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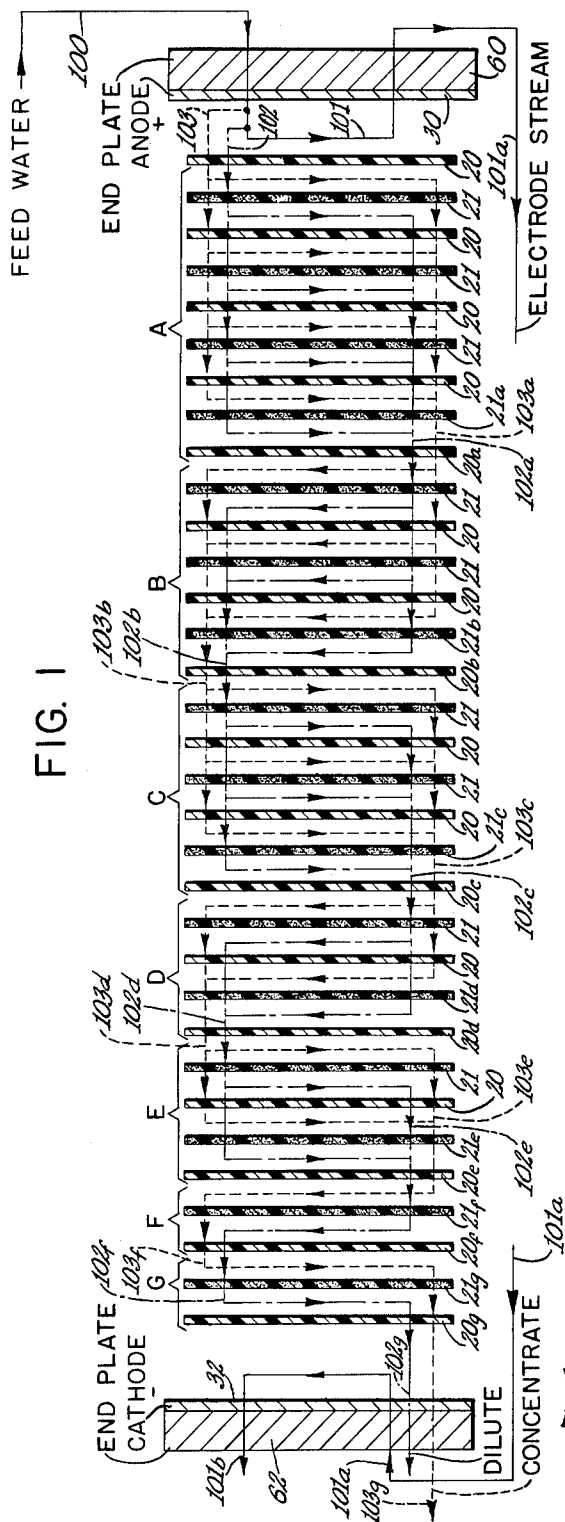
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3,223,606

ELECTRODIALYSIS DEVICE AND METHOD OF OPERATION

Original Filed May 22, 1959

6 Sheets-Sheet 1



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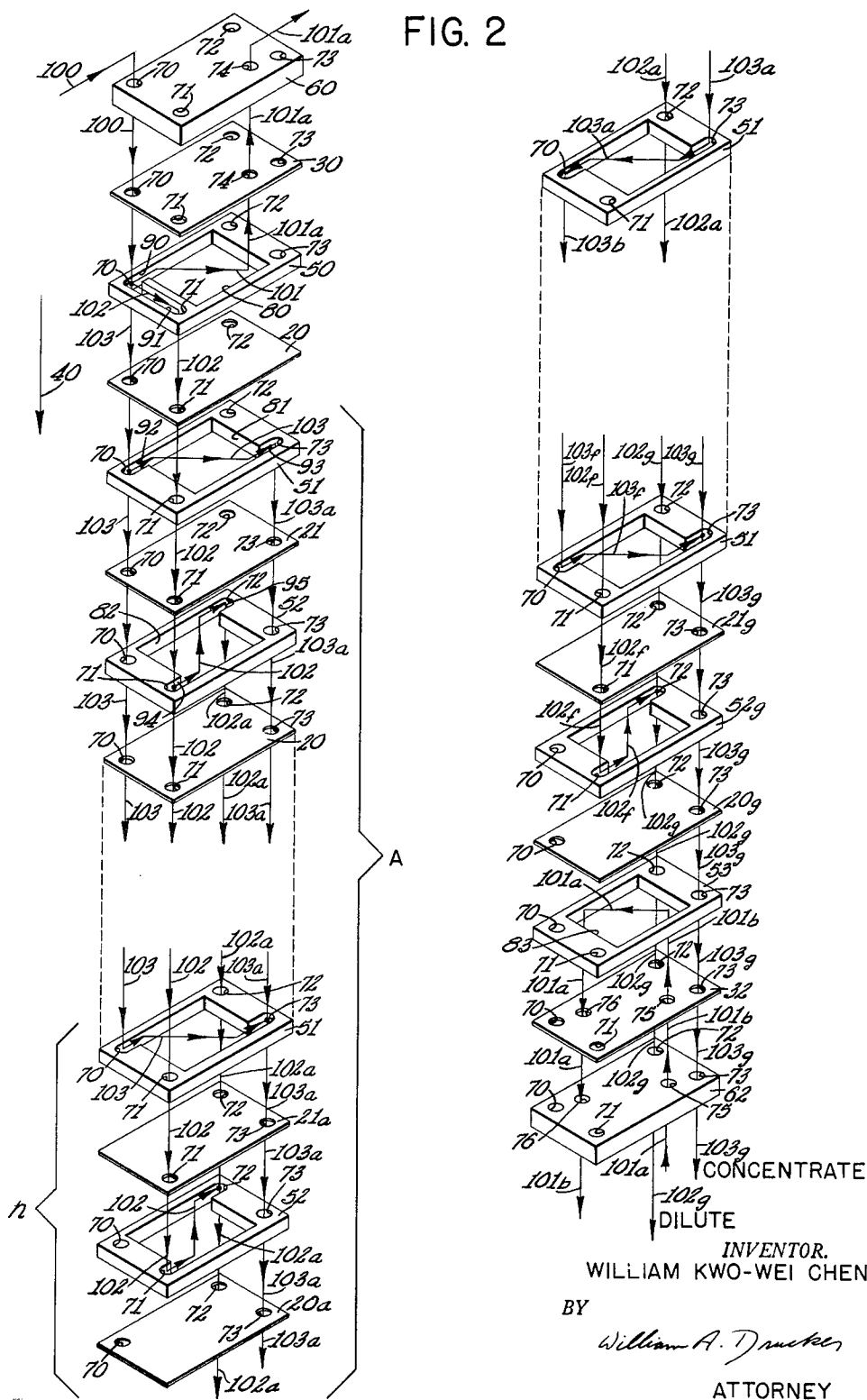
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ELECTRODIALYSIS DEVICE AND METHOD OF OPERATION

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FIG. 2



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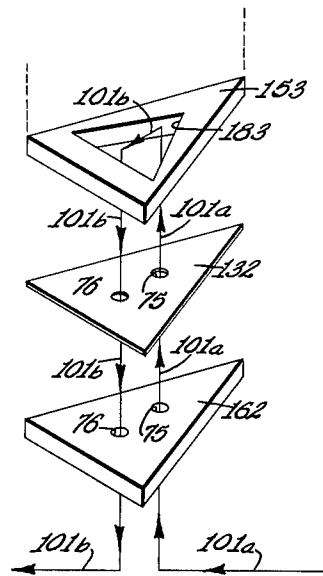
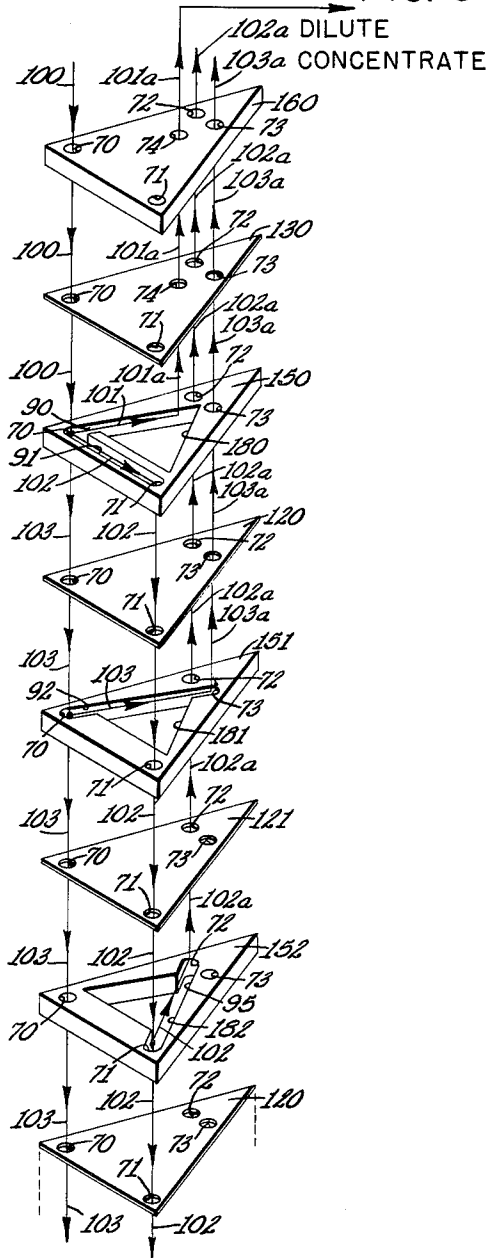
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ELECTRODIALYSIS DEVICE AND METHOD OF OPERATION

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6 Sheets-Sheet 3

FIG. 3



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ELECTRODIALYSIS DEVICE AND METHOD OF OPERATION

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6 Sheets-Sheet 4

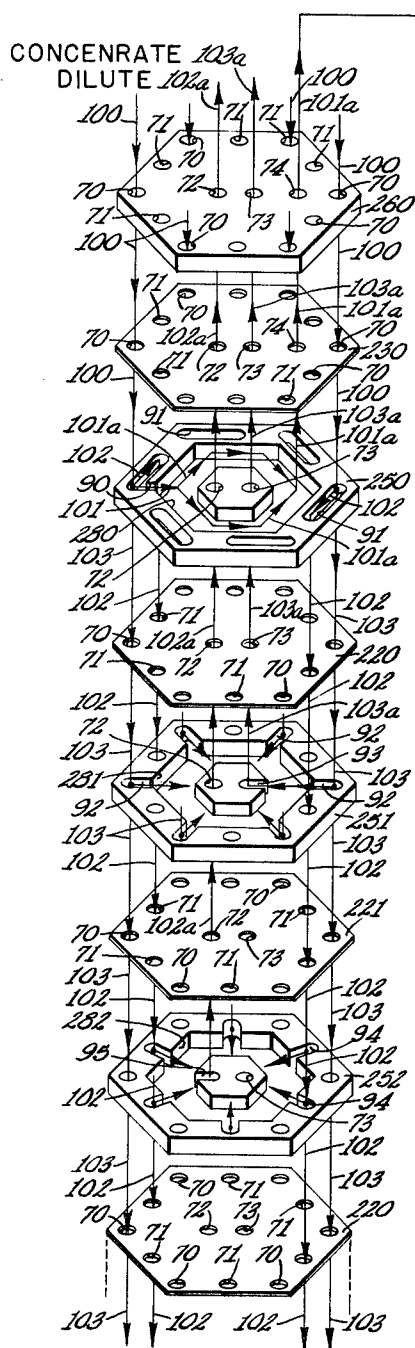
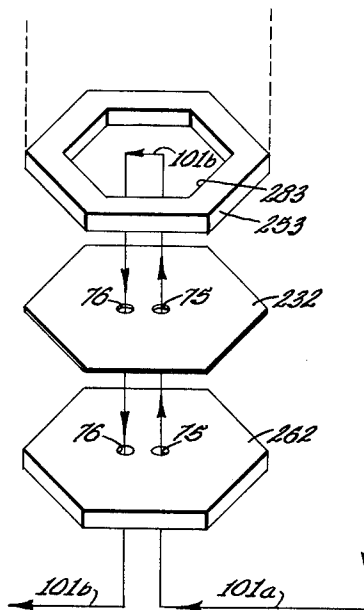


FIG. 4



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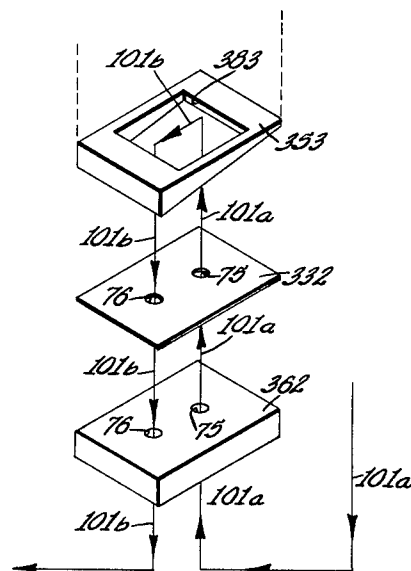
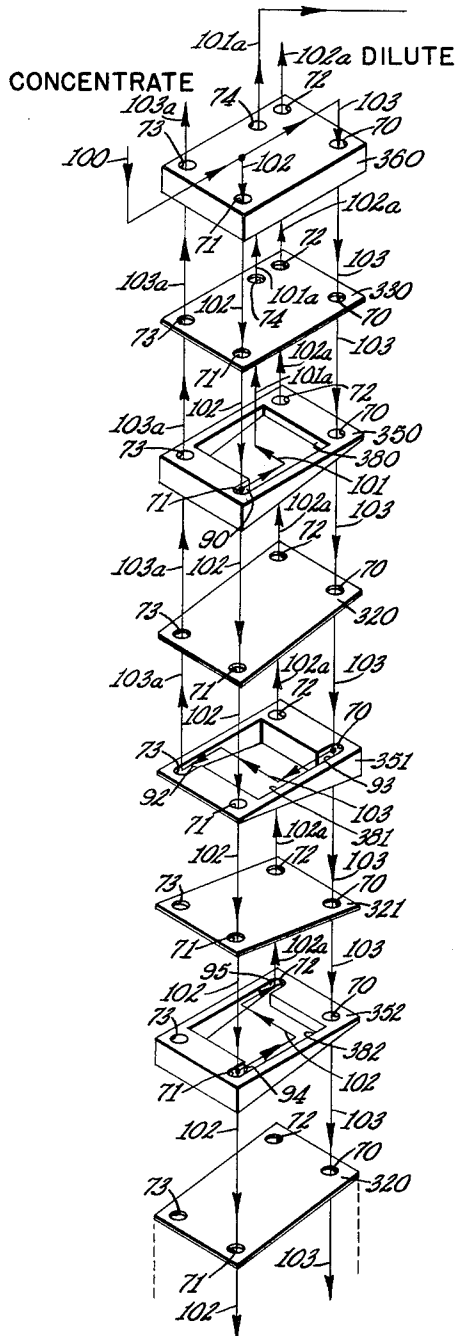
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ELECTRODIALYSIS DEVICE AND METHOD OF OPERATION

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6 Sheets-Sheet 5

FIG. 5



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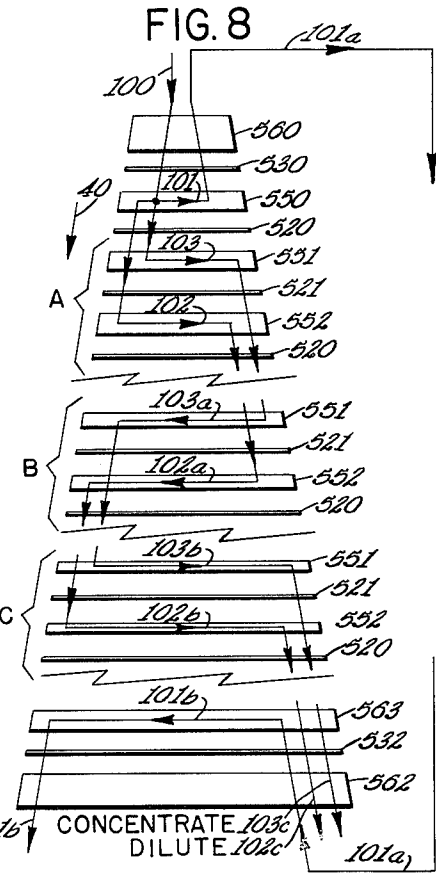
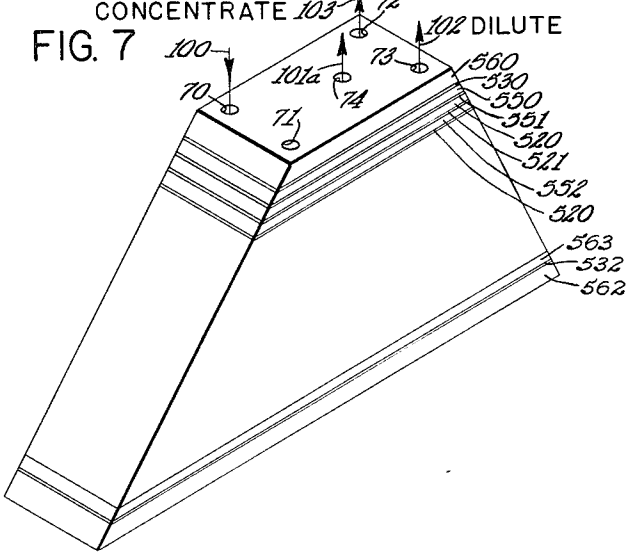
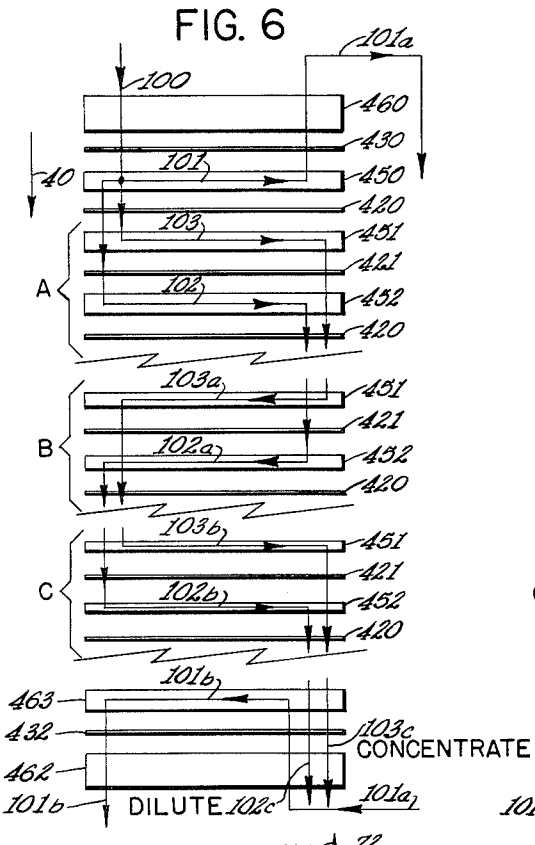
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ELECTRODIALYSIS DEVICE AND METHOD OF OPERATION

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6 Sheets-Sheet 6



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1

3,223,606

ELECTRODIALYSIS DEVICE AND METHOD OF OPERATION

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Continuation of applications Ser. No. 815,188, May 22, 1959, and Ser. No. 195,352, Mar. 30, 1962, which is a division of application Ser. No. 815,188, May 22, 1959.

This application Dec. 6, 1962, Ser. No. 243,217

5 Claims. (Cl. 204—180)

This application is a continuation of my copending applications S.N. 815,188 filed May 22, 1959, now abandoned, and Serial No. 195,352 filed March 30, 1962 as a division of application S.N. 815,188.

This invention relates to multiple chamber electro-dialysis devices and methods of operation of these devices. More particularly, the invention relates to electrodialysis with permselective membranes and to the hydraulic and electrical arrangement of such membranes in electro-dialysis devices.

In the operation of multicell electrodialysis apparatus, concentration polarization and consequent water dissociation at the membrane surfaces due to electrolysis effects have been difficult practical problems. Concentration polarization results in lower current efficiency and also causes pH changes in the solutions undergoing electrodialysis. Such pH changes may also induce precipitation of pH sensitive salts such as calcium carbonate, which, in turn, would cause scaling of membranes and obstruction of flow channels. High linear velocity of solutions has been used to reduce this difficulty. However, substantial residence time of the solution in contact with the membrane surface is desirable to achieve an economical degree of demineralization.

Various designs and methods have been proposed to combine both prolonged contact of solutions with ion exchange material and rapid solution circulation. One approach has been to recirculate product streams with the attendant disadvantage of extra recirculation pumps, valves, storage tanks and control devices. This method is described in British Patent No. 682,703.

Another approach to minimizing concentration polarization has been the use of tortuous hydraulic channels through the intermembrane spacers as described in U.S. Patent No. 2,708,658. This complex spacer design generally results in the inefficient use of membrane surface areas and high pumping energy requirements, and since the flow through these tortuous channels is in series, plugging at any point in the flow path would seriously reduce the rate of flow. The high pressures required for the operation of such a system also require the use of very rigid membranes to prevent their distortion under unbalanced pressure conditions.

Another method of dealing with polarization problems has been proposed in U.S. Patent No. 2,694,680. A number of independent electrodialysis assemblies are arranged in a staggered system of stages to achieve incremental demineralization from stage to stage. Solution velocities and electric current flow are adjusted in each stage for greatest efficiency. However, the use of individual electrodes, different direct current voltages and separate packaging leads to engineering complications and expensive systems. For small electrodialysis systems in particular, where the flow rates are less than about two gallons per minute, it would be very impractical to provide separate end blocks, end plates, electrodes and interconnecting piping systems for each small group of cells. Separate electrode streams for each assembly would be

2

especially impractical for low capacity systems because they require special handling.

Moreover, in Australian Patent No. 164,040 an electrodialysis process is described in which the stream to be demineralized is passed successively in series through every alternate compartment from one electrode to another to achieve a degree of demineralization higher than would result from parallel rather than series flow. The maximum electrical current that can be used in an electrodialysis cell without excessive concentration polarization is proportional to the concentration of electrolytes in the cell. In a series stack the limiting current through the stack is therefore governed by the concentration in the last product cell. This requirement automatically implies that the current flowing through the preceding product cells is lower than the maximum current which may be applied to these cells, and the process results in the uneconomical use of these cells. Since all compartments are in series, high hydraulic pressure requirements are also encountered at desirable solution velocities.

Therefore, it is an object of this invention to provide a method of electrodialysis which controls concentration polarization effects.

It is also an object of this invention to provide an electrodialysis device which incorporates means for producing high solution velocities in the sections of an electrodialysis device where such high velocities are required to reduce concentration polarization while maintaining lower solution velocities where maximum residence time (for higher degree of demineralization) and minimum hydraulic pressure drops are desired.

It is also an object of this invention to provide a multicell electrodialysis device which produces the desired gradation in solution velocity and reduces concentration polarization by incorporating a series of cascaded membrane cells between a single electrode pair so that the cross sectional area in succeeding cascades progressively decreases in the direction of flow.

It is also an object of this invention to provide a method of electrodialysis by which a solution can be demineralized with a minimum expenditure of hydraulic pumping energy and a minimum degree of polarization between a single pair of electrodes.

It is also an object of this invention to provide a method of fluid demineralization in a multi-compartment electrodialysis device characterized by the step of establishing a solution velocity gradient in inverse relation to the solution salinity gradient at constant current density between a single pair of electrodes.

It is also an object of this invention to provide a method of fluid demineralization in a multi-compartment electrodialysis device characterized by the step of establishing between a single pair of electrodes a current density gradient in direct relation to a solution salinity gradient at constant solution velocity.

It is also an object of this invention to provide a method of fluid demineralization in a multi-compartment electrodialysis device characterized by the step of establishing between a single pair of electrodes a graduated membrane area increasing in size in the direction of fluid flow.

These and other objects of the invention are described in the following detailed account of the invention and in the attached drawing in which reference characters of similar elements correspond and in which:

FIG. 1 is a schematic cross-section of an electrodialysis stack, the preferred form of the invention, showing alternate anion and cation permeable membranes arranged to form diluting and concentrating cells. The liquid flow patterns or hydraulic circuits are depicted by lines with arrows;

FIG. 2 is an exploded isometric view of the electrodialysis stack shown schematically in FIG. 1;

3

FIG. 3 is an exploded isometric view of a modified form of the invention, an electro dialysis stack employing membranes and intermembrane gaskets having a triangular cross-section;

FIG. 4 is an exploded isometric view of another form of the invention, an electro dialysis stack employing membranes and intermembrane gaskets of hexagonal cross-section and utilizing a radial flow pattern;

FIG. 5 is an exploded isometric view of an electro dialysis stack employing wedge-shaped intermembrane gaskets, another form of the invention;

FIG. 6 is a schematic cross-section of an electro dialysis stack employing intermembrane gaskets of varying thicknesses, another form of the invention;

FIG. 7 is an isometric view of an assembled electro dialysis stack employing membranes and intermembrane gaskets of varying cross-sectional area, another form of the invention;

FIG. 8 is a schematic cross-section of the electro dialysis stack shown in FIG. 7.

In general the various embodiments of the invention include a method of electro dialysis in which the velocity of liquid or the density of electric current flowing through an electro dialysis stack is adjusted along the solution flow path in relation to polarizing conditions established by a salinity gradient between the electrodes in the electro dialysis stack configurations in which these methods may be applied.

When raw feed solution enters the electro dialysis device, it is divided into two main parts. The first part becomes the "product" or dilute stream and the second part becomes the "waste" or concentrate stream. These descriptions apply when dilution rather than concentration is the desired effect.

As the dilute stream becomes more dilute in the course of flow through the device, a salinity gradient is established between the beginning and end of the dilute flow. Near the end of the dilute flow, as the stream is about to emerge from the device as a demineralized "product," the solution is no longer so good an electrical conductor as in its initial concentrated form. Polarization, or water dissociation at the membrane surfaces, as a result of too dilute (and therefore poorly conducting) a layer forming at the interfaces becomes particularly acute as the ratio of current density to salinity increases.

Increased solution velocity in relation to the decrease of salinity tends to overcome polarization, and the method of achieving velocity control is a first feature of this invention.

By decreasing the current density in particular parts of the device where the salinity has decreased, the ratio of current density to salinity is reduced and polarization is controlled. Reducing the current density as the salinity declines tends to maintain an optimum ratio of current density to salinity and controls polarization. Reduction of current density as salinity declines is a second feature of this invention.

Both control of fluid flow velocity and change of current density may be used separately or together in the same device in the practice of this invention, but for simplicity they are illustrated separately herein since both contribute to control of the same polarization effect.

According to a preferred form of this invention, as shown in FIG. 1, solution residence time is controlled by connecting groups of cells or cascades hydraulically in series between a single set of electrodes 30, 32. By using a single electrode pair, a saving is made in the cost of supplying multiple electrodes, and the power losses associated with electrode over-voltage are reduced.

Each hydraulic pass at right angles to the direction of electrical current as indicated by the arrow 40 becomes an electrical cascade, and the change in concentration per cascade is proportional to the number of diluting cells or chambers per cascade,

4

Thus, in FIG. 1 and FIG. 2, the first cascade A containing four diluting cells (a diluting cell is formed within a stack when it is bounded by an anion membrane on the anode side and a cation membrane on the cathode side) will have a dilute stream solution velocity only $\frac{3}{4}$ that of the second cascade B with only three diluting cells.

Since the change in concentration in each cell is proportional to the electric current (which is constant in this case) and inversely proportional to the flow rate in the cell, it is seen that the first cascade A will undergo $\frac{4}{3}$ the change in concentration of the second cascade B. Since all the cells are electrically in series, the average current density, CD, in all cells will be the same. In the diluting cells, however, the average electrolyte concentration will be progressively lower in each succeeding cascade. The ratio of current density to solution normality, CD/N , is an indication of polarizing conditions and will increase from the first cascade A to the seventh cascade G. In FIG. 1, cascades A, B, C, D, E, F, G contain 4, 3, 3, 2, 2, 1, 1 cells, respectively.

Since higher CD/N ratios are tolerable at higher solution velocities, successive electrical cascades comprise a progressively reduced number of diluting cells in parallel. This type of membrane stack configuration or array thereby facilitates the use of designs in which the flow rates through different cells are optimized for the electrochemical conditions which exist therein.

FIG. 2 is an exploded isometric view of the electro dialysis stack in FIG. 1 showing the components used in the preferred form of the invention and the methods used to direct the various streams through the components. The principal components are end blocks 60, 62, electrodes 30, 32, intermembrane gaskets 50, 51, 52, 53 and membranes 20, 21, 20A, 21A. A clamping device, used to hold these components in liquid-tight relationship, is not shown.

In operation the solution to be demineralized is fed as a single stream 100 to a manifold hole 70 which is formed by the alignment of symmetrical holes 70 in the various components comprising the stack. An intermembrane gasket 50 serves both to define a flow path 80 for the anode rinsing stream and to split the feed stream 100 into three auxiliary streams 101, 102, 103. The three-way split is achieved by slits 90, 91 in a gasket 50 which communicates with the flow path 80 in the intermembrane gasket 50 and manifold hole 71. The function of the anolyte stream 101 is to collect and carry away the products of electrolysis at the anode 30. Where this stream leaves the stack, it is given the designation 101A in the drawing to denote its change in composition.

The dilute stream 102 undergoes dilution in respect to the solute during electro dialysis. It is distributed from a manifold 71 via slits 94 to the alternately arrayed intermembrane gaskets 52. Where this stream leaves the flow paths 82 in the gaskets 52 via slits 95, it is designated 102A in the drawing to denote the change in composition due to electro dialysis. In subsequent cascades, this stream is designated 102B, 102C, 102D, etc. in the drawing to denote progressive additional changes in composition.

The concentrate stream 103 undergoes concentration in respect to the solute during electro dialysis. It is distributed from a manifold 70 to the alternately arrayed intermembrane gaskets 51 via slits 93. Where this stream leaves the flow paths 141 in intermembrane gaskets 51 via slits 93, it is designated 103A in the drawing to denote the change in composition due to electro dialysis. In subsequent cascades this stream is designated 103B, 103C, 103D, etc. in the drawing to denote progressive additional changes in composition.

When the first electrode 30 is an anode and the second electrode 32 is a cathode, the first and alternate membranes 20 are cation permselective membranes and the

intervening membranes **21** are anion permselective membranes. In the last parallel pass of each cascade (denoted in FIG. 2 as the n th pass of a cascade having n parallel passes) membranes **21A**, **20A** containing only three holes each are used in place of the anion and cation membranes **21**, **20**. Hydraulic blocks **70**, **71** are thereby created at the inlet stream manifolds. The dilute stream effluents **102A** from cascade A are thus recombined in the dilute stream manifold hole **72** and are redistributed through the alternately arrayed gaskets **52** in cascade B. Since cascade B contains fewer parallel passes and a smaller total cross-sectional flow path for the dilute stream **102A** than does cascade A, the average linear solution velocity will be greater in cascade B. It should also be noted that the direction of flow of the dilute stream is reversed by this process.

The concentrate stream effluents **103A** are similarly recombined in the concentrate stream manifold hole **73** and are redistributed through the alternately arrayed gaskets **51** in cascade B. Since cascade B contains fewer parallel passes for the concentrate stream **103A** than does cascade A, the average linear solution velocity will be greater in cascade B.

In the case of the concentrate stream progressively increasing solution velocity is not an essential feature. It is shown only for the sake of symmetry as an aid in the description of a cascaded structure. Similarly, the internal development of several process streams **101**, **102**, **103** from a single feed stream is not essential to the practice of this invention but shows the preferred method of construction.

The anolyte stream **101A** flowing out of the top of the stack through holes **74** in the anode and end block respectively is utilized again at the bottom of the stack where it is directed through holes **75** in the end block **62** and cathode **32** respectively into the flow path **83** of the intermembrane gasket **53**. Since the intermembrane gasket **53** has no slits communicating with the manifold holes **70**, **71**, **72**, **73**, the catholyte stream **101A** is isolated from the dilute and concentrate streams and leaves the stack through holes **76** in the cathode and end block respectively as stream **101B**. Alternately, raw feed solution from stream **100**, the concentrate stream effluent **103G** or a portion of the dilute stream effluent **102G** could be used for the cathode wash stream **101A-B**.

FIG. 3 is an exploded isometric view of another embodiment of this invention in which the end blocks **160**, **162**, electrodes **130**, **132**, the intermembrane gaskets **150**, **151**, **152**, **153** and membranes **120**, **121** are all of triangular shape. A clamping device, used to hold these components in liquid-tight relationship, is not shown.

In operation the solution to be demineralized is fed as a single stream **100** to the manifold hole **70** at the base of the triangle which is formed by the alignment of symmetrical holes **70** in the various components comprising the stack.

The intermembrane gasket **150** serves both to define a flow path **180** for the anode rinsing stream and to split the feed stream **100** into three auxiliary streams **101**, **102**, **103**. The three-way split is achieved through the use of slits **90**, **91** in the intermembrane gasket **150** which communicate with the flow path **180** in the intermembrane gasket **150** and the dilute inlet manifold hole **71**. The anode rinse stream **101** is called the anolyte. Its function is to collect and carry away the products of electrolysis at the anode **130**. Where this stream leaves the stack, it is given the designation **101A** to denote its change in composition.

The dilute stream **102** is distributed from the dilute stream manifold **71** via slits **94** to the alternately arrayed intermembrane gaskets **152** and is electrodialed in the same manner as that described for FIG. 1. In this case, however, all intermembrane gaskets **152** are connected hydraulically in parallel, and the desired increase in solution velocity along the salinity gradient and desalination

flow path **182** is achieved by the progressively decreasing cross-sectional flow area in the flow path.

The concentrate stream **103** is distributed from the concentrate stream manifold **70** via slits **92** to the alternately arrayed intermembrane gaskets **151** and undergoes electrodialytic concentration along its flow paths **181**. Parallel construction and parallel flow of the concentrate stream is illustrated for simplicity but is not essential for the operation of the apparatus.

The electrode rinse stream **101-101A-101B** flows in series through the anolyte flow path **180** and the catholyte flow path **183** in a manner similar to that described for FIG. 2.

Solution entering at the base of the triangle flows toward the apex. Thus, the flow path becomes constricted as solution flows toward the triangle apex, and velocity increases. The increase in velocity corresponds with a decrease in solution salinity. Decreased salinity is usually conducive to polarization problems, whereas increased velocity of flow has a correcting effect. As the polarization conditions become more acute, they are compensated by higher flow rates. The triangular flow path has been chosen as representative of the principle of this embodiment, but other flow path cross-sections such as trapezoids would serve as well, provided only that the solution is forced to increase its velocity progressively along the direction of flow.

FIG. 4 is an exploded isometric view of another embodiment of this invention in which end blocks **260**, **262**, electrodes **230**, **232**, intermembrane gaskets **250**, **251**, **252**, **253** and membranes **220**, **221** are all of hexagonal shape. A clamping device, used to hold these components in liquid-tight relationship, is not shown.

The functions of the various components are identical with corresponding components shown in FIG. 3. In this case, however, the desired increase in solution velocity along the desalination flow path **282** is achieved by radial flow of the dilute stream **102** from the multiple dilute stream inlet manifolds **70** located in the periphery of the intermembrane gaskets **252** toward the dilute stream outlet manifold **72** located in or near the center of the intermembrane gaskets **252**. Solution velocity increases as the liquid moves to the more constricted central flow path.

The concentrate stream **103** is similarly manifolded in alternately arrayed gaskets **251**, but this construction feature is not essential to the operation of apparatus.

The hexagonal shape of the various stack components has been chosen for simplified illustration. Other polygonal, circular or elliptical shapes having a radial symmetry would serve as well, provided only that the flow is directed from the periphery toward the center or core of the diluting compartments.

FIG. 5 is an exploded isometric view of another embodiment of this invention in which intermembrane gaskets **350**, **351**, **352**, **353** are wedge-shaped and so oriented in respect to one another that their complete assembly constitutes a right prismatic stack.

The dilute stream **102** is directed through flow path **382** in gaskets **352** so that the decrease in cross-sectional area in the gasket imparts a progressively increasing solution velocity along the desalination flow path.

In FIG. 5 the concentrate stream **103** originates outside the stack from the feed stream **100** and is directed separately into the concentrate stream manifold **70**. The concentrate stream thus flows countercurrent to the dilute stream and is also subjected to progressively increasing velocity. Countercurrent flow of the concentrate stream is not essential to the practice of this invention, however, and has been illustrated only to show its method of application to the invention.

FIG. 6 is a schematic cross-sectional view of an electrodialed stack depicting another embodiment of this invention in which the intermembrane gaskets **451**, **452**

7

have progressively reduced thicknesses in each of the successive electrical cascades A, B, C. The dilute stream 102 flowing in series through each cascade is thereby forced to increased linear velocity along the desalination flow path. The concentrate stream 103 is similarly affected by the configuration shown in FIG. 6, but this feature is not essential to the practice of the invention. The function of all other corresponding stack components is essentially the same as that described for FIG. 2.

FIG. 7 is an isometric view of an assembled electro-dialysis stack showing another embodiment of this invention in which the intermembrane gaskets 551, 552 have progressively larger areas in each of the successive electrical cascades A, B, C.

The dilute stream 102 and concentrate stream 103 flow paths through this stack are illustrated by reference to the schematic cross-section shown in FIG. 8. The solution velocity of the dilute stream 102 changes throughout the stack array from cascade A to B to C as is also seen in FIGURE 6 and the total electrical current flow 40 is distributed over a progressively larger area in the same general direction that the dilute stream 102 follows.

The current density (total current divided by the area through which it flows) is progressively, although not necessarily linearly, reduced along the desalination flow path and achieves the same depolarizing effect as that realized by increasing solution velocity at constant current density.

The concentrate stream 103 is shown in parallel and concurrent flow relative to the dilute stream 102, but this feature is not essential to the application of the invention. All other stack components have functions similar to those described for FIG. 2.

Example I

An electrodialysis stack constructed as shown in FIGS. 1 and 2 comprised seventeen cation membranes, sixteen anion membranes, sixteen diluting compartments, sixteen concentrating compartments, one anolyte compartment and one catholyte compartment. The diluting and concentrating compartments arranged in cascades from A to G comprised 4, 3, 3, 2, 2, 1 and 1 compartment each respectively. Each intermembrane gasket was five centimeters wide by thirty-eight centimeters long by one-tenth centimeter thick and had a flow path area two and one-half centimeters by thirty and one-half centimeters (seventy-six square centimeters). A turbulence promoting screen similar to that shown in U.S. Patent No. 1,972,433 was located within the flow path of each gasket.

At a pressure of 700 grams per square centimeter and a total flow rate of 5 ml./sec. to the stack, the flow rates to each stream were distributed as follows:

	ml./sec.
Dilute stream -----	2
Concentrate stream -----	2
Electrode streams (in series) -----	1

When the composition of the feed solution was adjusted to 3,000 parts per million of sodium chloride at pH 7 and a direct current potential of 18 volts applied, a steady state current of 0.55 ampere was observed. Under these conditions the product water (dilute stream) contained 450 parts per million of sodium chloride and its pH was 6.7. Thus a high degree of demineralization was achieved with control of polarization.

Since slight pH changes normally occur during electro-dialysis, the exact pH change at which polarization is considered undesirable is not critical. A change in pH of about three units in the range of pH 7, however, would be considered unacceptable, whereas a pH change of about 0.5 unit would be satisfactory in most cases.

Example II

An electrodialysis stack was constructed with the components used in Example I and comprised sixteen diluting compartments in series, sixteen concentrating compart-

8

ments in series, one anolyte compartment and one catholyte compartment. At a pressure of 4.2 kilograms per square centimeter and a total flow rate of 5 ml./sec., the flow rates to each stream were distributed as follows:

	ml./sec.
Dilute stream -----	2
Concentrate stream -----	2
Electrode streams (in series) -----	1

When the composition of the feed solution was adjusted to 3,000 parts per million of sodium chloride at pH 7 and a direct current potential of 15 volts applied, a steady state current of 0.55 ampere was observed. Under these conditions the product water (dilute stream) contained 470 parts per million of sodium chloride and the pH was 6.6.

Thus it is seen that a considerably higher pressure is required with all the cells in series to achieve the same degree of demineralization as that achieved in Example I.

After an overnight operation under the above described conditions, it was found that this excessively high pressure caused leakage and ultimate failure of the system.

There have thus been described methods and apparatus for electrodialysis in which the velocity of liquid and the density of electric current flowing through an electro-dialysis system are adjusted along the solution flow path in relation to polarization conditions and in particular in relation to the salinity gradient in the stacked membrane and gasket array, and the ratio of current density to salinity is controlled.

What is claimed is:

1. In an electrodialysis device, a pair of electrodes, means for imposing a direct current potential between said electrodes, a plurality of ion-permeable membranes and intermembrane gaskets arranged in substantially flat stacked array between the electrodes and providing alternate concentrating and diluting compartments, a dilute stream inlet and outlet for said device, said diluting compartments providing a serial hydraulic flow path from said inlet to said outlet, said membranes being progressively larger in surface area from said inlet to said outlet whereby the electric current density across said membranes between the electrodes is progressively decreased in the direction of flow of the dilute stream.

2. An electrodialysis device comprising:

- first and second electrode means;
- means for imposing a direct current potential between said electrode means;
- a plurality of permselective membranes arranged in said electrodialysis device between said first and second electrode means;
- gasket means forming concentrating and diluting chambers between said membranes;
- fluid inlet and outlet means for said chambers;
- means for connecting said diluting chambers in serial flow in a direction from the first electrode means to the second electrode means whereby an electrolyte stream is de-ionized in successive diluting chambers; and
- said gasket means forming diluting chambers having a decreasing membrane separation between at least some of the membranes in a direction from said first electrode means to said second electrode means whereby the linear velocity of the electrolyte stream increases with decreasing concentration of electrolyte.

3. The device of claim 2 further characterized by said chambers and membranes having an increasing cross-sectional area in a direction from said first electrode means toward said second electrode means, whereby the current density between electrodes decreases with decreasing concentration in said electrolyte stream.

4. A method of electrodialysis comprising:

- imposing a direct current potential between first and second electrode means;

- (b) arranging a plurality of permselective membranes between said electrode means;
- (c) forming concentrating and diluting chambers, having inlet and outlet means, between said membranes; 5
- (d) connecting said diluting chambers in serial flow in a direction from said first electrode means to said second electrode means;
- (e) deionizing an electrolyte stream by successive passage through said diluting chambers; and 10
- (f) increasing the linear velocity of said electrolyte stream as said stream is deionized by decreasing the separation between at least some of said membranes in a direction from said first electrode means toward said second electrode means. 15
5. The method of claim 4 further comprising: decreasing the current density between said electrode means as the concentration in said electrolyte stream is decreased by increasing the cross-sectional membrane area in a direction from said first electrode means toward said second electrode means.

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