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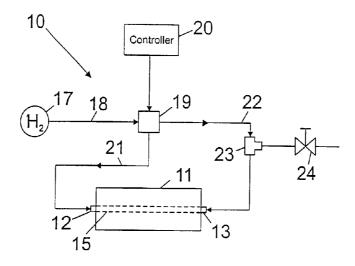
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(54) Title: PASSIVE AIR BREATHING FUEL CELL SYSTEM WITH SWITCHED FUEL GAS DELIVERY



(57) Abstract: A fuel cell system (10) includes a polymer electrolyte fuel cell (11) with a stack of fuel cell units having an internal fuel flow channel extending between a first fuel supply inlet (12) and a second fuel supply inlet (13). A distribution valve (9) is connected by supply lines to the two inlets of the fuel cell and to a source of fuel gas such as hydrogen in a canister under pressure (17). In a first position of the distribution valve, fuel is supplied to the fuel flow channel through the first supply inlet, and in a second position of the valve, fuel is supplied through the second fuel supply inlet. The valve is switched at a selected frequency to provide alternating flow of fuel gas through the fuel flow channel, equalizing the concentration of fuel gas in the fuel flow channel and mixing the water vapor and inert gases within the fuel flow channel to avoid localized concentrations.



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PASSIVE AIR BREATHING FUEL CELL SYSTEM WITH SWITCHED FUEL GAS DELIVERY

FIELD OF THE INVENTION

[0001] This invention pertains generally to fuel cells and particularly to passive air breathing polymer electrolyte fuel cells.

BACKGROUND OF THE INVENTION

[0002] Polymer electrolyte fuel cells (also known as proton exchange membrane fuel cells) are well suited to low power applications of the type now typically served by conventional batteries. Such fuel cells are usually designed to use hydrogen gas as the fuel and may be designed to operate with ambient air as the oxidant. Larger fuel cells are often actively humidified, cooled, or supplied with fuel or oxygen under pressure. However, for fuel cells intended to be used as portable power sources, it is highly desirable that the operation of the fuel cell be passive, with no requirement for forcing either hydrogen fuel or air through the fuel cell.

[0003] A suitable passive fuel cell which meets the requirements for a portable power supply is shown and described in U.S. patents 5,514,486 to Wilson and 5,595,834 to Wilson, et al. The fuel cells shown in these patents are formed of a stack of unit cells, distributed along a common axis. The fuel cell components include a polymer electrolyte membrane, an anode and a cathode contacting opposite sides of the membrane, and fuel and oxygen flow fields contacting the anode and cathode, respectively, with the components defining an annular region therethrough along the axis that acts as a fuel flow channel. A fuel distribution manifold is mounted within the annular region to distribute fuel to the flow field in each of the

unit cells. A single bolt through the annular region clamps the unit cells together. During operation of fuel cells of this type, the fuel cell consumes hydrogen supplied from a fuel source and oxygen supplied from the ambient air, and produces electricity and water. In a completely passive fuel cell of this type, there are no pumps to recirculate or remove the reactants or the reaction products. The hydrogen fuel is introduced into the hydrogen supply flow channel of the fuel cell through an inlet, but there is no outlet for the water vapor and other inert gases which accumulate within the fuel cell except through the active membranes of the cell units. Water vapor and inert gas tend to accumulate at the closed end of the hydrogen flow channel (or in the middle of the channel where hydrogen gas is fed in from both ends), with the result that the hydrogen gas is diluted in the region of the closed end (or middle) of the fuel cell and the voltages produced by the unit cells near the closed end (or middle) are consequently reduced. The inert gases most commonly include nitrogen, which can diffuse into the fuel cell through the polymer electrolyte membrane, and possibly also other impurity gases which are fed into the fuel cell with the hydrogen fuel gas.

[0004] A conventional method of removing the accumulated inert gases and water vapors from a passive fuel cell is to provide a manually operated purge valve at the closed end of the hydrogen supply channel, with the valve periodically being opened to release the inert gases and vapors. See, e.g., Larminie and Dicks, Fuel Cell Systems Explained (book), John Wiley & Sons, 2000, p. 101. The provision of a valve to be manually operated to allow purge of the fuel gases means that proper operation of the fuel cell is dependent on human intervention. If the opening of the valve to purge the system is not carried out often enough, the efficiency of the fuel cell will degrade, whereas if the system is purged too often an unnecessary volume of fuel gas will be wasted.

SUMMARY OF THE INVENTION

[0005] In accordance with the invention, a fuel cell system supplies fuel to a polymer electrolyte fuel cell to substantially equalize the concentration of fuel gas,

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such as hydrogen, through the flow channel of the fuel cell while mixing inert gases and water vapor accumulated in the flow channel to avoid localized concentrations of inert gases and water vapors. The supply of fuel gas to the fuel flow channel and the mixing of gases in the channel is carried out utilizing relatively simple and inexpensive components and without requiring fans or pumps to force fuel gas through the fuel cell.

190001 The fuel cell system of the invention includes a polymer electrolyte fuel cell having a plurality of fuel cell units arranged in a stack. The fuel cell units include a polymer electrolyte membrane, and the stack of fuel cell units have an inner periphery defining a fuel flow channel through which fuel can flow through the stack. A first fuel supply inlet is formed at one end of the stack and a second fuel supply inlet is formed at another end of the stack, each in communication with the fuel flow channel and through which fuel gas can be directed to the fuel flow channel. The fuel cell system further includes a distribution valve having an input and two outputs, one of the outputs connected to the first fuel supply inlet and the other of the outputs connected to the second fuel supply inlet. The input of the distribution valve can be connected to a source of fuel gas such as a canister of hydrogen under pressure. A controller is connected to the distribution valve to switch it at a selected frequency between two positions, a first in which the first of the outputs is connected to the input of the valve and a second in which a second of the outputs is connected to the input of the valve. When the distribution valve is in its first position, fuel is supplied to the first fuel supply inlet through the fuel flow channel to distribute fuel gas, e.g., hydrogen, throughout the entire fuel flow channel. Fuel gas is not provided to the second fuel supply inlet in this position. In the second position of the distribution valve, fuel gas is provided to the second fuel supply inlet and through the fuel flow channel and fuel gas is not provided to the first fuel supply inlet. The distribution valve is switched between its two positions at a frequency that may be selected to adequately provide distribution of fuel gas within the fuel supply channel and mixing of the inert gases and water vapor within the channel. The switching of the distribution valve may take place at a relatively

slow frequency (e.g., the valve may be maintained in each of its two positions for a minute or more), with the switching frequency preferably optimized for the particular fuel cell utilized in the system.

[0007] As the fuel gas is supplied under pressure alternately to the two inlets to the fuel cell, the fuel gas flows through the fuel supply channel first in one direction and then in another. This alternating flow of fuel gas helps to mix the gases within the internal fuel flow channel. This mixing may be enhanced by flow of gas entirely through the fuel flow channel of the fuel cell and into the fuel supply line which, at that time, is not supplying fuel gas to the fuel cell, with the capacity of the closed fuel supply line functioning as an accumulator for gas flow.

[0008] Further objects, features and advantages of the invention will be apparent from the following detailed description when taken in conjunction with the accompanying drawings.

BRIEF DESCRIPTION OF THE DRAWINGS

[0009] In the drawings:

- [0010] Fig. 1 is a schematic diagram of the fuel cell system of the invention with switched fuel gas delivery.
- [0011] Fig. 2 is a schematic diagram of the electrical components of the fuel cell system.
- **[0012]** Fig. 3 is an electrical circuit diagram of the controller for the fuel cell system.
- [0013] Fig. 4 is a graph illustrating the timing cycle for the switching of fuel gas delivery to the two ends of the fuel cell in accordance with the invention.
- [0014] Fig. 5 is a simplified cross-sectional view of an exemplary passive air breathing fuel cell which may be utilized in accordance with the invention.

- [0015] Fig. 6 is a detailed cross-sectional view of the layers forming a cell unit in the fuel cell of Fig. 5.
- [0016] Fig. 7 are graphs showing fuel cell output current over time with a constant fuel cell output voltage of 12 volts in a conventionally supplied fuel cell.
- [0017] Fig. 8 are graphs of output current at a constant output voltage of 12 volts for the fuel cell system with switched fuel gas delivery of the present invention.
- [0018] Fig. 9 are graphs of output current at a constant output voltage of 13 volts for a conventional fuel cell.
- [0019] Fig. 10 are graphs of output current at a constant output voltage of 13 volts for a fuel cell system with switched fuel gas delivery in accordance with the invention.
- [0020] Fig. 11 are graphs of output current at a constant output voltage of 13 volts for the fuel cell system of the invention run over a longer period of time.

DETAILED DESCRIPTION OF THE INVENTION

governments with reference to the drawings, a schematic diagram of a fuel cell system with switched fuel gas delivery is shown generally at 10 in Fig. 1. The fuel cell system 10 has a fuel cell 11 that may be of conventional design, an example of which is described further below, which has two fuel supply ports 12 and 13 located at the two opposite ends of a fuel supply channel or manifold 15 that runs through the fuel cell 11. A supply 17 of fuel gas, e.g., a canister of hydrogen gas under pressure, supplies the fuel gas through a supply line 18 to a switchable valve 19. The switching of the valve 19 is controlled by output signals from a controller 20, which controls the valve 19 to switch to supply gas from the supply line 18 either to first supply line 21 leading to the first input port 12 or to a second supply line 22 leading to the second input port 13. A T-fitting 23 may be mounted in the line 22

for connection to a manually operated purge valve 24 which can be opened to allow manual purge of the hydrogen within the fuel cell 11, if desired.

[0022] The valve 19 and the controller 20 are preferably electrically actuated utilizing power supplied from the fuel cell 11. As illustrated in the electrical diagram of Fig. 2, the fuel cell 11 provides DC output power on output lines 25 to a load 26, and the power on the lines 25 may be connected in parallel with the load 26 to the controller 20 which selectively activates a solenoid within the valve 19. An exemplary controller 20 for the system is shown schematically in Fig. 3 and includes a voltage regulator 27 (e.g., Tandy Corp. Radio Shack Part # 276-1771A; etc.), which receives a DC voltage from the fuel cell and supplies a regulated DC voltage to the solenoid valve 19 and to a timer 28 (e.g., a 555 timer such as SE555 from Phillips Semiconductors), the output waveform of which is controlled by the values of resistors R_a and R_b and a capacitor C₁, as illustrated by the waveform of Fig. 4. The output of the timer 28 is provided to the base of a transistor 29 which is connected to the valve coil 30 of the solenoid distribution valve 19. The solenoid valve 19 may be a commercially available three-way valve, (e.g., AL4312 three-way universal valve available from SmartLook). The present invention is not limited to the solenoid valve shown for illustration or to an electronic controller. A latching type solenoid valve may be used which switches positions when it receives an activation signal but does not continuously draw power in either position. An example of such a valve is the Angar Scientific AL4312L valve. A valve may also be utilized that uses the available fuel gas pressure to switch directions at selected intervals. This type of valve may utilize an external pneumatic control system or the controller may be built into the valve.

[0023] For exemplification, the present invention may be utilized with the type of passive air breathing fuel cell shown in the aforesaid U.S. Patents No. 5,514,486 and 5, 595,834, although it is understood that the invention may be utilized with other types of fuel cell structures. A simplified view of the type of fuel cell described in the aforesaid patents is shown in cross-section generally in Fig. 5.

The fuel cell 11 has a plurality of fuel cell units 31 mounted together in stacked relation, with each of the fuel cell units 31 having an (preferably circular) outer periphery 33 and an (preferably circular) inner periphery 34 which defines a fuel flow channel 15. An upper current collector plate 36 is electrically connected to the uppermost one of the fuel cell units 31, and a lower current collector plate 37 is electrically connected to the lowermost of the fuel cell units 31. An upper end plate 43 is engaged against the upper current collector plate 36 (preferably with a layer of electrical insulation between the end plate and the current collector), and a lower end plate 44 is engaged against the lower current collector 37 (preferably separated therefrom by a layer of insulation). The fuel cell units 31 are preferably annular in shape and symmetrical about a common axis. A bolt 46 extends through the flow channel 15 and through the upper end plate 43 and lower end plate 44, and an upper nut 47 and a lower nut 48 are threadingly engaged to the bolt 46 to press against the end plates 43 and 44, respectively, to compress the stacked fuel cell units 31 and diffusion cell units 39 together. The fuel supply inlet or port 12 is formed in the top nut 47 and extends to the top end of the fuel flow channel 15 to direct hydrogen fuel therethrough to the fuel flow channel 15. The lower fuel supply inlet or port 13 is formed in the lower nut 48 and extends to the bottom end of the fuel flow channel 15. A protective shell (not shown), with openings to allow gases to flow freely, may be mounted around the stack of fuel cell units to protect them from damage.

[0024] A more detailed view of the fuel cell units 39 of the fuel cell is shown in Fig. 6. As illustrated therein, each of the cell units 39 includes a membrane electrode assembly 55, a first upper layer of diffuser material 56, a second upper layer of diffuser material 57 above the layer 56 on the cathode side, and a lower layer of diffuser material 58 on the anode side of the membrane 55 opposite to the side at which the diffuser material layers 56 and 57 are located. The use of a thin layer 56 and a thick layer 57 of diffuser material is shown, but it is understood that a single layer (or three or more layers) may be utilized. Similarly, the diffuser layer 58 may be a single layer or multiple layers. An inner ring gasket 60 engages with the membrane electrode assembly 55 to seal off the inner periphery

of the cell unit from the flow channel 35 except at the exposed inner periphery of the lower diffuser layer 58. A ring shaped outer gasket 61 is engaged with the membrane 55 at the outer periphery of the diffusion cell unit and seals off the bottom diffuser layer 58 from the outside air. A gas impermeable (e.g., a metal) cell divider 63 is mounted between adjacent cell units and engaged with the top diffuser layers 57 and 58 to separate the cell units and prevent flow therebetween.

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[0025] The membrane 55 may be formed as a polymer electrolyte membrane of the type described in the aforesaid patents Nos. 5,514,486 and 5,595,834, e.g., a polymer electrolyte membrane catalyzed with a thin film platinum catalyst layer (e.g., about 0.15 mg Pt/cm²/electrode, as described in patents Nos. 5,211,984 and 5,234,777) sandwiched between uncatalyzed ELAT backings (E-Tek, Inc., Natwick, Massachusetts) as the diffuser layers 56 and 58. The material of the diffuser layer 58 may also be formed, e.g., of macroporous electrically conductive material such as carbon fiber based paper available under the name Spectracarb 2050 available from Spectracorp, Inc., Lawrence, Massachusetts.

[0026] The fuel cell stack may be held in compression using mechanical means other than the threaded center bolt 46. An example is a conventional clamp external to the cell units or threaded bolts, etc., which are capable of holding the cell units together. If desired, a center manifold (not shown) may be mounted within the fuel flow channel 15 around the bolt 46 to distribute fuel and to help distribute moisture evenly through the length of the flow channel 15. The center manifold may be formed as described in patents Nos. 5,514,486 and 5,595,834, or in other manners as desired.

[0027] When a conventional fuel cell having the configuration of Fig. 5 is operated, the hydrogen gas travels through the central channel of the stack end to the closed end if gas is fed in from only one end, diffusing radially into the various cells. These cells consume hydrogen to make electricity and produce water as a byproduct. Some of this water diffuses back into the central flow channel and then gets carried along with the flow of hydrogen to the closed end of the stack. This

water slowly diffuses out through the lowermost cells into the atmosphere. As the load (current per cell area) is increased, the rate of water production by the cells exceeds the rate at which it can diffuse out of the cells. The cells which see the most diluted hydrogen tend to drop in performance. This is commonly referred to as "flooding." Flooding will occur at any point in the central channel where there is stagnation of the supply of hydrogen or a build-up of inert species. Where hydrogen is fed in from one end only of the stack, the flooding tends to occur at the bottom or closed end of the flow channel. If the hydrogen is fed in from both ends of the stack (e.g., through hydrogen end feeds in both of the nuts 47 and 48), the flooding would occur at cell units approximately in the middle of the stack. The conventional solution to this problem has been to provide a manually operated valve such as the purge valve 24 which can be periodically opened to discharge accumulated water vapor and inert gas. As noted above, the use of a purge valve has the significant disadvantage that it relies on the user of the fuel cell to operate the valve at appropriate times.

[0028] Fig. 7 shows the decline in output current over time in a conventional fuel cell of the type shown in Figs. 5 and 6 when supplied with fuel constantly at one of the input ports. As illustrated therein, when the fuel cell is operated at a constant output voltage of 12 volts, the output current declines significantly over a relatively short period of time (15% decline in ½ hour as shown).

[0029] Fig. 8 illustrates output current over time for a fuel cell system of the invention operated at a constant current of 12 volts. As illustrated therein, the switching of the fuel gas from one input port to the other of the fuel cell at a rate of approximately one cycle every two minutes virtually eliminates the deterioration of the output current as a function of time. A small cyclic fluctuation of the load current is seen in Fig. 8 which is due to the current that the solenoid valve requires when it is in the electrically activated position. When the controller current is off, the valve is in its un-activated position, allowing the output load to draw the

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maximum current. When the controller current is on, the valve is in the actuated condition and the amount of current available to the output load is reduced by the amount drawn by the solenoid valve. Even when current is being drawn by the valve, it is seen from a comparison of Figs. 7 and 8 that the output load current utilizing the invention (Fig. 8) is higher than the output load current without the invention (Fig. 7). As noted above, a latching valve may be utilized that does not draw power except when it is switched.

[0030] Fig. 9 shows output current as a function of time with a constant output voltage of 13 volts from the conventional fuel cell over a period of approximately one hour, showing a continuous significant decline in output current. Fig. 10 illustrates the output current of the fuel cell system of the invention at a constant output voltage of 13 volts over a period of over two hours, showing that the output current remains substantially constant. Fig. 11 illustrates the output current over an even longer period of time from the fuel cell system of the invention, showing that the fuel cell output current remains substantially constant after an initial small decline, over a period of time of almost one day, indicating that the output current from the device has completely stabilized.

[0031] It is understood that the invention is not limited to the embodiments described herein, but embraces all such forms thereof as come within the scope of the following claims.

1

CLAIMS

What is claimed is:

1.

2	(a) a polymer electrolyte fuel cell comprising a plurality of fuel cell units
3	defining an outer periphery and arranged in a stack, the fuel cell units including a
4	polymer electrolyte membrane, wherein the stacked fuel cell units have an inner
5	periphery defining a fuel flow channel through which fuel can flow through the
6	stack, an electrically conductive current collector electrically connected to a fuel cell

A fuel cell system comprising:

- 7 unit at one end of the stack of fuel cell units and an electrically conductive current
- 8 collector electrically connected to a fuel cell unit at another end of the stack of fuel
- 9 cell units, and a first fuel supply inlet at one end of the stack and a second fuel
- supply inlet at another end of the stack through which fuel can be directed to the
- 11 fuel flow channel;
- 12 (b) a distribution valve having an input and two outputs and switchable
- between two positions, a first position in which a first of the outputs is connected to
- 14 the input and a second position in which a second of the outputs is connected to the
- input, and a fuel supply line extending from a first of the outputs to the first fuel
- supply inlet and another fuel supply line extending from the second of the outputs to
- the second fuel supply inlets of the fuel cell, the input of the distribution valve
- 18 connectable to the source of fuel gas; and
- 19 (c) a controller connected to the distribution valve to switch it between
- 20 its two positions at a selected frequency.
- 1 2. The fuel cell system of Claim 1 wherein the distribution valve is
- 2 electrically operated in response to an electrical control signal, and wherein the
- 3 controller provides an electrical output signal that switches at a selected rate to
- 4 switch the position of the distribution valve.
- 1 3. The fuel cell system of Claim 2 wherein the distribution valve is a
- 2 solenoid operated three-way valve.

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1 4. The fuel cell system of Claim 2 wherein the controller includes a 2 timer that produces an output signal with a high and low voltage output that switches 3 with a selected period and a selected waveform.

- 5. The fuel cell system of Claim 2 wherein the controller switches the distribution valve at a regular cycle having a period of about two minutes.
- 1 6. The fuel cell system of Claim 1 further including a manually operated 2 purge valve connected in one of the supply lines leading from the distribution valve to one of the fuel supply inlets of the fuel cell.
- 7. The fuel cell system of Claim 1 wherein the controller switches the distribution valve with a regular cycle having a selected period with the valve being in each of its positions approximately an equal length of time during each cycle.
- 8. The fuel cell system of Claim 1 wherein the fuel cell units are annular in shape, having a circular outer periphery and a circular inner periphery defining walls of the fuel flow channel, the fuel cell units stacked along a central axis extending between the fuel supply inlets at each end of the stack.
- 9. 1 The fuel cell system of Claim 1 wherein the fuel cell units each 2 include a polymer electrolyte membrane between top and bottom layers of diffuser 3 material, a gasket mounted at the outer periphery of the fuel cell unit to prevent air 4 flow from outside the fuel cell to one of the top or bottom diffuser layers and a 5 gasket mounted at the inner periphery of the fuel cell unit to prevent fuel flow from 6 the flow channel to the other of the top and bottom diffuser layers, a cathode at one 7 side of the polymer electrolyte membrane and an anode at the other side of the 8 polymer electrolyte membrane, one of the top or bottom diffuser layers in contact 9 with one of the anode or cathode and the other of the top and bottom diffuser layers 10 in contact with the other of the anode or cathode.

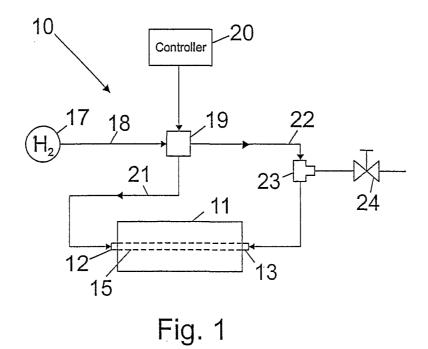
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1	10.	The fuel cell system of Claim 1 further including a canister of
2	hydrogen fuel	gas connected by a fuel supply line to the input of the distribution
3	valve.	

- 1 11. A method of operating a polymer electrolyte fuel cell of the type
 2 having a plurality of fuel cell units arranged in a stack, with the stacked fuel cell
 3 units having an inner periphery defining a fuel flow channel through which fuel can
 4 flow, with first and second fuel supply inlets formed at ends of the stack in
 5 communication with the fuel flow channel, comprising:
- 6 (a) directing fuel gas from a source of gas to the first fuel supply inlet at
 7 one end of the stack to direct fuel therethrough to the fuel flow channel while not
 8 providing flow of fuel to the second fuel supply inlet for a selected length of time;
- 9 (b) then supplying fuel gas from the source to the second fuel supply inlet 10 for a selected length of time while not providing fuel gas to the first fuel supply 11 inlets.
- 1 12. The method of Claim 11 wherein steps (a) and (b) are repeated at a selected frequency.
- 1 13. The method of Claim 12 wherein the period of each cycle of the repetition of steps (a) and (b) is about two minutes.



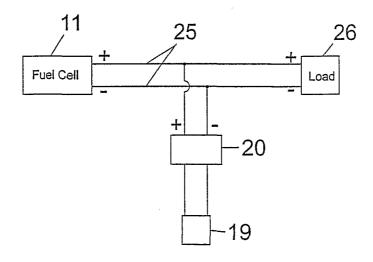
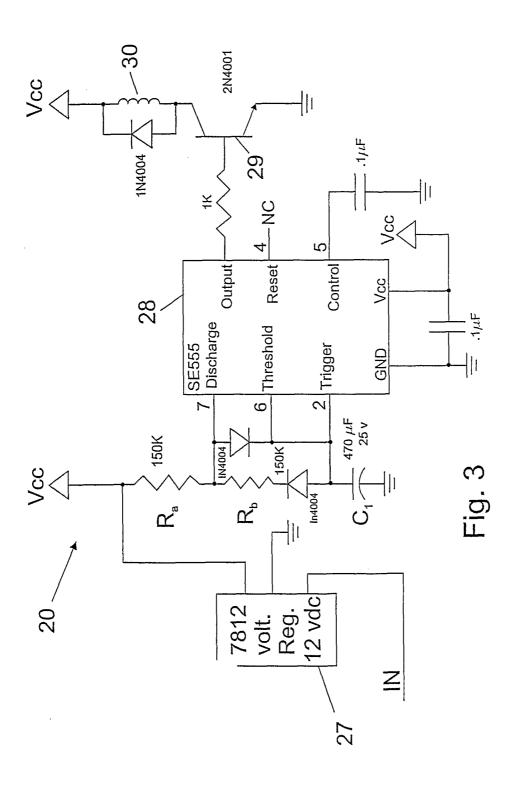
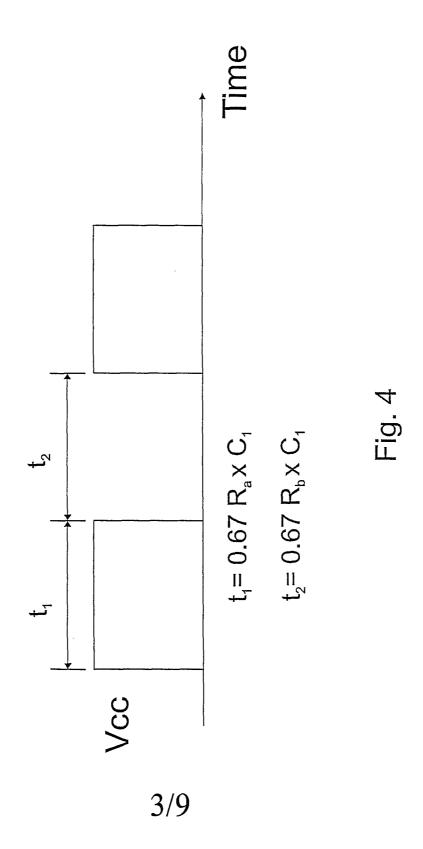


Fig. 2





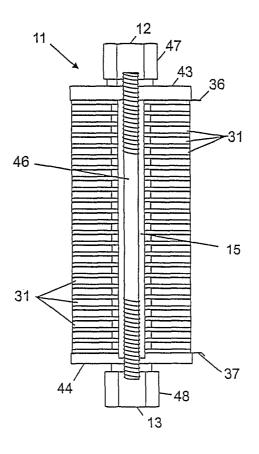


Fig. 5

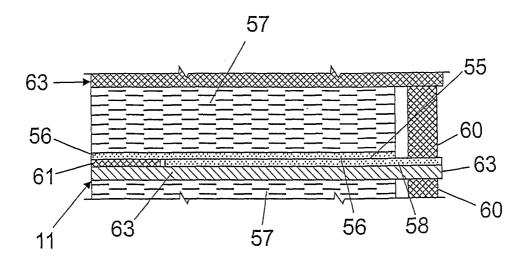
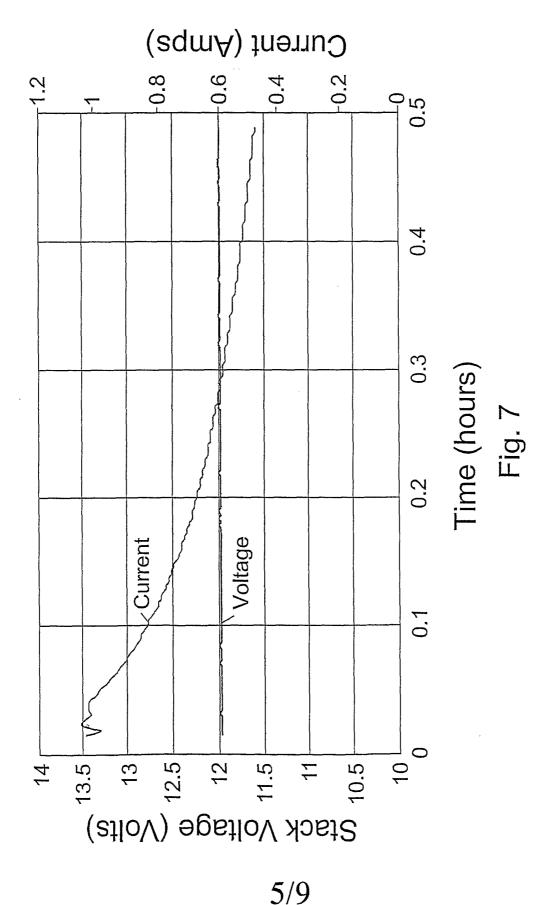
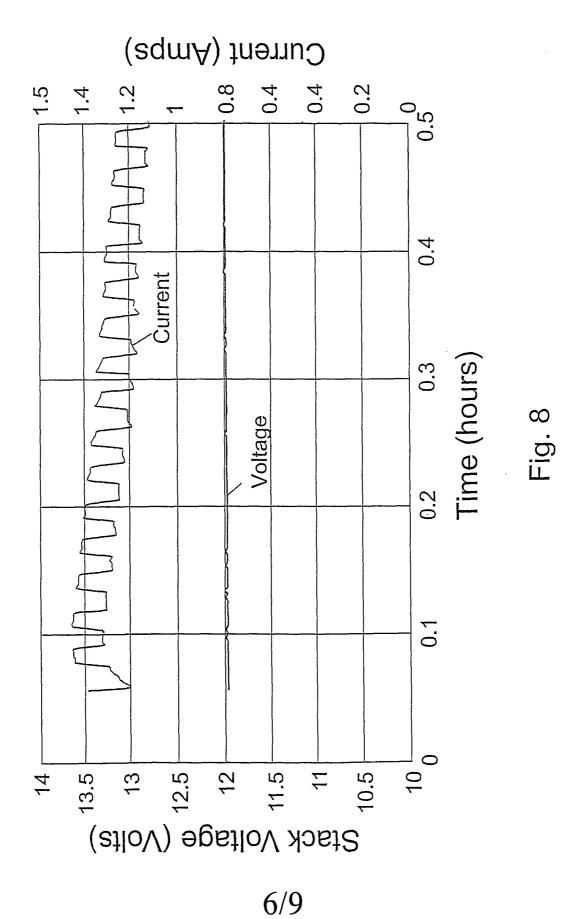
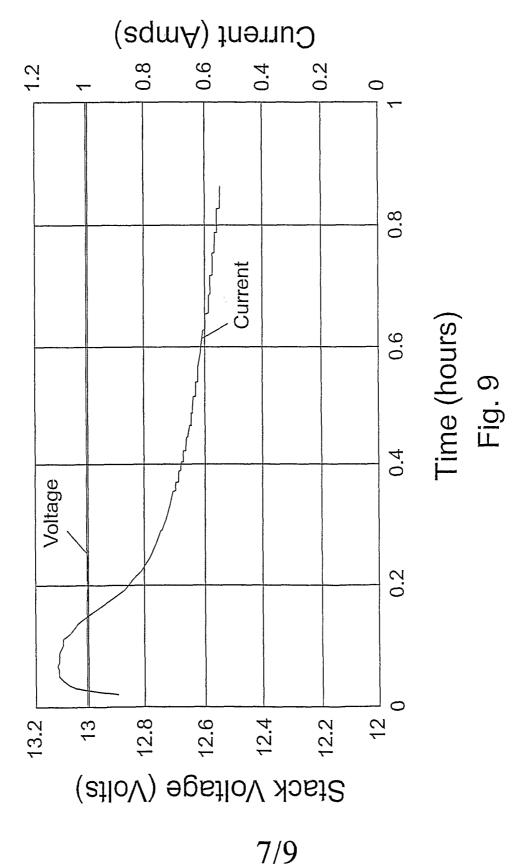


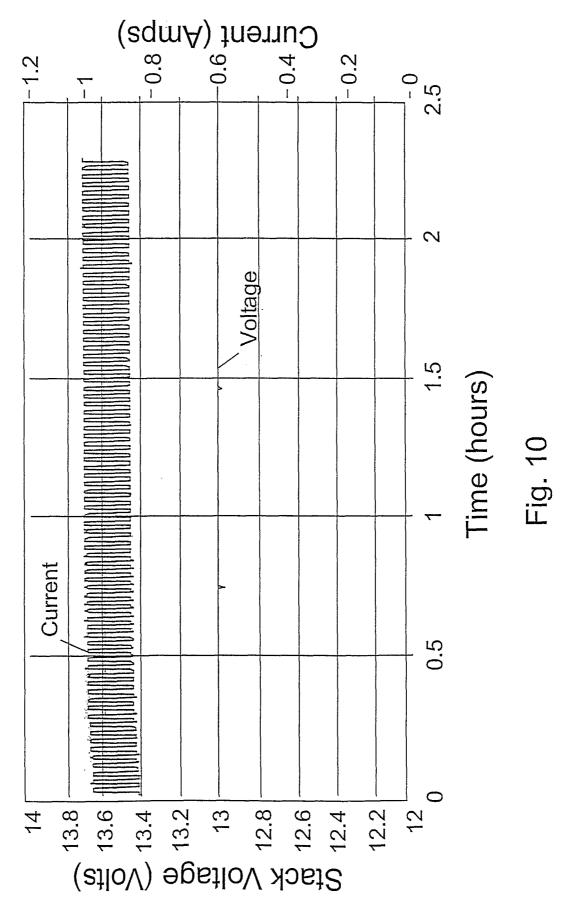
Fig. 6





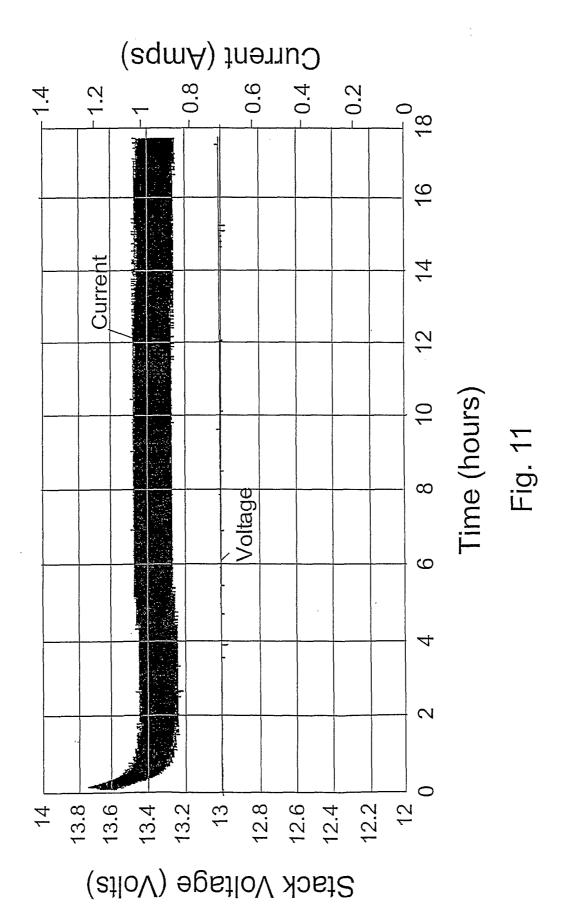


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INTERNATIONAL SEARCH REPORT

International application No.

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A. CLASSIFICATION OF SUBJECT MATTER IPC(7) : H01M 08/00, 08/04, 08/12, 08/10, 02/14 US CL : 429/12, 13, 22, 30, 39 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) U.S.: 429/12, 13, 22, 30, 39							
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) Non-Patent Literature							
C. DOCUMENTS CONSIDERED TO BE RELEVANT							
Category *	Citation of document, with indication, where a		Relevant to claim No.				
Y	US 5,595,834 A (WILSON et. al.) 21 January 1997 column 5, line 6.	(21.01.1997), column 3, line 49 -	1-13				
Y	US 6,063,161 A (KEEFER et. al.) 16 May 2000 (16 14, line 23.	1-13					
Y	US 5,316,869 A (PERRY JR. et. al.) 31 May 1994 column 5, line 47.	1-13					
A	US 6,127,057 A (GORMAN) 03 October 2000 (03.	1-13					
A	US 6,103,411 A (MATSUBAYASHI et. al.) 15 Aug Document	gust 2000 (15.08.2000), The Whole	1-13				
A,P	US 6,376,114 B1 (BONVILLE, JR. et. al.) 23 April Document	1 2002 (23.04.2002), The Whole	1-13				
Further	documents are listed in the continuation of Box C.	See patent family annex.					
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