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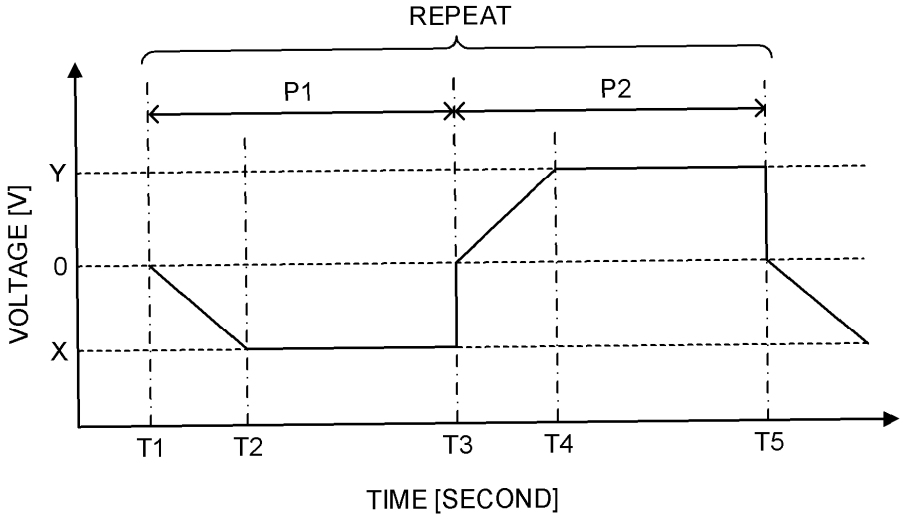
(54) **VOLTAGE CONTROL METHOD**

(57) A voltage control method of an embodiment is for applying a voltage to an electrochemical device that has an electrode with a catalyst containing a noble metal and periodically reversing a polarity of the voltage to perform an electrolytic reaction and recover a valuable resource from the electrochemical device. The voltage control method includes continuously increasing an absolute value of the voltage and then holding the value while periodically reversing the polarity of the voltage.

FIG. 5



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Description

FIELD

[0001] Embodiments relate to a voltage control method.

BACKGROUND

[0002] As global transition to a carbon neutral society progresses, hydrogen is being focused on as an alternative to fossil energy. In a society where hydrogen is used for various purposes, electrochemical devices such as a water electrolysis device that produces hydrogen by electrolysis of water and a fuel cell that generates power by using obtained hydrogen are important. There are various types of water electrolysis devices and fuel cells, depending on differences in operating temperature and device construction. Among those devices, a PEM-type water electrolysis device and a polymer electrolyte fuel cell (PEFC) using a solid polymer membrane as an electrolyte and a noble metal as an electrode catalyst have merits such as possibilities of size reduction and low-temperature operation, and are expected to become further widespread. Structures of the PEM-type water electrolysis device and the PEFC are quite similar, and both use a membrane electrode assembly (MEA) in which electrocatalysts are coated on both sides of a solid polymer membrane.

[0003] As transition to the hydrogen society progresses in the future, the number of PEM-type water electrolysis devices and PEFCs on the market increases, and a total amount of noble metals used for electrode catalysts also increases. Thus, in order for those devices to become widely used, it is necessary to secure a sufficient amount of noble metals. As a method for securing the amount of noble metals, there is cited recycling of a used PEM-type water electrolysis device and PEFC to recover noble metals. Procurement costs are expected to be decreased by recovering noble metals that are present in high concentration in waste products than by recovering noble metals contained in quite low concentration in ores. Further, recovering unevenly existing resources from waste articles enables stable row material procurement to thereby allow sustainable product manufacturing.

[0004] As a method of recovering a noble metal from a fuel cell, generally, after wastes are burned in an incinerator or the like, a noble metal component contained in ash is dissolved with a strongly acidic solution such as aqua regia, separated, and recovered (Patent Document 1). Further, as a method simpler and low in environmental load, a technology for dissolving and recovering a noble metal from a fuel cell by using electrolysis (Patent Documents 2, 3, and 4) is also developed, and development of a fuel cell recycling technology is expected to progress further in the future.

[0005] The technology for dissolving a noble metal from a MEA by electrolysis, the technology described in Patent Documents 2, 3, and 4, enables easy dissolution of a normally indissoluble noble metal by using dilute hydrochloric acid at a room temperature and is expected to be implemented in society as a method with a low environmental load. In the electrolysis method used here, the noble metal is dissolved by periodically reversing a polarity of an applied voltage, unlike in ordinary electrolysis that is performed at a constant voltage or constant current.

[0006] When a voltage is applied to an electrochemical device from the outside, there are observed a nonfaradaic current associated with charging of an electric double layer at an interface between an electrolytic solution and an electrode and a faradaic current associated with progress of an electrochemical reaction. The nonfaradaic current flows at a moment when the voltage is applied and does not flow when and after charging at the interface is completed, and only the faradaic current by the electrochemical reaction is observed (Non-patent Document 1). When electrolysis is performed such that the polarity of the applied voltage is periodically reversed, a nonfaradaic current flows at every moment of polarity switching. Since an electricity quantity saved at the interface increases as a polarity area becomes large, a large nonfaradaic current flows intermittently in a device such as a PEM-type water electrolysis device and a PEFC for which a porous body with a large electrode area is used as an electrode material. Since the nonfaradaic current flowing immediately after reversing of the polarity is quite large compared with the faradaic current, it is necessary to prepare a power supply with a large output size that can withstand a current larger than the current actually used for noble metal dissolution when noble metal dissolution by the electrolytic method is performed.

[0007] Patent Document 4 shows an example of performing electrolysis by applying a rectangular wave voltage. Though noble metal dissolution is possible also by this method, when considering a size increase of a device, a large current flows at a time of polarity reversal, and there is required an increase in a device cost and a safety mechanism against a the large current.

REFERENCE

[0008]

Patent Document 1: Japanese Patent No. 1626036
Patent Document 2: Japanese Patent No. 6652518

Patent Document 3: Japanese Patent No. 6652454
Patent Document 4: Japanese Patent No. 6109769
Non-Patent Document 1: "Chronoamperometry" by Fusao KITAMURA, Electrochemistry, 68, No. 1, p. 63-68 (2000)

SUMMARY

[0009] A problem to be solved by the present invention is to provide a voltage control method for recovering a valuable resource from an electrochemical device, the method being able to suppress generation of a large current.

[0010] A voltage control method of an embodiment is for applying a voltage to an electrochemical device that has an electrode with a catalyst containing a noble metal and periodically reversing a polarity of the voltage to perform an electrolytic reaction and recover a valuable resource from the electrochemical device. The voltage control method includes continuously increasing an absolute value of the voltage and then holding the value while periodically reversing the polarity of the voltage.

BRIEF DESCRIPTION OF THE DRAWINGS

[0011]

FIG. 1 is a block diagram illustrating a configuration example of an electrochemical system.
FIG. 2 is a block diagram illustrating a configuration example of an electrochemical system.
FIG. 3 is a block diagram illustrating a configuration example of an electrochemical system.
FIG. 4 is a graph illustrating a temporal variation of a voltage applied to an electrochemical device in order to recover a valuable resource by conventional electrolysis.
FIG. 5 is a graph illustrating a temporal variation of a voltage in an embodiment.
FIG. 6 is a schematic view illustrating an electric circuit that represents the inside of an electrochemical device.
FIG. 7 is a diagram illustrating a temporal variation of a voltage E.
FIG. 8 is a diagram illustrating a temporal variation of a current i.
FIG. 9 is a diagram illustrating an example of a temporal variation of a voltage E.
FIG. 10 is a diagram illustrating an example of a temporal variation of a current i.
FIG. 11 is a graph illustrating another example of a temporal variation of a voltage wave in the embodiment.
FIG. 12 is a graph illustrating a temporal variation of a current in an example.

DETAILED DESCRIPTION

[0012] Hereinafter, embodiments will be explained with reference to the drawings. In each embodiment below, the same codes may denote substantially the same component parts to partially omit explanation thereof. The drawings are schematic ones in which a relationship between a thickness and a planar dimension, a thickness ratio among components, and so on may be different from actual ones.

[0013] Note that in this specification "connecting" may include not only connecting directly but also connecting indirectly, unless otherwise specified.

[0014] This embodiment relates to a voltage control method for recovering a valuable resource from an electrochemical device by applying a voltage to the electrochemical device that has an electrode with a catalyst containing a noble metal, thereby reversing a polarity of the voltage periodically and performing an electrolytic reaction. The catalyst has a function to accelerate the electrolytic reaction such as an oxidation reaction and a reduction reaction by the electrochemical device and the electrolytic reaction of an electrolyte or a solute. Examples of the noble metal include platinum (Pt), iridium (Ir), ruthenium (Ru), rhodium (Rh), palladium (Pd), gold (Au), rhenium (Re), and so on. The catalyst may include an oxide that contains at least one of the aforementioned noble metals. The electrolytic reaction is performed by bringing an object (for example, an electrode having an oxide catalyst, or the like) into contact with a solution containing at least one of ion species and chemical species that can form a complex with the noble metal, applying a voltage to the object in contact with the solution, and periodically reversing a polarity of the voltage. In other words, the electrolytic reaction involves alternately performing the oxidation reaction and the reduction reaction. The solution contains, for example, hydrochloric acid (HCl).

[0015] FIG. 1, FIG. 2, and FIG. 3 are block diagrams illustrating configuration examples of an electrochemical system capable of performing the aforementioned method of controlling the voltage. The electrochemical system has an electrochemical device 100, a power supply 200, and a voltage control device 300.

[0016] The electrochemical device 100 has an electrode with a catalyst containing a noble metal. The noble metal is a valuable resource being an object to be recovered. As the electrochemical device 100, it is possible to use, for example, a fuel cell and a PEM-type water electrolysis device which are described in Patent Documents 2 and 3, and it is also possible to use an electrolysis tank provided with an electrode with a valuable resource inclusion, such as an electrode for soda

electrolysis and an insoluble electrode for plating.

[0017] The power supply 200 can generate a voltage for performing electrolysis by the electrochemical device 100. The power supply 200 is connected directly or indirectly to the electrochemical device 100, for example.

[0018] The voltage control device 300 can control a value of a voltage supplied from the power supply 200 to the electrochemical device 100, for example. The voltage control device 300 is connected to the power supply 200, for example.

[0019] The voltage control device 300 may be a system that controls a value of a voltage output from the power supply 200 by supplying a control signal to the power supply 200 as shown in FIG. 1, or may be a system that directly changes the value of the voltage output from the power supply 200 as shown in FIG. 2. Further, the power supply 200 and the voltage control device 300 may be provided independently or may be configured to have respective functions of the power supply 200 and the voltage control device 300 within a single power supply device 400 as shown in FIG. 3.

[0020] In dissolution of catalysts of noble metals such as Pt and Ru which is reported in Patent Documents 2, 3, and 4, a solution containing a component capable of forming a complex with a target noble metal is injected into the electrochemical device, and an extremely thin noble metal oxide film existing on a metal catalyst surface is converted to a metal by a reduction treatment, and thereafter, the exposed metal is dissolved by an oxidation treatment. At the time of oxidation treatment, not only dissolution but also formation of an oxide film proceeds simultaneously, so that a dissolution reaction stops when the metal is covered by the oxide film. Since the oxide film is removed by performing the reduction treatment again, performing reduction and oxidation alternately makes it possible to dissolve the noble metal that is normally indissoluble. In other words, a conventional method is a technique for dissolving a noble metal with a thin oxide on a surface thereof.

[0021] FIG. 4 is a graph illustrating a temporal variation of a voltage applied to an electrochemical device in order to recover a valuable resource by conventional electrolysis. A horizontal axis of the graph represents a time [second]. A vertical axis of the graph represents a voltage [V] applied to the electrochemical device. In the conventional recovery of the valuable resource described in Patent Documents 2, 3, and 4, voltages X and Y are periodically applied to the electrochemical device in a rectangular wave as illustrated in FIG. 4, to thereby dissolve a platinum group metal such as platinum and ruthenium. The voltage X has a value lower than the voltage Y. The voltage Y has a value higher than the voltage X, that is, a positive value, for example. However, in the above method of controlling the voltage, a spike-like nonfaradaic current flows at the moment of switching from the voltage X to the voltage Y and from the voltage Y to the voltage X, and there is a problem that electrolysis operation cannot be performed if an output of the electrochemical device is insufficient. As described above, though noble metal dissolution is possible by the aforementioned method, considering a size increase of the device, a large current flows at the time of polarity reversal, and there is required an increase in a device cost and a safety mechanism against a large current.

[0022] Thus, hereinafter, there will be explained a voltage control method which is capable of dissolving a noble metal while suppressing a large nonfaradaic current that flows at the time of polarity reversal. In this embodiment, a polarity of a voltage is periodically reversed not by a rectangular wave but by continuously increasing an absolute value of a voltage and thereafter holding that value as illustrated in FIG. 5. Thereby, it is possible to suppress instantaneous flowing of a large nonfaradaic current. In other words, in this embodiment, when reversing the polarity of the voltage, an inclination of a voltage wave from a value being a reference such as 0 V to a target value of an applied voltage is made larger than a rectangular wave. Examples of such a voltage wave include a trapezoidal wave.

[0023] FIG. 5 is a graph illustrating a temporal variation of the voltage in the embodiment. A horizontal axis of the graph represents a time [second]. A vertical axis of the graph represents a voltage [V] applied to the electrochemical device 100. In the embodiment, since an electrolytic reaction is performed by applying the voltage to the electrochemical device 100, thereby reversing the polarity of the voltage periodically, there are a period P1 of a first polarity and a period P2 of a second polarity reverse to the first polarity. In recovering the valuable resource, the period P1 and the period P2 are alternately switched and repeated. The voltage is applied, for example, to the electrode with the aforementioned catalyst.

[0024] In the period P1, a value of the voltage applied to the electrochemical device 100 is decreased over time from a time T1 to a time T2 to a value equal to or lower than a value of a voltage X. The voltage X is lower than a voltage Y. Note that the voltage X is not limited to have a negative value lower than 0 V, but may have a positive value. In FIG. 5, an example of decreasing the voltage over time from 0 V to the voltage X is shown, but the voltage may be decreased over time from a value different from 0 V to the voltage X.

[0025] Next, the value of the voltage applied to the electrochemical device 100 is held equal to or lower than the value of the voltage X from the time T2 to a time T3. FIG. 5 illustrates an example of holding the voltage at the value of the voltage X as an example, but the value is not necessarily required to be a constant value. The voltage held until the time T3 then changes to 0 V by stopping of application. On this occasion, the value of the voltage may be increased over time from the value of the voltage X to 0 V.

[0026] Next, in the period P2, the value of the voltage applied to the electrochemical device 100 is increased over time from the time T3 to a time T4 to a value equal to or higher than the voltage Y. The voltage Y is higher than 0 V, for example. In FIG. 5, an example of increasing the voltage from 0 V to the voltage Y over time is shown, but the voltage may be increased

over time from a value different from 0 V to the voltage Y.

[0027] Next, the value of the voltage applied to the electrochemical device 100 is held at the value equal to or higher than the value of the voltage Y from the time T4 to a time T5. In FIG. 5, an example of holding the value at the value of the voltage Y is shown as an example, but the value is not necessarily required to be a constant value. The voltage whose value is held until the time T5 then changes to 0 V by stopping of application. On this occasion, the value of the voltage may be decreased over time from the value of the voltage Y to 0 V.

[0028] Here, it is explained that a spike-shaped nonfaradaic current can be suppressed by changing the voltage wave from the rectangular wave to the trapezoidal wave. FIG. 6 is a schematic view illustrating an electric circuit (equivalent circuit) that represents the inside of an electrochemical device. When the inside of the electrochemical device is represented by an electric circuit, the electric circuit is an RC circuit where a resistor R_s of an electrolytic solution and an electric double layer (capacitor) C_d are connected as illustrated in FIG. 6. FIG. 6 further illustrates a switch SW that controls supply of a voltage E to the electrochemical device and a current i flowing inside the electrochemical device.

[0029] Formula (1) holds for a voltage E_c applied to the electric double layer, a charged electricity quantity q, and a capacitor C_d.

[Formula 1]

$$q = C_d E_c \qquad (1)$$

[0030] If the voltage applied to the electrochemical device is represented by E when the switch SW is in a state of ON, a total sum of a voltage drop iR_s in the electrolytic solution and the voltage E_c applied to the electric double layer is required to be equal to E, so that Formula (2) holds.

[Formula 2]

$$E = i R_s + E_c = i R_s + \frac{q}{C_d} \qquad (2)$$

[0031] Further, the current i is the electricity quantity flowing per unit time and thus can be expressed by Formula (3).

[Formula 3]

$$i = \frac{dq}{dt} \qquad (3)$$

[0032] By substituting Formula (3) into Formula (2), Formula (4) can be obtained.

[Formula 4]

$$\frac{dq}{dt} = \frac{-q}{R_s C_d} + \frac{E}{R_s} \qquad (4)$$

[0033] By solving a differential equation of Formula (4) under the condition that q = 0 when t = 0 for differentiation, a variation of the current i in relation to the time t can be obtained as shown in Formula (5). The current i in this case is a nonfaradaic current.

[Formula 5]

$$i = \frac{E}{R_s} \exp\left(-\frac{t}{R_s C_d}\right) \qquad (5)$$

[0034] FIG. 7 and FIG. 8 are qualitative illustration of Formula (5). FIG. 7 illustrates an example of a temporal variation of the voltage E. FIG. 8 illustrates an example of a temporal variation of the current i. It is found from FIG. 7 and FIG. 8 that a large current flows at the time of voltage application.

[0035] Next, a case where the voltage is applied to the electrochemical device in the trapezoidal wave will be explained. In this case, the voltage E increases gradually with an inclination v in relation to the time t, which can be expressed by Formula (6).

[Formula 6]

$$E = v\,t \qquad (6)$$

[0036] By substituting Formula (6) into Formula (4), Formula (7) can be obtained.
[Formula 7]

$$v\,t = R_s \frac{dq}{dt} + \frac{q}{R_s} \qquad (7)$$

[0037] By solving the above under the condition that q = 0 when t = 0 for further differentiation, Formula (8) can be obtained.
[Formula 8]

$$i = v C_d \left[1 - \exp\left(-\frac{t}{R_s C_d} \right) \right] \qquad (8)$$

[0038] FIG. 9 and FIG. 10 are qualitative illustration of Formula (8). FIG. 9 illustrates an example of the temporal variation of the voltage E. FIG. 10 illustrates an example of the temporal variation of the current i. As obvious from FIG. 9 and FIG. 10, unlike the case of the rectangular wave, a nonfaradaic current gradually increases and finally reaches a constant value determined by the voltage control condition and the electrochemical device. Therefore, compared with the case of applying a voltage in a rectangular wave, it is possible to suppress the large nonfaradaic current that flows instantaneously due to an increase of an absolute value of the voltage over time when reversing the polarity. Note that in FIG. 5, the trapezoidal wave where the voltage is increased linearly in relation to the time is explained, but instantaneous flowing of a large nonfaradaic current can be suppressed also in the case where the voltage is changed curvilinearly. Further, if at least a part of the voltage increases over time, instantaneous flowing of a large nonfaradaic current can be suppressed, for example, also in the case where the absolute value of the voltage is changed in a staircase shape.

[0039] In this embodiment, if the absolute value of the voltage can be increased over time in reversing the polarity of the voltage, a period in which a voltage is not applied may be provided, for example, before the voltage is decreased or increased over time. FIG. 11 is a graph illustrating another example of the temporal variation of the voltage in the embodiment. A horizontal axis of the graph represents a time [second]. A vertical axis of the graph represents a voltage [V] applied to the electrochemical device 100. FIG. 11 illustrates, as an example, an example with a period P3 between the period P1 and the period P2 in FIG. 5, but without being limited to the above, the period P3 is provided before the period P1 or before the period P2. These periods are repeated in the order of the period P1, the period P3, and the period P2 or in the order of the period P2, the period P3, and the period P1. In the period P3, application of the voltage is halted, for example, from the time T3 to a time TX, and the value of the voltage is then changed over time from the time TX to the time T4. By providing the period P3, an electric charge having been charged to an electrode can be suppressed from flowing back to the power supply 200.

[0040] A time necessary for decreasing from 0 V to a value equal to or lower than a value of the voltage X in the period P1 and a time necessary for increasing from 0 V to a value equal to or higher than a value of the voltage Y in the period P2 are preferably 0.6 seconds or longer and further preferably 1 second or longer, respectively. The time of 0.6 seconds or longer can suppress instantaneous flowing of a large nonfaradaic current. An upper limit of the aforementioned time is not limited in particular, but is 60 seconds or shorter, for example. The aforementioned time can be controlled by adjusting a rate of increase of the absolute value of the voltage (inclination of the voltage wave), for example, by the voltage control device 300.

[0041] As described above, in this embodiment, when recovering the valuable resource by the electrolytic reaction, the absolute value of the voltage is increased over time and then held to thereby reverse the polarity of the voltage, so that instantaneous flowing of the large nonfaradaic current can be suppressed. Thus, the valuable resource can be recovered without a power supply having a large output size.

EXAMPLES

(Comparative Example 1, Example 1, 2)

[0042] FIG. 12 illustrates a variation per hour of a current at the time of a test (voltage application test) in which voltages of a rectangular wave and a trapezoidal wave are applied to a cell stack of a fuel cell. A horizontal axis of a graph of FIG. 12 represents a time [second]. A vertical axis of the graph of FIG. 12 represents a current [A] in the cell stack. The voltage

application test was carried out without removing an electrode containing a noble metal from the cell stack.

[0043] In Comparative Example 1, when a voltage of 18.2 V was applied to the cell stack in a rectangular wave at an inclination of smaller than 30.3 V/s for less than 0.6 seconds, a current of 91 A flowed immediately after voltage application, followed by a gradual current decrease to a stationary current of about 20 A. In such a case, a power supply 200 capable of outputting a current of about 100 A is required even though electrolysis operation is performed at the stationary current of 20 A. In contrast, in Example 1, a voltage was applied to the aforementioned cell stack while the voltage was increased at an inclination of 18.2 V/s in a trapezoidal wave for 1 second and then held at 18.2 V, a current gradually increased from 0 A to a maximum current of 35 A, and finally a stationary current of about 20 A the same as in the case of the rectangular wave flowed. Further, in Example 2, a voltage was applied to the aforementioned cell stack while the voltage was increased at an inclination of 30.3/s in a trapezoidal wave for 0.6 seconds and then held at 18.2 V, a current gradually increased from 0 A to a maximum current of 37.5 A, and finally a stationary current of about 20A the same as in the case of the rectangular wave flowed. From the results above, it was confirmed that reversing the polarity of the voltage by increasing the absolute value of the voltage over time and then holding that voltage can suppress instantaneous flowing of a large nonfaradaic current and that a power supply 200 capable of outputting a current of 40 to 50 A is sufficient if a wave voltage is applied in a trapezoidal wave, for example.

[0044] While certain embodiments have been described, these embodiments have been presented by way of example only, and are not intended to limit the scope of the inventions. Indeed, the novel embodiments described herein may be embodied in a variety of other forms; furthermore, various omissions, substitutions and changes in the form of the embodiments described herein may be made without departing from the spirit of the inventions. The accompanying claims and their equivalents are intended to cover such forms or modifications as would fall within the scope and spirit of the inventions.

Claims

1. A voltage control method for applying a voltage to an electrochemical device that has an electrode with a catalyst containing a noble metal and periodically reversing a polarity of the voltage to perform an electrolytic reaction and recover a valuable resource from the electrochemical device,

   the voltage control method comprising
   continuously increasing an absolute value of the voltage and then holding the value while periodically reversing the polarity of the voltage.

2. The method according to claim 1, comprising:

   a first period in which the value of the voltage is decreased linearly or curvilinearly from a first value to equal or lower than a second value that is lower than the first value and then held; and
   a second period in which the value of the voltage is increased linearly or curvilinearly from a third value the same as or different from the first value to equal to or higher than a fourth value that is higher than the third value and then held.

3. The method according to claim 2, comprising:
   a third period in which the voltage is not applied between the first period and the second period.

4. The method according to claim 2, wherein

   a time necessary for the value of the voltage to reach the second value from the first value is 0.6 seconds or longer, in the first period, and
   a time necessary for the value of the voltage to reach the fourth value from the third value is 0.6 seconds or longer, in the second period.

5. The method according to claim 1, wherein
   the catalyst contains at least one noble metal selected from the group consisting of platinum, iridium, ruthenium, rhodium, palladium, gold, and rhenium.

FIG. 1

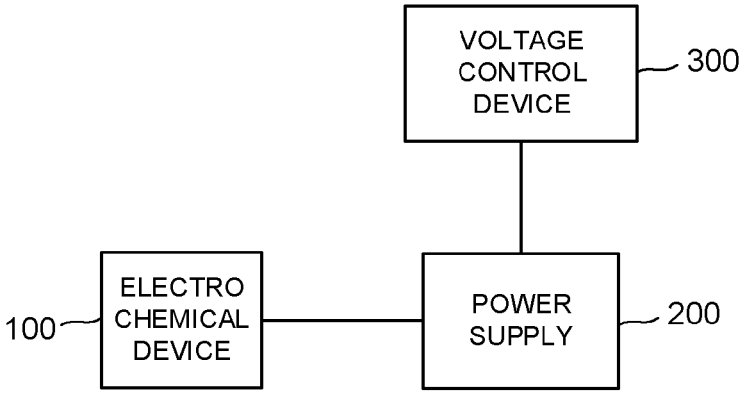


FIG. 2

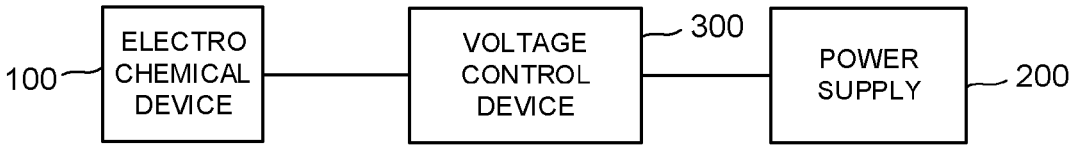


FIG. 3

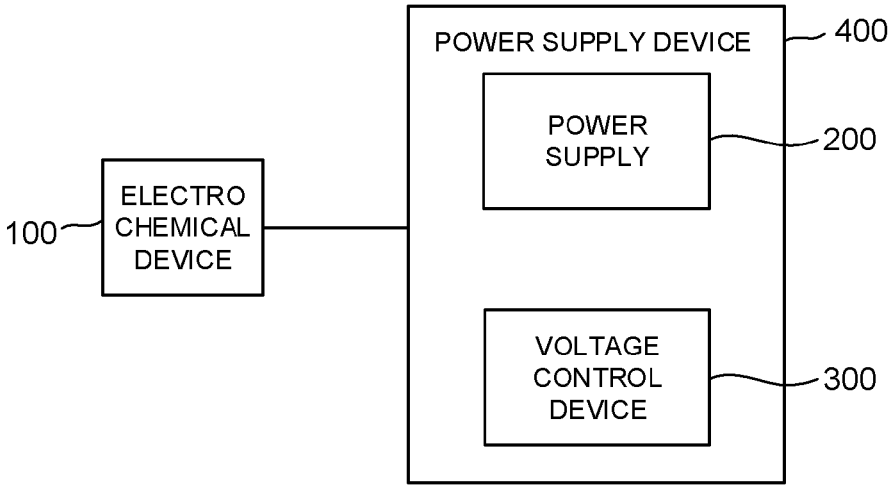
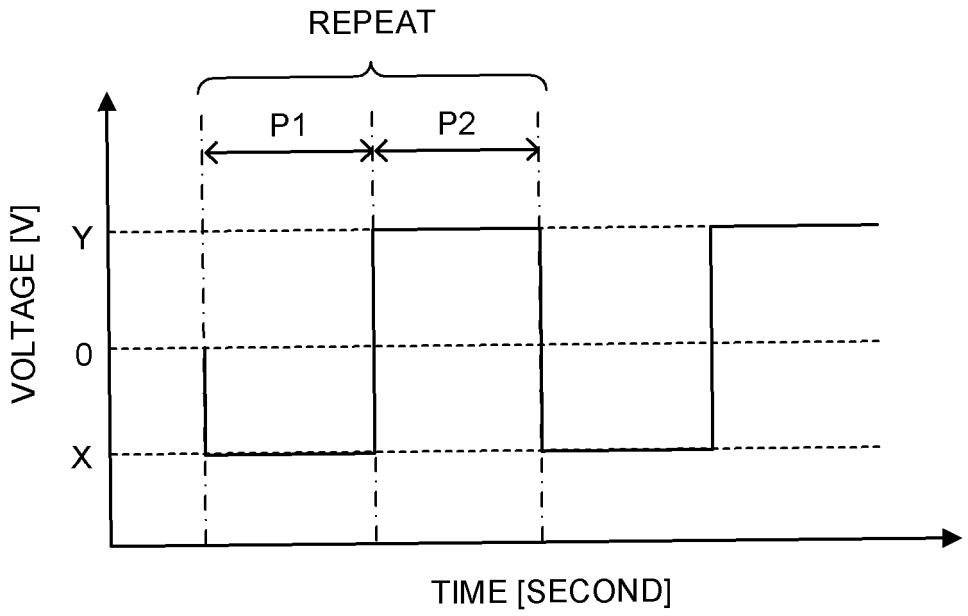


FIG. 4


REPEAT
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P2
VOLTAGE [V]
Y
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X
TIME [SECOND]

FIG. 5

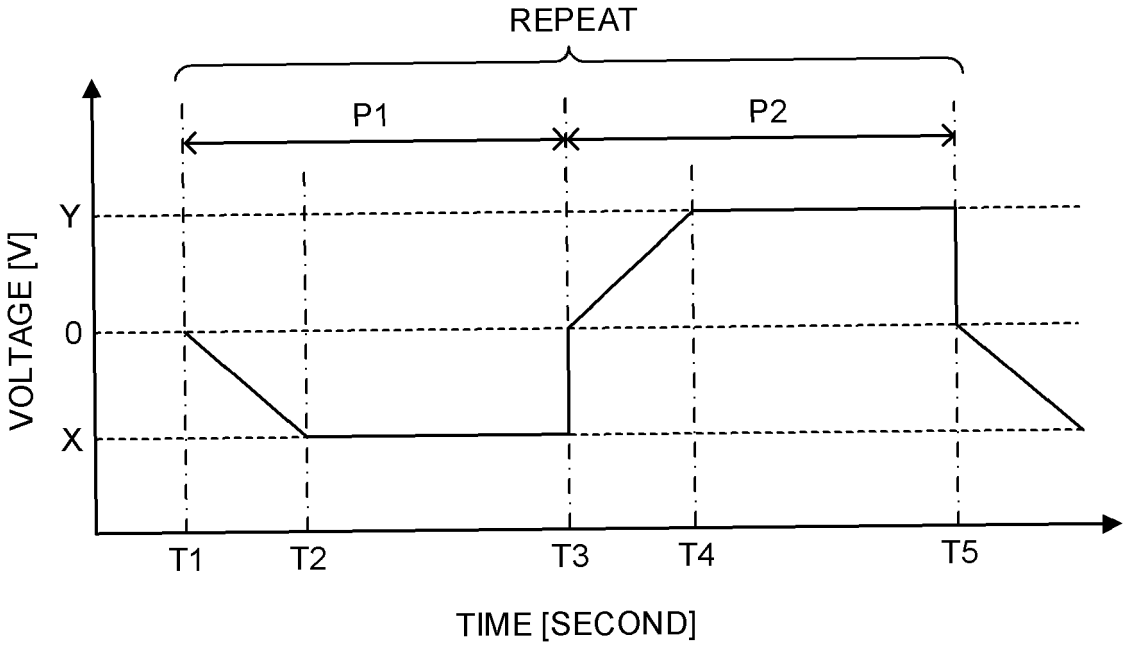

REPEAT
P1
P2
VOLTAGE [V]
Y
0
X
T1
T2
T3
T4
T5
TIME [SECOND]

FIG. 6

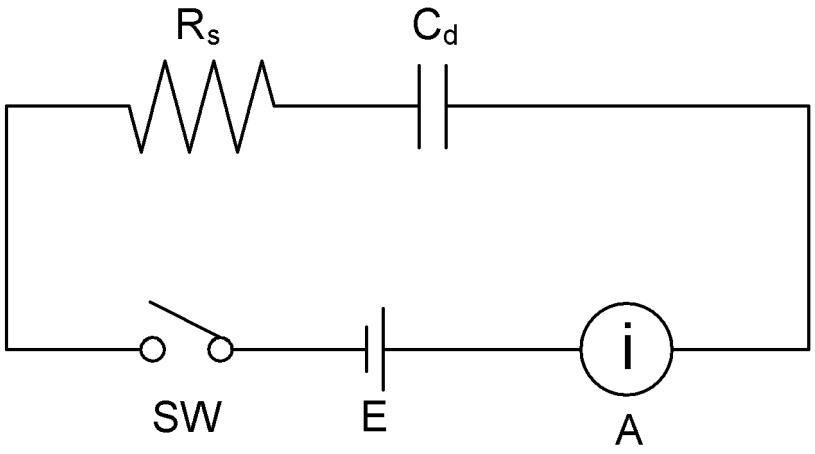


FIG. 7

FIG. 8

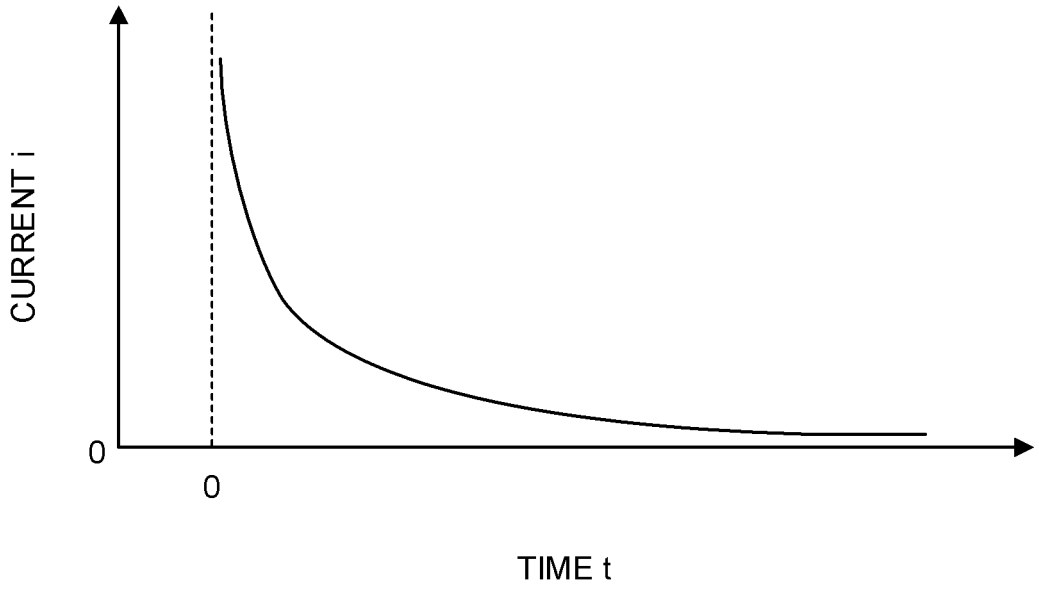


FIG. 9

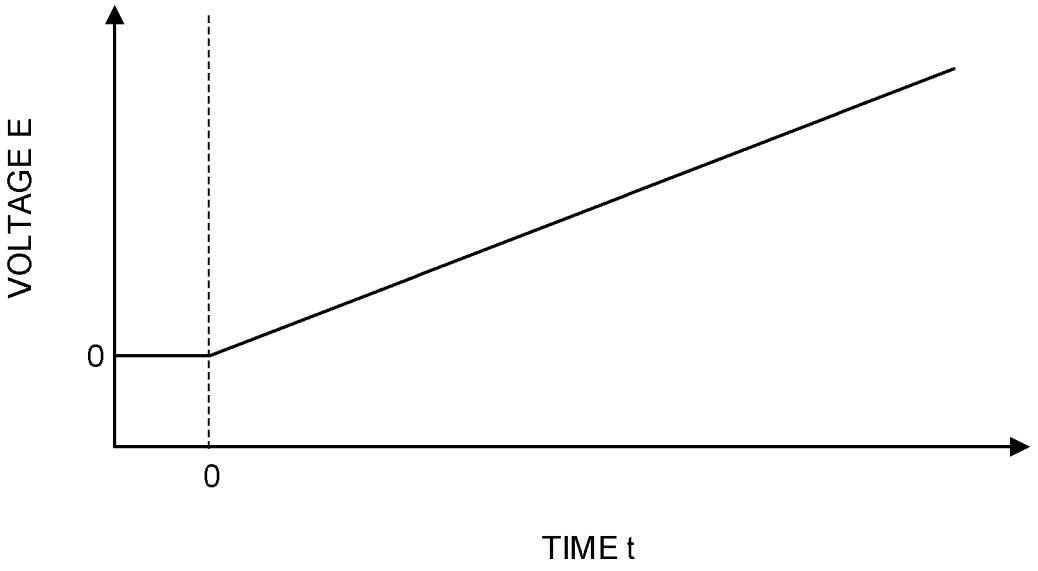


FIG. 10

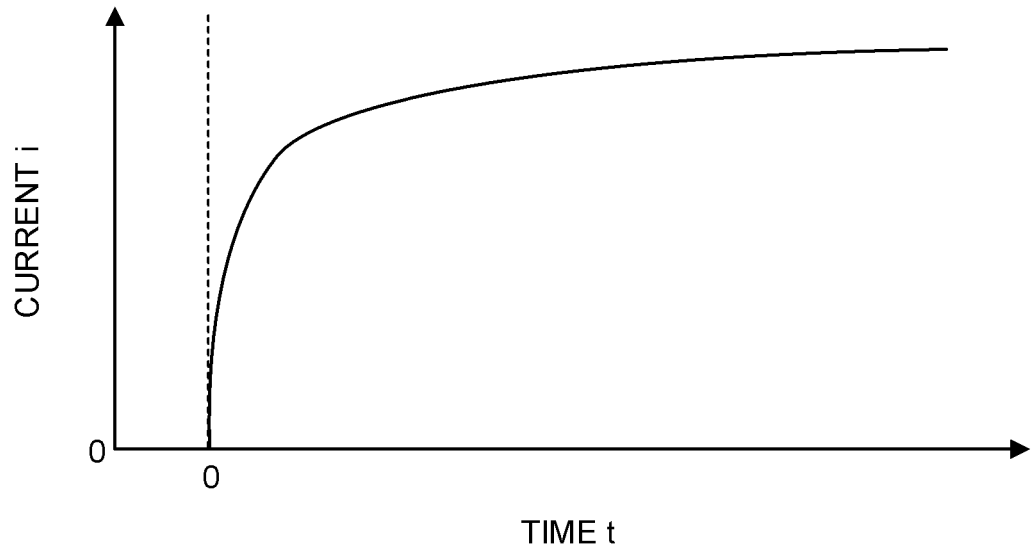


FIG. 11

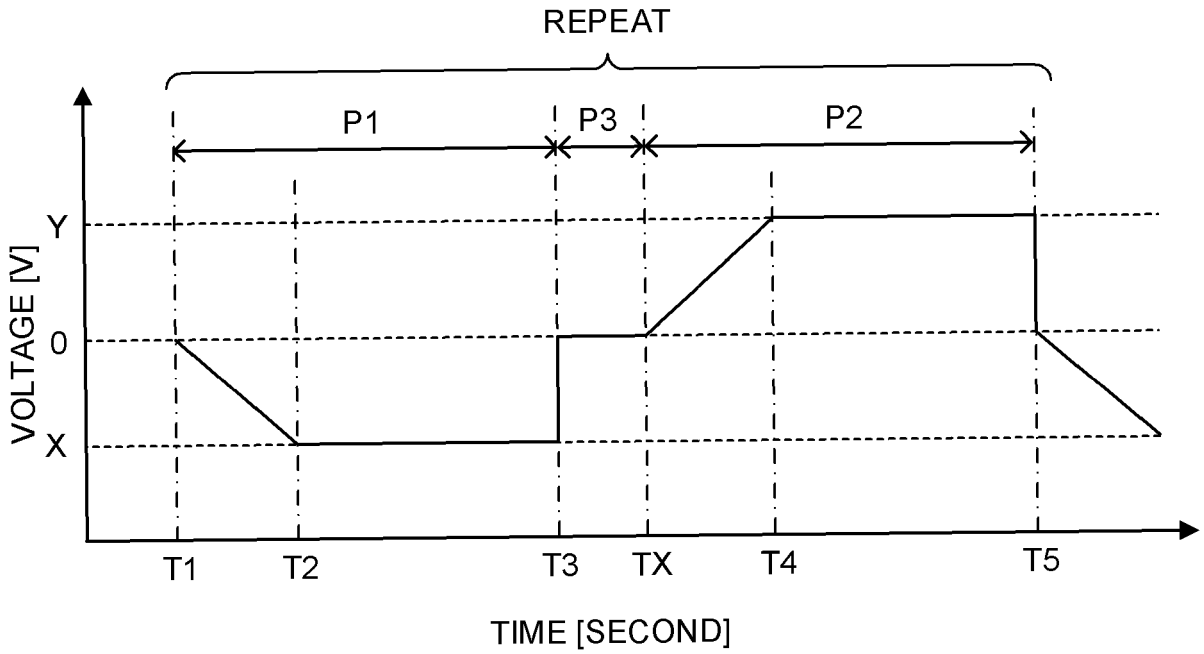
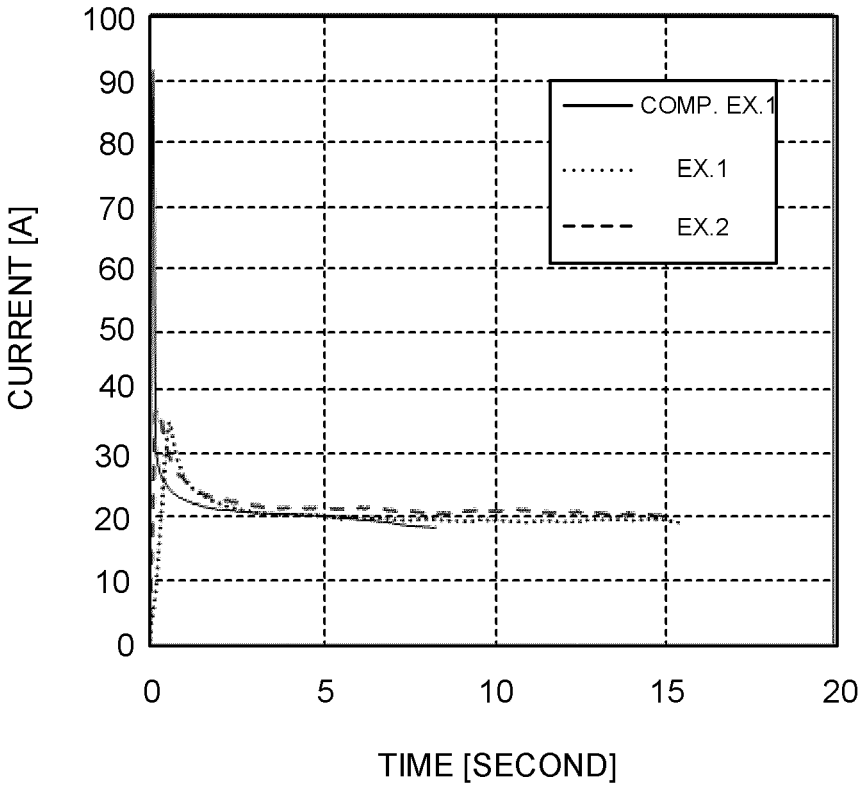


FIG. 12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80
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50
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20
10
0
CURRENT [A]
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5
10
15
20
TIME [SECOND]
COMP. EX.1
EX.1
EX.2

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EUROPEAN SEARCH REPORT

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