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Published:

— *with international search report (Art. 21(3))*

(54) Title: PEM-TYPE ELECTROLYZER STACK FOR OPERATION AT HIGH PRESSURE

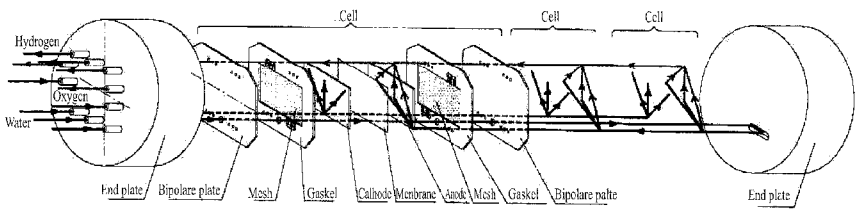

FIG. 6

(57) Abstract: Electrolyzer module comprising a front plate, end plate enclosing an anode cavity, and a cathode cavity.

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PEM-TYPE ELECTROLYZER STACK FOR OPERATION AT HIGH PRESSURE

FIELD OF THE INVENTION

A PEM Electrolysis module is a filter- pressure design (STACK) collected from the cells, sandwiched between the end plates with bolts. An Electrolyzer has an internal communication system associated with the external system of water regime providing and gas separators. Internal communication is facilitated by channels that are formed by the connections of individual parts by doing in these parts a number of coaxial holes. Elements of the cells are sealed with gaskets from made of fluorine rubber.

STATE OF THE ART

The improvement and optimization of a PEM electrolyzer efficiency and operative life operating at high pressures is linked to the following:

- Membranes with low ion-exchange capacity
- The technology of the MEA manufacturing with structural improvements in order to increase contact with the current collectors.
- Optimization of the mesh elements design, gaskets, bipolar plates, as well as system of supply and distribution of reagents to the cell contours.
- Providing a more uniform distribution of water on the active surface of the electrolyzer cells
- Providing rapid removal of electrolysis gases from the cathode and anode spaces.
- Providing a minimum electrical contact resistance of electrolysis cell elements and minimum degradation

Hydrogen occurrence in oxygen may in some cases accompanied by decreasing of a current efficiency (up to 50% at smaller currents) relative to theoretical value, which is obtained at the assumption of losses from a crossover. Hence, processes, which lead to hydrogen formation in anode area, may be electrochemical character

and directly accompany anode processes of water decomposition. Hydrogen concentration in electrolytic oxygen cannot be caused only membranes gas permeability; It is possible that the hydrogen in electrolyzer anode space is formed as a result of electrochemical processes directly in anode catalyst layer or corrosion processes on electrolyzer design elements. It should be noted that the specific reasons for such processes to the end and not yet clarified the issue continues to be relevant.

Uniform distribution of water on the active surface of the cell provides a uniform release rate of electrolysis gases formation. This eliminates the possibility of the formation of stagnant zones, and increases efficiency and life of work, improves the purity of electrolysis gases and prevents accidents related to local heat, causing destruction of the elements of MEA.

Quickly remove of electrolysis gases from the cathode and anode spaces provides increased water content in the cathode and the anode space of the electrolyzer, which contributes to improving the efficiency of electrolysis.

The minimum contact resistance of electrolysis cell design elements at operating conditions provides the best electrical characteristics of the cell and, consequently, increases its efficiency as a whole.

For obtain of pure electrolytic gases it is necessary to improve MEA manufacturing techniques (improve the homogeneity of the catalytic layers structure, clarification of requirements for the degree of

Dependence of membrane gas permeability from temperature has exponential increase and increases approximately by 5 times at increase temperature from 40°C to 80°C while hydrogen concentration in anode area almost does not depend from temperature.

BRIEF DESCRIPTION OF THE DRAWINGS

To complete the description and in order to provide for a better understanding of the invention, a set of drawings is provided. Said drawings illustrate a preferred embodiment of the invention, which should not be interpreted as restricting the

scope of the invention, but just as an example of how the invention can be embodied.

Fig.1 Shows the electrolysis module in the backpressure case where the electrolyzer stack. is located.

Fig. 2 Shows the end plate with a handing out channels to be implemented in the present invention

Fig. 3. Shows a gasket with the holes and channels with conical to be implemented in the present invention

Fig. 4. Is a representation of a mesh insert to be implemented in the present invention

Fig. 5. Show the mesh insert with dividing walls to be implemented in the present invention.

Fig .6. Show the wáter and gases flows in the electrolisis stack with the components of the present invention

DESCRIPTION OF THE INVENTION

The invention is related to several improvements in the design and construction of components and manufacture techniques of the PEM electrolyzer stack components that solve the majority of the technical issues that have influence in the gas production at high pressures up to 500 bars.

The invention provides a PEM electrolyzer working at high pressure, placed in a backpressure case **(fig.1).** The case is filled by an inert gas (or hydrogen) at pressure, which corresponds to the pressure inside the electrolyzer. This design and operating mode has been described in our Patent P**200900163.**

Because the distribution and collection of reagents in the electrolysis module is made from the front side, there is a need to direct flows towards the rear of the electrolysis module. To do this, the invention presents an end plate with milled grooves**. (fig. 2)** with this end plate design, distribution and collection of reagents is performed from the two sides of the plate.

In order to obtain a more uniform distribution of water on the active surface of the cell and more efficient removal of electrolysis gases from the cathode and anode space by optimization of sizes, amount and configuration of the input and output of gases and water, the invention presents in a particular embodiment, a configuration of the gaskets with conical windows for inserts of a conical shape. **(fig. 3).** The configuration of distributing holes in gaskets, depends on the amount of generated gas (ie, the number of cells in the electrolysis module and pressure) can be made from two to eight over-sized holes. The size of the slot channels of the upper and lower inserts made different: in the upper groove gasket are installed inserts with E = 1.00 mm tp 2,8 mm, in the lower grooves are installed inserts with E = 0,70 mm to 1.50 mm. This allows to provide the necessary backpressure to improve the uniformity of reagents and flow distribution on the cross sections of electrolysis cells.

To provide uniform water distribution in the inner cavities of the electrolysis module cells and to reduce the "stagnant" zones in gasket construction in a particular embodiment, the invention is provided with slotted holes that form additional cross-cutting channels. To reduce dead zones, two to four channels , depending on the amount of generated gases, for distribution and collection reagents are provided, and the holes have a shape such as to direct the flow towards the gaskets corners. **(fig.3)**

To reduce corrosion at the center of the bipolar plates and the corners of porous current collectors, the inner corners of the gasket windows are rounded, with a predetermined curvature radius

Deposition of the catalytic layer directly on the membrane results in improvements in the homogeneity of the catalytic layer's structure, highly degree of purity of reagents and materials, specially when the process is carried on with a current density of 1A/cm^2 and voltage lowered to 100mV for the cell, or at a current density of 2A/cm^2– for 200mV. Hydrogen concentration in electrolytic oxygen falls under this conditions approximately a 10%.

In order to lower the electrical contact resistance of the mesh inserts of the electrolyzer (fig. 4)

The first and third layer of the grids are immediately rolled "in size" and have a smooth surface, and the second layer, located between them, has a different thickness without smooth surfaces After welding of the grid layers "rolled on rollers to the desired size", which allows improving the contact resistance between the layers of grids and reduces the contact resistances. With the use of this technology object of the invention we reduce the contact electrical resistance of the electrolysis cell construction is reduced from 25 mΩ/cm^2 to 15 mΩ/cm^2, which corresponds to the decrease in cell voltage at a current of 1 A/ cm^2 at 10 mV.

Another variant of the mesh inserts construction described above **(fig.5)** may be with dividing walls for optimization of water distribution. The presence of walls directs the water flow to the right direction and eliminates the appearance of stagnant zones. This design is more effective in cells with a large work surface.

In **fig 6.** is presented the scheme of water and gas flows inside the electrolysis stack manufactured with the elements of the present invention. Water is supplied to the anode channel through 3 lower front flange fittings,. After that, water is decomposed on the membrane and oxygen and part of remaining water passes to the rear end plate at its upper part, and through channels made in the end plate, turns to the side of the front flange and is expelled through two upper lateral connections. Water is supplied through 2 bottom side connections to the cathode cavity of the electrolysis stack ,then passes to the back plate where the flow turns in the opposite direction and is distributed over the cathode cavity. The resulting decomposition hydrogen input into the upper hole of the electrolyzer stack components and comes out through the top 3 fittings in the front flange.

CLAIMS

1.-PEM Electrolyzer stack in which the water is supplied to the **anode cavity** through 3 lower front flange fittings,. After that, water is decomposed on the membrane and oxygen and part of remaining water, passes to the rear end plate at its upper part, and through channels made in the end plate, turns to the side of the front flange and is expelled through two upper lateral connections. Water is supplied through 2 bottom side connections to **the cathode cavity** of the electrolysis stack, and then passes to the back plate where the flow turns in the opposite direction and is distributed over the cathode cavity. The resulting decomposition hydrogen flows into the upper hole of the electrolyzer stack components and comes out through the top 3 fittings in the front flange. **(fig 6) .**

2.- PEM Electrolyzer stack in accordance with claim 1. further comprising gaskets made of Fluor rubber or with Fluor rubber with a cover of Teflon film, the gaskets further comprising windows, slots and channels at their sides with conical windows for inserts of a conical shape, each channel having 3 distributing holes, the wholes being of a bigger size as the windows, the configuration of number of distributing holes, depends on the amount of generated gases and can be from two to eight. The size of the slot channels of the upper and lower inserts have different size: in the upper groove gasket inserts with E = 1.00 mm tp 2,8 mm, in the lower grooves inserts with E = 0,70 mm to 1.50 mm. **(fig 3),**

3.- PEM Electrolyzer Stack in accordance with any of the previous claims in which the end plates made with titanium with milled grooves.**(fig 2)**

4.- PEM Electrolyzer module in accordance with any of the previous Claims , Claim 1. in which Gas Distribution layers made with Titanium or Composites of Titanium and Carbon in the cathode region, in which the first and third grids layer immediately rolled "in size" and have a smooth surface and a second layer, located between the two, has a margin thickness. **(fig 4)**

5.- Electrolyzer module in accordance with Claim 4. in which Gas Distribution layers incorporated dividing walls **(fig.5)**

6- Electrolyzer module in accordance with any of the previous claims, further comprising current collectors and bipolar plates made with titanium or Composites of Titanium and Carbon which present a curvature radius on the inner corner.

7.- Electrolyzer module in accordance with any of the previous claims , in which the bipolar plates made with titanium or Composites of Titanium and Carbon have a curvature radius on the inner corner.

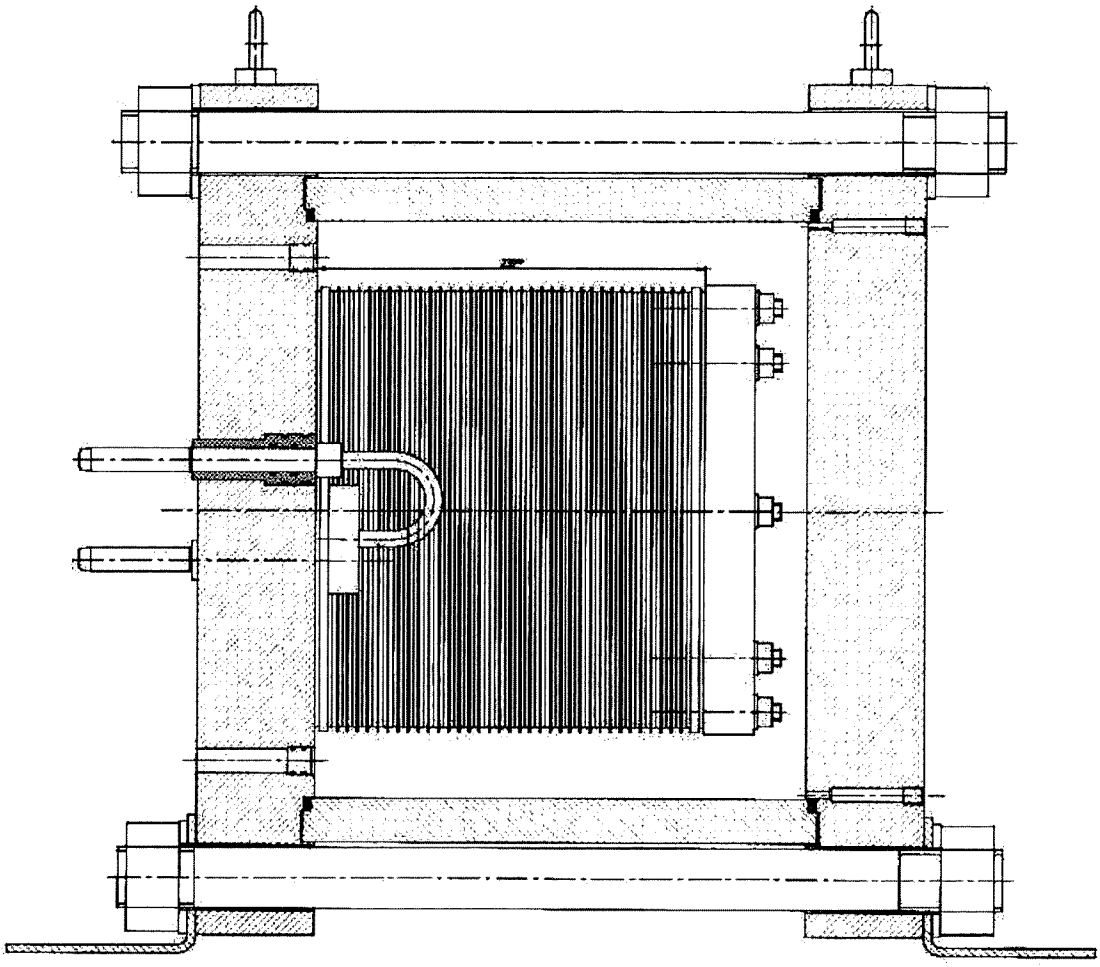

FIG. 1

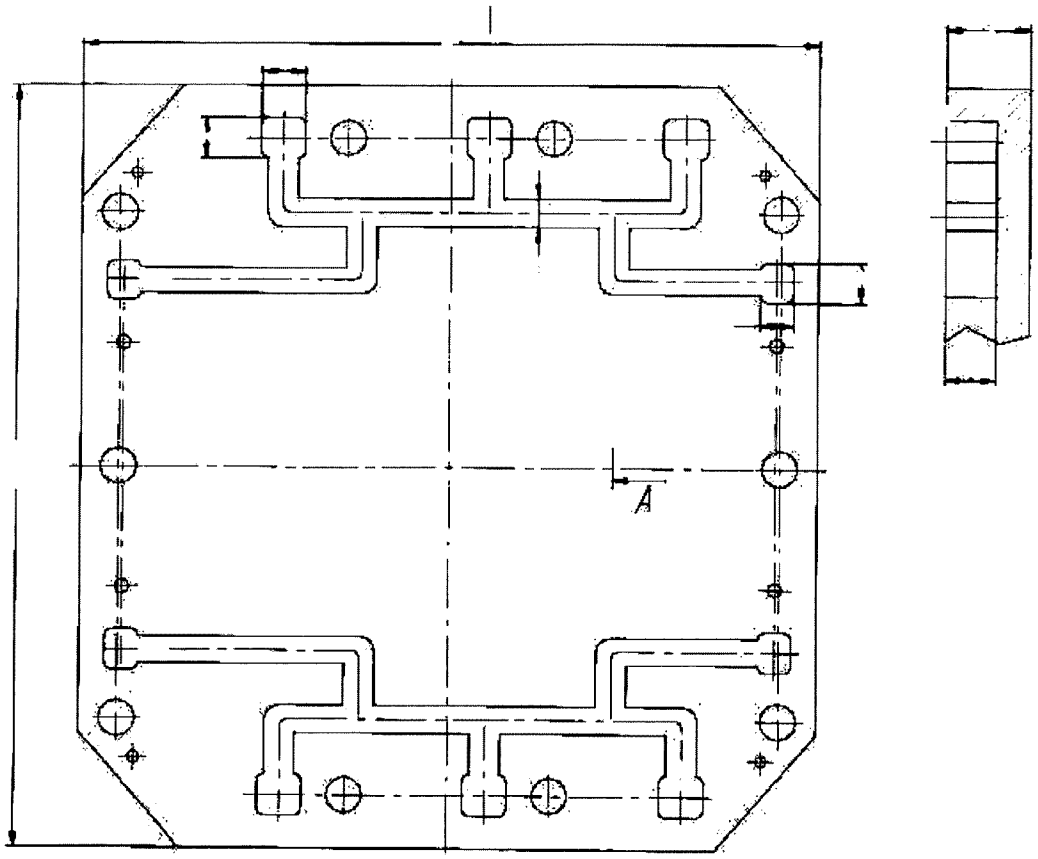

FIG. 2

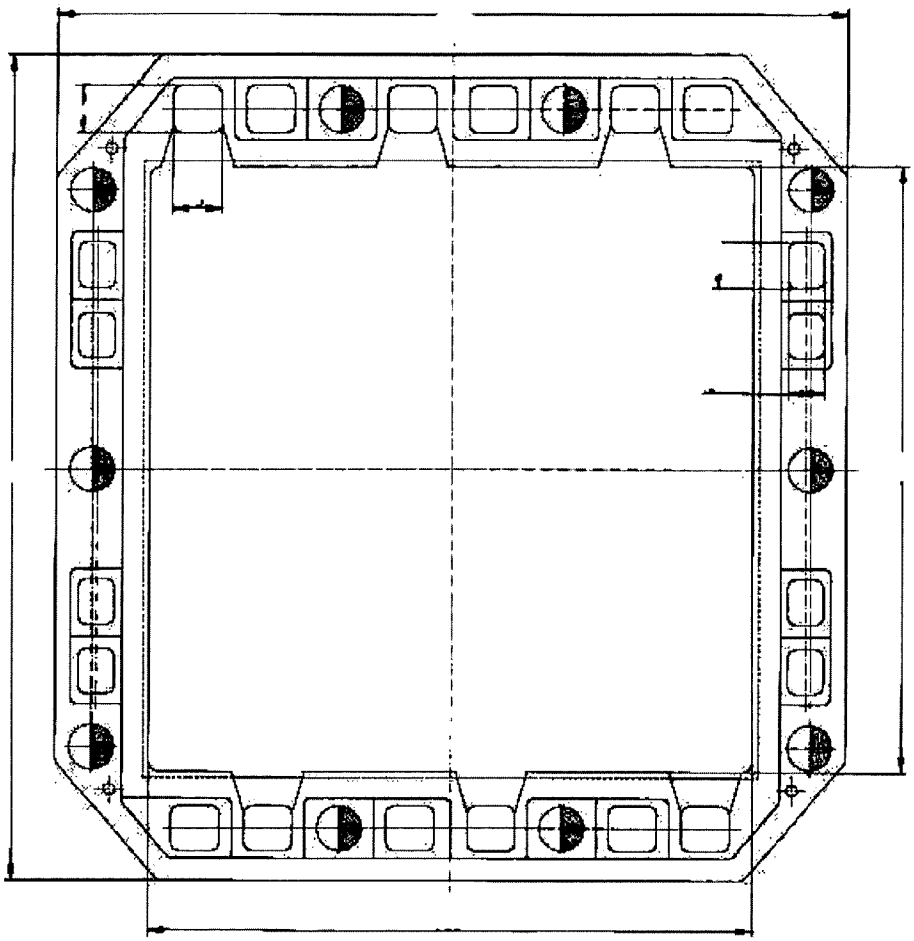

FIG. 3

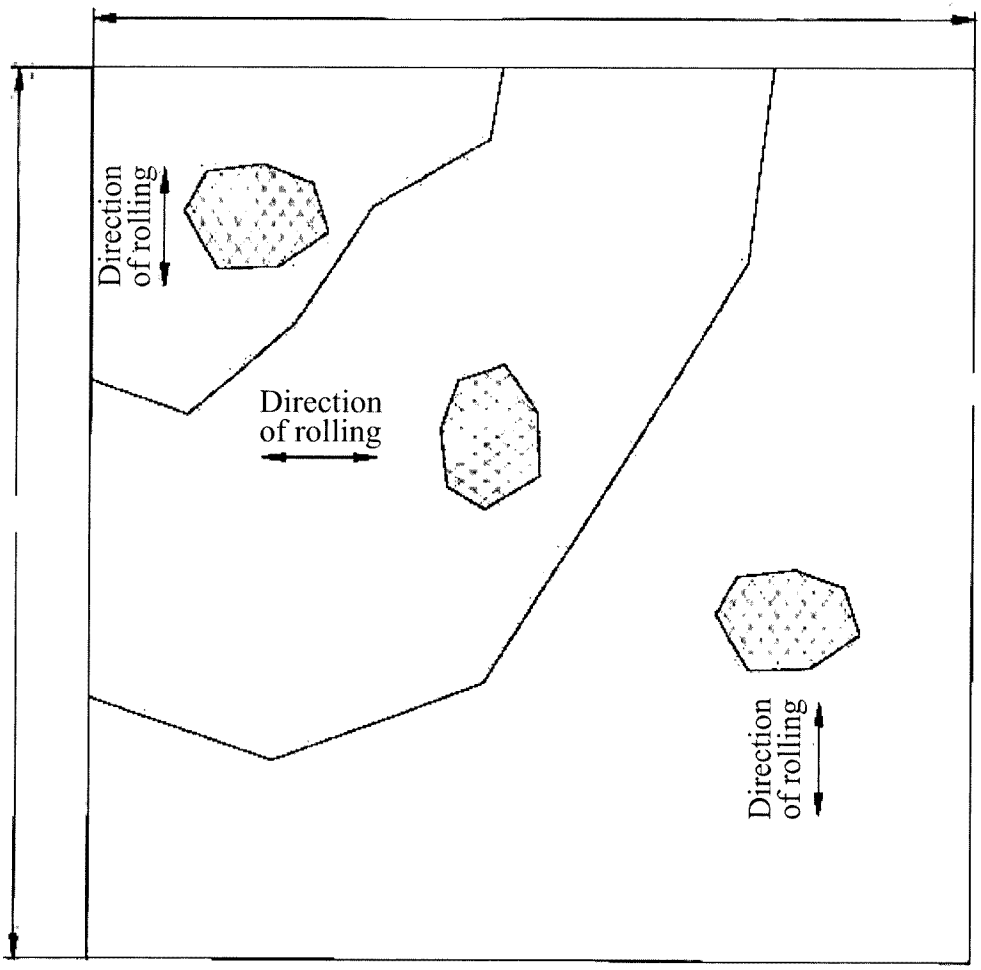
Direction of rolling
Direction of rolling
Direction of rolling

FIG. 4

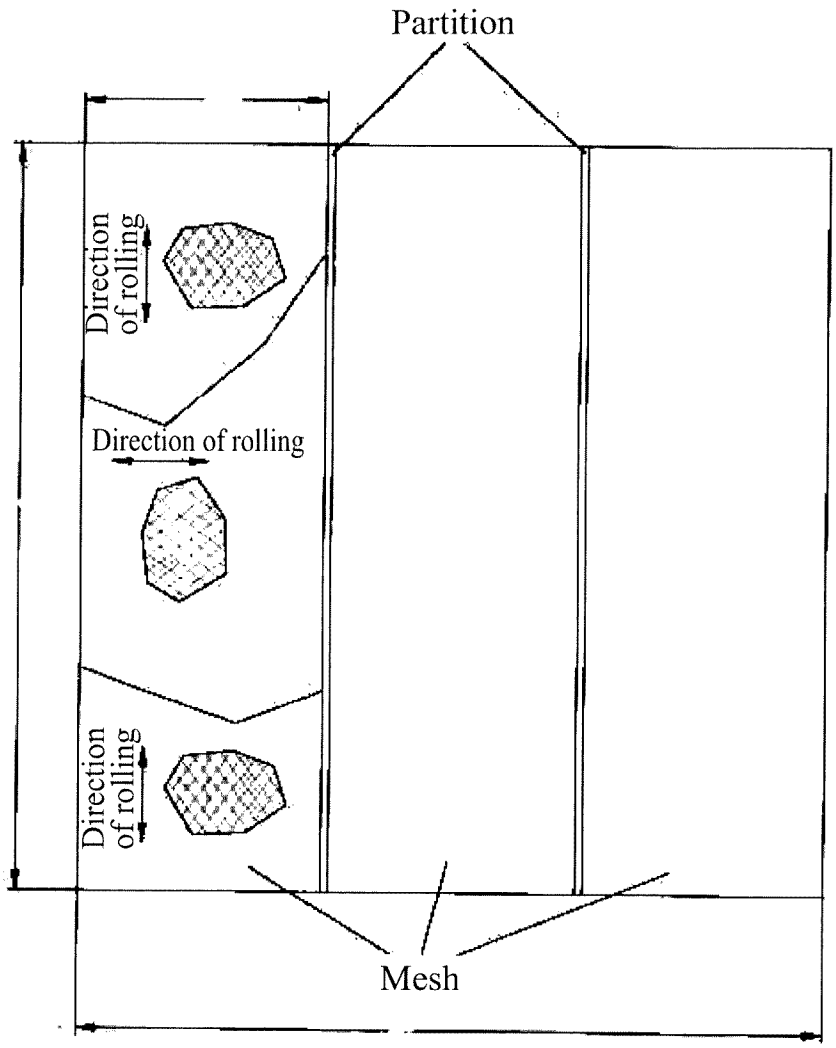
Partition
Direction of rolling
Direction of rolling
Direction of rolling
Mesh

FIG. 5

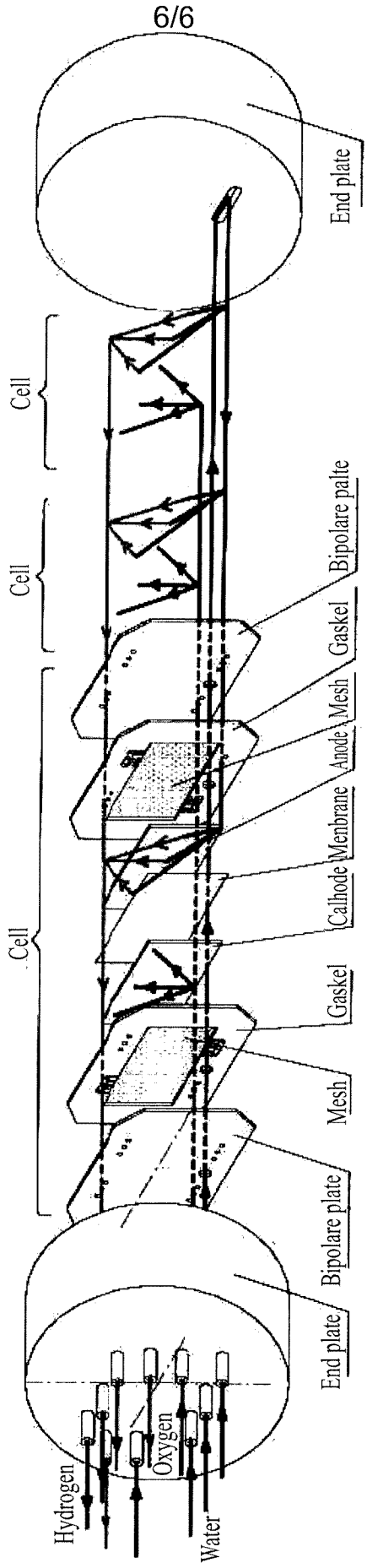
End plate
Cell
Cell
Cell
Bipolare palte
Gaskel
Mesh
Anode
Menbrane
Calhode
Gaskel
Mesh
Bipolare plate
End plate
Hydrogen
Oxygen
Water

FIG. 6

INTERNATIONAL SEARCH REPORT

International application No.
PCT/EP2012/072982

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.: 1-7(partially)
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:
see FURTHER INFORMATION sheet PCT/ISA/210

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:

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.:

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/EP2012/072982

A. CLASSIFICATION OF SUBJECT MATTER
INV. C25B15/08 C25B1/04
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

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)
EPO-Internal, WPI Data

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	US 5 401 371 A (OSHIMA YUJIRO [JP] ET AL) 28 March 1995 (1995-03-28) claims 1-3 column 2, lines 31-47 column 8, lines 17-19 -----	1-7
X	EP 0 229 473 A1 (ICI PLC [GB]) 22 July 1987 (1987-07-22) claims 24,26 figure 7 column 9, line 45 - column 10, line 10 column 11, lines 17-26 column 14, line 24 - column 15, line 52 -----	1-7
X	US 4 720 331 A (BILLINGS ROGER E [US]) 19 January 1988 (1988-01-19) claims 18,24,30-34 figures 1-2 -----	1-7

-/--

[X] Further documents are listed in the continuation of Box C.

[X] 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

"L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)

"O" document referring to an oral disclosure, use, exhibition or other means

"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

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

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

"&" document member of the same patent family

Date of the actual completion of the international search	Date of mailing of the international search report
8 July 2013	16/07/2013

Name and mailing address of the ISA/	Authorized officer
European Patent Office, P.B. 5818 Patentlaan 2 NL - 2280 HV Rijswijk Tel. (+31-70) 340-2040, Fax: (+31-70) 340-3016	Perednis, Dainius

INTERNATIONAL SEARCH REPORT

International application No
PCT/EP2012/072982

C(Continuation). DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	DE 10 2009 044144 A1 (ALFRED WALTHER METALLWARENFABRIKATION KUNSTHANDWERK E K [DE]; STRAEUSL) 7 April 2011 (2011-04-07) claims 1,33,34,36 figure 4 paragraph [0060] -----	1-7

INTERNATIONAL SEARCH REPORT

Information on patent family members

International application No
PCT/EP2012/072982

Patent document cited in search report	Publication date	Patent family member(s)	Publication date
US 5401371 A	28-03-1995	JP 3278909 B2	30-04-2002
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EP 0229473 A1	22-07-1987	AU 585104 B2	08-06-1989
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		ZA 8609109 A	26-08-1987
US 4720331 A	19-01-1988	NONE	
DE 102009044144 A1	07-04-2011	NONE	

Continuation of Box II.2

Claims Nos.: 1-7(partially)

The language used in the claims 1-7 is not clear to such an extent that it causes difficulty in determining the scope of claims. The description and drawings do not contribute to the clarification of claims. Furthermore, most of the features in the apparatus claim 1 relate to a method of using the apparatus rather than clearly defining the apparatus in terms of its technical features. The terms "immediately rolled in size", "a margin thickness", and "curvature radius on the inner conner" used in claims 4, 6 and 7 are vague and unclear. This makes it impossible to compare the claims 1-7 to the prior art. As a result, the application does not comply with the requirement of clarity under Article 6 PCT.

The applicant's attention is drawn to the fact that claims relating to inventions in respect of which no international search report has been established need not be the subject of an international preliminary examination (Rule 66.1(e) PCT). The applicant is advised that the EPO policy when acting as an International Preliminary Examining Authority is normally not to carry out a preliminary examination on matter which has not been searched. This is the case irrespective of whether or not the claims are amended following receipt of the search report or during any Chapter II procedure. If the application proceeds into the regional phase before the EPO, the applicant is reminded that a search may be carried out during examination before the EPO (see EPO Guidelines C-IV, 7.2), should the problems which led to the Article 17(2) declaration be overcome.