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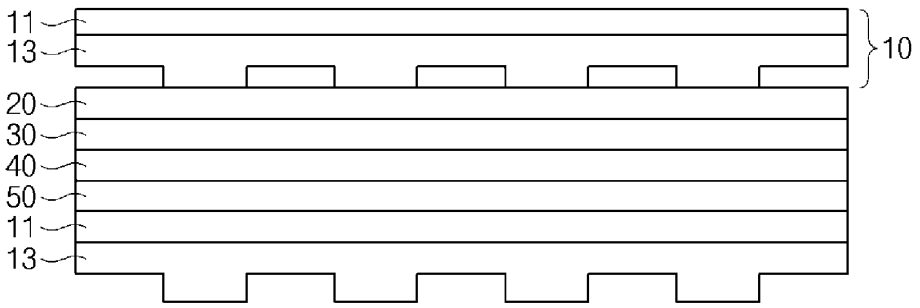
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(54) SEPARATOR PLATE AND ELECTROLYSIS APPARATUS INCLUDING SAME

(57) The present invention relates to a separator plate including a clad metal plate containing a titanium layer and a stainless-steel layer bonded to one side of the titanium layer, and a titanium porous diffusion layer disposed on the other side of the titanium layer, wherein the stainless-steel layer includes a flow path to reduce difficulty in processing the flow path, and the thickness of the separator plate is reduced to decrease electrical resistance and improve electrolysis efficiency.

[FIG. 1]



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Description

TECHNICAL FIELD

[0001] The present invention relates to a separator plate and an electrolysis device including the same.

BACKGROUND ART

[0002] Carbon dioxide is a greenhouse gas causing global warming, which needs to be reduced. As the method of reducing carbon dioxide, capturing, chemical conversion or electrochemical conversion, or the like are widely known. Among these methods, the electrochemical conversion may be more economically advantageous than simply removing carbon dioxide as it allows precise adjustment of the ingredients so as to prepare another synthetic gas. In addition, by electrolyzing carbon dioxide with water, an organic material such as carbon monoxide, ethylene, methane, formic acid, formate, various hydrocarbon and aldehyde, or alcohol may be obtained.

[0003] The process by which carbon dioxide electrochemically decomposes or is reduced is classified according to the ion exchange method. In case of using a cation exchange membrane among the methods, when an anode is supplied with water and applied with a current, water decomposes in the anode to generate oxygen, electrons, and hydrogen ions, and the hydrogen ions move to a cathode through the cation exchange membrane. In the cathode, carbon dioxide reacts with the electrons, generated in the anode, and the hydrogen ions having passed through the cation exchange membrane to cause a reduction reaction, and is converted into another material.

[0004] In addition, in case of using an anion exchange membrane, since the activity of electrochemical reaction improves in a strong alkaline atmosphere, a KOH solution at a certain concentration is generally used as an electrolyte solution. When carbon dioxide and water are supplied together and a current is applied to the cathode, hydroxide ions (OH^-) are generated, and the hydroxide ions pass through the anion exchange membrane and move to the anode. The moved hydroxide ions are converted into water and oxygen in the anode, and in this process, electrons are generated. The electrons move to the cathode along a conducting wire, and the moved electrons react with carbon dioxide and water in the cathode, and decompose into carbon monoxide and hydroxide ions.

[0005] Through the above process, electrochemical decomposition reaction of carbon dioxide is complete. At this time, the water supplied with carbon dioxide reacts with the moved electrons, irrelevantly to the reaction of generating carbon monoxide, and is electrolyzed, thereby generating hydrogen gas as well as generating hydroxide ions. This reaction of water and electrons may be in a competing reaction relationship with the reaction of generating carbon monoxide. Since the above reactions are electrochemical reactions, the amount of generating carbon monoxide and the ratio of hydrogen/carbon dioxide may be easily adjusted by adjusting the voltage.

[0006] Meanwhile, a carbon dioxide electrolysis device is manufactured by stacking unit cells, and at this time, a separator plate having a bipolar surface, which is disposed between a plurality of membrane electrode assemblies, is required. In a conventional separator plate, a different type of flow path was formed on each side of the bipolar surface according to the purpose, and therefore, a processing process is complex, and there was difficulty selecting a material satisfying such processibility. In addition, a titanium material was used for a surface of the separator plate facing the anode due to corrosive environment, and in this case, the surface was bent or denatured by heat during the processing process of the separator plate, so that a thick separator plate was inevitably used. However, in case of setting the large thickness for the separator plate, there was a problem of increasing electrical resistance and reducing electrolysis efficiency.

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DISCLOSURE OF THE INVENTION

TECHNICAL PROBLEM

[0007] A task of the present invention is to provide a separator plate including a clad metal plate containing a titanium layer and a stainless-steel layer, and a titanium porous diffusion layer on a surface of the titanium layer, thereby decreasing the thickness to reduce electrical resistance, reducing difficulty of processing a flow path, and improving electrolysis efficiency, and to provide an electrolysis device including the same.

TECHNICAL SOLUTION

[0008] The present invention provides a separator plate and an electrolysis device including the same.

(1) The present invention provides a separator plate including a clad metal plate containing a titanium layer and a

stainless-steel layer bonded to one side of the titanium layer, and a titanium porous diffusion layer disposed on the other side of the titanium layer, wherein the stainless-steel layer includes a flow path.

(2) The present invention provides the separator plate of (1) above, wherein the titanium porous diffusion layer is a titanium felt.

(3) The present invention provides the separator plate of (1) or (2) above, wherein the clad metal plate is formed of the titanium layer and the stainless-steel layer as one body by forced rolling.

(4) The present invention provides the separator plate of any one of (1) to (3) above, wherein the clad metal plate has a thickness of 0.5 mm or greater and 1.5 mm or less.

(5) The present invention provides the separator plate of any one of (1) to (4) above, wherein a ratio of a thickness of the titanium layer and a thickness of the stainless-steel layer is 1:5 to 20.

(6) The present invention provides the separator plate of any one of (1) to (5) above, wherein the titanium porous diffusion layer has a thickness of 0.6 mm or greater and 0.8 mm or less.

(7) The present invention provides the separator plate of any one of (1) to (6) above, wherein the titanium layer and the titanium porous diffusion layer include no flow path.

(8) The present invention provides the separator plate of any one of (1) to (7) above, wherein the separator plate has an area of 100 cm^2 or greater and 5000 cm^2 or less.

(9) The present invention provides an electrolysis device including a membrane electrode assembly in which an anode, a separator, a cathode, and a gas diffusion layer are disposed in sequence, the separator plate according to any one of (1) to (8) above disposed between the plurality of membrane electrode assemblies, a first gasket disposed between the anode and the separator plate, and a second gasket disposed between the cathode and the separator plate, wherein a thickness of the first gasket is larger than a thickness of the second gasket.

(10) The present invention provides the electrolysis device of (9) above, wherein a ratio of the thickness of the first gasket and the thickness of the second gasket is 1:0.2 to 0.3.

(11) The present invention provides the electrolysis device of (9) or (10) above, wherein the membrane electrode assembly further includes a porous layer on one side of the anode on which the separator is not disposed.

(12) The present invention provides the electrolysis device of (11) above, wherein the first gasket is disposed surrounding side surfaces of the porous layer and the titanium porous diffusion layer, and the second gasket is disposed surrounding a side surface of the gas diffusion layer.

(13) The present invention provides the electrolysis device of any one of (10) to (12) above, wherein the electrolysis device electrolyzes carbon dioxide to generate one or more materials selected from the group consisting of carbon monoxide, ethylene, methane, formic acid, hydrocarbon, aldehyde, and alcohol.

ADVANTAGEOUS EFFECTS

[0009] According to the separator plate of the present invention, by decreasing the thickness of the separator plate, as well as securing the current-collecting area and the cross-sectional area of a flow path, to reduce electrical resistance, electrolysis efficiency may be improved when the separator plate is applied to an electrolysis device.

[0010] In addition, according to the separator plate of the present invention, since a separate flow path is not formed on a surface facing a cathode, complication in flow path processing may be reduced, and at the same time, material and processing costs may be reduced.

[0011] In addition, according to the electrolysis device of the present invention, by applying gaskets with different thicknesses, a gap between the separator plate and the membrane electrode assembly may be reduced, thereby decreasing the electrical resistance and improving the electrolysis efficiency.

BRIEF DESCRIPTION OF THE DRAWINGS

[0012]

FIG. 1 is a side view of a separator plate of the present invention.

FIG. 2 is a graph showing resistance values measured according to Example 1 and Comparative Example 1.

FIG. 3 is a table listing the values of measuring carbon monoxide faraday efficiency, hydrogen faraday efficiency, carbon dioxide conversion rate, and overvoltage according to Example 1 and Comparative Example 2.

FIG. 4 is a side view of a separator plate of the present invention including gaskets.

MODE FOR CARRYING OUT THE INVENTION

[0013] Hereinafter, the present invention will be described in more detail in order to help understanding of the present invention. At this time, it will be understood that terms or words used in the specification and claims shall not

be interpreted as the meaning defined in commonly used dictionaries, and it will be further understood that the terms or words should be interpreted as having a meaning that is consistent with their meaning in the context of the relevant art and the technical idea of the invention, based on the principle that an inventor may properly define the meaning of the terms or words to best explain the invention.

[0014] The terms used in this specification are only used to illustrate example embodiments, and are not intended to limit the present invention. The singular forms are intended to include the plural forms as well unless the context clearly indicates otherwise.

[0015] It will be further understood that the terms 'include', 'provide', 'have' or the like, when used in this specification, specify the presence of stated features, integers, steps, elements, or a combination thereof, but do not preclude the presence or addition of one or more other features, integers, steps, elements, or a combination thereof.

Separator Plate

[0016] The present invention provides a separator plate including a clad metal plate 10 containing a titanium layer 11 and a stainless-steel layer 13 bonded to one side of the titanium layer 11, and a titanium porous diffusion layer 50 disposed on the other side of the titanium layer 11, wherein the stainless-steel layer includes a flow path.

[0017] The separator plate included in a carbon dioxide electrolysis device of the present invention may not only serve to supply or separate a reaction gas, but also serve to physically support a membrane electrode assembly 30 and a gas diffusion layer when a cell is connected, and may also serve to release products, and control heat inside the cell through electrical conductivity and electrochemical reaction.

[0018] A separator plate included in a conventional electrochemical conversion device was manufactured by stacking separate metal plates each of which had a flow path formed individually. For example, a metal plate in which a cathode flow path is formed and a metal plate in which an anode flow path is formed may be sequentially stacked, and in some cases, a separator plate may be further disposed between the metal plates. In this case, the stacking of the metal plates may be performed in the method such as laser welding, brazing, resistance spot welding, or adhesion with an adhesive, in such methods, the separator plate was manufactured through a separate bonding process for each metal plate, which was highly complex due to the manufacturing process, causing a problem of increasing cost and time. Also, in the case that the metal plates were simply stacked as above, electrical resistance on the stacked interface increased, causing a problem of decreasing efficiency of the electrochemical conversion device. In addition, the total thickness of the stacked separator plate becomes large, which may also cause increased electrical resistance.

[0019] To solve the above problems, the separator plate of the present invention includes the clad metal plate 10. In particular, the clad metal plate 10 may be formed of the titanium layer 11 and the stainless-steel layer 13 as one body by forced rolling. The clad metal plate 10 refers to a plate including a material made by joining dissimilar metals through atomic diffusion to form a single metal, and for example, by rolling a titanium metal and a stainless-steel metal under strong pressure, the metal tissues penetrate each other, destroying each other's tissues, thereby becoming the plate including a single metal having stabilized tissues on the whole. Accordingly, the separator plate including the clad metal plate 10 of the present invention may correspond to a material different from a conventional separator plate in which two metal plates are bonded or stacked simply by bonding, welding, or the like, and since the clad metal plate 10 of the present invention is not detached even through it is left for a long time and the adhesion becomes rather stronger, the clad metal plate 10 may be formed in a very small thickness, compared to the separator plate of two or more separate metal plates being stacked. In addition, due to dissimilar joining of the layers, an electrolysis cell may be constituted only with a single separator plate even for a bipolar (cathode and anode) environment, so that the interface resistance between materials of the electrical conversion device may be reduced, thereby improving the efficiency in electrical conversion.

[0020] The clad metal plate may include a titanium layer 11 and a stainless-steel layer 13, the titanium layer 11 may face an anode layer when applied to the electrolysis device, and the stainless-steel layer 13 may face a cathode layer. In the anode layer, hydroxide ions generate water and oxygen, which corresponds to a moisture environment, so that titanium, resistant to corrosion, is preferred.

[0021] According to an embodiment of the present invention, the titanium porous diffusion layer 50 may be a titanium felt, and the titanium layer 11 and the titanium porous diffusion layer 50 may not include a flow path.

[0022] A separator plate, used for a conventional electrochemical conversion device such as an electrolyzer or a fuel cell, may include a plurality of flow paths each on a surface facing the anode and a surface facing the cathode, and the flow path may transfer various fluids including carbon dioxide, and diffuse reaction materials to a catalyst layer. In addition, in the flow path, an oxygen-generating reaction of the anode may occur, and a carbon dioxide-reduction reaction of the cathode may occur. However, in order to form the flow path, the manufacturing was carried out in a method of processing such that both the flow path and a supply manifold were included in the original plate, and in particular, the flow path had to be formed through sheet forming, stamping, or etching. Accordingly, there was difficulty

in forming the flow path while maintaining the small thickness of the separator plate, a constant current-collecting area and cross-sectional area of the flow path was difficult to form due to the process characteristics of the sheet forming, etching, or the like, and a phenomenon of bending occurred in the original plate, due to heat applied to the original plate. Such phenomenon may result in imbalanced transfer of the materials through the flow path, or result in an increase of electrical resistance, thereby causing a problem of reducing electrolysis efficiency.

[0023] Therefore, in order to maintain smooth transfer of the materials, while maintaining the small thickness of the separator plate, and to reduce electrical resistance, the present invention solved the problem by positioning the titanium porous diffusion layer 50 on the titanium layer 11 without forming a separate flow path. The titanium porous diffusion layer 50 may be a titanium felt, and the titanium porous diffusion layer 50 may improve the contact force and reduce the gap between the separator plate and the membrane electrode assembly 30 when the electrolysis device is connected, thereby reducing processing errors that occur when the electrolysis device is processed. In addition, the titanium porous diffusion layer 50 may serve as a flow path of the anode, and at the same time, may be well in contact with the catalyst layer, serving to reduce battery resistance and overvoltage. In addition, since a separate flow path is not formed in the titanium layer, costs and processing time, associated with a process of forming the flow path, may be reduced.

[0024] In addition, in the separator plate of the present invention, the titanium porous diffusion layer is positioned on the titanium layer without a separate flow path, the stainless-steel layer 13 includes a flow path on a surface not bonded to the titanium layer 11. The flow path included in the stainless-steel layer 13 is a layer facing the cathode, which may have a serpentine or interdigitated-type structure, and in the flow path of the above structure, a rotating section is present, so that the flow rate at which fluid is transferred may increase, and products may thus be easily released, and since the fluid is evenly distributed, larger amounts of reactants may react with the catalyst layer. If the flow path is not formed in the stainless-steel layer, the material-transfer resistance may increase, resulting in local depletion of carbon dioxide. In addition, since a separate flow path is included in the stainless-steel layer, the supplied carbon dioxide may be evenly distributed in the entire active area.

[0025] According to an embodiment of the present invention, the clad metal plate 10 may have a thickness of 0.5 mm or greater and 1.5 mm or less. For example, the thickness of the clad metal plate 10 may be 0.50 mm or greater, 0.52 mm or greater, 0.55 mm or greater, 0.57 mm or greater, 0.59 mm or greater, 0.60 mm or greater, 0.63 mm or greater, 0.65 mm or greater, 0.67 mm or greater, 0.69 mm or greater, 0.70 mm or greater, 1.5 mm or less, 1.45 mm or less, 1.4 mm or less, 1.35 mm or less, 1.3 mm or less, 1.25 mm or less, 1.2 mm or less, 1.15 mm or less, 1.1 mm or less, 1.05 mm or less, 1.0 mm or less, 0.97 mm or less, 0.95 mm or less, 0.93 mm or less, 0.90 mm or less, 0.87 mm or less, 0.85 mm or less, 0.83 mm or less, 0.80 mm or less, 0.77 mm or less, 0.75 mm or less, 0.73 mm or less, and 0.71 mm or less. In particular, the thickness of the clad metal plate 10 may be 0.6 mm or greater, and 0.8 mm or less, and when the thickness of the clad metal plate 10 falls within the above ranges, excellent durability and physical strength may be maintained, and at the same time, the small thickness may be formed, so that the connection may be improved when the electrolysis device is manufactured, and accordingly, the interface resistance between the components in the device may be reduced, thereby improving the electrolysis efficiency.

[0026] According to an embodiment of the present invention, the ratio of the thickness of the titanium layer 11 and the thickness of the stainless-steel layer 13 may be 1:5 to 20. For example, the ratio of the thickness of the titanium layer 11 and the thickness of the stainless-steel layer 13 may be 1:5 or greater, 5.5 or greater, 5.7 or greater, 5.9 or greater, 6 or greater, 6.3 or greater, 6.5 or greater, 6.7 or greater, 7.0 or greater, 20 or less, 19.5 or less, 19 or less, 18.5 or less, 18 or less, 17.5 or less, 17 or less, 16.5 or less, 16 or less, 15.5 or less, and 15 or less. In particular, the ratio may be 1:5 to 19, and when the ratio of the thickness of the titanium layer 11 and the thickness of the stainless-steel layer 13 falls within the above ranges, material cost, and costs for manufacturing the separator plate and processing the flow path may be reduced, and at the same time, corrosion resistance of the separator plate may be improved, and overvoltage may be reduced, thereby improving the electrolysis efficiency.

[0027] In particular, the thickness of the titanium layer 11 may be 0.03 mm or greater, 0.04 mm or greater, 0.05 mm or greater, 0.15 mm or less, 0.14 mm or less, 0.13 mm or less, 0.11 mm or less, 0.1 mm or less, 0.09 mm or less, 0.08 mm or less, 0.07 mm or less, 0.06 mm or less, and the thickness of the stainless-steel layer 13 may be 0.5 mm or greater, 0.51 mm or greater, 0.53 mm or greater, 0.55 mm or greater, 0.57 mm or greater, 0.59 mm or greater, 1.45 mm or less, 1.4 mm or less, 1.35 mm or less, 1.3 mm or less, 1.25 mm or less, 1.2 mm or less, 1.15 mm or less, 1.1 mm or less, 1.05 mm or less, 1.0 mm or less, 0.95 mm or less, 0.9 mm or less, 0.85 mm or less, 0.8 mm or less, 0.75 mm or less, 0.7 mm or less, 0.65 mm or less, and 0.6 mm or less. In the separator plate of the present invention, the thicknesses of the titanium layer 11 and the stainless-steel layer 13 are adjusted, within the respective ranges, to the thickness ratio of the titanium layer 11 and the stainless-steel layer 13 previously described, thereby maintaining excellent physical durability and strength, and at the same time, reducing the costs and improving resistance characteristics.

[0028] According to an embodiment of the present invention, the titanium porous diffusion layer 50 may have a thickness of 0.6 mm or greater and 0.8 mm or less. For example, the thickness of the titanium porous diffusion layer 50

may be 0.6 mm or greater, 0.62 mm or greater, 0.64 mm or greater, 0.65 mm or greater, 0.67 mm or greater, 0.69 mm or greater, 0.7 mm or greater, 0.8 mm or less, 0.78 mm or less, 0.75 mm or less, 0.73 mm or less, and 0.72 mm or less. In particular, the thickness may be 0.6 mm or greater, and 0.7 or less, when the thickness of the titanium porous diffusion layer 50 falls within the above ranges, the titanium porous diffusion layer may take on the role of flow path without a separate flow path being formed on the titanium layer 11, and oxygen, generated by reaction, may be effectively released through an optimal space area, thereby reducing the overvoltage. In addition, due to excellent strength of titanium, it is highly difficult to perform processing and etching for forming a flow path on the titanium layer 11 of the separator plate, and the cost and time is required, but the titanium porous diffusion layer 50 in the above thickness may be an excellent substitute for the flow path when it is in contact with the titanium layer 11, thus the supply and release of the fluid may be carried out smoothly, thereby reducing the overvoltage and improving the electrolysis efficiency.

[0029] According to an embodiment of the present invention, the area of the separator plate may be 100 cm^2 or greater, and 5000 cm^2 or less. For example, the area of the separator plate may be 100 cm^2 or greater, 200 cm^2 or greater, 300 cm^2 or greater, 500 cm^2 or greater, 700 cm^2 or greater, 1000 cm^2 or greater, 1200 cm^2 or greater, 1500 cm^2 or greater, 1700 cm^2 or greater, 2000 cm^2 or greater, 5000 cm^2 or less, 4500 cm^2 or less, 4000 cm^2 or less, 3500 cm^2 or less, 3000 cm^2 or less, and 2500 cm^2 or less.

[0030] Since a fuel cell is actually used for a vehicle, etc., it is difficult to form the fuel cell as a large-area electrochemical cell or stack in consideration of the specification when it is used, but the cell or stack included in the electrolysis device may be formed in large area in order to increase the conversion amount of carbon dioxide or water per unit time. At this time, in case of the electrolysis cell or stack, it is important to maintain physical/chemical durability and high electrolysis efficiency as well as maintain the large area for a long time. However, as the electrode area of the cell or stack becomes larger, the area of the separator plate to be applied also increases, and in case that such large-area separator plate is manufactured by stacking separate metal plates, a required supply of a molding machine and processing difficulty may increase, and in particular, processing errors may occur during a molding for forming a flow path, and detachment may occur due to long-term exposure.

[0031] On the contrary, since the clad metal plate 10 including the titanium layer 11 and the stainless-steel layer 13 is used in the separator plate of the present invention, and the titanium porous diffusion layer 50 is included on the surface of the titanium layer 11, it may be easy to manufacture the large-area separator plate, and physical durability may be maintained and detachment may be prevented in long-term exposure, and interface resistance may be reduced due to the small thickness.

Electrolysis Device

[0032] The present invention provides an electrolysis device including a membrane electrode assembly 30 in which an anode, a separator, a cathode, and a gas diffusion layer 20 are sequentially disposed, the separator plate according to the present invention disposed between the plurality of membrane electrode assemblies, a first gasket 61 disposed between the anode and the separator plate, and a second gasket 63 disposed between the cathode and the separator plate, wherein a thickness of the first gasket 61 is larger than a thickness of the second gasket 63. In addition, the membrane electrode assembly may further include a porous layer on one side of the anode on which the separator is not disposed.

[0033] According to an embodiment of the present invention, the first gasket 61 may be disposed surrounding side surfaces of the porous layer and the titanium porous diffusion layer 50, and the second gasket 63 may be disposed surrounding a side surface of the gas diffusion layer. In particular, the first gasket 61 may be positioned on the anode side, and the second gasket 63 may be positioned on the cathode side.

[0034] When the electrolysis device, including the separator plate, operates, the gaskets may prevent reactants or products from deviating from the corresponding flow path out of the device, or prevent the reactants or products from flowing out of each flow path to be mixed with each other in the device. That is, the gaskets may serve to prevent the reactants or products from flowing to a region other than a designated flow path.

[0035] A conventional gasket to be used was manufactured as thin plates of a rubber or elastic material, and stacked on one side of the separator plate, and in this case, when the area of the gasket increases or the number of stacks increases, the gasket may be compressed unevenly, thereby causing a phenomenon of outflow of reactants or products.

[0036] The separator plate, included in the carbon dioxide electrolysis device of the present invention, may include a line-shape gasket, not a surface-shape, in order to solve the conventional limitation. The gasket of the present invention may be formed in a structure that surrounds a region including the flow path by being spaced apart from the region, and the gasket in the form of a plurality of lines may be formed in two layers, or three or more layers.

[0037] According to an embodiment of the present invention, the ratio of the thickness of the first gasket 61 and the thickness of the second gasket 63 may be 1:0.2 to 0.3. For example, the ratio of the thickness of the first gasket 61

and the thickness of the second gasket 63 may be 1:0.2 or greater, 0.21 or greater, 0.22 or greater, 0.23 or greater, 0.24 or greater, 0.3 or less, 0.29 or less, 0.28 or less, 0.27 or less, 0.26 or less, and 0.25 or less. In particular, the thickness ratio may be 1:0.25 or greater and 0.3 or less. When the thicknesses fall within the above range, it may be possible to reduce gaps between the gasket, the separator plate, and the porous layer, and the titanium porous diffusion layer 50 and the gas diffusion layer, and it may be possible to effectively prevent internal materials from flowing out. For example, the first gasket 61 may be 1.0 mm or greater, 1.1 mm or greater, 1.2 mm or greater, 1.3 mm or greater, 1.35 mm or greater, 1.5 mm or less, 1.45 mm or less, and 1.4 mm or less, and the second gasket 63 may be 0.2 mm or greater, 0.25 mm or greater, 0.3 mm or greater, 0.35 mm or greater, 0.4 mm or greater, 0.6 mm or less, 0.55 mm or less, 0.5 mm or less, and 0.45 mm or less.

[0038] The separator plate may be disposed on at least one side of the membrane electrode assembly, and in particular, may be included between the membrane electrode assemblies. In addition, in the membrane electrode assembly including the separator between the anode and the cathode, the separator plate may be closely disposed on one side or both sides of the membrane electrode assembly to form an electrolysis unit cell.

[0039] According to an embodiment of the present invention, both of the separator plate and the membrane electrode assembly may be used in an electrochemical conversion device, and the electrochemical conversion device may include a fuel cell, a device capable of producing useful chemical materials through electrochemical conversion such as water electrolysis, and a device that may be used for reduction and conversion of carbon dioxide and NOx. In particular, the electrolysis cell may be included in an electrolysis device that converts carbon dioxide to carbon monoxide and ethylene.

[0040] The electrolysis refers to decomposing a material through oxidation reduction reaction by applying direct current voltage for a decomposition reaction that does not occur itself. The anode is an oxidation electrode, which oxidizes water to generate oxygen, and at this time, hydrogen ions are generated. The hydrogen ions generated in the anode may be transferred to the cathode through the separator, and the cathode is a reduction electrode in which a reactant added to the cathode may react with electrons and hydrogen ions moved from the anode to generate a product. In addition, the separator may be disposed between the anode and the cathode. The separator may be made of an inert material not involved in the electrochemical reaction as it is, but may provide a path through which the ions may move between the anode and the cathode, and may serve to prevent a physical contact between the anode and the cathode.

[0041] In addition, the anode and the cathode of the membrane electrode assembly of the carbon dioxide electrolysis device of the present invention may each include a catalyst layer. In addition, in the cathode region, vapors, supplied together with the carbon dioxide, generate a reduction product on a surface of the cathode by the electrical reduction reaction. Therefore, the cathode may include a gas diffusion layer 20 in order to evenly supply carbon dioxide gas humidified in the cathode region. In the case that the cathode includes the gas diffusion layer 20 that is hydrophobic, the supplied carbon dioxide may be smoothly diffused, distributed, and supplied to the catalyst layer of the cathode. In addition, the hydrophobic gas diffusion layer 20 may prevent moisture condensation effectively, and therefore, the supply of carbon dioxide may continue uniformly, and at the same time, the electrolysis reaction may be smoothly carried out. In addition, the catalyst layer may have a surface in a porous structure, etc., such that gas permeation characteristics are well exhibited on the surface.

[0042] According to an embodiment of the present invention, the anode may include a catalyst active in water electrolysis, and the catalyst layer of the anode may include one or more selected from the group consisting of Pt, Au, Pd, Ir, Ag, Rh, Ru, Ni, Al, Mo, Cr, Cu, Ti, W, an alloy thereof, or a mixed metal oxide, for example, Ta_20_5 and $Ir0_2$, in order for oxygen-generating reaction. In particular, in the carbon dioxide electrolysis device of the present invention, the anode may include titanium (Ti) coated with iridium oxide (IrO_2). The titanium may include a titanium mesh 40 or a titanium paper as a porous layer. In case of using the titanium mesh 40 or paper, diffusion of materials may be carried out smoothly. Meanwhile, the titanium mesh or paper may include a plurality of pores, and the titanium mesh 40 or paper may have a porosity of about 50 to about 80%. The titanium mesh 40 or paper may have a thickness of 60 to 500 μm. When the thickness is less than 60 μm, the pores of the titanium mesh 40 or paper may become clogged after a compression process, causing performance reduction, and when the thickness is greater than 500 μm, a material transfer path of water may be longer, causing performance reduction.

[0043] In addition, since a carbon dioxide reduction reaction, occurred in the cathode, competes with a hydrogen-generating reaction, a high voltage is required for the hydrogen-generating reaction, and a catalyst which is active in the carbon dioxide reduction reaction may be included. The catalyst layer of the cathode may include one or more selected from the group consisting of Sn, a Sn alloy, Al, Au, Ag, C, Cd, Co, Cr, Cu, a Cu alloy, Ga, Hg, In, Mo, Nb, Ni, $NiCo_2O_4$, a Ni alloy, a Ni-Fe alloy, Pb, Rh, Ti, V, W, Zn, and a mixture thereof for the hydrogen-generating reaction. In particular, in the carbon dioxide electrolysis device of the present invention, the cathode may include silver (Ag).

[0044] In addition, as the separator, a cation exchange membrane (CEM) or an anion exchange membrane (AEM) may be included. In particular, the cation exchange membrane may serve as a conjunctive membrane preventing reduction materials, generated by catalysis in the cathode, from moving to the anode to be oxidized, and may serve as

a separating image impermeable to anions and permeable to cations such as hydrogen ions (H+).

[0045] In addition, in the anode, water may be oxidized to generate hydrogen ions (H^+), the hydrogen ions may move to the cathode in large amounts and saturate an active site of the catalyst where carbon dioxide is converted, causing a problem of decreasing the conversion rate of carbon dioxide, and at this time, the anion exchange membrane may decrease the amount of the hydrogen ions that move to the cathode. The anion exchange membrane may prevent deterioration in conversion performance of carbon dioxide in the cathode by blocking the movement of hydrogen ions, and may serve as a separating image permeable to the anions such as OH-, HCO_3-, and CO_3^{2-}.

[0046] In addition, as the gas diffusion layer 20, a porous body with a carbon material being used, such as carbon fiber cloth, carbon fiber felt, and carbon fiber paper, or a metal porous body composed of a thin metal plate in a net structure, such as an expanded metal and a metal mesh, may be used, and in the carbon dioxide electrolysis cell of the present invention, the carbon fiber cloth may be used for the gas diffusion layer 20.

[0047] According to an embodiment of the present invention, the electrolysis device may electrolyze carbon dioxide to generate one or more materials selected from the group consisting of carbon monoxide, ethylene, methane, formic acid, hydrocarbon, aldehyde, and alcohol.

[0048] Hereinafter, examples of the present invention will be described in detail so that those skilled in the art to which the present invention pertains may easily carry out the present invention. However, the present invention may be implemented in several different forms, and is not limited to the examples to be described herein.

<Example 1>

[0049] In the operational conditions described below, a carbon dioxide electrolysis unit cell was operated.

- Reaction current density: 200 mA/cm^2 (constant current operation)
- Reaction voltage: 1 to 4 V
- Reaction temperature: 40 °C
- Reaction pressure: 1 atm (normal pressure)
- Anode catalyst: IrO_2 on Ti mesh
- Cathode catalyst: Ag powder
- Separator: porous separator (polyethersulfone, PES)
- Electrode area: 100 cm^2
- Gas diffusion layer: Sigracet 39 BB
- Anode electrolyte: 0.25 M Cs_2CO_3 (200 ml/min)
- Cathode reactant: 40 °C humidified CO_2 gas (200 ccm)
- Separator plate material: a clad metal (thickness: 0.6 mm) made of titanium (Ti) (thickness: 0.05 mm) and stainless steel (STS) (thickness: 0.55 mm), and a titanium felt (thickness: 0.6 mm)
- Separator plate (stainless-steel metal layer) flow path structure: serpentine type
- Separator plate (titanium metal layer) flow path structure: no flow path

<Comparative Example 1>

[0050] A carbon dioxide electrolysis unit cell was operated in the same conditions as those of Example 1 above, except that a multi-layer separator plate, manufactured by stacking a titanium (Ti) metal plate (thickness: 0.05 mm) and a stainless steel (STS) metal plate (thickness: 0.55 mm) each of which had a flow path in a serpentine type formed through a mechanical processing, was applied instead of the separator plate according to Example 1.

<Comparative Example 2>

[0051] A carbon dioxide electrolysis unit cell was operated in the same conditions as those of Example 1 above using the separator plate made of the same clad metal materials as those of Example 1 above, except that both of the stainless-steel metal layer and the titanium metal layer did not include a flow path structure.

Experimental Example 1

[0052] Through the carbon dioxide electrolysis unit cells according to the example and comparative examples, products according to carbon dioxide reduction reaction were analyzed with gas chromatography. At this time, impedance (resistance), carbon monoxide faraday efficiency, and overvoltage were measured according to the measurement method in the following.

[0053] The impedance (resistance) values according to Example 1 and Comparative Example 1 above were mea-

sured and compared, and the data were shown in FIG. 1, and the carbon monoxide faraday efficiency and overvoltage values according to Examples 1 and 2, and Comparative Examples 2 and 3 above were measured and compared, and the data were shown in FIG. 2.

(1) CO Faraday Efficiency (%)

[0054] The composition of gas released to the outside of the carbon dioxide electrolysis unit cell was measured through gas-chromatography (GC) analysis. In addition, the faraday efficiency was calculated through the equation below.

[Mathematical Equation 1]

$$FE_{product}(\%) = \frac{i_{product}}{i_{total}} \times 100 = \frac{V_{product} \times Q \times \frac{2Fp}{RT}}{i_{total}} \times 100$$

[0055] In Mathematical Equation 1 above, Q is a flow amount of a flow path in which products are released out of the carbon dioxide electrolysis unit cell, F is a faraday constant, p is a pressure, T is a measurement temperature, and R is an ideal gas constant. The total current (i_{total}) is a total current value applied over time, the current for the product ($i_{product}$) is a value calculated from the volume ($V_{product}$) of the gas measured through the GC analysis.

(2) Impedance (mΩ)

[0056] The Ohmic resistance was measured at 0.2 V and a frequency of 100 KHz to 10 Hz using a VSP potentiostat of BioLogic.

(3) Voltage (V)

[0057] Applying the current and measuring the voltage were performed through the VSP potentiostat of BioLogic. A booster of 80 A was installed, and the applying of the current for the large area was carried out. The applying of the current was maintained for a certain period of time at 200 mA/cm^2, and then the voltage when 30 minutes had passed was recorded. At this time, gas-chromatography (GC) analysis was carried out simultaneously.

[0058] FIG. 2 shows the values of impedance measured according to Example 1 and Comparative Example 1, and as a result of the experiment, the resistance was about 2.85 mΩ in Example 1 where the separator plate including the clad metal plate was used, and in Comparative Example 1 where a conventional separator plate having two metal plates being stacked was used, the resistance was about 6.10 mΩ. Through the results above, it can be seen that the clad metal plate corresponding to a single material through dissimilar joining had excellent characteristics in terms of resistance, compared to the separator plate formed simply by two metal plates being stacked. In addition, in case of a device in which cells, other than a unit cell, are stacked, voltage rise caused by the resistance may overlap, and the overvoltage may increase in the device and system, exhibiting a significant difference in terms of electrolysis efficiency between Example 1 and Comparative Example 1.

Experimental Example 2

[0059] For each of the electrolysis devices according to Example 1 and Comparative Example 2, a conversion rate of carbon dioxide, carbon monoxide faraday efficiency, hydrogen faraday efficiency, and overvoltage were measured in the condition of 300 mA/cm^2 in addition to the conditions of the electrode area of 1,000 cm^2, the carbon dioxide supply flow rate of 6 LPM and 8 LPM, the current density of 200 mA/cm^2. The conversion rate (%) of carbon dioxide was calculated as the ratio of the generated carbon monoxide (CO) to the amount of carbon dioxide (CO_2) gas added per time. In addition, the carbon monoxide faraday efficiency and overvoltage were measured in the same manner as that of Experimental Example 1, and the hydrogen faraday efficiency was measured in the same manner as that of the carbon monoxide faraday efficiency. The measurement results were listed in FIG. 3 below.

[0060] FIG. 3 is a table listing the result values of measuring carbon monoxide faraday efficiency, hydrogen faraday efficiency, carbon dioxide conversion rate, and overvoltage according to Example 1 and Comparative Example 2. Referring to FIG. 3, it can be seen that Example 1 where the flow path was included in the stainless-steel layer of the clad metal plate showed excellent level in all aspects of characteristics, compared to Comparative Example 2 where the clad metal plate was used as it is without a flow path. It can be seen that, without a flow path structure on the

stainless-steel layer, the material transfer resistance increased, causing a carbon dioxide depletion phenomenon to occur locally, thereby reducing the carbon monoxide faraday efficiency and the carbon dioxide conversion rate, and increasing the hydrogen faraday efficiency and the overvoltage.

[0061]

10: Clad metal plate
11: Titanium layer
13: Stainless-steel layer
20: Gas diffusion layer
30: Membrane electrode assembly
40: Titanium mesh
50: Titanium porous diffusion layer
61: First gasket
63: Second gasket

Claims

1. A separator plate comprising:

a clad metal plate including a titanium layer and a stainless-steel layer bonded to one side of the titanium layer; and
a titanium porous diffusion layer disposed on the other side of the titanium layer,
wherein the stainless-steel layer includes a flow path.

2. The separator plate of claim 1, wherein the titanium porous diffusion layer is a titanium felt.

3. The separator plate of claim 1, wherein the clad metal plate is formed of the titanium layer and the stainless-steel layer as one body by forced rolling.

4. The separator plate of claim 1, wherein the clad metal plate has a thickness of 0.5 mm or greater, and 1.5 mm or less.

5. The separator plate of claim 1, wherein a ratio of a thickness of the titanium layer and a thickness of the stainless-steel layer is 1:5 to 20.

6. The separator plate of claim 1, wherein the titanium porous diffusion layer has a thickness of 0.6 mm or greater, and 0.8 mm or less.

7. The separator plate of claim 1, wherein the titanium layer and the titanium porous diffusion layer comprise no flow path.

8. The separator plate of claim 1, wherein the separator plate has an area of 100 cm^2 or greater, and 5000 cm^2 or less.

9. An electrolysis device comprising:

a membrane electrode assembly in which an anode, a separator, a cathode, and a gas diffusion layer are disposed in sequence;
the separator plate according to claim 1 disposed between the plurality of membrane electrode assemblies;
a first gasket disposed between the anode and the separator plate; and
a second gasket disposed between the cathode and the separator plate,
wherein a thickness of the first gasket is larger than a thickness of the second gasket.

10. The electrolysis device of claim 9, wherein a ratio of the thickness of the first gasket and the thickness of the second gasket is 1:0.2 to 0.3.

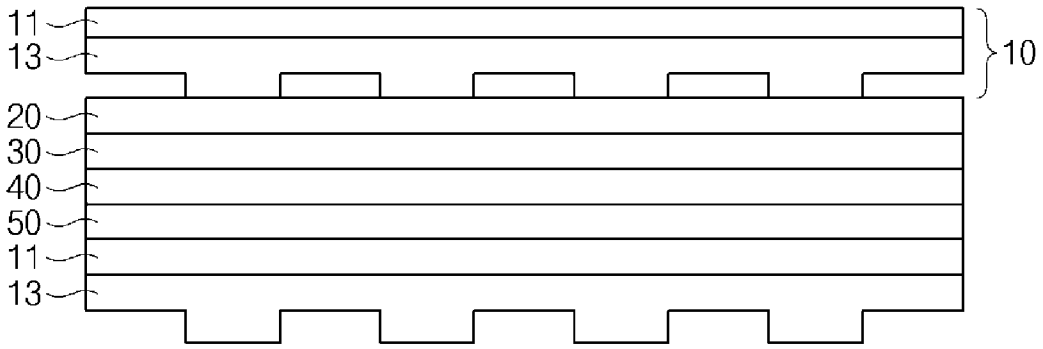
11. The electrolysis device of claim 9, wherein the membrane electrode assembly further comprises a porous layer on one side of the anode on which the separator is not disposed.

12. The electrolysis device of claim 11,

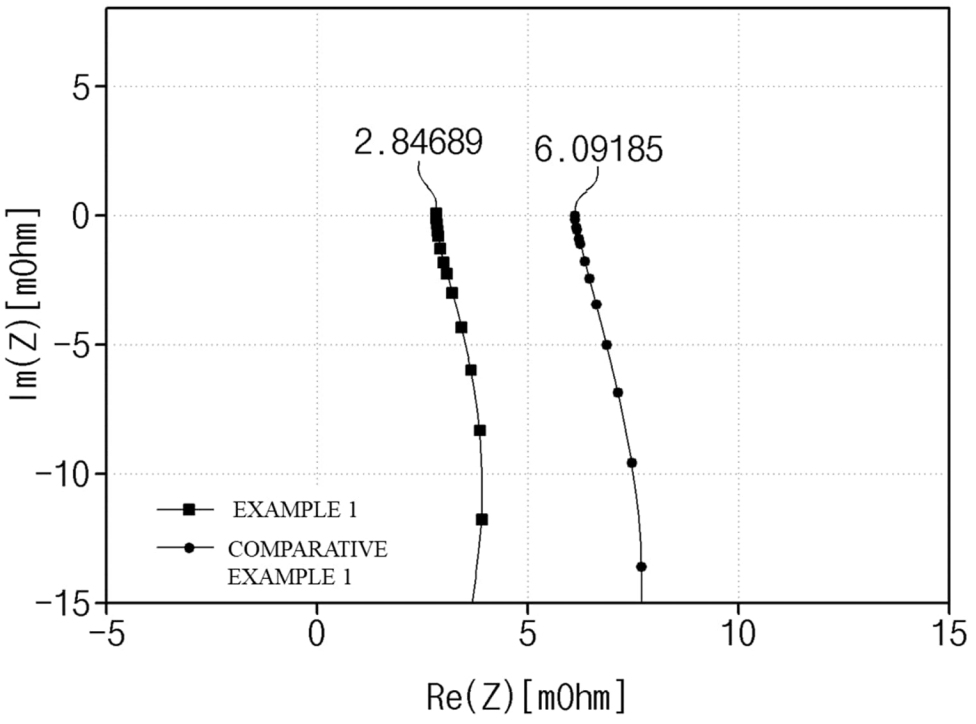
wherein the first gasket is disposed surrounding side surfaces of the porous layer and the titanium porous diffusion layer, and
the second gasket is disposed surrounding a side surface of the gas diffusion layer.

13. The electrolysis device of claim 9, wherein the electrolysis device electrolyzes carbon dioxide to generate one or more materials selected from the group consisting of carbon monoxide, ethylene, methane, formic acid, hydrocarbon, aldehyde, and alcohol.

[FIG. 1]


11
13
10
20
30
40
50
11
13

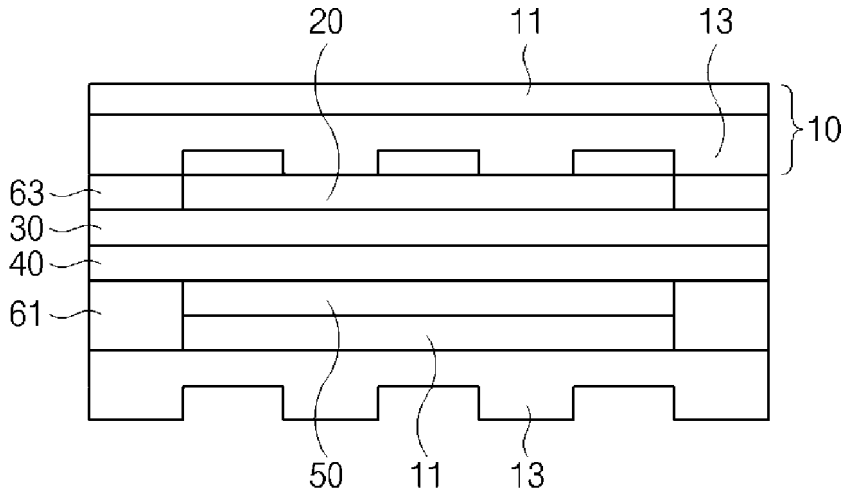
[FIG. 2]


2.84689
6.09185
EXAMPLE 1
COMPARATIVE EXAMPLE 1
Im(Z)[mOhm]
Re(Z)[mOhm]
5
0
-5
-10
-15
-5
0
5
10
15

[FIG. 3]

CLASSIFICATION		200mA/cm^2 (6LPM/Cell)	300mA/cm^2 (6LPM/Cell)	300mA/cm^2 (8LPM/Cell)
FE_{CO} (%)	EXAMPLE 1	98.78	92.36	96.92
	COMPARATIVE EXAMPLE 2	88.92	73.04	85.58
FE_{H2} (%)	EXAMPLE 1	1.08	6.98	2.81
	COMPARATIVE EXAMPLE 2	10.28	25.91	13.72
CONVERSION RATE (%)	EXAMPLE 1	22.13	33.97	23.69
	COMPARATIVE EXAMPLE 2	19.16	28.33	21.36
Vol.(V/Cell)	EXAMPLE 1	3.61	3.76	3.65
	COMPARATIVE EXAMPLE 2	3.85	4.07	3.92

[FIG. 4]



INTERNATIONAL SEARCH REPORT

International application No.
PCT/KR2025/000930

A. CLASSIFICATION OF SUBJECT MATTER

C25B 9/19(2021.01)i; **C25B 11/032**(2021.01)i; **C25B 3/26**(2021.01)i

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

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

C25B 9/19(2021.01); C25B 1/10(2006.01); C25B 11/031(2021.01); C25B 15/08(2006.01); C25B 3/04(2006.01); C25B 9/00(2006.01); C25B 9/77(2021.01); H01M 8/0208(2016.01); H01M 8/0228(2016.01); H01M 8/0232(2016.01); H01M 8/0243(2016.01); H01M 8/10(2006.01)

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Korean utility models and applications for utility models: IPC as above
Japanese utility models and applications for utility models: IPC as above

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)

eKOMPASS (KIPO internal) & keywords: 전기분해 장치(electrolysis device), 분리판(separator), 티타늄(titanium, Ti), 스테인리스강(stainless steel), 클래드 금속판(clad metal plate), 다공성 확산층(porous diffusion layer), 유로(flow path)

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
Y	KR 10-2023-0081020 A (KOREA INSTITUTE OF SCIENCE AND TECHNOLOGY) 07 June 2023 (2023-06-07) See claims 1-5 and 7-13; paragraphs [0020], [0021], [0025], [0030]-[0035], [0049], [0053]-[0055], [0058], [0062] and [0091]; and figures 4 and 5.	1-13
Y	JP 2017-179601 A (IXTG NIPPON OIL & ENERGY CORP. et al.) 05 October 2017 (2017-10-05) See claims 1 and 2; paragraphs [0037]-[0046], [0050], [0051], [0059] and [0060]; and figures 2 and 6.	1-13
Y	KR 10-2017-0069596 A (LG CHEM, LTD.) 21 June 2017 (2017-06-21) See claims 1-4 and 12; paragraphs [0058], [0061]-[0063], [0107]-[0109] and [0127]; and figures 4 and 5.	9-13
A	KR 10-2539783 B1 (HONG, Won Gi et al.) 07 June 2023 (2023-06-07) See claim 1; and paragraph [0037].	1-13

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

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"P" document published prior to the international filing date but later than the priority date claimed
"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention
"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone
"Y" 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 member of the same patent family

Date of the actual completion of the international search	Date of mailing of the international search report
18 April 2025	**23 April 2025**
Name and mailing address of the ISA/KR **Korean Intellectual Property Office** **Government Complex-Daejeon Building 4, 189 Cheongsa-ro, Seo-gu, Daejeon 35208** Facsimile No. **+82-42-481-8578**	Authorized officer Telephone No.

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INTERNATIONAL SEARCH REPORT

International application No.
PCT/KR2025/000930

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	KR 10-2022-0161633 A (SEOJIN INDUSTRIAL CO., LTD.) 07 December 2022 (2022-12-07) See abstract; claims 1-5; and figures 1 and 2.	1-13
A	US 2020-0002822 A1 (KABUSHIKI KAISHA TOSHIBA) 02 January 2020 (2020-01-02) See abstract; and claims 1-15.	1-13

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INTERNATIONAL SEARCH REPORT
Information on patent family members

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REFERENCES CITED IN THE DESCRIPTION

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