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 Applicant: **THE DOW CHEMICAL COMPANY**  
**2030 Dow Center Abbott Road P.O. Box 1967**  
**Midland, MI 48640(US)**


 Designated Contracting States:  
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 Inventor: **Beaver, Richard N.**  
**10 Forest Lane**  
**Angleton Texas 77515(US)**  
 Inventor: **Dang, Hiep D.**  
**511 Southern Oaks**  
**Lake Jackson Texas 77566(US)**  
 Inventor: **Morris, Gregory J.E.**  
**123 Sequoia**  
**Lake Jackson Texas 77566(US)**  
 Inventor: **Pimlott, John R.**  
**1410 Weekes**  
**Sweeny Texas 77480(US)**


 Representative: **Huber, Bernhard, Dipl.-Chem.**  
**et al**  
**Patentanwälte H. Weickmann, Dr. K. Fincke**  
**F.A. Weickmann, B. Huber Dr. H. Liska, Dr. J.**  
**Prechtel Möhlstrasse 22 Postfach 860 820**  
**D-8000 München 86(DE)**


**Structural frame for an electrochemical cell.**


 The present invention is a structural frame particularly suitable for use in an electrochemical cell. The frame comprises a planar member (12) made of a polymeric material with a plurality of horizontally and vertically spaced-apart shoulders (14) protruding outwardly from opposing generally coplanar surfaces of the planar member. Each of the shoulders annularly encircles and supports an electrically conductive insert (26) extending from an exterior face of a shoulder (14) on one surface of the planar member, through the planar member (12), to an exterior face of a shoulder (14) on the opposite surface of the planar member. An electrically conductive, substantially completely hydraulically impermeable liner (22), (24) resistant to the corrosive effects of an electrolyte in said cell matingly contacted with at least one of said surfaces or opposing surfaces of said planar member and adapted to minimize con-

tact between the electrolyte in said cell in said planar member.

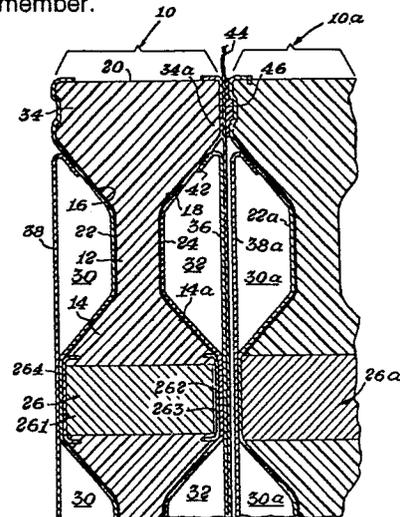


Fig. 1

EP 0 282 614 A1

## STRUCTURAL FRAME FOR AN ELECTROCHEMICAL CELL

This invention relates to an electrochemical cell and in particular to a structural frame for use in an electrochemical cell.

It is well established that various chemicals can be produced in an electrochemical cell containing an anode and a cathode. For example, alkali metal chlorates, such as sodium chlorate, have been formed electrolytically from a sodium chloride brine in cells without a separator positioned between the anode and the cathode.

When a separator, such as a liquid permeable asbestos or polytetrafluoroethylene diaphragm or a substantially liquid impervious ion exchange membrane, is used in a cell to electrolyze a sodium chloride brine, the electrolytic products will normally be gaseous chlorine, hydrogen gas, and an aqueous solution containing sodium hydroxide.

For a number of years gaseous chlorine was produced in electrolytic cells wherein an asbestos diaphragm was interposed between finger-like, anodes and cathodes which were interleaved together. During the past several years it has become apparent that the use of a substantially liquid impermeable cation exchange membrane may be preferable to the more well established diaphragm in instances where a higher purity, for example a lower sodium chloride content, higher sodium hydroxide product is desired. It was found to be more convenient to fabricate ion exchange type electrochemical cells from relatively flat or planar sheets of ion exchange membrane rather than to interleave the membrane between the anode and cathode within the older finger-like cells used with asbestos diaphragms.

The newer, so-called flat plate electrochemical cells using a planar piece of ion exchange membrane to separate the anolyte from the catholyte compartments also have a plurality of solid, liquid impervious frames adapted to support the anode on one side and the cathode on the opposite side. These frames have previously been constructed of materials such as metal and plastic, but neither of these materials has been found to be entirely satisfactory. In any electrochemical cell, including both monopolar and bipolar cells, there is a possibility that electrolyte may leak from within the cell to the exterior. In instances where such leakage has occurred in cells with iron or other ferrous type frames, it was found that the iron frame corroded or was itself electrolytically attacked. Plastic frames are not generally subject to the electrolytic attack, but are normally not resistant to the anolyte and/or catholyte within the cell under operating conditions for extended periods of time, for example, several years.

It is desired to provide a structural frame for use in electrochemical cells which would minimize the corrosion problems and would increase the relatively short useful life attendant with those frames used by the prior art.

The present invention particularly resides in a structural frame adapted for use in an electrochemical cell comprising:  
a central cell element in the form of a generally planar member made of a polymeric material having a plurality of horizontally and vertically spaced-apart shoulders protruding outwardly from at least one surface of said planar member;  
at least one electrically conductive insert extending from an exterior face of a shoulder on one surface of the planar member, through the planar member, to an exterior face of a shoulder on the opposite surface of the planar member, wherein each of said shoulders annularly encircles and supports each of said inserts,  
an electrically conductive, substantially completely hydraulically impermeable liner cover resistant to the corrosive effects of an electrolyte in said cell matingly contacted with at least one of said surfaces of said planar member and adapted to minimize contact between the electrolyte in said cell and said planar member.

In one embodiment, an anolyte cover is matingly affixed to the anolyte surface of the planar member and adapted to minimize contact between the anolyte and the planar member. The anolyte cover is resistant to the corrosive effects of the anolyte. A catholyte cover is matingly affixed to the opposite, catholyte surface of the planar member and adapted to minimize contact between the catholyte and the planar member. The catholyte cover is resistant to the corrosive effects of the catholyte. Both the anolyte cover and the catholyte cover may be made from a metal or, from another material which is provided with metallic inserts molded into the non-metallic liner at the points where the metallic inserts in the liner contact the metallic inserts which pass through the planar member.

The invention further includes an electrochemical cell utilizing a plurality of the above described structural frames removably and sealably positioned in a generally coplanar relationship with each other and with each of the planar members being spaced apart by an anode on one or opposite sides of the planar member and a cathode on an opposing side of the planar member or by an anode on one side and a cathode on the opposite side of the planar member.

The accompanying drawing further illustrates the invention;

Figure 1 is a cross-sectional view of one embodiment of the invention,

Figure 2 is an exploded, isometric view of another embodiment of the structural frame in combination with an anode, cathode, and ion exchange membrane,

Figure 3 is a cross-sectional side view of another embodiment of the electrochemical cell of the present invention.

Identical numbers, distinguished by a letter suffix within the several figures represent parts having a similar function within the different embodiments.

In Figure 1 are shown structural frames 10 and 10a, which achieve the above objects. It is illustrated for use in an electrochemical cell for producing gaseous chlorine in aqueous alkali metal hydroxide solution. Although the present invention can be beneficially employed to produce chlorine and various alkali metal hydroxide solutions, it is preferred to use sodium chloride as the primary salt in the starting brine since this particular salt is readily available commercially and there are many well established uses for sodium hydroxide produced electrolytically.

The cell structure 10 includes a generally planar member 12 comprising a planar barrier portion, a peripheral flange portion (34), and anode and cathode standoff means or bosses for maintaining the anode and cathode of adjacent cell structures at a predetermined distance from the planar barrier portion. The planar member can be produced by commercial and known procedures into a shape with a plurality of horizontally and vertically spaced apart shoulders 14 and 14a (bosses) protruding outwardly from cathode and anode sides 16 and 18, respectively. The peripheral surface 20 of the planar member 12 defines the outer surface of the electrochemical cell when a plurality of the planar members are positioned together as shown in the drawing. The peripheral configuration of the planar members 12 is optional and can be varied to suit the particular configuration of the electrochemical cell shape desired.

The number, size, and shape of the shoulders 14 and 14a may be an important consideration in both the design and operation of the present invention. They may be square, rectangular, conical, cylindrical, or any other convenient shape when viewed in sections taken either parallel or perpendicular to the central portion. The shoulders may have an elongated shape to form a series of spaced ribs distributed over the surface of the plastic member.

A number of polymeric materials are suitable for use in the present invention for the construction of the planar member. Without intending to be limited by the specific materials hereinafter delineated, examples of such suitable materials include

polyethylene; polypropylene; polyvinylchloride; chlorinated polyvinyl chloride; acrylonitrile, polystyrene, polysulfone, styrene acrylonitrile, butadiene and styrene copolymers; epoxy; vinyl esters; polyesters; and fluoroplastics and copolymers thereof. It is preferred that a polymeric material such as polypropylene be used for the planar member since it produces a shape with adequate structural integrity at elevated temperatures, is readily available, and is relatively inexpensive with respect to other suitable materials.

It is surprising that the planar member 12 can be produced by any of a number of processes known well to those skilled in the art of plastic molding. Such molding processes include, for example, injection molding, compression molding, transfer molding, and casting. Of these processes, injection molding has been found to satisfactorily produce a structure with adequate strength for use in an electrochemical cell. Preferably, the polymeric material is injected into a mold containing the desired number of inserts (discussed later). In this manner, the planar member is a one-piece member which fits tightly around the inserts holds them in place, and provides a high degree of support to them. Such a configuration minimizes the likelihood that the inserts will separate from the planar member and become loose. The ease of molding relatively complex shapes and the strength of the finished injection molded article contribute to making this process preferred for making the herein described planar member. This is a considerable advantage over the prior art where the polymeric member was molded first and then the electrical conductors were subsequently installed.

When the planar member 12 is employed in an electrochemical cell for producing chlorine, the temperature of the cell and the planar member will frequently reach, or be maintained at, temperatures of from 60° to 90°C. At these temperatures polymeric materials, as do most materials, expand a measurable amount. Any expansion and later contraction on cooling of the planar member could result in electrolyte seeping from within the plurality of cells when joined together or, more importantly, could result in distortion of the anode and cathode which are made of metallic expanded mesh or perforated sheets. Furthermore, the differential expansion between the planar member 12 an electrolyte linear or cover 22 and/or 24 would create stress on the welds which affix these covers to the inserts which are themselves molded in the planar member.

To reduce, and preferably minimize, the difference in expansion between the covers 22 and 24 and the planar member 12, it is preferred to incorporate an additive to reduce thermally induced expansion of the planar member. More preferably,

the additive will also increase the structural strength of the finished article. Such additive can be, for example, fiberglass, graphite fibers, carbon fibers, talc, glass beads, pulverized mica, asbestos, and the like, and combinations thereof. It is preferred that the polymer contain from 5 to 75 weight percent and more preferably from 10 to 40 weight percent of the additive. Glass fibers can be readily mixed with polypropylene to produce an injectable material suitable for use in the present invention which results in a solid, physically strong body with a coefficient of expansion less than polypropylene not containing glass fibers. Of greater importance is the need to minimize the difference in expansion between the planar member, the electrodes, and the current collector, since these elements are welded together and it is critical that they remain substantially flat and parallel.

It has been determined that the use of commercially available polypropylene which has been specially formulated to afford bonding with the glass fibers works particularly well. This results in a composite having a lower coefficient of expansion than a mixture of polypropylene and glass fibers. Such chemically-combined glass fiber reinforced polypropylene is available from, for example, Hercules, Inc., Wilmington, Delaware, as Pro-fax PC072 polypropylene.

At least one electric conducting element, such as insert(s) 26, is positioned and preferably molded into the planar member 12. The insert(s) 26 extends through the planar member from one electrolyte surface, e.g. the catholyte surface 16 to an opposite electrolyte surface, e.g. the anolyte surface 18. The inserts 26 and 26a are preferably retained within the planar member 12 by means of friction between the polymeric material and the insert. It is more preferable to increase the friction between these two bodies by having an additional means to restrain the insert within the polymeric material. Such additional means include, for example grooves (one or more) around the circumference of the insert(s), keys welded to the insert, hole(s) extending into and/or through the insert, slots, rings, collars, studs, or bosses.

The insert(s) 26 can be any material which will permit flow of an electric current between the catholyte cover 22 and the anolyte cover 24. Since the covers 22 and 24 are preferably made of a metal, it is convenient to fabricate the insert from a metal, such as aluminum, copper, iron, steel, nickel, titanium, and the like, or alloys or physical combinations including such metals.

The shoulders and inserts should be spaced so that they provide a somewhat uniform and low electrical potential gradient across the face of the electrode to which they are attached. They should be spaced so that they allow free fluid circulation

from any unoccupied point within their respective electrolyte compartment to any other unoccupied point within that compartment. Thus the shoulders will be somewhat uniformly spaced apart from one another in their respective compartments.

To improve the flow of DC electric current between the covers 22 and 24, the insert 26 is preferably made of a material weldably compatible with the particular cover it contacts. For example, the insert 26 may be a welded assembly of a steel rod 261 with a vanadium disk 262 interposed between and welded to both the rod 261 and a titanium cup-like member 263 on the anode facing portion of the structure 10. A similar nickel cup-like member 264 may be welded directly to the rod 261 on the cathode facing portion of the insert. The titanium and nickel members 263 and 264, respectively, are then readily weldable to the titanium anolyte cover 24 and the nickel catholyte cover 22 and 22a preferred for use in an electrochemical cell producing chlorine and an aqueous sodium hydroxide solution.

To prevent catholyte from contacting the planar member within the electrochemical cell and cause deterioration of the polymeric material and/or leakage of electrolyte between the polymeric material and the insert 26 from cathode compartment 30 to anode compartment 32 the cover 22 is in mating contact with the catholyte surface 16 and the anolyte cover 24 is in mating contact with the anolyte surface 18. As is shown in Figure 1, both the anolyte and the catholyte covers are so shaped to correspond closely to the exterior surface of the planar member 12. In some instances, the covers 22 or 24 may abut the frame 10 in one or more locations. It is important that the portions of both the covers 22 and 24 which are exposed to the anolyte or catholyte and span the planar member contain no openings through which electrolyte or electrolytic products can pass during operation of the electrochemical cell. The freedom from openings through the covers minimizes the likelihood that electrolyte will leak or seep through holes or spaces around gaskets of other seals and come into contact with the planar member.

The anolyte cover 24 is made of a material which is resistant to the anolyte during operation of the cell. Normally, this material is not electrolytically active, but the invention is still operable if the material does become or is active electrolytically. Suitable materials are, for example, titanium, tantalum, zirconium, tungsten, and other valve metals not materially affected by the anolyte. Titanium is preferred as the anolyte cover.

The catholyte cover 22 is resistant to attack by the catholyte under the conditions present in the electrochemical cell. Suitable materials for the catholyte cover include, for example, iron, steel,

stainless steel, nickel, lead, molybdenum, and cobalt and alloys, including major portions of these metals. Nickel, including nickel base alloys, is preferably used for the catholyte cover, since nickel and nickel alloys are generally resistant to the corrosive effects of the catholyte, especially an aqueous catholyte solution containing up to at least about 35 weight percent sodium hydroxide. Steel has also been found to be suitable, and relatively inexpensive, for use in a cell as a catholyte cover in the presence of a dilute (i.e., up to about 22 weight percent) aqueous solution of sodium hydroxide.

To assist in assembling a plurality of the structural frames 10 into an electrochemical cell it is desirable, although not essential, to have flanges 34 and 34a extending outwardly from the main structural portion of the planar member 12 along the periphery of the planar member. In a preferred embodiment the flanges extend outwardly from the planar member about the same distance as the insert 26. Alternatively, but not preferred, separate spacer elements (not shown) could be utilized to build up to the planar member 12 sufficiently to permit a number of the planar members to be combined into a cell series without having electrolyte, either anolyte or catholyte, leak from within the catholyte and anolyte compartments 30 and 32, respectively, to an exterior portion of the cell.

Figure 1 further shows an anode 36 which is positively charged during operation of the cell from an external power source (not shown), electrically connected to the anolyte cover 24. Such electrical connection is readily achieved by welding the anode 36 to the anode cover where the anode cover comes into physical contact with the insert 26. For improved electrical contact, the anolyte cover 24 is welded to the insert 26 and the anode 36 is welded to the anolyte cover 24 adjacent to the insert 26. Various means of welding can be utilized in the present invention, but it has been found highly satisfactory to use resistance or capacitance discharge welding techniques. The anode 36 can additionally be welded to the cover 24 at anode end portion 42 by, for example, resistance or capacitance discharge welding. Other suitable welding techniques include tungsten inert gas (TIG) and metal inert gas (MIG) welding. This welding serves a primary purpose of retaining the anode in position and not for electrical flow, although electric current will naturally pass through the welded areas.

The anode 36 is a metal, such as one of the common film-forming metals, which is resistant to the corrosive effects of the anolyte during the operation of the cell. Suitable metals are well known to include tantalum, tungsten, columbium, zirconium, molybdenum, and preferably, titanium and

alloys containing major amounts of these metals, coated with an activating substance, for example, an oxide of a platinum group metals, such as ruthenium, iridium, rhodium, platinum, palladium, either alone or in combination with an oxide of a film-forming metal. Other suitable activating oxides include cobalt oxide either along or in combination with other metal oxides. Examples of such activating oxides are found in U.S. Patents 3,632,498; 4,142,005; 4,061,549; and 4,214,971.

The cathode 38 and 38a, which has a negative electric potential during operation of the cell, is electrically connected to the catholyte cover 22 and 22a, respectively, in substantially the same manner as above described for the anode 36. The cathode 38 and 38a should be constructed of a material which is resistant to the corrosive effects of the catholyte during operation of the cell. Materials suitable for contact with the catholyte will depend upon the concentration of the alkali metal hydroxide in the aqueous solution and may be readily determined by one skilled in the art. Generally, however, materials such as iron, nickel, lead, molybdenum, cobalt, and alloys including major amounts of these metals, such as low carbon stainless steel, are suitable for use as the cathode. The cathodic electrodes may optionally be coated with an activating substance to improve performance of the cell. For example, a nickel substrate could be coated with oxides of nickel and a platinum group metal, such as, ruthenium, or nickel and a platinum group metal, or oxide thereof such as ruthenium oxide, to reduce hydrogen overvoltage. U. S. Patent 4,465,580 describes the use of such cathodes.

As is apparent from the drawing, both the anode and the cathode are permeable to the respective electrolyte. The electrodes can be made permeable by several means including, for example, using a punched sheet or plate, an expanded mesh, or woven wire. The anode should be sufficiently porous to permit anolyte and chlorine to pass therethrough and the cathode should be sufficiently porous to permit catholyte to pass therethrough and hydrogen to pass therethrough.

The electrochemical cell of Figure 1 also shows the anode 36 and the cathode 38 spaced apart by an ion exchange membrane 44 which is in contact with the anode 36. If desired, however, although not preferred, the membrane 44 could be in contact with the cathode 38 or be suspended between the two electrodes. It is important, that the ion exchange membrane 44 separate the anode compartment 32 from the cathode compartment 30a.

To minimize leakage of electrolyte from the cell after assembling a number of the structural frames 10 together, at least one gasket 46 is positioned between the frames 10 and 10a. During

assembly of the frames a compressive force is applied to the extremes of the frames to compress the gasket material 46 so that it both seals the ion exchange membrane 44 in position and minimizes leakage of electrolyte from within the final cell series to the exterior of the cells. Preferably, the membrane 44 is positioned to substantially entirely prevent leakage of electrolyte from within the final cell series to the exterior of the cells. Various gaskets materials can be used including, for example, fluorocarbon, chlorinated polyethylene rubber, and ethylene propylene diene terpolymer rubber.

Figure 2 is an exploded, partially cross-sectioned isometric view of the structural frame 10b, including a planar member 12a with a plurality of frustoconical shoulders 14b, with inserts 26a encased therein, extending outwardly from the generally planar anolyte surface 18a. Identical shoulders 14c extend outwardly from the catholyte surface of the planar member 12a in a mirror image relationship with the shoulders 14b on the anolyte surface. A conduit or opening 48 is provided in the planar member 12a to permit exit of production produced in the electrochemical cell during operation. Preferably, a pipe, tube, or shaped metal conduit is positioned within the opening 48 and affixed to the cover 24b to facilitate substantially leak free removal of the product from the cell. A similar opening and conduit (not shown) is provided in, for example, a wall portion of the planar member at a location generally diagonally opposed to the opening 48 to permit an aqueous sodium chloride solution to be fed through the conduit into the anode compartment. Similar openings and conduits are provided on the cathode side of the planar member to permit feeding, for example, water into the cathode compartment and removal of products, such as a solution containing sodium hydroxide, and optionally hydrogen, therefrom. The anolyte cover 24b and catholyte cover 22a are adapted to closely fit over the respective surface of the planar member 12a and prevent entrance of electrolyte from the respective electrode compartments into the space, if any, between the cover and the planar member. The covers 22a and 24b also have conduits therein for exit of the brine solution and product produced in the respective electrolyte compartment and feeding of starting solutions to the respective compartments. For example, a shaped pipe 50, in the cover 24b corresponds to the opening 48 in the planar member to afford ready exit of the product chlorine and spent anolyte from the anode compartment. An expanded mesh anode 36b and an expanded mesh cathode 38a are adapted to fit within the respective anolyte and catholyte covers substantially the same as shown in Figure 1. An ion exchange membrane is shown in sheet 44a. A leak minimizing gasketing

material 46a is placed between structural frame members prior to the assembly of an electrochemical cell series.

In Figure 3 is shown a partially assembled cell series containing three sets of structural frame members with anodes and cathodes spaced apart by ion exchange membranes as shown in the previous figures. In this embodiment the inserts 26b, 26c, 26d, and 26e are of different configurations than those shown in Figures 1 and 2. In particular, the insert 26d is a tubular member with a roughened exterior surface and an electrically conducting end portion 52 physically and electrically connected to end covering the entire cross-section of the tubular insert 26d. Such electrical and physical connection can be obtained by welding or other known bonding techniques as known to those skilled in the particular art. The peripheral portions of the cover 24b may optionally contain expansion grooves (not shown) to minimize any effects of thermal expansion of the covers upon the operation of the cell.

In operating the cell series as an electrolysis cell series for NaCl brine, certain operating conditions are preferred. In the anode compartment a pH of from about 0.5 to about 5.0 is desired to be maintained. The feed brine preferably contains only minor amounts of multivalent cations (less than about 80 parts per billion when expressed as calcium). More multivalent cation concentration is tolerated with the same beneficial results if the feed brine contains carbon dioxide in concentrations lower than about 70 ppm when the pH of the feed brine is lower than 3.5.

Operating temperatures range from 0° to 110°C, preferably from 60°C to 80°C. Brine purified from multivalent cations by ion-exchange resins after conventional brine treatment has occurred is particularly useful in prolonging the life of the membrane. A low iron content in the feed brine is desired to prolong the life of the membrane. Preferably the pH of the brine feed is maintained at a pH below 4.0 by the addition of hydrochloric acid. Preferably the operating pressure is maintained at less than 7 atmospheres.

Usually the cell is operated at a current density of from 1.0 to 4.0 amperes per square inch, but in some cases operating above 4.0 amps/in.<sup>2</sup> is quite acceptable.

## Claims

1. A structural frame adapted for use in an electrochemical cell comprising:  
a central cell element in the form of a generally planar member made of a polymeric material having a plurality of horizontally and vertically spaced-

apart shoulders protruding outwardly from at least one surface of said planar member;

at least one electrically conductive insert extending from an exterior face of a shoulder on one surface of the planar member, through the planar member, to an exterior face of a shoulder on the opposite surface of the planar member, wherein each of said shoulders annularly encircles and supports each of said inserts,

an electrically conductive, substantially completely hydraulically impermeable liner resistant to the corrosive effects of an electrolyte in said cell matingly contacted with at least one of said surfaces of said planar member and adapted to minimize contact between the electrolyte in said cell and said planar member.

2. The structural frame of Claim 1, wherein said liner is in contact with an anolyte surface of said planar member, said anolyte liner comprising a metal selected from titanium, tantalum, zirconium, tungsten, and alloys thereof.

3. The structural frame of Claim 1, wherein said liner is in contact with a catholyte surface of said planar member, said catholyte surface of said planar member, said catholyte liner comprising a metal selected from iron, steel, stainless steel, nickel, lead, molybdenum, cobalt, and alloys thereof.

4. The structural frame of Claim 1 or 2, wherein a liner is attached to opposite surfaces of said planar member, and said opposite surfaces are both anolyte surfaces.

5. The structural frame of Claim 1 or 3, wherein a liner is attached to opposite surfaces of the planar member, and said opposite surfaces are both catholyte surfaces.

6. The structural frame of Claim 1, 2 or 3, wherein a liner is attached to opposite surfaces of said planar member, one of said surfaces being an anolyte surface and the opposite side being a catholyte surface.

7. The structural frame of Claim 2 wherein the anolyte liner is titanium, or an alloy thereof; at least some of said inserts are made of a ferrous metal; and said liner is attached, by welding, to at least some of the said ferrous metal inserts through an intermediate metal which is weldable compatible with said titanium cover and said ferrous metal inserts.

8. The structural frame of any one of the preceding claims, wherein said inserts are made of a metal selected from aluminium, copper, iron, steel, nickel, titanium, alloys of these metals or physical combination of said metals.

9. The structural frame of anyone of the preceding claims wherein the polymeric material of the planar member is selected from polyethylene, polypropylene, polyvinylchloride, polystyrene, poly-

sulfone, styrene acrylonitrile, chlorinated polyvinylchloride, acrylonitrile, butadiene and styrene copolymers, epoxy, vinyl esters, polyesters, and fluoroplastics.

10. The structural frame of anyone of the preceding claims, wherein the polymeric material of the planar member contains from 5 to 75 weight percent of an additive selected from fiberglass, graphite fibers, carbon fibers, talc, glass beads, asbestos, and pulverized mica.

11. An electrochemical cell comprising: a plurality of the structural frames of any one of the preceding claims, wherein said frames are removably and sealably positioned in a generally coplanar relationship with each other and each of said planar members is spaced apart by an electrode selected from an anode on one or opposite sides of each of said frames or a cathode on one or opposite sides of each of said frames.

12. The cell of Claim 9 wherein each of said liners are welded to at least a portion of said inserts, and said electrodes are welded to the respective liners at locations adjacent to said inserts.

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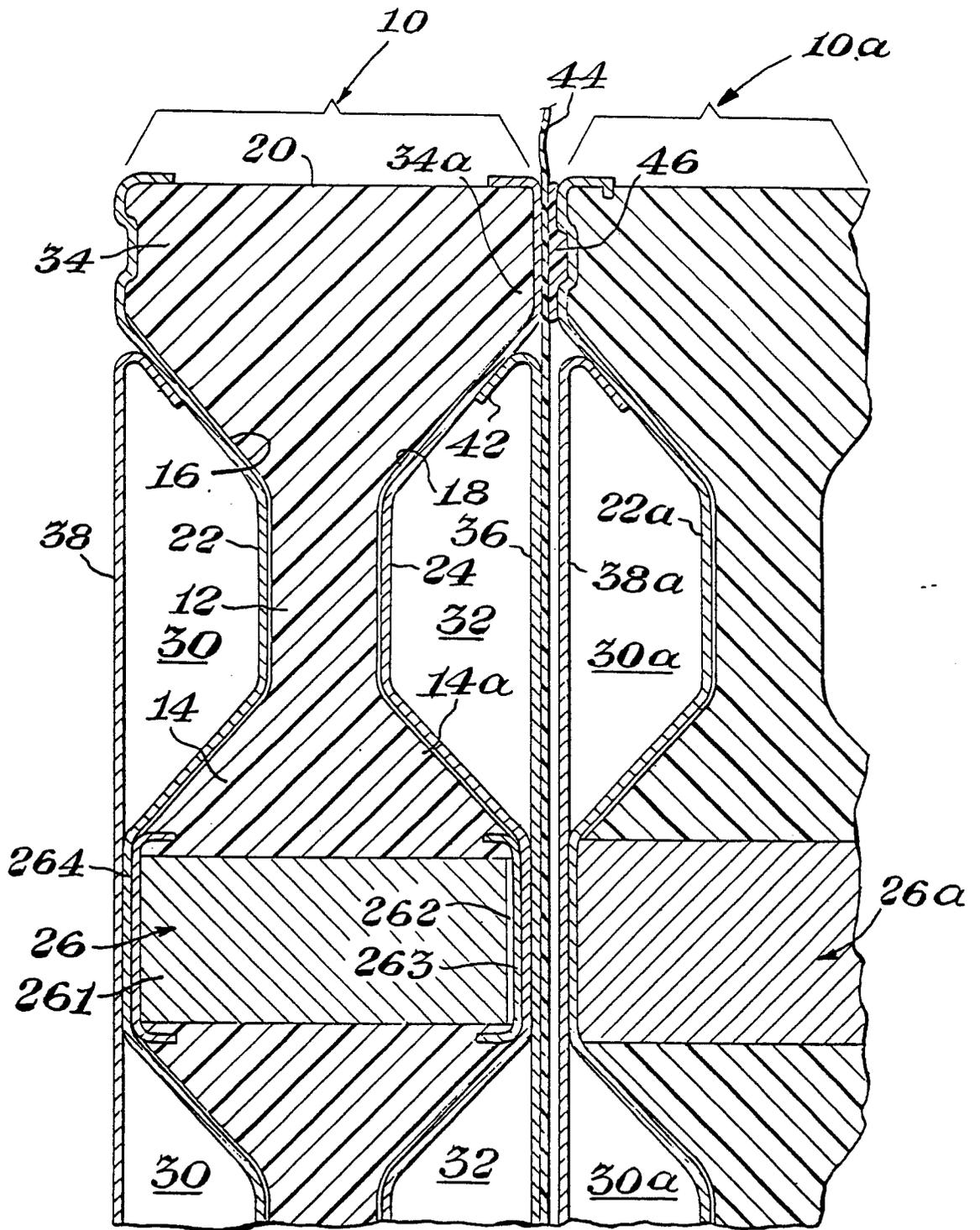


Fig. 1

Fig. 3

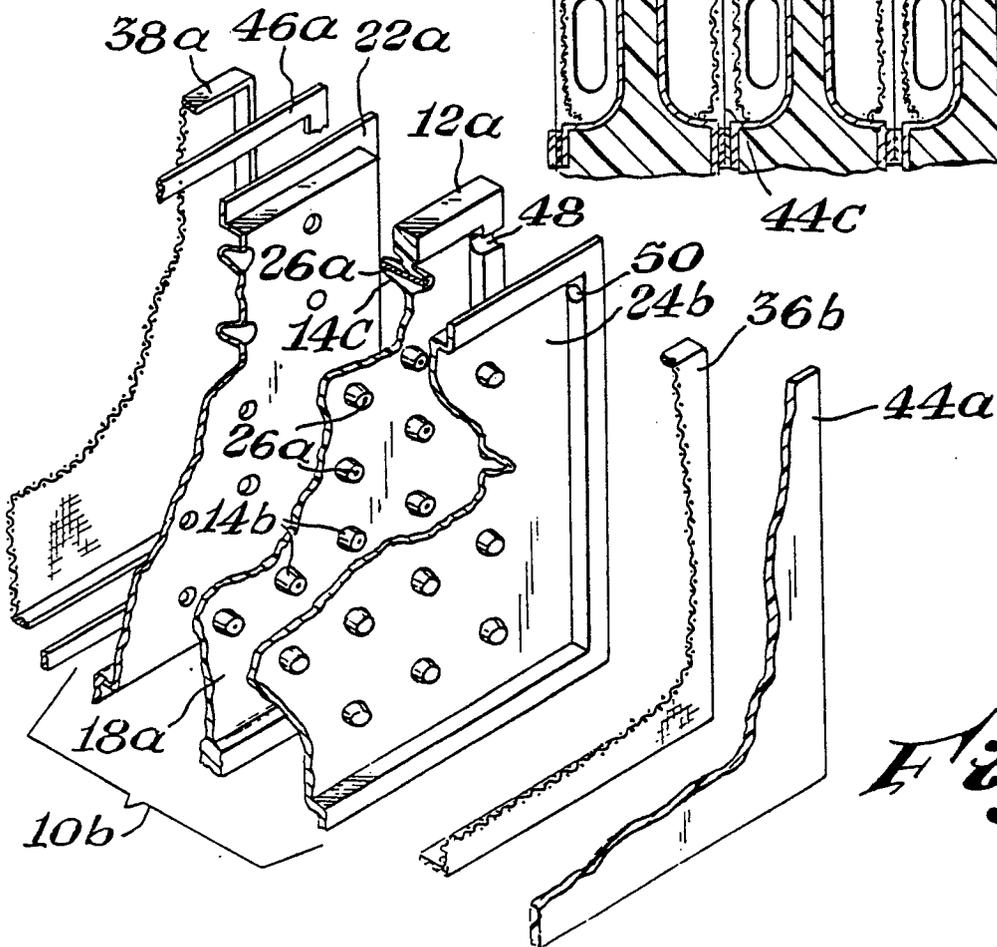
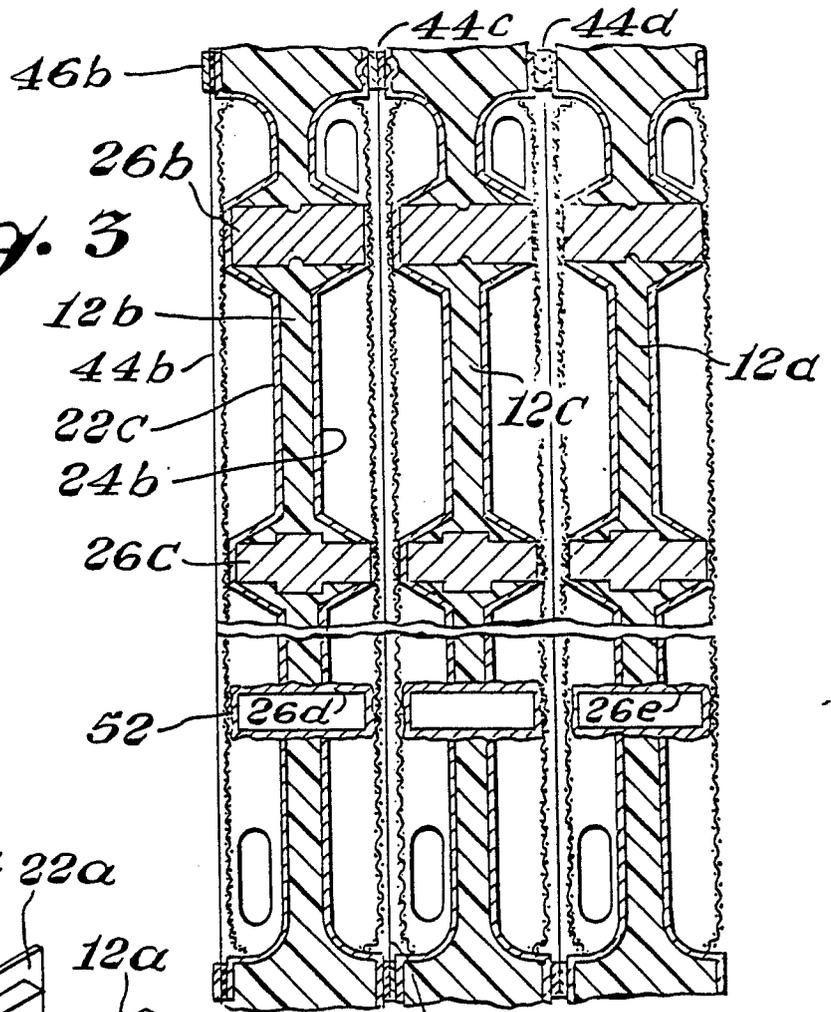


Fig. 2



| DOCUMENTS CONSIDERED TO BE RELEVANT   |  |   |   |
|---|--|---|---|
| Category  | Citation of document with indication, where appropriate, of relevant passages                            | Relevant to claim   | CLASSIFICATION OF THE APPLICATION (Int. Cl.4) |
| E   | US-A-4 666 580 (THE DOW CHEMICAL CO.)<br>* Whole document *<br>---                                       | 1-3,6-12  | C 25 B 9/00                                   |
| Y   | DE-A-2 600 345 (H.T. HYDROTECHNIK GmbH)<br>* Page 4, lines 21-25; page 7, lines 19-27; figure 4 *<br>--- | 1-6,11  |   |
| Y   | EP-A-0 185 270 (THE DOW CHEMICAL CO.)<br>* Whole document *<br>-----                                     | 1-6,11  |   |
|   |  |   | TECHNICAL FIELDS SEARCHED (Int. Cl.4)         |
|   |  |   | C 25 B 9                                      |
| The present search report has been drawn up for all claims  |  |   |   |
| Place of search<br>THE HAGUE  |  | Date of completion of the search<br>12-01-1988  | Examiner<br>GROSELLER PH.A.                   |
| CATEGORY OF CITED DOCUMENTS   |  | T : theory or principle underlying the invention<br>E : earlier patent document, but published on, or after the filing date<br>D : document cited in the application<br>L : document cited for other reasons<br>.....<br>& : member of the same patent family, corresponding document |   |
| X : particularly relevant if taken alone<br>Y : particularly relevant if combined with another document of the same category<br>A : technological background<br>O : non-written disclosure<br>P : intermediate document |  |   |   |