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(54) **ELECTROCHEMICAL HYDROGEN COMPRESSOR**

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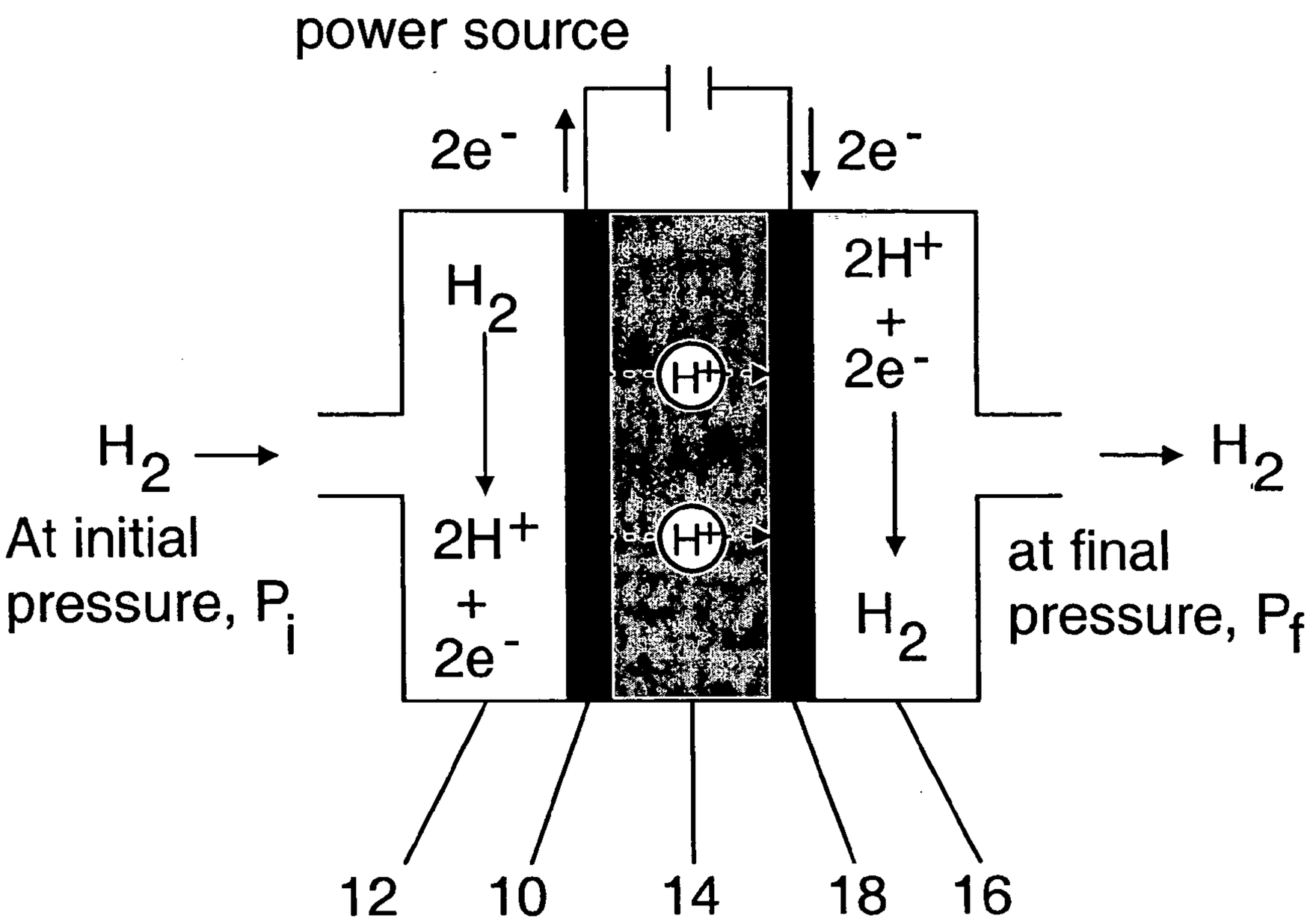
(57) **ABSTRACT**

The invention disclosed relates to an apparatus and process for electrochemical compression of hydrogen. The apparatus comprises a membrane electrolyte cell assembly (MEA), including planar gas distribution plates sandwiching the MEA, the assembly being held together by end-plates, the end-plates having complementary peripheral grooves for seating an intervening seal between the end-plates and the MEA, the end-plate on the anode side further including a hydrogen supply inlet and the end-plate on the cathode side further including a compressed hydrogen outlet. Both single cell and multi-cell assemblies are disclosed. The multi-cell assemblies comprise a plurality of such single cells connected in series, such that the compressed hydrogen from the outlet of a first cell is connected to the hydrogen outlet of the next cell in series, where each cell is electrically isolated from the adjacent cell in the series. The process involves the electrochemical compression of hydrogen in such cells, whereby pressures of up to 12,000 psi are achieved by multi-cell assemblies.



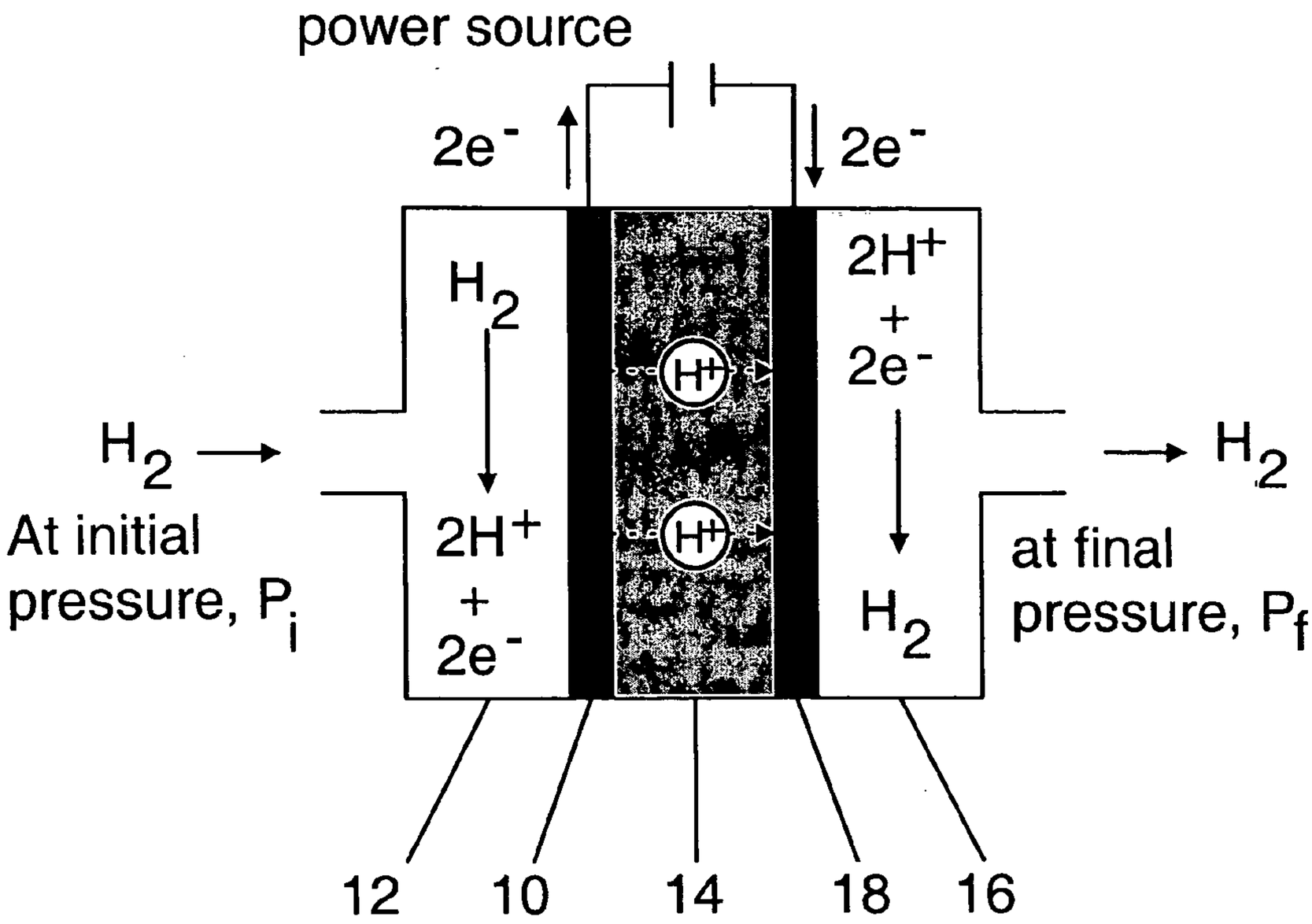

power source
2e-
2e-
H2
2H+
+
2e-
H+
H+
H2
At initial
pressure, Pi
2H+
+
2e-
H2
at final
pressure, Pf
12
10
14
18
16

Fig. 1

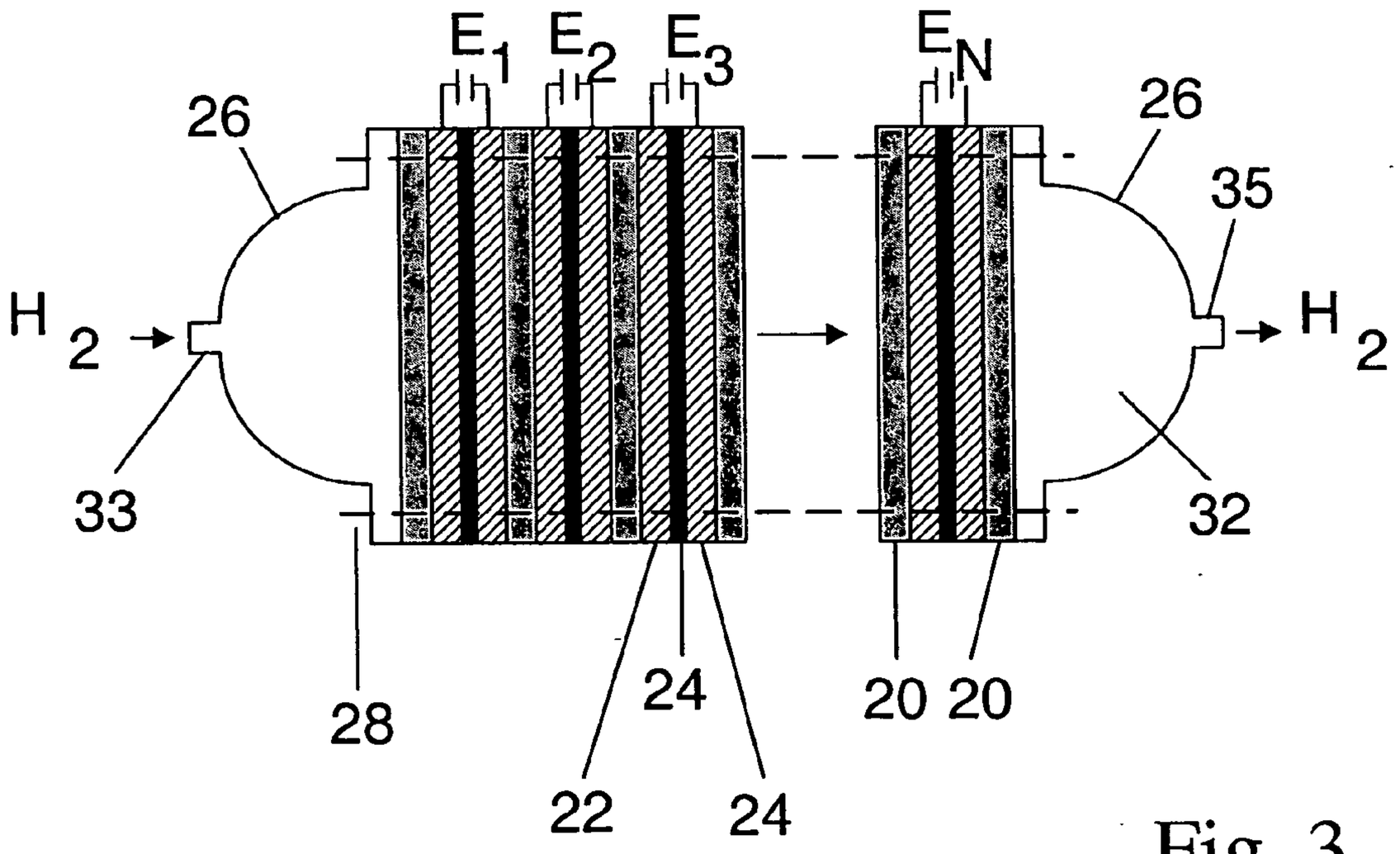

E1
E2
E3
EN
26
26
35
H2
H2
33
32
28
24
20
20
22
24

Fig. 3

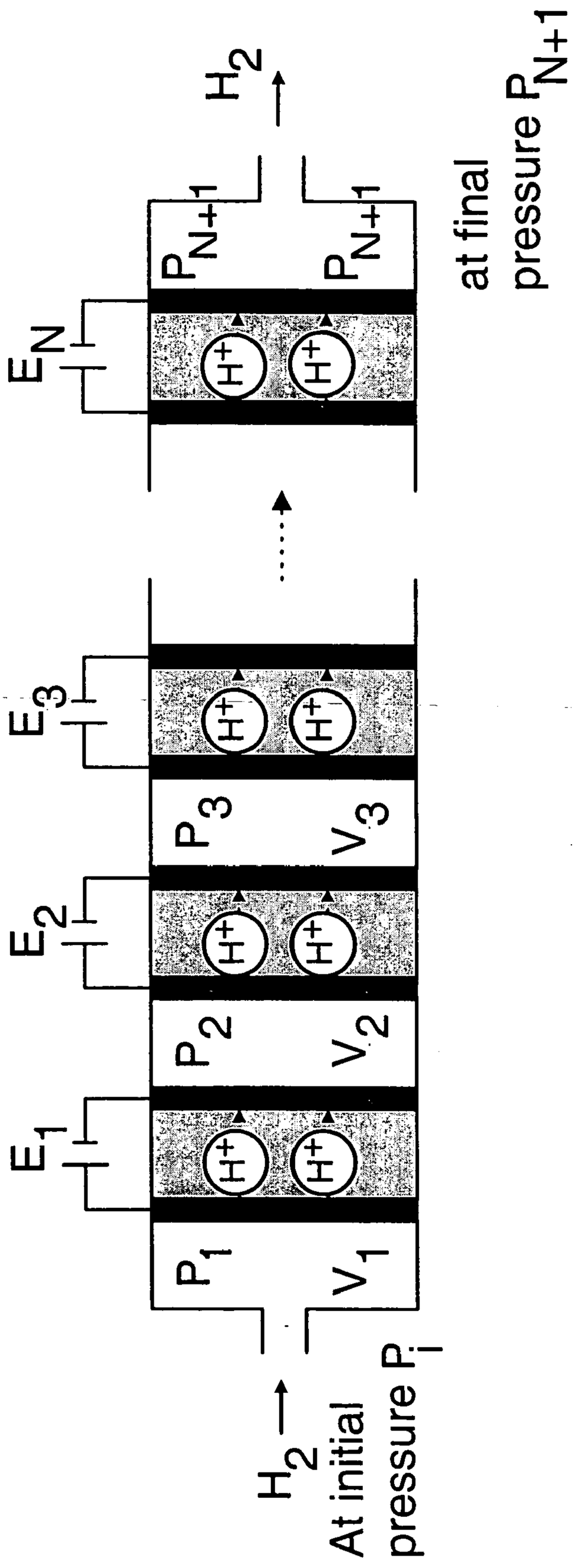
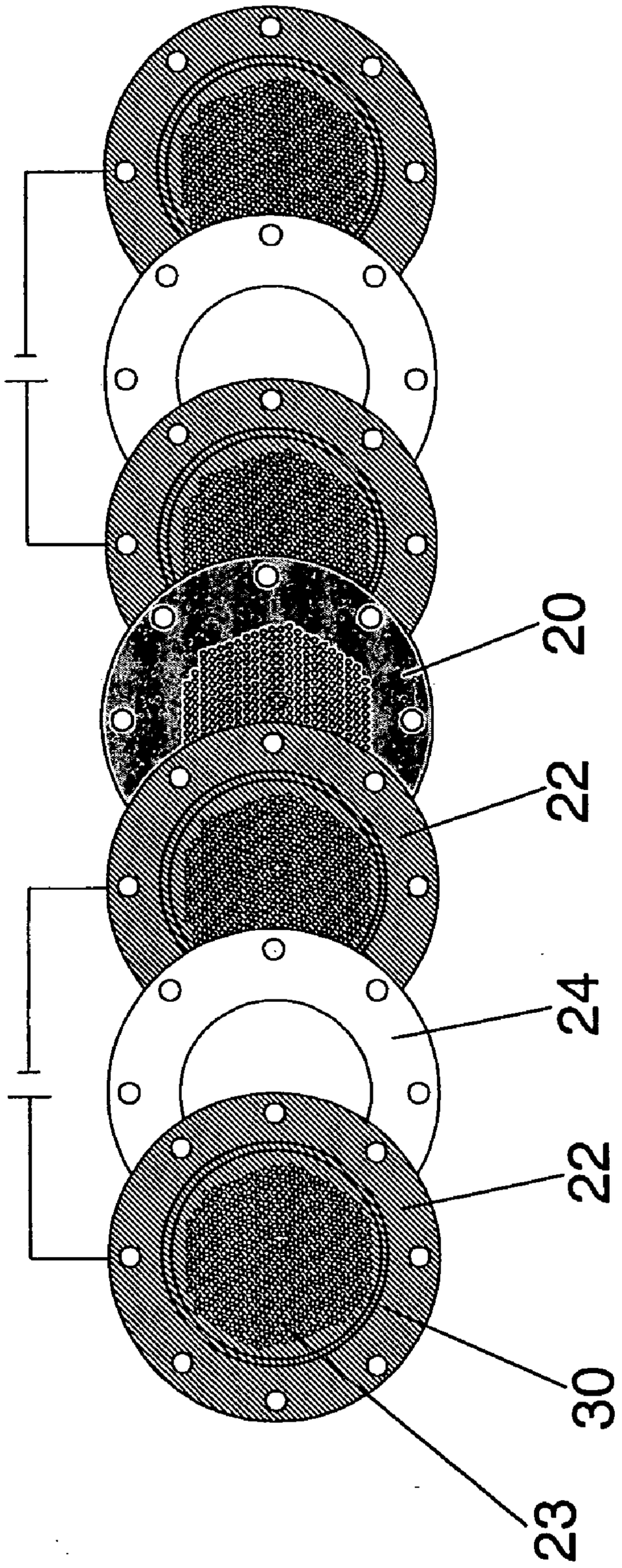

H_2
P_{N+1}
P_{N+1}
at final
pressure P_{N+1}
E_N
H^+
H^+
E_3
P_3
V_3
E_2
P_2
V_2
E_1
P_1
V_1
H_2
At initial
pressure P_I

Fig. 2

Fig. 4



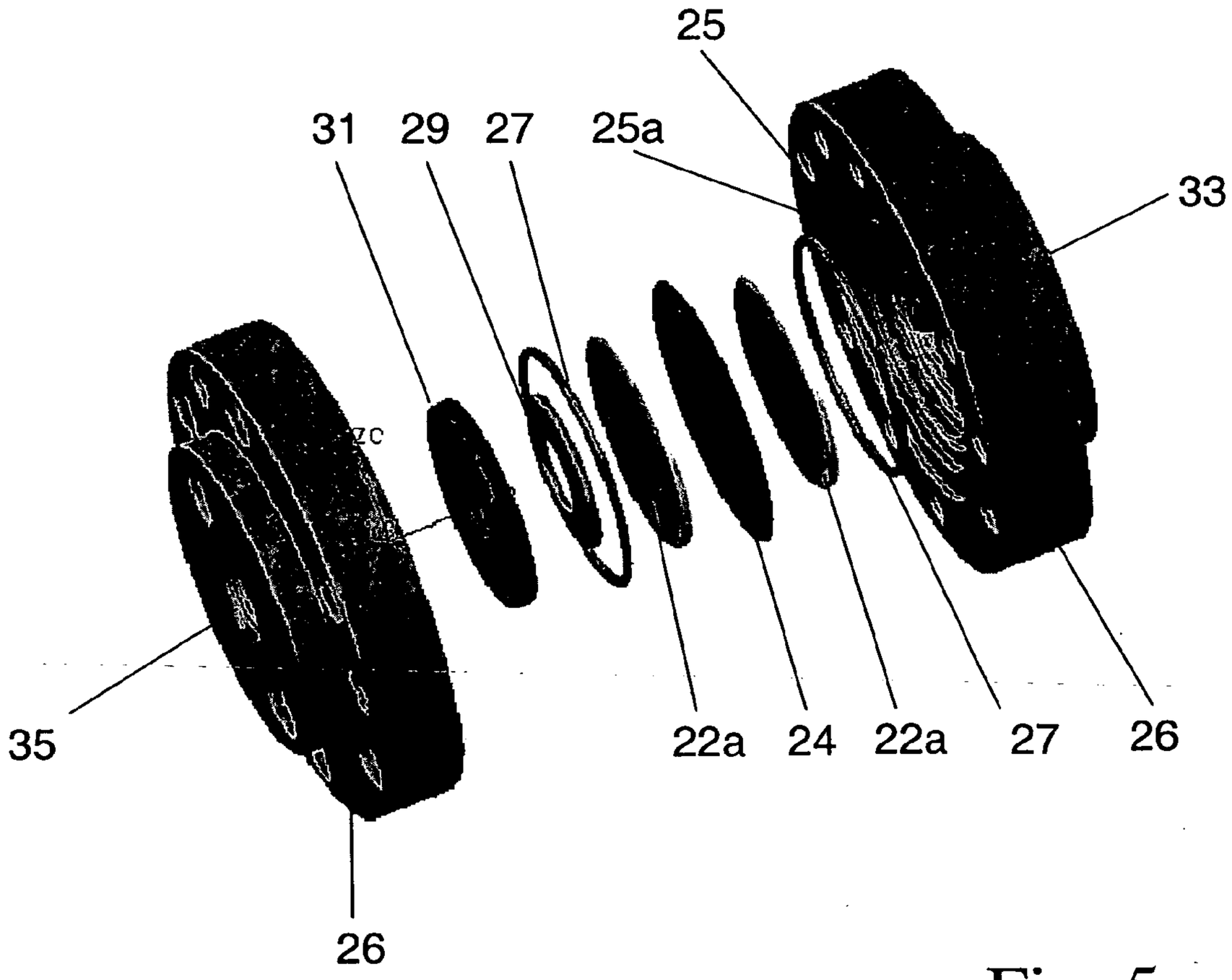


Fig. 5

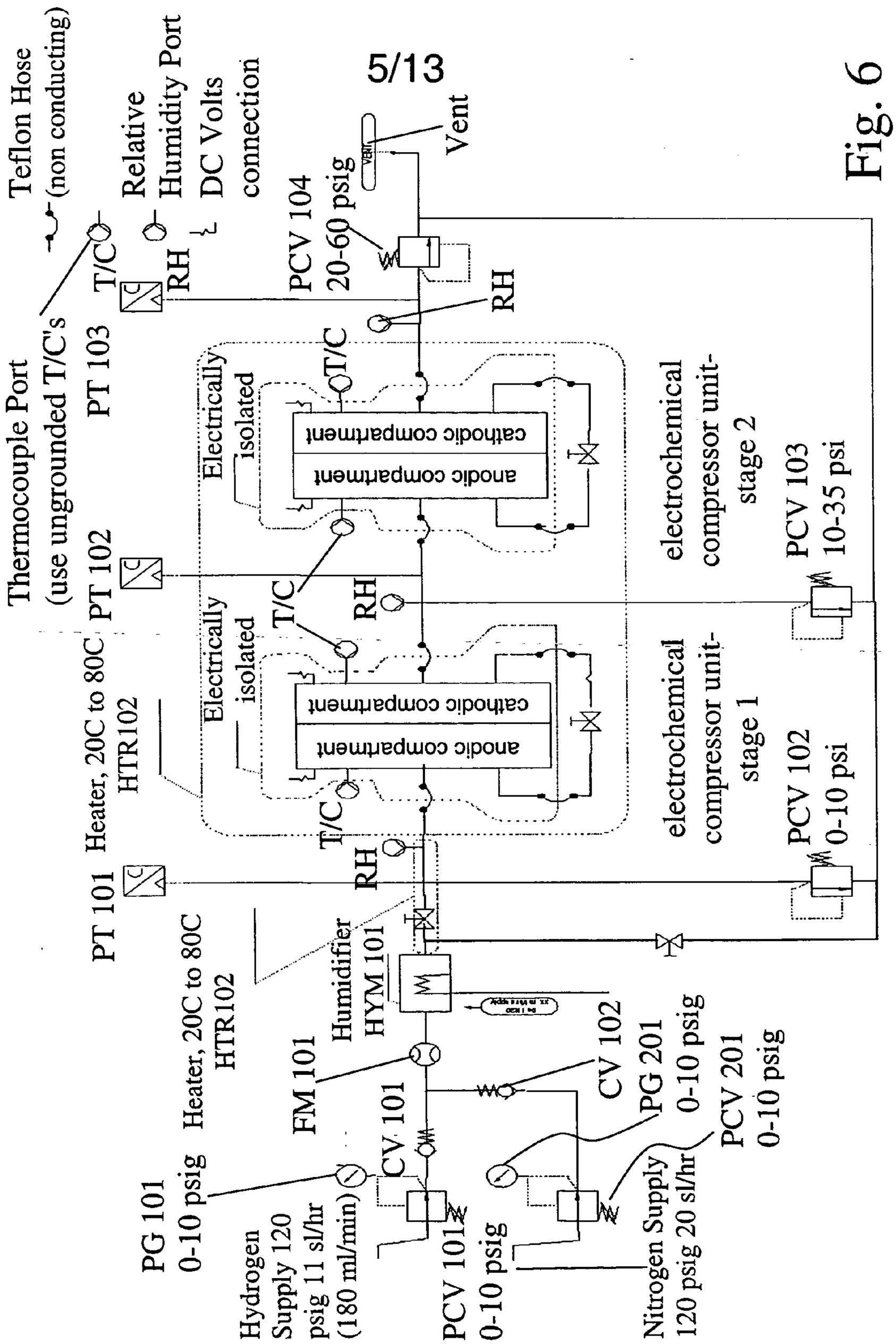

5/13
Teflon Hose
(non conducting)
Thermocouple Port
(use ungrounded T/C's)
PT 103
PT 102
PT 101
T/C
RH
Relative
Humidity Port
DC Volts
connection
Heater, 20C to 80C
HTR102
Heater, 20C to 80C
HTR102
PCV 104
20-60 psig
Vent
Electrically
isolated
anodic compartment
cathodic compartment
electrochemical
compressor unit-
stage 2
electrochemical
compressor unit-
stage 1
PCV 103
10-35 psi
PCV 102
0-10 psi
Humidifier
HYM 101
FM 101
CV 101
CV 102
PG 101
0-10 psig
Hydrogen
Supply 120
psig 11 sl/hr
(180 ml/min)
PCV 101
0-10 psig
PG 201
0-10 psig
Nitrogen Supply
120 psig 20 sl/hr
PCV 201
0-10 psig

Fig. 6

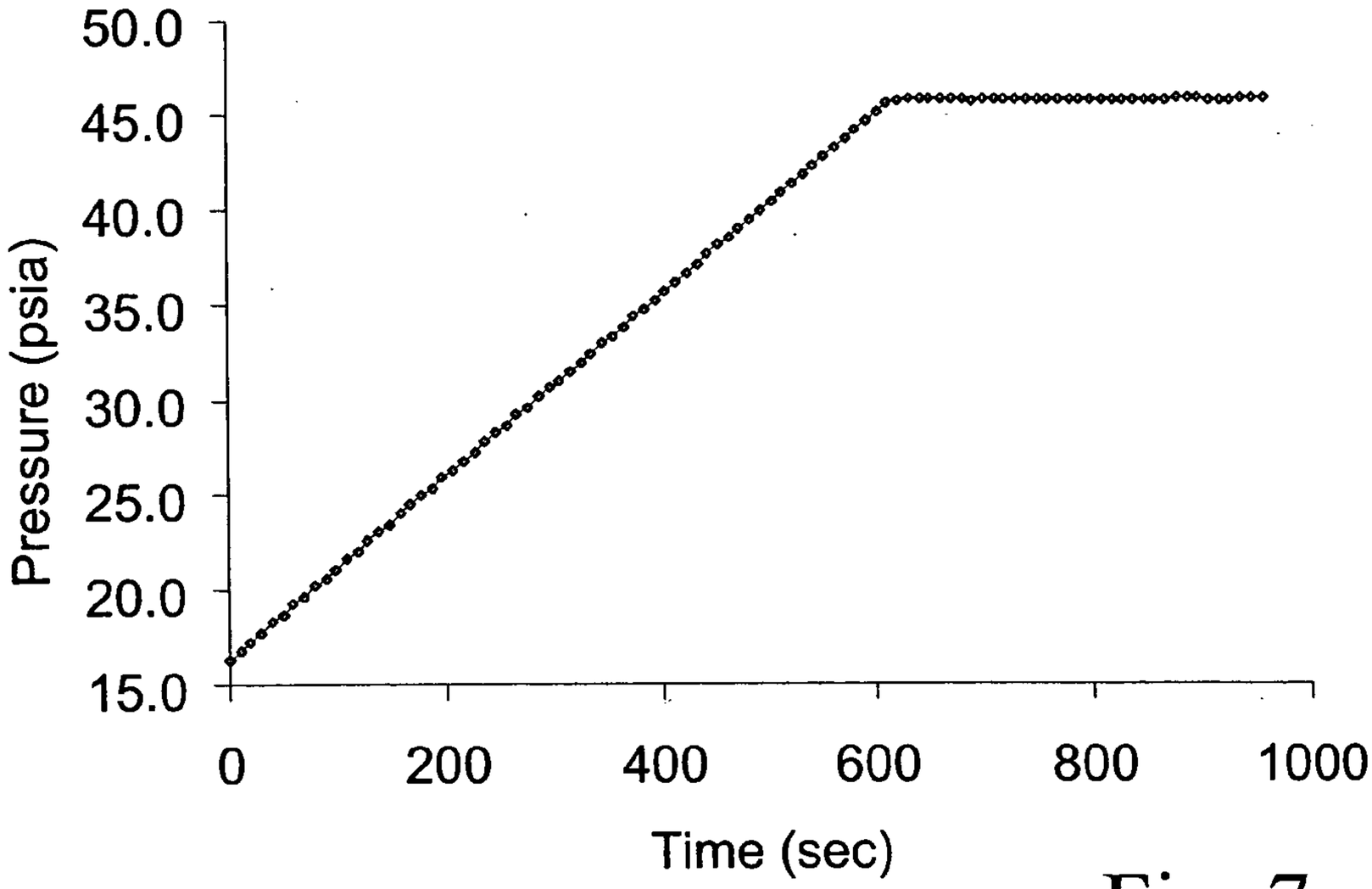

50.0
45.0
40.0
35.0
30.0
25.0
20.0
15.0
Pressure (psia)
0
200
400
600
800
1000
Time (sec)

Fig. 7

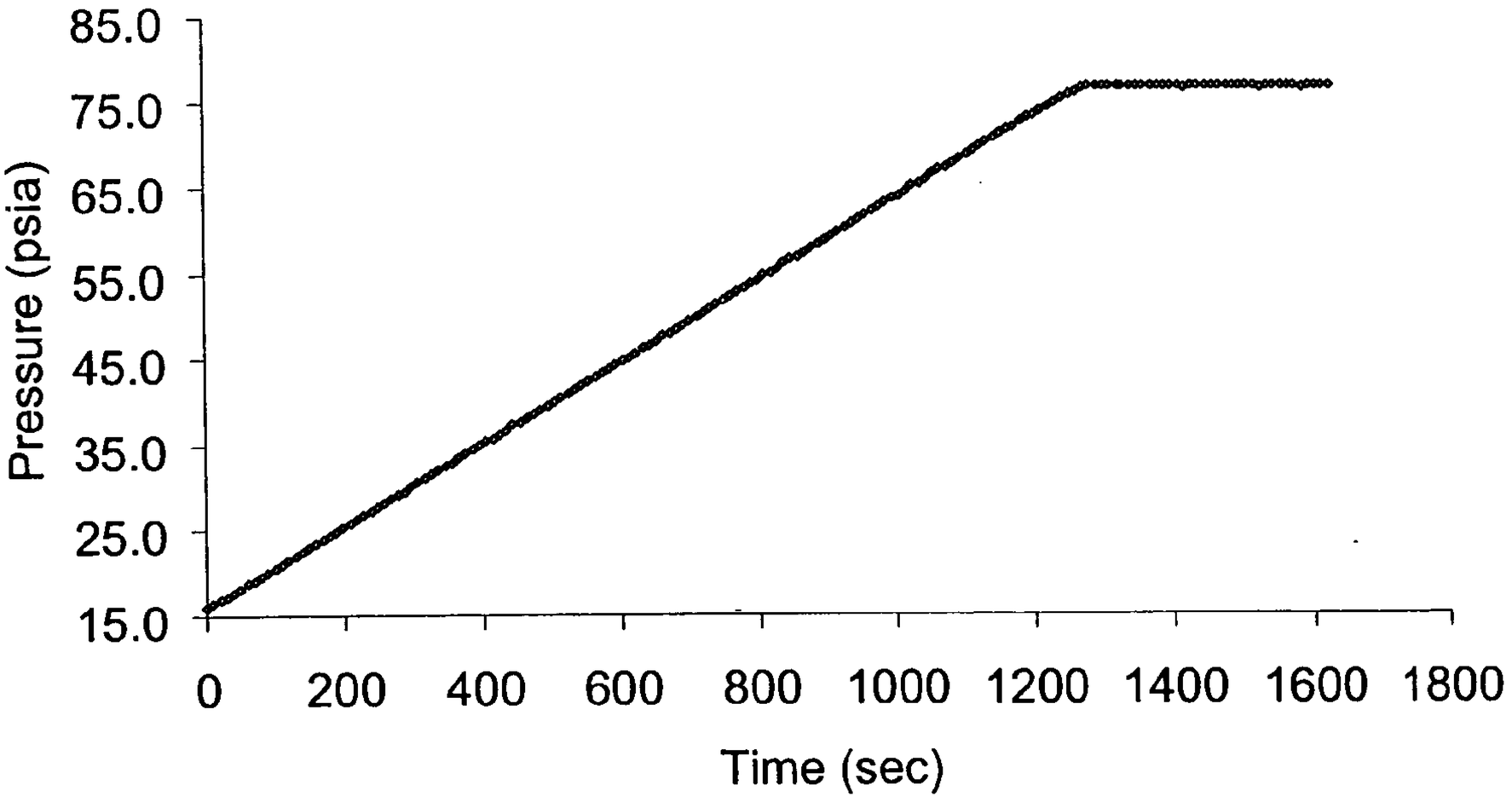

85.0
75.0
65.0
55.0
45.0
35.0
25.0
15.0
Pressure (psia)
0
200
400
600
800
1000
1200
1400
1600
1800
Time (sec)

Fig. 8

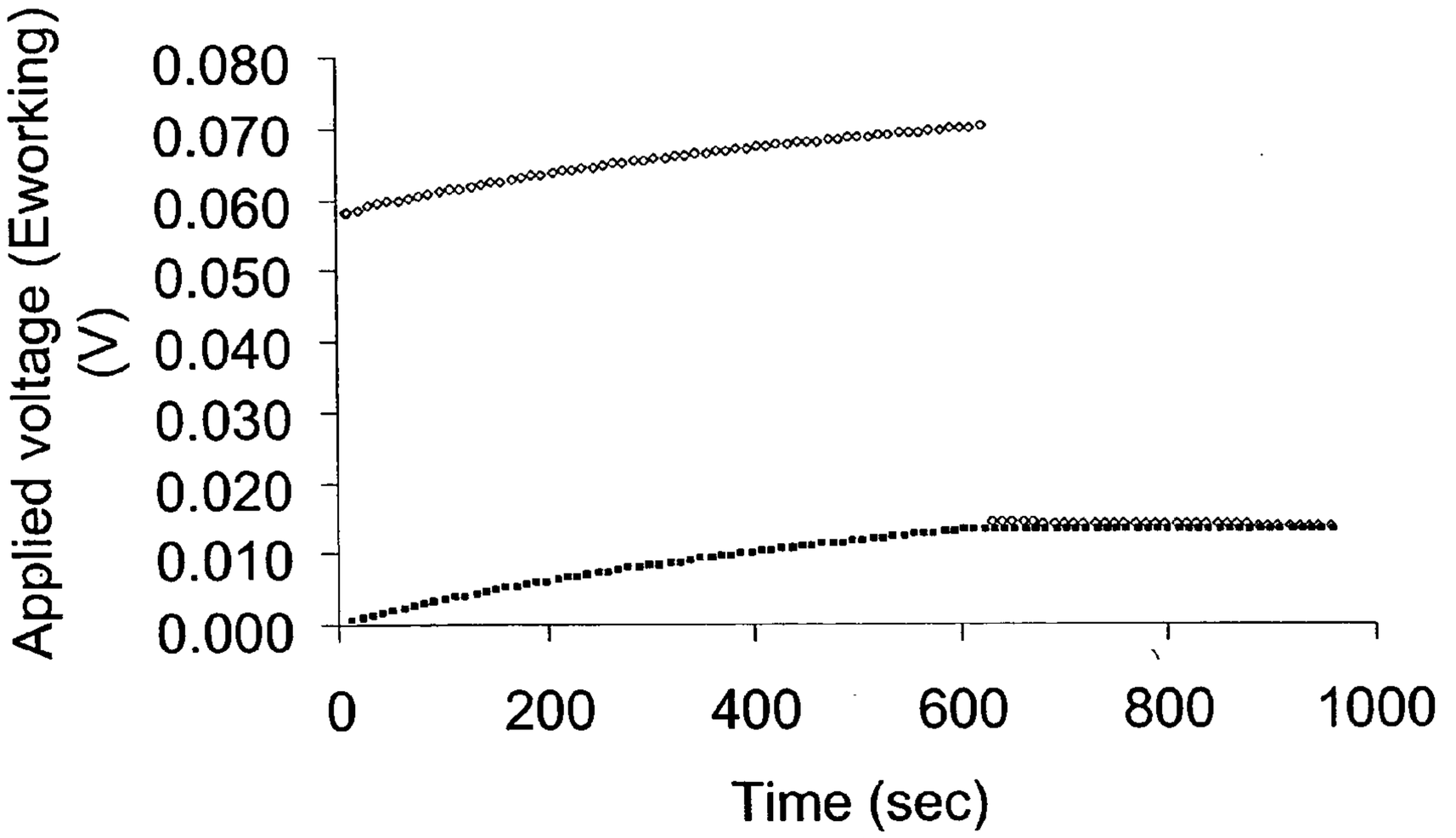

Applied voltage (Eworking) (V)
0.080
0.070
0.060
0.050
0.040
0.030
0.020
0.010
0.000
0
200
400
600
800
1000
Time (sec)

Fig. 9

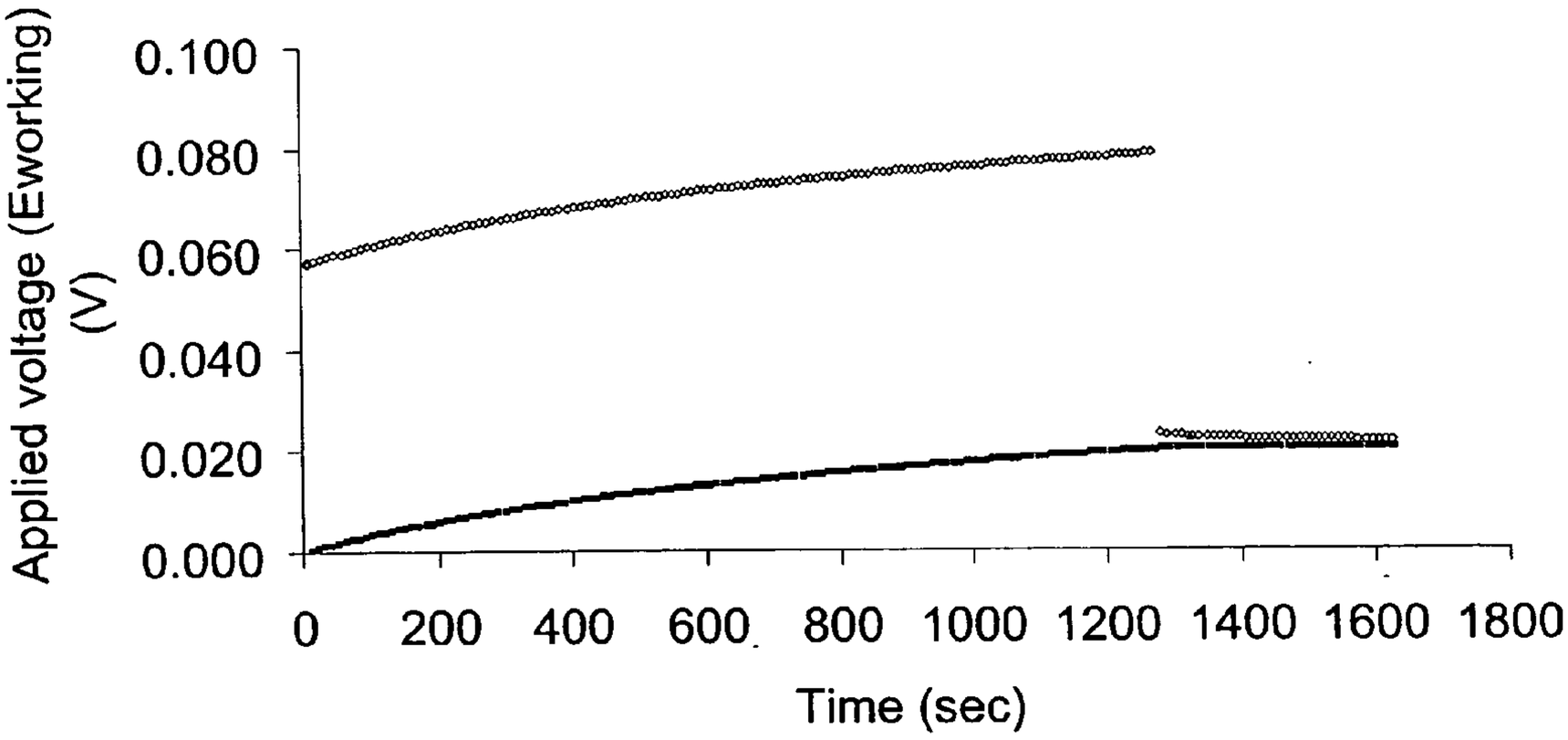

Applied voltage (Eworking) (V)
0.100
0.080
0.060
0.040
0.020
0.000
0
200
400
600
800
1000
1200
1400
1600
1800
Time (sec)

Fig. 10

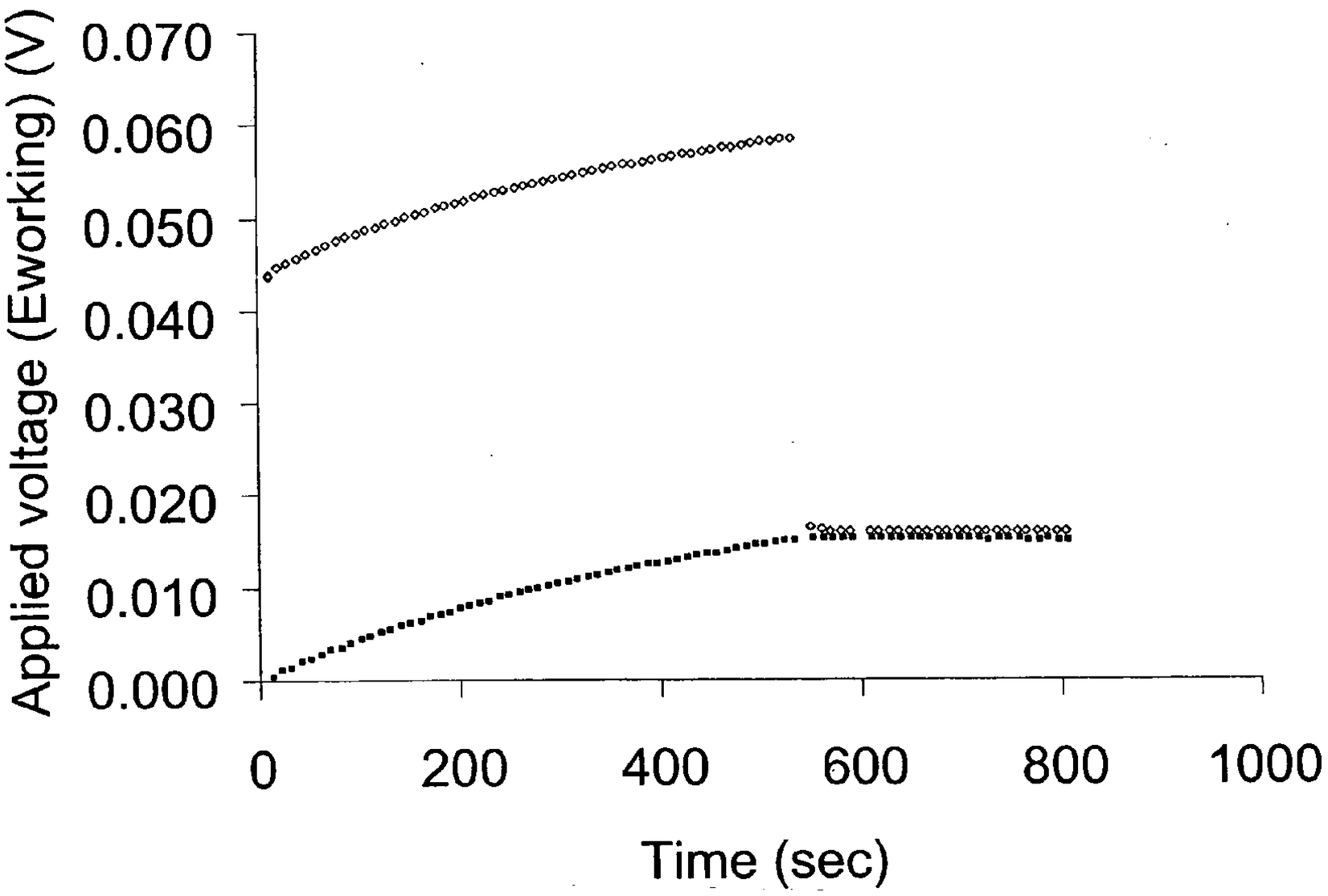

Applied voltage (Eworking) (V)
0.070
0.060
0.050
0.040
0.030
0.020
0.010
0.000
0
200
400
600
800
1000
Time (sec)

Fig. 11

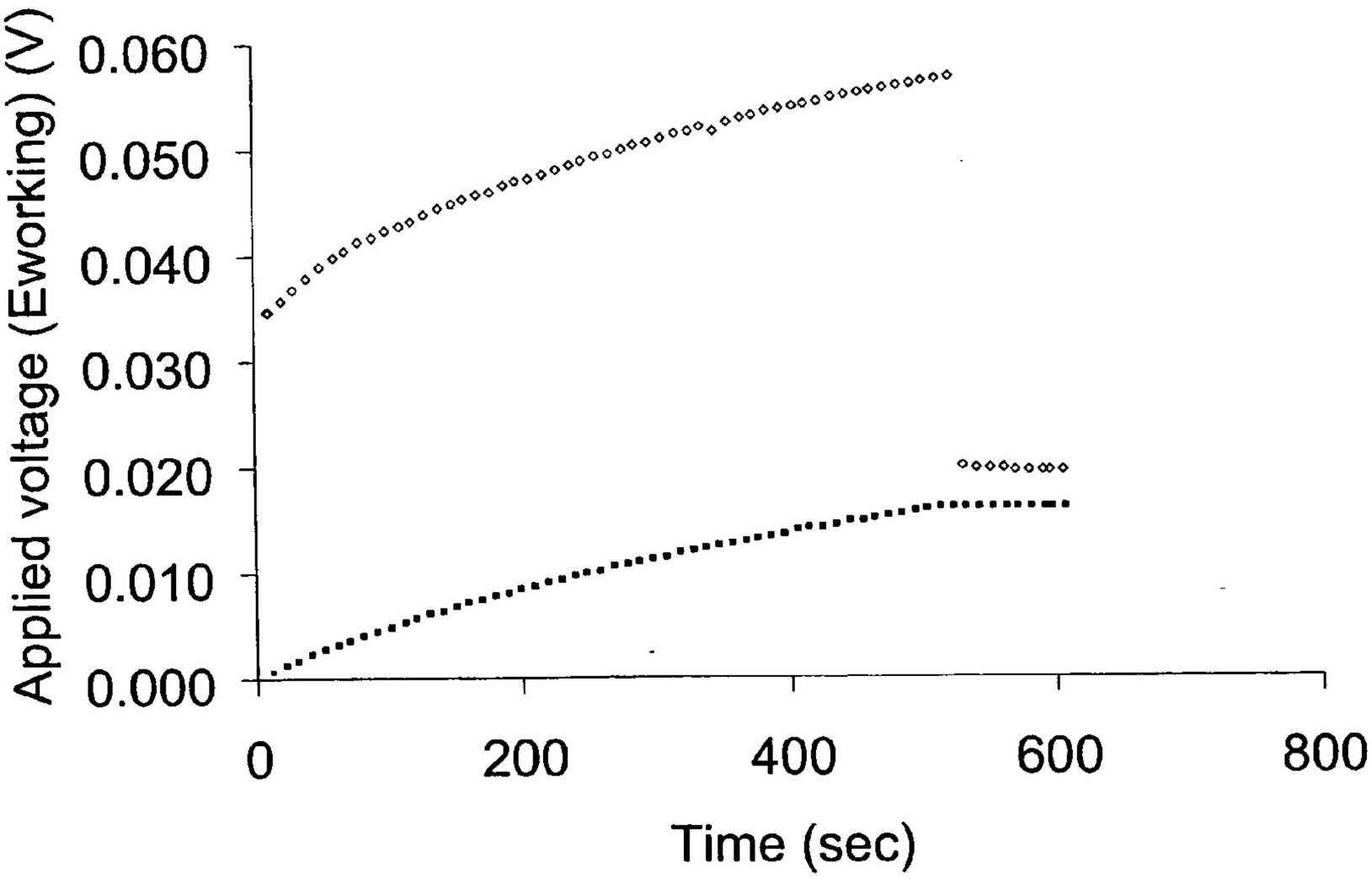

Applied voltage (Eworking) (V)
0.060
0.050
0.040
0.030
0.020
0.010
0.000
0
200
400
600
800
Time (sec)

Fig. 12

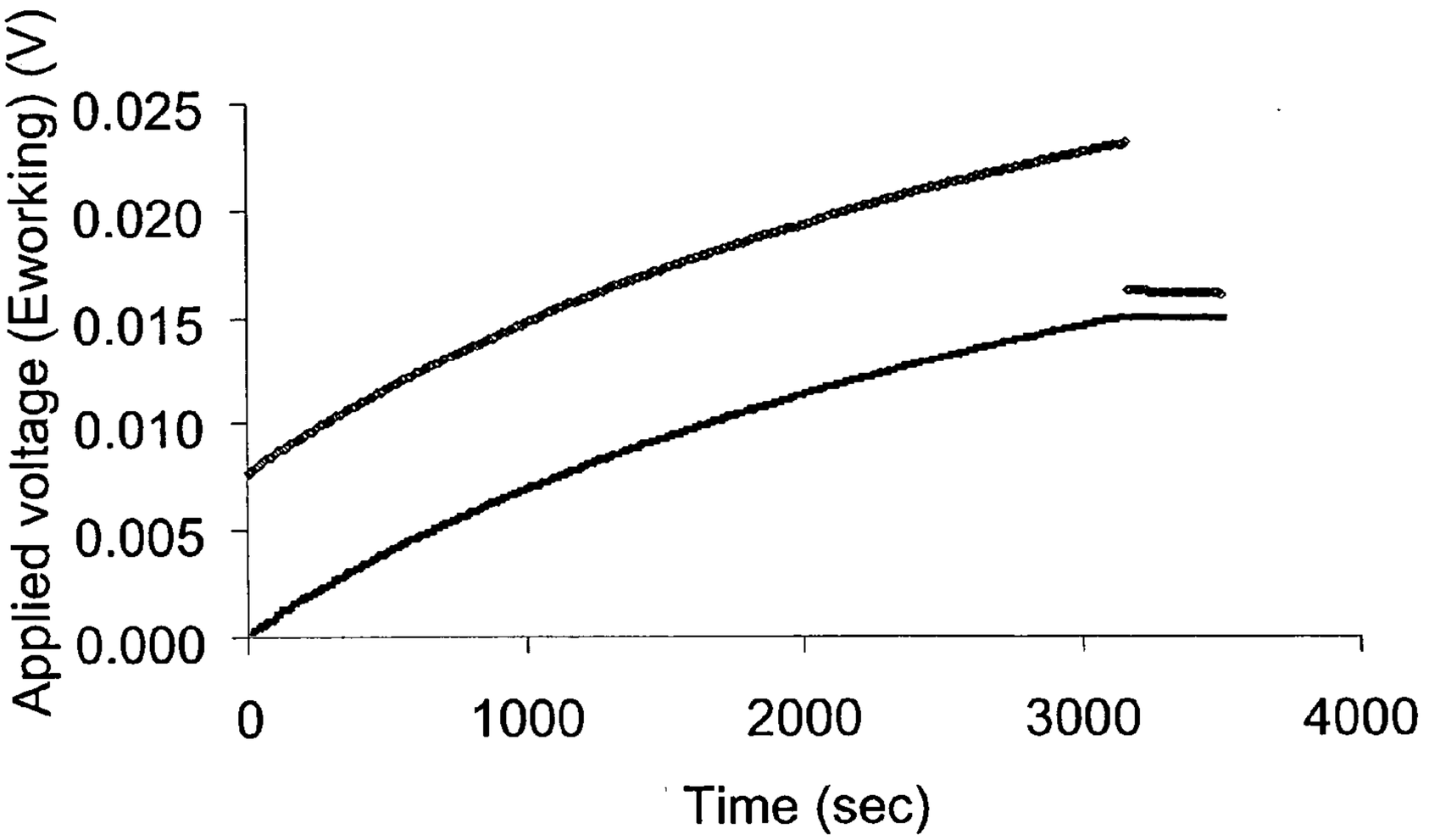

Applied voltage (Eworking) (V)
0.025
0.020
0.015
0.010
0.005
0.000
0
1000
2000
3000
4000
Time (sec)

Fig. 13

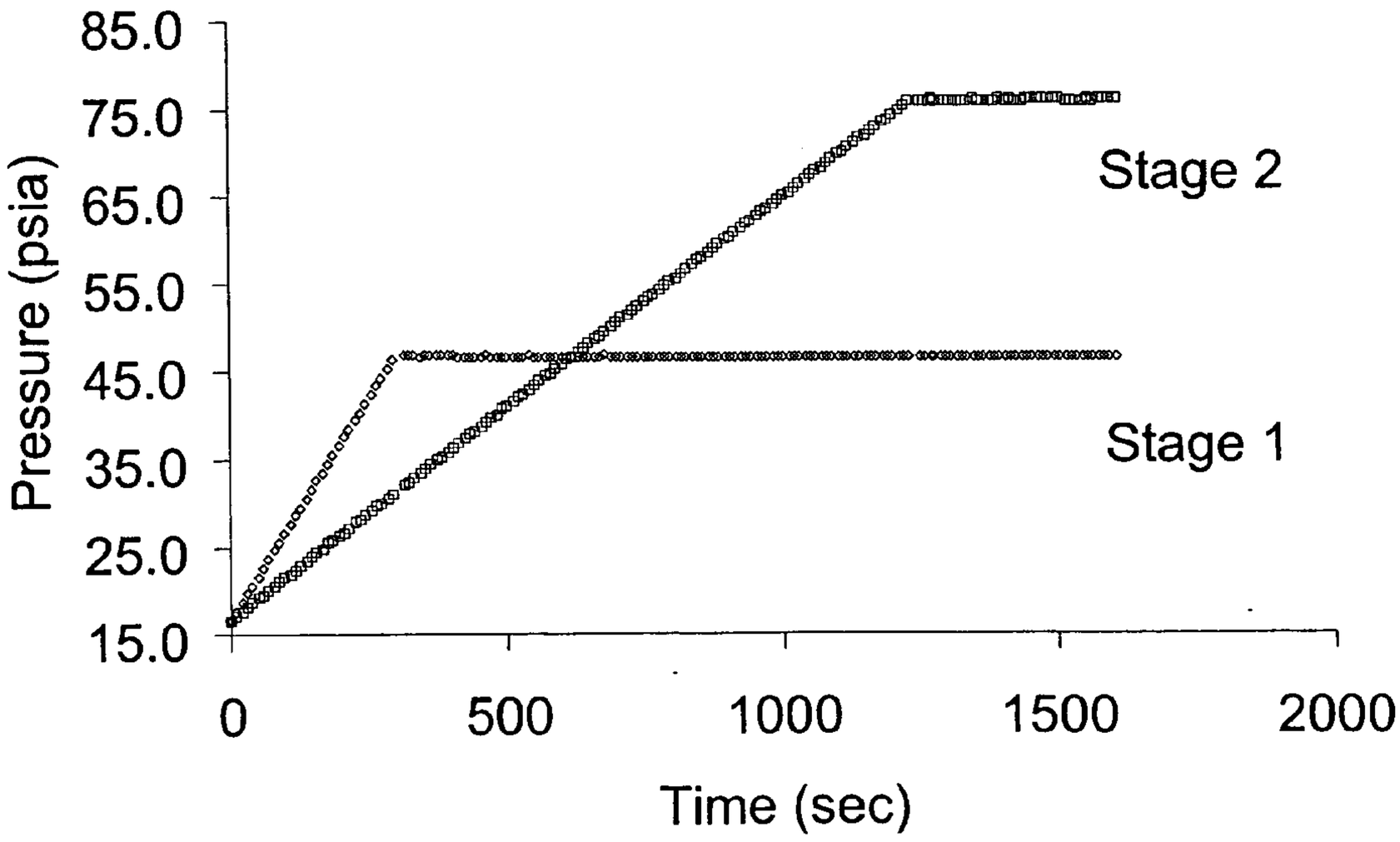

Pressure (psia)
85.0
75.0
65.0
55.0
45.0
35.0
25.0
15.0
Stage 2
Stage 1
0
500
1000
1500
2000
Time (sec)

Fig. 14

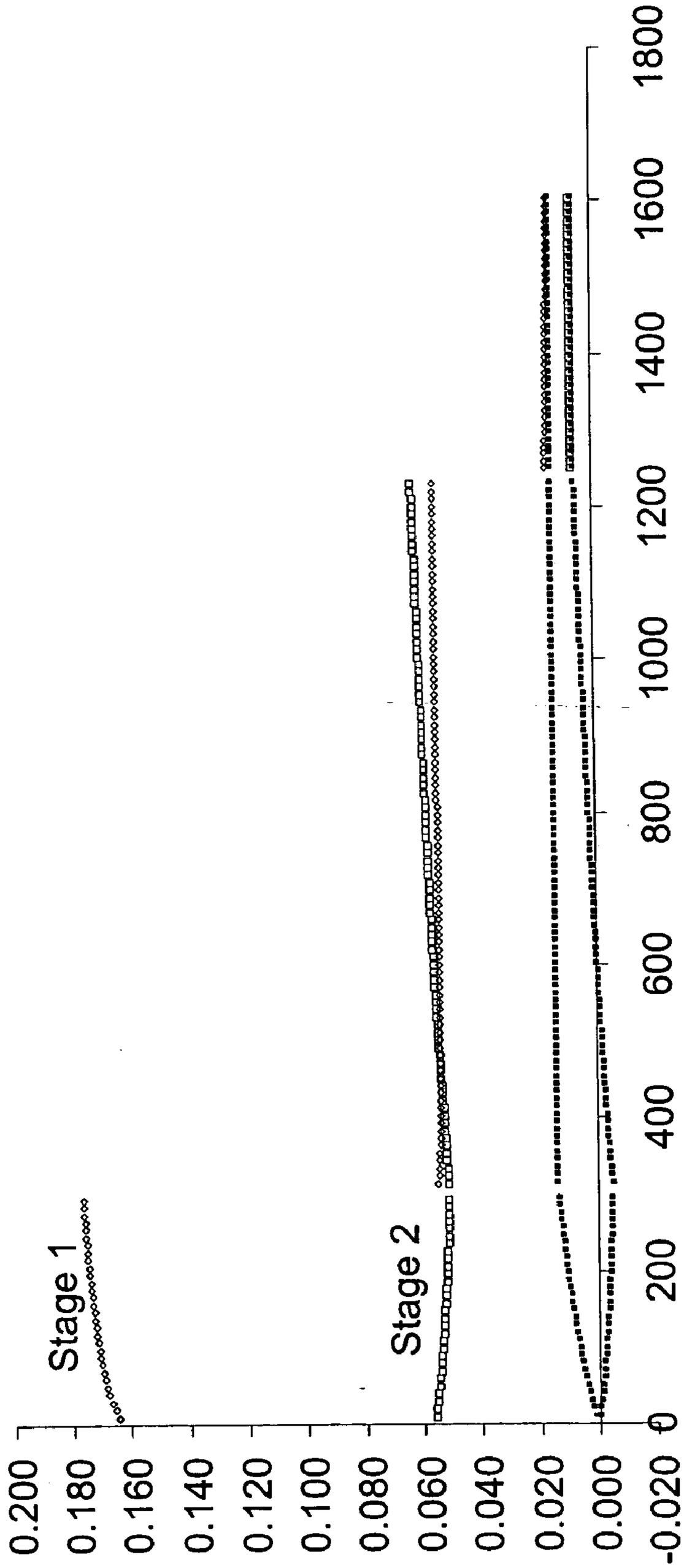

Applied voltage (Eworking) (V)
0.200
0.180
0.160
0.140
0.120
0.100
0.080
0.060
0.040
0.020
0.000
-0.020
0
200
400
600
800
1000
1200
1400
1600
1800
Time (sec)
Stage 1
Stage 2

Fig. 15

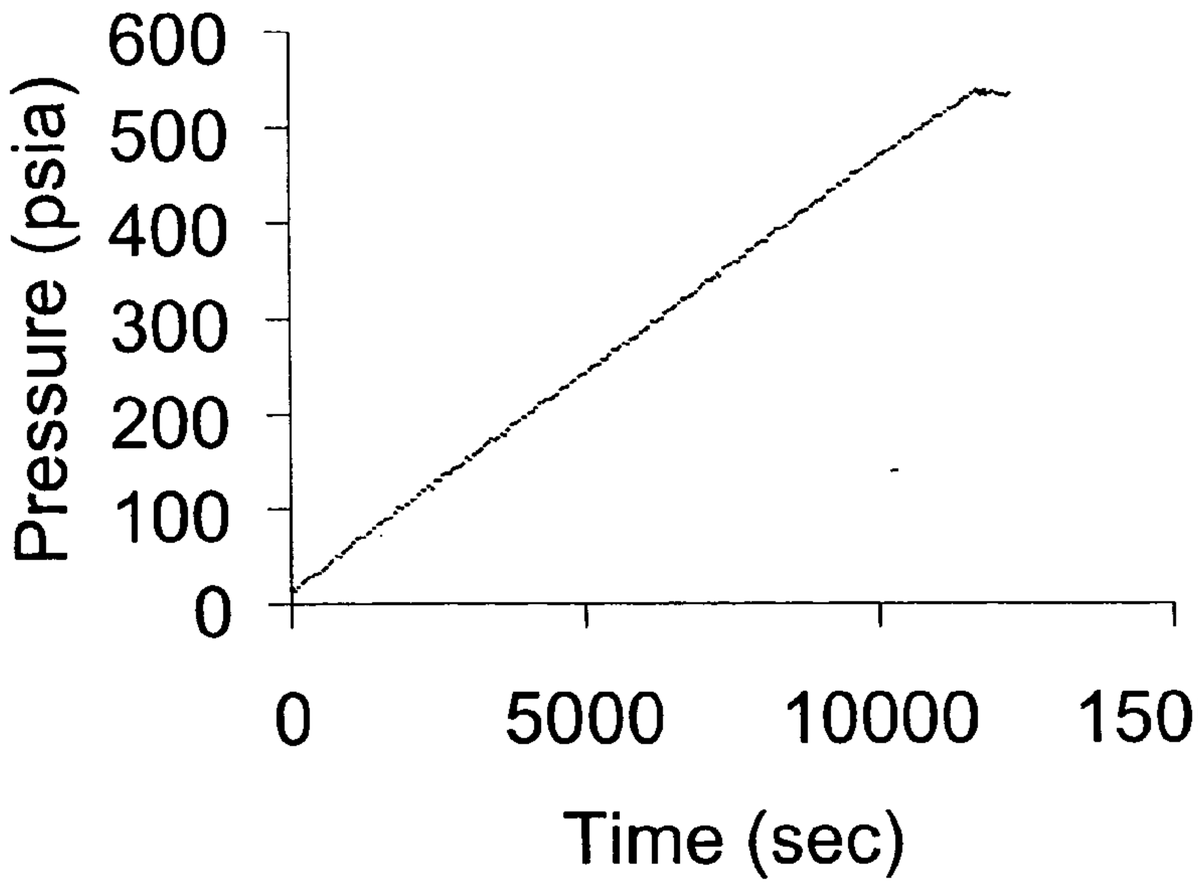

Pressure (psia)
600
500
400
300
200
100
0
0
5000
10000
15000
Time (sec)

Fig. 16

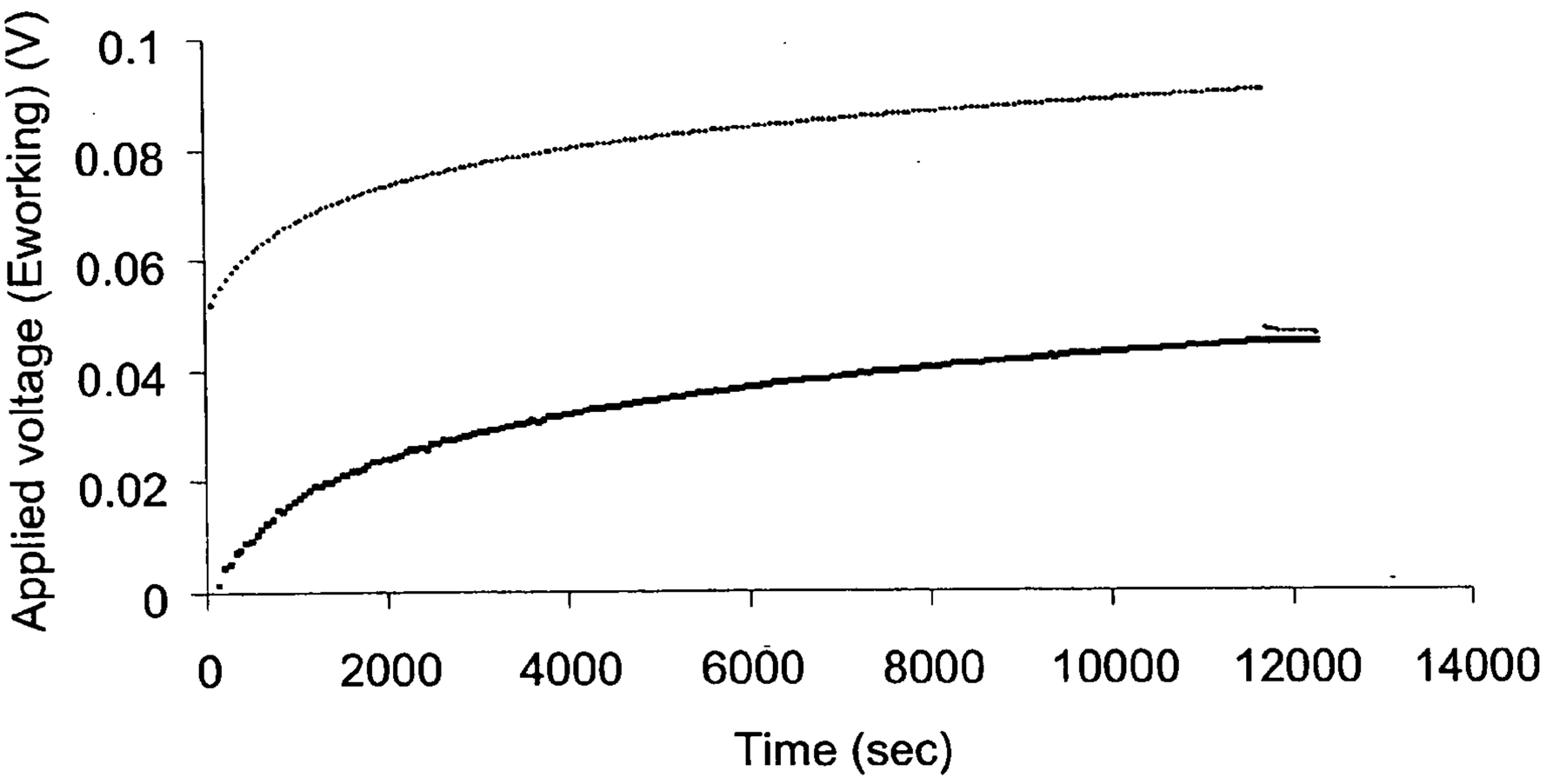

Applied voltage (Eworking) (V)
0.1
0.08
0.06
0.04
0.02
0
0
2000
4000
6000
8000
10000
12000
14000
Time (sec)

Fig. 17

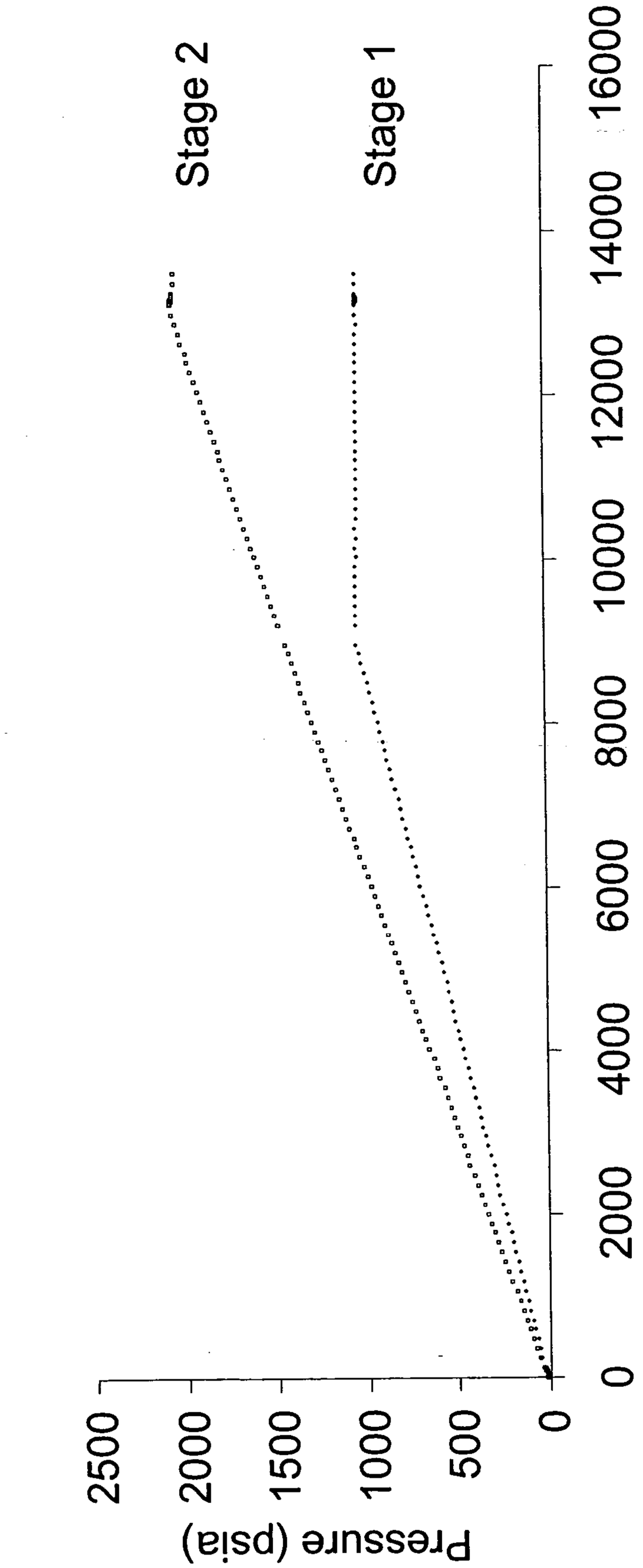

Stage 2
Stage 1
Pressure (psia)
2500
2000
1500
1000
500
0
0
2000
4000
6000
8000
10000
12000
14000
16000
Time (sec)

Fig. 18

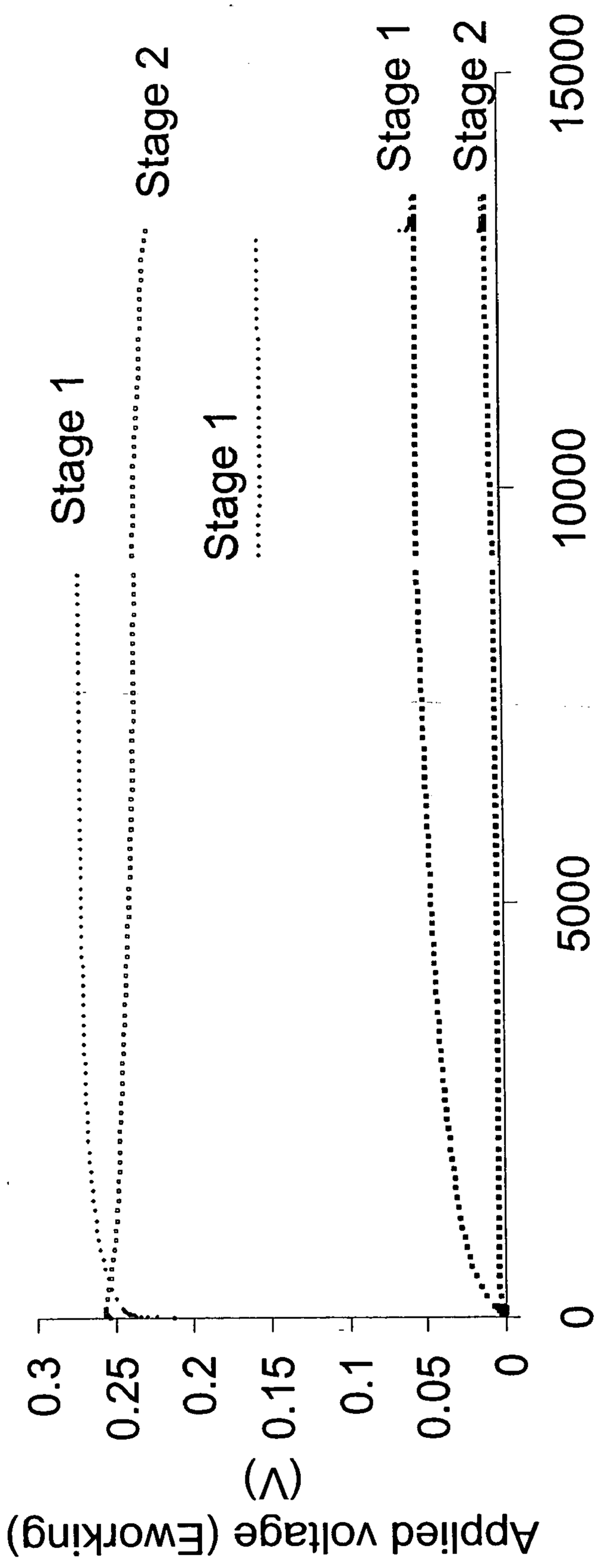

Stage 2
Stage 1
Stage 1
Stage 2
15000
10000
5000
Time (sec)
0
0.05
0.1
0.15
0.2
0.25
0.3
(V)
Applied voltage (Eworking)

Fig. 19

ELECTROCHEMICAL HYDROGEN COMPRESSOR

BACKGROUND OF THE INVENTION

[0001] This invention relates to an apparatus and process for electrochemical compression of hydrogen.

[0002] Fuel cells offer an environmentally friendly method of efficient energy generation, and the use of hydrogen as the fuel of choice is attractive as the conversion to electrical energy is emissions-free, with water and heat being the only by-products. The delivery of hydrogen in gaseous or liquid form or as an absorbed species (e.g. metal hydride, on activated carbon, or in carbon nanotubes) depends on the fuel cell application, and re-fueling frequency and related autonomy are important factors to consider in the selection of the appropriate mode of fuel storage. As liquid hydrogen involves energy-intensive and sophisticated cryogenic technologies and has a high boil-off rate and absorption technology is still in its infancy, gaseous hydrogen is a convenient and common form for storage, usually by pressurized containment for increased energy density. To this end, mechanical compression is the most common means by which to achieve pressurization; however, it suffers from limitations due to 1) intensive energy use, 2) wear-and-tear of moving parts, 3) hydrogen embrittlement, 4) excessive noise, 5) bulky equipment, and 6) contamination of the gas usually by compressor lubricants. Non-mechanical pressurization by thermal cycling is possible, but this is also energy intensive and not commercially practical yet.

[0003] Electrochemical transfer of hydrogen through proton-conductive materials is known, and fundamental studies on single-stage transfer applications can be found reported in the literature. For example, the use of thin perovskite-type oxide proton-conducting ceramics is well documented for single-stage separation of hydrogen from gas mixtures [1-3]. In these applications, the single cell operates at elevated temperatures (500-1000° C.) in order to maintain sufficiently high protonic conductivity through the separator. Reports on electrochemical hydrogen compression are scarce and most describe the use of single cells with a polymer electrolyte membrane (PEM), i.e. Nafion®, as the proton-conductive separator and Pt as the electrocatalyst on carbon electrodes (both anode and cathode) [4-7]. Operation of these cells to pressure differentials of ~43 atm (with anodic compartment pressure=1 atm) occurs without excessive energy demands; at greater differentials, however, rapid loss of H_2 due to leakage around the cell seals causes an exponential increase in power consumption. Use of Nafion PEM in electrochemical transfer of H_2 has also been reported [8]; in this case, H_2 was directed to the cathodic compartment filled with water in order to de-oxygenate the water by reaction of the H_2 with the dissolved O_2. It is important to note that in all these applications, electrochemical transport is selective to H_2 only due to the proton conductive nature of the separator and, in a gas mixture, hydrogen is not only concentrated (pressurized) but also purified by such means.

[0004] In U.S. Pat. No. 6,361,896 of Eberle et al. [10], the disclosure indicates that for single cell devices, differential pressures of up to about 10 bar can be achieved. This compares with earlier prior art devices that can only achieve a 5 bar differential pressure. Also disclosed is the use of a second cell to increase the differential pressure theoretically to "more than about 15 bar". The higher pressure differential is said to be achieved by means of a planar porous gas distribution support layer on the anode side (see col. 2). However, it is significant that there is no experimental proof provided that this was achieved. Moreover, no specific structure is described

[0005] Also described in Ströbel et al. [5] is a multi-cell stack. It is noted that the cells in the stack are connected in-parallel, so that there is no H_2 transport from one cell to the next. Accordingly, the H_2 output pressure from each cell is the same. The maximum pressure differential achieved was about 54 bar.

SUMMARY OF THE INVENTION

[0006] According to the invention, an apparatus and process are provided for pressurizing hydrogen electrochemically.

[0007] As will be discussed later, this technology targets the hydrogen supply and gas storage industries as well as the emerging fuel cell industry. With potential application of the technology in the fuel cell industry, high-pressure compression is desired and, more specifically, pressurization up to 12,000 psi is targeted, as this level is deemed necessary by the transportation industry for practical implementation of fuel cell vehicles.

[0008] According to one aspect of the invention, an apparatus is provided for compression of hydrogen, comprising a membrane electrolyte cell assembly (MEA), including a proton-conducting electrolyte membrane, an anode on one side of the membrane and a cathode on the other side of the membrane, the anode having an electrochemically active material for oxidizing hydrogen to protons, the cathode having an electrochemically active material for reducing protons to hydrogen, and further comprising next to the anode and cathode, planar gas distribution and support plates sandwiching the MEA, the assembly being held together by end-plates, the end-plates having complementary peripheral grooves for seating an intervening seal between the end-plates and the MEA, the end-plate on the anode side further including a hydrogen supply inlet and the end-plate on the cathode side further including a compressed hydrogen outlet.

[0009] According to another aspect of the invention, a process is provided for the compression of hydrogen by means of the apparatus described in the preceding paragraph, wherein hydrogen is compressed electrochemically by the MEA cell by oxidation of the hydrogen to protons at the anode, which having passed through the membrane to the cathode side are reduced back to hydrogen and discharged under pressure.

[0010] According to yet another aspect of the invention, in order to achieve even higher total or system pressure, we provide a plurality of such cells connected in series. The more cells connected in series, the higher the final outlet (overall) pressure that is achieved. By connection in series, we mean connected such that there is a progressive increase in pressure from cell to cell in the series. It would be expected by those skilled in the art that compression to higher pressures than those achievable by a single cell design according to our invention are achievable by includ-

ing additional cells connected in series. For example, by setting a pressure differential of 1000 psi per stage (at each cell), compression to 10,000 psi overall would require ten cells.

BRIEF DESCRIPTION OF THE DRAWING

[0011] **FIG. 1** is a diagram showing the concept of electrochemical hydrogen compression.

[0012] **FIG. 2** is a diagram showing the concept of multi-stage electrochemical hydrogen compression.

[0013] **FIG. 3** is a concept diagram showing a cross-sectional view of a multi-stage electrochemical hydrogen compressor with an overall cylindrical configuration.

[0014] **FIG. 4** is a diagram showing the design of planar gas distribution and support plates according to the invention with complementary grooves for intervening seals to provide a leak-free seal between the MEA and the plates.

[0015] **FIG. 5** is a diagram showing the unassembled view of a single-stage electrochemical hydrogen compressor unit according to the invention.

[0016] **FIG. 6** is a schematic circuit design for a two-stage electrochemical hydrogen compressor system according to the invention.

[0017] **FIG. 7** is a graph showing the results of electrochemical hydrogen compression to 45 psia (T=22° C.; i=0.6 A).

[0018] **FIG. 8** is a graph showing the results of electrochemical hydrogen compression to 75 psia (T=22° C.; i=0.6 A).

[0019] **FIG. 9** is a graph showing the voltage applied during electrochemical hydrogen compression to 45 psia (T=22° C.; i=0.6 A) (---ΔE derived using equation 4).

[0020] **FIG. 10** is a graph showing the voltage applied during electrochemical hydrogen compression to 75 psia (T=22° C.; i=0.6 A) (---ΔE derived using equation 4).

[0021] **FIG. 11** is a graph showing the voltage applied during electrochemical hydrogen compression to 45 psia (T=65° C.; i=0.6 A) (---ΔE derived using equation 4).

[0022] **FIG. 12** is a graph showing the voltage applied during electrochemical hydrogen compression to 45 psia (T=80° C.; i=0.6 A) (---ΔE derived using equation 4).

[0023] **FIG. 13** is a graph showing the voltage applied during electrochemical hydrogen compression to 45 psia (T=65° C.; i=0.1 A) (---ΔE derived using equation 4).

[0024] **FIG. 14** is a graph showing the results of dual-stage, electrochemical hydrogen compression to 75 psia (T=22° C.).

[0025] **FIG. 15** is a graph showing the voltage applied during dual-stage, electrochemical hydrogen compression to 75 psia (T=22° C.) (---ΔE derived using equation 4).

[0026] **FIG. 16** is a graph showing the results of electrochemical hydrogen compression to 535 psia (T=22° C.; i=0.6 A).

[0027] **FIG. 17** is a graph showing the voltage applied during electrochemical hydrogen compression to 535 psia (T=22° C.; i=0.6 A) (---ΔE derived using equation 4).

[0028] **FIG. 18** is a graph showing the results of dual-stage, electrochemical hydrogen compression to 2000 psia (T=22° C.).

[0029] **FIG. 19** is a graph showing the voltage applied during dual-stage, electrochemical hydrogen compression to 2000 psia (T=22° C.) (---ΔE derived using equation 4).

DETAILED DESCRIPTION OF THE INVENTION

[0030] Electrochemical compression of hydrogen is accomplished by the application of an electric potential across a proton-conductive polymer electrolyte material separating anode and cathode compartments to effect the transport of hydrogen from one side to the other. The process is based on the following anodic and cathodic reactions:

$$H_2 \rightarrow 2H^+ + 2e^- \text{ anodic (oxidation)} \quad (1)$$

$$2H^+ + 2e^- \rightarrow H_2 \text{ cathodic (reduction)} \quad (2)$$

[0031] The use of electrocatalysts facilitates these reactions, and the principle of operation can be illustrated as shown in **FIG. 1**. Oxidation of H_2 at the anode **10**, located in anodic compartment **12** generates hydrogen ions (protons) and electrons; the hydrogen ions migrate across the proton-conductive polymer electrolyte separator **14** while the electrons travel via an external circuit to the cathodic compartment **16** where reduction back to H_2 takes place at the cathode **18**. From thermodynamic considerations using the Nemst equation, the theoretical applied potential to effect a desired final pressure of H_2 exiting the cathodic compartment can be determined. For example, the thermodynamic cell potential is represented by the following equation:

$$E_{cell} = E_c - E_a = E^\circ_{cell} - \frac{RT}{2F} \ln \frac{a_{H_2,c}}{a_{H_2,a}} \quad (3)$$

[0032] E_{cell}=thermodynamic cell potential, V

[0033] E_c=cathode half-cell potential, V

[0034] E_a=anode half-cell potential, V

[0035] E_{cell}°=thermodynamic cell reference potential, 0.00 V

[0036] $a_{H_2,c}$=activity of H_2 at the cathode

[0037] $a_{H_2,a}$=activity of H_2 at the anode

[0038] R=gas constant, 8.3144 mol^{-1} K^{-1} L kPa

[0039] T=temperature, K

[0040] F=Faraday constant, 96487 C/mol e^-

[0041] With hydrogen as a gas, a_{H_2} equates to pressure, P_{H_2}, and the applied potential, ΔE, is determined from the following equation:

$$\text{applied potential} = \Delta E = |E_{cell}| = \frac{RT}{2F} \ln \frac{P_{H_2,c}}{P_{H_2,a}} \quad (4)$$

[0042] For a more rigorous mathematical treatment, the thermodynamic property, fugacity (f), is used and is the

effective pressure when the non-ideality of gases is taken into consideration. Fugacity relates to P by the following equation:

$$\text{fugacity, } f=\phi P \tag{5}$$

[0043] where ϕ is the fugacity coefficient, akin to the activity coefficient (γ) in the thermodynamic treatment of non-ideal solutions. Fugacity coefficients have been tabulated for a number of gases and, for hydrogen, ϕ is essentially 1.0 for pressures up to 1000 psia (68 atm) [9]; at higher pressures, f becomes significant. The applied potential is then determined more accurately from the following equation:

$$\text{applied potential} = \Delta E = |E_{cell}| = \frac{RT}{2F}\ln\frac{f_{H_2,c}}{f_{H_2,a}} = \frac{RT}{2F}\ln\frac{\phi_{H_2,c}}{\phi_{H_2,a}} + \frac{RT}{2F}\ln\frac{P_{H_2,c}}{P_{H_2,a}} \tag{6}$$

[0044] ϕ_c=cathodic compartment fugacity coefficient

[0045] ϕ_a=anodic compartment fugacity coefficient

[0046] For example, for a ten-fold increase in pressure with $P_{H_2,a}$=1 atm and $P_{H_2,c}$=10 atm at room temperature (25° C.), the applied potential is ΔE=29.7 mV (with ($\phi_{H_2,a}$=1.000 and $\phi_{H_2,c}$=1.006). For a further ten-fold increase with $P_{H_2,a}$=10 atm and $P_{H_2,c}$=100 atm, ΔE=30.3 mV ($\phi_{H_2,a}$=1.006 and $\phi_{H_2,c}$=1.063) and, with $P_{H_2,a}$=1 atm and $P_{H_2,c}$=100 atm, ΔE=59.9 mV ($\phi_{H_2,a}$=1.000 and $\phi_{H_2,c}$=1.063). As evident, a relatively small applied potential results in significant pressurization, and the device functions essentially as a concentration cell. In actuality, the required applied voltage, $E_{working}$, would be higher due to electrode overpotentials and resistance (circuit and ohmic drop across the separator) and due to application of an electric current to effect a timely increase in pressure:

$$E_{working}=\Delta E+E_{polarzation}$$
$$E_{polarization}=|\eta_a|+|\eta_c|+iR_{separator}+iR_{circuit} \tag{7}$$

[0047] η_a=overpotential of the anode, V

[0048] η_c=overpotential of the cathode, V

[0049] i=applied current, A

[0050] $R_{separator}$=resistance across the proton-conductive separator, ohm

[0051] $R_{circuit}$=resistance of the electrical circuit, ohm

[0052] The overpotentials of the anode and cathode represent chemical kinetic barriers, i.e. the energy required for electron transfer during the anodic and cathodic electrochemical reactions, and the use of electrocatalysts (e.g. Pt) and/or higher temperatures can reduce these values. For resistance, the ohmic drop across the separator can be minimized, for instance, by the use of thinner materials and, across the circuit, with appropriate electrical materials.

[0053] With the initial condition, $P_{H_2,a}=P_{H_2,c}=P_i$, the energy consumption for single-stage compression, from $P_{H2,c}=P_i$ to P_f, can be determined from the following equation:

$$\text{energy} = w = \Delta nRT\ln\frac{P_f}{P_i} + E_{polarization}(i)(\Delta t) \tag{8}$$

[0054] Δn=no. of moles of H_2 transferred, mol

[0055] Δt pressure increase time period, sec

[0056] $\Delta nRT\ln(P_f/P_i)=w_t$, thermodynamic work of compression,

[0057] The efficiency (%) of electrochemical hydrogen compression is referenced to the applied voltage and is a measure of the deviation from thermodynamic work:

$$\text{efficiency} = \frac{w_t}{w}\times 100 \tag{9}$$

[0058] In practice, multi-stage compression is preferred for high-pressure applications whereby pressure differentials between stages can be set to reasonable values in order to accommodate material limitations (e.g. structural integrity, effective sealing, and H_2 back-diffusion phenomenon [5]). For an electrochemical compressor with N stages with P_1, P_2 . . . P_N initially established and kept constant (**FIG. 2**), the total energy consumption can be determined simply from summation of the compression energies at each stage:

$$\text{total energy} = \sum_1^N w_N = \sum_1^N E^N_{polarization}(i)(\Delta t) \tag{10}$$

[0059] Methodology

[0060] The design of an electrochemical hydrogen compressor is similar to that of a fuel cell, and it is proposed that a multi-stage unit be modeled after a PEM fuel cell stack. Nafion® is employed as the proton-conductive polymer membrane separator with Pt as the electrocatalyst dispersed on carbon to function as the anode and cathode electrodes in the overall membrane-electrode-assembly (MEA).

[0061] As shown in **FIG. 3**, an overall cylindrical multi-cell stack configuration, having, for example, hemispherical end-plates **26** provides good mechanical stability. Hydrogen supply inlet **33** is provided in the end-plate on the anode side of the first cell and compressed hydrogen outlet **35** in the other end-plate on the cathode side of the last cell. The plates are connected by tie-bolts **28**. The design of a multi-stage unit is as illustrated where electrically non-conductive separators **20** ensure electrical separation of compression stages. It will be appreciated by those skilled in the art that other configurations will also work effectively. As with fuel cells, graphite support plates **22** could be used sandwiching the MEA's **24**, but these require separate charge collectors for good electrical conductivity (cf. copper endplates in a fuel cell stack).

[0062] As best seen in **FIG. 4**, porous stainless steel support plates **22** are used, which are positioned adjacent to the MEA **24** with seals (e.g. in the form of an elastomeric o-ring) disposed in grooves **30** to ensure a leak-free seal

between the plate and the membrane of the MEA (i.e. the peripheral area outside of the active area). Unlike a fuel cell stack, complex serpentine flow fields are not necessary, and access of H_2 to the MEA's is simply achieved by perforating the plates **22** e.g. in a central area **23** of the plate, or by use of sintered frit plates. The sintered metal frit plates are made of a powdered metal material such as stainless steel, which is compressed into the form of a plate. Such material provides a structurally strong, yet porous material to provide for passage of gases to and from the active area of the MEA.

[**0063**] It is believed that the high differential pressures are achieved by means of the porous supporting plate **22** on the anode side and its seating in socket **25***a*. On the cathode side, the porous plate maintains contact during pressurization with the active area of the MEA via use of a spring means, including a spring **29** and spring support **31**, (i.e. see **FIG. 5**) for ensuring adequate electrical contact. High-pressure stability is provided because the plates **22** immobilize the MEA during pressurization, such that the membrane does not rupture due to a ballooning effect.

[**0064**] Commercially available materials (MEA, stainless steel plating, and seals) are used in the construction of single- and two-stage compressors (see later). Examples of other proton-conductive membranes include sulfonated-polystyrene and the partially fluorinated ionomeric membranes, IonClad R-1010 and R-4010 (Pall Co.), as these represent more economical alternatives to Nafion. Also, as H_2 is the only species of interest, complications of slow membrane deterioration, as reported in fuel cells and attributed to the formation of hydrogen peroxide (from reaction of H_2 with O_2) within the membrane, is not expected to be a problem, and the use of non-fluorinated materials such as sulfonated-polystyrene will suffice in electrochemical compression applications.

[**0065**] The design of supporting plates **22** incorporates porosity or perforation characteristics in order to allow sufficient exposure of H_2 to catalytic active sites on the surface of the MEA and, at the same time, permit the plates to give sufficient structural support to the membrane, thus minimizing its deformation under conditions of high-pressure differentials.

[**0066**] In the embodiment shown in **FIGS. 3 and 4**, the design of the supporting plates **22** also incorporates complementary peripheral grooves **30** for disposition of seals, e.g. an elastomeric o-ring to insure a leak-free seal between the MEA and the plates.

[**0067**] **FIG. 5** shows the unassembled view of a single stage of the working system responsible for establishing proof-of-concept, multi-stage electrochemical compression. This electrochemical compressor unit comprises a membrane-electrode-assembly (MEA) **24** supported by stainless steel sintered frit plates **22***a* and contained within cylindrical stainless steel housing **26** that make up the anodic and cathodic compartments. The stainless steel housing **26** is a high-pressure filter holder (Fisher Scientific, cat. no. 09-753-13M) adapted for its present use. The membrane-electrode-assembly **24** (Palcan Fuel Cell Co. Ltd., Vancouver, Canada) is circular in design (**FIG. 4**) with an active area of 11.34 cm^2 and comprises of gas-diffusion electrodes (anode and cathode), comprising Pt (1 mg/cm^2) as the electrocatalyst supported on carbon (40 wt. % Pt/C), and Nafion® 115 as the electrolyte. Use of this unit is documented in examples described below for single- and dual-stage hydrogen compression.

[**0068**] As shown in **FIG. 5**, complementary pairs of grooves **25** and pocket **25***a* are machined into the inside face of both end-plates **26**. Upon assembly, the seal between the end-plates **26** and the MEA **24**, is provided by an o-ring **27** of an elastomeric material, disposed in the grooves **25**, and the frit plate **22***a* is seated in pocket **25***a*.

[**0069**] A spring **29** and spring support **31** are provided on the cathode side. Both the spring and spring support are conveniently made of stainless steel. This spring and spring support arrangement provides for equalization of the force exerted on the MEA by the frit plate on the cathode side of the MEA **24**, regardless of the pressure differential across the MEA, such that the MEA can move together, i.e. without separating as a result of the high pressure.

[**0070**] As H_2 is a small molecule able to permeate through many types of materials, the selection and design of appropriate sealing material is important. Examples include Viton®, Santoprene®, and PTFE.

[**0071**] The multi-stage compressor embodiment includes a plurality of PEM cells connected in series, such that the compressed hydrogen from the outlet of a first cell in the series is fed to the hydrogen inlet of the next cell in series, wherein each cell is electrically isolated from the adjacent cell in the series. Note that while it is apparent that in the Strobel et al. publication, the cells are clearly shown to be connected in parallel, U.S. Pat. No. 6,361,896 states that the cells are connected "in series". However, it is apparent that by "in series" the authors mean that the cells are arranged or placed adjacent to each other, i.e. as illustrated in the publication, for increased hydrogen flux. However, the hydrogen outputs and inputs are not connected in series and, therefore, progressive increases in pressure from cell to cell are not possible.

[**0072**] The circuit diagram for a two-stage unit connected in series showing the balance-of-plant is illustrated in **FIG. 6**, wherein PG refers to pressure gauges; PCV refers to pressure check valves; CV refers to check valves; FM refers to flow meters; PT refers to pressure transducers; HUM refers to the gas humidifier; HTR refers to the heater; RH refers to the relative humidity ports; and T/C refers to the thermocouple ports. Separate power supplies are used for each electrochemical compressor unit. The system is purged with nitrogen prior to hydrogen compression. Hydrogen is humidified by HUM101 and initially introduced to the entire system at atmospheric pressure. Thereafter, power is applied to the electrochemical compressor unit(s), and the pressure is monitored via PT101, PT102, and PT103. The system temperature is monitored via thermocouples at all T/C ports. In the circuit diagram, the stages are electrically isolated by use of electrically insulating (e.g. Teflon®) tubing, or by Swagelok dielectric fittings. This provides electrical isolation of stage 1 from stage 2.

[**0073**] In single-stage compression, one electrochemical compressor unit is employed and, as examples of its performance, **FIGS. 7 and 8** show temporal plots for compression from atmospheric pressure (15.9 psia) to approx. 45 and 75 psia hydrogen, respectively. **FIGS. 9 and 10** show corresponding temporal plots of the applied voltages along

with the thermodynamic applied potential (ΔE) as determined from equation 4. For compression to 45 psia, 0.6 A was applied galvanostatically, and a linearly increase in pressure in the cathodic compartment (volume=18.2 mL) was effected. At t=610 sec, the pressure reached 45.7 psia (**FIG. 7**), and the voltage applied during compression increased from 58.2 to 70.3 mV (**FIG. 9**). The current source was then discontinued (i=0 A), and the equilibrium potential across the cell was measured with $\bigstar E_{cell}|$=14.1 mV. For compression to 75 psia, a linear pressure increase in the cathodic compartment also took place and, at t=1270 sec, $P_{H_2,c}$=76.4 psia. The applied voltage during compression increased from 57.1 to 78.4 mV (**FIG. 11**). The current source was then discontinued, and the equilibrium cell potential measured was $|E_{cell}|$=22.0 mV. In both studies, the system temperature recorded was 22.0° C. There is excellent agreement in the profile of the plots of the applied voltages and ΔE ($E_{polarization}$=58 mV), and between ΔE and the measured $|E_{cell}|$. For compression to 45 psia, the energy consumption, determined using equation **8**, was 25.1 J (with w_t=3.9 J). The efficiency, as determined from equation **9**, was 16%. For compression to 75 psia, w=56.1 J (with w_t=11.9 J), and the efficiency was 21%. It is noted that efficiency improves with increasing temperature and with decreasing applied current (due to lower i^2R losses), and upwards of 80% has been reported for single-stage electrochemical compressors [4,5]. For the present system, compression to 45 psia at 65 and 80° C. with i=0.6 A yields the applied voltage profiles as illustrated in **FIGS. 11 and 12** ($E_{polarization}$≈43 mV at 65° C.; $E_{polarization}$=33 mV at 80° C.), and the respective efficiencies were 22 and 28%. Likewise, with i=0.1 A and T=65° C., the profile of the applied voltage for compression to 45 psia is shown in **FIG. 13** ($E_{polarization}$= 7.8 mV), and the efficiency here was 60%.

[**0074**] In dual-stage compression, two electrochemical compressor units connected in series are employed, and **FIG. 14** shows an example of the pressure change at each stage (unit) from application of electrical power. Here, 45 and 75 psia were chosen as final pressures for the first and second stages, respectively, both stages initially at atmospheric pressure (15.9 psia). A current of 2.4 A was applied galvanostatically to stage 1 and 0.6 A to stage 2. A linear increase in pressure was effected at both stages; 46.7 psia was reached at t=315 sec for stage 1 (volume=27.2 mL), and 75.7 psia was attained after 1230 sec for stage 2 (volume= 18.2 mL). At stage 1, the current was reduced to 0.6 A to maintain the pressure until the final pressure at stage 2 was reached. Afterwards, the current source was discontinued at both stages (i=0 A), and the equilibrium cell potential was measured at both stages, with $|E_{cell}|$=15.0 mV for stage 1 and $|E_{cell}|$=6.9 mV for stage 2.

[**0075**] **FIG. 15** shows the temporal plots of the applied voltages with comparison to those for ΔE (as determined from equation 4), and there is good agreement of the profile of the plots ($E_{polarization}$=40 mV (0.6 A) and 158 mV (2.4 A) for stage 1, and $E_{polarization}$=57 mV (0.6 A) for stage 2). The system temperature was 22.0° C. The energy consumption for priming the dual-stage compressor to the chosen stage pressures was 400.7 J, as determined using equation 8.

[**0076**] Single-stage compression to higher pressures was also performed. For example, compression to 535 psia (**FIG. 16**) was carried out at T=22° C. and using i=0.6 A. The profile of the applied voltage is shown in **FIG. 17**, and there is good agreement with that of ΔE ($E_{polarization}$≈52 mV). The energy consumption, as determined from eq. 8, was 362.4 J (with W_t=231.4 J), and the efficiency, as determined from eq. 9, was 39%.

[**0077**] Dual-stage compression to higher to pressures has also been carried out. For example, at T=22° C., compression to 2000 psia, with stage 1 at 1000 psia, is illustrated in **FIG. 18**. The profile of the applied voltages is shown in **FIG. 19**, and there is good agreement with ΔE derived from eq. 4. The applied current was 4.0 A for stage 1 and 2.0 A for stage 2. The equilibrium cell potentials were $|E_{cell}|$=53.0 mV for stage 1 and $|E_{cell}|$=8.7 mV for stage 2.

[**0078**] For the fuel cell industry, the electrochemical hydrogen compressor can be applied interfacing: 1) a hydrogen production device (i.e. fuel processor, electrolyzer, etc.) and a fuel cell; 2) a hydrogen production device and a hydrogen storage device; and 3) a hydrogen storage device and a fuel cell. For industries concerned with hydrogen supply and storage, the compressor can be applied interfacing a hydrogen production device and a hydrogen storage device.

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1. An apparatus for compression of hydrogen, comprising a membrane electrolyte cell assembly (MEA), including a proton-conducting electrolyte membrane, an anode on one side of the membrane and a cathode on the other side of the membrane, the anode having an electrochemically active material for oxidizing hydrogen to protons, the cathode having an electrochemically active material for reducing protons to hydrogen, and further comprising next to the anode and cathode, planar gas distribution and support plates sandwiching the MEA, the assembly being held together by end-plates, the end-plates having complementary peripheral grooves for seating an intervening seal

between the end-plates and the MEA, the end-plate on the anode side further including a hydrogen supply inlet and the end-plate on the cathode side further including a compressed hydrogen outlet.

2. The apparatus according to claim 1, wherein the gas distribution and support plates include a central gas distribution area.

3. The apparatus according to claim 2, wherein the gas distribution area is in the form of pores.

4. The apparatus according to claim 1, the gas distribution and support plates are made of a porous sintered metal frit material.

5. The apparatus according to claim 4, wherein the metal frit is stainless steel frit.

6. The apparatus according to claim 2, wherein the end-plates each include an additional complementary pocket for seating the metal frit plates.

7. The apparatus according to claim 6, additionally comprising on the cathode side between the gas distribution and support plate and the end plate, a spring means for ensuring adequate electrical contact.

8. The apparatus according to claim 7, wherein both the spring and spring support are made of stainless steel.

9. The apparatus according to claim 7, wherein the proton conducting membrane is of a material selected from the group consisting of Nafion®, sulfonated-polystyrene and the partially fluorinated ionomeric membranes, lonClad® R-1010 and R-4010.

10. The apparatus according to claim 1, additionally comprising means for applying an electric potential to the cell, wherein the applied potential to effect a final pressure of hydrogen exiting the cathode side of the cell is determined by the Nernst equation.

11. The apparatus according to claim 1, comprising a plurality of MEA cells connected in series, such that the compressed hydrogen from the hydrogen outlet of a first cell in the series is fed to the hydrogen inlet of the next cell in series, wherein each cell is electrically isolated from the next cell in the series.

12. A process for the compression of hydrogen by means of the apparatus according to claim 1, wherein hydrogen is compressed electrochemically by the MEA by oxidation of the hydrogen to protons at the anode, which having passed through the membrane to the cathode side are reduced back to hydrogen and discharged under pressure.

13. The process according to claim 12, wherein hydrogen is pressurized to 12,000 psi or greater.

14. The process according to claim 12, wherein a plurality of MEA cells are connected in series, each cell being electrically isolated from the next cell in the series, such that hydrogen discharged under pressure from the hydrogen outlet of a first cell in the series is fed to the hydrogen inlet of the next cell in series, and hydrogen is discharged from the hydrogen outlet of the next cell at a higher pressure.

15. An apparatus for compression of hydrogen, comprising a membrane electrolyte cell assembly (MEA), including a proton-conducting electrolyte membrane, an anode on one side of the membrane and a cathode on the other side of the membrane, the anode having an electrochemically active material for oxidizing hydrogen to protons, the cathode having an electrochemically active material for reducing protons to hydrogen, and further a hydrogen supply inlet and a compressed hydrogen outlet.

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