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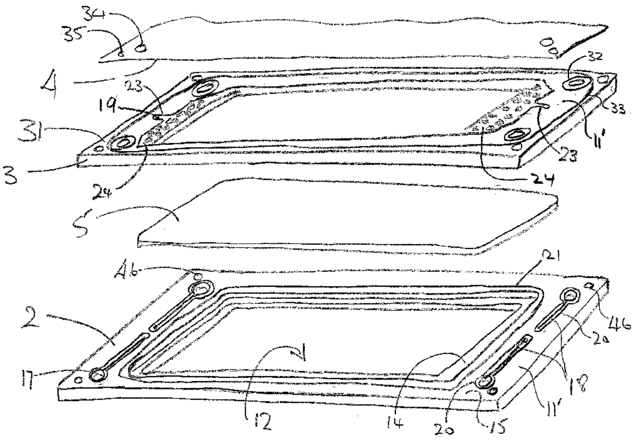
(54) Title: ELECTROCHEMICAL CELL STACK WITH FRAME ELEMENTS



(57) Abstract: A redox fuel cell stack (1) comprises a plurality of essentially similar half-cell frames (2,3) of moulded polymer. Interleaved between them are semi -permeable membranes (4) and bipolar plate electrodes. The frames (2, 3) are rectangular, with margins (11) around central voids (12). At the voids, they have rebates (14) in abutting faces (15) for locating the plate electrodes. At their corners, they have apertures (17) for forming ducts throughout the stack for flow of electrolyte to and from the cell cavities provided by the voids (12). The frames (2) have electrolyte flow passages (18) open in their faces 15 abutting the frames 3 and leading towards each other. The passages stop short of each other and are surrounded by grooves containing sealing O-rings (20). Diagonally opposite ones of the passages (18) end at openings (22) passing through the frames (2). The other passages have no openings in the frames (2), but the frames (3) have openings through them in register with the ends of the passages. On the other side of the frames (2) and (3), the openings (21, 22) open into short passages (23) directed towards the central voids and debouching into electrolyte distribution rebates (24). Thus electrolyte can flow from one duct aperture (17) in one corner, via the passage (18) from the aperture, either through the frame (2) via the opening (21) or the frame (3) via the opening (22), through short passage (23) and the respective distribution rebate and into the central void to whichever side of the plate electrode it was directed by the opening (21, 22). From the opposite end of the central void, the electrolyte is lead back into the diagonally opposite duct aperture

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ELECTROCHEMICAL CELL STACK

The present invention relates to a stack of electrochemical or electrolytic cell, in particular though not exclusively to a regenerative reduction/oxidation (redox) fuel cell stack.

Electrochemical cells are known which consist of typically between two and fifty alternate positive and negative half cells, although greater numbers are not unknown; since the cells components are stacked together, such plurality of half cells is typically known as an electrochemical stack or an electrolytic cell stack, often shortened simply to "a stack". Significant factors in the design of such a cell stack are the method of construction and thickness of the individual cells. Typical arrangements use what is known as a filter press design comprising within each cell successive layers of a non-conductive gasket material. The layers comprise frames, which provide accommodation for electrode material and also contain within their thickness electrolyte flow distribution passages. Each frame is assembled into one of two types of one half cell – positive and negative; it is noted that in general the design of frames for both positive and negative half cells is essentially similar and their assignment as either is a consequence of the overall construction and use of the stack rather than any inherent characteristic. These frames are typically interleaved alternately with sheets of a suitable electrode material and a suitable membrane separator. This construction produces a succession of half-cell pairs in series with electrodes common to two half cells, whence the electrodes are referred to as bipolar electrodes. It is also possible and desirable in some applications to connect electrically to the intermediate electrodes and, depending on the internal electrolyte distribution arrangement, operate the cells in various other series and/or parallel manners when some or all of the electrodes may be unipolar rather than bipolar.

Since the frames must provide a number of different features, including hydraulic sealing, mechanical strength, accommodation of the electrode and flow distribution passages, these passages being required to provide both isolation against internal shunt currents and conversely minimal flow resistance and uniform flow distribution, a design compromise between features is usually required. In particular, it is known to be desirable to achieve high linear flow velocity of electrolyte within the cell, which implies small cell spacing but since the frame thickness defines the spacing this in turn has the undesired effect of reducing the depth available for the distribution passages which are typically indented into one or other surface of the frames. Furthermore, it is known that for efficient and reliable cell performance, closure of the distribution channels within such frames must be achieved such as to prevent undesirable and potentially damaging paths for both hydraulic and electrical current leakage

The object of the present invention is to provide an improved electochemical cell stack.

According to the invention there is provided an electrochemical stack cell comprising a plurality of cells arranged side-by-side in a stack, each cell having:

- a membrane,
- a first half cell cavity on one side of the membrane and a second half cell cavity on the other side of the membrane,
- a respective electrode plate at the side of each half cell opposite from the membrane, each electrode plate providing contact between adjacent cells at least for intermediate ones of the cells,
- a pair of frames, one for one half cell and the other for the other, the frames:
    - captivating the membrane between themselves,
    - locating the electrode plates and
    - having:
        - continuous margins around central voids providing the half cell cavities,
        - apertures in the continuous margins providing ducts for flow of electrolyte through the stack for distribution to the cells,
        - electrolyte distribution rebates at opposite inside edges of the margins and
        - passages in the continuous margins for electrolyte flow from one of the duct apertures, into and out of the half cell at the distribution rebates and to another of the duct apertures,

wherein:

- each plate electrode is captivated between a frame from one cell and a frame from an adjacent cell with at least two portions of the margins of these frames extending outside respective edges of the plate electrode, the adjacent cell frames having faces which abut at the portions;
- the flow passages are formed in the faces of the margins and are closed by abutting opposite frame faces; and
- through-frame openings are provided in the frames for extending the passages from the abutting faces of the frames to the other, membrane side of the frames into distribution rebates.

Although other configurations are possible particularly curved or polygonal having rectilinear opposite margins with the electrolyte duct apertures arranged at the corners, normally the frames will be rectangular, i.e. having four straight margins, with the electrolyte duct apertures arranged at the corners. The flow passages can be distributed into all four margins, however, they are preferably provided in two opposite margins only. It is possible to provide all the passages in the face of one of each pair of abutting face frames, i.e. with two passages in each marginal portion having passages with one through frame opening in the portion at the end of one of the passages and another said opening in the other frame opposite the end of the other passage. Alternatively, one passage only can be provided in each marginal portion having a said flow passage. Conveniently each flow passage then has an opening through the frame; or all openings can be provided in the opposite marginal portion. The passages can be provided such that the frames have symmetry about a central axis transverse the plane of their abutting faces; or the passages can be arranged to extend from two duct apertures at neighbouring corners of the frame, with the passages extending in the marginal portions extending away from the margin interconnecting the neighbouring corners.

Conveniently, the electrodes are captivated at rebates in the abutting faces of the frames extending around the entire continuity of the margins around the central void

Whilst it is envisaged that frames could be held together with sufficient compression to seal the cavities, the ducts and the passage ways, particularly where the frames are of elastomeric material. However it is preferred to provide seals around the ducts and the passages radiating from them and around the electrodes. The seals can be of gasket material, but are preferably O-rings set in grooves in frames.

In the preferred embodiment, passage extensions are provided in the opposite faces of the frames from the abutting faces, the extensions extending from the through-frame openings to the respective electrolyte distribution rebates. Preferably, the electrolyte distribution rebates are wider than the electrode captivation rebates.

To help understanding of the invention, a specific embodiment thereof will now be described by way of example and with reference to the accompanying drawings, in which:

Figure 1 is a perspective view of part of a cell stack of the invention, a full stack in practice having more cells than shown;

Figure 2 is an exploded view of two frames, an electrode and a membrane of the stack of Figure 1;

Figure 3 is a cross-sectional side view of the stack of Figure 1 on the plane III-III shown in Figure 1;

Figure 4 is another cross-sectional side view of fewer frames on the plane IV-IV shown in Figure 1;

Figure 5 is a scrap cross-sectional side view on the plane V-V shown in Figure 1;

Figure 6 is a view similar to Figure 2 of alternative frames, having an alternative passage layout; and

Figure 7 is another view similar to Figure 2 showing another alternative passage layout.

Referring to the drawings, a redox fuel cell stack 1 comprises a plurality of half cell frames 2,3 which are essentially the same, although differing slightly. They are of moulded polymer. Interleaved between them are semi-permeable membranes 4 and graphite plate electrodes 5 (which are of polymer heavily filled with graphite powder or flakes). In use the electrodes act as bipolar electrodes for respectively different reagents and reactions on either side. The membranes equally separate the reagents and allow passage of selected ions and electrons as the reaction progresses. The present invention is concerned with the physical arrangement of the features of the cell stack, although it should be noted that a complete electro-chemical cell is present between each pair of electrodes and includes a membrane and half cell spaces provided by the voids about to be described.

The frames 2,3 are both rectangular, with margins 11 around central voids 12. At the voids, they have rebates 14 in abutting faces 15 for locating the plate electrodes. Closest to their corners, they have small holes 46 for location rods 16 and set in from these, apertures 17 are provided for forming ducts throughout the stack for flow of electrolyte to and from the cell cavities provided by the voids 12. With reference to Figures 2, 4 & 5, the frames 2 have electrolyte flow passages 18 open in their faces 15 abutting the frames 3 and leading from the duct apertures 17 towards each other in end parts 11' of the margins of the frames. The passages stop short of each other and are surrounded by grooves containing sealing O-rings 20. These latter seal with the opposite face of the abutting frame. Also in the faces 15 are grooves for O-rings 21 sealing the frames around the electrode plates 5. The O-rings 20,21 seal the frames against leakage of electrolyte out from between them. Diagonally opposite ones of the passages 18 end at openings 22 passing through the frames 2. The other passages have no openings in the frames 2, but the frames 3 have openings 19 through them in register with the ends of the passages.

On the other side of the frames 2 and 3, the openings 22,19 open into short passages 23 directed towards the central voids and debouching into electrolyte distribution rebates 24, which extend the full width of the central voids at the margin end parts 11'. These rebates have dimples 25 for locating a membrane 4 between them, insofar as a rebate in one frame 2 is adjacent another in frame 3 and so on. Thus electrolyte can flow from one duct aperture 17 in one corner, via the passage 18 from the aperture, either through the frame 2 via the opening 22 or the frame 3 via the opening 19, through short passage 23 and the respective distribution rebate 24 and into the central void to whichever side of the plate electrode it was directed by the opening 19,22. From the opposite end of the central void, the electrolyte is lead back into the diagonally opposite duct aperture 17.

The half cells which the central voids define are closed by the membranes 4, captivated between the frames 2,3 at their faces opposite from those abutting at the electrode plates. These faces 31 have O-rings 32 in grooves around the apertures 17 and O-rings 33 in grooves around the entirety of the central void and the apertures 17. These seal with the membrane. In order to avoid O-rings pressing against O-rings via the membrane, ones O-rings in the frames 2 are set at a smaller diametral dimension D than those D + d in the frames 3, whereby the O-rings are offset from each other, as can be seen in Figure 4. Similarly, the O-rings 33 on opposite sides of the membranes are staggered. It should be noted that the membranes are apertured at 34 & 35 for flow of electrolyte in the ducts 17 through them and the location rods 16.

As shown in Figure 3, the half cells can include three dimensional electrodes in the form of graphite felt pads 41. These fill the central voids, from the electrode plates to the membranes. However, the felt is open in the sense of having appreciable spaces around the individual fibres. Thus the felt provides little resistance to flow of the electrolyte through the cell.

At the end of the stack, a copper collector plate 51 is provided across the end one of the plate electrodes for collection of current from it. The collector plate is set in an insulating carrier 52 and the whole stack is held in compression by an end plate 53. This is clamped in position by non-shown studs acting between it and another compression plate at the other end of the stack.

Figures 6 & 7 show alternative passage layouts. In Figure 6, the frame 102 has a passages 118 from the duct apertures 117 at one long side only of the frame. At the end of each passage there is a through opening 122 to further passages and distribution rebates on the other side. These 123,124 are shown in frame 103, which has the same layout of passages, as can be envisaged as rotation of the frame 103 about the longitudinal axis L. It should be noted that short passages 123 are both on the same side of the longitudinal axis L. In Figure 7, the passages 218 are arranged symmetrically about the central transverse axis A, as well as the layout being symmetrical about the axis L. Otherwise, the arrangement is essentially similar. Whilst it is desirable for production purposes that the two frames 102 & 103, 202 &

203 respectively should be identical, the provision of the O-rings at the membrane with differing diametral dimensions causes the frames to be different and mitigates against them being otherwise identical. The arrangement shown in Figure 7 has moulded recesses 246 in both faces of the frames 203 in place of the holes 46 and lugs 216 moulded on both faces of the frames 202. This arrangement ensures that the frames are assembled with the O-rings within each other as intended, even although the passage layout is identical.

With either of these alternatives, or with the embodiment of Figures 1 to 5, the frames 2,3; 102,103; 202,203 can be assembled together in pairs with their electrode plates and O-rings, as sub-assemblies. The sub-assemblies are then stacked together with a membrane sandwiched between each sub-assembly. This is a more convenient manufacturing process than assembling the stack from a successive selection of four components.

It should be noted that the above described stacks have the following additional advantages:

- The flow passages are defined in one rigid frame face, closed by another. Thus the passages are dimensionally stable, electrically isolated from the plate electrodes and not bounded by the membrane. This arrangement gives more predictable properties to the finished stack for instance in terms of the loss due to ohmic connection of one electrode to the next by the electrolyte columns in the passages connecting each electrode to its neighbour via the electrolyte flow passages;
- The O-rings provide a high degree of sealing integrity;
- The cell thickness, in terms of the separation from the membranes to the electrode plates is independent of the thickness of the frames. For instance very thin cells can be constructed, which would provide difficulties in terms of flow passage depth, with the flow passages being accommodated in that part of the frames accommodating the thickness of the electrode plates.
- The frames can be moulded in simple insert-less moulds and the only additional parts required are the electrodes, membranes and seals. (In our prior cell stack, numerous location washers were required.)

The cell stack is equally suitable for cells used for generating electricity by electrochemical reaction as for cells in which electrochemical reaction is brought about by application of electricity. For this reason, no details of the chemicals nor the reactions are given. However, the chemicals are likely to be corrosive, and as such the materials of the cell need to be as resistant to chemical reaction as reasonably possible. For instance, the electrode plate is preferably of graphite filled polypropylene, with the same polymer being used for the frames. The O-rings can be of fluoroelastomer, typically Viton™ material from DuPont. The membranes can be of conventional electrochemical membrane material.

The invention is not intended to be restricted to the details of the above described embodiment. For instance, where the chemistry of the reaction in the cell is suited to very thin cells without three dimensional electrodes, the felt electrode can be omitted. Further, it is envisaged that at least at the abutting faces 15, the frames may be bonded together to captivate and seal the electrodes and seal the passages and duct apertures. Figure 8 shows adhesive 350 for this. In the variant shown in this Figure, the O-rings 21 sealing the frames to each other peripherally of the electrode plates 5 have been replaced by O-rings 321 sealing the frames to the electrode plates 305, in inwards of the adhesive 350. This variant shows an end, copper current collector plate 351 located in a special end frame 352 have a rectangular cut-out 3521 for the collector plate 351 and a groove 3522 for a contact tongue 3511 of the collector plate. A back-up plate 3523 insulates the collector plate from a clamp plate 353.

CLAIMS:

1. An electrochemical stack cell comprising a plurality of cells arranged side-by-side in a stack, each cell having:

- a membrane,
- a first half cell cavity on one side of the membrane and a second half cell cavity on the other side of the membrane,
- a respective electrode plate at the side of each half cell opposite from the membrane, each electrode plate providing contact between adjacent cells at least for intermediate ones of the cells,
- a pair of frames, one for one half cell and the other for the other, the frames:
  - captivating the membrane between themselves,
  - locating the electrode plates and
  - having:
    - continuous margins around central voids providing the half cell cavities,
    - apertures in the continuous margins providing ducts for flow of electrolyte through the stack for distribution to the cells,
    - electrolyte distribution rebates at opposite inside edges of the margins and
    - passages in the continuous margins for electrolyte flow from one of the duct apertures, into and out of the half cell at the distribution rebates and to another of the duct apertures,

wherein:

- each plate electrode is captivated between a frame from one cell and a frame from an adjacent cell with at least two portions of the margins of these frames extending outside respective edges of the plate electrode, the adjacent cell frames having faces which abut at the portions;
- the flow passages are formed in the faces of the margins and are closed by abutting opposite frame faces; and
- through-frame openings are provided in the frames for extending the passages from the abutting faces of the frames to the other, membrane side of the frames into distribution rebates.

2. An electrochemical stack cell as claimed in claim 1, having rectilinear opposite margins with the electrolyte duct apertures arranged at the corners.

3. An electrochemical stack cell as claimed in claim 2, wherein the flow passages are provided in two opposite margins.

4. An electrochemical stack cell as claimed in claim 3, wherein all the passages in the face of one of each pair of abutting face frames, i.e. with two passages in each marginal portion having passages with one through-frame opening in the portion at the end of one of the passages and another said opening in the other frame opposite the end of the other passage.

5. An electrochemical stack cell as claimed in claim 3, wherein one passage only is provided in each marginal portion having a said flow passage.

6. An electrochemical stack cell as claimed in claim 5, wherein each said passage has an opening through its frame.

7. An electrochemical stack cell as claimed in claim 5, wherein all through-frame openings are provided in the marginal portion opposite from the end of the respective passages.

8. An electrochemical stack cell as claimed in claim 5, claim 6 or claim 7, wherein the flow passages are provided such that the frames have symmetry about a central axis transverse the plane of their abutting faces

9. An electrochemical stack cell as claimed in claim 5, claim 6 or claim 7, wherein the passages are arranged to extend from two duct apertures at neighbouring corners of the frame, with the passages extending in the marginal portions extending away from the margin interconnecting the neighbouring corners.

10. An electrochemical stack cell as claimed in any preceding claim, wherein the electrodes are captivated at rebates in the abutting faces of the frames extending around the entire continuity of the margins around the central void.

11. An electrochemical stack cell as claimed in claim 10, wherein the electrolyte distribution rebates are wider than the electrode captivation rebates.

12. An electrochemical stack cell as claimed in claim 11, including flow spreading elements in the electrolyte distribution rebates.

13. An electrochemical stack cell as claimed in any preceding claim, wherein the frames are held together with sufficient compression to seal the cavities, the ducts and the passage ways, the frames being of elastomeric material.

14. An electrochemical stack cell as claimed in any one of claims 1 to 12, wherein seals are provided around the ducts and the passages radiating from them, around the electrodes and around the half cell cavities between the frame and the membranes.

15. An electrochemical stack cell as claimed in claim 14, wherein the seals are O-rings set in grooves in frames, the O-rings on one side of the membrane being of set at a smaller diametral dimension than those on the other side, whereby the O-rings are offset from each other.

16. An electrochemical stack cell as claimed in claim 14 or claim 15, wherein O-ring seals are provided on opposite sides of the electrode plates, sealing them to the frames captivating them.

17. An electrochemical stack cell as claimed in any preceding claim, wherein at least some adjacent half cell frames are bonded together.

18. An electrochemical stack cell as claimed in any preceding claim, including passage extension in the opposite faces of the frames from the abutting faces, the extensions extending from the through-frame openings to the respective electrolyte distribution rebates.

19. An electrochemical stack cell as claimed in any preceding claim, including three dimensional electrodes extending from the plate electrodes into the respective half cells.

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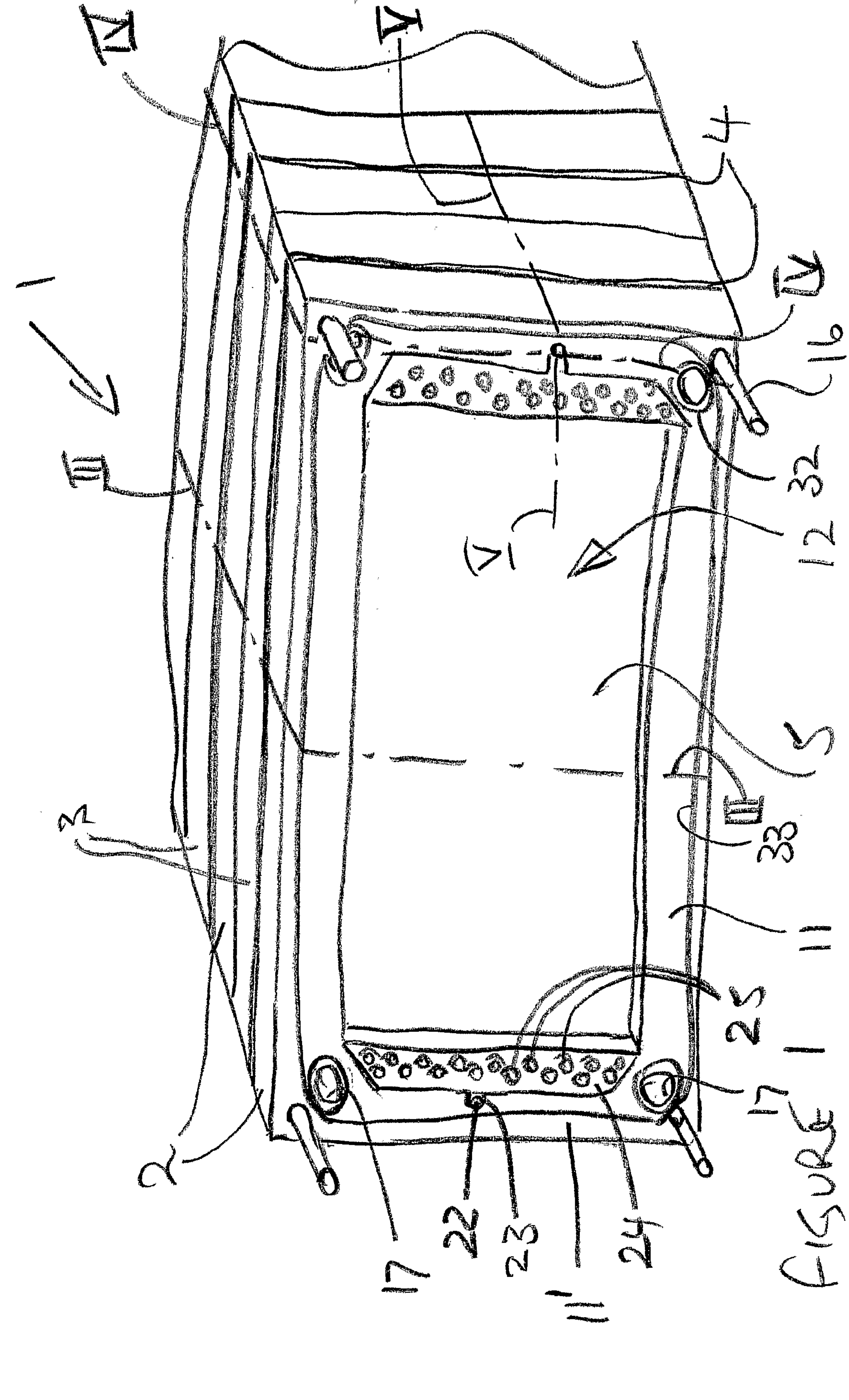

FIGURE 1

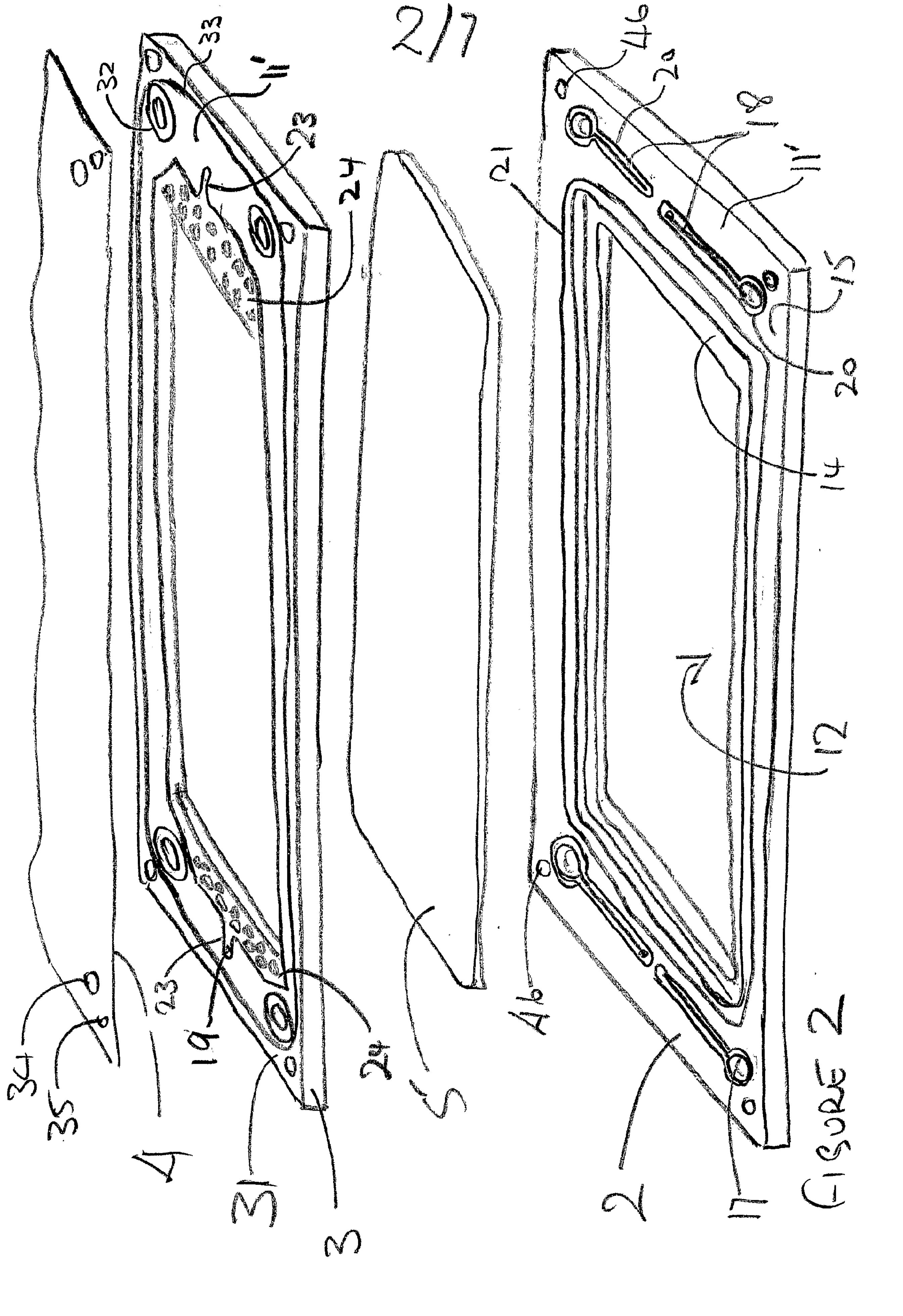

FIGURE 2

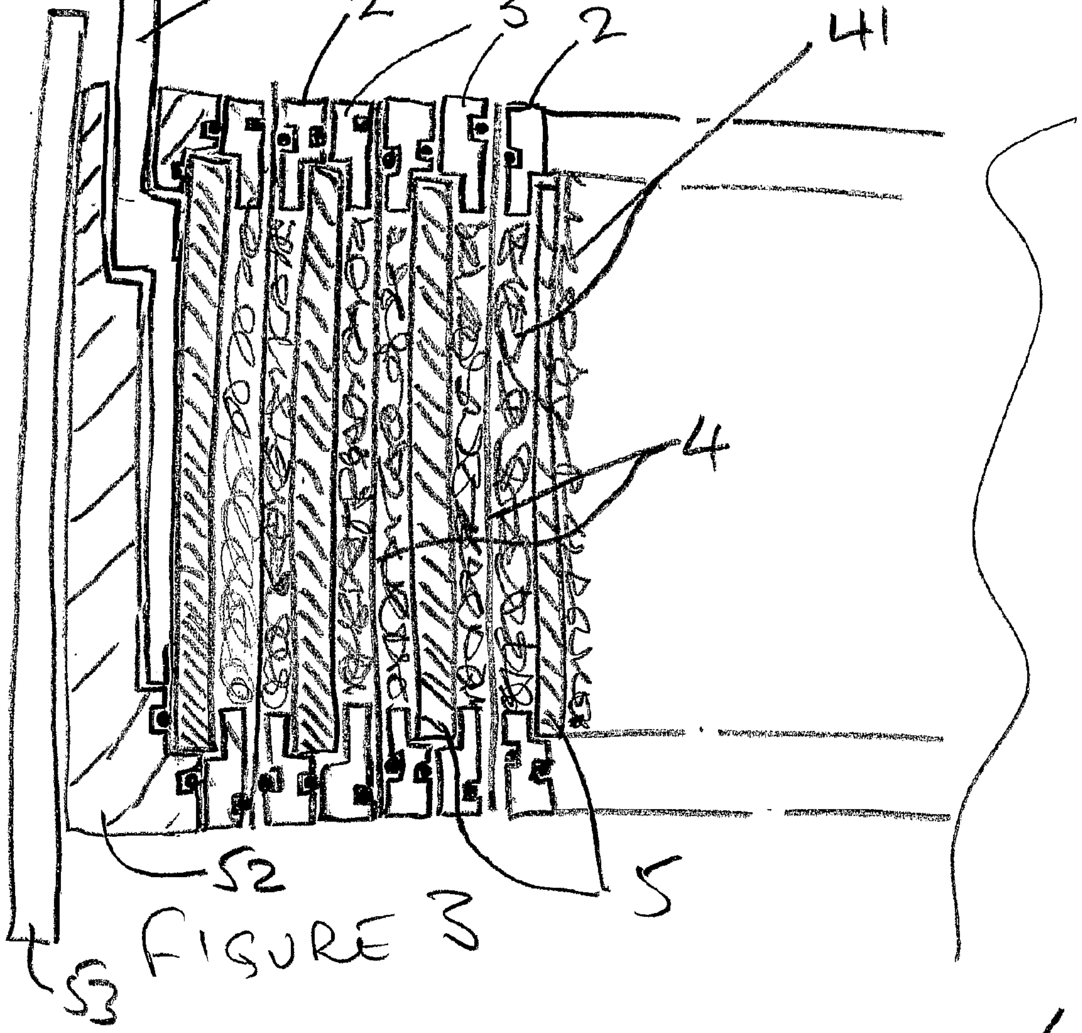

FIGURE 3

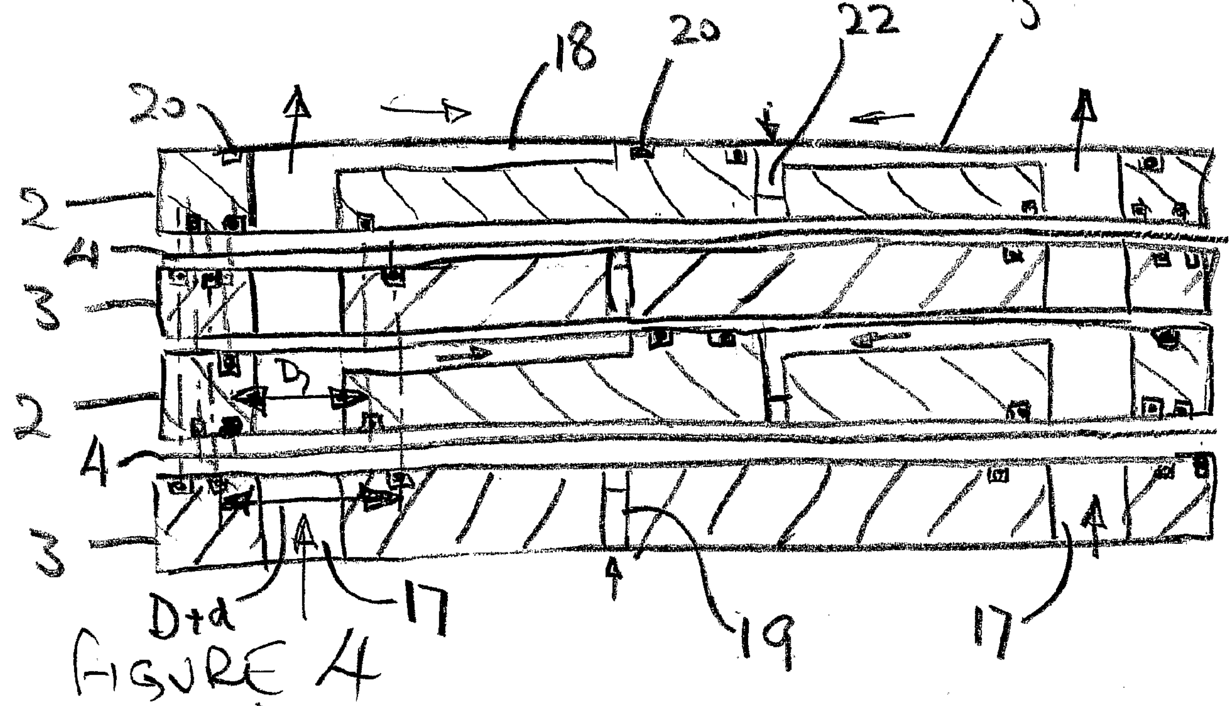

FIGURE 4

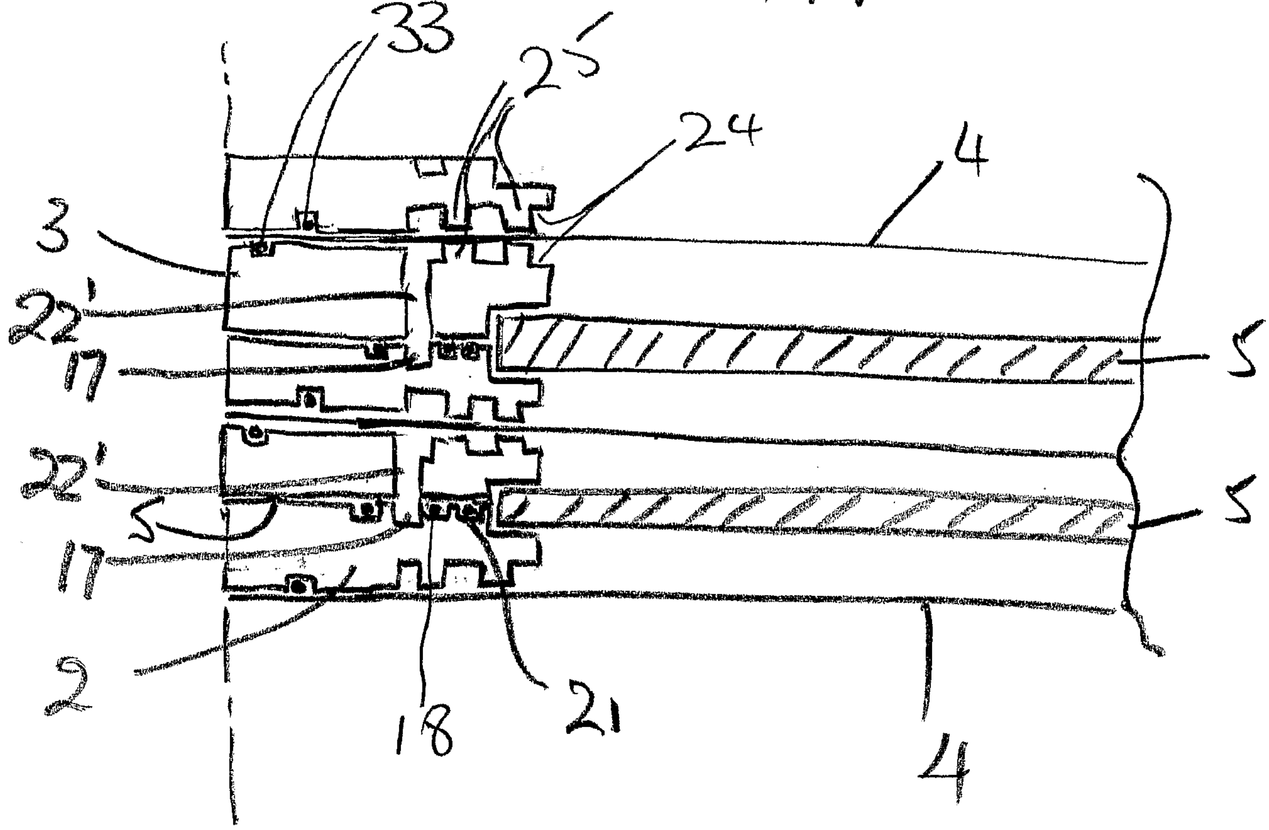

FIGURE 5

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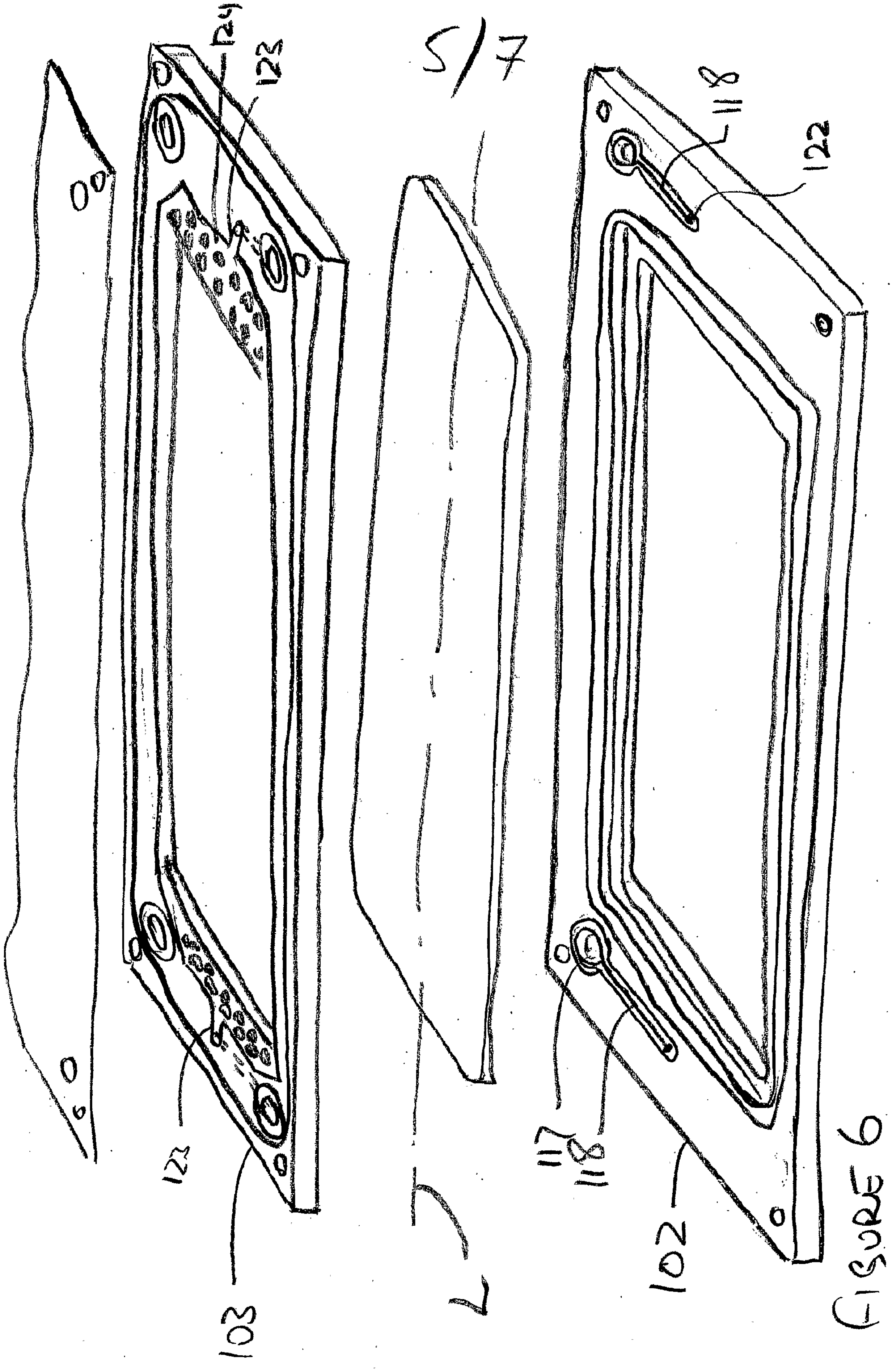

FIGURE 6

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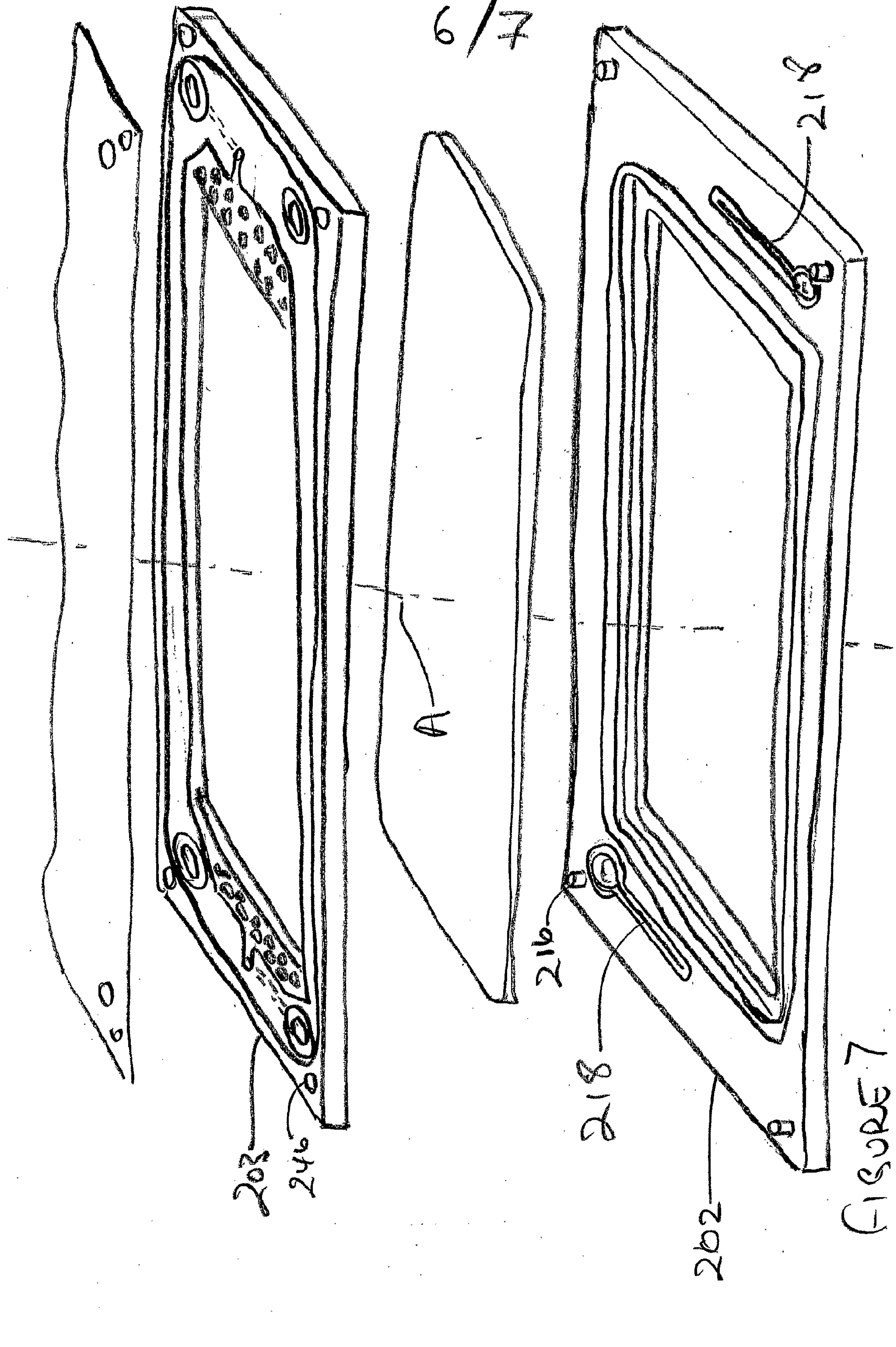

FIGURE 7

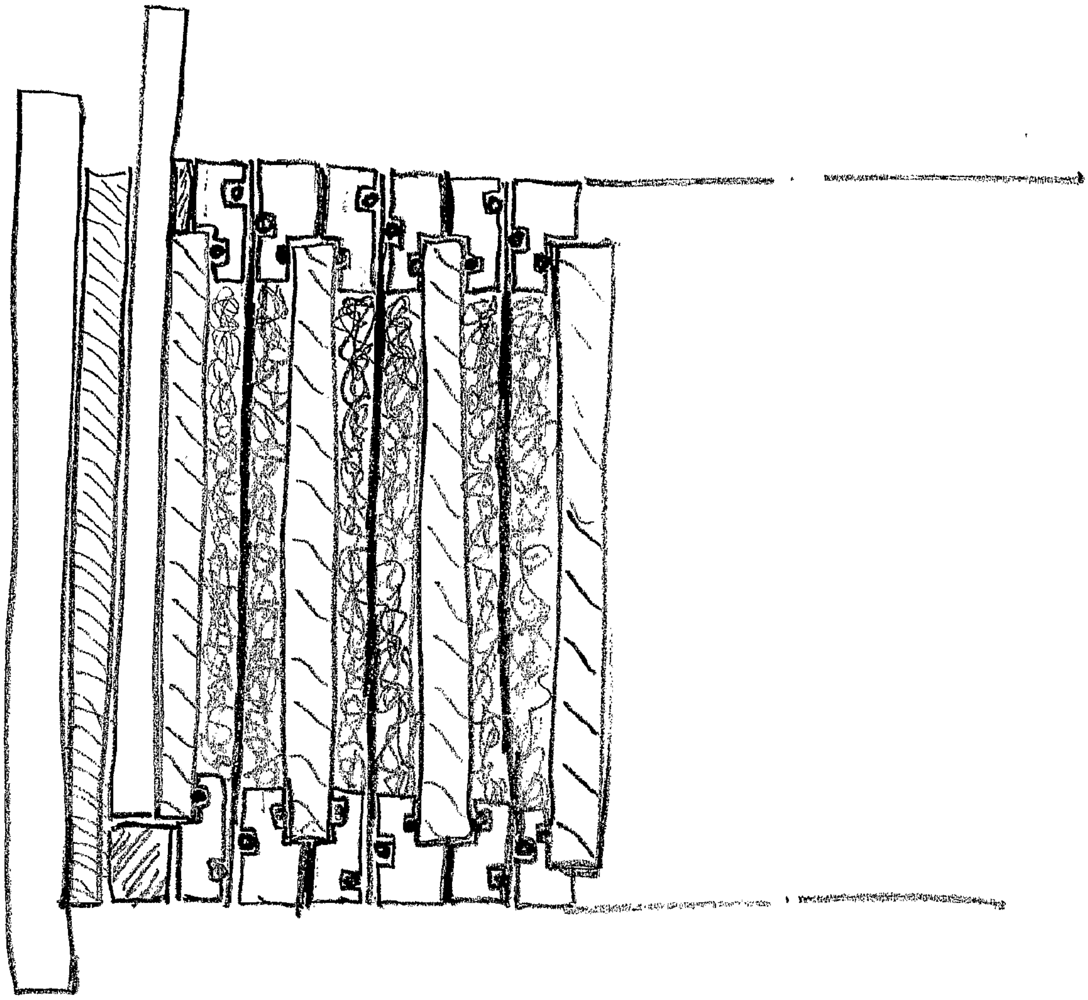

Figure 8

PATENT COOPERATION TREATY

PCT

INTERNATIONAL SEARCH REPORT

(PCT Article 18 and Rules 43 and 44)

Applicant's or agent's file reference	**FOR FURTHER ACTION** see Form PCT/ISA/220 as well as, where applicable, item 5 below.	
2675/PCT		
International application No. PCT/GB2006/001256	International filing date *(day/month/year)* 05/04/2006	(Earliest) Priority Date *(day/month/year)* 16/04/2005
Applicant RE-FUEL TECHNOLOGY LIMITED		

This international search report has been prepared by this International Searching Authority and is transmitted to the applicant according to Article 18. A copy is being transmitted to the International Bureau.

This international search report consists of a total of 5 sheets.

[X] It is also accompanied by a copy of each prior art document cited in this report.

1. **Basis of the report**
   a. With regard to the **language**, the international search was carried out on the basis of:
      [X] the international application in the language in which it was filed
      [ ] a translation of the international application into ____________, which is the language of a translation furnished for the purposes of international search (Rules 12.3(a) and 23.1(b))
   b. [ ] With regard to any **nucleotide and/or amino acid sequence** disclosed in the international application, see Box No. I.
2. [ ] **Certain claims were found unsearchable** (See Box No. II)
3. [ ] **Unity of invention is lacking** (see Box No III)
4. With regard to the **title**,
   [ ] the text is approved as submitted by the applicant
   [X] the text has been established by this Authority to read as follows:

   ELECTROCHEMICAL CELL STACK WITH FRAME ELEMENTS

5. With regard to the **abstract**,
   [ ] the text is approved as submitted by the applicant
   [X] the text has been established, according to Rule 38.2(b), by this Authority as it appears in Box No. IV. The applicant may, within one month from the date of mailing of this international search report, submit comments to this Authority
6. With regard to the **drawings**,
   a. the figure of the **drawings** to be published with the abstract is Figure No. 2
      [ ] as suggested by the applicant
      [X] as selected by this Authority, because the applicant failed to suggest a figure
      [ ] as selected by this Authority, because this figure better characterizes the invention
   b. [ ] none of the figures is to be published with the abstract

INTERNATIONAL SEARCH REPORT	International application No. PCT/GB2006/001256

Box No. IV Text of the abstract (Continuation of item 5 of the first sheet)

A redox fuel cell stack (1) comprises a plurality of essentially similar half cell frames 2,3 of moulded polymer. Interleaved between them are semi-permeable membranes (4) and bipolar plate electrodes. The frames (2, 3) are rectangular, with margins (11) around central voids (12). At the voids, they have rebates (14) in abutting faces (15) for locating the plate electrodes. At their corners, they have apertures (17) for forming ducts throughout the stack for flow of electrolyte to and from the cell cavities provided by the voids (12). The frames (2) have electrolyte flow passages (18) open in their faces 15 abutting the frames 3 and leading towards each other. The passages stop short of each other and are surrounded by grooves containing sealing O-rings (20). Diagonally opposite ones of the passages (18) end at openings (22) passing through the frames (2). The other passages have no openings in the frames (2), but the frames (3) have openings through them in register with the ends of the passages. On the other side of the frames (2) and (3), the openings (21, 22) open into short passages (23) directed towards the central voids and debouching into electrolyte distribution rebates (24). Thus electrolyte can flow from one duct aperture (17) in one corner, via the passage (18) from the aperture, either through the frame (2) via the opening (21) or the frame (3) via the opening (22), through short passage (23) and the respective distribution rebate and into the central void to whichever side of the plate electrode it was directed by the opening (21, 22). From the opposite end of the central void, the electrolyte is lead back into the diagonally opposite duct apertur

INTERNATIONAL SEARCH REPORT

International application No
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A. CLASSIFICATION OF SUBJECT MATTER
INV. H01M8/02 H01M8/18 C25B9/20
ADD. C25B9/06 C25B9/02

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

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)
H01M 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 practical, search terms used)

EPO-Internal, PAJ, WPI Data

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	US 6 555 267 B1 (BROMAN BARRY MICHAEL ET AL) 29 April 2003 (2003-04-29) column 1, line 8 - line 14; figures 3,5 column 6, line 6 - line 58 column 7, line 33 - line 58 column 8, line 10 - line 19	1-19
A	US 4 339 324 A (HAAS ET AL) 13 July 1982 (1982-07-13) column 1, line 5 - line 9 column 6, line 9 - line 67; figures 5,6,8-10 column 7, line 64 - column 8, line 7	1-19

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

[X] See patent family annex.

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Date of the actual completion of the international search	Date of mailing of the international search report
29 June 2006	19/07/2006

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, Tx. 31 651 epo nl, Fax: (+31-70) 340-3016	Raimondi, F

INTERNATIONAL SEARCH REPORT

International application No
PCT/GB2006/001256

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Information on patent family members

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