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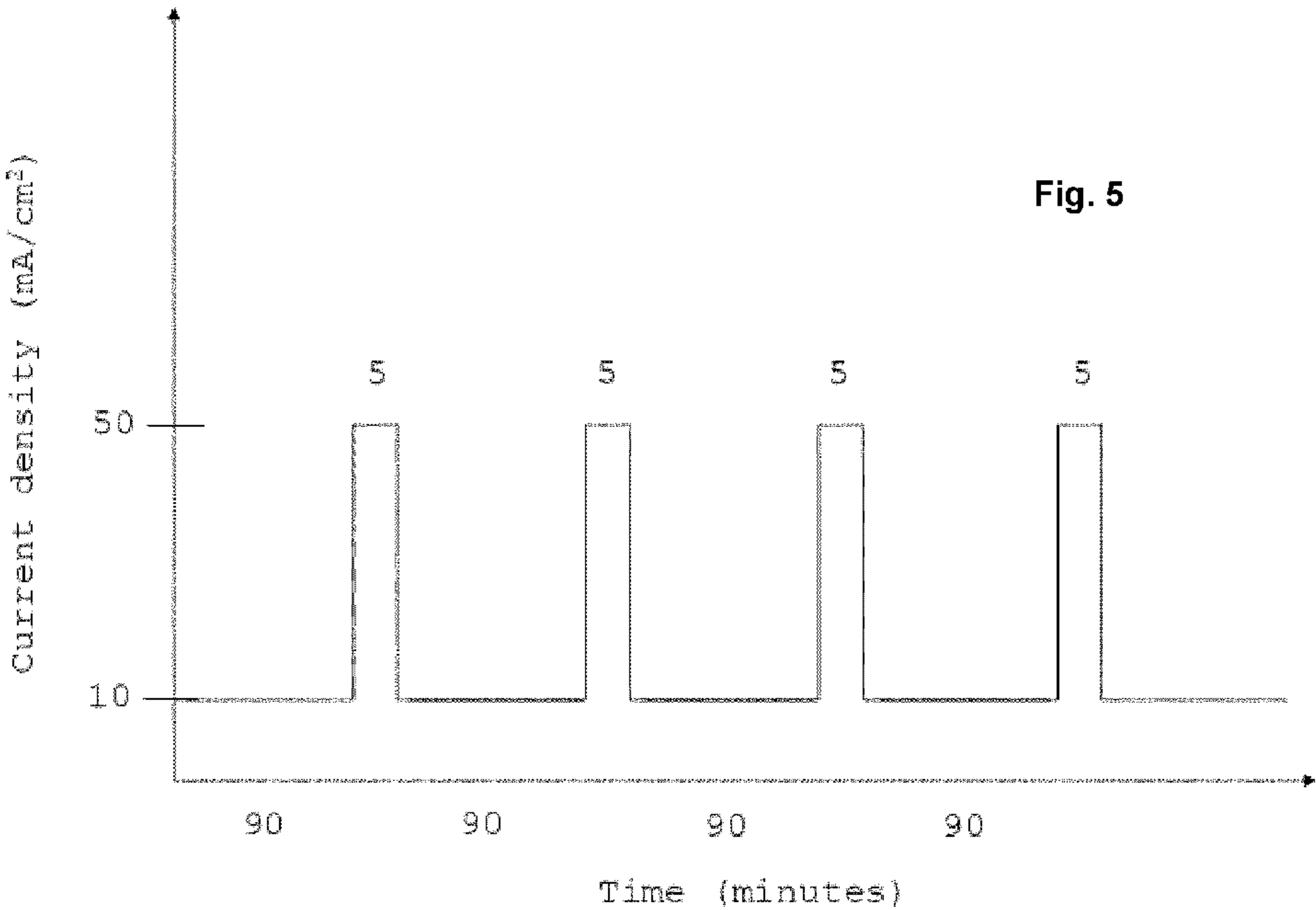
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(57) Abstract: In a method for electrochemical disinfection of water, especially potable water, with no addition of disinfecting chemicals to the water, an electropolished stainless steel plate-shaped cathode (mirror-like surface finish) sandwiched between two anodes on each side of the cathode plate is used. A low current density is applied to electrodes during normal disinfection and a higher current density is used during de-scaling of the cathode.

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A CATHODE FOR WATER DISINFECTION APPLICATIONS

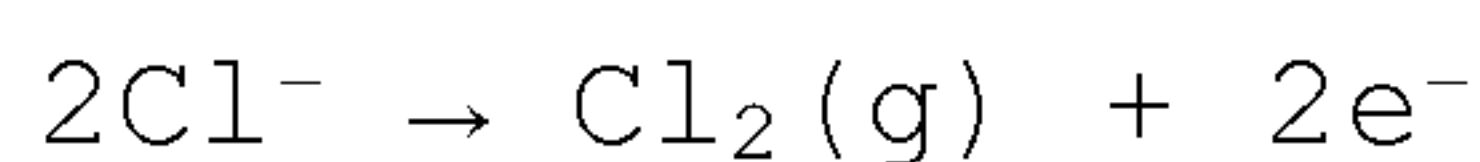
5 The present invention relates to a novel cathode for water disinfection applications. More specifically, the invention relates to a combination between an optimized cathode surface structure and a cyclic current operation that prevents deposition of calcareous deposits.

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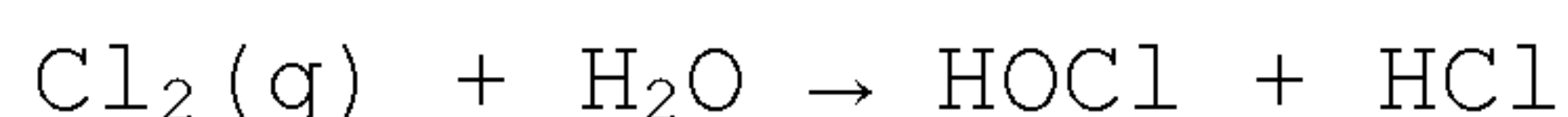
Electrochemical disinfection of potable water has the advantage that the disinfecting chemical species is produced directly in the electrolysis process, either from the water or from substances dissolved in the water. In the electrolysis process, both oxidizing and disinfecting substances are possible to be formed. Oxidizing substances are e.g. oxygen, ozone, hydrogen peroxide and hydroxyl radicals produced from the water, and disinfecting substances are e.g. hypochlorite/hypochlorous acid from dissolved chloride and peroxodisulfate from dissolved sulfate. The most important chemical species for disinfection of potable water are in most cases hypochlorous acid and hypochlorite, which are both formed from chlorine $\text{Cl}_2(\text{g})$ when it dissolves in water. The basic reaction equations are listed and shortly explained below:

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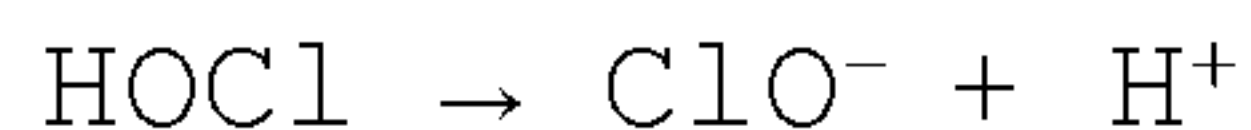
Oxidation of chloride to chlorine gas (anode):



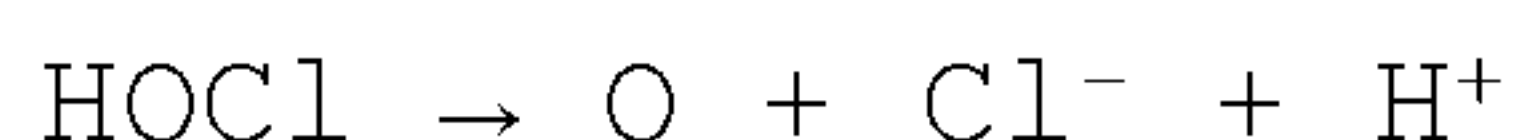
30 Dissolution of chlorine gas in water:



Dissociation of hypochlorous acid into hypochlorite:



Disinfection effect by releasing atomic oxygen from hypochlorous acid and hypochlorite:



Thus, during the actual disinfection, chloride ions consumed by the electrolysis process are released back again, leaving the water without changes to its original chemical composition.

Another advantage of this method is that, in most cases, addition of disinfecting chemicals is not required and, therefore, handling of toxic substances, such as chlorine gas, is becoming redundant. It also enables a cheap and alternative method for decentralized production of disinfected potable water, both in the existing grid but also in areas where it is difficult or inappropriate to transport hazardous chemicals (since only electricity is required for the electrolysis process).

Electrochemical production of free chlorine from the natural occurring chloride content in potable water are therefore known to be an effective disinfecting method, as both hypochlorite and hypochlorous acid, as mentioned, are produced. The electrolysis process often uses electrodes with mixed oxides of ruthenium and iridium ($\text{RuO}_2/\text{IrO}_2$). It is especially the high electrocatalytic activity of ruthenium oxide (RuO_2) that provides the electrodes with a low overpotential to form free chlorine $\text{Cl}_2(\text{g})$ from chloride, that

has made this type of coating relevant for the application. The electrodes are commonly known as mixed metal oxide (MMO) electrodes, and it is especially the mixed $\text{RuO}_2/\text{IrO}_2$ type of coatings that has shown superior performance compared to other types of electrode coatings, such as e.g. platinum for water disinfection.

Henri Bernard Beer developed and patented the first successful MMO anodes for the chlor-alkali electrolysis industry back in the 1960s, but since then they have found use in many other applications as well. Examples of other applications are e.g. corrosion protection in seawater (cathodic protection), electro-plating, electro-winning and anodizing of aluminium, to mention a few. Because of the proven long-term performance of $\text{RuO}_2/\text{IrO}_2$ coatings with low overpotential for chlorine, it has also been intensively investigated for water treatment applications such as disinfection.

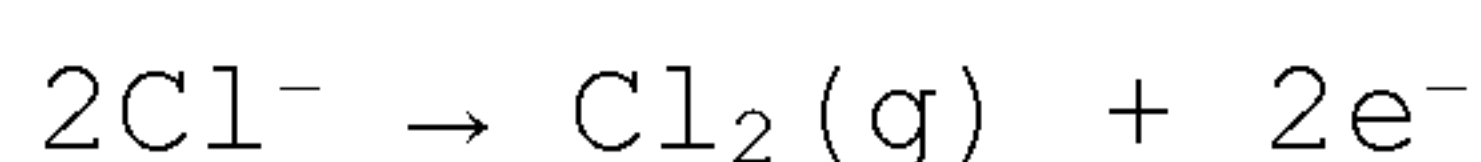
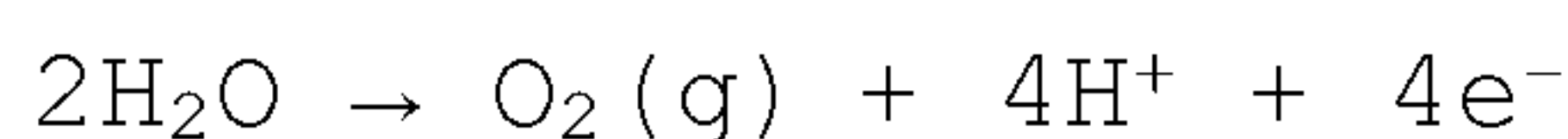
An MMO anode consists of a corrosion resistant base material, typically made from titanium (Ti) or niobium (Nb), that is subsequently coated with mixtures of metal oxides from the platinum group and metal oxides from group IVB and VB, e.g. Ti, Nb, V, Hf, Ta and Zr, also known as the valve metals. Most often, these coatings are obtained using the thermal decomposition method. The method can be explained as follows: Salts of the respective metals are dissolved and mixed into a liquid and then - by spraying or by other means - applied to the surface of the base material. Then the mixture is thermally decomposed onto the base material by heating in air at temperatures typically in the range of 400 - 550 °C. As a consequence of the method, the procedure

has to be repeated several times before a layer with appropriate thickness and electro-catalytic performance is obtained.

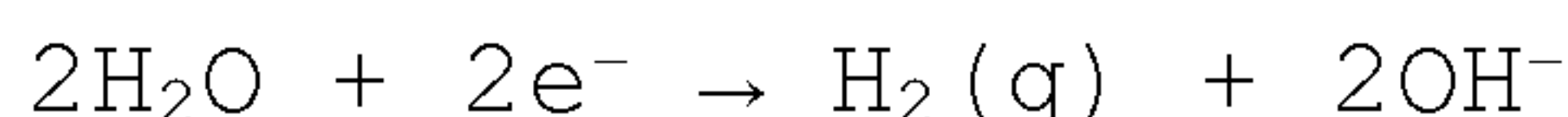
5 In electrolysis of potable water, both anodic and cathodic reactions are taking place at the electrodes. At the anode, water oxidizes into oxygen gas and dissolved chlorides oxidize into chlorine gas. At the cathode, water is reduced into hydrogen gas and hydroxyl ions. The basic reaction equations are explained below:

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At the anode:

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At the cathode:



As a consequence of the electrolysis process, the pH of the water near the electrodes is affected. This means that in the vicinity of the anode, the water becomes acidic, and in the vicinity of the cathode, the water becomes alkaline. However, potable water also contains dissolved ions of magnesium (Mg^{2+}), calcium (Ca^{2+}) and hydrogen carbonate (HCO_3^-), and the alkalinity near the cathode causes both magnesium hydroxide ($\text{Mg}(\text{OH})_2$) and calcium carbonate (CaCO_3) to precipitate and deposit at the surface. This, are explained thermodynamically with Pourbaix diagrams which are seen in figure 1-3. These "calcareous deposits" will eventually create problems due to coverage of the cathode surface, which reduces the active surface area for the reactions to take place. Since the water is continuously flowing in the electrolysis process, the calcareous deposit will continue

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to grow and eventually cause blockage, low production of disinfecting substances or other failures that make the process infeasible or unattractive at system levels. This means that the hardness of the water to be treated has a great impact on how fast the calcareous deposits will grow on the cathode and should, therefore, always be considered. However, the problem with calcareous deposits can partly be avoided if the polarity of each electrode is reversed. This technique, which is often referred to as "polarity reversal", means that the former cathode becomes anode and the former anode becomes cathode. The technique advantageously exploits that the water near the anode surface is acidic, and therefore can assist dissolution of the calcareous deposits adhering on the former cathode. By this method, the coverage of calcareous deposits can be minimized, but not totally avoided. Since the calcareous deposit grows rapidly (depending on the water hardness), there is a need for a frequent constant change of polarity (typically every 5-30 minutes) in order to dissolve the calcareous deposit layer before it starts growing in thickness.

Since calcareous deposits, in most cases, cannot be fully avoided, there will be a need to descale the electrodes with regular intervals. This descaling is normally carried out by an acid wash that dissolves the calcareous deposits from the electrodes. Acid wash or descaling is an unwanted process that causes system downtime, and therefore the need for it should be kept as low as possible.

Today, the polarity reversal technique, as described above, is known as being one of the most effective methods for

avoiding calcareous deposits on the cathode during electrochemical water disinfection. This method, however, has one major downside, which is that the electrodes need to be identical. This means, in turn, that each electrode needs -
5 as described - to act as both anode and cathode, which is problematic from a material science point of view. The MMO electrodes are designed to act as anodes, not as cathodes, but they can in principle be used as both. If, however, the polarity is not reversed regularly, calcareous deposits
10 will build up on the cathode, but at the same time polarity reversal causes loss of the active MMO coating on the electrodes. Often, this phenomenon is described as wear of the coating. As a consequence of this wear, the electrodes are becoming electrochemically inactive, due to loss of the
15 coating, thus leaving the base material (Ti, Nb) unprotected from oxidation, which causes passivation of the metal. As a result, this will create a very high voltage between the electrodes, since the current cannot pass the non-conductive oxides of the base material. Therefore, it
20 is known that electrochemical disinfection of potable water - due to the hardness of the water - only has a very limited lifetime, which makes the technique infeasible from an economic perspective (cost of electrodes).

25 The invention concerns a novel cathode that prevents any deposition of calcareous deposits, so that polarity reversal of the electrodes no longer is needed for electrochemical water treatment.

30 The invention addresses the need for a long-term stable cathode that prevents calcareous deposits from adhering to

the electrochemically active surface. Having a cathode surface that does not allow deposits to build up solves the main problem with the electrode configuration, which is the need for identical electrodes, and allows the RuO₂/IrO₂ based MMO electrode to function as anode only. As mentioned, the polarity reversal is the main degradation mechanism and has to be avoided before the lifetime perspective can be improved to an extent where the technology is feasible on the market. This problem is solved with the novel cathode of the invention which does not degrade due to coverage with calcareous deposits. Another problem addressed by the invention is that the cathode can be made from very cheap materials compared to a standard MMO electrode that - besides titanium or niobium as core metal - contains mixtures of expensive platinum group metals such as Ru and Ir. Therefore, these MMO electrodes are costly to install in large scale water treatment plants, and that is why the lifetime and reduction in number of electrodes is valuable. By using the newly developed cathode of the invention, the use of precious metals can be lowered by 50 % and also spare the costly processing route (thermal decomposition of several painted layers) and titanium or niobium metal as coating substrate.

A number of prior art documents deal with electrolytic or electrochemical purification and disinfection of water. Thus, US 10.301.728 describes an electrolysis mechanism for deployment in a tank containing water, said mechanism having at least one rotating cathode configured to rotate during an electrolysis process, at least one stationary cathode cleaning element and at least one stationary anode deployed adjacent to the rotating cathode.

US 2011/0198300 deals with electrochemical elimination or reduction of microbial impurities of liquids. The treated liquids may i.a. include wastewater, industrial process water and water intended for human consumption. The device comprises an electrode stack of two or more perforated electrode plates made of a conductive material.

In US 2005/0087484, an electrolytic system for treating raw surface water sources and disinfecting them for *Cryptosporidium* to produce water of a drinkable quality for humans is described. The system can be stand-alone, or it can be a sub-system of municipal water-treatment plants serving up to 10.000 people.

So far, the problem with calcareous deposits on the active cathode has mostly been addressed using the polarity reversal technique, since it is the easiest known way to avoid the calcareous build-up on the cathode. Over time, many attempts have been made to avoid this calcareous build-up: For example, US 4.048.030 describes an electrolytic cell for water treatment comprising a closed container with a lower inlet opening and an upper outlet opening for the water, and a cathode and an anode. In the interior of the electrolytic cell, particles are located whose density is higher than that of water, and which are fluidized by the water flowing through the cell. The movement of the fluidized bed of spherical particles leads to a mechanical cleaning of the electrodes in the cell.

The use of a cathode consisting of an elastically deformable, electrically conductive body has been proposed in US

4.169.035 for use in a scaling-free electrode arrangement. The cathode undergoes elastic oscillations, whereby the formation of undesired deposits is prevented. In a similar approach, described in US 3.378.479, a spiral or helical metal cathode equipped with a heater is used. The alternate expansion and contraction of the cathode, which is obtained during alternate heating and cooling, is reported to shed the scale. Other attempts to solve the problem are described in section 9 of *Electrochemical Water Disinfection* by Alexander Kraft et al., Recent Res. Devel. Electrochem. 6, 27-55 (2003).

The main feature of the present invention is that it is in fact possible to avoid using the polarity reversal technique, because the cathode is designed to have a very low adherence of calcareous deposits. The function of the cathode in the context of the invention is to assist the anodic reaction, i.e. the oxidation of chloride ions to free chlorine. At the cathode, hydrogen gas and hydroxyl ions are produced as side reactions from water splitting, and it is - as mentioned - because of the local increase in pH near the cathode surface that calcium carbonate (CaCO_3) and magnesium hydroxide ($\text{Mg}(\text{OH})_2$) are favoured to nucleate and grow on the active surface. Now it has, however, turned out that the deposition of calcareous deposits not only can be lowered, but decidedly avoided, if the cathode is made from electropolished stainless steel and combined with cyclic high current intervals. Thus, during operation at higher current densities, an extra and more vigorous hydrogen evolution will take place at the cathode surface and cause de-attachment of loosely adhering calcareous deposits. So, the combined effect of a smooth electropolished stainless

steel surface and a time period with extra hydrogen gas
evolvment can in fact prevent calcareous deposits to grow
on the surface, and hereby the need for polarity reversal
and identical MMO electrodes is avoided. However, at the
5 anode, more free chlorine is produced when operating at
higher current densities, which should be taken into ac-
count when designing larger systems.

It should further be emphasized that high surface roughness
10 of an MMO electrode normally is considered as a beneficial
effect, due to the increased surface area. Especially, the
obtained "mud-cracked" surface structure of a thermally de-
composed MMO coating in combination with a sandblasted ti-
tanium/niobium base material is known to improve both the
15 coating adhesion and the catalytic surface area, which are
important parameters when the electrode is used as anode.
On the contrary, when the MMO electrode acts as a cathode,
the high surface roughness of both coating and substrate is
a major disadvantage for avoiding deposition and growth of
20 calcareous deposits, especially in the case of electrochem-
ical water treatment. It is well documented in literature
that surface roughness is a critical parameter concerning
the growth rate of calcareous deposits and that the problem
can be counteracted by creating more smooth surface struc-
25 tures, i.e. surfaces with low roughness or R_a value (see
e.g. "*Biomimetic Approach to Anti-fouling Surfaces*" by Wen
Choo Chiang, Doc. Phil. thesis, University of Leeds, and
Declét et al., Rev.Adv.Mater.Sci.44 (2016), 87-107) . For
example, this effect is well known in the heat exchanger
30 industry.

Electrochemical polishing or electropolishing can be explained as an electrolytic process that uses electric current to remove material from a metallic surface immersed in an electrolyte. The metal object to be electropolished is coupled as anode (+), and the cathodes (-) are primarily used to assist the current distribution on the surface of the object. At the anode, metal dissolves preferentially at the micro-peaks of the surface, resulting in a reduced surface roughness. The polishing effect is controlled by an anode-film created by the electrolyte in the electrolysis process, where surface-metal in areas with high current densities is dissolved more rapidly than in areas with low current densities. The electropolishing therefore results in metal surfaces with very low roughness caused by anodic dissolution of the metal in the electrolytic process. Many different metals, such as for example stainless steels, carbon steels, aluminum, copper, nickel, nickel alloys, titanium and zirconium, can be electropolished, and the difference between the processes is mainly the electrolyte, the temperature and the current densities used. Stainless steels are very often electropolished with the main purpose of obtaining smooth and burr-free surfaces with a low roughness. These features are interesting to obtain for many reasons, such as (to mention a few):

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- Improvement of corrosion properties
- Removal of burrs from edges (de-burring)
- Appearance for decorative purposes
- Lower friction
- 30 - Hygienic surfaces
- Improving fatigue properties (removal of scratches)

So electropolishing of stainless steels is, in fact, a surface treatment process that has many advantages and, therefore, also is well established in the industry. As mentioned, it is primarily the chemical constituents in the electrolyte of the electropolishing process that control the anodic dissolution of the metal. For stainless steels, a large variety of electrolytes has been developed to cover the field of different series of alloys. For the 300 (austenitic) and 400 (ferritic) series, an example of a standard electrolyte composition is listed together with operating parameters in the following table 1:

Table 1

Typical composition of an electropolishing electrolyte for the 300 and 400 series of stainless steels

Chemical constituents:	Standard composition in %:
Sulfuric acid H_2SO_4 (conc.)	41%
Phosphoric acid H_3PO_4 (80%)	45%
Chromic acid CrO_3	7%
Water H_2O	7%
Operating parameters:	Operating conditions:
Current density	30-60 A/dm ²
Temperature	65 °C

So, the present invention relates to a novel cathode for water disinfection applications, said cathode being characterized in that it is

- made from a stainless steel substrate and subsequently polished/electropolished to a mirror finish according to a

1P/2P surface finish (according to EN 10088-2) with typical R_a values below $0.1\mu\text{m}$,

- shaped as a massive plate in order to minimize any geometrical areas where the calcareous deposits can adhere, and
- assisted by an MMO anode on each side to cover both sides of the cathode plate fully with an electrical field, ensuring hydrogen evolution over the full surface area.

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In an embodiment of the invention the anodes on each side of the cathode plate, each has an area which is larger than the area of the cathode, to ensure that the cathode plate is fully covered with an electrical field. The anode area may be 1% larger than the cathode area or even larger. The anode area may thus be 2%, 3%, 4%, 5%, 6%, 7% or 8% or between 8% and 20% larger than the cathode area. It is to be understood that when speaking of 1%, 2%, 3%, 4%, 5%, 6%, 7%, 8% or 20%, it is not exact numbers, but numbers with tolerances which are normally occurring in the manufacture, such as $\pm 0.5\%$.

20

In a further embodiment of the invention the plate-shape of the anodes corresponds to the plate-shape of the cathodes in order to fully cover the cathode plates with an electrical field. Thus, if the cathode plate or plates are square, also the anode plates are square. If the cathode plate(s) are rectangular, circular etc., also the anode plates are rectangular, circular etc. corresponding the shape of the cathode plate(s).

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In at further embodiment of the invention, the anode plates are arranged with their centre located on the same centre line as the cathode plate or plates, to ensure that the cathode plate(s) are fully covered with an electrical field when in operation. Thus, in the embodiment where the anode plates have a larger area than the cathode plate(s), the anode area "overlapping" the cathode area has an equal width along the edge of the cathode plate(s).

It is, as mentioned, the combination of a smooth surface, i.e. a surface with a very low surface roughness, and the evolving hydrogen gas that prevents the calcareous deposit to adhere to the cathode surface. This is also what has been observed during the experimental work described later in the examples.

In the method, in which the new cathode is used, calcareous deposits might eventually create problems anyhow. These are often caused by build-up on non-conductive spacers between the stacked electrodes, or other non-conducting material such as chamber walls etc. For that reason, it may still be necessary to de-scale the electrodes from time to time. This, however, is equally necessary for the polarity reversal technique, and de-scaling with acid needs to be performed on a more or less regular basis, depending on water hardness, for the same reasons as mentioned above. In the present case, where the cathode of the invention is made of electropolished stainless steel, it is important to choose the right de-scaling agent, which should preferably be diluted sulfamic acid (aminosulfonic acid, H_3NSO_3) or another acid, such as (but not restricted to) citric acid ($\text{C}_6\text{H}_8\text{O}_7$)

or acetic acid ($\text{C}_2\text{H}_4\text{O}_2$), that does not initiate corrosion of the electropolished stainless steel surface.

5 The main advantage of the novel cathode of the invention is that electrochemical water disinfection can be performed with different electrodes, i.e. with an MMO anode and a cathode according to the invention where there will be no need to perform polarity reversal to avoid growth of calcareous deposits. This will enhance the lifetime of the
10 electrodes significantly, simply because the MMO electrode does not fail due to loss of the active coating (wear) which produces the free chlorine, $\text{Cl}_2(\text{g})$, in the electrolysis process.

15 As another advantage of the invention can be mentioned the cost savings, since the use of expensive platinum group metals, i.e. ruthenium and iridium, can be reduced by 50% and replaced with electropolished stainless steel.

20 The cathode of the invention can also be obtained by other means, such as bright nickel coatings made by electrodeposition methods. These coatings are known to provide smooth and mirror-like surfaces by material deposition, preferentially in the micro-valleys of the surface, in contrast to
25 electropolishing, as explained, where surface roughness is reduced by material dissolution. Bright nickel coatings can therefore provide the same features as electropolishing with a "reverse" process that reduces the surface roughness of the material by levelling. Nickel metal, compared to
30 stainless steels, is also more electrocatalytic, which means that the overpotential to evolve hydrogen at the cathode surface is reduced. It is also expected that the

corrosion properties will be improved by nickel coatings due to the excellent resistance of the metals in alkaline solutions. However, these extra features of electrodeposited bright nickel coatings are challenged by the additional cost, compared to electropolished stainless steel, thus considering its impact on performance in larger systems.

Experimental

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In the following, reference is made to the figures 4-5.

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Working of the invention can be performed by designing a cathode that is made from a stainless steel substrate and subsequently electropolished to a 1P/2P mirror-like surface finish. It is important that the cathode is shaped as a massive plate to minimize the presence of geometrical areas to which the calcareous deposits can adhere. Further, it is important that the cathode (100) is assisted by an anode (101) on each side to ensure that both sides of the plates are fully covered with an electrical field as illustrated in Figure 4. This means that, when stacking the electrodes, the first and last electrode in the stack should preferably be an anode. Advantageously, the anodes are shaped at least 1% by area larger than the cathode in order to minimize calcareous build-up along the edges of the cathode. It is equally important that the plate-shape of the anode and the cathode are the same throughout the stack e.g. square, rectangular, circular etc. The surface of the cathode could beneficially be improved further by bright nickel coatings to create a more electrocatalytic and corrosion resistant surface.

In order to inhibit the growth of calcareous deposits on the active cathode surface, high current intervals creating extra and more vigorous hydrogen gas evolution are necessary to perform during the electrolysis process, as already mentioned. Such high current intervals can, as an example, be performed by raising the current from the normal set point by a factor of 5 over a time period of 5 minutes. An example of such current operation is seen in figure 5. However, it is always important that the anode current densities are kept in an optimal range, i.e. that the targeted level of chlorine produced is adjusted accordingly to the anode area. If the current densities used in the electrolysis process are too high, the MMO anodes can be damaged, causing shortening of the lifetime. This applies both during normal and especially during high current operation.

An example of optimal ranges of current density and time for electrochemical water disinfection during cyclic current operation is given in table 2 below.

Table 2

Operation mode	Anode current density (mA/cm ²)	Cathode current density (mA/cm ²)	Time (minutes)
Normal (disinfection)	5-30	5-30	30-120
High (de-scaling)	30-150	30-150	5-15

It should, however, be highlighted that there are many combinations, in which it is optimal to perform the normal and high current cycles. Especially the chloride content and hardness of the water to be treated, respectively, have a major impact on the settings for current used for disinfection i.e. production of chlorine and the high current used for hydrogen evolution i.e. de-scaling. However, the most important feature is that the MMO electrodes are consistently operating as anodes and not as cathodes in the electrolysis process, hereby eliminating the need for polarity reversal.

Claims:

1. A method for electrochemical disinfection of water,
5 especially potable water, with no addition of disinfecting
chemicals to the water, wherein
- a plate-shaped cathode is used, which is assisted by an
anode on each side to fully cover both sides of the plate
10 with an electrical field,
 - the need for polarity reversal of the electrodes during
use is eliminated, and
 - 15 - a low current density of the electrodes is used during
normal disinfection operation, while a higher current den-
sity of the electrodes is used during de-scaling of the
cathode,
 - 20 said cathode being characterized in that it is
 - made from a stainless steel substrate and subsequently
electropolished to a 1P/2P mirror-like surface finish,
 - 25 - shaped as a massive plate in order to minimize any geo-
metrical areas where the calcareous deposits can adhere,
and
 - assisted by an anode on each side to cover both sides of
30 the plate fully with an electrical field.

2. Method according to claim 1, wherein the current density of the electrodes is 5-30 mA/cm² during 30-120 minutes of disinfection operation and 30-150 mA/cm² during 5-15 minutes of de-scaling operation.

3. Cathode according to claim 1, wherein the surface is further protected with bright nickel deposits to provide additional smoothness and corrosion protection.

4. Cathode according to claim 1, wherein the anodes on each side of the plate-shaped cathode each has an area which is larger than the area of the cathode, to fully cover both sides of the plate-shaped cathode with an electrical field.

5. Cathode according to claim 4, wherein the anodes on each side of the plate-shaped cathode each has an area which is 1% larger than the area of the cathode.

6. Cathode according to claim 4, wherein the anodes on each side of the plate-shaped cathode each has an area which is 2%, 3%, 4%, 5%, 6%, 7% or 8% larger than the area of the cathode.

7. Cathode according to claim 4, wherein the anodes on each side of the plate-shaped cathode each has an area which is between 8% and 20% larger than the area of the cathode.

8. Cathode according to any of the preceding claims,
wherein shape of the anodes corresponds to the plate-shape
of the cathode to fully cover both sides of the plate-
shaped cathode with an electrical field.

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9. Cathode according to any of the preceding claims,
wherein the shape of the cathode is square, and the shape
of the anodes are square.

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10. Cathode according to any of the preceding claims,
wherein the shape of the cathode is rectangular, and the
shape of the anodes are rectangular.

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11. Cathode according to any of the preceding claims,
wherein the shape of the cathode is circular, and the shape
of the anodes are circular.

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12. Cathode according to any of the preceding claims,
wherein the anodes plates are arranged with their centre
located on the same centre line as the centre line of the
cathode, to fully cover both sides of the plate-shaped
cathode with an electrical field.

25

13. Use of the cathode according to any of the claims
claim 3 - 4, where the cathode is assisted by an anode on
each side to ensure that both sides of the plates are fully
covered with an electrical field.

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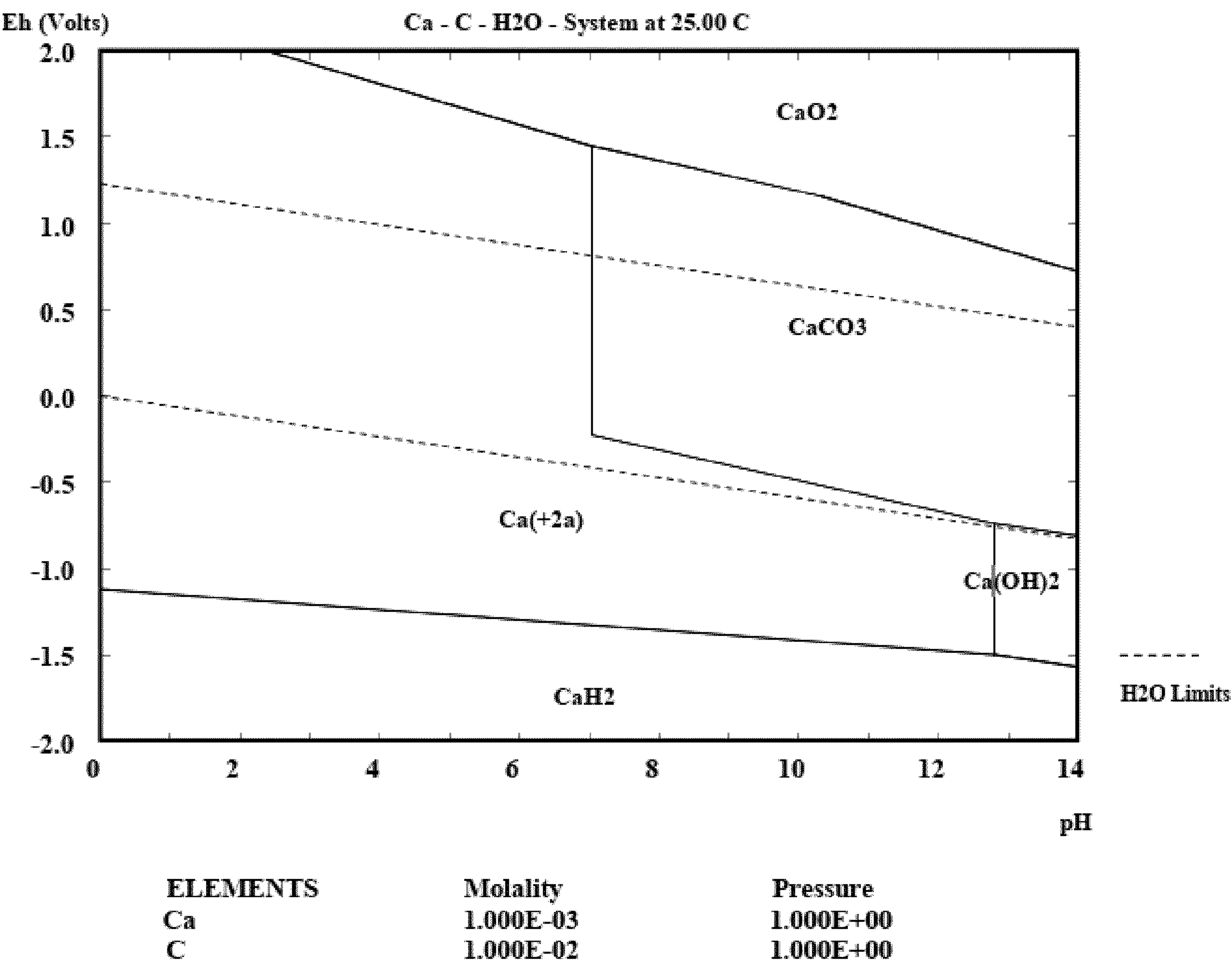
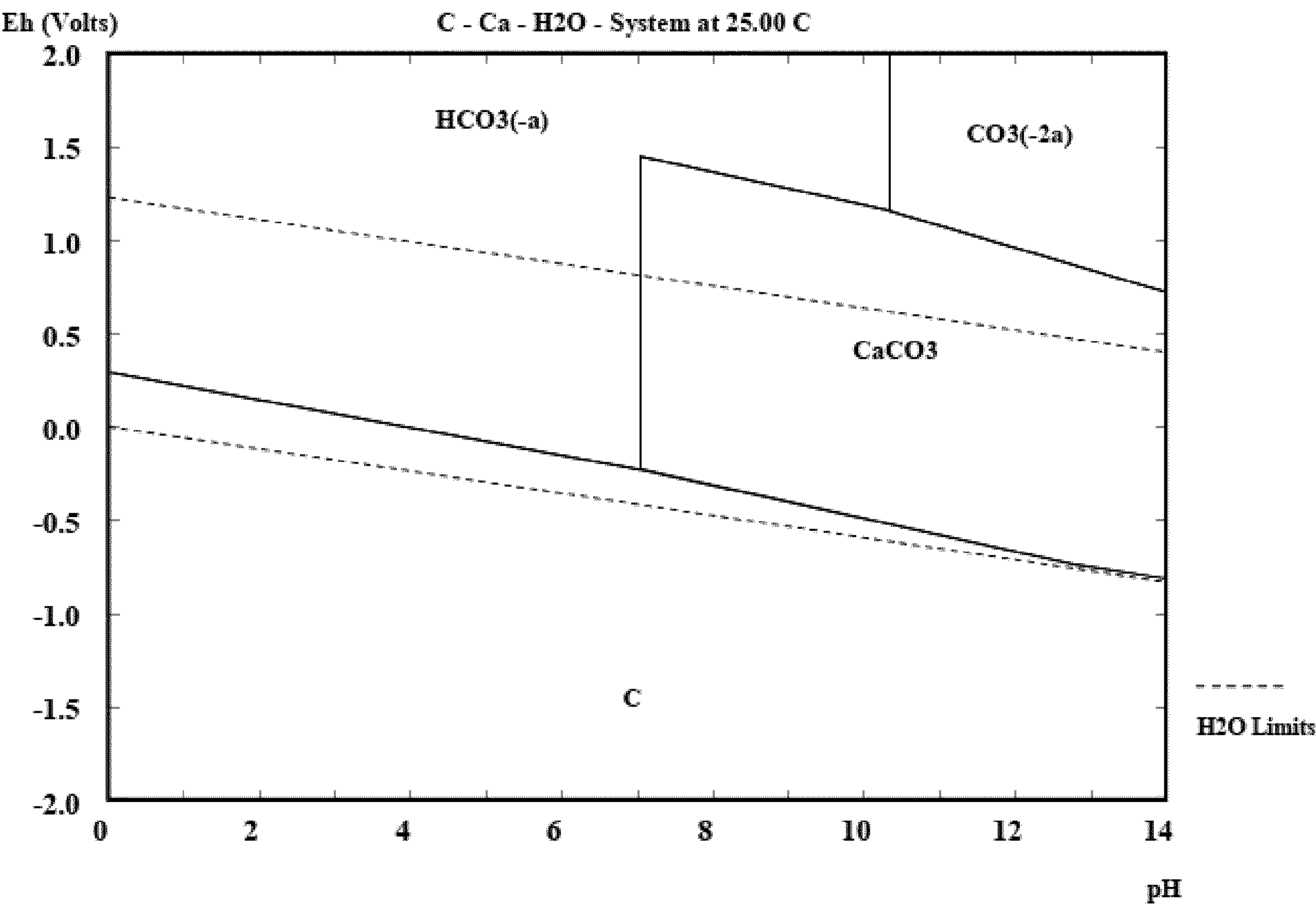


Fig. 1

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ELEMENTS	Molality	Pressure
Ca	1.000E-03	1.000E+00
C	1.000E-02	1.000E+00

Fig. 2

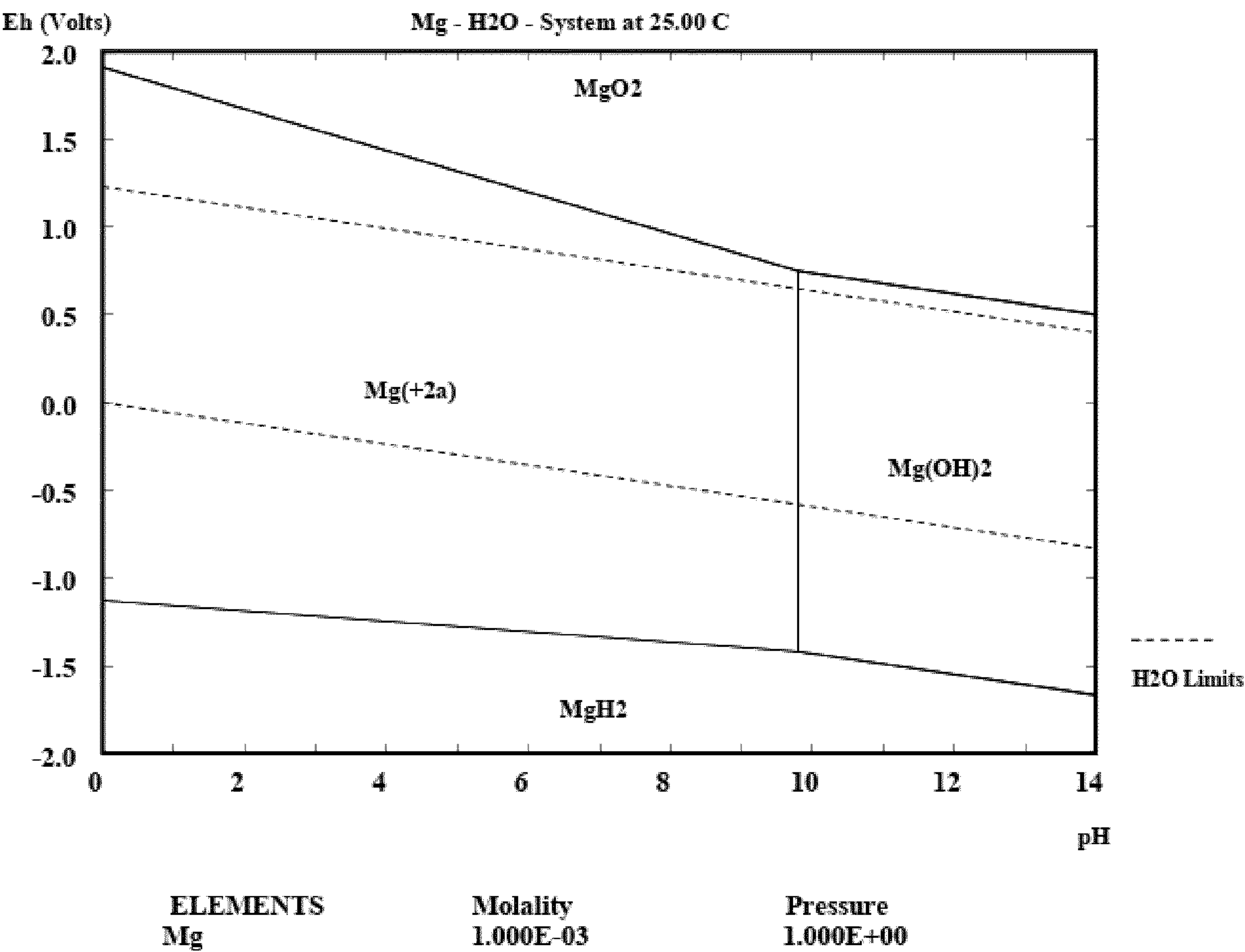


Fig. 3

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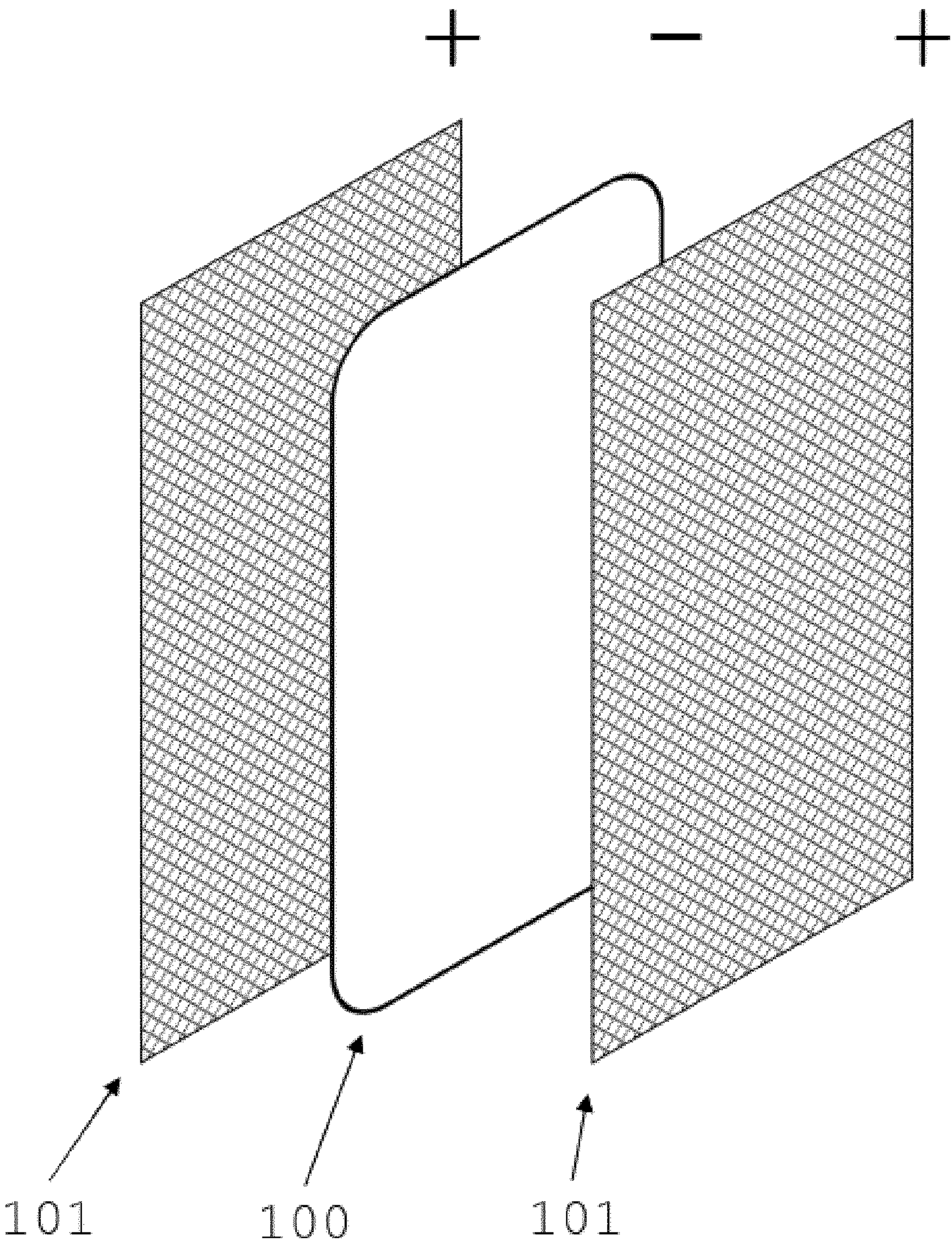


Fig. 4

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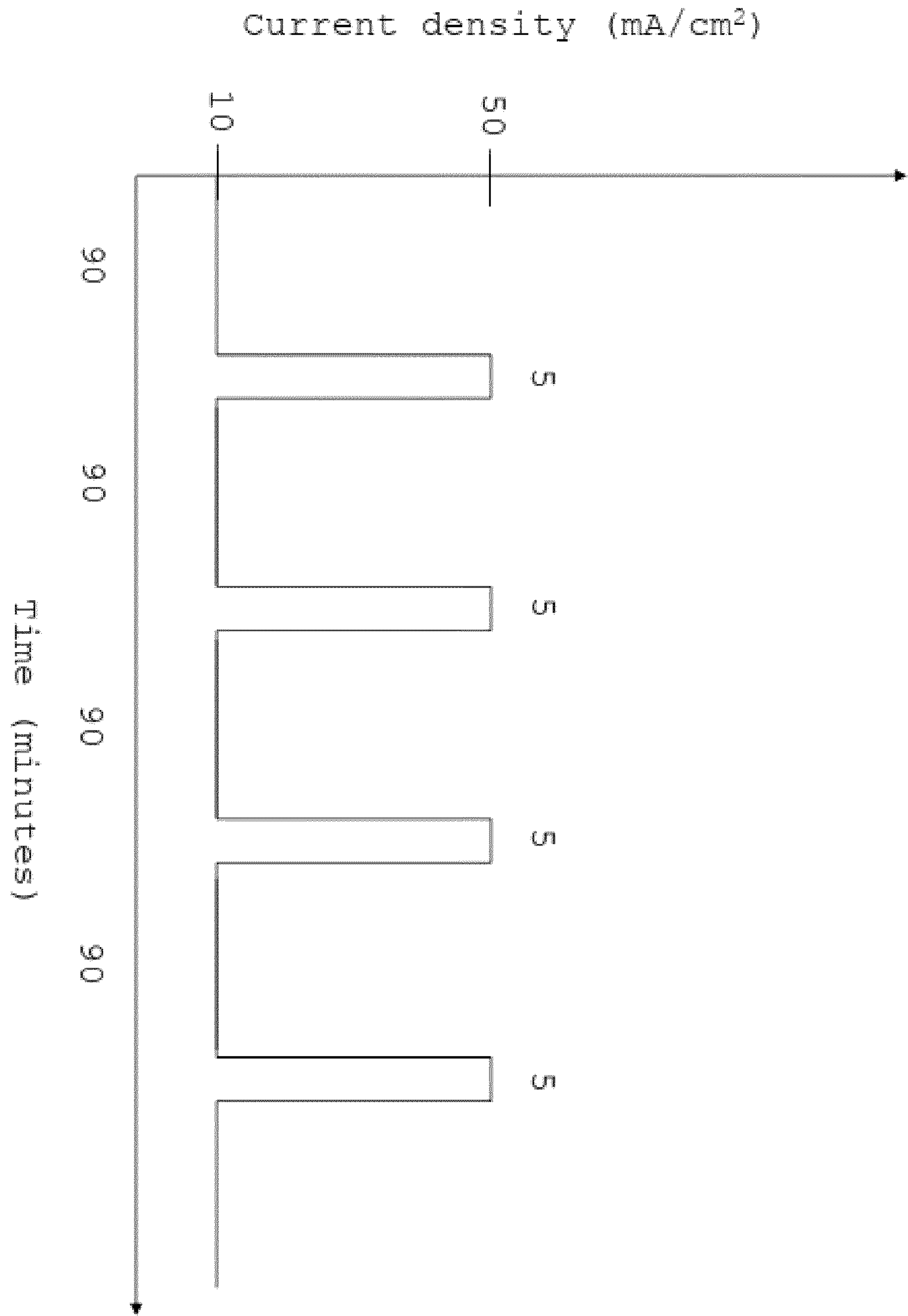


Fig. 5

INTERNATIONAL SEARCH REPORT

International application No

PCT/EP2020/079762

A. CLASSIFICATION OF SUBJECT MATTER INV. C02F1/46 C02F1/467 ADD. C02F103/02 C02F1/461 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) C02F Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched Electronic data base consulted during the international search (name of data base and, where practicable, search terms used) EPO-Internal, WPI Data		
C. DOCUMENTS CONSIDERED TO BE RELEVANT		
Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	CN 107 879 488 A (UNIV ZHEJIANG) 6 April 2018 (2018-04-06) paragraphs [0001], [0010], [0019] - [0021], [0027] - [0030], [0033]; figures 1,2 -----	1-13
A	WO 96/30307 A1 (BIOQUEST [US]) 3 October 1996 (1996-10-03) claims 22,23 -----	1-13
A	US 2013/001099 A1 (PULLIAINEN MARTTI [FI] ET AL) 3 January 2013 (2013-01-03) page 8, lines 19-35; figure 3 -----	1-13
A	AU 2006 284 520 A1 (POOLRITE EQUIPMENT PTY LTD [AU]) 1 March 2007 (2007-03-01) page 6, lines 15-22 ----- -/-	1-13
☒ Further documents are listed in the continuation of Box C. ☒ See patent family annex.		
* Special categories of cited documents : "A" document defining the general state of the art which is not considered to be of particular relevance "E" earlier application or patent but published on or after the international filing date "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) "O" document referring to an oral disclosure, use, exhibition or other means "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 18 December 2020		Date of mailing of the international search report 14/01/2021
Name and mailing address of the ISA/ European Patent Office, P.B. 5818 Patentlaan 2 NL - 2280 HV Rijswijk Tel. (+31-70) 340-2040, Fax: (+31-70) 340-3016		Authorized officer Fiocchi, Nicola

INTERNATIONAL SEARCH REPORT

International application No
PCT/EP2020/079762

C(Continuation). DOCUMENTS CONSIDERED TO BE RELEVANT		
Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
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A	----- GB 2 409 684 A (MEDIPURE LTD [GB]) 6 July 2005 (2005-07-06) page 4, lines 1-16 -----	1-13

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International application No

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