Nov. 5, 1974 J. F. GIFFORD 3,846,274

ELECTROPERISTATIC ION PUMP

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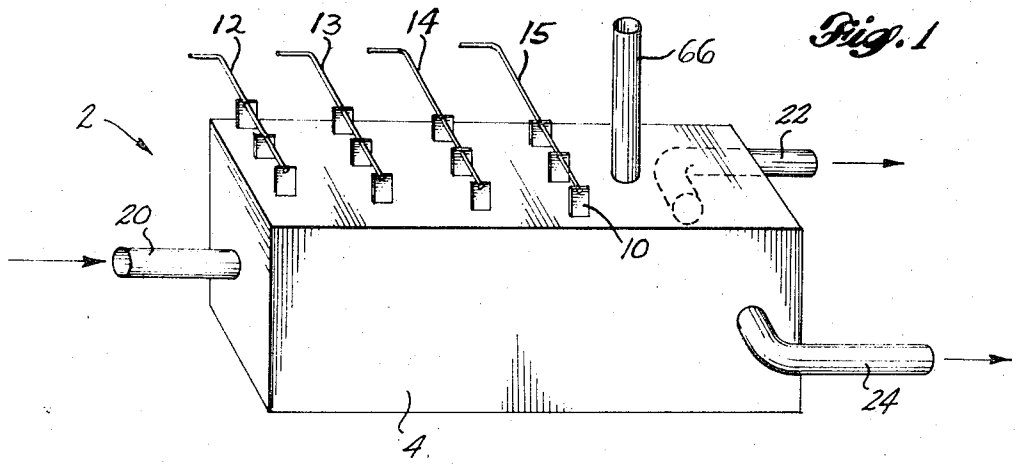


Fig. 1

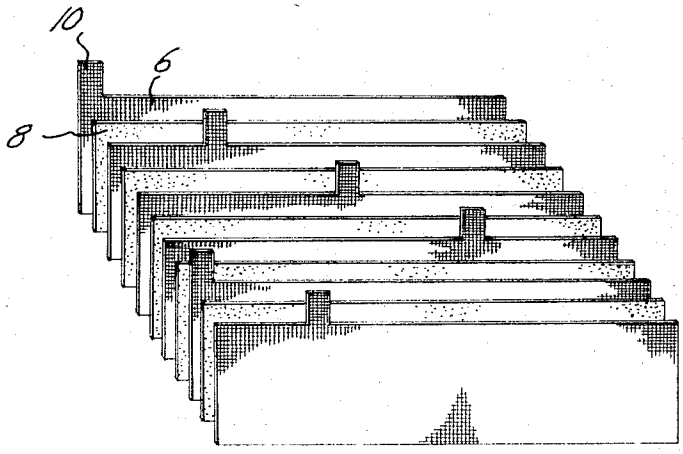


Fig. 2.

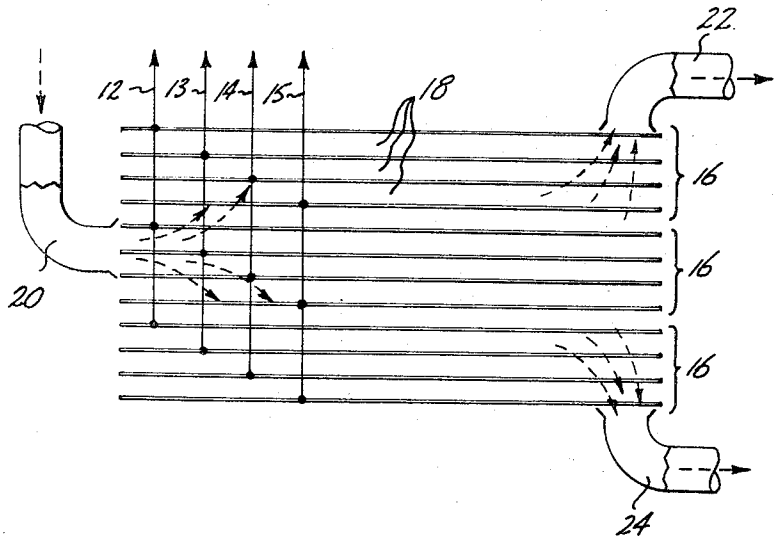


Fig. 3.

ELECTROPERISTATIC ION PUMP

Filed April 10, 1972 9 Sheets-Sheet 2

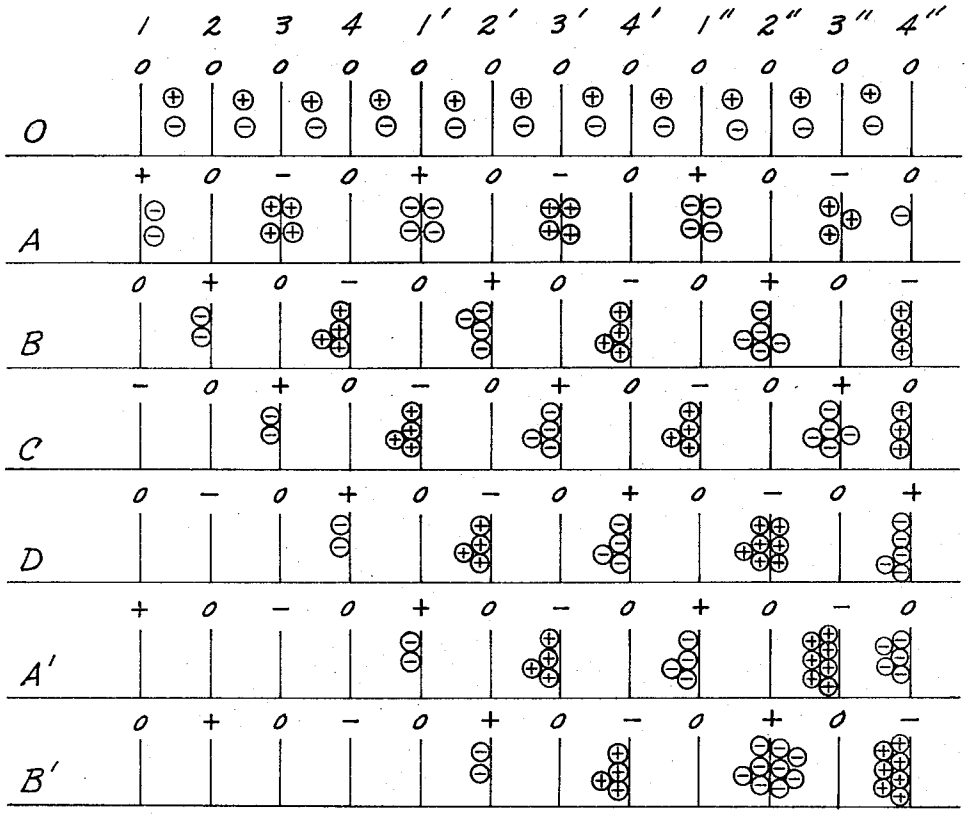


Fig. 5.

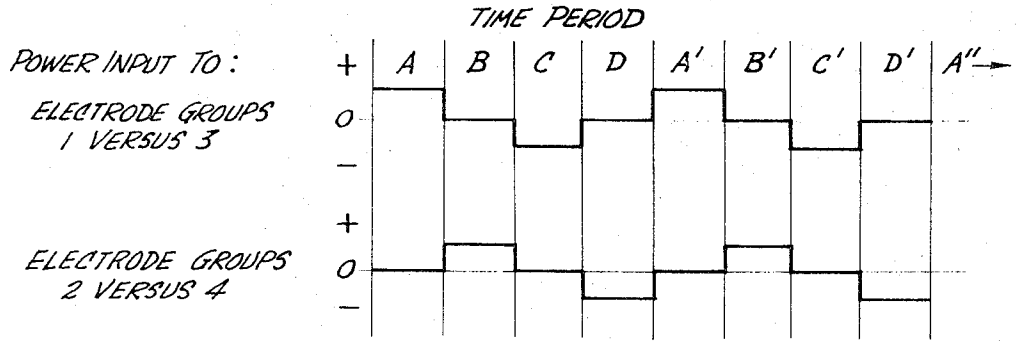
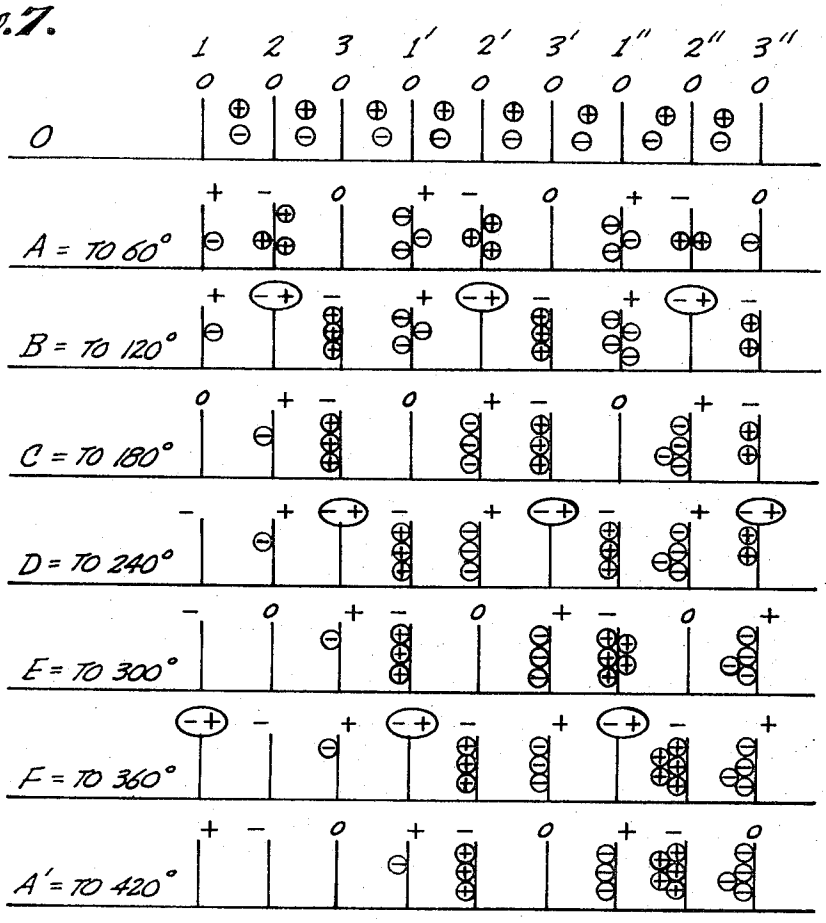


Fig. 4.

ELECTROPERISTATIC ION PUMP

Filed April 10, 1972 9 Sheets-Sheet 3

Fig. 7.



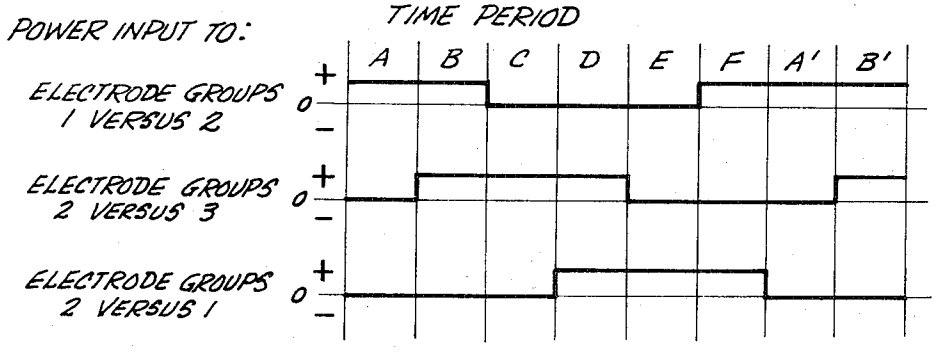
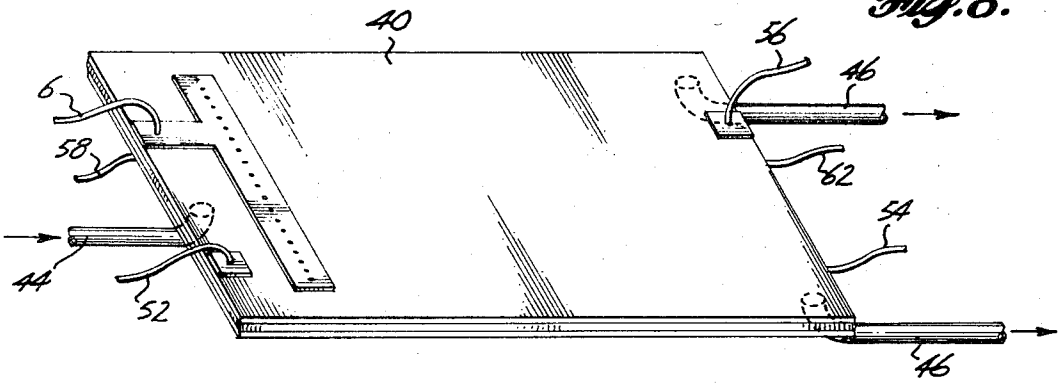


Fig. 6.

ELECTROPERISTATIC ION PUMP

Filed April 10, 1972 9 Sheets-Sheet 4

Fig. 8.



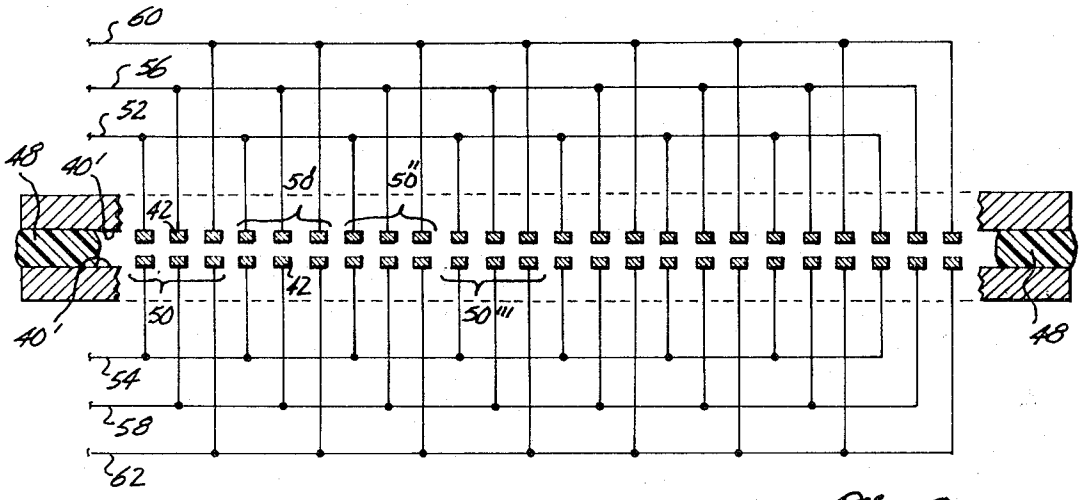


Fig. 9.

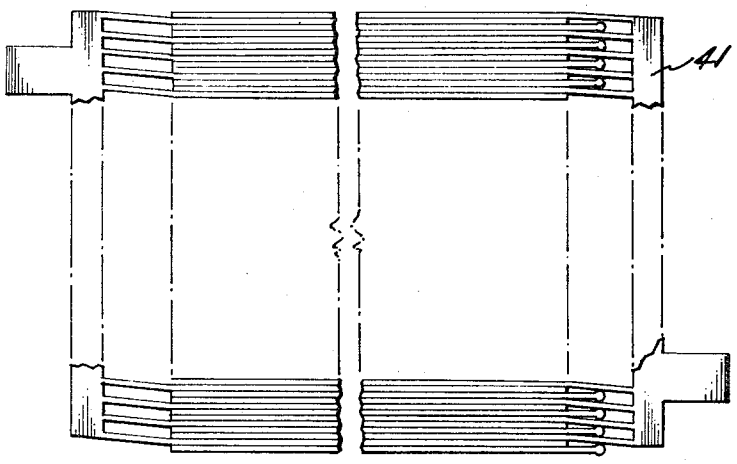


Fig. 10.

ELECTROPERISTATIC ION PUMP

Filed April 10, 1972 9 Sheets-Sheet 5

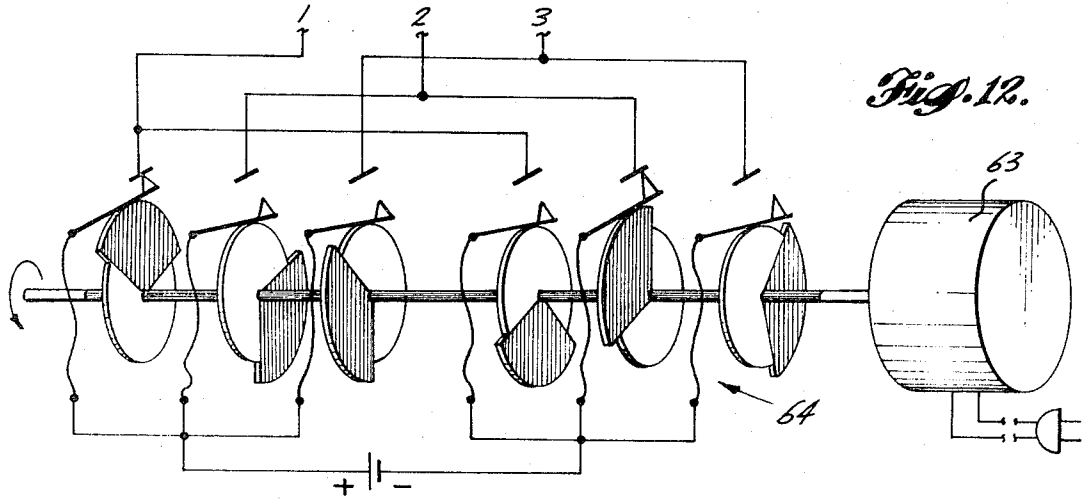


Fig. 12.

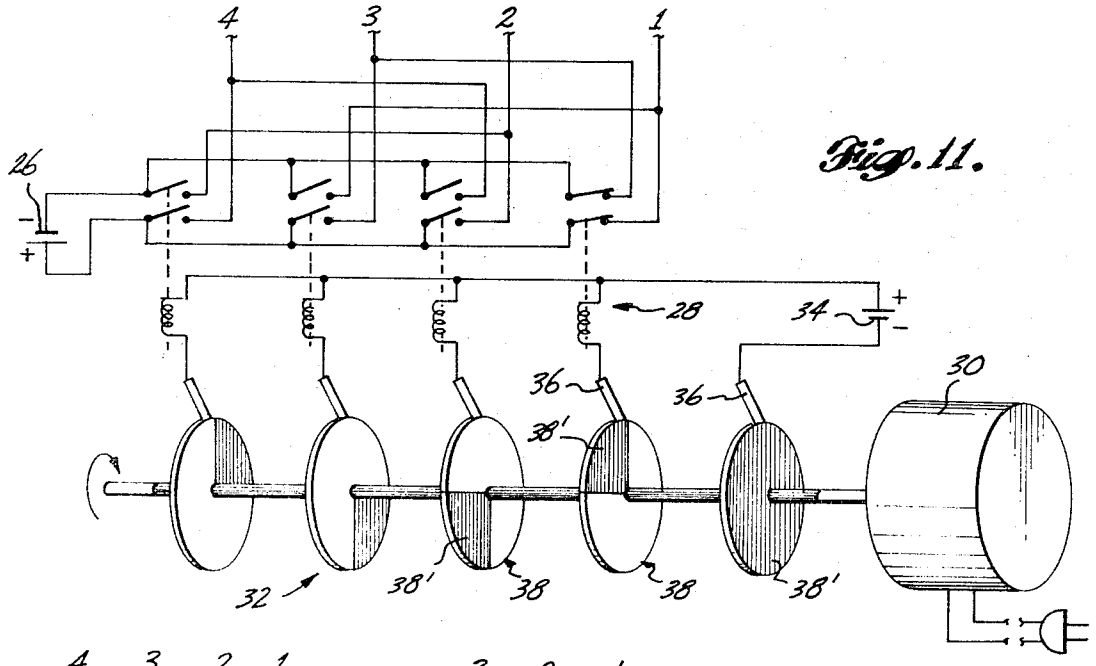


Fig. 11.

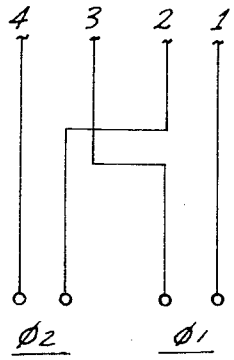


Fig. 13.

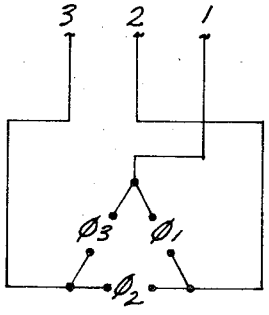


Fig. 14.

ELECTROPERISTATIC ION PUMP

Filed April 10, 1972 9 Sheets-Sheet 6

Fig. 16.

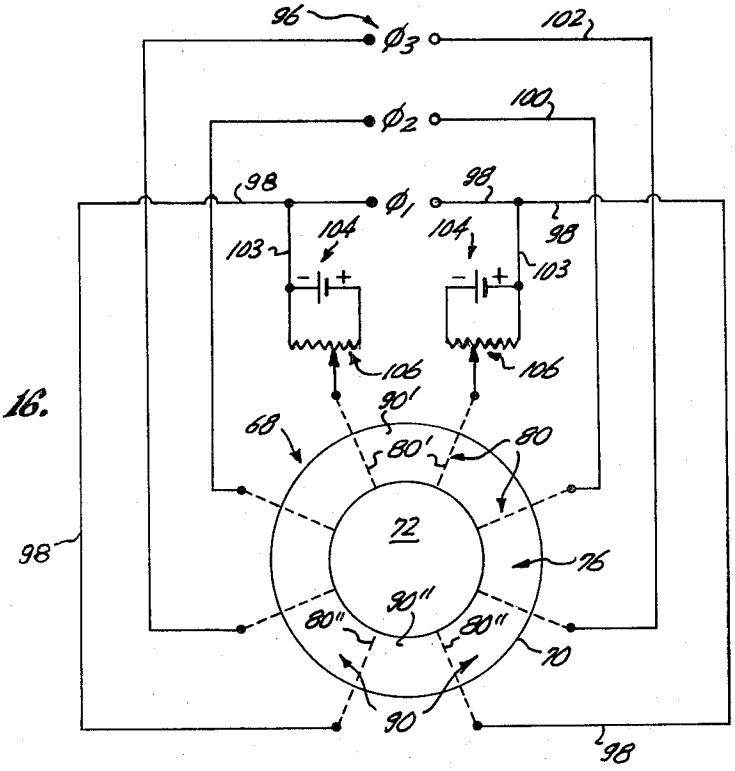
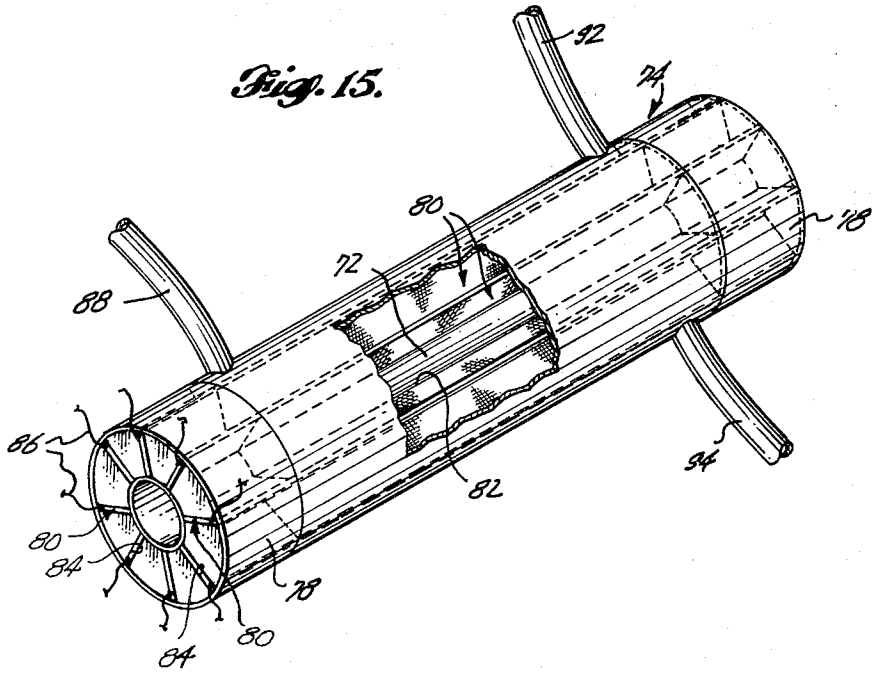


Fig. 15.



ELECTROPERISTATIC ION PUMP

Filed April 10, 1972 9 Sheets-Sheet 7

Fig. 17.

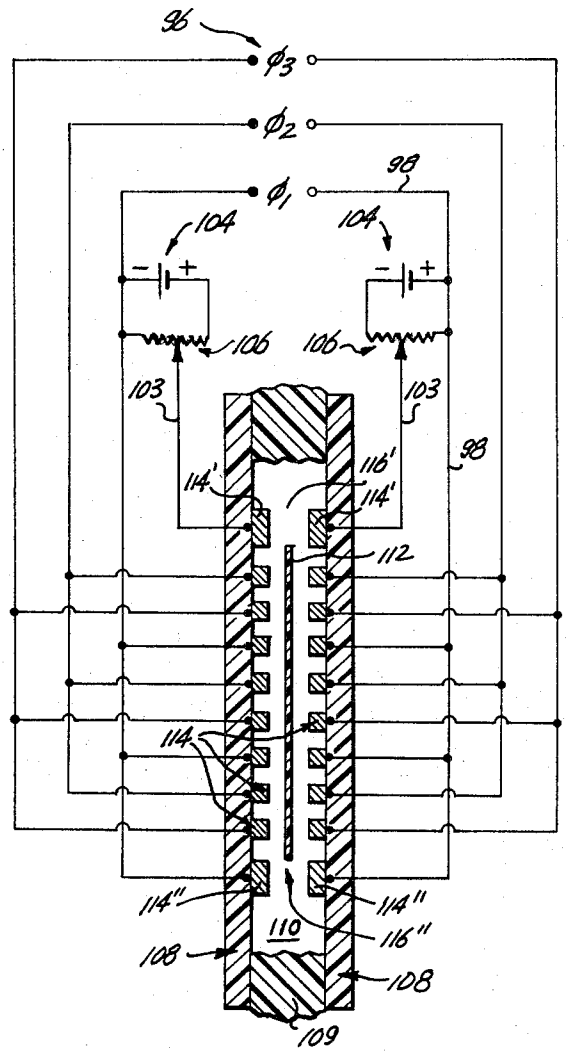
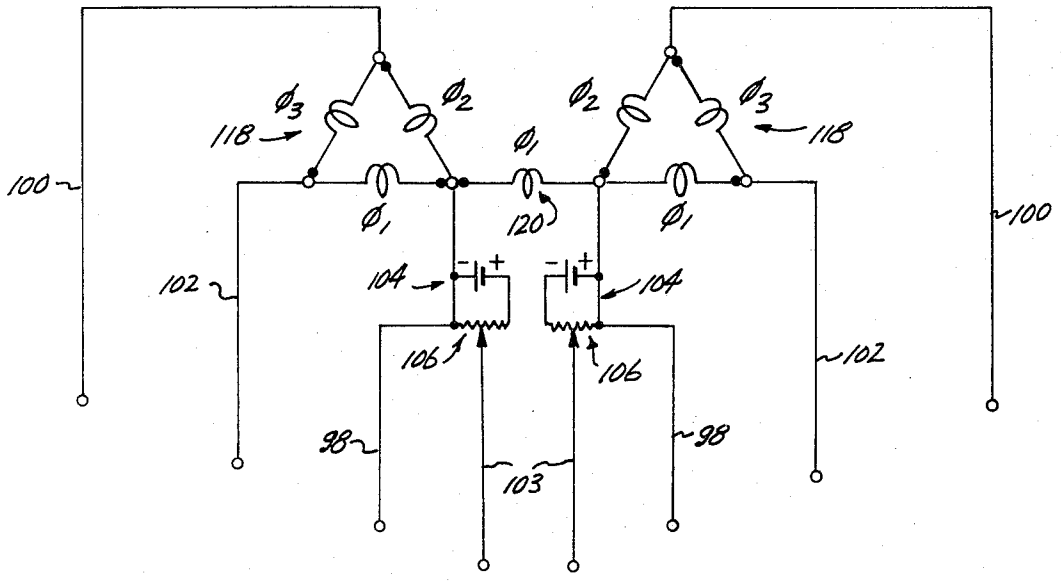


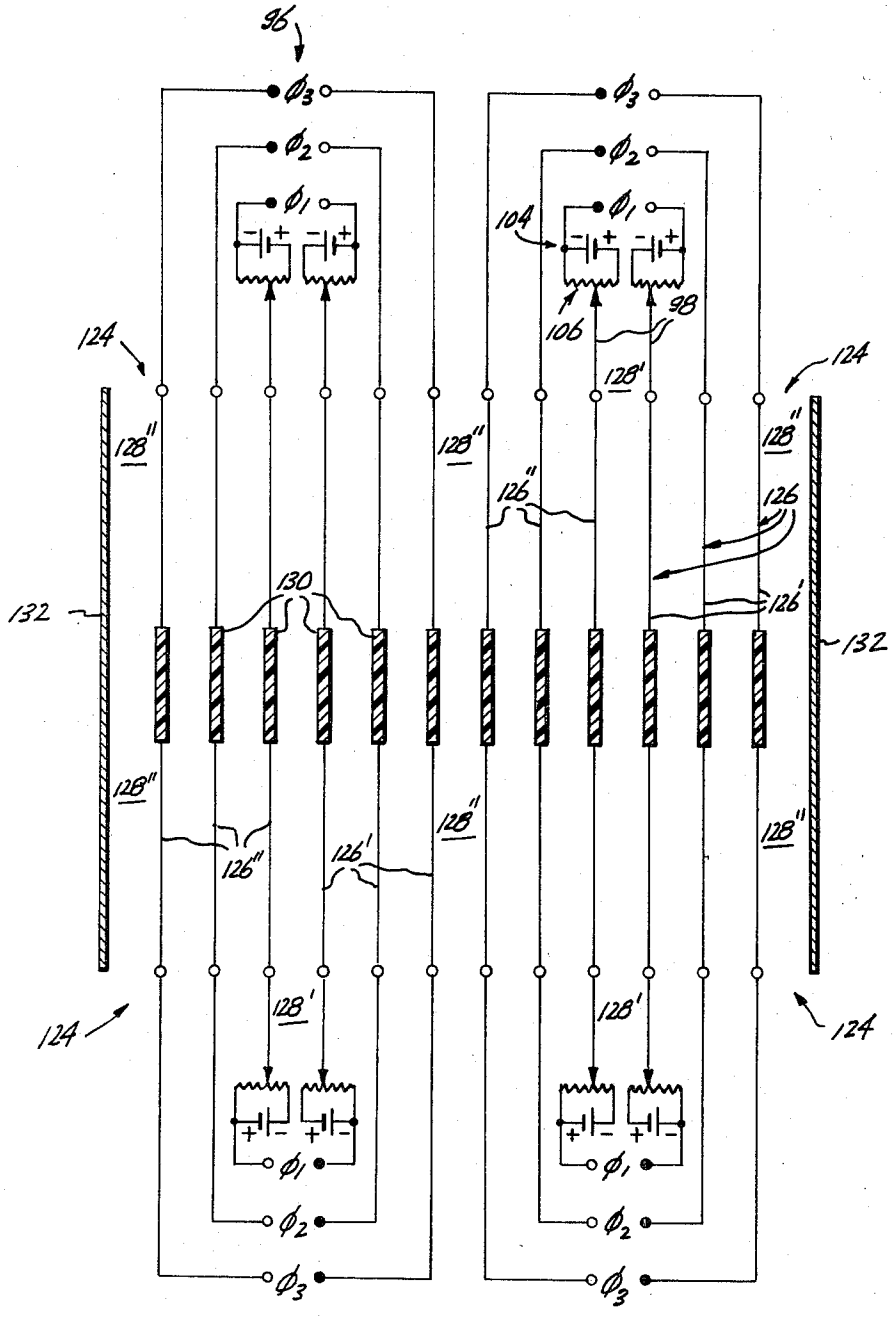
Fig. 18.



ELECTROPERISTATIC ION PUMP

Filed April 10, 1972 9 Sheets-Sheet 8

Fig. 19.



ELECTROPERISTATIC ION PUMP

Filed April 10, 1972 9 Sheets-Sheet 9

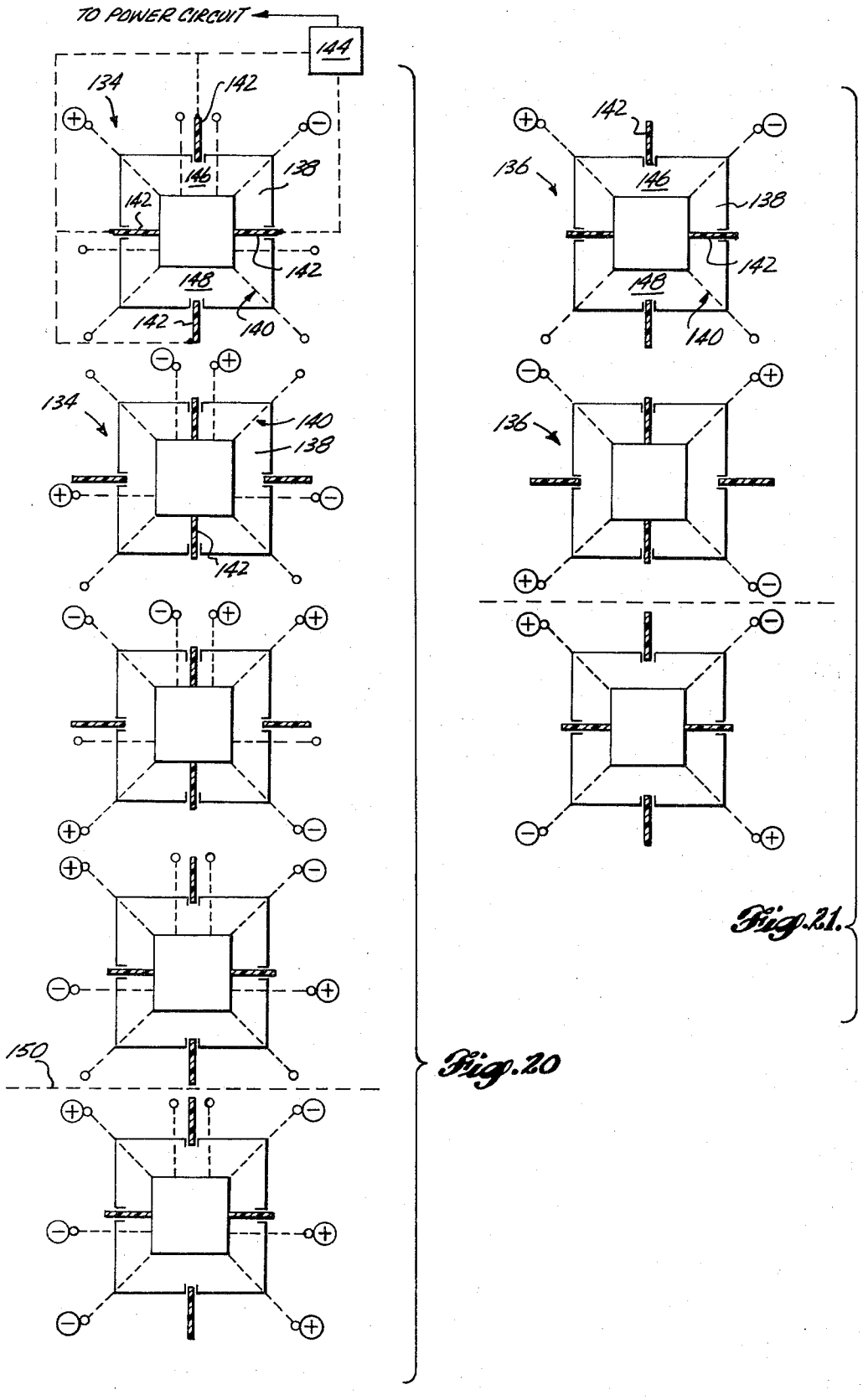


Fig. 21.

Fig. 20

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3,846,274

ELECTROPERISTALTIC ION PUMP

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Int. Cl. B01k 5/00; C02b 1/82
U.S. Cl. 204—299 27 Claims

ABSTRACT OF THE DISCLOSURE

A means and technique are disclosed for achieving an electroperistaltic pumping effect among electrically charged matter mobilely suspended in a fluid medium, such as among the ions of an ionized electrolyte such as sea water. In practicing the method, a series of successively timed electrical pulses are applied across a series of successively interconnected flow regions which are formed between a series of spaced electrodes in a fluid flow cell. The pulses are applied between the individual electrodes of pairs of electrodes whose respective electrodes lie on opposite sides of successive regions in one transverse direction of the series of electrodes. Between the successive time phases of the pulses, the polarity of the relatively forward electrode of the pair of electrodes on the opposite sides of one flow region, in the aforesaid transverse direction of the series of electrodes, is neutralized, while a polarity is established in the relatively forward electrode of the pair of electrodes on the opposite side of the next successive flow region, which is identical to the polarity which characterized the aforesaid relatively forward electrode of the aforesaid one flow region, during the last preceding pulse. Due to the characteristic differences in mobility among the ions of different chemical nature in an ionized electrolyte, the pump can be employed not only as a means of deionizing the electrolyte, but also as a means of segregating two or more ion fractions of different nature.

FIELD OF THE INVENTION

This invention relates to a means and technique for achieving a pumping effect among electrically charged matter mobilely suspended in a fluid medium, and in particular to a means and technique of this nature wherein the pumping effect is achieved by an electrolytic phenonomenon which can be best described as "electroperistaltic." The pumping effect in turn produces a change in the distribution of the electrically charged matter in the fluid medium, so that the invention is especially applicable to the deionization of ionized electrolytic solutions such as sea water, either for the purpose of achieving a simple demineralization of the solution, or for the purpose of achieving a fractionalization of the various chemical constituents of the solute or solvent, or for the purpose of doing both. However, the invention is also useful for pumping ions in an electrolyte other than water, and for pumping charged particulate or colloidal matter dispersed or suspended in a nonelectrolytic fluid other than water, or in which water, if present, is not the continuous medium.

The invention is equally applicable to the pumping of charged ions or particles in either a gaseous medium or a liquid medium.

BACKGROUND OF THE INVENTION INCLUDING CERTAIN OBJECTS THEREOF

One object of the invention is to provide an electrolytic apparatus and an electrolytic technique of this nature which are free from the disadvantages that accompanied previous attempts to use the electrodialytic process in this connection. In particular, the present invention employs no membranes or other filtration means in segregating the

minerals or other charged particles from the fluid medium. Thus, it is capable of far higher throughput rates than those of the electrodialtytic process, and can be installed and operated at considerably lower expense, since it does not require the large volumetric dimensions of the electrodialytic cell. Also, in contrast to the electrodialytic process, any acidic and alkaline factors produced in accordance with the present invention, are intimately mixed, so that the likelihood of precipitate formation is greatly reduced. Where precipitates do form, moreover, they can be readily chemically or electrochemically purged from the system, because of the absence of membranes therein. Going further, the fluid flow cell employed in the present invention also has a far lower internal resistance than the electrodialytic cell, since the membranes in the latter cell constitute areas of high electrical resistivity, and in addition, the use of a central "deionized" compartment in the latter cell requires increasingly higher voltages in the later stages of the operation to drive the ions therethrough.

Another object is to provide a membraneless apparatus and technique of this nature which are free from the disadvantages of the electrode adsorption techniques which have been previously used in this connection. In particular, while the present invention employs one or more series of electrodes in the apparatus thereof, the electrodes may be more widely spaced, and may have a far lower adsorption capacity and/or adsorption area than those required in the prior art adsorption techniques. Also the electrodes in the present apparatus need not be constructed of expensive and delicate, highly differentiated anion-selective and cation-selective materials; and in fact, may be constructed from more ordinary materials such as metal screen, carbon paper, felt, and the like.

Still another object of the invention is to provide a membraneless apparatus and technique of this nature using an electrolytic cell which can be operated over a wide range of voltages, including at voltages below the decomposition potential of the electrolyte solution, so as to avoid the evolution of gases, and/or the incidence of lasting acidic or alkaline conditions therein. A further object is to provide an apparatus and technique of this nature wherein the cell can be operated at voltages above the direct current decomposition potential of the solution, if desired, but nevertheless in such a manner as to minimize gas evolution, and to assure that those acidic and/or alkaline factors which are produced, are so intimately mixed as to minimize the production of acidic or alkaline solution in the cell. Other objects include the provision of an apparatus and technique of this nature wherein the electrolytic cell can be programmed to achieve certain additional effects when equipped with anion-selective and cation-selective electrodes, or specific-ion-selective electrodes; or when equipped with perm-selective membranes, or limited-permeability non-selective membranes. Still further objects will become apparent from the description of the invention which follows hereafter.

SUMMARY OF THE INVENTION

These objects and advantages are realized by an electrolytic apparatus and technique of my invention wherein the ions of the electrolyte undergo a pumping effect which may be called "electroperistaltic" in nature, by analogy to the pumping effect in biological organs such as the esophagus which generate the effect by means of successively applied, successively spatially offset muscular pulses. According to the invention, a series of successively timed electrical pulses are applied across a series of successively interconnected flow regions formed between a series of spaced electrodes in a fluid flow cell. The pulses are applied between the individual electrodes of pairs of electrodes whose respective electrodes lie on opposite sides

of successive regions in one transverse direction of the series of electrodes. Between the successive time phases of the pulses, the polarity of the relatively forward electrode of the pair of electrodes on the opposite sides of one flow region, in the aforesaid transverse direction of the series of electrodes, is neutralized, while a polarity is established in the relatively forward electrode of the pair of electrodes on the opposite sides of the next successive flow region, which is identical to the polarity which characterized the aforesaid relatively forward electrode of the aforesaid one flow region, during the last preceding pulse. In this way, the ions are driven or advanced in one crosswise direction of the cell, by a series of short "pushes," each of which is brief in duration but advances the ions a little bit further than the one before. And since ions of different chemical nature react differently to the "pushes," i.e., have different mobilities, the process can be employed as a means of discriminating between the ions of the solvent and the ions of the solute, as for example, in the case of demineralizing sea water, or as a means of discriminating among the ions within either the solvent or the solute, as for example, where it is desired to segregate a specific ion fraction or fractions within one or the other.

The electroperistaltic pumping effect described above may be achieved in one of a number of ways. For example, the pulses may be applied across successively adjacent pairs of successively adjacent flow regions while each of the aforesaid pairs of flow regions has a neutralized electrode therebetween. Or the pulses may have overlapping time phases and may be applied across successively adjacent flow regions during their overlapping time phases so as to neutralize the electrode therebetween. In each instance, the series of pulses is normally applied on a repetitive basis, and in the case of the first mentioned mode of operation above, the relative polarity of the relatively forward and trailing electrodes on the opposite sides of each of the aforesaid pairs of successively adjacent flow regions is reversed each time that the series of pulses is repeated. On the other hand, in the case of the second-mentioned mode of operation employing overlapping time phases, the relative polarity of the relatively adjacent electrodes on the opposite sides of each flow region is maintained each time that the series of pulses is repeated.

One also has the option of using a common electrical source or separate electrical sources in powering the series of pulses. When using a common source, the second-mentioned overlapping time phase embodiment may be powered by a DC current source which is accompanied by suitable switch means for generating square wave pulses. Or, isolated three-phase rectified AC may be used; or three-phase delta may be used unrectified. When the first-mentioned embodiment is powered by a single current source, the source may be a DC source which is accompanied by suitable commutator-actuated relay means to pulse it as needed; or a two phase sine wave AC source. Neither the two phases or the three phase AC system will operate properly as an unrectified square wave source, since no dwell time at neutral voltage exists in a normal + — square wave current.

Typically the cell has an inlet therein which is disposed adjacent one end of the flow regions. At least one outlet is disposed adjacent the other end of the flow regions, on the relatively remote side of the series of flow regions, in the aforesaid transverse direction of the series of electrodes. In addition, where the cell is employed as a deionization device for electrolytic solutions such as sea water, another outlet may be disposed adjacent the other end of the flow regions, on the relatively near side of the series of flow regions, considering this direction.

The electrodes may or may not be fluid permeable. For example, in certain embodiments of the invention, the electrodes are formed by conductive metal wire screens, which may or may not be spaced apart by porous electrically insulative spacer elements. In other embodiments, the electrodes are formed by solid, elongated conductive metal strips which are interconnected to a common insulative backing plate. The backing plate is one of a pair of walls which are disposed in spaced parallel array with one another so that the series of conductive metal strips on the plate, faces the other of the walls. This latter wall may also carry a series of electrode strips, as for example, where the walls are comprised of a pair of printed circuit boards arranged in spaced parallel, face to face array with one another.

The electrodes may or may not be ion-absorptive. Carbon electrodes such as those as described in U.S. Pat. 3,244,612, have the advantage that they can hold a considerable number of ions at saturation, whereas flat metal surfaces can hold only a thin "sheath" of ions, based solely on electrostatic attraction, and perhaps of the order of 100 Angstroms in thickness. Yet both function in accord with the invention, since upon reversal of the polarity, the ions adsorbed on carbon electrodes are released to the same degree as those held on purely ion-adsorptive electrodes.

The invention also contemplates the use of the various specialized anion-selective and cation-selective electrodes known in the art; or the use of electrodes which are especially selective toward only a single species of ion, such as calcium or lithium, and which will thus aid in selectively "pumping" that particular ion. Similarly, various ion-selective, semipermeable membranes, or nonselective membranes of limited permeability or porosity, may be used in various combinations, located for example, between the various electrodes to suit certain performance criteria required for a particular cell or cells.

Where twin boards are used, one of the boards may have a grid of cation responsive carbon electrodes thereon, while the other may have a grid of anion responsive carbon electrodes thereon, so as to achieve the benefit of both electrode absorption characteristics.

Preferably, the fluid flow cell has two spaced series of electrodes disposed therein, each of which forms a series of the recited flow regions; and further comprises means for applying an electrical bias to the space between the two series of electrodes, adjacent the relatively near sides of the series of flow regions in the aforesaid transverse directions of the series of electrodes, so that ions of differing polarities travel through differing series of flow regions. The respective ions are then neutralized when they reach the relatively remote sides of the series of flow regions in the aforesaid transverse directions of the series of electrodes.

The electrical biasing means may include a source of DC potential connected to the relatively first electrode of each series of electrodes, adjacent the relatively near sides of the series of flow regions in the aforesaid directions of the series of electrodes. The biasing means may also include means for varying the DC bias on each of the aforesaid first electrodes. Preferably, the first electrodes are connected to the pulse applying means through the sources of DC potential. Also, the pulse applying means preferably includes a source of isolated three phase sine wave AC potential, and the first electrodes are connected to the first phase thereof.

The two series of electrodes may be in unidirected, juxtaposed array with one another, with the two series of flow regions separated by a liquid impermeable baffle which terminates adjacent the relatively remote sides thereof in the aforesaid directions of the series of electrodes, so that the ions of one series of regions are neutralized by those of the other. Alternatively, the two series of electrodes may be in oppositely directed, tandem array with one another, with the two series of flow regions interconnected to a pair of similar cells adjacent the relatively remote sides thereof in the aforesaid directions of the series of electrodes, so that the ions of one cell are neutralized by those of the other. In the latter instance, moreover, one of the additional cells may be disposed back to back with the recited cell, and the relatively last

electrodes adjacent the relatively remote sides of the series of flow regions in the aforesaid directions of the series of electrodes of the respective one additional cell and the recited cell, may have an unpowered electrode thereopposite to reduce the electrical resistance therebetween. Preferably, the unpowered electrode is rotatable.

In another feature of the invention, the fluid flow cell has mechanical valves interposed between the pairs of electrodes in each series of electrodes, and further comprises means for opening and closing the valves in relation to the pulses applied between the respective pairs of electrodes.

BRIEF DESCRIPTION OF THE DRAWINGS

These features will be better understood by reference to the accompanying drawings which illustrate certain preferred embodiments of the invention.

In the drawings, FIG. 1 is a perspective view of a fluid flow cell for practicing the inventive technique;

FIG. 2 is an exploded perspective view of the series of electrodes embodied in the cell;

FIG. 3 is a plan view of the series of electrodes showing the arrangement in which they are electrically interconnected when the technique is practiced under the first mentioned mode of operation above;

FIG. 4 is a graphical representation of the sequence of electrical pulses applied to the arrangement in FIG. 3, using separate power sources for the respective stages of the sequence;

FIG. 5 is a schematic representation of the ion flow when the pulses are applied across the arrangement in the sequence of FIG. 4;

FIG. 6 is a graphical representation of the sequence of electrical pulses, when the electrodes are interconnected to function under the second mentioned mode of operation, and separate power sources are used;

FIG. 7 is a schematic representation of the ion flow in the modified arrangement when the pulses are applied in the manner of FIG. 6;

FIG. 8 is a perspective view of a fluid flow cell which is constructed from a pair of matching printed circuit boards;

FIG. 9 is a schematic cross-sectional view of the cell in FIG. 8 showing the modified electrical arrangement as it is employed therein;

FIG. 10 is a plan view of the printed circuit pattern on the face of one of the printed circuit boards in the cell of FIG. 8;

FIG. 11 is a schematic representation of a commutator-actuated relay system whereby the arrangement of FIG. 3 can be operated from a single DC power source;

FIG. 12 is a schematic representation of a multiple-cam cycling timer by which the arrangement of FIG. 9 can be operated from such a source;

FIG. 13 is a schematic representation of a powering system whereby the arrangement of FIG. 3 can be operated from a two-phase sine wave AC current source;

FIG. 14 is a similar representation whereby the arrangement of FIG. 9 can be operated from a three-phase sine wave AC current source;

FIG. 15 is a part cut-away perspective view of a fluid flow cell for practicing the dual flow concept of the invention;

FIG. 16 is a schematic cross-sectional view of the dual flow cell, showing the arrangement therein whereby the two series of electrodes are electrically interconnected and powered by a three-phase sine wave AC current source, to operate under the second mentioned mode of operation above;

FIG. 17 is a cross-sectional view of another dual flow cell using the same electrical arrangement and power source as that in FIG. 15;

FIG. 18 is a schematic representation of an alternate power circuit for use with the embodiments of FIGS. 15, 16 and 17;

FIG. 19 is a schematic representation of the electrically interconnected electrodes and power sources of a stacked multi-cell dual flow arrangement;

FIG. 20 is a schematic representation of the stage by stage operation of a dual flow cell which is equipped with mechanical valving between electrodes; and

FIG. 21 is a schematic representation of a similar type of flow cell having a lesser number of electrodes therein.

DESCRIPTION OF THE PREFERRED EMBODIMENTS

Referring first to FIGS. 1–3, it will be seen that the fluid flow cell **2** is comprised of a liquid-tight, corrosion-resistant casing **4** which is electrically insulative and equipped with a series of thin, closely spaced, parallel platinum wire-mesh electrode sceens **6** that are upstanding therein. The electrode screens **6** are retained in upright position by means of electrically insulative spacers **8** of glass or non-woven synthetic fiber cloth therebetween which also operate to maintain a uniform spacing between the screens. Both the screens **6** and the spacers **8** are highly porous in nature, such as approximately 10–200 mesh to the inch; and the screens have tabs **10** upstanding along their upper edges which project through the shell of the casing **4** to enable the screens to be interconnected in certain groupings by electrical leads **12–15** which emanate from a pair of DC current sources (not shown). Referring for example to FIG. 3, it will be seen that the first lead **12** on the left-hand side of the cell interconnects the uppermost electrode with every fourth electrode thereafter in the series; whereas the next lead **13** to the right interconnects the second electrode from the top with every fourth electrode after it; the next or third lead **14** interconnects the third electrode with every fourth electrode after it; and finally, the right-hand lead **15** connects the fourth electrode with every fourth electrode after it in the series. Thus, the electrodes are grouped together into successive sub-cells **16** of four electrodes each; and under the first mode of operation mentioned above, the first and third leads **12** and **14** are connected to the terminals of one current source, while the second and fourth leads **13** and **15** are connected to the terminals of the other source. Then, in use, the two current sources are switched alternately on and off at the same time so that the source interconnecting electrode groups one versus three is pulsed in alternate sequence to the source interconnecting electrode groups two versus four, and the current takes on a square wave form in which the polarity of every alternate pair of pulses is reversed as the series of pulses is repeated. See FIG. 4 wherein the pulse periods are divided into coordinate cycles lettered A through D, A′ through D′, etc.

The effect of the pulse sequence is illustrated in FIG. 5. The electrodes **6** are vertically arranged and numbered across the top of the Figure in accord with the groupings in which they are interconnected by the leads **12–15** in FIG. 3. The pulse periods of the current sources are indicated along the left-hand margin, with the relative polarities of the electrodes in each period being indicated in conventional manner, and a zero stage being included to indicate the condition of the cell before power is applied. Anions and cations in the fluid are indicated as $\ominus$ and $\oplus$, respectively, and for ease of illustration, only one anion and one cation is indicated in the flow region **18** between each pair of electrodes.

When an electrolytic solution such as salt water is introduced through the cell inlet **20** adjacent the left-hand end of the flow regions **18** in FIGS. 1 and 3 (or the top of the regions in FIG. 5), and at a central location within the series of electrodes, all of the ions in the solution, including the hydrogen and hydroxyl ions, are subject to the electrical forces of the pulses. Moreover, the spacing between electrodes, the interelectrode voltage, and the frequency of the pulses are all adapted so that during each pulse period following the initial pulse period A, the ions

collected on one face of an electrode are driven past that electrode into the next successive flow region where they are given time to collect on the corresponding face of the next successive electrode. For example, starting with the period A in FIG. 5, it will be seen that the cation in the flow region between electrodes 1 and 2 is driven past electrode 2 to the left-hand face of electrode 3 where it collects with the cation in the flow region between electrodes 2 and 3. Simultaneously, the cations in the flow regions between electrodes 3 and 4 and electrodes 4 and 1', collect on the right-hand face of electrode 3. However, during pulse period B, the polarity of electrode 3 is neutralized, while a polarity is established in electrode 4 which is identical to that which electrode 3 had during pulse period A. As a consequence, the cations on the left-hand face of electrode 3 travel to the same face of electrode 4, together with the cations which were on the right-hand of electrode 3. In pulse period C, moreover, the polarity of electrode 4 is now neutralized, while electrode 1' takes on the polarity which electrode 4 had during the last preceding pulse period B. Therefore, the ions continue to travel in the right-hand direction across the series of electrodes, and eventually into the next sub-cell during period D, so that the process can repeat itself from that sup-cell to the next as the series of pulses is recycled. Simultaneously, the anions in the flow regions between electrodes 3 and 4 and electrodes 4 and 1, collect on the left-hand face of electrode 1' in pulse period A, then travel to the left-hand face of electrode 2' in period B, together with the anions which collected on the right-hand face of electrode 1' from the flow regions between electrodes 1' and 2' and electrodes 2' and 3', during period A. In progressive fashion, therefore, the left-hand flow regions in FIG. 5 become depleted of ions while the right-hand regions become enriched in ions.

Turning again to FIG. 3, it will be seen that a relatively ion enriched effluent can be withdrawn through an outlet 22 at the upper or remote side of the series of electrodes, while an ion depleted effluent is available at an outlet 24 on the lower or near side of the Figure. Or alternately, two or more ion fractions can be withdrawn at different points along the remote side of the series of electrodes, based on the difference in their mobility. In very dilute aqueous solutions, for example, mobilities range from 4×10^{-4} for the lithium ion, to 36×10^{-4} for the hydrogen ion, in units of centimeters per second per volt per centimeter. Thus, two or more ion fractions can be readily separated from one another in such a medium, as well as in many other media.

The parameters of electrode thickness, interelectrode voltage, and pulse frequency, are usually empirically obtained. In practice so far, I have found that when working with 3% aqueous solutions of sodium chloride containing additional salts in smaller amounts, electrode spacings in the range of .010–.020" and very low pulse frequencies, produce the best results. But in any case, considering ion mobilities in general, one may use an order of magnitude of 1×10^{-3} centimeters per second per volt per centimeter to make a rough estimate. For example, using 80 mesh platinum metal screens .010" thick, with .010" thick glass cloth spacers between them, the distance between two adjacent left-hand electrode faces is .02" or .05 centimeter. If the pulse voltage is 1.5 volts, the interelectrode potential gradient averages 1.5 v./.075 centimeter, or 20 volts per centimeter. The ion velocity then averages 20×10^{-3} centimeters per second, and the estimated quarter cycle time period for ion travel from one left-hand face to the next is .05 centimeter/20×10^{-3} centimeters per second, or 2.5 seconds. On the other hand, an electrode width and spacing of .001" produces an interelectrode potential gradient of 200 volts per centimeter and an estimated frequency of ten cycles per second. Other types of electrodes such as engine-ruled metal films made by the methods of manufacturing diffraction gratings, permit even smaller electrode widths and spacings and therefore the use of much higher frequencies.

FIG. 11 illustrates a powering system whereby the 4-electrode sub-cell arrangement of FIG. 3 can be pulsed from a single DC current source. The current source takes the form of a battery 26, and the battery 26 is pulsed by four sealed mercury relays 28 which are actuated in turn by a motor 30 driven, four gang commutator 32. The relays 28 are powered from a second battery 34, and include brushes 36 which make contact with appropriately angularly positioned conductive metal portions 38' of the commutator discs 38.

Alternatively, the arrangement of FIG. 3 may be powered by a two-phase sine wave AC source in the manner of FIG. 13.

Referring now to FIGS. 6–10, it will be seen that the embodiment illustrated therein comprises a pair of matching printed circuit boards 40 which are disposed in spaced, parallel face to face array with one another. Each board 40 has a series of spaced parallel conductive metal strips 42 relatively raised on the inside face 40' thereof, in spaced registry with the series of strips raised on the face 40' of the opposing board. Together the boards 40 form a fluid flow cell with interconnecting flow regions between the pairs of opposing electrodes. The cell has an inlet 44 and a pair of outlets 46 in the manner of FIG. 1, and the periphery of the cell is closed and sealed by an annular gasket 48 of silicone resin caulking material, or a formed rectangular ring of neoprene. The boards 40 themselves are fabricated by the usual commercial techniques from copper-clad sheets of fiberglass resin material patterned from a photographic negative template which is a photographic reduction 41 of a hand-drawn master, as seen in FIG. 10. The copper electrodes may be plated with gold, nickel, tin, rhodium, palladium, and other such corrosion resistant metals.

The electrodes 42 on each board are interconnected in sub-cells 50 of three electrodes each. Referring to FIG. 9, it will be seen that the first, fourth, seventh, tenth, etc., electrodes are interconnected by a common lead 52 to one board, and similarly, the same electrodes are interconnected by a common lead 54 to the other board. Also, the second, fifth, eighth, etc., electrodes on each board are interconnected with one another in a similar manner by leads 56 and 58; and the third, sixth, ninth, etc., electrodes are interconnected on each board by leads 60 and 62. As seen in FIG. 6, the pulses in this instance have overlapping time phases and are applied across successively adjacent flow regions during their overlapping time phases, so as to neutralize the electrodes therebetween. The pulses are applied either by means of a DC power source and a standard motor 63 driven multiple-cam cycling timer 64 working therewith, as in FIG. 12, or by isolated, rectified three-phase sine wave sources; or the three phase delta circuit of FIG. 14 may be used without rectification.

In FIG. 7, the ion flow is indicated more simply when the pulses are applied by means of a pair of DC sources, switched on and off in appropriate sequence, for example, by a standard continuous interval timer, or at higher frequencies, i.e., one cycle to forty cycles per second, by fast-response, sealed mercury relays actuated by the switching of a two phase/three phase commutator which is driven by a variable speed motor. In pulse period A, wherein a pulse is applied between electrodes 1 and 2 only, the cations collect at electrode 2; but in overlapping pulse period B, when a pulse is also supplied between electrodes 2 and 3, the result is to neutralize the intermediate electrode 2 so that the cations travel past the electrode to the near face of electrode 3. In pulse period C, the pulse between electrodes 1 and 2 is discontinued so that the anions are, so to speak, "drawn up behind" the cations in their course of travel. In overlapping pulse period D, electrode 3 is neutralized so that the cations advance into the first electrode of the next sub-cell 50'

(FIG. 9) in consequence of the pulse between electrode **3** and electrode **1'**. And by the end of period F and a full cycle of pulses, both cations and anions are advanced into sub-cell **50'** so that the series of pulses can be recycled to advance them into sub-cells **50''** and so on.

Typically, the electrodes **42** in FIGS. 8–10 are .020'' wide and spaced .020'' apart, although electrode widths and spacings down to about .002'' are possible. In fact, for a cell to operate below the decomposition potential of most saline water solutions, the electrodes are desirably as narrow as possible, and also as closely spaced as possible so that the ion sheaves collected on the electrodes with each pulse are extremely thin, while a high potential gradient in volts per centimeter is available from a low interelectrode voltage pulse.

The two boards **40** in the cell of FIG. 8 are also typically spaced apart at about .020''. A group of metal washers (not shown) may be employed to maintain the clearance. The electrodes **42** need not register with one another from one board to the next. Also, the two boards may be powered at different voltages, frequencies, and/or phase orders to provide higher discrimination in ion mobilities. For example, one board may be operated in phase order 1, 2, 3 at one voltage and one frequency, while the other is operated in phase order 3, 2, 1 at a different voltage and frequency to provide a higher degree of discrimination between ions of different mobilities. Several effluent outlets at different positions along the exit side of the cell can serve to collect the different ion fractions.

A printed circuit board cell also has the advantage that once a photographic master template is made, boards can be inexpensively produced with great precision and uniformity of electrode width and spacing. Moreover, the uniform electrode spacing initially provided in the cell is relatively unaffected by cell operating conditions, such as gas evolution, heat convection or the like. Furthermore, due to the rigid unitized construction of the cell, change and replacement of electrodes is readily possible.

Gas evolution is relieved by a gas vent **66**, as in FIG. 1.

Referring next to FIGS. 15–21, it will be seen that the dual flow cell **68** of FIGS. 15 and 16 comprises a liquid-tight, corrosion resistant shell or casing **70** of tubular configuration, which is electrically insulative and enclosed about the neck **72** of a liquid impermeable, "dumbbell"-shaped inner core **74** which is also corrosion resistant and electrically insulative. The core **74** is concentrically disposed within the casing **70**, and is of considerably smaller diameter at the neck **72** so as to form an elongated, annularly cross-sectioned flow chamber **76** within the casing, the ends of which are closed by the cylindrical polls **78** of the core so as to render the chamber **76** liquid tight. The chamber is subdivided about the circumferential extent thereof, by a series of thin platinum wire-mesh electrode screens **80** which are installed in symmetrically radially oriented, longitudinally extending slots **82** in the neck **72** of the core, and sized to project radially outwardly therefrom across the full radial extent of the chamber **76**. The electrodes **80** are also received in a corresponding number of radially oriented, longitudinally mutually aligned slots **84** in the polls **78** of the core, where they extend to points adjacent the ends of the polls so as to be accessible for the attachment of electrical leads **86** thereto. To preserve the liquid tight character of the cell, however, a silicone resin sealant is applied in and around the slots **84**, as well as around the junctures between the polls **78** and the casing **70**.

The chamber **76** is supplied with electrolytic solution by means of a feed tube **88** communicating with the top of the chamber **76** at the left-hand end thereof in FIG. 15; and the solution is withdrawn from the chamber, after flowing lengthwise thereof in the regions **90** between electrodes, by means of a pair of discharge tubes **92** and **94** communicating with the top and bottom of the chamber at the other end thereof.

Operatively speaking, the neck **72** of the core serves as a liquid impermeable baffle that is interposed between all but the first and fourth electrodes **80'** and **80''**, respectively, of the clockwise series of electrodes **80** on the right-hand side of the cell in FIG. 16, and all but the first and fourth electrodes **80'** and **80''**, respectively, of the counterclockwise series of electrodes **80** on the left-hand side of the cell. Moreover, the electrodes in the respective series are interconnected with a cource of isolated three-phase sine wave AC potential **96**, to be operated in a manner consistent with the principles of the embodiments in FIGS. 6–10, although in the present instance, the ion flow is relatively circumferential of the cell, and relatively through the bodies of the electrodes, as shall be explained.

Referring again to FIG. 16, it will be seen that the first and fourth eectrodes **80'** and **80''** of the respective series, are interconnected with one phase of the source by means of a pair of leads **98**; the second electrodes in the respective clockwise and counterclockwise directions of the series, are interconnected with the second phase of the source by leads **100**; and the third electrodes in these directions are interconnected with the third phase of the source by leads **102**. Following the principles of the embodiments of FIGS. 6–10, therefore, pulsing the electrodes in the respective series, causes the ions in the solution to flow in the respective clockwise and counterclockwise directions thereof; although as shall be explained, the ions separate and flow around different sides of the baffle **72**, depending on their polarity in the solution.

This follows from the fact that the first electrodes **80'** in the respective series, are interconnected to their respective leads **98**, through a pair of branch leads **103**, and a pair of independent sources **104** of DC bias, which in turn are equipped with variable potentiometers **106** that enable the bias to be varied. Together, the sources **104** apply a DC bias to that flow region **90'** which is disposed between the first electrodes **80'** adjacent the upper end of the baffle **72**, with the result that the cations are collected in pulses onto one of the electrodes **80'**, such as the right-hand electrode, and are pumped clockwise through the right-hand series of electrodes; whereas the anions are collected in pulses on the left-hand electrode **80'** and are pumped in corresponding fashion but in counterclockwise direction through the left-hand series of electrodes. Ultimately, both sets of ions arrive in the flow region **90''** adjacent the lower end of the baffle **72**, where they are recombined by the unbiased sine wave potential across the electrodes **80''**.

Thus, when an electrolytic solution is introduced to the cell through the inlet **88**, it is gradually demineralized as it flows toward the opposite end of the chamber **76**, and at this latter end two effluents are discharged through the outlets **92** and **94**, one being an ion-enriched effluent leaving the cell through the outlet **94** interconnected with the region **90''**, and the other being an ion-depleted effluent leaving the cell through the outlet **92** connected with the region **90'**.

The most effective demineralization occurs when the sum of the two DC biases **104** is such that the potential between electrodes **80'** never reverses itself by an appreciable amount. For example, assuming that each phase has a peak of 1.0 volt, the DC bias at the left-hand potentiometer **106** would be plus 0.55 volt, and the DC bias at the right-hand potentiometer, minus 0.55 volt. Thus, the potential at the respective elecrode pairs **80** spaced clockwise and counerclockwise from electrodes **80'**, would range between plus 1.0 volt and minus 1.0 volt, while the potential across the electrode pair **80'** would range between plus 2.1 volt and plus 0.1 volt. Also, consistent with this, the ion flow in region **90'** would always be anions toward the left-hand electrode **80'**, and cations toward the right-hand electrode **80'**.

The separated biasing of the left-hand electrode **80'** versus the right-hand electrode **80'**, with respect to the

terminals of the first phase of source **96**, also permits simultaneous control of the separate bias between the left-hand electrode **80'** and the left-hand electrode **80''** and similarly that between the right-hand electrode **80'** and the right-hand electrode **80''**. This in turn permits the introduction of a higher average potential gradient in one series of electrodes versus the other, in order to compensate for the differing mobilities of the anions and the cations in the solution. In fact, such an operation may be necessary to assure that an equal charge of anions and cations reaches the region **90''** to be neutralized, since otherwise the buildup of an incipient charge in region **90''** would inhibit effective pulsing of the ions in the chamber.

The frequency of the three-phase sine wave potential **96**, as well as the voltages and the phase angles, depends on the cell configuration, the spacing between electrodes, the concentration of the electrolytic feed solution, the electrode thickness, and the electrode porosity.

In FIG. 16, only one pair of electrodes is subjected to the DC bias. Successive pairs of electrodes in the respective series of the same, may also be DC biased, progressively or regressively, according to the effect desired. Furthermore, differing phase angles, and phase voltages may be used if desired.

In FIG. 17, the cell comprises a pair of face to face printed circuit boards **108**, with an annular silicone resin gasket **109** therearound, forming a rectangularly cross-sectioned chamber **110** therebetween in the manner of the embodiment in FIGS. 8–10. However, a thin, liquid impermeable, electrically insulative baffle **112** is interposed between the two boards, and the baffle is sized to separate all but the top and bottom electrodes **114'** and **114''**, respectively, in the two series of electrodes **114** thereon. Moreover, the cell is electrically arranged and powered in the manner of the embodiment in FIGS. 15 and 16, in that the electrodes **114** are interconnected with an isolated three-phase sine wave AC power source **96**, and are arranged in three-electrode pulsing cycles, with a DC bias **104**, **106** across the top of first electrodes **114'** in the series of the same.

The operation of the cell is, therefore, also the same as that of FIGS. 15 and 16, in that the anions are pumped relatively downward from region **116'** adjacent the upper edge of the baffle **112**, through the left-hand side of the cell; while cations are pumped relatively downward through the right-hand side thereof, the two ions then recombining with one another in the region **116''** adjacent the lower edge of the baffle.

The operation of the cell differs from that of FIGS. 15 and 16, however, in that the ions are pumped across the faces of the electrodes **114**, whereas in the embodiment of FIGS. 15 and 16, they are pumped through the electrodes **80**.

Preferably, the electrodes **114** are superposed on a copper base and are a composite of rhodium plated over nickel, although other materials such as carbon and platinum may be used. The baffle **112** is a thin but rigid sheet of polyethylene.

Since the cell in FIG. 17 provides a wide area of separation between the ion depleted and ion enriched regions **116'** and **116''**, it is particularly useful in segregating or separating ions of differing mobilities, such as for example sodium and lithium. For this purpose, however, a second three-phase power supply (not shown) of different voltage and frequency, is superimposed on the three-phase power supply **96**, and operated in the opposite direction of phase progression, that is, from the enrichment zone **116''** to the depletion zone **116'**. For example, in one such arrangement, phase three of the second supply might be isolated and in series with phase one of the supply **96**; phase two of the second supply might also be isolated and in series with phase two of the supply **96**; and phase one of the second supply might be isolated and in series with phase three of the supply **96**. Operationally, ions of higher mobility would be pumped from the depletion zone **116'** to the enrichment zone **116''**, while ions of lower mobility would be continuously restrained in the direction of the depletion zone **116'**.

FIG. 18 shows an alternate power circuit for use with the embodiments of FIGS. 15–17. The coils **118** of the input phases, are typically secondary coils of appropriate power transformers, in order to achieve the proper degree of isolation. As indicated by the black dots, the delta on the right is, at each phase, of opposite polarity, or 180° out of phase with respect to the correspondingly numbered phase in the delta on the left.

Using the circuit of FIG. 18, the interelectrode voltages may be set at fixed values, rather than allowed to float as in FIGS. 16 and 17. Also due to their isolation, all three phases of the right-hand delta may be advanced or retarded with respect to the center phase **120**; and likewise, all three phases of the left-hand delta may be advanced or retarded with respect to the center phase **120**.

The invention is also applicable to cells using four electrodes per electrode cycle, powered by two phase/four phase current.

FIG. 19 shows an arrangement in which four cells **124** are stacked in juxtaposition to one another, and openly interconnected with one another, so that the anions of one cell are combined with the cations of another, and vice versa, in the process of demineralizing or differentiating the electrolyte flow. Each cell **124** comprises two sets **126'** and **126''** of electrodes **126** which are arranged in tandem and interconnected with a three phase source **96** which is subjected to DC bias **104**, **106** at the first phase, as in the earlier embodiments of FIGS. 15–17. Thus, in each cell, the ion flow is to the right or to the left with respect to the region **128'** between the relatively biased electrodes on leads **98**, and the latter region **128'** is ultimately relatively depleted of ions. Similarly, the ion flow in either direction, terminates in an enrichment zone **128''**; and because of the openly interconnected arrangement of the cells, the two cells at the top share a common enrichment zone **128''** therebetween, and the two cells at the bottom also share a common enrichment zone **128''** therebetween. In addition, the two right-hand, vertically related cells share a common enrichment zone **128''** at the right-hand ends thereof, whereas the two left-hand, vertically related cells share a common enrichment zone **128''** at the left-hand ends thereof. The electrodes of the adjacent upper and lower cells are separated by non-conductive partitions **130** between the respective upper and lower edges thereof, and these partitions prevent mixing of the electrolyte between adjacent electrodes in the vertical direction.

Again, the ion flow is crosswise the faces of the electrodes **126**, that is, into the plane of the drawing; and in short, the composite arrangement represents a continuous circle of back-to-back electrode cycles, accompanied by a pair of end electrodes **132** which are unpowered and operate to reduce the effective electrical resistance between the adjacent right- and left-hand powered electrodes **126'** and **126''** of the adjacent cells. Preferably, the unpowered end electrodes **132** take the form of circular discs which are mounted to be slowly rotated in relation to the arrangement, so as to continuously neutralize the separated cations and anions, on the continuously changing upper and lower surface portions of each disc.

A wide variety of liquid flow paths is possible in the arrangement of FIG. 19, including a partial counterflow arrangement wherein the depleted effluent from one cell **124**, is used to flush the enriched region **128''** of another cell, in order to reduce the concentration gradient across the electrodes **126** within the second mentioned cell.

In the simplest flow pattern, the feed solution is introduced through a pair of ports (not shown) coinciding with the depletion zones **128'** at the top of the arrangement; and the depleted solution leaves the arrangement at the opposite end, by way of a pair of ports (not shown) at the bottom of the arrangement, coinciding with the bottom depletion zones **128'**; while enriched effluent leaves

the bottom of the arrangement through a pair of ports (not shown) coinciding with the bottom enriched zones **128''**.

In lieu of the plurality of powder supplies **96**, a single power supply may be connected to all of the electrodes in correspondence with the sequence shown.

If a greater electrical spread of ion travel is desired, the number of electrodes per cycle may be greatly increased.

For simplicity of illustration the outer shell of the multi-cell arrangement is omitted.

In the arrangements of FIGS. 15–19, the success of each demineralization operation depends on developing an ion pumping rate which is greater than the ion back diffusion rate, so as to achieve the depletion and enrichment intended. This relationship in turn depends on the factors of providing an adequate number of electrodes, and an adequate power level, as well as the factors of electrode porosity, and cell geometry.

In the embodiment of FIGS. 20 and 21, the same pumping action is employed, but in separate discreet phase steps, and accompanied by positive mechanical valving action at each step, so as to assure that if the power progression is stopped at any step, ions cannot diffuse backwards in the cell to undo the ion pumping action which had been previously effected. Each cell **134** or **136** has an annular flow chamber **138** consistent with the arrangement of FIGS. 15 and 16, but plain square wave DC powering is used according to the polarity sequences shown in the four time phases of FIG. 20. All electrodes **140** without a designated polarity, are left open circuited and floating, rather than short circuited or grounded.

The mechanism used to open and close the radially reciprocable partition gates **142** in proper sequence, is seen at **144** in connection with the first phase. Ion depletion occurs at zone **146**, and ion enrichment at zone **148**, as in the previous embodiments.

In FIG. 20 a four step cycle is shown, with the time phase below the dashed line **150**, beginning the second cycle of the operation. The two-step cell of FIG. 21 does not have the disadvantage of requiring eight electrodes, and instead uses only four electrodes.

Rather than simultaneously pulsing the two series of electrodes of each cell in each of the arrangements in FIGS. 15–21, it may be desirable instead to generate a "zigzag" pattern of pulses, by effecting a phase-displacement between the pulses of one series of electrodes and the pulses of the other series. In FIG. 20, for example, the transition from step one to step two, may be achieved in two steps, that is, a left-hand step, then a right-hand step, or vice versa, and so on through the entire set of steps. Or in the power circuit of FIG. 18, the three phases in the left-hand delta may be advanced a number of degrees from the angle of the center phase **120**, while the three phases in the right-hand delta are retarded a number of degrees from the angle of the center phase, or vice versa.

The "zigzag" pattern may be useful in compensating for such factors as differing ion mobilities, differing ion adsorptive capacities of the various electrodes, and differing internal cell structure.

The invention also contemplates that the foregoing means and technique may be used to form an electrode monolayer over the hull of an ocean vessel, either for propulsion effect or for braking effect; or at a minimum, as a means of reducing hull skin-resistance so that less power is required to propel the vessel.

What is claimed is:

1. In a fluid flow cell defining a fluid flow passage having a series of spaced electrodes disposed therein, forming a series of successively interconnected flow regions therebetween, electrical pulse generating means connected to apply a series of successively timed electrical pulses across the series of flow regions, between the individual electrodes of pairs of electrodes whose respective electrodes lie on opposite sides of successive regions in one transverse direction of the series of electrodes, means operative between the successive time phases of the pulses, to neutralize the polarity of the relatively forward electrode of the pair of electrodes on the opposite sides of one flow region, in the aforesaid transverse direction of the series of electrodes, while establishing a polarity in the relatively forward electrode of the pair of electrodes on the opposite sides of the next successive flow region, which is identical to the polarity which characterized the aforesaid relatively forward electrode of the aforesaid one flow region, during the last preceding pulse, and means for passing the fluid into the passage and recovering a portion thereof, adjacent opposite ends of the flow regions.

2. The fluid flow cell according to claim **1** wherein the pulses are applied across successively adjacent pairs of successively adjacent flow regions while each of the aforesaid pairs of flow regions has a neutralized electrode therebetween.

3. The fluid flow cell according to claim **2** wherein the series of pulses is applied on a repetitive basis, and the relative polarity of the relatively forward and trailing electrodes on the opposite sides of each of the aforesaid pairs of successively adjacent flow regions is reversed each time that the series of pulses is repeated.

4. The fluid flow cell according to claim **1** wherein the pulses have overlapping time phases and are applied across successively adjacent flow regions during their overlapping time phases so as to neutralize the electrode therebetween.

5. The fluid flow cell according to claim **4** wherein the series of pulses is applied on a repetitive basis, and the relative polarity of the relatively adjacent electrodes on the opposite sides of each flow region is maintained each time that the series of pulses is repeated.

6. The fluid flow cell according to claim **1** wherein each pulse in the series is applied by a separate power source.

7. The fluid flow cell according to claim **1** wherein the cell has an outlet therein which is disposed adjacent one end of the flow regions and on the relatively remote side of the series of flow regions in the aforesaid transverse direction of the series of electrodes.

8. The fluid flow cell according to claim **7** wherein the cell has an inlet therein which is disposed adjacent the other end of the flow regions.

9. The fluid flow cell according to claim **8** wherein the cell has a second outlet therein which is disposed adjacent the one end of the flow regions and on the relatively near side of the series of flow regions in the aforesaid transverse direction of the series of electrodes.

10. The fluid flow cell according to claim **1** wherein the electrodes are formed by conductive metal wire screens.

11. The fluid flow cell according to claim **10** wherein the wire screens are spaced apart by porous, electrically insulative spacer elements.

12. The fluid flow cell according to claim **1** wherein the electrodes are formed by solid, elongated conductive metal strips.

13. The fluid flow cell according to claim **12** wherein the metal strips are connected to insulative backing plates on the opposite sides of the aforesaid series of the same.

14. The fluid flow cell according to claim **1** having two spaced series of electrodes disposed therein, each of which forms a series of the recited flow regions, and further comprising means for applying an electrical bias to the space between the two series of electrodes, adjacent the relatively near sides of the series of flow regions in the aforesaid transverse directions of the series of electrodes, so that ions of differing polarities travel through differing series of flow regions, and means for neutralizing the respective ions when they reach the relatively remote sides of the series of flow regions in the aforesaid transverse directions of the series of electrodes.

15. The fluid flow cell according to claim 14 wherein the electrical biasing means includes a source of DC potential connected to the relatively first electrode of each series of electrodes, adjacent the relatively near sides of the series of flow regions in the aforesaid directions of the series of electrodes.

16. The fluid flow cell according to claim 15 wherein the electrical biasing means also includes means for varying the DC bias on each of the aforesaid first electrodes.

17. The fluid flow cell according to claim 15 wherein the first electrodes are connected to the pulse applying means through the sources of DC potential.

18. The fluid flow cell according to claim 17 wherein the pulse applying means includes a source of isolated three-phase sine wave AC potential, and the first electrodes are connected to the first phase thereof.

19. The fluid flow cell according to claim 14 wherein the two series of electrodes are in unidirected, juxtaposed array with one another, and the series of flow regions are separated by a liquid impermeable baffle which terminates adjacent the relatively remote sides thereof in the aforesaid directions of the series of electrodes, so that the ions of one series of regions are neutralized by those of the other.

20. The fluid flow cell according to claim 14 wherein the two series of electrodes are in oppositely directed, tandem array with one another, and the series of flow regions are interconnected to a pair of similar cells adjacent the relatively remote sides thereof in the aforesaid directions of the series of electrodes, so that the ions of one cell are neutralized by those of the other.

21. The fluid flow cell according to claim 20 wherein one of the additional cells is disposed back to back with the recited cell, and the relatively last electrodes adjacent the relatively remote sides of the series of flow regions in the aforesaid directions of the series of electrodes of the respective one additional cell and the recited cell, have an unpowered electrode thereopposite to reduce the electrical resistance therebetween.

22. The fluid flow cell according to claim 21 wherein the unpowered electrode is rotatable.

23. The fluid flow cell according to claim 14 having mechanical valves interposed between the pairs of electrodes in each series of electrodes, and further comprising means for opening and closing the valves in relation to the pulses applied between the respective pairs of electrodes.

24. Electrochemical deionization apparatus for ionized fluids comprising a successively spaced series arrangement of immersion electrodes exceeding two in number and associated with means for positioning the same in such a fluid, a first pair of the electrodes defining a first interelectrode field region in the fluid, and a second pair of said electrodes defining a second interelectrode field region in the fluid, the electrodes of said second pair being located in such position in the series relative to the electrodes of the first pair that at least a portion of said second field region effectively continues spatially from and beyond the first field region, and electric energy source means connected and operable to impress across the first pair of electrodes a first electrical impulse having a duration effective to produce electrochemical migration in said first interelectrode field region of a first group of negative fluid ions substantially to one of said first two electrodes and of a second group of positive fluid ions substantially to the other of such first two electrodes, said source means being also connected and operable to impress a second electrical impulse across said second pair of electrodes having its inception effectively delayed by a period permitting the aforesaid ionic migrations, causing a relative polarity of said second pair of electrodes to produce further migration of said migrated ions in one of said groups into the second interelectrode region during said second electrical impulse, and having a duration effecting migration of ions in the first and second groups participating in the first migration substantially to respective electrodes of the second pair.

25. Apparatus as defined in claim 24, wherein the series arrangement of electrodes includes additional electrode pairs, alternate ones of which sequentially in the series are connected to the energy source with said first and second pairs of electrodes respectively so that the first electrical impulse is impressed on certain other pairs spatially recurrent along the series so as to effect similar ionic migration at a plurality of points along the electrode series.

26. Apparatus as defined in claim 25, wherein the first and second pulses recur periodically and the electric energy source means is operable to reverse the polarity of the impressed electrical impulses on alternate recurrences of each.

27. Apparatus as defined in claim 25, wherein each of the first pairs of electrodes includes successively adjacent electrodes in the series, each of the respectively related second pairs of electrodes comprises the electrode next adjacent the respectively related first pair and that electrode of the related first pair which is separated from the related second pair by the other electrode of the first pair, thereby to constitute the related first and second pairs from spatially recurrent groups of first, second and third electrodes in the series, and wherein the electric energy source means is connected and operable to impress impulses on the electrodes consisting of periodically recurring, first, second, and third time-sequential impulses, said first impulse being impressed across the first and second electrodes, said second impulse across said second and third electrodes, and said third impulse across said third and first electrodes, the inception of the second impulse occurring at a time intermediate the times of inception of the first and third impulses.

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