

DESCRIPTION

Title of the Invention: Electrolytic cell

5 Technical Field:

[0001]

The present invention relates to an electrolytic cell. More specifically, the present invention relates, but is not limited, to an electrolytic cell that can be
10 suitably used to produce an aqueous solution of quaternary ammonium hydroxide from an aqueous solution of quaternary ammonium salt.

Background Art:

15 [0002]

Patent Documents 1 to 4 below disclose a method for producing an aqueous solution of quaternary ammonium hydroxide from an aqueous solution of quaternary ammonium salt. Such a production method involves the use of an
20 electrolytic cell including a cathode frame, a cathode plate fixed to the inner surface of the cathode frame, an anode frame, and an anode plate fixed to the inner surface of the anode frame. At least one cation exchange membrane is provided between the cathode plate and the anode plate.
25 A cathode compartment is defined between the cathode frame and the cation exchange membrane, and an anode compartment is defined between the anode frame and the cation exchange membrane. The electrolytic cell further includes a cathode side fluid supply path for supplying a cathode side fluid
30 to the cathode compartment, a cathode side fluid discharge path for discharging the cathode side fluid from the cathode compartment, an anode side fluid supply path for supplying an anode side fluid to the anode compartment, and an anode side fluid discharge path for discharging the
35 anode side fluid from the anode compartment. More

specifically, the cathode frame includes a plurality of upper flow paths extending respectively from a plurality of upper openings arranged at intervals in the width direction and a plurality of lower flow paths extending respectively from a plurality of lower openings arranged at intervals in the width direction. Either the upper or lower flow paths serve as the cathode side fluid discharge path, and the other flow paths serve as the cathode side fluid supply path. Similarly, the anode frame includes a plurality of upper flow paths extending respectively from a plurality of upper openings arranged at intervals in the width direction and a plurality of lower flow paths extending respectively from a plurality of lower openings arranged at intervals in the width direction. Either the upper or lower flow paths serve as the anode side fluid discharge path, and the other flow paths serve as the anode side fluid supply path. The cathode side fluid, that is, an aqueous solution of quaternary ammonium hydroxide, is circulated through the cathode side fluid supply path, the cathode compartment, and the cathode side fluid discharge path. The anode side fluid, that is, an aqueous solution of quaternary ammonium salt, is circulated through the anode side fluid supply path, the anode compartment, and the anode side fluid discharge path.

Prior Art Documents:

Patent Documents:

[0003]

Patent Document 1: JP-A-62-142792

Patent Document 2: JP-B-8-16274

Patent Document 3: JP-B-8-19539

Patent Document 4: JP-A-2009-13477

Summary of the Invention:

Problems to be solved by the Invention:

[0004]

In the conventional electrolytic cell as described above, the cathode side fluid supply path and the cathode side fluid discharge path are set to have substantially the same effective flow path cross-sectional area, and the anode side fluid supply path and the anode side fluid discharge path are set to have substantially the same effective flow path cross-sectional area. In particular, in an electrolytic cell as proposed by the present inventors in Japanese Patent Application No. 2022-33651 (that is, an electrolytic cell in which the cathode plate includes at least one upper through opening in its upper end part that matches with the upper opening in the cathode frame, and at least one lower through opening in its lower end part that matches with the lower opening in the cathode frame; the anode plate includes at least one upper through opening in its upper end part that matches with the upper opening in the anode frame, and at least one lower through opening in its lower end part that matches with the lower opening in the anode frame; and the upper and lower through openings formed in the cathode and anode plates determine the effective flow path cross-sectional areas of the fluid flow paths), the effective flow path cross-sectional areas are set to the minimum necessary to achieve maximum electrolysis efficiency. However, after repeated intensive studies and experiments on the operation of electrolytic cells, the present inventors have found the following problem. Gases produced in the cathode compartment and the anode compartment caused by electrolytic action flow through the cathode side fluid discharge path and the anode side fluid discharge path together with the cathode side fluid and the anode side fluid, respectively. This increases the flow velocity in the cathode side fluid discharge path and the anode side fluid discharge path, as compared to that in the cathode side fluid supply path and

the anode side fluid supply path, so that back pressure is generated. As a result, the electrolytic voltage is increased and, accordingly, a stable operation of electrolytic cells tends to be inhibited.

5 [0005]

The present invention has been made in view of the aforementioned fact, and a primary technical object of the invention is to provide a novel and improved electrolytic cell that can be operated with ensured stability without causing an increase in electrolytic voltage.

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Means for solving the Problems:

[0006]

In light of the above-described fact, the present inventors have found that the above-described primary technical object can be achieved by: allowing a cathode side fluid discharge path to have an effective flow path cross-sectional area CD larger than an effective flow path cross-sectional area CS of a cathode side fluid supply path and allowing an anode side fluid discharge path to have an effective flow path cross-sectional area AD larger than an effective flow path cross-sectional area AS of an anode side fluid supply path.

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[0007]

More specifically, in order to achieve the primary technical object, the present invention provides an electrolytic cell including a cathode frame with a cathode plate fixed to its inner surface, an anode frame with an anode plate fixed to its inner surface, an ion exchange membrane provided between the cathode frame and the anode frame, a cathode side fluid supply path for supplying a cathode side fluid to a cathode compartment defined between the cathode frame and the ion exchange membrane, a cathode side fluid discharge path for discharging the cathode side fluid from the cathode compartment, an anode side fluid

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supply path for supplying an anode side fluid to an anode compartment defined between the anode frame and the ion exchange membrane, and an anode side fluid discharge path for discharging the anode side fluid from the anode

5 compartment, wherein

the cathode side fluid discharge path has an effective flow path cross-sectional area CD larger than an effective flow path cross-sectional area CS of the cathode side fluid supply path, and the anode side fluid discharge path has an effective flow path cross-sectional area AD larger than an effective flow path cross-sectional area AS of the anode side fluid supply path.

[0008]

It is preferable that the effective flow path cross-sectional area CD is 1.1 to 3.0 times as large as the effective flow path cross-sectional area CS (i.e., $CD = 1.1$ to $3.0CS$), and the effective flow path cross-sectional area AD is 1.1 to 3.0 times as large as the effective flow path cross-sectional area AS (i.e., $AD = 1.1$ to $3.0AS$). In particular, it is preferable that the effective flow path cross-sectional area CD is 1.5 to 2.5 times as large as the effective flow path cross-sectional area CS (i.e., $CD = 1.5$ to $2.5CS$), and the effective flow path cross-sectional area AD is 1.5 to 2.5 times as large as the effective flow path cross-sectional area AS (i.e., $AD = 1.5$ to $2.5AS$).

[0009]

In a preferred embodiment, the cathode frame includes at least one upper flow path extending from an upper opening located in an upper end part of the inner surface and at least one lower flow path extending from a lower opening located in a lower end part of the inner surface,

the anode frame includes at least one upper flow path extending from an upper opening located in an upper end part of the inner surface and at least one lower flow path extending from a lower opening located in a lower end part

of the inner surface,

the cathode plate extends continuously from above the upper opening of the cathode frame to below the lower opening of the cathode frame,

5 the cathode plate includes at least one upper through opening in its upper end part that matches with the upper opening in the cathode frame, and at least one lower through opening in its lower end part that matches with the lower opening in the cathode frame,

10 the anode plate extends continuously from above the upper opening of the anode frame to below the lower opening of the anode frame,

the anode plate includes at least one upper through opening in its upper end part that matches with the upper opening in the anode frame, and at least one lower through opening in its lower end part that matches with the lower opening in the anode frame,

the upper through opening in the cathode plate and the upper flow path in the cathode frame constitute either the cathode side fluid discharge path or the cathode side fluid supply path, and the lower through opening in the cathode plate and the lower flow path in the cathode frame constitute the other path, which is the cathode side fluid discharge path or the cathode side fluid supply path, and

25 the upper through opening in the anode plate and the upper flow path in the anode frame constitute either the anode side fluid discharge path or the anode side fluid supply path, and the lower through opening in the anode plate and the lower flow path in the anode frame constitute the other path, which is the anode side fluid discharge path or the anode side fluid supply path.

[0010]

Preferably, the cathode frame includes a plurality of the upper flow paths extending respectively from a plurality of the upper openings arranged at intervals in a

width direction in the upper end part of the inner surface,
and a plurality of the lower flow paths extending
respectively from a plurality of the lower openings
arranged at intervals in the width direction in the lower
5 end part of the inner surface,

the anode frame includes a plurality of the upper
flow paths extending respectively from a plurality of the
upper openings arranged at intervals in the width direction
in the upper end part of the inner surface, and a plurality
10 of the lower flow paths extending respectively from a
plurality of the lower openings arranged at intervals in
the width direction in the lower end part of the inner
surface,

the cathode plate includes a plurality of the upper
15 through openings in its upper end part that are formed in
alignment with the plurality of respective upper openings
in the cathode frame, and a plurality of the lower through
openings in its lower end part that are formed in alignment
with the plurality of respective lower openings in the
20 cathode frame,

the anode plate includes a plurality of the upper
through openings in its upper end part that are formed in
alignment with the plurality of respective upper openings
in the anode frame, and a plurality of the lower through
25 openings in its lower end part that are formed in alignment
with the plurality of respective lower openings in the
anode frame,

the upper through openings in the cathode plate and
the upper flow paths in the cathode frame or the lower
30 through openings in the cathode plate and the lower flow
paths in the cathode frame that serve as the cathode side
fluid discharge path are present in larger numbers than the
lower through openings in the cathode plate and the lower
flow paths in the cathode frame or the upper through
35 openings in the cathode plate and the upper flow paths in

the cathode frame that serve as the cathode side fluid supply path, and

the upper through openings in the anode plate and the upper flow paths in the anode frame or the lower through openings in the anode plate and the lower flow paths in the anode frame that serve as the anode side fluid discharge path are present in larger numbers than the lower through openings in the anode plate and the lower flow paths in the anode frame or the upper through openings in the anode plate and the upper flow paths in the anode frame that serve as the anode side fluid supply path.

[0011]

It is suitable that the upper openings and the lower openings in the cathode frame and the upper through openings and the lower through openings in the cathode plate are circular in cross-section, and the upper openings and the lower openings in the anode frame and the upper through openings and the lower through openings in the anode plate are circular in cross-section. It is desirable that the cathode plate and the anode plate are formed of a rectangular plate.

[0012]

According to a preferred embodiment of the present invention, the above-described electrolytic cell including at least one cation exchange membrane between the cathode plate and the anode plate is used to produce an aqueous solution of quaternary ammonium hydroxide from an aqueous solution of quaternary ammonium salt as a raw material.

Effects of the Invention:

[0013]

In the electrolytic cell of the present invention, the cathode side fluid discharge path has the effective flow path cross-sectional area CD larger than the effective flow path cross-sectional area CS of the cathode side fluid

supply path, and the anode side fluid discharge path has the effective flow path cross-sectional area AD larger than the effective flow path cross-sectional area AS of the anode side fluid supply path. Thus, the electrolytic cell
5 can be operated with ensured stability without causing an increase in electrolytic voltage, as will be clearly understood from Examples below.

Brief Description of the Drawings:

10 [0014]

[Fig. 1]: a schematic cross-sectional view showing a preferred embodiment of an electrolytic cell constituted in accordance with the present invention;

[Fig. 2]: a schematic cross-sectional view showing a
15 cathode frame and a cathode plate in the electrolytic cell shown in Fig. 1, taken along the line II-II in Fig. 1;

[Fig. 3]: a partially enlarged cross-sectional view showing an upper opening in the cathode frame, an upper through opening formed in the cathode plate, and an upper
20 communicating opening formed in a gasket, in the electrolytic cell shown in Fig. 1; and

[Fig. 4]: a schematic cross-sectional view showing, similarly to Fig. 2, a cathode frame and a cathode plate in a modification of the electrolytic cell constituted in
25 accordance with the present invention.

Mode for Carrying out the Invention:

[0015]

Hereinafter, a preferred embodiment of an
30 electrolytic cell constituted in accordance with the present invention will be described in further detail with reference to the accompanying drawings.

[0016]

Referring to Figs. 1 and 2, the illustrated
35 electrolytic cell constituted in accordance with the

present invention includes a hollow rectangular parallelepiped housing 2 constructed from a cathode frame 4 (Fig. 1), an anode frame 6 (Fig. 1), a cathode side upper wall member 8, an anode side upper wall member 10, a cathode side lower wall member 12, an anode side lower wall member 14, a cathode side front wall member 16 (Fig. 2), an anode side front wall member (not shown), a cathode side back wall member 18 (Fig. 2), and an anode side back wall member (not shown). The cathode frame 4, the anode frame 6, the cathode side front wall member 16, the anode side front wall member, the cathode side back wall member 18, and the anode side back wall member extend substantially vertically, while the cathode side upper wall member 8, the anode side upper wall member 10, the cathode side lower wall member 12, and the anode side lower wall member 14 extend substantially horizontally. The base end surface (i.e., the right end surface in Fig. 1) of the cathode side upper wall member 8 is connected to an upper end edge part of the inner surface of the cathode frame 4 by appropriate connecting means such as a fastening screw or an adhesive, and the base end surface (i.e., the right end surface in Fig. 1) of the cathode side lower wall member 12 is connected to a lower end edge part of the inner surface of the cathode frame 4 by appropriate connecting means. Similarly, the base end surface (i.e., the left end surface in Fig. 1) of the anode side upper wall member 10 is connected to an upper end edge part of the inner surface of the anode frame 6 by appropriate connecting means, and the base end surface (i.e., the left end surface in Fig. 1) of the anode side lower wall member 14 is connected to a lower end edge part of the inner surface of the anode frame 6 by appropriate connecting means. The cathode side front wall member 16 is connected to the front surface of the cathode frame 4 and the anode side front wall member is connected to the front surface of the anode frame 6 by appropriate

connecting means. The cathode side back wall member 18 is connected to the back surface of the cathode frame 4 and the anode side back wall member is connected to the back surface of the anode frame 6 by appropriate connecting means. The upper wall member 8, 10 and the lower wall member 12, 14 on the cathode or anode side, as well as the front wall member 16 and the back wall member 18 are connected to the cathode frame 4 or the anode frame 6 in the above-described manner. Alternatively, these wall members may be integrally formed in advance before being connected to the cathode frame 4 or the anode frame 6. Alternatively, the wall members may be integrally formed with the cathode frame 4 or the anode frame 6. When the upper wall member 8, 10 and the lower wall member 12, 14 as well as the front wall member 16 and the back wall member 18 are integrally formed, they may be fixed to the cathode frame 4 or the anode frame 6 via a seal member. In such a case, a gasket 38 (described later), which corresponds in size to the cathode plate 32 or an anode plate 56, may be extended to correspond to the cathode frame 4 or the anode frame 6 so that the extended portion can serve as the seal member. Each of the cathode frame 4, the anode frame 6, the cathode side upper wall member 8, the anode side upper wall member 10, the cathode side lower wall member 12, the anode side lower wall member 14, the cathode side front wall member 16, the anode side front wall member, the cathode side back wall member 18, and the anode side back wall member may be in the form of a solid block or plate except for openings and flow paths (described later), and can be made of an appropriate synthetic resin, such as an olefin-based resin (e.g., polypropylene or polyethylene), a vinyl chloride-based resin, or a fluorine-based resin. An appropriate sealing member (not shown), such as a gasket, can be placed in each interconnection region between the cathode frame 4, the anode frame 6, the cathode side upper

wall member 8, the anode side upper wall member 10, the cathode side lower wall member 12, the anode side lower wall member 14, the cathode side front wall member 16, the anode side front wall member, the cathode side back wall member 18, and the anode side back wall member.
[0017]

Still referring to Figs. 1 and 2 as well as Fig. 3, in an upper end part of the inner surface (i.e., the left surface in Fig. 1) of the cathode frame 4, a plurality of (e.g., six in the drawing) upper openings 20 (one of which is shown by a dashed line and a solid line in Figs. 1 and 3, respectively) are formed at regular intervals in the width direction (i.e., the direction perpendicular to the paper plane in Fig. 1; the horizontal direction in Fig. 2). The cathode frame 4 includes a plurality of (e.g., six in the drawing) upper flow paths 22 (one of which is shown by a dashed line and a solid line in Figs. 1 and 3, respectively), each extending substantially horizontally from each of the upper openings 20 to penetrate through the cathode frame 4. Similarly, in a lower end part of the inner surface (i.e., the left surface in Fig. 1) of the cathode frame 4, a plurality of (e.g., three in the drawing) lower openings 24 (one of which is shown by a dashed line in Fig. 1) are formed at regular intervals in the width direction (i.e., the direction perpendicular to the paper plane in Fig. 1; the horizontal direction in Fig. 2). The cathode frame 4 includes a plurality of (e.g., three in the drawing) lower flow paths 26 (one of which is shown by a dashed line in Fig. 1), each extending substantially horizontally from each of the lower openings 24 to penetrate through the cathode frame 4. The upper openings 20 and the lower openings 24 may be circular, and the upper flow paths 22 and the lower flow paths 26 may be circular in cross-section to match the circular shape of the upper openings 20 and the lower openings 24. The upper

flow paths 22 and the lower flow paths 26 are connected to each other by an outside flow path (not shown) provided on the outside of the housing 2. An outside flow path 31 (partially shown in Fig. 2) is provided with a circulating pump, a product storage tank, a plurality of valve members for flow control, and the like. (A detailed description of the outside flow path and the aforementioned components provided therein will be omitted herein as they are well known to those skilled in the art.)

10 [0018]

The cathode plate 32 is fixed to the inner surface of the cathode frame 4. In the illustrated embodiment, the cathode plate 32 extends continuously from above the upper openings 20 formed in the cathode frame 4 to below the lower openings 24 formed in the cathode frame 4. The cathode plate 32, which is constituted from a rectangular plate formed of appropriate conductive metal such as nickel, is located such that: its upper end surface abuts or is close to the inner surface (i.e., the lower surface) of the cathode side upper wall member 8; its lower end surface abuts or is close to the inner surface (i.e., the upper surface) of the cathode side lower wall member 12; its front side surface abuts or is close to the inner surface (i.e., the back surface) of the front wall member 16; and its back side surface abuts or is close to the inner surface (i.e., the front surface) of the back wall member 18. The cathode plate 32 can be fixed to the cathode frame 4 suitably by, for example, inserting a fastening screw (not shown) in each corner of the cathode plate 32 so that the cathode plate 32 is screwed to the cathode frame 4. In an upper end part of the cathode plate 32, a plurality of (e.g., six in the drawing) upper through openings 34 are formed at regular intervals in the width direction (i.e., the direction perpendicular to the paper plane in Fig. 1; the horizontal direction in Fig. 2). In a

lower end part of the cathode plate 32, a plurality of (e.g., three in the drawing) lower through openings 36 are formed at regular intervals in the width direction (i.e., the direction perpendicular to the paper plane in Fig. 1; the horizontal direction in Fig. 2). (The upper through openings 34 and the lower through openings 36 will be further referred to later.) The upper through openings 34 formed in the cathode plate 32 are located in alignment with the respective upper openings 20 formed in the upper end part of the cathode frame 4. In the illustrated embodiment, the upper through openings 34 have the same shape (i.e., a circular shape) and dimensions as the upper openings 20. Similarly, it is important for the lower through openings 36 formed in the cathode plate 32 to be located in alignment with the respective lower openings 24 formed in the lower end part of the cathode frame 4. In the illustrated embodiment, the lower through openings 36 have the same shape (i.e., a circular shape) and dimensions as the lower openings 24. It is preferable that the upper through openings 34 (and the upper openings 20) are located close to the upper end of the cathode plate 32 and that the lower through openings 36 (and the lower openings 24) are located close to the lower end of the cathode plate 32.

[0019]

Still referring to Figs. 1 and 3, the gasket 38 is suitably interposed between the cathode frame 4 and the cathode plate 32. The gasket 38 helps to stably fix the cathode plate 32 to the cathode frame 4 and to prevent, for example, corrosion resulting from liquid infiltration at the interface between the cathode frame 4 and the cathode plate 32. The gasket 38 may be formed of a rectangular plate with substantially the same dimensions as the cathode plate 32 (or alternatively, the gasket 38 can also be sized to correspond to the cathode frame 4 as mentioned above).

The gasket 38 can be made of an appropriate elastomer, such

as silicone rubber, ethylene propylene rubber, chloroprene rubber, soft PVC, butyl rubber, butadiene rubber, or fluoro rubber. When the gasket 38 is interposed between the cathode frame 4 and the cathode plate 32, a fastening screw (not shown) may be inserted in each corner of the cathode plate 32 so that the cathode plate 32 together with the gasket 38 can be screwed to the cathode frame 4. In an upper end part of the gasket 38, a plurality of (e.g., six in the drawing) upper communicating openings 40 are formed, through which the upper through openings 34 formed in the cathode plate 32 and the upper openings 20 formed in the cathode frame 4 communicate respectively with each other. In a lower end part of the gasket 38, a plurality of (e.g., three in the drawing) lower communicating openings 42 are formed, through which the lower through openings 36 formed in the cathode plate 32 and the lower openings 24 formed in the cathode frame 4 communicate respectively with each other. As is clearly understood from Fig. 3, the upper communicating opening 40 and the lower communicating opening 42 formed in the gasket 38 are desired to be larger than the upper through opening 34 and the upper opening 20, as well as the lower through opening 36 and the lower opening 24. When the upper communicating opening 40 and the lower communicating opening 42 formed in the gasket 38 have substantially the same dimensions as the upper through opening 34 and the upper opening 20, as well as the lower through opening 36 and the lower opening 24, the communication between the upper through opening 34 and the upper opening 20 and the communication between the lower through opening 36 and the lower opening 24 tend to be insufficient or impaired after continued operation of the electrolytic cell, because the gasket 38 is somewhat swollen and causes the upper communicating opening 40 and the lower communicating opening 42 to be reduced in size and displaced. The upper communicating opening 40 and the

lower communicating opening 42 may be in the shape of a circle with a diameter larger by a predetermined amount than that of the upper through opening 34 and the upper opening 20, as well as the lower through opening 36 and the lower opening 24. The upper communicating opening 40 and the lower communicating opening 42 do not always have to be concentric but can be eccentric with respect to the upper through opening 34 and the upper opening 20, and the lower through opening 36 and the lower opening 24, respectively. The diameter of the upper communicating opening 40 and the lower communicating opening 42 and the degree of their eccentricity with respect to the upper through opening 34 and the upper opening 20, and the lower through opening 36 and the lower opening 24, respectively, can be set on an experimental basis based on the swelling and displacement of the gasket 38 caused by continued operation of the electrolytic cell. When the upper communicating opening 40 and the lower communicating opening 42 formed in the gasket 38 are larger, the communication between the respective openings is less likely to be insufficient or impaired. However, the larger upper communicating opening 40 and lower communicating opening 42 increase the area of contact with a liquid on the rear surface of the cathode plate 32, which results in galvanic or metallic corrosion, so that an increased amount of electrode metal is contained in the liquid, as described later. Accordingly, each preferred size of the upper communicating opening 40 and the lower communicating opening 42 formed in the gasket 38 is provided as follows: The size of the upper communicating opening 40 is larger than each size of the upper through opening 34 and the upper opening 20 by 30 mm or less, preferably 20 mm or less, and more preferably 10 mm or less; and the size of the lower communicating opening 42 is larger than each size of the lower through opening 36 and the lower opening 24 by 30 mm or less, preferably 20 mm or

less, and more preferably 10 mm or less. Meanwhile, when the gasket 38 is made of a material that hardly swells, the communication between the respective openings is less likely to be insufficient or impaired even when the upper communicating opening 40 and the lower communicating opening 42 are close in size to the upper through opening 34 and the upper opening 20, as well as the lower through opening 36 and the lower opening 24. As such, the material for the gasket 38 preferably has a coefficient of linear expansion of 3×10^{-4} ($1/^{\circ}\text{C}$) or less, more preferably 1.5×10^{-4} ($1/^{\circ}\text{C}$) or less, and most preferably 1×10^{-4} ($1/^{\circ}\text{C}$) or less. [0020]

In the illustrated embodiment, the anode frame 6 is substantially the same as the above-described cathode frame 4. More specifically, the cathode frame 4 and the anode frame 6 are symmetric with respect to a virtual plane extending therebetween perpendicularly to the paper plane in Fig. 1. As such, the anode frame 6 includes upper openings 44, upper flow paths 46, lower openings 48, and lower flow paths 50. To avoid duplication of description, the upper openings 44, the upper flow paths 46, the lower openings 48, and the lower flow paths 50 will not be described in detail. The upper flow paths 46 and the lower flow paths 50 are connected to each other by an outside flow path (not shown) provided on the outside of the housing 2. The outside flow path is provided with a circulating pump, a product storage tank, a plurality of valve members for flow control, and the like. (A detailed description of the outside flow path and the aforementioned components provided therein will be omitted herein as they are well known to those skilled in the art.) [0021]

The anode plate 56 is fixed to the inner surface of the anode frame 6. In the illustrated embodiment, the anode plate 56 is substantially the same as the above-

described cathode plate 32, except that it is made of an appropriate conductive metal suitable for an anode, such as titanium with an indium oxide plated surface. More specifically, the cathode plate 32 and the anode plate 56 are symmetric with respect to a virtual plane extending therebetween perpendicularly to the paper plane in Fig. 1. As such, the anode plate 56 has a rectangular shape that extends continuously from above the upper openings 44 formed in the anode frame 6 to below the lower openings 48 formed in the anode frame 6. The anode plate 56 includes upper through openings 58 and lower through openings 60 located in matching with the upper openings 44 and the lower openings 48, respectively, formed in the anode frame 6. To avoid duplication of description, the anode plate 56 will not be described in detail.

[0022]

In the illustrated embodiment, the gasket 62 is interposed also between the anode frame 6 and the anode plate 56. The gasket 62 is also substantially the same as the gasket 38 interposed between the cathode frame 4 and the cathode plate 32. More specifically, the gasket 38 and the gasket 62 are symmetric with respect to a virtual plane extending therebetween perpendicularly to the paper plane in Fig. 1. As such, the gasket 62 includes upper communicating openings 64 through which the upper openings 44 formed in the anode frame 6 and the upper through openings 58 formed in the anode plate 56 communicate respectively with each other, and lower communicating openings 66 through which the lower openings 48 formed in the anode frame 6 and the lower through openings 60 formed in the anode plate 56 communicate respectively with each other. To avoid duplication of description, the gasket 62, the upper communicating opening 64, and the lower communicating opening 66 will not be described in detail.

[0023]

As is clearly understood from Fig. 1, a cation exchange membrane 68 is provided between the cathode plate 32 and the anode plate 56 in the illustrated embodiment. The cation exchange membrane 68, which may be in a form known per se, is in the shape of a rectangular plate, and is located such that: its upper end edge part is held between the cathode side upper wall member 8 and the anode side upper wall member 10; its lower end edge part is held between the cathode side lower wall member 12 and the anode side lower wall member 14; its edge part on the front surface side of the cathode frame 4 and the anode frame 6 is held between the cathode side front wall member 16 and the anode side front wall member; and its edge part on the back surface side of the cathode frame 4 and the anode frame 6 is held between the cathode side back wall member 18 and the anode side back wall member. An appropriate seal member (not shown) can be placed between the cation exchange membrane 68 and each of the cathode side upper wall member 8, the anode side upper wall member 10, the cathode side lower wall member 12, the anode side lower wall member 14, the cathode side front wall member 16, the anode side front wall member, the cathode side back wall member 18, and the anode side back wall member.

[0024]

In the electrolytic cell as described above, a cathode or product compartment 70 is defined between the cathode plate 32 and the cation exchange membrane 68, and an anode, that is, material compartment 72 is defined between the anode plate 56 and the cation exchange membrane 68. At an early stage, a dilute aqueous solution of quaternary ammonium hydroxide (or pure water) is circulated through the product compartment 70. More specifically, the solution flows into the product compartment 70 either through the lower flow paths 26 or the upper flow paths 22 formed in the cathode frame 4 and flows out of the product

compartment 70 through the other flow paths. At the same time, an aqueous solution of quaternary ammonium salt is circulated through the material compartment 72. More specifically, the solution flows into the material

5 compartment 72 either through the lower flow paths 50 or the upper flow paths 46 formed in the anode frame 6 and flows out through the other flow paths. An electrolytic voltage is applied between the cathode plate 32 and the anode plate 56. Thus, the aqueous solution of quaternary ammonium hydroxide circulated through the product
10 compartment 70 gradually increases in concentration. A detailed description of such an electrolytic action will be omitted herein as it is well known to those skilled in the art. According to the illustrated embodiment of the
15 electrolytic cell constructed in accordance with the present invention, the cathode plate 32 and the anode plate 56 extend continuously from above the upper openings 20 and 44 to below the lower openings 24 and 48 formed in the cathode frame 4 and the anode frame 6, respectively. The
20 cathode plate 32 and the anode plate 56 include the upper through openings 34 and 58 and the lower through openings 36 and 60 formed in alignment with the upper openings 20 and 44 and the lower openings 24 and 48, respectively. As such, the cathode plate 32 and the anode plate 56 extend
25 over approximately the entire inner surfaces of the cathode frame 4 and the anode frame 6, respectively. Thus, the current-carrying area of the cathode plate 32 and the anode plate 56 is relatively large for the size of the electrolytic cell, with the result that electrolysis is
30 conducted with increased electrolysis efficiency.

[0025]

In the electrolytic cell constructed in accordance with the present invention, it is important that a cathode side fluid discharge path has an effective flow path cross-
35 sectional area CD larger than an effective flow path cross-

sectional area CS of a cathode side fluid supply path and similarly that an anode side fluid discharge path has an effective flow path cross-sectional area AD larger than an effective flow path cross-sectional area AS of an anode side fluid supply path. The term "effective flow path cross-sectional area" as used herein refers to the smallest cross-sectional area of the flow paths. In the illustrated embodiment, the cathode side fluid discharge path CD is determined from the six upper through openings 34 formed in the cathode plate 32, as well as the upper communicating openings 40, the upper openings 20, the upper flow paths 22, and a part of the outside flow path 31 that communicate with the upper through openings 34. The cross-sectional areas of the upper communicating opening 40, the upper opening 20, the upper flow path 22, and a part of the outside flow path 31 are equal to or larger than the cross-sectional area of the upper through opening 34. Thus, the effective flow path cross-sectional area CD of the cathode side fluid discharge path is defined by the sum of the cross-sectional areas of the six upper through openings 34. Meanwhile, the effective flow path cross-sectional area CS of the cathode side fluid supply path is determined from the three lower through openings 36 formed in the cathode plate 32, as well as the lower communicating openings 42, the lower openings 24, the lower flow paths 26, and a part of the outside flow path 31 that communicate with the lower through openings 36. The cross-sectional areas of the lower communicating opening 42, the lower opening 24, the lower flow path 26, and a part of the outside flow path 31 are equal to or larger than the cross-sectional area of the lower through opening 36. Thus, the effective flow path cross-sectional area CD of the cathode side fluid discharge path is defined by the sum of the cross-sectional areas of the three lower through openings 36. In particular, from the viewpoint of suppressing an increase in back pressure

induced by the fluid being discharged, the cross-sectional area of a part of the outside flow path 31 is preferably larger than those of the upper through opening 34 or the lower through opening 36. Specifically, the cross-sectional area of a part of the outside flow path 31 is preferably 1.0 times or more, more preferably 1.5 times or more, larger than those of the upper through opening 34 and the lower through opening 36. The cross-sectional area of a part of the outside flow path 31 relative to the cross-sectional areas of the upper through opening 34 or the lower through opening 36 is determined as appropriate according to the effective flow path cross-sectional areas and the supply of the fluid.

[0026]

In the illustrated embodiment, the six upper through openings 34 are formed at regular intervals in the width direction (i.e., the direction perpendicular to the paper plane in Fig. 1; the horizontal direction in Fig. 2), while the three lower through openings 36 are formed at regular intervals in the width direction (i.e., the direction perpendicular to the paper plane in Fig. 1; the horizontal direction in Fig. 2). The cross-sectional area of each of the six upper through openings 34 is the same as the cross-sectional area of each of the three lower through openings 36. Accordingly, the effective flow path cross-sectional area CD of the cathode side fluid discharge path is twice as large as the effective flow path cross-sectional area CS of the cathode side fluid supply path. As will be understood from Examples below, the effective flow path cross-sectional area CD of the cathode side fluid discharge path is preferably 1.1 to 3.0 times (i.e., $CD = 1.1$ to $3.0CS$), particularly 1.5 to 2.5 times (i.e., $CD = 1.5$ to $2.5CS$), as large as the effective flow path cross-sectional area CS of the cathode side fluid supply path.

[0027]

The cross-sectional area of each of the upper through openings 34 and the lower through openings 36 formed in the cathode plate 32 is desired to be set to the minimum necessary to achieve maximum electrolysis efficiency. From the above, after repeated intensive studies and experiments on the operation of electrolytic cells, the present inventors have found the following problem. Gases produced in the cathode or product compartment 70 caused by electrolytic action flow through the cathode side fluid discharge path together with the cathode side fluid. Thus, if the effective flow path cross-sectional area CD of the cathode side fluid discharge path is set to be equal to the effective flow path cross-sectional area CS of the cathode side fluid supply path, the flow velocity increases in the cathode side fluid discharge path, as compared to that in the cathode side fluid supply path, so that back pressure is generated. As a result, the electrolytic voltage is increased and, accordingly, a stable operation of the electrolytic cell tends to be inhibited. From the above, the electrolytic cell constructed in accordance with the present invention is such that the effective flow path cross-sectional area CD of the cathode side fluid discharge path is set to be larger than the effective flow path cross-sectional area CS of the cathode side fluid supply path, whereby the electrolytic cell can be operated with ensured stability without causing an increase in electrolytic voltage, as will be clearly understood from Examples below.

[0028]

In the above-described embodiment shown in Figs. 1 to 3, the number (six) of the upper through openings 34 is larger than the number (three) of the lower through openings 36, so that the effective flow path cross-sectional area CD of the cathode side fluid discharge path is larger than the effective flow path cross-sectional area

CS of the cathode side fluid supply path. Alternatively, if desired, the number of the upper through openings 34 may be the same as that of the lower through openings 36, and instead the cross-sectional area of each of the upper through openings 34 can be set to be larger than that of each of the lower through openings 36, so that the effective flow path cross-sectional area CD of the cathode side fluid discharge path is larger than the effective flow path cross-sectional area CS of the cathode side fluid supply path. In a modification shown in Fig. 4, the three upper through openings 34 are formed at regular intervals in the width direction (i.e., the horizontal direction in Fig. 4) in an upper end part of the cathode plate 32. (Accordingly, there are the three upper communicating openings 40, the three upper openings 20, and the three upper flow paths 22.) In a lower end part of the cathode plate 32, the three lower through openings 36 are formed at regular intervals in the width direction (i.e., the horizontal direction in Fig. 4). The cross-sectional area of each of the upper through openings 34 is set to be twice as large as that of each of the lower through openings 36. However, according to the experience of the present inventors, providing the upper through openings 34 in larger numbers than the lower through openings 36 tends to be more effective in facilitating the flow of discharge, rather than allowing the upper through openings 34 to have a larger cross-sectional area than the lower through openings 36.

[0029]

In the above-described embodiment and modification, the cathode plate 32 includes the plurality of upper through openings 34 and lower through openings 36 formed at regular intervals in the width direction. Alternatively, if desired, the cathode plate 32 can include a single upper through opening and a single lower through opening that are

elongated in the width direction such that the upper through opening 34 has a larger cross-sectional area than the lower through opening 36.

[0030]

5 Further, in the above-described embodiment and modification, the cathode plate 32 extends continuously from above the upper openings 20 formed in the cathode frame 4 to below the lower openings 24 formed in the cathode frame 4, and the cathode plate 32 includes the
10 plurality of upper through openings 34 in its upper end part that are formed at regular intervals in the width direction (i.e., the direction perpendicular to the paper plane in Fig. 1; the horizontal direction in Figs. 2 and 4) and the plurality of lower through openings 36 in its lower
15 end part that are formed at regular intervals in the width direction (i.e., the direction perpendicular to the paper plane in Fig. 1; the horizontal direction in Figs. 2 and 4). Alternatively, if desired, the cathode plate 32 can extend from below the upper openings 20 formed in the
20 cathode frame 4 to above the lower openings 24 formed in the cathode frame 4, and the cathode side fluid discharge path can be such that its upstream end is defined by the upper openings 20 formed in the cathode frame 4, while the cathode side fluid supply path can be such that its
25 downstream end is defined by the lower openings 24 formed in the cathode frame 4, with no upper through opening 34 and no lower through opening 36 formed in the cathode plate 32, for example. In such a case, the effective flow path cross-sectional area of the cathode side fluid discharge
30 path is defined by the sum of the cross-sectional areas of the upper openings 20, and the effective flow path cross-sectional area of the cathode side fluid supply path is defined by the sum of the cross-sectional areas of the lower openings 24.

35 [0031]

The detailed description has been given of the relationship between the effective flow path cross-sectional area CD of the cathode side fluid discharge path and the effective flow path cross-sectional area CS of the cathode side fluid supply path. The same applies to the relationship between the effective flow path cross-sectional area AD of the anode side fluid discharge path and the effective flow path cross-sectional area AS of the anode side fluid supply path. To avoid duplication of description, the relationship between the effective flow path cross-sectional area AD of the anode side fluid discharge path and the effective flow path cross-sectional area AS of the anode side fluid supply path will not be described in detail.

[0032]

Examples 1 to 3 and Comparative Example 1

The electrolytic cells as shown in Fig. 1 to 3 or Fig. 4 were prepared and operated in constant current control mode, and the average of the cell voltage (i.e., the voltage applied between the anode plate and the cathode plate) during operating time was detected. In each of Examples 1 to 3, the effective flow path cross-sectional area CD of the cathode side fluid discharge path was larger than the effective flow path cross-sectional area CS of the cathode side fluid supply path, and the effective flow path cross-sectional area AD of the anode side fluid discharge path was larger than the effective flow path cross-sectional area AS of the anode side fluid supply path. On the other hand, in Comparative Example 1, the effective flow path cross-sectional area CD of the cathode side fluid discharge path was equal to the effective flow path cross-sectional area CS of the cathode side fluid supply path, and the effective flow path cross-sectional area AD of the anode side fluid discharge path was equal to the effective flow path cross-sectional area AS of the anode side fluid

supply path. The average cell voltage values were as shown in Table 1.

[0033]

Component details and operating conditions of the
5 electrolytic cells were as follows.

Operating time: 30 days

Cathode: Nickel plate (thickness: 2 mm; surface
dimensions: 1 m × 1 m)

Anode: Titanium plate with an indium oxide
10 plated surface (thickness: 2 mm; surface dimensions: 1 m ×
1 m)

Cation exchange membrane: Exchange membrane
(thickness: 1 mm) marketed by The Chemours Company under
the product name of "N324"

15 Gasket: EPDM (the upper communicating opening
and the lower communicating opening were 2 mm larger in
diameter than the corresponding upper through opening and
lower through opening, respectively)

Raw material (anode side fluid): Aqueous
20 solution of methylammonium chloride

Product (cathode side fluid): Aqueous solution
of methylammonium hydroxide

Current: 1000 A (10 A/dm²)

Operating temperature: 70°C

25 Temperature during assembly of electrolytic
cell: 20°C

Flow rate of raw material (anode side fluid):
20 L/min

Flow rate of product (cathode side fluid): 20
30 L/min

[0034]

[Table 1]

	Upper through opening (discharge path)		Lower through opening (supply path)		Cell voltage (V)
	Number of openings	Diameter of opening (mm)	Number of openings	Diameter of opening (mm)	
Example 1	12	10	10	10	8.9
Example 2	20	10	10	10	8.4
Example 3	10	16	10	10	8.6
Comparative Example 1	10	10	10	10	9.8

[0035]

The preferred embodiment of the present invention has been described in detail with reference to the accompanying drawings. However, the present invention is not limited to this embodiment, and various modifications and alterations can be made without departing from the technical scope of the present invention. For example, in the illustrated embodiment, the single cation exchange membrane 68 is provided between the cathode plate 32 and the anode plate 56. However, the present invention is also applicable to an electrolytic cell including a plurality of exchange membranes (i.e., a cation exchange membrane and an anion exchange membrane) between the cathode plate 32 and the anode plate 56.

Explanations of Letters or Numerals:

[0036]

20 2: Housing of electrolytic cell
4: Cathode frame
6: Anode frame
8: Cathode side upper wall member
10: Anode side upper wall member
25 12: Cathode side lower wall member
14: Anode side lower wall member
16: Cathode side front wall member

18: Cathode side back wall member
20: Upper opening
22: Upper flow path
24: Lower opening
5 26: Lower flow path
31: Outside flow path
32: Cathode plate
34: Upper through opening
36: Lower through opening
10 38: Gasket
40: Upper communicating opening
42: Lower communicating opening
44: Upper opening
46: Upper flow path
15 48: Lower opening
50: Lower flow path
56: Anode plate
58: Upper through opening
60: Lower through opening
20 62: Gasket
64: Upper communicating opening
66: Lower communicating opening
68: Cation exchange membrane
70: Product compartment (cathode compartment)
25 72: Material compartment (anode compartment)

Claims

[Claim 1]

An electrolytic cell comprising a cathode frame with
5 a cathode plate fixed to its inner surface, an anode frame
with an anode plate fixed to its inner surface, an ion
exchange membrane provided between the cathode frame and
the anode frame, a cathode side fluid supply path for
supplying a cathode side fluid to a cathode compartment
10 defined between the cathode frame and the ion exchange
membrane, a cathode side fluid discharge path for
discharging the cathode side fluid from the cathode
compartment, an anode side fluid supply path for supplying
an anode side fluid to an anode compartment defined between
15 the anode frame and the ion exchange membrane, and an anode
side fluid discharge path for discharging the anode side
fluid from the anode compartment, wherein

the cathode side fluid discharge path has an
effective flow path cross-sectional area CD larger than an
20 effective flow path cross-sectional area CS of the cathode
side fluid supply path, and the anode side fluid discharge
path has an effective flow path cross-sectional area AD
larger than an effective flow path cross-sectional area AS
of the anode side fluid supply path.

[Claim 2]

The electrolytic cell according to claim 1, wherein
the effective flow path cross-sectional area CD is 1.1 to
3.0 times as large as the effective flow path cross-
30 sectional area CS (i.e., $CD = 1.1$ to $3.0CS$), and the
effective flow path cross-sectional area AD is 1.1 to 3.0
times as large as the effective flow path cross-sectional
area AS (i.e., $AD = 1.1$ to $3.0AS$).

[Claim 3]

The electrolytic cell according to claim 2, wherein the effective flow path cross-sectional area CD is 1.5 to 2.5 times as large as the effective flow path cross-sectional area CS (i.e., $CD = 1.5 \text{ to } 2.5CS$), and the effective flow path cross-sectional area AD is 1.5 to 2.5 times as large as the effective flow path cross-sectional area AS (i.e., $AD = 1.5 \text{ to } 2.5AS$).

[Claim 4]

10 The electrolytic cell according to any one of claims 1 to 3, wherein

the cathode frame includes at least one upper flow path extending from an upper opening located in an upper end part of the inner surface and at least one lower flow path extending from a lower opening located in a lower end part of the inner surface,

the anode frame includes at least one upper flow path extending from an upper opening located in an upper end part of the inner surface and at least one lower flow path extending from a lower opening located in a lower end part of the inner surface,

the cathode plate extends continuously from above the upper opening of the cathode frame to below the lower opening of the cathode frame,

25 the cathode plate includes at least one upper through opening in its upper end part that is formed in alignment with the upper opening in the cathode frame, and at least one lower through opening in its lower end part that is formed in alignment with the lower opening in the cathode frame,

the anode plate extends continuously from above the upper opening of the anode frame to below the lower opening of the anode frame,

the anode plate includes at least one upper through opening in its upper end part that is formed in alignment

with the upper opening in the anode frame, and at least one lower through opening in its lower end part that is formed in alignment with the lower opening in the anode frame,

the upper through opening in the cathode plate and
5 the upper flow path in the cathode frame constitute either the cathode side fluid discharge path or the cathode side fluid supply path, and the lower through opening in the cathode plate and the lower flow path in the cathode frame constitute the other path, which is the cathode side fluid
10 discharge path or the cathode side fluid supply path, and

the upper through opening in the anode plate and the upper flow path in the anode frame constitute either the anode side fluid discharge path or the anode side fluid supply path, and the lower through opening in the anode
15 plate and the lower flow path in the anode frame constitute the other path, which is the anode side fluid discharge path or the anode side fluid supply path.

[Claim 5]

20 The electrolytic cell according to claim 4, wherein the cathode frame includes a plurality of the upper flow paths extending respectively from a plurality of the upper openings arranged at intervals in a width direction in the upper end part of the inner surface, and a plurality
25 of the lower flow paths extending respectively from a plurality of the lower openings arranged at intervals in the width direction in the lower end part of the inner surface,

the anode frame includes a plurality of the upper
30 flow paths extending respectively from a plurality of the upper openings arranged at intervals in the width direction in the upper end part of the inner surface, and a plurality of the lower flow paths extending respectively from a plurality of the lower openings arranged at intervals in
35 the width direction in the lower end part of the inner

surface,

the cathode plate includes a plurality of the upper through openings in its upper end part that are formed in alignment with the plurality of respective upper openings in the cathode frame, and a plurality of the lower through openings in its lower end part that are formed in alignment with the plurality of respective lower openings in the cathode frame,

the anode plate includes a plurality of the upper through openings in its upper end part that are formed in alignment with the plurality of respective upper openings in the anode frame, and a plurality of the lower through openings in its lower end part that are formed in alignment with the plurality of respective lower openings in the anode frame,

the upper through openings in the cathode plate and the upper flow paths in the cathode frame or the lower through openings in the cathode plate and the lower flow paths in the cathode frame that constitute the cathode side fluid discharge path are present in larger numbers than the lower through openings in the cathode plate and the lower flow paths in the cathode frame or the upper through openings in the cathode plate and the upper flow paths in the cathode frame that constitute the cathode side fluid supply path, and

the upper through openings in the anode plate and the upper flow paths in the anode frame or the lower through openings in the anode plate and the lower flow paths in the anode frame that constitute the anode side fluid discharge path are present in larger numbers than the lower through openings in the anode plate and the lower flow paths in the anode frame or the upper through openings in the anode plate and the upper flow paths in the anode frame that constitute the anode side fluid supply path.

[Claim 6]

The electrolytic cell according to claim 5, wherein
the upper openings and the lower openings in the
cathode frame and the upper through openings and the lower
5 through openings in the cathode plate are circular in
cross-section, and

the upper openings and the lower openings in the
anode frame and the upper through openings and the lower
through openings in the anode plate are circular in cross-
10 section.

[Claim 7]

The electrolytic cell according to claim 4, wherein
the cathode plate and the anode plate are constituted from
15 a rectangular plate.

[Claim 8]

A method for producing an aqueous solution of
quaternary ammonium hydroxide from an aqueous solution of
20 quaternary ammonium salt as a raw material using the
electrolytic cell according to any one of claims 1 to 3
arranged with least one cation exchange membrane between
the cathode plate and the anode plate.