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## (54) ELECTROLYSIS ELECTRODE

(57) An electrolysis electrode with improved durability is to be provided. An electrolysis electrode (1) includes a conductive substrate (2), a catalyst layer (4), and a tantalum oxide layer (5). The conductive substrate (2)

includes at least titanium. The catalyst layer (4) is provided on the conductive substrate (2). The catalyst layer (4) includes platinum and iridium oxide. The tantalum oxide layer (5) is provided on the catalyst layer (4). In the electrolysis electrode (1), the catalyst layer (4) is partially exposed.

# FIG. 1 A

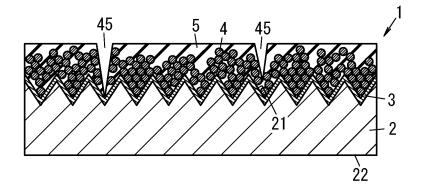
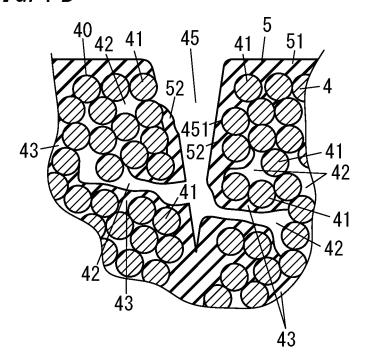


FIG. 1B



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#### **Technical Field**

**[0001]** The present disclosure relates to electrolysis electrodes and specifically relates to an electrolysis electrode including iridium oxide and platinum.

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#### **Background Art**

**[0002]** There is known a technology that produces hypochlorous acid by causing reaction between water and chlorine produced through electrolysis of a diluted sodium chloride solution obtained by adding salt to tap water (Patent Literature 1).

[0003] Patent Literature 1 discloses an electrode for electrolysis that includes: an electrode body made from titanium or titanium alloy; a titanium oxide layer provided on the electrode body; an intermediate oxide layer provided on the titanium oxide layer, the intermediate oxide layer being made of a composite that contains iridium oxide within a range of 3 to 30 mol% and tantalum oxide within a range of 70 to 97 mol% in metal conversion; and a composite body provided on the intermediate oxide layer, the composite body containing rhodium oxide within a range of 2 to 35 mol%, iridium oxide within a range of 30 to 80 mol%, tantalum oxide within a range of 6 to 35 mol%, and platinum within a range of 12 to 62 mol% in metal conversion.

**[0004]** Electrolysis electrodes are desirably improved in durability.

#### **Citation List**

#### **Patent Literature**

[0005] Patent Literature 1: JP 2009-52069 A

## **Summary of Invention**

[0006] It is an object of the present disclosure to provide an electrolysis electrode with improved durability. [0007] An electrolysis electrode according to an aspect of the present disclosure includes a conductive substrate, a catalyst layer, and a tantalum oxide layer. The conductive substrate includes at least titanium. The catalyst layer is provided on the conductive substrate. The catalyst layer includes platinum and iridium oxide. The tantalum oxide layer is provided on the catalyst layer. In the electrolysis electrode, the catalyst layer is partially exposed.

## **Brief Description of Drawings**

## [8000]

FIG. 1A is a sectional view of an electrolysis electrode according to an embodiment;

FIG. 1B is an illustrative view of a main part of the

electrolysis electrode;

FIG. 2 is an illustrative view of particles included in a catalyst layer of the electrolysis electrode;

FIGS. 3A to 3D are sectional views illustrating respective steps in a manufacturing method of the electrolysis electrode;

FIG. 4 is a sectional view of an electrolysis electrode according to Comparative Example 2; and

FIG. 5 is a graph of results of a durability test conducted on an electrolysis electrode according to Example of the embodiment, an electrolysis electrode according to Comparative Example 1, and an electrolysis electrode according to Comparative Example 2.

#### **Description of Embodiments**

**[0009]** FIGS. 1A, 1B, 2, 3A to 3D, and 4 described in the following embodiment and the like are schematic views, and the ratio of sizes and the ratio of thicknesses of components in the figures do not necessarily reflect actual dimensional ratios.

(Embodiment)

**[0010]** An electrolysis electrode 1 according to an embodiment will be described below with reference to FIGS. 1A to 3D.

(1) Overview

[0011] The electrolysis electrode 1 is an electrode used to produce chlorine by electrolyzing salt water. In this case, the salt water is, for example, a sodium chloride solution. Suppose the electrolysis electrode 1 is used for an application of electrolyzing salt water, and in this case, the electrolysis electrode 1 is used as an anode, of a cathode and the anode to which a direct-current voltage is applied from a power supply, thereby electrolyzing a sodium chloride solution to produce chlorine, and through reaction of the chlorine with water, hypochlorous acid water can be produced.

## (2) Components of Electrolysis Electrode

**[0012]** As illustrated in FIG. 1A, the electrolysis electrode 1 includes a conductive substrate 2, a catalyst layer 4, and a tantalum oxide layer 5. The catalyst layer 4 is provided on the conductive substrate 2. The tantalum oxide layer 5 is provided on the catalyst layer 4. The electrolysis electrode 1 further includes an intermediate layer 3 provided between the conductive substrate 2 and the catalyst layer 4.

[0013] These components of the electrolysis electrode 1 will be described in further detail below.

#### (2.1) Conductive Substrate

[0014] The conductive substrate 2 has a first principal surface 21 and a second principal surface 22 located opposite the first principal surface 21. The shape of the conductive substrate 2 in plan view (the outer peripheral shape of the conductive substrate 2 when viewed in the thickness direction defined with respect to the conductive substrate 2) is rectangular. The thickness of the conductive substrate 2 is, for example, greater than or equal to 100  $\mu m$  and less than or equal to 2 mm and is, for example, 500  $\mu m$ . The size of the conductive substrate 2 in plan view is, for example, 25 mm  $\times$  60 mm.

**[0015]** The conductive substrate 2 includes at least titanium. The conductive substrate 2 is, for example, a titanium substrate. The material for the conductive substrate 2 is titanium or an alloy including titanium as a main component (hereinafter referred to as a titanium alloy). The titanium alloy is, for example, a titanium-palladium alloy, a titanium-nickel-ruthenium alloy, a titanium-tantalum alloy, a titanium-aluminum alloy, or a titanium-aluminum-vanadium alloy.

[0016] The first principal surface 21 of the conductive substrate 2 is preferably a rough surface to improve adhesiveness to the intermediate layer 3. In the electrolysis electrode 1 according to the embodiment, the first principal surface 21 of the conductive substrate 2 is roughened before the intermediate layer 3 is provided. Regarding the surface roughness of the first principal surface 21 of the conductive substrate 2, an arithmetic mean roughness Ra is, for example, 0.7  $\mu$ m, and a maximum height Rz is 7  $\mu$ m. The arithmetic mean roughness Ra and the maximum height Rz are specified in, for example, JIS B 0601-2001 (ISO 4287-1997). The arithmetic mean roughness Ra and the maximum height Rz are values measured from, for example, a Crosssectional Scanning Electron Microscope (SEM) Image.

## (2.2) Intermediate Layer

[0017] The intermediate layer 3 is provided on the conductive substrate 2. More specifically, the intermediate layer 3 is provided on the first principal surface 21 of the conductive substrate 2. The electrolysis electrode 1 has an interface between the conductive substrate 2 and the intermediate layer 3. The intermediate layer 3 is preferably made of a material having corrosion resistance against salt water and chlorine and having higher corrosion resistance than the conductive substrate 2. Moreover, to increase the electrical conductivity of the electrolysis electrode 1 as a whole, the material for the intermediate layer 3 is preferably a conductive material having high electrical conductivity. The material for the intermediate layer 3 is, for example, transition metal or a mixture including the transition metal and is, for example, platinum; a mixture of tantalum, platinum, and iridium; iridium; iridium oxide; or nickel. The material for the intermediate layer 3 is, for example, platinum. The thickness of the

intermediate layer 3 is, for example, greater than or equal to 0.2  $\,\mu$ m and less than or equal to 5  $\,\mu$ m, and is, for example, 0.6  $\,\mu$ m.

#### (2.3) Catalyst Layer

**[0018]** The catalyst layer 4 is provided on the intermediate layer 3. The electrolysis electrode 1 has an interface between the catalyst layer 4 and the intermediate layer 3. That is, the catalyst layer 4 is provided on the intermediate layer 3 on the conductive substrate 2.

[0019] The catalyst layer 4 includes platinum and iridium oxide. As shown in FIG. 1B, the catalyst layer 4 is a porous layer including a plurality of composite particles 41 and a plurality of pores 42. As shown in FIG. 2, each of the plurality of composite particles 41 includes a platinum particle 411 and iridium oxide particles 412. In each of the plurality of composite particles 41, for example, a plurality of iridium oxide particles 412 are bonded to one platinum particle 411. In the catalyst layer 4, the iridium oxide is dispersed by the platinum. The iridium oxide functions as a catalyst for producing chlorine. In the catalyst layer 4, the molar ratio of the platinum to the iridium oxide is, for example, but not limited to, 8:5. To suppress iridium from agglomerating over time as the electrolysis electrode 1 is used, the molar quantity of the iridium oxide is preferably less than or equal to the molar quantity of the platinum. The catalyst layer 4 may include iridium in addition to the platinum and the iridium oxide. In this case, each of the composite particles 41 may include, in addition to the iridium oxide particles 412, at least one iridium particle bonded to the platinum particle 411. Moreover, in the catalyst layer 4, the platinum particles 411 may be bonded to each other. The bonding state in the catalyst layer 4 is not particularly limited.

[0020] The catalyst layer 4 has a plurality of recesses 45 recessed from a principal surface 40 on an opposite side of the catalyst layer 4 from the conductive substrate 2. In the electrolysis electrode 1, the catalyst layer 4 is partially exposed in the plurality of recesses 45. Each of the plurality of recesses 45 is, for example, a crack formed in the catalyst layer 4. More specifically, each of the plurality of recesses 45 is a crack which is linear in plan view in the thickness direction defined with respect to the catalyst layer 4. The plurality of cracks (recesses 45) have different shapes. Moreover, each crack may be formed along the thickness direction defined with respect to the catalyst layer 4 or may have a bent on the way in the thickness direction defined with respect to the catalyst layer 4.

[0021] The depth of each of the plurality of recesses 45 is, for example, greater than or equal to 0.1  $\mu m$ . The depth of each of the plurality of recesses 45 may be a depth reaching the intermediate layer 3 or may be a depth not reaching the intermediate layer 3. In the electrolysis electrode 1 according to the embodiment, the plurality of recesses 45 do not extend through the intermediate layer 3, and the entirety of the first principal surface 21 of the

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conductive substrate 2 is covered with the intermediate layer 3. In plan view in the thickness direction defined with respect to the conductive substrate 2, the width of each of the plurality of recesses 45 is greater than or equal to 0.1  $\mu m$  and less than or equal to 10  $\mu m$  and is preferably greater than or equal to 0.3  $\mu m$  and less than or equal to 3  $\mu m$ . The width of each recess 45 in plan view in the thickness direction defined with respect to the conductive substrate 2 is an opening width in a short direction (in a direction orthogonal to the length direction) on the principal surface 40 of the catalyst layer 4. In plan view in the thickness direction defined with respect to the conductive substrate 2, the length of each of the plurality of recesses 45 is shorter than the length of each side of the conductive substrate 2.

[0022] The thickness of the catalyst layer 4 is, for example, within a range of 0.1  $\mu$ m to 10  $\mu$ m.

[0023] Moreover, in plan view in the thickness direction defined with respect to the conductive substrate 2, the percentage of S2 to (S1+S2) is, for example, higher than or equal to 5% and lower than or equal to 50%, where S1 is the area of the principal surface 40 of the catalyst layer 4, and S2 is the total area of opening areas of the plurality of recesses 45 in the principal surface 40 of the catalyst layer 4. The percentage of S2 to (S1+S2) is preferably higher than or equal to 5% to improve the production efficiency of chlorine. Moreover, the percentage of S2 to (S1+S2) is preferably lower than or equal to 50%, more preferably lower than or equal to 20%, to suppress, for example, peel-off of the catalyst layer 4. That is, the percentage of S2 to (S1+S2) is more preferably higher than or equal to 5% and lower than or equal to 20%.

## (2.4) Tantalum Oxide Layer

**[0024]** The tantalum oxide layer 5 has a function of suppressing elution of the iridium oxide of the catalyst layer 4.

[0025] As shown in FIG. 1B, the tantalum oxide layer 5 has a first portion 51 provided on the principal surface 40 of the catalyst layer 4 and a second portion 52 provided on an inner surface 451 of at least one recess 45 of the plurality of recesses 45 in the catalyst layer 4. The tantalum oxide layer 5 preferably has the second portion 52 on the inner surface 451 of each of the plurality of recesses 45 in the catalyst layer 4.

**[0026]** The molar quantity of tantalum in the tantalum oxide layer 5 and iridium in the iridium oxide is preferably lower than or equal to 60% of the total molar quantity of the molar quantity of the platinum.

#### (2.5) Tantalum Oxide

**[0027]** The electrolysis electrode 1 further includes tantalum oxide 43 provided in least one pore 42 of the plurality of pores 42, and the tantalum oxide 43 is in contact with the catalyst layer 4. The tantalum oxide 43 is

formed, for example, at the time of forming the tantalum oxide layer 5. The tantalum oxide 43 is in contact with the composite particles 41 of the catalyst layer 4.

(3) Manufacturing Method of Electrolysis Electrode

**[0028]** With reference to FIGS. 3A to 3D, an example of the manufacturing method of the electrolysis electrode 1 will be described.

10 [0029] In the manufacturing method of the electrolysis electrode 1, the conductive substrate 2 is prepared at first, and then, a surface roughening step, an intermediate layer forming step, a catalyst layer forming step, and a tantalum oxide layer forming step are sequentially performed.

[0030] The surface roughening step includes immersing, for example, the conductive substrate 2 in an oxalic acid aqueous solution, thereby roughening the first principal surface 21 of the conductive substrate 2 (see FIG. 3A). The surface roughening step is not an essential step. Regarding the surface roughness of the first principal surface 21 of the conductive substrate 2 after the surface roughening step, an arithmetic mean roughness Ra is, for example, 0.7  $\mu m$ , and a maximum height Rz is 7  $\mu m$ . The arithmetic mean roughness Ra and the maximum height Rz may be values measured with a surface roughness meter of Zygo Co.

[0031] The intermediate layer forming step includes forming the intermediate layer 3 on the first principal surface 21 of the conductive substrate 2 (see FIG. 3B). The intermediate layer 3 is, for example, a platinum layer. The intermediate layer forming step includes applying a solution which will be the intermediate layer 3 and then performing a heating process, and thereafter, baking the solution, thereby forming the intermediate layer 3. The solution is a solution obtained by dissolving a platinum compound in a solvent. The solvent is, for example, liquid obtained by mixing ethylene glycol monoethyl ether and hydrochloric acid and ethanol. The platinum compound is, for example, but not limited to, chloroplatinic acid, and the platinum compound may be, for example, platinum chloride. The formation method of the intermediate layer 3 is not limited to the examples described above but may be, for example, a vapor-deposition method, a sputtering method, a CVD method, or a plating method.

**[0032]** The catalyst layer forming step includes forming the catalyst layer 4 on the intermediate layer 3 (see FIG. 3C). The catalyst layer forming step includes a first step and a second step.

[0033] The first step of the catalyst layer forming step includes performing an application step at least once and a drying step at least once, thereby forming a catalyst material layer which will be the catalyst layer 4 on the intermediate layer 3 on the conductive substrate 2. The number of times of performing the application step and the drying step is determined based on, for example, a prescribed thickness of the catalyst layer 4. Regarding the number of times of performing the application step

and the drying step, the number of times of performing the application step and the drying step is at least increased as the prescribed thickness of the catalyst layer 4 increases. For example, in the catalyst layer forming step, a first specified number of times (e.g., eight times) of application steps and the first specified number of times of drying steps are alternately repeated one by one, thereby forming the catalyst material layer which will be the catalyst layer 4 on the intermediate layer 3 on the conductive substrate 2.

[0034] In the first step of the catalyst layer forming step, a solution (hereinafter referred to as a first solution) including the platinum compound which will be the catalyst layer 4 and the iridium compound is directly or indirectly applied onto the intermediate layer 3 on the conductive substrate 2 (the application step is performed), and then, a heating process of drying the first solution by heating under a first condition (the drying step) is performed at least once (e.g., eight times), thereby forming the catalyst material layer which will be the catalyst layer 4. The first solution is a solution obtained by dissolving the platinum compound and the iridium compound in a solvent (hereinafter referred to as a first solvent). The first solvent is, for example, liquid obtained by mixing ethylene glycol monoethyl ether and hydrochloric acid and ethanol. The platinum compound is, for example, but not limited to, chloroplatinic acid, and the platinum compound may be, for example, platinum chloride. The chloroplatinic acid is, for example, hydrogen hexachloroplatinate(IV) n-hydrate. The iridium compound is, for example, but not limited to, chloroiridic acid, and the iridium compound may be, for example, iridium chloride or iridium nitrate. The chloroiridic acid is, for example, hexachloroiridate(IV) nhydrate. The metal concentration (the total concentration of platinum and iridium) of the first solution is, for example, 50 mg/mL. Moreover, the application quantity of the first solution is, for example, 2 μL/cm<sup>2</sup>. The first condition includes a heat process temperature and a heat process time. The heat process temperature in the first condition is within a range of 100°C to 400°C, for example, and may be 220°C as an example. Moreover, the heat process time in the first condition is within a range of 5 minutes to 15 minutes, for example, and may be 10 minutes as an example.

[0035] In the second step of the catalyst layer forming step, a heat process of baking the catalyst material layer under a prescribed baking condition is performed, thereby forming the catalyst layer 4 and the plurality of cracks (the recesses 45) (see FIG. 3C). The baking condition includes a baking temperature and a baking time. The baking temperature is within a range of 500°C to 700°C, for example, and may be 560°C as an example. The baking time is within a range of 5 minutes to 20 minutes, for example, and may be 10 minutes as an example.

**[0036]** The tantalum oxide layer forming step includes forming the tantalum oxide layer 5 on the catalyst layer 4 (see FIG. 3D). The tantalum oxide layer forming step includes a first step and a second step.

[0037] The first step of the tantalum oxide layer step includes performing an application step at least once and a drying step at least once, thereby forming a material layer which will be the tantalum oxide layer 5 on the catalyst layer 4. The number of times of performing the application step and the drying step is determined based on, for example, a prescribed thickness of the tantalum oxide layer 5. Regarding the number of times of performing the application step and the drying step, the number of times of performing the application step and the drying step is at least increased as the prescribed thickness of the tantalum oxide layer 5 increases. For example, in the tantalum oxide layer forming step, the application step is performed a second specified number of times (e.g., once) and the drying step is performed the second specified number of times, thereby forming the material layer which will be the tantalum oxide layer 5 on the catalyst layer 4.

[0038] In the first step of the tantalum oxide layer forming step, a solution (hereinafter referred to as a second solution) including a tantalum compound which will be the tantalum oxide layer 5 is applied onto the catalyst layer 4 (that is, the application step is performed), and then a heat process of drying the second solution by heating under the second condition (the drying step) is performed at least once (e.g., once), thereby forming the metal layer which will be the tantalum oxide layer 5. The second solution is a solution obtained by dissolving the tantalum compound in a solvent (hereinafter referred to as a second solvent). The second solvent is, for example, liquid obtained by mixing ethylene glycol monoethyl ether and hydrochloric acid and ethanol. The tantalum compound is, for example, but not limited to, tantalum chloride, and the tantalum compound may be, for example, tantalum ethoxide. The metal concentration (tantalum concentration) of the second solution is, for example, 50 mg/L. Moreover, the application quantity of the second solution is, for example, 1 µL/cm<sup>2</sup>. The second condition includes a heat process temperature and a heat process time. The heat process temperature in the second condition is within a range of 100°C to 400°C and may be 220°C as an example. Moreover, the heat process time in the second condition is within a range of 5 minutes to 15 minutes, for example, and may be 10 minutes as an example.

**[0039]** In the second step of the tantalum oxide layer forming step, the heat process of baking the material layer under a prescribed baking condition is performed, thereby forming the tantalum oxide layer 5 (see FIG. 3D). The baking condition includes a baking temperature and a baking time. The baking temperature is within a range of 500°C to 700°C, for example, and may be 560°C as an example. The baking time is within a range of 5 minutes to 20 minutes, for example, and may be 10 minutes as an example.

**[0040]** In the manufacturing method of the electrolysis electrode 1 described above, the tantalum oxide 43 in at least one of the pores 42 in the catalyst layer 4 is formed

in the tantalum oxide layer forming step.

#### (4) Example and Comparative Examples

**[0041]** FIG. 5 is a graph of results of a durability test conducted on an electrolysis electrode 1 according to Example of the embodiment, an electrolysis electrode according to Comparative Example 1, and an electrolysis electrode 1r (see FIG. 4) according to Comparative Example 2.

[0042] The electrolysis electrode according to Comparative Example 1 is different from the electrolysis electrode 1 according to Example in that Comparative Example 1 does not include the tantalum oxide layer of the electrolysis electrode 1 according to Example. The electrolysis electrode 1r according to Comparative Example 2 includes 15 tantalum oxide layers 6 and 15 catalyst layers 7 alternately stacked one by one instead of the catalyst layer 4 and the tantalum oxide layer 5 of the electrolysis electrode 1 according to the embodiment. In FIG. 4, only three of the tantalum oxide layers 6 and only three of the catalyst layers 7 are shown. In the electrolysis electrode 1r according to Comparative Example 2, the total catalyst amount of the 15 catalyst layers 7 is the same as the catalyst amount of the electrolysis electrode 1 according to Example. The catalyst layer 7 includes platinum and iridium oxide. In the electrolysis electrode 1r according to Comparative Example 2, a composite layer including the 15 tantalum oxide layers 6 and the 15 catalyst layers 7 has a plurality of cracks.

[0043] The durability test is accelerated testing. The durability test was performed in which two electrolysis electrodes 1 (or two electrolysis electrodes or two electrolysis electrodes 1r) formed under the same condition were adopted as pair of electrodes, and the pair of electrodes were immersed in salt water in an electrolytic bath in a durability test facility. In the durability test, polarity reversal was performed each time the pair of electrodes are energized for a predetermined time (3 minutes). In this case, the polarity reversal means that a combination of the anode and the cathode in the pair of electrodes is reversed. In other words, the polarity reversal means that an electrode of the pair of electrodes which is on a highpotential side is changed such that an electrode used as the anode and an electrode used as the cathode are respectively used as the cathode and the anode. The electrolytic bath in the durability test facility has a water inlet and a water outlet for salt water. In the durability test, salt water is added so that the electric conductivity of the salt water in the electrolytic bath is  $1650\pm165~\mu\text{S/cm}$ . Moreover, in the durability test, the electrolytic bath in the durability test facility is drained while tap water is constantly supplied to the electrolytic bath at a flow rate of 2 L/min. The salt water supplied to the electrolytic bath in the durability test facility is a sodium chloride solution obtained by dissolving salt (sodium chloride) in tap water. The current value of an energization current in the durability test is 400 mA. For measuring the hypochlorous acid water

concentration, the electrodes were taken out of the electrolytic bath in the durability test facility at the time of measuring the hypochlorous acid water concentration, and the hypochlorous acid water concentration was measured as described below. As salt water in an electrolytic bath for measuring a hypochlorous acid water concentration, salt water produced by dissolving 4.5 g salt (sodium chloride) in 800 mL pure water was used. The current value of an energization current for measuring the hypochlorous acid water concentration is 400 mA. Moreover, in initial aging, the polarity reversal was performed each time the pair of electrodes were energized for a predetermined time (3 minutes), and in this way, the pair of electrodes were energized for a total of 12 minutes. After the initial aging, electrolysis was performed for 12 minutes under the same condition as the initial aging, and then, some of the electrolysis water was taken out every 3 minutes, and the hypochlorous acid water concentration was measured. For the hypochlorous acid water concentration, the free chlorine concentration (HOCl, OCl-) was measured by a pocket residual chlorine meter (HACH, Pocket Colorimeter II 58700-00) based on the DPD method. In this case, the polarity reversal means that a combination of the anode and the cathode in the pair of electrodes is reversed. In other words, the polarity reversal means that an electrode of the pair of electrodes which is on a high-potential side is changed such that an electrode used as the anode and an electrode used as the cathode are respectively used as the cathode and the anode.

**[0044]** The abscissa in FIG. 5 represents a durability test time (elapsed time). The ordinate in FIG. 5 represents the hypochlorous acid water concentration measured after the energization for a unit time (3 minutes) was performed at the time. In this case, chlorine produced in the vicinity of the anode contributes to production of hypochlorous acid, and therefore, the hypochlorous acid water concentration is substantially determined based on the amount of the chlorine produced per unit time.

[0045] From FIG. 5, it can be seen that in the electrolysis electrode 1 according to Example, the hypochlorous acid water concentration is higher and a time until the hypochlorous acid water concentration decreases to or less than a prescribed value (e.g., 5 mg/L) is longer (the durability is improved more) than in the electrolysis electrode according to Comparative Example 1 and the electrolysis electrode 1r according to Comparative Example 2. Note that the durability is determined based on elution caused by consumption of the catalyst layer 4, peel-off of the catalyst layer 4, or the like. In the electrolysis electrode 1r according to Comparative Example 2, the tantalum oxide layers 6 and the catalyst layers 7 are alternately stacked, and therefore, a conduction path and a path of gas are narrow, the amount of chlorine produced per unit time is small, and the number of active points not used is large, and therefore, the electrolysis electrode 1r is presumed to have reduced service life. In the electrolysis electrode according to Comparative Example 1,

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chlorine is more easily produced than in the electrolysis electrode 1r according to Comparative Example 2, but the catalyst is more likely to desorb than in the electrolysis electrode 1 according to Example, and therefore, the electrolysis electrode according to Comparative Example 1 is presumed to have shorter service life than the electrolysis electrode 1 according to Example. In other words, it can be seen from FIG. 5 that the electrolysis electrode 1 according to Example is capable of producing a larger amount of chlorine and thus has a longer service life than the electrolysis electrode according to Comparative Example 1 and the electrolysis electrode 1r according to Comparative Example 2.

#### (5) Effects

[0046] The electrolysis electrode 1 according to the embodiment includes the tantalum oxide layer 5 provided on the catalyst layer 4 including platinum and iridium oxide, and the catalyst layer 4 is partially exposed, which enables the durability to be improved. This enables the electrolysis electrode 1 according to the embodiment to make the catalyst layer 4 contribute to production of chlorine and to have improved durability compared to the case where the entirety of the principal surface 40 of the catalyst layer 4 is in contact with salt water. The electrolysis electrode 1 according to the embodiment includes the tantalum oxide layer 5 and the tantalum oxide 43, which enables platinum iridium to be suppressed from being excessively consumed (eluted) from the catalyst layer 4 during use, thereby suppressing a rapid structural change in the catalyst layer 4, and partial desorption of the catalyst layer 4 and peel-off of the catalyst layer 4 can be suppressed. Moreover, in the electrolysis electrode 1 according to the embodiment, the agglomeration of iridium can be suppressed.

**[0047]** Further, the electrolysis electrode 1 according to the embodiment includes the tantalum oxide 43 which is provided in the plurality of pores 42 in the catalyst layer 4 and which is in contact with the catalyst layer 4, which enables the mechanical intensity of the catalyst layer 4 to be improved and enables excessive consumption of iridium oxide, agglomeration of the iridium oxide, and the like to be suppressed.

**[0048]** The embodiment is a mere example of various embodiments of the present disclosure. Various modifications may be made to the embodiment depending on design and the like as long as the object of the present disclosure is achieved.

**[0049]** For example, the shape of the conductive substrate 2 in plan view is not limited to a rectangular shape, but may be, for example, a square shape.

**[0050]** Moreover, the catalyst layer 4 is not limited to a porous layer but may be a non-porous layer.

**[0051]** Further, the plurality of recesses 45 may have the same shape. In this case, for example, in the manufacturing method of the electrolysis electrode 1, the plurality of recess 45 may be formed by an etching tech-

nique, a laser processing technique, or the like. Using these techniques provides the advantages that the degree of freedom of design in terms of the layout and the size of the plurality of recesses 45 is increased and the reproducibility of formation locations of the plurality of recesses 45 is increased.

**[0052]** Moreover, in the electrolysis electrode 1, the catalyst layer 4 does not have to have the plurality of recesses 45, and in this case, for example, the tantalum oxide layer 5 has at least a plurality of holes (e.g., pin holes or cracks) through which the principal surface 40 of the catalyst layer 4 is partially exposed.

[0053] Moreover, in the electrolysis electrode 1, even in the case of the catalyst layer 4 having the plurality of recesses 45, the tantalum oxide layer 5 may have a plurality of cracks through which the catalyst layer 4 is partially exposed. In the manufacturing method of the electrolysis electrode 1 described above, if the thickness of the tantalum oxide layer 5 is greater than or equal to 50 nm, cracks through which the catalyst layer 4 is partially exposed may be formed in the tantalum oxide layer 5 in the second step of the tantalum oxide layer forming step. Moreover, in the manufacturing method of the electrolysis electrode 1 described above, cracks may be formed in the tantalum oxide layer in the second step of the tantalum oxide layer forming step, and in addition, cracks continuous with the cracks in the tantalum oxide layer may be formed in the catalyst layer 4. The plurality of holes in the tantalum oxide layer 5 may be formed by an etching technique, a laser processing technique, or the like.

**[0054]** The electrolysis electrode 1 may include a titanium oxide layer provided between the conductive substrate 2 and the intermediate layer 3.

**[0055]** The tantalum oxide layer 5 may include tantalum in addition to the tantalum oxide. In other words, the tantalum oxide layer 5 may be a layer in which tantalum oxide and tantalum are included.

**[0056]** Moreover, the electrolysis electrode 1 may further include, on the second principal surface 22 of the conductive substrate 2, a structural component similar to a structural component including the intermediate layer 3, the catalyst layer 4, and the tantalum oxide layer 5 at the side of the first principal surface 21.

(Summary)

**[0057]** The embodiment and the like described above disclose the following aspects in the present specification.

[0058] An electrolysis electrode (1) of a first aspect includes a conductive substrate (2), a catalyst layer (4), and a tantalum oxide layer (5). The conductive substrate (2) includes at least titanium. The catalyst layer (4) is provided on the conductive substrate (2). The catalyst layer (4) includes platinum and iridium oxide. The tantalum oxide layer (5) is provided on the catalyst layer (4). In the electrolysis electrode (1), the catalyst layer (4) is

partially exposed.

**[0059]** The electrolysis electrode (1) of the first aspect has improved durability.

**[0060]** In an electrolysis electrode (1) of a second aspect referring to the first aspect, the catalyst layer (4) is a porous layer including: a plurality of composite particles (41) each including platinum (a platinum particle 411) and iridium oxide (iridium oxide particles 412); and a plurality of pores (42). The electrolysis electrode (1) further includes tantalum oxide (43) provided in at least one pore (42) of the plurality of pores (42), and the tantalum oxide (43) is in contact with the catalyst layer (4).

**[0061]** The electrolysis electrode (1) according to the second aspect has improved production efficiency of chlorine with improved durability.

[0062] In an electrolysis electrode (1) of a third aspect referring to the first or second aspect, the catalyst layer (4) has a plurality of recesses (45) recessed from a principal surface (40) on an opposite side of the catalyst layer (4) from the conductive substrate (2). The tantalum oxide layer (5) has: a first portion (51) provided on the principal surface (40) of the catalyst layer (4); and a second portion (52) provided on an inner surface (451) of at least one recess (45) of the plurality of recesses (45) in the catalyst layer (4).

**[0063]** The electrolysis electrode (1) according to the third aspect has improved production efficiency of chlorine with improved durability.

**[0064]** In an electrolysis electrode (1) of a fourth aspect referring to the third aspect, the catalyst layer (4) is partially exposed in the plurality of recesses (45).

**[0065]** In the electrolysis electrode (1) of the fourth aspect, salt water easily enters the catalyst layer (4) in an in-plane direction of the catalyst layer (4) through the inner surface (451) of the at least one recess (45), the inner surface (451) being exposed through the at least one recess (45). Thus, it is presumed that in the electrolysis electrode (1) of the fourth aspect, the catalyst layer (4) readily contributes to production of chlorine, which enables the durability to be improved.

**[0066]** An electrolysis electrode (1) of a fifth aspect referring to any one of the first to fourth aspects further includes an intermediate layer (3). The intermediate layer (3) is provided between the conductive substrate (2) and the catalyst layer (4). The intermediate layer (3) includes platinum.

**[0067]** In the electrolysis electrode (1) of the fifth aspect, peel-off of a plurality of catalyst layers (4) is suppressed, which enables the durability to be improved.

**[0068]** In an electrolysis electrode (1) of a sixth aspect referring to the fifth aspect, the conductive substrate (2) has a principal surface (a first principal surface 21) facing the catalyst layer (4), and the principal surface is a rough surface.

**[0069]** In the electrolysis electrode (1) of the sixth aspect, adhesiveness between the conductive substrate (2) and the intermediate layer (3) is improved, which enables the catalyst layer (4) to be suppressed from peeling off

from a side of the conductive substrate (2), thereby improving the durability.

#### **Reference Signs List**

#### [0070]

- 1 Electrolysis Electrode
- 2 Conductive Substrate
- 21 First Principal Surface
  - 3 Intermediate Layer
  - 4 Catalyst Layer
  - 40 Principal Surface
  - 41 Composite Particle
- 411 Platinum Particle
- 412 Iridium Oxide Particle
- 42 Pore
- 43 Tantalum Oxide
- 45 Recess
- 9 451 Inner Surface
  - 5 Tantalum Oxide Layer
  - 51 First Portion
  - 52 Second Portion

#### Claims

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1. An electrolysis electrode, comprising:

a conductive substrate including at least titani-

a catalyst layer provided on the conductive substrate and including platinum and iridium oxide; and

a tantalum oxide layer provided on the catalyst layer.

the catalyst layer being partially exposed.

2. The electrolysis electrode of claim 1, wherein

the catalyst layer is a porous layer including

a plurality of composite particles each including platinum and iridium oxide and a plurality of pores, and

the electrolysis electrode further includes tantalum oxide provided in at least one pore of the plurality of pores, the tantalum oxide being in contact with the catalyst layer.

**3.** The electrolysis electrode of claim 1 or 2, wherein

the catalyst layer has a plurality of recesses recessed from a principal surface on an opposite side of the catalyst layer from the conductive

the tantalum oxide layer has

a first portion provided on the principal surface in the catalyst layer and a second portion provided on an inner surface of at least one recess of the plurality of recesses in the catalyst layer.

**4.** The electrolysis electrode of claim 3, wherein the catalyst layer is partially exposed in the plurality of recesses.

5. The electrolysis electrode of any one of claims 1 to 4, further comprising an intermediate layer provided between the conductive substrate and the catalyst layer, the intermediate layer including platinum.

**6.** The electrolysis electrode of claim 5, wherein the conductive substrate has a principal surface facing the catalyst layer, the principal surface being a rough surface.

FIG. 1 A

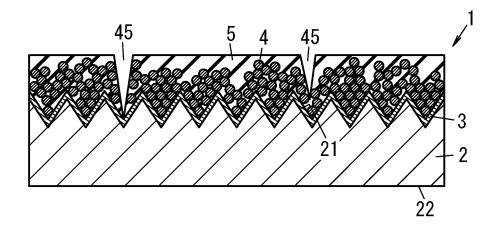
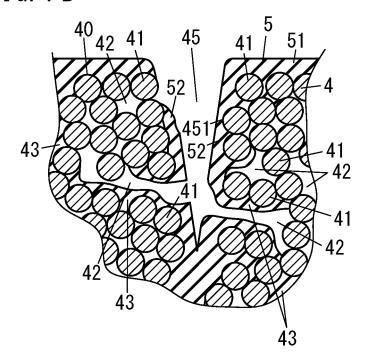


FIG. 1B





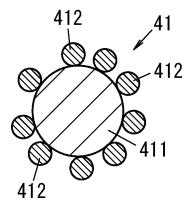


FIG. 3A

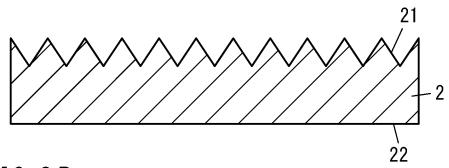


FIG. 3B

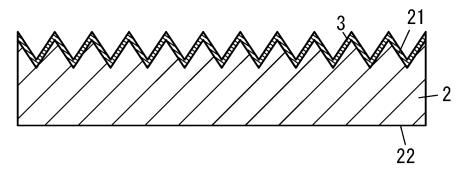


FIG. 3C

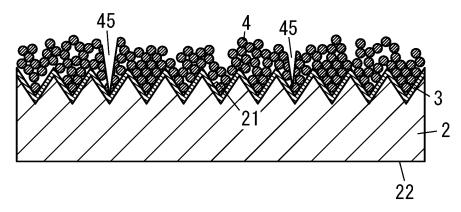
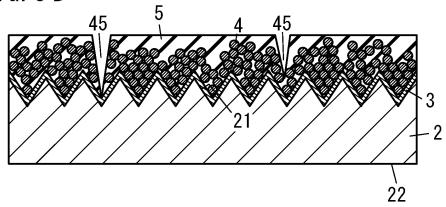
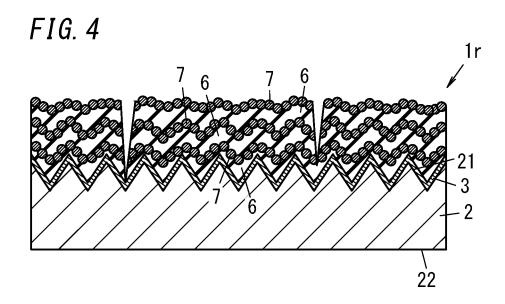
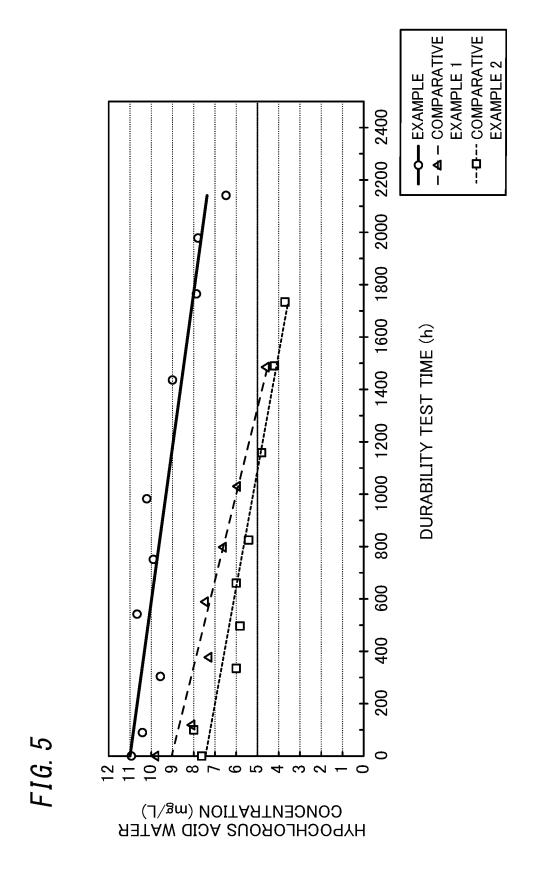


FIG. 3D







5 INTERNATIONAL SEARCH REPORT International application No. PCT/JP2020/035612 A. CLASSIFICATION OF SUBJECT MATTER Int.Cl. C25B11/04(2021.01)i, C25B11/03(2021.01)i FI: C25B11/08A, C25B11/04A, C25B11/03 10 According to International Patent Classification (IPC) or to both national classification and IPC FIELDS SEARCHED Minimum documentation searched (classification system followed by classification symbols) Int.Cl. C25B11/08, C25B11/03, C25B11/04 15 Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched Published examined utility model applications of Japan 1922-1996 Published unexamined utility model applications of Japan 1971-2020 Registered utility model specifications of Japan 1996-2020 Published registered utility model applications of Japan 1994-2020 20 Electronic data base consulted during the international search (name of data base and, where practicable, search terms used) C. DOCUMENTS CONSIDERED TO BE RELEVANT Category\* Citation of document, with indication, where appropriate, of the relevant passages Relevant to claim No. 25 JP 2018-111874 A (KAIGEN PHARMA CO., LTD.) 19 July 2018 1-3, 5-6 Χ Υ (2018-07-19), claims, paragraphs [0001]-[0059], fig. 2-3, 5-6 Α 1, 2 JP 2004-323955 A (WAKO SANGYO KK) 18 November 2004 2-3, 5-6 Υ 30 (2004-11-18), claim 5, paragraph [0010] 35 40  $\bowtie$ See patent family annex. Further documents are listed in the continuation of Box C. Special categories of cited documents: 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 document defining the general state of the art which is not considered to be of particular relevance "A" "E" earlier application or patent but published on or after the international document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive filing date step when the document is taken alone 45 "L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified) document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art document referring to an oral disclosure, use, exhibition or other means document published prior to the international filing date but later than document member of the same patent family the priority date claimed Date of mailing of the international search report Date of the actual completion of the international search 50 09 November 2020 17 November 2020 Name and mailing address of the ISA/ Authorized officer Japan Patent Office 3-4-3, Kasumigaseki, Chiyoda-ku, Telephone No. Tokyo 100-8915, Japan 55

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