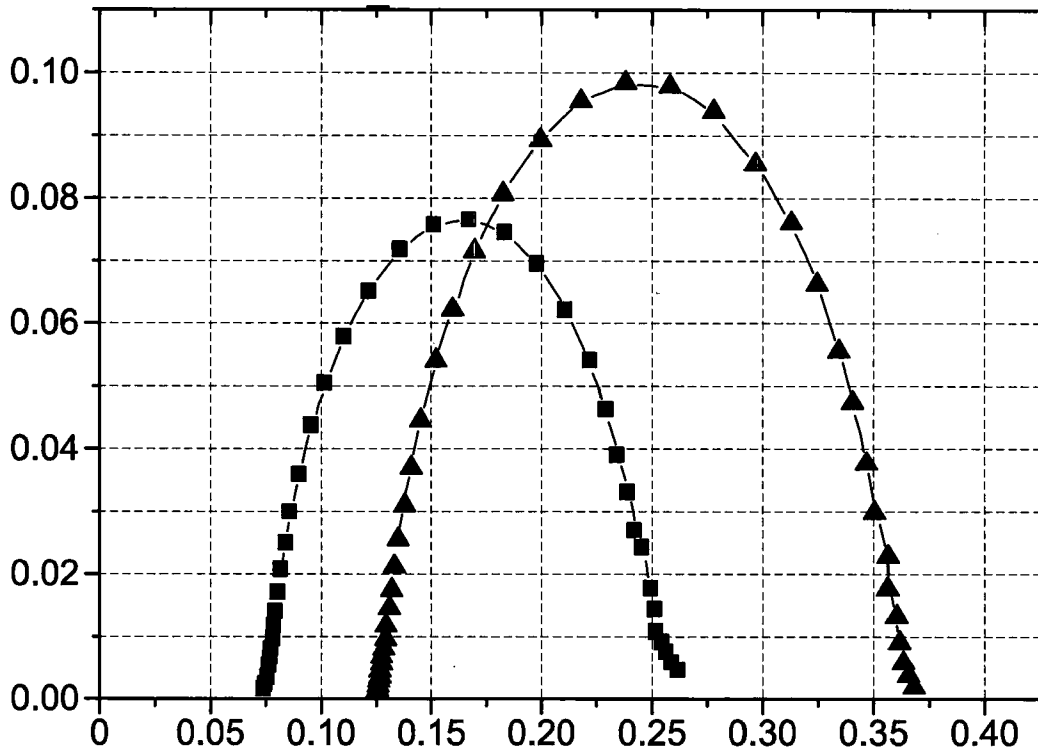


Fig. 9

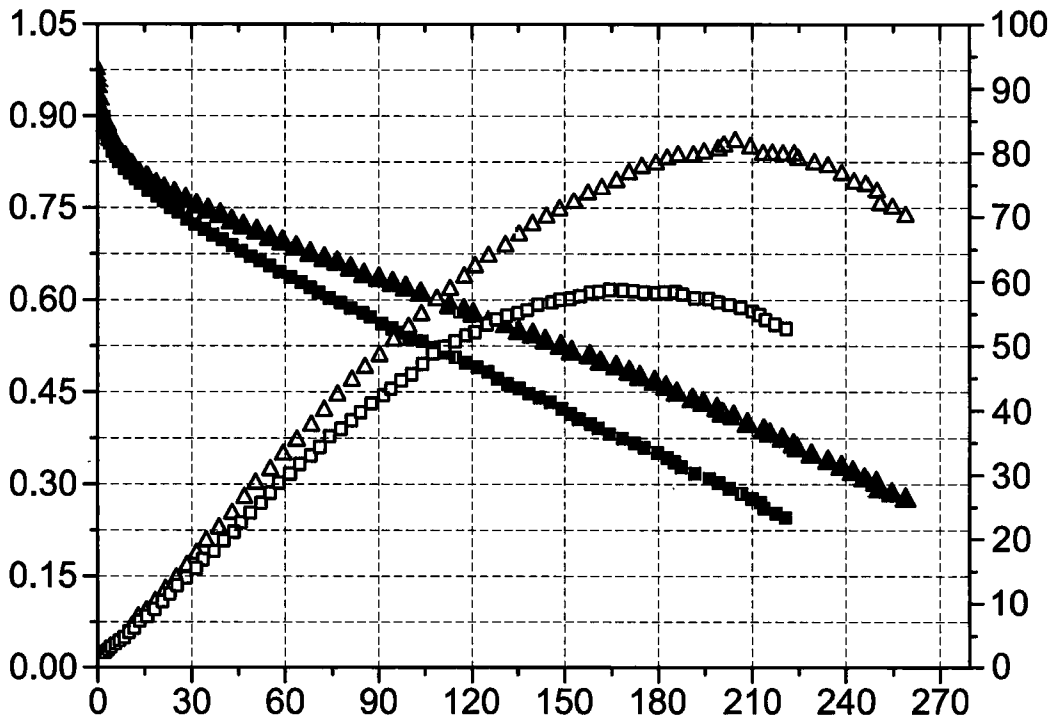


Fig. 10

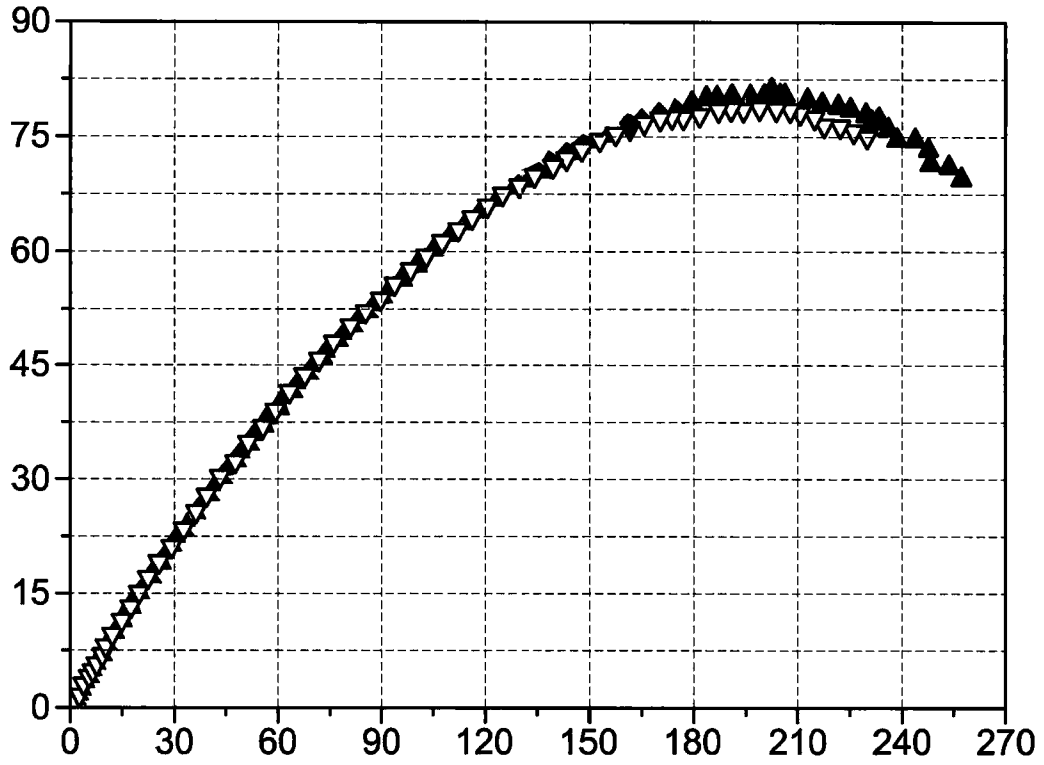


Fig. 11

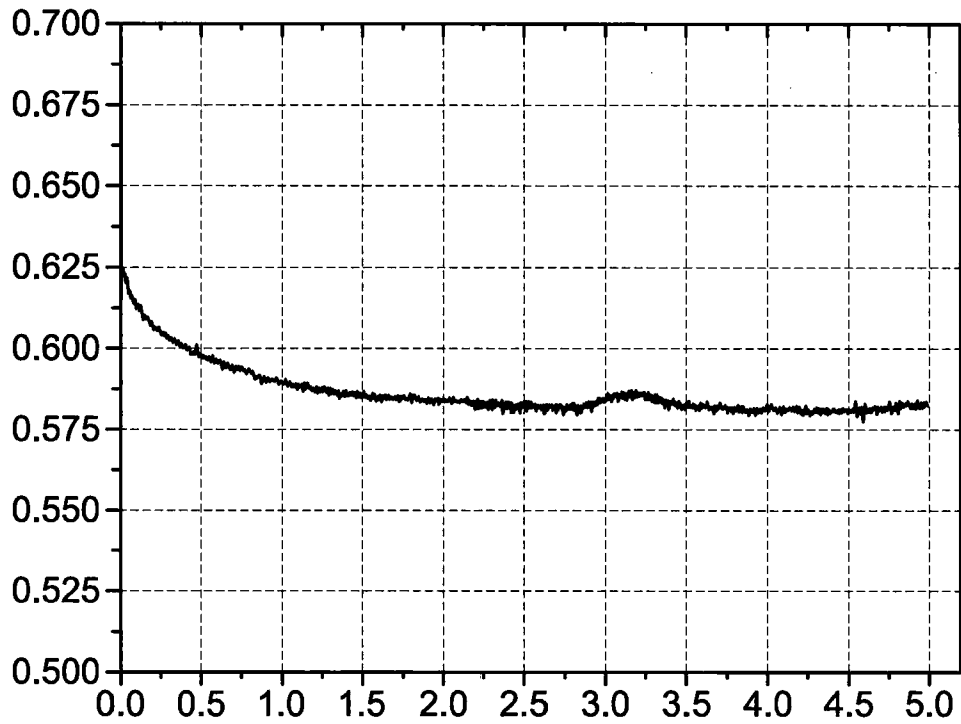


Fig. 12

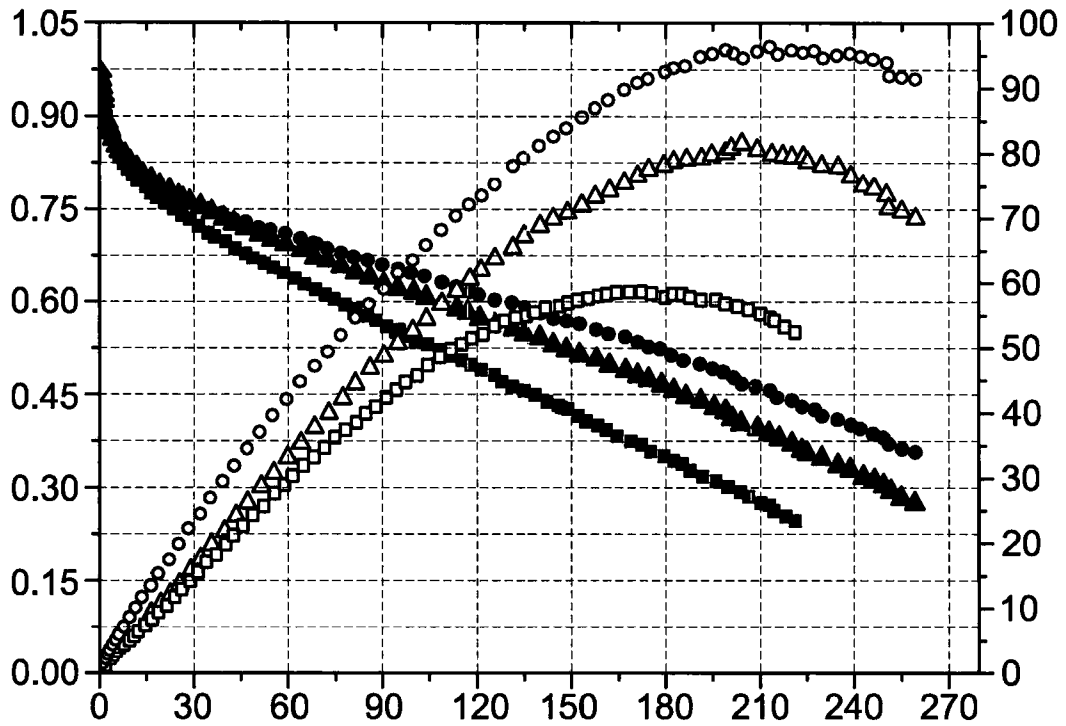


Fig. 13

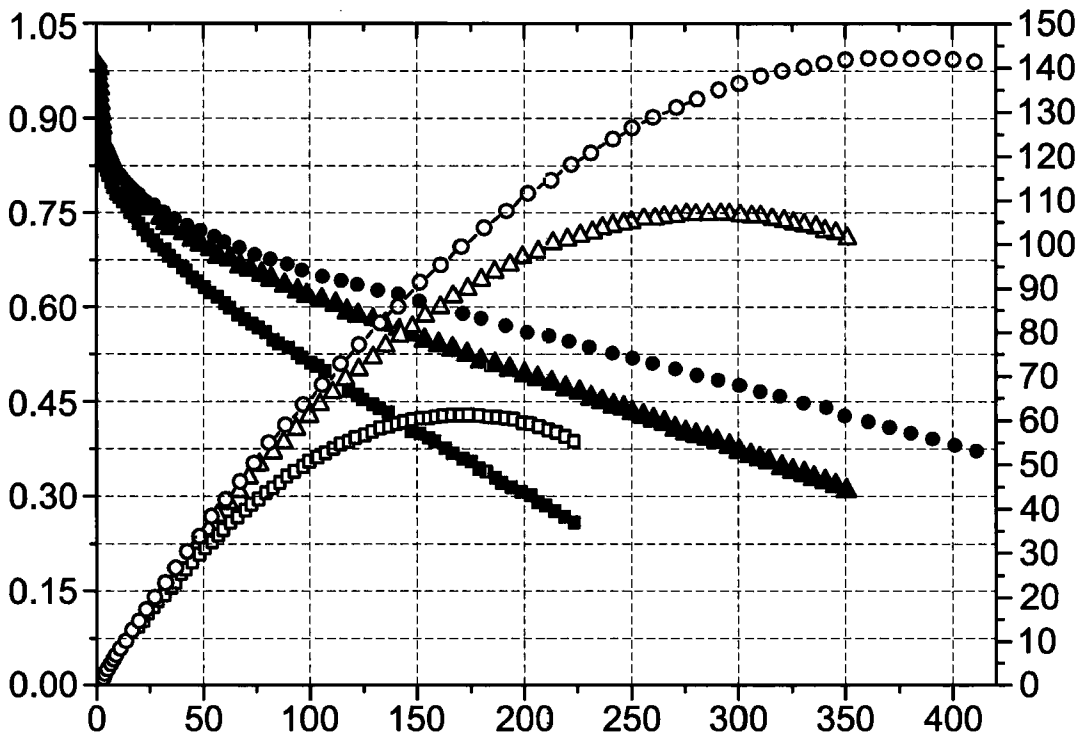


Fig. 14

INTERNATIONAL SEARCH REPORT

International application No

PCT/GB2013/052941

A. CLASSIFICATION OF SUBJECT MATTER

INV. H01M8/02
ADD.

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

H01M

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)

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 Further documents are listed in the continuation of Box C. See patent family annex.

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"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention

"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone

"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art

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Date of the actual completion of the international search

27 February 2014

Date of mailing of the international search report

05/03/2014

Name and mailing address of the ISA/

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Authorized officer

Reich, Claus

INTERNATIONAL SEARCH REPORT

International application No

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C(Continuation). DOCUMENTS CONSIDERED TO BE RELEVANT

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A	WO 2006/063163 A2 (PRAXAIR TECHNOLOGY INC [US]; CHRISTIE GERVAISE MAXWELL [US]; LANE JONAT) 15 June 2006 (2006-06-15) claims 1-18 -----	1-37

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