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(54) Title: DC POWER SOURCE FOR ELECTROLYSIS DEVICES AND ELECTRODE SYSTEM WITHOUT CATALYSTS

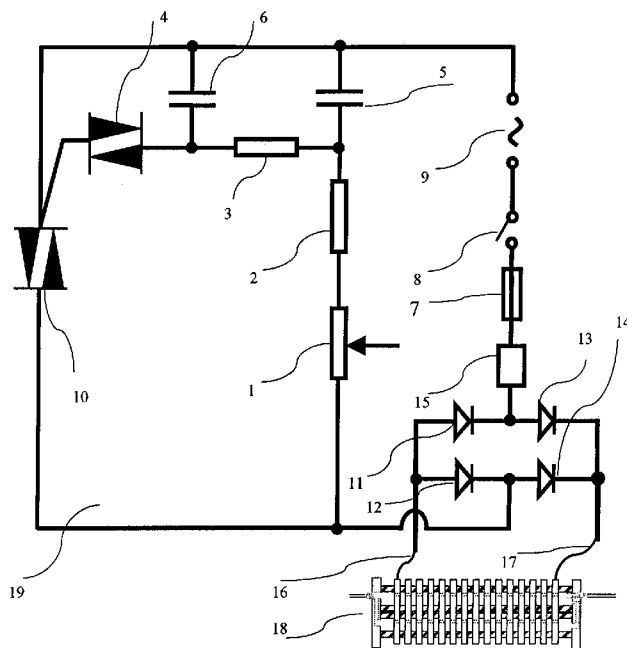


FIGURE 1

(57) Abstract: A power source which converts the alternat-
ive current from 220V mains to direct current for hydrogen
producing water fuelled electrolysis devices is invented.
The invented power source can be used both in classical
pressurized alkaline type and catalyst loaded proton ex-
change electrolyte membrane systems. A compressed elec-
trode system which separates the stainless steel electrodes
from butyl rubber gaskets is invented in the produced elec-
trolysis device which is using the invented power source.
The gas tightness is accomplished by special flexibility ad-
justed rubber gaskets. Also, a filter system is invented to
prevent the mixing of produced hydrogen and oxygen in
anode and cathode of alkaline electrolysis systems. The pre-
vention of gas mixing is achieved by using microporous
poly ether sulfone and butyl gaskets in the filter system.

DC POWER SOURCE FOR ELECTROLYSIS DEVICES AND ELECTRODE SYSTEM WITHOUT CATALYSTS

This invention is related to the electrochemically hydrogen and oxygen producing electrolysis devices and their power sources. The field of invention is about decomposition of water, methanol and some other organic materials in liquid electrochemical cells by applying electrochemical potential. The invented power source generally can be applied to direct current (DC) pressurized electrolysis devices.

An electrolysis device which uses distilled or de-ionized water to produce pure hydrogen and oxygen in stoichiometric ratios by using 220V mains without a transformer was developed. The power source of this design is related to the pulsed and square waved DC producing electronic circuits, which are known as switch mode power supplies (SMPS) in the field of industrial electronics. The invented power source and electrode design was generally designed for pressurized alkaline electrolysis systems. However, the same power source can be used in generating hydrogen from ethanol and methanol with modifications in cell design and electrode materials. The invented power source can be used in platinum catalyst loaded PEM type as well as the solid oxide electrolyte type electrolysis cells. The invented pressurized alkaline electrolysis device can also produce hydrogen and oxygen as a mixture. In that case, the stoichiometrically produced hydrogen and oxygen can be used for a welding application known as hydrogen welding, silver brazing and hard soldering in some industrial applications as an alternative to the acetylene torch welding. The produced oxy-hydrogen flame from manufactured electrolysis device can also be used in cutting thick steel slabs. Hydrogen produced purely from the device can be stored as energy carrier and it can be used as fuel directly in fuel cell research. Produced pure oxygen can be stored and it has an economical value.

Since fossil fuel based hydrogen production methods are more economical, the hydrogen production from water using classical type electrolysis devices has not been in large proportions. It is estimated that less than 5 % of hydrogen in the world is produced by electrolysis. The most part of this production takes place as a side product during the production of chlorine from salt water process in electrolysis plants. However, the technology of pressurized and high pressurized electrolysis systems has long been in use. The devices made for hydrogen production using catalyst loaded polymer electrolyte membranes or proton conducting membranes (PEM) has increased recently parallel to the developments in fuel cell industry. But, the production with these type designed membrane (Nafion) systems is not growing in industrial scale since it is less economic than the hydrogen production from natural gas, lignite and other fossil fuel sources. There have been various research and developments in hydrogen production recently. Solid oxide fuel cell technology related electrolysis is one of the most important and interesting research subjects. Similarly, high temperature water electrolysis has been studied for long time. Methods of hydrogen production from fossil Fuels using bacteria or other chemical reactions has also become more significant recently.

The subject of invention in technology of classical pressurized water electrolysis devices and systems have not experienced so many changes for long time. In general, there have not been major differences in alkaline modern industrial cells except various applications such as the

electrode separation distance optimization and coating nickel on steel electrodes and usage of more corrosion resistant materials. Asbestos diaphragms are mostly used as gas separators in industrial alkaline electrolysis devices. Asbestos membranes are usually supported mechanically by gasket frames and compressed between the electrodes to minimize current loss in the tight cell design. Ceramic and other polymeric based membrane separators have also been applied and studied as an alternative to asbestos. Two different electrode designs are generally used in industrial water decomposition applications. In general, a conductive electrolyte in liquid form along with water is used in cells to enable electrical conduction between parallel or serially placed metal electrodes in terms of power connection. It is important that the standard electrode potential of anion of electrolyte is to be higher than that of hydroxide anion and the standard electrode potential of cation of electrolyte to be lower than hydrogen. NaOH, KOH and H₂SO₄, which are some of the suitable salts of Li^+ , Rb^+ , K^+ , Cs^+ , Ba^{2+} , Sr^{2+} , Ca^{2+} , Na^+ , and Mg^{2+} , are generally used in industrial applications. The compositions of these electrolytes are in the concentration ranges usually in which the maximum electrical conductivity is achieved also with respect to temperature. The metal electrodes with different surface patterns and forms are mostly used in solid electrode selections. It is assumed that the nickel coated steel electrodes have one of the best corrosion resistances in alkaline media. Different types of power sources have been used for long time according to electrode placing and cell orientation. A potential difference, which is slightly above the thermodynamic threshold value of 1.23 volts, is usually applied between 1.8-2 volts for distilled water electrolysis. This potential difference is usually adjusted in parallel designed cells using a transformer type power source which applies a fully rectified DC current. However, voltage controlled power circuits which can adjust the potential drop in each cell are used in series type connected electrode systems. Power sources made with no transformer switch mode power supply (SMPS) technology can now be used in such high power and current demanding applications parallel to the recent advances in the electronic part manufacturing.

The invented power source is a fully rectified but not filtered DC voltage generator. The voltage can be adjusted between 100-220 volts and current can be applied up to 20 Amps without any problems. The problem of using large volume heavy transformer is solved with this invention. The gases produced in the cathode and anode compartments are separated using asbestos filters or membrane-gasket assemblies in the state of the art designs. Another related invention is also made on the gas separation electrode system design. The invention of a mechanically butyl rubber gasket supported fiber glass or rock wool frame membrane system which can be used instead of asbestos membrane is delineated in Figure 4. This invention can both be used in parallel and serial type pressurized electrolysis devices. The combination of the invented power source and a serial electrode connected alkaline electrolysis device which produces nonseparating hydrogen and oxygen mixture is described in Figure 2. The invention of metal electrode gasket system which can be used in that system is seen in Figure 3.

The produced devices in order to achieve the objects of the invention are detailed in figures below.

Figure 1. Power source and the general view of the electrolysis device

Figure 2. Serial pressurized type nonseparating electrolysis device

Figure 3. The electrode gasket design invention for nonseparating electrolysis devices

Figure 4. Membrane filter (gas separating filter) mechanism.

The parts in the figures are numbered and the definitions are given below.

- 1- Potentiometer (variable resistor) $1M\Omega$
- 2- R1 resistor 1000 ohm
- 3- R2 resistor 4700 ohm
- 4- Diac SM DB3
- 5- K1- capacitor 22nF/ 300v
- 6- K2- capacitor 22nF/ 300v
- 7- Circuit breaker- 20 A
- 8- Switch
- 9- 220 volt AC mains
- 10- Triac- bt136 or bta20
- 11- D1- diode 30 A/600V
- 12- D2- diode 30 A/600V
- 13- D3- diode 30 A/600V
- 14- D4- diode 30 A/600V
- 15- Ground fault current interrupter (GFCI) 40 A
- 16- Diode bridge lead
- 17- Diode bridge lead
- 18- Electrolyzer 15a, 3.5 kW-220V DC
- 19- DC power source circuit
- 20- Welding torch
- 21- Flashback arrestor
- 22- Gas washing bottle
- 23- Electrolyte overfill tank
- 24- Rubber gas hose
- 25- Type 316 stainless steel electrodes
- 26- Flexible butyl rubber gasket
- 27- Liquid ionic conducting electrolyte
- 28- Plexiglass support
- 29- Pressure regulating and liquid transfer openings
- 30- Gas transfer openings
- 31- Electrolyte filling lid
- 32- Stainless steel 316 type plates
- 33- Butyl rubber gasket
- 34- Gas transfer holes
- 35- Gas and liquid pressure level adjustment openings
- 36- "Poly ether sulfone" microporous membrane
- 37- Glass fiber- stone wool filter system
- 38- Butyl rubber flexible gasket
- 39- Gas separating filter system

The power source circuit given in Figure 1 operates with AC mains voltage at 50 Hz frequency. The electronic circuit adjusts the current intensity with potentiostat P(1). This circuit was applied successfully in a 4 kilowatt (kW) alkaline potassium hydroxide containing pure water electrolyzer. The triac was connected to a heat sink at these power levels. The power consumed by the load which is connected to the power circuit is controlled by conduction of triac with the pulse signals which are applied at the gate terminal. This is controlled by the diac which is connected to the gate terminal of the triac. The logarithmic potentiometer (1) operates as to adjust the voltage from the circuit. The potentiometer (1) is an adjustable 1 mega-ohm variable resistor. The R1 (2) which is a fixed value 1000 ohm linear resistor is serially connected to the potentiometer (1). The R2 (3) which is a fixed value 4700 ohm linear resistor is parallelly connected to the R1 (2).

The voltage from the potentiometer which is applied to the diac is adjusted by these resistors. Diac, R2 (3), K1 (5) and K2 (6) capacitors are connected in parallel. A circuit breaker (7) which is used as a fuse and safety switch is serially connected to a 220 V on/off switch (8) and the mains voltage (9). The two parallelly connected 300 V and 22 nano farad capacitors are connected to the mains (9) and the triac (10). Four equivalent D1 (11), D2 (12), D3 (13) and D4 (14) diodes in bridge configuration are connected to the 40 Amps ground fault current interrupter (GFCI) (15), the potentiometer (1) and the triac (10) terminal lead. The leads of diode bridge rectifier (16) and (17) are connected to a 3.5-4 kW power drawing electrolyzer (18).

The bridge rectifier provides full-wave rectification from an AC input. The adjustment of the resistance in potentiometer (1) causes diac (4) to apply its break over voltage to the gate terminal of triac (10) with different time delays. Triac (10) conducts when the break over voltage is being applied to its gate electrode causes a trigger. The time interval of power applied to or drawn by the electrolyzer is determined by the frequency of triggering pulses.

When power is given to the circuit, the charging and discharging times of the capacitor are governed by the $T = R \cdot C$ equation which are determined by the resistances of P potentiometer (1) and R1 (2) resistor. This time interval determines the triggering angle. This angle can not be higher than 90 degrees for a single capacitor. The second K2 (6) capacitor is used in the circuit to delay the triggering angle a little more. The total delay can not reach 180 degree. For this reason a diac (4) is added to the circuit. In this way the triac can be triggered between 0 and 180 degrees. The triggering angle changes with the adjustment of the dial of P potentiometer (1).

When the resistance of P potentiometer (1) is decreased, the voltage needed to trigger the diac is reached at K2 (6) in early stages of alternate. When the resistance of P potentiometer (1) is increased, the voltage needed to trigger the diac is reached at K2 (6) in latter stages of alternate. The current passes through the serial electrolysis cells when the voltage is a little lower than the maximum value of alternating mains voltage of 220 volts which is expected when 2 volts or less voltage drop is experienced for every cell according to the number of electrolytic cells. For example, a setup of 110 plate serial electrolysis device needs a voltage maximum of $110 \cdot 1.8 = 198$ volts because each cell requires approximately 1.8 volts. In other words, the electrolyzer will work when the power source allows the alternating voltages between 200 and 220 volts. Thermodynamically speaking, there is no electrolysis taking place in cells for the states when the alternating voltages are below 135 volts. However, the

electrolysis reactions don't happen up until 198 volts due to the electrical losses. When the potentiometer (1) is adjusted and the gas outflow is suddenly seen, it is understood that the required potential difference is reached and it is assumed that the direct currents are applied to the cells as DC pulse signals with certain time intervals. It is tested with experiments that application of time interval adjusted pulsating currents are effective in dispersion of gas bubbles from the surface without sticking or accumulating.

The electrolyzer shown in Figure 1 (18) is a water electrolyzer which includes alkaline or acidic electrolytes. It is made with roughly up to 110 equivalent serially placed 316 type stainless steel plates since the power source can be adjusted between 0-220 V ranges. This type of stainless steel materials is one of the most resistant steels to withstand alkaline environment. The stainless electrodes can be assumed as bi-polar electrically with opposite polarization at each surface. In fact, one side of the bi-polar plates has a higher electrical potential than the other because of a "IR" potential drop between the two faces. Because this potential drop occurs in every cell, every serial cell behaves as a single electrolysis cell. Actually, the voltage and the current taken from the power source is intermittent and spaced although it is the unidirectional. This intermittent voltage and current appears to be helping formation and dispersion of gas bubbles between the electrodes. Because of the ohmic and other losses, the evident generation of hydrogen and oxygen is observed approximately above 1.8 V at the surface of catalyst uncoated 316 types of stainless steel electrodes which is higher than the thermodynamic value of 1.23 V. The potentiometer in the power circuit is adjusted to apply 1.8 V and above to the cells in order to increase the gas production.

The device (18) which produces hydrogen and oxygen from 30 % potassium hydroxide with pure water by using the energy from the power source (19) is given in Figure 2. Gas separating membranes are not used in this device and oxygen and hydrogen which are produced in anode and cathode leave the system as a mixture. There is a permanent pressure of 0.7-1 atm in the device and the outflow gas from the torch tip has 33.3 % oxygen and 66.7 % hydrogen by volume in stoichiometric ratios. This gas mixture can be passed through a zeolite filter to separate hydrogen from oxygen for using in fuel cell applications. The separated oxygen can also be used purely in various applications.

The part which is called torch (20) is a metallic material with a mechanism which can hold steady flame for hydrogen welding. A cylindrical tube shaped part which is called flashback arrestor (21) is attached to the torch directly. This part prevents the flame on the torch tip from coming back. There are various types of this commercial product especially made for hydrogen. The part which is defined as gas washing bottle (22) is also used as an additional safety precaution to prevent the flame on torch tip from coming back inside to the pressurized electrolyzer unit. This part can be made out of metals and other alloys. Pure water is usually kept inside the gas washing bottle; however, the part must be isolated electrically from the electrodes or power circuit and the device chassis for safety reasons. The part which is called electrolyte overfill tank (23) is connected to the electrolyzer gas outlet and gas washing bottle (22) with rubber welding hoses (24) The functions of the electrolyte overfill tank are to hold excessive amount of liquid electrolyte material during the first filling of electrolysis cells and store the liquid electrolyte which may be spewed out from the cells because of the inside pressure in early stages of device operation. This part is also electrically isolated. A potassium hydroxide (27) 30 % electrolyte solution is filled in the spaces between rectangular fin like stainless steel electrodes (25) which are compressed from both sides and separated by butyl

rubber gasket frames (26). The plexiglass (28) support lid holds electrodes and rubber gaskets together compressed and allows the level of the liquid inside the electrolyzer to be checked. The holes (29)-(30) and grooves in metal electrodes (25) and plexiglass supports (28) are applied to control pressure inside the device and regulate the gas flow and the electrolyte (27) passage between the electrolytic cells. A pressure sensor can also be integrated with the alkaline electrolyte filling lid (31) which is a leak-proof Teflon lid which is used to add fresh electrolyte solution and pure water.

The invented stainless steel plate (32) butyl rubber (33) electrode system is given in Figure 3. This electrode system can be used in serial or parallel electrolysis systems. Butyl rubber gaskets are compressed carefully between 316 type stainless steel electrodes to prevent air and liquid leaks. The stainless steel electrodes are electrically isolated in a way by the rubber gaskets (33) that the plates do not touch each other. The electron transfer between the metal plates can only be made by the potassium hydroxide (27) ions in the liquid media. The gas transfer holes (34) in the invented design allow homogenous pressure distribution and free gas transfer between the electrolysis cells. The liquid transfer holes (35) at the bottom are designed in such a way to regulate gas and liquid pressure and homogeneous liquid distribution between the electrolysis cells.

The invented electrode membrane gasket assembly or the filter system which can be used in both parallel and serial connected electrolyzer systems is seen in Figure 4. This invention solves the problem of necessity for using asbestos or polymeric membranes. The invention is an alternative to the various polymeric and asbestos state of the art membranes as a filter (37) which is prepared by sandwiching glass fiber and/or stone wool between a Poly Ether Sulfone (PES) membrane (36). This apparatus, which is prepared by compressing a fiber material between hydrophilic or PES type membranes, prevents the passage of oxygen or hydrogen bubbles to other electrode side. Since the 1-2 micron micro pores of PES membrane allows electrical conductivity but not the passage of gas bubbles through the PES membrane, hydrogen and oxygen generated in anode and cathode leave the system separately without mixing. In order to support the filter system, flexible butyl rubber gasket frames (38) are designed to be sandwiched between the metal electrodes and the filter system on the both sides of PES membrane to achieve water-tightness. This invention can be used in both serial and parallel type design electrolysis systems. PES polymeric membrane (36) and 316 type stainless steel plates are very resistant to the alkaline environments. Similar hydrophilic membranes with 1-2 micron micro pore sizes which are also resistant to alkaline environments can be used for the same purposes.

What is Claimed is:

1. It is a DC power source (19) circuit system for pressurized alkaline or acidic electrolyzers which delivers power from mains AC to parallel or serial type electrodes in terms of pulsating unidirectional waves without using a transformer and adjusts the high current and voltage to the electrolysis cells by potentiometer and diac-triac.
2. It is a water electrolysis device (18) which uses alkaline electrolyte (27) and 316 type stainless steel electrodes (32) to generate hydrogen and oxygen electrochemically by using the power source (19) in Claim 1.
3. It is a electrode gasket design for water electrolysis devices without gas separation mentioned in Claim 2, which includes gas transfer holes (34), and liquid level and gas pressure regulating holes (35) and the design of separated stainless steel electrodes with butyl rubber gaskets in pressurized cells.
4. It is a filter system (39) for devices made by using Claim1 and Claim2 which prevents from mixing of generated hydrogen and oxygen in anode and cathode by using a system which places 1-2 micron micro porous PES membrane sandwiched glass fiber or stone wool and butyl rubber gaskets from both sides of the membrane in between the metal electrodes

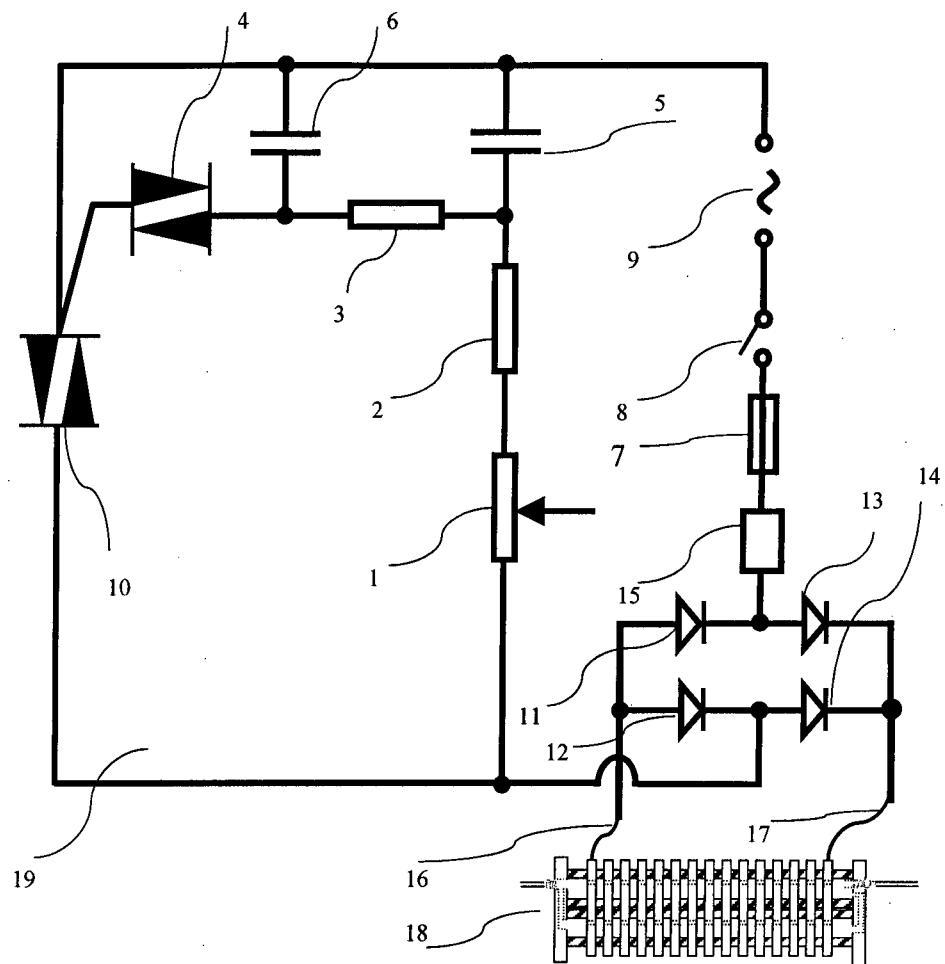


FIGURE 1

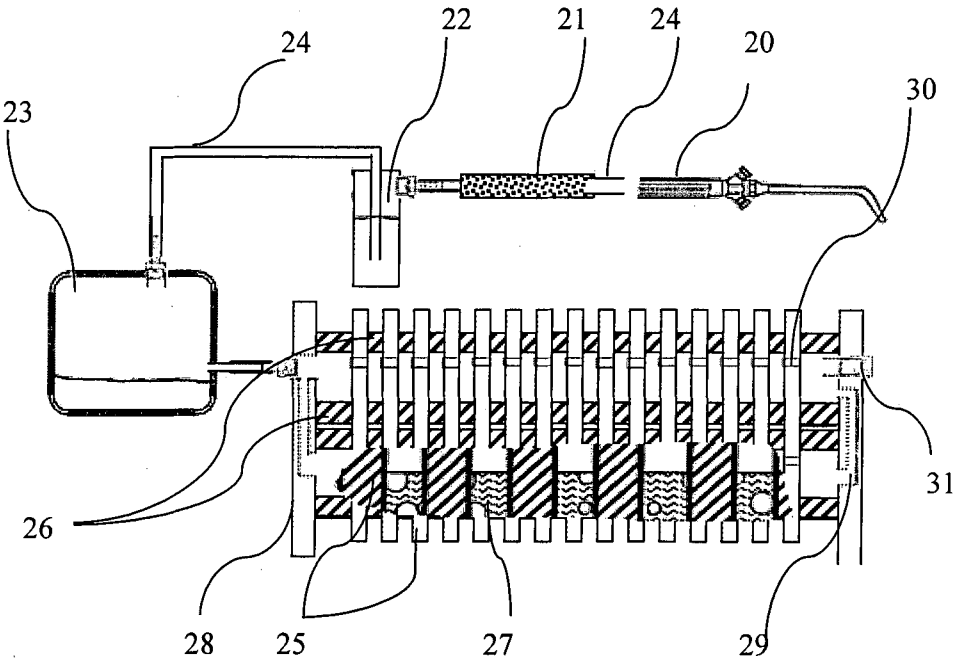


FIGURE 2

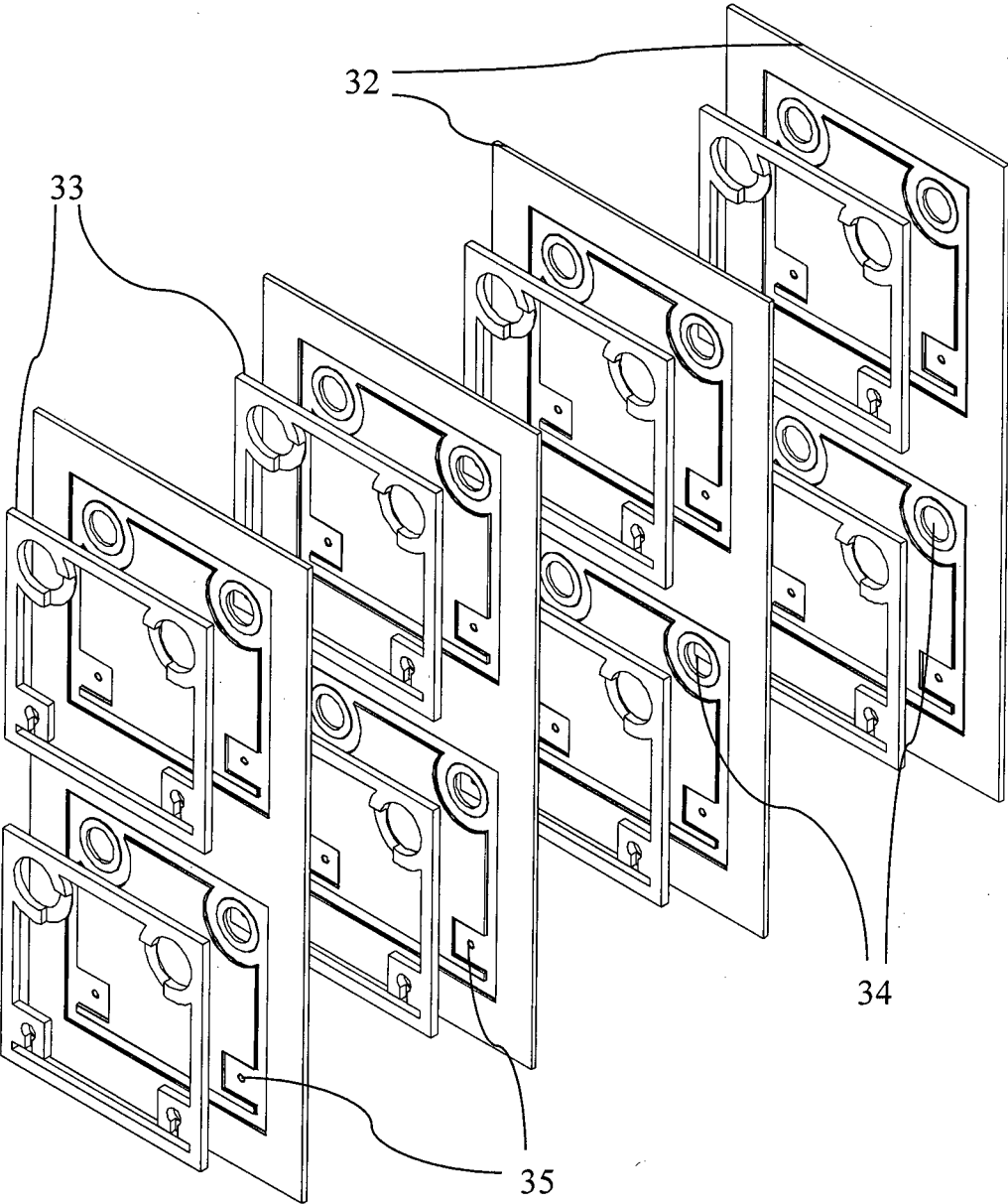


FIGURE 3

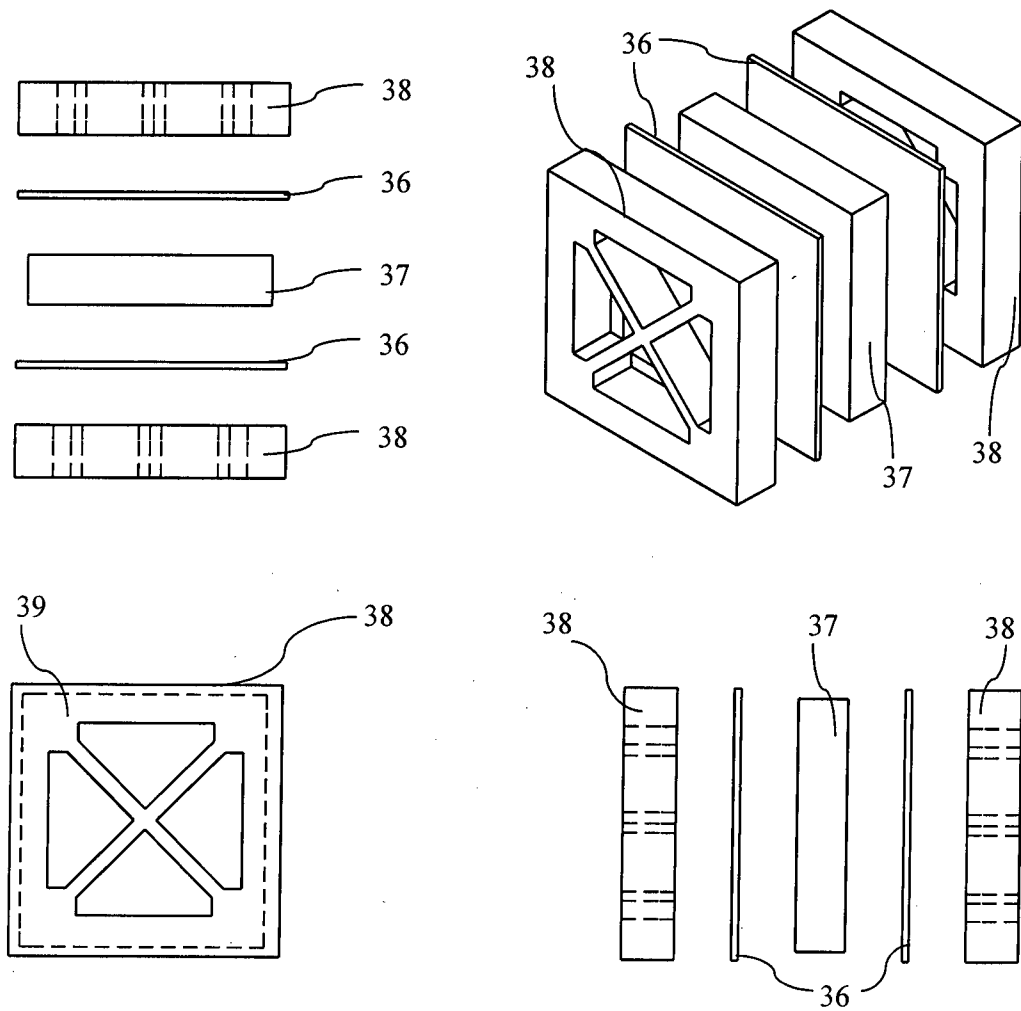


FIGURE 4

INTERNATIONAL SEARCH REPORT

International application No
PCT/TR2014/000067

A. CLASSIFICATION OF SUBJECT MATTER

INV. C25B13/04 C25B15/02 C25B1/12 C25B9/02
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)

C25B H02M

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)

EP0-Internal

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	CN 101 289 748 A (BAOCHI ZHANG [CN]) 22 October 2008 (2008-10-22)	1
Y	abstract; figure 1 -----	2
X	CN 2 795 234 Y (XU ZHIHENG [CN]) 12 July 2006 (2006-07-12)	1
Y	abstract; figure 1 -----	2
Y	US 4 235 694 A (HALL FREDERICK F [US]) 25 November 1980 (1980-11-25) column 1, line 40 - column 2, line 26 -----	2
A	US 2007/251830 A1 (CONRAD WAYNE E [CA]) 1 November 2007 (2007-11-01) paragraph [0221] - paragraph [0223] paragraph [0249]; figure 7 ----- -/--	1,2



Further documents are listed in the continuation of Box C.



See patent family annex.

* Special categories of cited documents :

"A" document defining the general state of the art which is not considered to be of particular relevance

"E" earlier application or patent but published on or after the international filing date

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"P" document published prior to the international filing date but later than the priority date claimed

"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

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"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

"&" document member of the same patent family

Date of the actual completion of the international search

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Date of mailing of the international search report

04/02/2015

Name and mailing address of the ISA/

European Patent Office, P.B. 5818 Patentlaan 2
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Authorized officer

Hammerstein, G

INTERNATIONAL SEARCH REPORT

International application No
PCT/TR2014/000067

C(Continuation). DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	US 5 843 292 A (SPIROS SPIRO ROSS [AU]) 1 December 1998 (1998-12-01) column 8, line 66 - column 9, line 3; claim 1; figure 9 -----	1,2

INTERNATIONAL SEARCH REPORT

International application No.
PCT/TR2014/000067

Box No. II Observations where certain claims were found unsearchable (Continuation of item 2 of first sheet)

This international search report has not been established in respect of certain claims under Article 17(2)(a) for the following reasons:

1. ☐ Claims Nos.:
because they relate to subject matter not required to be searched by this Authority, namely:
2. ☐ Claims Nos.:
because they relate to parts of the international application that do not comply with the prescribed requirements to such an extent that no meaningful international search can be carried out, specifically:
3. ☐ Claims Nos.:
because they are dependent claims and are not drafted in accordance with the second and third sentences of Rule 6.4(a).

Box No. III Observations where unity of invention is lacking (Continuation of item 3 of first sheet)

This International Searching Authority found multiple inventions in this international application, as follows:

see additional sheet

1. ☐ As all required additional search fees were timely paid by the applicant, this international search report covers all searchable claims.
2. ☐ As all searchable claims could be searched without effort justifying an additional fees, this Authority did not invite payment of additional fees.
3. ☐ As only some of the required additional search fees were timely paid by the applicant, this international search report covers only those claims for which fees were paid, specifically claims Nos.:
4. ☒ No required additional search fees were timely paid by the applicant. Consequently, this international search report is restricted to the invention first mentioned in the claims; it is covered by claims Nos.:

1, 2

Remark on Protest

- ☐ The additional search fees were accompanied by the applicant's protest and, where applicable, the payment of a protest fee.
- ☐ The additional search fees were accompanied by the applicant's protest but the applicable protest fee was not paid within the time limit specified in the invitation.
- ☐ No protest accompanied the payment of additional search fees.

INTERNATIONAL SEARCH REPORT

International Application No. PCT/ TR2014/ 000067

FURTHER INFORMATION CONTINUED FROM PCT/ISA/ 210

This International Searching Authority found multiple (groups of) inventions in this international application, as follows:

1. claims: 1, 2

Claims 1 and 2 are directed towards a DC power source circuit and a water electrolysis device comprising the said power source circuit.

2. claims: 3, 4

Claims 3 and 4 are directed towards an electrode gasket and filter system for a water electrolysis device.

INTERNATIONAL SEARCH REPORT

Information on patent family members

International application No

PCT/TR2014/000067

Patent document cited in search report	Publication date	Patent family member(s)	Publication date
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