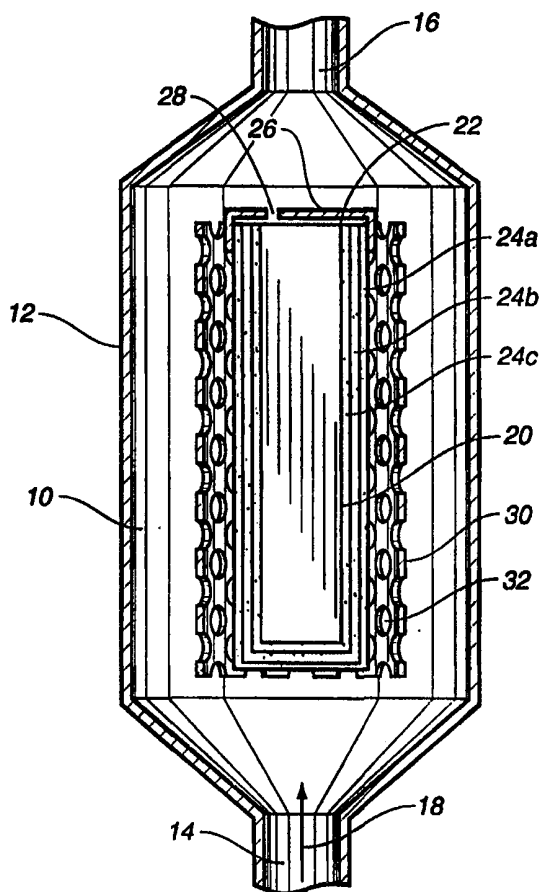




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(21) International Application Number: PCT/US99/03064 (22) International Filing Date: 12 February 1999 (12.02.99) (30) Priority Data: 09/025,120 17 February 1998 (17.02.98) US (71) Applicant: POTABLE WATER SYSTEMS LTD. [US/US]; Suite 120, 952 Echo Lane, Houston, TX 77024 (US). (72) Inventor: MILLER, Jorge; Apartment 9204, 3300 Sage Road, Houston, TX 77056 (US). (74) Agent: BROOKHART, Walter, R.; Browning Bushman, Suite 1800, 5718 Westheimer Road, Houston, TX 77057 (US).		(81) Designated States: AL, AM, AT, AU, AZ, BA, BB, BG, BR, BY, CA, CH, CN, CU, CZ, DE, DK, EE, ES, FI, GB, GE, GH, GM, HR, HU, ID, IL, IS, JP, KE, KG, KP, KR, KZ, LC, LK, LR, LS, LT, LU, LV, MD, MG, MK, MN, MW, MX, NO, NZ, PL, PT, RO, RU, SD, SE, SG, SI, SK, SL, TJ, TM, TR, TT, UA, UG, UZ, VN, YU, ZW, ARIPO patent (GH, GM, KE, LS, MW, SD, SZ, UG, ZW), Eurasian patent (AM, AZ, BY, KG, KZ, MD, RU, TJ, TM), European patent (AT, BE, CH, CY, DE, DK, ES, FI, FR, GB, GR, IE, IT, LU, MC, NL, PT, SE), OAPI patent (BF, BJ, CF, CG, CI, CM, GA, GN, GW, ML, MR, NE, SN, TD, TG). Published <i>With international search report.</i> <i>With amended claims.</i>
(54) Title: ELECTROLYTIC CELL WITH POROUS MEMBRANES TO CONCENTRATE ANIONS		
(57) Abstract The present invention is directed to an apparatus (10) and method for the treatment of water by generating a purifying gas from anions in the water. Preferably, the apparatus (10) and method of the present invention provide for the purification of water by the generation of chlorine at the anode (20) from naturally occurring chloride in the water. The electrolytic cell (10) of the present invention includes a closed electrolytic chamber (12) having a lower inlet opening (14) and an upper outlet opening (16) for directing water to be treated through the chamber. Disposed within the chamber (12) are an anode (20) and a cathode (30) in electrical contact with a conventional direct current power source. Disposed about the anode (20), preferably substantially surrounding the anode (20) and forming the walls of an anode compartment are a plurality of porous membranes (24) disposed so that water must pass sequentially through each membrane before contacting the anode. In the presently preferred embodiment, these porous membranes (24) comprise alternating layers of polypropylene felt and microporous polytetrafluoroethylene film. Most preferably, the cell (10) includes 2-6 sequentially disposed porous membranes (24).		



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ELECTROLYTIC CELL WITH POROUS MEMBRANES TO CONCENTRATE ANIONS

Background of the Invention

I. Field of the Invention

The present invention generally relates to an electrolytic cell for the treatment of water. More specifically, the present invention is directed to
5 methods and apparatus for purifying and sterilizing water by concentrating anions from which a purifying gas may be generated, e.g., chloride ions, in the water which contacts the anode.

II. Description of the Background

Many various types of electrolytic cells for the purification and sterilization
10 of water have proposed. In some of these cells, consumable or non-consumable electrodes of iron, aluminum, copper, silver, platinum, carbon and the like have been used to remove contaminating materials dissolved or suspended in the water to be treated. However, these cells have suffered from many operational problems. For example, during the electrolysis of water, and particularly of hard
15 water, a layer of calcium carbonate will rapidly cover the cathode. This carbonate layer inhibits the flow of current and, thus, the operation of the cell. Metal anodes, particularly silver, copper, iron and aluminum, become covered with an oxide layer during electrolysis. These conductive oxides inhibit dissolution of the anode metal and encourage production of oxygen, rather than
20 the preferred purifying gas, at the anode.

These systems have also encounter problems resulting from the normally low concentration of anions in the water. For example, in order to purify water with chlorine, the concentration of chloride in the water contacting the anode should be at least 2,000 ppm. Below this level, chlorine generation is
5 insufficient to produce an acceptable level of purification. The concentration of chloride found in most water sources is significantly below this threshold. Accordingly, additional chloride must be added to the cell or the concentration of chloride in the water contacting the anode must be increased.

Electrolytic cells have long been used to produce chlorine for purification
10 of the water in swimming pools and spas. While it is theoretically possible to add sufficient chloride to the pool water, e.g., by the addition of common table salt, i.e., sodium chloride, to increase the chloride concentration to the desired range, the required concentrations would result in an undesirable salty taste being imparted to the pool water. Further, at the required concentrations, the
15 deposition of calcium on the cathode and the resulting interference with the flow of current in the electrolysis circuit would be increased to unacceptable levels. Accordingly, swimming pool chlorination systems have typically employed a separate electrolysis cell containing a high concentration of salt in which chlorine is produced for introduction into the pool water.

20 Wellwater, surface water and sewage water typically contains only about 40-400 ppm chloride. Taste and environmental discharge regulations prevent the addition of chloride to such waters to achieve the desired chloride concentration. Alternatively, it has been suggested that chloride ions may be

concentrated in an anode compartment by the use of anion selective membranes. Such membranes permit the passage of anions into the anode compartment while minimizing back diffusion due to the concentration gradient across the membrane. Once the chloride ion concentration in the anode compartment has reached a sufficient level, e.g., 2,000 ppm, the electrolysis cell can produce sufficient chlorine to effectively purify the water. Chlorine gas generated at the anode may escape from the anode compartment and mix with the water to be treated through an appropriate aperture in the top of the anode compartment. An exemplary system is disclosed in United States Patent No. 4,121,991 which is incorporated herein by reference.

Anion selective membranes used in such systems have most often used tertiary, quaternary and polyamines to provide the desired amine exchange capacity. Unfortunately, these amines react with the chlorine generated at the anode to liberate nitrogen. Thus, the exchange capacity of these anion selective membranes is continuously destroyed by the chlorine generated at the anode. In fact, because of the continuous destruction of the exchange capacity of the anion selective membranes, it has been necessary to regularly replace the anion selective membranes. Accordingly, prior art electrolytic cells using anion selective membranes to concentrate chloride in the anode compartment have suffered from unnecessary down time and high operating costs.

If the anion selective membrane were replaced by a porous membrane, the back diffusion of the concentrated chloride ions which depend on the concentration difference across the membrane and on the porosity of the

membrane normally would be so pronounced that insufficient concentration in the anode apartment could be achieved. Accordingly, systems which used a porous membrane were also unsuccessful.

Accordingly, those skilled in the art have long sought a more acceptable
5 means for concentrating the desired anions in the water contacting the anode. Thus, there has been a long felt but unfulfilled need for a more economical and more efficient means for concentrating the anions contacting the anode of an electrolytic cell used to purify water. The present invention solves those needs.

Summary of the Invention

10 The present invention is directed to an apparatus and method for the treatment of water using an electrolytic cell including a series of porous membranes for concentrating anions from which a purifying gas may be generated at the anode.

The electrolytic cell of the present invention includes a closed electrolytic
15 chamber having a lower inlet opening and an upper outlet opening for directing water to be treated. Disposed within the chamber are an anode and a cathode in electrical contact with a direct current power source for driving the electrolytic cell. In a presently preferred embodiment, the anode is disposed along the axis of the chamber and surrounded by a concentrically disposed cathode
20 constructed of an expanded or perforated metal screen.

The electrolytic cell further includes a plurality of porous membranes disposed about the anode so that water must pass sequentially through each membrane before contacting the anode. In the presently preferred embodiment,

these membranes form substantially all of the walls of a closed anode compartment within which the anode is disposed. The anode compartment surrounds the anode with the exception of a small aperture at the top thereof to permit escape of fluids which permeate through the porous membranes and
5 gases generated at the surface of the anode. The presently preferred embodiment employs 2-6 sequentially disposed porous membranes selected from the group of materials consisting of porous porcelain and microporous plastics. In the most presently preferred embodiment, alternating layers of polypropylene felt and polytetrafluoroethylene (PTFE) film are employed.

10 In the method of the present invention, the water to be treated is directed into and through an electrolytic chamber having a cathode and an anode. All water which contacts the anode is directed sequentially through each of a plurality of porous membranes surrounding the anode in order to concentrate anions from which a purifying gas may be generated. In the presently preferred
15 embodiment, water contacting the anode is directed sequentially through a plurality of porous membranes comprising alternate layers of polypropylene felt and microporous polytetrafluoroethylene film. By forcing the water which will contact the anode sequentially through a series of porous membranes, back diffusion across each membrane is minimized and the anion concentrated to a
20 level sufficient to permit generation at the anode of an effective concentration of purifying gas. The method of the present invention is particularly appropriate for the generation of chlorine from chloride ions naturally found in the water to be treated.

Thus, the long felt, but unfulfilled need for an improved method for generating a purifying gas, preferably chlorine, in an electrolytic cell has been met. These and other meritorious features and advantages of the present invention will be more fully appreciated from the following description and
5 claims.

Brief Description of the Drawings

Other features and intended advantages of the present invention will be more readily apparent by the references to the following detailed description in connection with the accompanying drawings, wherein:

10 Fig. 1 is a cross-sectional illustration of an electrolytic cell in accord with the present invention and useful for concentrating anions and generating a purifying gas in accord with the method of the present invention; and

Fig. 2 is a cross-sectional illustration of a composite porous membrane comprising a polypropylene felt and polytetrafluoroethylene film for use in an
15 electrolytic cell in accord with the present invention.

While the invention will be described in connection with the presently preferred embodiments, it will be understood that it is not intended to limit the invention to those embodiments. On the contrary, it is intended to cover all alternatives, modifications and equivalents as may be included in the spirit of the
20 invention as defined in the appended claims.

Detailed Description of the Preferred Embodiments

The present invention provides an improved, more efficient and more economical apparatus and method for generating a purifying gas from anions

occurring naturally in water to be treated. The present invention provides an electrolytic cell capable of concentrating anions, particularly chloride ions, naturally found in the water to be treated and of electrolyzing them to chlorine gas for purification of the water. In the absence of chloride ions, other anions
5 such as carbonate and sulfate found in the water may concentrate within the anode compartment. Electrolysis of these anions produces percarbonate and persulfate, respectively, each providing an alternative oxidizing agent for purifying the water.

The present invention overcomes the deficiency caused by the high back
10 diffusion gradient across a single, porous membrane by providing means for concentrating the anions in an anode compartment to a level sufficient to permit purifying concentrations of gas, preferably chlorine, to be generated. Since back diffusion is proportional to the concentration difference across the membrane, use of a plurality of sequentially disposed membranes minimizes the
15 concentration gradient across each membrane, thus minimizing back diffusion. The effect is similar to that achieved by using a series of labyrinths in steam turbines to prevent the loss of steam.

The electrolytic cell of the present invention includes a closed electrolytic chamber having a lower inlet opening and an upper outlet opening for directing
20 water to be treated through the chamber. Also disposed within the chamber are an anode and a cathode which are in electrical contact with a direct power source to drive the electrolytic cell. At least partially surrounding the anode are

a plurality of porous membranes disposed so that water must pass sequentially through each membrane before contacting the anode. In the presently preferred embodiment, these porous membranes comprise the walls of an anode compartment completely surrounding the anode with the exception of a small
5 aperture in the top of the compartment to permit the escape of gases formed at the anode and water which has permeated through the porous membranes.

The presently preferred embodiment includes 2-6 sequentially disposed porous membranes selected from the group of materials consisting of porous porcelain and the microporous plastics. In the presently most preferred
10 embodiment, the membranes comprise alternating layers of polypropylene felt and microporous polytetrafluoroethylene (PTFE) film. Exemplary are polypropylene felts having a thickness of about 0.1-0.5 inch. Those having a thickness of about 0.25 inch are particularly useful.

The anode may be a single anode or multiple anodes and may be
15 constructed with any desired configuration. Anodes may be constructed from any appropriate conductive material. Exemplary appropriate materials include the corrosion resistant metals of the platinum family, platinized titanium, niobium, graphite, carbon and metal oxides. The anode may be solid or may be formed of a screen, grid or expanded or perforated metal sheet to provide greater
20 surface area. While less desirable, corrodible anodes constructed of aluminum, iron, copper or silver may alternatively be employed.

Any cathode deemed appropriately by those skilled in the art may be employed. In one exemplary embodiment, the cathode may take the form of a

plurality of electrodes disposed radially about an anode placed along the longitudinal axis of the electrolytic chamber. Another exemplary configuration includes a cathode comprising an electrode concentrically disposed about an anode. The cathode may be constructed from any suitable conductive material, e.g., stainless steel, copper and the like. While the cathode may be solid, it may also be formed of a screen, grid or expanded or perforated metal sheet.

The chamber of the electrolytic cell may be constructed of many materials known to those skilled in the art. Exemplary materials include plastics, porcelain, glass, hard rubber and concrete. In fact, if constructed in part of a conductive metal, the metal of the chamber might serve as the cathode of the electrolytic cell.

The present invention will now be described in connection with the exemplary electrolytic cell illustrated in Fig. 1. The electrolytic cell 10 includes closed electrolytic chamber 12 having inlet opening 14 and an outlet opening 16 to direct water to the treated in the cell upwardly as illustrated by direction arrow 18.

Electrolytic cell 10 includes an anode 20 disposed along the longitudinal axis of chamber 12 within anode compartment 22. Compartment 22 is formed by a plurality of sequentially disposed porous membranes 24 substantially surrounding anode 20 together with a top wall 26. Within wall 26 is small aperture 28 designed to permit gases generated at anode 20 to escape from anode compartment 22 to mix with and purify the water flowing through cell 10.

Aperture 28 also permits escape of water which has permeated through membranes 24 into anode compartment 22.

Concentrically disposed about anode 20 and surrounding porous membranes 24 is cathode 30 formed of a perforated metal screen to facilitate
5 passage of water and dissolved anions through perforations 32 and into contact with porous membranes 24.

The electrolytic cell 10 is driven by an appropriate direct current power source (not shown) electrically connected across anode 20 and cathode 30. Due to the electrical potential difference between these electrodes, a
10 concentration gradient is established which serves to drive dissolved anions, e.g., chloride, successively through porous membranes 24a, 24b and 24c into anode compartment 22 and toward anode 20. Thus, the concentration of anions within the anode compartment may be increased to a level, e.g., greater than 2,000 ppm chloride, where free chlorine in sufficient quantities to purify the water
15 can be generated.

Fig. 2 illustrates in cross-section a preferred porous membrane combination comprising a polypropylene felt 40 together with a polytetrafluoroethylene (PTFE) film 42. The combination porous membrane illustrated in Fig 2. may, in a preferred embodiment, be employed to form each
20 layer 24a, 24b and 24c of the wall of anode compartment 22 in electrolytic cell 10, thus, providing a cell with six (6) porous membranes through which water and dissolved anions must pass before contacting anode 20.

The foregoing description has been directed in primary part to a particular preferred embodiment in accord with the requirements of the Patent Statutes and for purposes of explanation and illustration. It will be apparent, however, to those skilled in the art that many modifications and changes in the specifically
5 described apparatus and methods may be made without departing from the true scope and spirit of the invention. For example, while the invention has been illustrated with a single, axially disposed anode substantially surrounded by three porous membranes, many other configurations and combinations may be used. Therefore, the invention is not restricted to the preferred embodiment
10 described and illustrated but covers all modifications which may fall within the scope of the following claims.

What is claimed is:

1. An electrolytic cell for the treatment of water, comprising:
a closed electrolytic chamber having a lower inlet opening and an
upper outlet opening for directing water to be treated through said chamber;
5 a cathode disposed in said chamber for contact with said water;
an anode compartment disposed within said chamber, said
compartment formed at least in part by a plurality of porous membranes
disposed so that water must pass sequentially through each said membrane
before entering into said compartment; and
10 an anode disposed within said compartment.
2. The electrolytic cell of Claim 1 comprising 2-6 sequentially
disposed porous membranes through which said water must pass.
3. The electrolytic cell of Claim 1 wherein said porous membranes
are selected from the group consisting of porous porcelain and microporous
15 plastics.
4. The electrolytic cell of Claim 3 wherein said microporous plastics
include porous polypropylene felt and microporous polytetrafluoroethylene film.

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5. The electrolytic cell of Claim 1 wherein said porous membranes comprise a microporous polytetrafluoroethylene film together with a polypropylene felt having a thickness of about 0.1-0.5 inch.

6. The electrolytic cell of Claim 1 further comprising an aperture in the
5 top of said compartment to permit the escape of gases formed at said anode.

7. An electrolytic cell for the treatment of water, comprising:
a closed electrolytic chamber having a lower inlet opening and an
upper outlet opening for directing water to be treated through said chamber;
an anode and a cathode disposed within said chamber for contact
10 with said water; and

a plurality of porous membranes disposed about said anode so
that water must pass sequentially through each said membrane before
contacting said anode.

8. The electrolytic cell of Claim 7 comprising 2-6 sequentially
15 disposed porous membranes through which said water must pass.

9. The electrolytic cell of Claim 7 wherein said porous membranes
are selected from the group consisting of porous porcelain and microporous
plastics.

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10. The electrolytic cell of Claim 7 wherein said porous membranes comprise alternating layers of polypropylene felt and microporous polytetrafluoroethylene.

11. The electrolytic cell of Claim 7 wherein said chamber has a longitudinal axis, said anode being disposed along said axis and said cathode comprising a plurality of electrodes disposed radially about said anode.

12. The electrolytic cell of Claim 7 wherein said chamber has a longitudinal axis, said anode being disposed along said axis and said cathode comprising an electrode concentrically disposed about said anode.

13. The electrolytic cell of Claim 12 wherein said cathode is an expanded metal screen.

14. The electrolytic cell of Claim 7 wherein said anode is disposed in an anode compartment.

15. The electrolytic cell of Claim 14 wherein said porous membranes form at least a part of the walls of said compartment.

16. The electrolytic cell of Claim 14 further including an aperture in the top of said compartment to permit the escape of gases generated at said anode.

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17. The electrolytic cell of Claim 7 further comprising a direct current source connected across said anode and said cathode.

18. A method for treating water, comprising:
directing water into an electrolytic chamber having a cathode and
5 an anode;
directing at least a portion of said water sequentially through each of a plurality of porous membranes at least partially surrounding said anode so that all water contacting said anode must pass sequentially through said plurality of membranes; and
10 applying a direct current across said anode and said cathode to generate at said anode a gas for treating said water.

19. The method of Claim 18 wherein said water includes chloride ions and said gas is chlorine.

20. The method of Claim 18 further comprising increasing the
15 concentration of chloride in the water contacting said anode to at least 2,000 ppm by driving chloride ions sequentially through each of said porous membranes by application of said current across said anode and cathode.

21. The method of Claim 18 wherein said porous membrane is selected from the group consisting of porous porcelain and microporous plastics.

22. The method of Claim 20 wherein said porous membranes comprise alternating layers of polypropylene felt and microporous polytetrafluoroethylene film.

23. The method of Claim 18 wherein said electrolytic cell is selected
5 from the group consisting of the cells defined in Claims 1-17.

AMENDED CLAIMS

[received by the International Bureau on 17 June 1999 (17.06.99);
original claims 1, 7 and 18-20 amended;
remaining claims unchanged (3 pages)]

What is claimed is:

1. (Amended) An electrolytic cell for the treatment of water, comprising:
a closed electrolytic chamber having a lower inlet opening and an upper outlet opening for directing water to be treated through said chamber;
a cathode disposed in said chamber for contact with said water;
an anode compartment disposed within said chamber, said compartment formed at least in part by a plurality of porous membranes sequentially disposed so that all of the water entering said compartment must pass sequentially through each said membrane before entering into said compartment, said plurality of membranes sequentially disposed to reduce the rate of back diffusion from said compartment to said chamber of an anion found in said water so that said anion is concentrated within said compartment to a level sufficient to permit a purifying concentration of an oxidizing agent for purifying said water to be produced; and
an anode disposed within said compartment for generating said oxidizing agent from said anion.
2. The electrolytic cell of Claim 1 comprising 2-6 sequentially disposed porous membranes through which said water must pass.
3. The electrolytic cell of Claim 1 wherein said porous membranes are selected from the group consisting of porous porcelain and microporous plastics.
4. The electrolytic cell of Claim 3 wherein said microporous plastics include porous polypropylene felt and microporous polytetrafluoroethylene film.

5. The electrolytic cell of Claim 1 wherein said porous membranes comprise a microporous polytetrafluoroethylene film together with a polypropylene felt having a thickness of about 0.1-0.5 inch.

6. The electrolytic cell of Claim 1 further comprising an aperture in the top of said compartment to permit the escape of gases formed at said anode.

7. (Amended) An electrolytic cell for the treatment of water, comprising:
a closed electrolytic chamber having a lower inlet opening and an upper outlet opening for directing water to be treated through said chamber;
an anode and a cathode disposed within said chamber for contact with said water; and

a plurality of porous membranes sequentially disposed and forming an anode compartment about said anode so that all of the water contacting said anode must pass sequentially through each said membrane, said plurality of membranes sequentially disposed to reduce the rate of back diffusion and increase the concentration of an anion found in said water to a level within said anode compartment sufficient to produce at said anode a purifying concentration of an oxidizing agent for purifying said water.

8. The electrolytic cell of Claim 7 comprising 2-6 sequentially disposed porous membranes through which said water must pass.

9. The electrolytic cell of Claim 7 wherein said porous membranes are selected from the group consisting of porous porcelain and microporous plastics.

17. The electrolytic cell of Claim 7 further comprising a direct current source connected across said anode and said cathode.

18. (Amended) A method for treating water, comprising:
directing water into an electrolytic chamber having a cathode and an anode;

directing a portion of said water sequentially through each of a plurality of porous membranes forming at least part of an anode compartment surrounding said anode so that all water contacting said anode has passed sequentially through said plurality of membranes;

concentrating within said compartment to a level sufficient to permit a purifying concentration of an oxidizing agent to be produced at least one anion found in said water by reducing the rate of back diffusion of said anion from said compartment to said chamber; and

applying a direct current across said anode and said cathode to generate at said anode from said anion a gas comprising an oxidizing agent in a concentration sufficient for purifying said water.

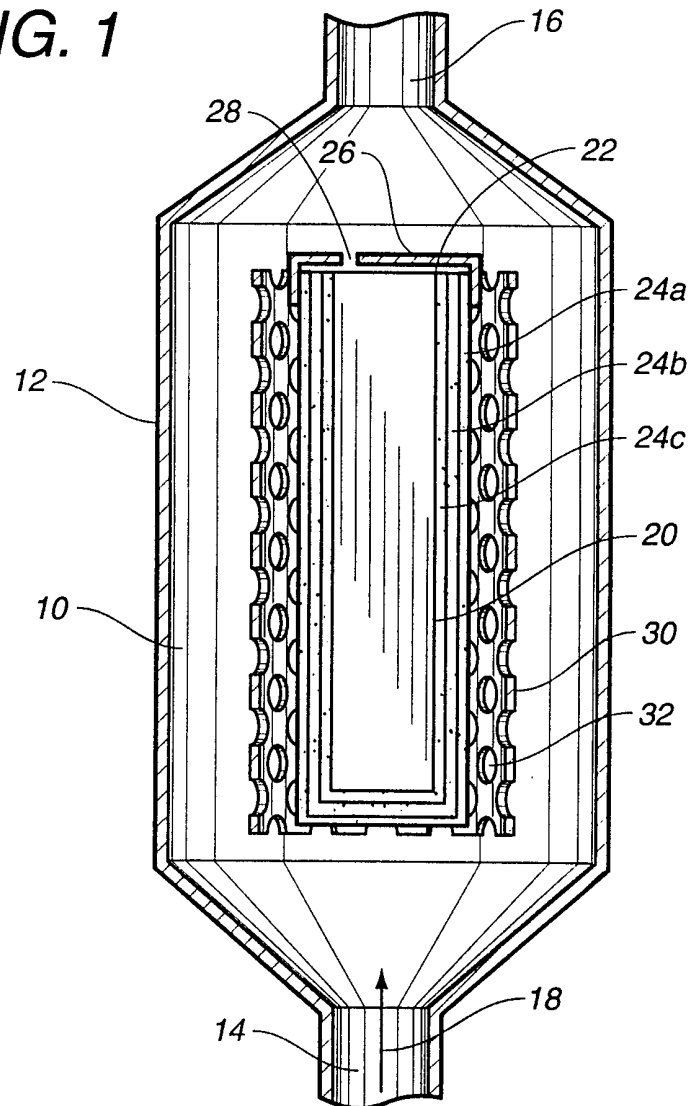
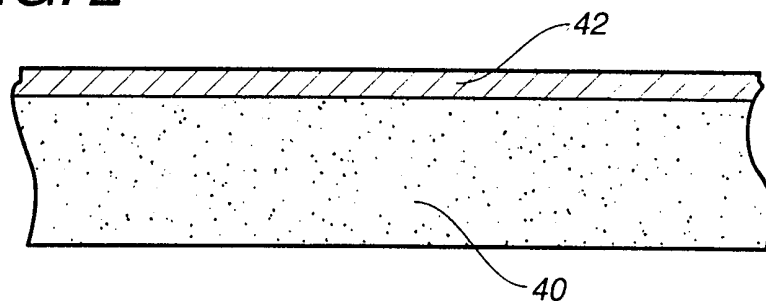
19. (Amended) The method of Claim 18 wherein said anion is chloride and said gas is chlorine.

20. (Amended) The method of Claim 19 further comprising increasing the concentration of chloride in the water contacting said anode in said compartment to at least 2,000 ppm by driving chloride ions sequentially through each of said porous membranes by application of said current across said anode and cathode.

21. The method of Claim 18 wherein said porous membrane is selected from the group consisting of porous porcelain and microporous plastics.

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FIG. 1**FIG. 2**

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INTERNATIONAL SEARCH REPORT

International application No.
PCT/US99/03064

A. CLASSIFICATION OF SUBJECT MATTER

IPC(6) :C02F 1/461, 1/467

US CL :205/701, 703, 748, 770; 204/260, 263, 272, 275

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

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

U.S. : 205/701, 703, 748, 770; 204/260, 263, 272, 275

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched
NONE

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)
NONE

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
Y	US 4,119,518 A (MILLER) 10 October 1978, see figs. 4 and 5 and claims 1-22.	1-23
Y	US 4,121,991 A (MILLER et al.) 24 October 1978, see figs. 1 and 2 and claims 1-6.	1-23

☐ Further documents are listed in the continuation of Box C. ☐ See patent family annex.

* Special categories of cited documents:	*T* later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention
A document defining the general state of the art which is not considered to be of particular relevance	*X* document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone
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Date of the actual completion of the international search
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13 APR 1999

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