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(54) Title: A METHOD OF GENERATING OXYGEN BY ELECTROCHEMICAL WATER SPLITTING AT OPTIMIZED CONDITIONS OF PH, TEMPERATURE, AND PRESSURE

(57) Abstract: The present disclosure relates to an efficient method of water splitting for large/industrial scale production of oxygen at the anode in an electrolyzer, while at the same time permitting hydrogen production at the cathode of the same electrolyzer. More particularly, the present disclosure provides optimal operating conditions such as temperature, pressure, and pH for a range of potentials for an electrochemical water splitting reactor to generate the maximum amount of oxygen at a given voltage while supplying the minimum possible amount of alkali.

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A METHOD OF GENERATING OXYGEN BY ELECTROCHEMICAL WATER SPLITTING AT OPTIMIZED CONDITIONS OF pH, TEMPERATURE, AND PRESSURE

STATEMENT REGARDING FEDERALLY SPONSPORED RESEARCH FOR DEVELOPMENT

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TECHNICAL FIELD

The present disclosure relates to the field of electrolysis and electrochemical water splitting into oxygen and hydrogen. In particular, the present disclosure relates to an efficient method of water splitting for large/industrial scale production of oxygen at the anode in an electrolyzer, while simultaneously permitting the large/industrial scale production of hydrogen at the cathode. More particularly, the present disclosure provides optimal operating conditions such as temperature, pressure, and pH for an electrochemical water splitting reactor to generate the maximum amount of oxygen at a given applied potential.

BACKGROUND OF THE DISCLOSURE

Oxygen (O_2) is a major component of the Earth's atmosphere and is also one of the most abundant elements on Earth. Oxygen, apart from keeping us alive, has a variety of applications. In particular, the relatively pure form of oxygen (liquid or compressed) has numerous industrial and medical applications. For instance, pure oxygen finds application in steel manufacturing, the paper-pulp industry, metal production and fabrication, glass manufacturing, the petrochemical industry, as the breathing gas in space suits, etc. Furthermore, oxygen has several use cases in healthcare, including in hospitals, outpatient treatment centers, and people's homes. Indeed, supplemental oxygen supply at home and in hospitals is critical for the survival of people suffering from breathing disorders such as emphysema, bronchitis, chronic obstructive pulmonary disease, etc.

The primary method for the industrial production of oxygen is the fractional distillation of liquefied air. In recent years, however, another alternative has emerged – the electrolysis of water into molecular oxygen and hydrogen. The electrolysis of water is considered an

environmentally friendly method to produce oxygen and hydrogen gases. Electrolysis involves the passage of electric current through two electrodes – known as the anode and the cathode – connected via an electric circuit, resulting in various chemical reactions involving constituents of the electrolyte solution. In water electrolysis, the electrolyte is an aqueous solution containing an acid or an alkali. Further, the electrodes are made using carefully chosen materials, with different types of materials considered for the anode and the cathode. The reactions taking place at the anode and the cathode are, respectively, the oxidation and reduction of various aqueous species within the electrolyte. Upon the application of a potential difference across the electrodes, the negatively charged hydroxide ions (OH^-) migrate to the positively charged anode where water/OH^- oxidation, i.e., the oxygen evolution reaction (OER) takes place and oxygen gas (O_2) evolves. Simultaneously, the positively charged protons (H^+) diffuse to the negatively charged cathode where water/proton reduction, i.e., the hydrogen evolution reaction (HER) takes place and hydrogen gas (H_2) evolves. The overall reaction during water electrolysis is: $H_2O \rightarrow H_2(g) + \frac{1}{2}O_2(g)$.

The electrode potential, solution pH, temperature, and pressure are key operating conditions for any (photo)electrochemical water splitting reactor, as depicted in Figure 1A. Several studies have investigated the influence of the electrode potential and solution pH on OER kinetics. The temperature-dependent oxygen evolution activity of various electrocatalysts also has been investigated experimentally. Fewer experimental studies have dealt with the pressure-dependence of OER kinetics. One study investigated the oxygen partial pressure's role in modulating the oxygen vacancy concentration in $NdNiO_3$, and thereby the OER activity. A second one studied the effect of pressure on the OER mechanism. Others have focused on the combined effect of temperature and pressure on water electrolysis. However, most of the studies of the thermodynamics and kinetics of the OER only consider conditions of room temperature (25°C) and pressure (1.013 bar) and there are no studies/research or experimentations examining the pressure-dependence of OER thermodynamics and kinetics under optimized reactor operating conditions which would result in improving the commercial water splitting reaction conditions. Thus, there is a need to understand the gaps pertaining to the direct effect of temperature and pressure on OER thermodynamics and kinetics and deployment of such understanding of the operating conditions to commercial water splitting reactions.

Further, commercial water splitting electrolyzers need to operate at elevated temperatures (~90 °C) and pressure (~100 bar) to achieve faster kinetics and produce oxygen and hydrogen in commercially viable quantities. Between the two-half reactions, i.e., the OER and HER constituting electrochemical water splitting, the OER is significantly more sluggish and presents a thermodynamic and kinetic bottleneck to the electrolysis of water. Since the OER is favoured under alkaline operating conditions, researchers have examined the possibility of alkaline water electrolysis using aqueous solutions of potassium hydroxide (KOH) and sodium hydroxide (NaOH) as the electrolyte. However, achieving high current densities of > 1000 mA/cm^2 in an environmentally friendly and cost-effective manner is one of the key challenges that needs to be surmounted before such a technology can be commercialized. To increase the current density in the water electrolysis process, one needs to optimise the key operating conditions used in the electrochemical water splitting reactor, i.e., the pH, temperature, pressure, and potential. This objective needs to be met while at the same time reducing the concentration of the alkali used in the electrolyte, so as to make the process more environmentally friendly, less corrosive to the reactor, and more economically viable via the use of smaller quantities of alkali. The present disclosure tries to address these needs outlined above.

SUMMARY OF THE DISCLOSURE

The present disclosure provides an efficient, eco-friendly, and cost-effective method for the production of pure oxygen and hydrogen by electrochemical water splitting in an electrolyzer. In particular, the present disclosure provides optimized operating conditions of an electrochemical water oxidation reaction such as optimal temperature, pressure, and pH (i.e., alkali content), at a given voltage to drive the oxygen evolution reaction (OER) in the presence of a metal-doped oxide, hydroxide, or oxyhydroxide catalyst (or electrocatalyst) for the production of oxygen at the anode.

Additionally, the present disclosure provides optimized operating conditions of an electrochemical water oxidation reaction, such as optimal temperature, pressure, pH, and potential, that result in a significant reduction in the required alkali concentration in the aqueous solution and improvement in the OER current density. In particular, the method of the present disclosure optimizes the pH of the electrolyte as a function of temperature and pressure, while consequently resulting in a significant reduction in the concentration of the alkali required in the electrolyte solution.

Accordingly, the present disclosure provides a method of generating oxygen by electrochemical water splitting at optimized conditions of pH, temperature, and pressure for an oxygen evolution reaction (OER) current density ranging from about 10 mA/cm^2 to about 10000 mA/cm^2, said method comprising:

a) contacting an anode of an electrolyzer with an electrolyte comprising water and alkali, in the presence of a metal-doped oxide, hydroxide, or oxyhydroxide catalyst; and

b) applying a potential ranging from about 1.45 V to about 1.75 V versus (vs.) the reversible hydrogen electrode (RHE) at the anode to allow said generation of the oxygen;

wherein pH of the electrolyte ranges from about 7.4 to about 10.9;

wherein the pH is a function of temperature T at a given pressure P; and

wherein the temperature T ranges from about 28 °C to 151 °C and is less than the boiling point of the electrolyte at the given pressure P.

In some embodiments of the present disclosure, the pH of the electrolyte is determined based on an unconstrained minimization function applied to the negative of an oxygen evolution reaction (OER) current density (equivalent to the maximization of the OER current density), and wherein the OER current density is a function of the applied potential.

In some embodiments of the present disclosure, the pH of the electrolyte is maintained in a manner that allows concentration of OH^- ions in the electrolyte to be less than 1 M.

In some embodiments of the present disclosure, the OER current density is improved by at least twofold when compared to any method performed outside the stated potential ranging from about 1.45 V to about 1.75 V versus the reversible hydrogen electrode (RHE) at the anode, stated pH range of about 7.4 to about 10.9 and the stated temperature range from about 28 °C to about 151 °C at a given pressure P, wherein the temperature is less than the boiling point of the electrolyte at the given pressure.

In some embodiments of the present disclosure, the OER current density is improved by up to 1000-fold when compared to a method performed outside the stated potential ranging from about 1.45 V to about 1.75 V versus the reversible hydrogen electrode (RHE) at the anode,

stated pH range of about 7.4 to about 10.9 and the stated temperature range from about 28 °C to about 151 °C at a given pressure P, wherein the temperature is less than the boiling point of the electrolyte at the given pressure.

In some embodiments of the present disclosure, the OER current density improves by at least about 80% when the method is carried out at said optimized conditions of pH, temperature and pressure for the potential range as described herein.

In some embodiments of the present disclosure, the OER current density improves by up to 200% when the method is carried out at said optimized conditions of pH, temperature and pressure for the potential range as described herein.

In some embodiments of the present disclosure, the potential is about 1.5 V vs. the RHE at the anode, the pH of the electrolyte ranges from about 7.4 to about 10.7, and the temperature *T* ranges from about 28 °C to about 151 °C and is less than the boiling point of the electrolyte at the given pressure *P*.

In some embodiments of the present disclosure, the potential is about 1.6 V vs. the RHE at the anode, the pH of the electrolyte ranges from about 8.5 to about 10.9, and the temperature *T* ranges from about 28 °C to about 92 °C and is less than boiling point of the electrolyte at the given pressure *P*.

In some embodiments of the present disclosure, the potential is about 1.7 V vs. the RHE at the anode, the pH of the electrolyte ranges from about 10.2 to about 10.9, and the temperature *T* ranges from about 28 °C to about 41 °C and is less than boiling point of the electrolyte at the given pressure *P*.

In some embodiments of the present disclosure, for maintaining the pH range of about 7.4 to about 10.9 of the present disclosure, the concentration or amount of the required alkali in the electrolyte range from about 0.07 mM to about 1.38 mM, including all values and ranges therebetween.

In some embodiments of the present disclosure, the alkali is selected from a group comprising potassium hydroxide (KOH), sodium hydroxide (NaOH), calcium hydroxide ($Ca(OH)_2$), magnesium hydroxide ($Mg(OH)_2$), or any other alkali or alkaline earth hydroxide.

In some embodiments of the present disclosure, the pH reduces from about 10.7 to about 7.4 with an increase in temperature and the corresponding increase in the OER current density at the potential of about 1.5 V vs. the RHE; and wherein the concentration or amount of the alkali required for maintaining the stated pH reduces from about 1.38 mM to about 0.07 mM at the given pressure *P*.

In some embodiments of the present disclosure, the pH reduces from about 10.9 to about 8.5 with an increase in temperature and the corresponding increase in the OER current density at the potential of about 1.6 V vs. the RHE; and wherein the concentration or amount of the alkali required for maintaining the stated pH reduces from about 1.37 mM to about 0.14 mM at the given pressure *P*.

In some embodiments of the present disclosure, the pH reduces from about 10.9 to about 10.2 with an increase in temperature and the corresponding increase in the OER current density at the potential of about 1.7 V vs. the RHE; and wherein the concentration or amount of the alkali required for maintaining the stated pH reduces from about 1.37 mM to about 0.47 mM at the given pressure *P*.

In some embodiments of the present disclosure, the metal-doped oxide, hydroxide, or oxyhydroxide catalyst is an iron-doped nickel oxide, hydroxide, or oxyhydroxide catalyst, which is also referred to as a mixed iron/nickel oxide, hydroxide, or oxyhydroxide material.

In some embodiments of the present disclosure, the potential applied across the electrolyzer ranges from about 1.45 V to about 1.75 V, including all values and ranges therebetween.

In some embodiments of the present disclosure, the electrolyzer further comprises a cathode within the electrolyte, and wherein application of a potential across the electrolyzer generates oxygen at the anode and hydrogen at the cathode.

In some embodiments of the present disclosure, the method comprises an electrolyte comprising water and KOH or NaOH having a concentration of about 0.15 mM, and wherein the catalyst is a mixed iron/nickel oxide, hydroxide, or oxyhydroxide material, the potential applied at the anode is 1.51 V versus the reversible hydrogen electrode (RHE), the temperature is about 90 ºC, the pressure is about 1 bar, the pH of the electrolyte is maintained at 8.6, and the OER current density is about 997 mA/cm^2.

In some embodiments of the present disclosure, the method comprises electrolyte comprising water and KOH or NaOH having a concentration of about 0.15 mM, the catalyst is of a mixed iron/nickel oxide, hydroxide, or oxyhydroxide material, the potential applied at the anode is 1.5 V versus the RHE, the temperature is about 90 ºC, the pressure is about 1 bar, the pH of the electrolyte is maintained at 8.6, and the OER current density is about 770 mA/cm^2.

In some embodiments of the present disclosure, the method comprises electrolyte comprising water and alkali (KOH or NaOH) having a concentration of about 0.26 mM, the catalyst is a of a mixed iron/nickel oxide, hydroxide, or oxyhydroxide material, the potential applied at the anode is 1.5 V versus the RHE, the temperature is about 100 ºC, the pressure is about 100 bar, the pH of the electrolyte is maintained at 8.6, and the OER current density is about 1241 mA/cm^2.

In some embodiments of the present disclosure, the method comprises electrolyte comprising water and KOH or NaOH having a concentration of about 0.27 mM, the catalyst is of a mixed iron/nickel oxide, hydroxide, or oxyhydroxide material, the potential applied at the anode is 1.5 V versus the RHE, the temperature is about 110 ºC, the pressure is about 200 bar, the pH of the electrolyte is maintained at 8.5 and the OER current density is about 1960 mA/cm^2.

BRIEF DESCRIPTION OF THE ACCOMPANYING FIGURES

In order that the present disclosure may be readily understood and put into practical effect, reference will now be made to exemplary embodiments as illustrated with reference to the accompanying figures. The figures together with the detailed description below, are incorporated in and form part of the specification, and serve to further illustrate the embodiments and explain various principles and advantages, where:

Figure 1 depicts – **(A)** a schematic diagram of a water splitting electrolyzer with the OER and the HER occurring at the anode and the cathode, respectively. pH, applied potential (U), temperature (T), and pressure (p) constitute the main operating conditions of the electrolyzer. **(B)** a graph of the standard Gibbs free energies of H_2O(l), H_2(g), and O_2(g) as a function of temperature, referenced to their respective values at room temperature (T_r =10 °C). **(C)** a graph of the standard Gibbs free energies of the four adsorbed species in the OER mechanism on Fe-doped β-NiOOH, referenced to their respective values at T_r, calculated using the surface normal modes from another reference. **(D)** a graph of the standard reduction potentials of the OER and the four elementary steps in the OER mechanism on Fe-doped β-NiOOH($\bar{1}2\bar{1}1$) as a function of temperature, with the thermodynamic overpotential (η_{thermo}) indicated.

Figure 2 illustrates graphs of the vapor-phase oxygen mole fraction, y_{O_2}, as a function of pressure at two different temperatures, 25 °C and 90 °C, and a fixed pH of 12.0. **(A)** depicts the vapor-phase oxygen mole fraction (y_{O_2}), **(B)** depicts the aqueous activity of oxygen (a_{O_2}).

Figure 3 depicts graphs of: **(A)** The equilibrium constant for water dissociation, Kw, as a function of temperature. **(B)** Water, **(C)** OH^-, and **(D)** K^+ activity as a function of pH under alkaline conditions at 25 °C and 90 °C (green). The system pressure is 1 bar in all cases.

Figure 4 illustrates a plot of standard pH with respect to OH^-, i.e., the pH at which $a_{OH^-} =$ 1 M, denoted as pH^0, as a function of temperature. The optimal pH for maximal OER current density, pH^{opt}, calculated at an applied potential of 1.51 V vs. the RHE, is shown as a function of temperature.

Figure 5 illustrates the OER current density as a function of temperature at an electrolyte pH of pH^0(T) (blue) or pH^{opt}(T) (red) at an applied potential of 1.51 V vs. the **(A)** RHE and **(B)** standard hydrogen electrode (SHE). The vertical axis in panel A is in a linear scale while in panel B it is in a log scale. The system pressure is 1 bar in both cases.

Figure 6 illustrates the K^+ activity, i.e., the initially supplied KOH concentration, required to maintain the solution at the optimal pH (Fig. 4) as a function of temperature.

Figure 7 illustrates **(A)** The potential versus the SHE required to obtain the maximum current density at pH^{opt}. **(B)** The difference between the absolute potentials (i.e., versus the SHE) at pH^{opt} and pH^{0} to obtain the same current density as at pH^{0}.

Figure 8 illustrates graphs of **(A)** Reaction Gibbs free energies of water splitting, the OER, and the HER as a function of pressure at a fixed temperature of 90 °C, assuming pure reactant and product species. Note the split vertical axis on the plot. Only the free energies at a pressure of 1 bar are standard values. O_2 and H_2 mole fractions assumed as 1.0 in their respective gas-evolution compartments. **(B)** Reduction potentials of the OER and the four elementary steps in the OER mechanism on Fe-doped β-NiOOH as a function of pressure at a fixed temperature of 90 °C.

Figure 9 illustrates a graph of the solubility of oxygen in water as a function of the applied pressure with the vapor-phase oxygen mole fraction maintained at unity, at 10 °C and 90 °C.

Figure 10 illustrates a graph of the OER current density as a function of pressure at an electrolyte pH of pH^{opt} and pH^{0} and applied potentials of (A) 1.46 V and (B) 1.51 V vs. the RHE. Note that the reactor (system) temperature, T, in both panels is 90 °C.

DESCRIPTION OF THE DISCLOSURE

Unless otherwise defined, all terms used in the disclosure, including technical and scientific terms, have meaning as commonly understood by one of ordinary skill in the art to which this invention belongs. By means of further guidance, definitions for some terms are included for better understanding of the present disclosure.

As used herein, the singular forms 'a', 'an' and 'the' include both singular and plural referents unless the context clearly dictates otherwise.

The term 'comprising', 'comprises' or 'comprised of' as used herein are synonymous with 'including', 'includes', 'containing' or 'contains' and are inclusive or open-ended and do not exclude additional, non-recited members, elements or method steps.

The recitation of numerical ranges by endpoints includes all numbers and fractions subsumed within the respective ranges, as well as the recited endpoints.

The term 'about' as used herein when referring to a measurable value such as a parameter, an amount, a temporal duration, and the like, is meant to encompass variations of ±10% or less, preferably ±5% or less, more preferably ±1% or less and still more preferably ±0.1% or less of and from the specified value, insofar such variations are appropriate to perform the present disclosure. It is to be understood that the value to which the modifier 'about' refers is itself also specifically, and preferably disclosed.

As used herein, the term/phrase 'electrolysis of water', or 'electrochemical water oxidation', or 'electrochemical water splitting' are used interchangeably and refers to the process of using electricity to decompose water into oxygen and hydrogen gas as described in this disclosure. The overall process of water electrolysis consists of two half-cell reactions: the hydrogen evolution reaction (used hereinafter as "HER") and the oxygen evolution reaction (used hereinafter as "OER"). In the HER, water/protons is/are reduced at the cathode to produce H_2, and in the OER, water/OH^- ions is/are oxidized at the anode to produce O_2. Currently, three main types of electrolysis technologies are being examined: (1) proton exchange membrane (PEM) electrolysis, (2) alkaline electrolysis, and (3) high-temperature solid oxide water electrolysis.

As used herein, the term/phrase 'electrolyzer' or 'electrochemical water splitting reactor' are interchangeable and refer to a system that uses electric current to decompose water into hydrogen and oxygen molecules in a process called electrolysis, as defined above. In its most basic form, an electrolyzer consists of a cathode (negatively charged electrode), an anode (positively charged electrode), and a membrane. The system also contains ancillary components, including, but not limited to, pumps, vents, measuring devices, storage tanks, a power supply, and a separator.

As used herein, the term/phrase 'current density' refers to the measure of the amount of electric charge per unit time that flows through a unit area of the cross-section. The area is measured in the direction perpendicular to the flow of current, and the resultant current density is expressed in amperes per square meter (or in other derived units, such as milliamperes per square centimeter). Usually, the more the current in a conductor, the higher will be the current density. In the context of the present disclosure, the conductor is the catalyst material as defined

above, and the higher the current density associated with the water splitting process, the higher will be the generation rate of oxygen and hydrogen.

As used herein, the term/phrase 'SHE' or 'standard hydrogen electrode' refers to an electrode that is used as a reference for measuring the potential of a working electrode. The value of the SHE potential is, by convention, zero at all temperatures and forms the basis with respect to which cell potentials are measured. The SHE consists of a 1.0 M H^+(aq) solution in a tube with a square piece of platinized platinum immersed in it. The SHE is connected via a platinum wire to the working electrode.

As used herein, the term 'RHE' or 'reversible hydrogen electrode' refers to an electrode that is used as a reference for measuring the potential of a working electrode. Unlike the SHE, the electrolyte in the RHE is not a 1.0 M H^+ (aq) solution, but is instead the same electrolyte in which the working electrode is immersed.

Accordingly, with an aim to overcome the drawbacks associated with the currently existing methods of electrochemical water splitting, the present disclosure provides an improved method for production of oxygen by water electrolysis.

Therefore, an objective of the present disclosure is to develop an efficient, eco-friendly, and cost-effective industrial/large scale method for the production of pure oxygen, while producing hydrogen as a by-product.

Another objective of the present disclosure is to optimize the operating conditions of pH, temperature, and pressure for a range of potentials for electrochemical water splitting reaction for production of oxygen at an industrial scale.

Another objective of the present disclosure is to optimize the pH as a function of temperature and pressure to produce the maximum possible amount of oxygen gas for a given applied potential.

Another objective of the present disclosure is to reduce the concentration of the alkali solution required for maintaining the optimized pH as mentioned above and develop an eco-friendly, economical electrochemical water oxidation method.

Still another objective of the present disclosure is to understand the gaps pertaining to the direct effect of temperature and pressure on OER thermodynamics and kinetics, rather than through indirect mechanisms such as phase stability and defect concentrations.

Yet another objective of the present disclosure is to develop a method of water electrolysis that results in generation of oxygen with minimal impact on the kinetics of production by means of achieving a maximal current density.

Another objective of the present disclosure is to develop a method for the production of oxygen by electrochemical water splitting that can be carried out in conjunction with a counter-electrode for the carbon-dioxide-emission-free production of hydrogen gas.

Yet another objective of the present disclosure is to provide a device and a process for generation of oxygen by electrochemical water oxidation, that employ the method developed in the present disclosure, at optimal conditions of pH, temperature, and pressure.

To achieve the aforesaid objectives, the present disclosure provides an efficient, eco-friendly, and cost-effective method for production of pure oxygen by electrochemical water oxidation in an electrolyzer. In particular, the present disclosure provides optimized operating conditions of an electrochemical water oxidation reaction such as optimal temperature, pressure, and pH (i.e., alkali content), at a given voltage to drive the oxygen evolution reaction (OER) in the presence of mixed iron/nickel oxide, hydroxide, or oxyhydroxide catalyst for the production of oxygen at the anode.

More particularly, the present disclosure provides optimized operating conditions of an electrochemical water oxidation reaction such as optimal temperature, pressure, pH, and potential that results in a significant reduction in the required alkali concentration in the electrolyte and an improvement in the OER current density. In particular, the method of the present disclosure optimizes the pH of the electrolyte as a function of temperature and pressure, that consequently also results in significant reduction of the optimal alkali concentration in the electrolyte.

The present inventors have analyzed the coupled effects of temperature, pressure, and pH on the thermodynamics and kinetics of the OER electrocatalyzed by a mixed iron/nickel oxyhydroxide catalyst, in order to understand overall electrochemical water-splitting cycles. To this end, the present inventors investigated the thermodynamics and kinetics of elementary steps in electrochemical water splitting and determined the temperature variations of the free energies of molecular and adsorbed species, respectively, to calculate the OER free energy as a function of temperature.

In some embodiments, the present disclosure provides a method of generating oxygen by electrochemical water splitting at optimized conditions of pH, temperature, and pressure for an oxygen evolution reaction (OER) current density ranging from about 10 mA/cm^2 to about 10000 mA/cm^2, said method comprising:

a) contacting an anode of an electrolyzer with an electrolyte comprising water and alkali, in presence of a iron-doped nickel oxide, hydroxide, or oxyhydroxide catalyst; and

b) applying a potential ranging from about 1.45 V to about 1.75 V versus the reversible hydrogen electrode (RHE) at the anode to allow said generation of the oxygen;
wherein pH of the electrolyte ranges from about 7.4 to about 10.9;
wherein the pH is a function of temperature T at a given pressure P; and
wherein the temperature T ranges from about 28 °C to 151 °C and is less than boiling point of the electrolyte at the given pressure P.

In some embodiments of the present disclosure, the pH of the electrolyte ranges from about 7.4 to about 10.9, including all values and ranges therebetween.

In some embodiments of the present disclosure, the optimal pH of the electrolyte is about 7.4, about 7.5, about 7.6, about 7.7, about 7.8, about 7.9, about 8.0, about 8.1, about 8.2, about 8.3, about 8.4, about 8.5, about 8.6, about 8.7, about 8.8, about 8.9, about 9.0, about 9.1, about 9.2, about 9.3, about 9.4, about 9.5, about 9.6, about 9.7, about 9.8, about 9.9, about 10.0, about 10.1, about 10.2, about 10.3, about 10.4, about 10.5, about 10.6, about 10.7, about 10.8, or about 10.9, including all values and ranges therebetween.

In some embodiments of the present disclosure, the pH of the electrolyte is a function of temperature T at a given pressure P of the electrolyzer and a given potential applied at the anode.

In some embodiments of the present disclosure, the pH of the electrolyte changes with the change in the temperature T at a given pressure P and a given potential applied at the anode.

In some embodiments of the present disclosure, the pH of the electrolyte decreases with the increase in the temperature T at a given pressure P and a given potential applied at the anode.

In some embodiments of the present disclosure, pH of the electrolyte is determined based on an unconstrained minimization function applied to the negative of an oxygen evolution reaction (OER) current density (equivalent to the maximization of the OER current density), and wherein the OER current density is a function of the applied potential.

In some embodiments of the present disclosure, the pH of the electrolyte is maintained in a manner that allows concentration of OH^- ions in the electrolyte to be less than 1 M.

In some embodiments, the electrolyte is a fluid comprising water and alkali.

In some embodiments of the present disclosure, the optimal temperature T ranges from about 28 °C to about 151 °C, including all values and ranges therebetween.

In some embodiments of the present disclosure, the optimal temperature T is less than the boiling point of the electrolyte at a given pressure P.

In some embodiments of the present disclosure, the optimal temperature T ranges from about 28 °C to about 151 °C, including all values and ranges therebetween, and is less than the boiling point of the electrolyte at a given pressure P.

In some embodiments of the present disclosure, the optimal temperature T ranges from about 28 °C to about 151 °C, including all values and ranges therebetween, and is less than the boiling point of the electrolyte at a given pressure P, and does not include temperatures below 28 °C.

In some embodiments of the present disclosure, the optimal temperature *T* is about 28 °C, about 30 °C, about 35 °C, about 40 °C, about 45 °C, about 50 °C, about 55 °C, about 60 °C, about 65 °C, about 70 °C, about 75 °C, about 80 °C, about 85 °C, about 90 °C, about 95 °C, about 100 °C, about 105 °C, about 110 °C, about 115 °C, about 120 °C, about 125 °C, about 130 °C, about 135 °C, about 140 °C, about 145 °C, about 150 °C, or about 151 °C, including all values and ranges therebetween.

In some embodiments of the present disclosure, the optimal temperature *T* ranges from about 40 °C to about 151 °C, including all values and ranges therebetween, and is less than the boiling point of the electrolyte at a given pressure *P*.

In some embodiments of the present disclosure, the optimal temperature *T* ranges from about 50 °C to about 151 °C, including all values and ranges therebetween, and is less than the boiling point of the electrolyte at a given pressure *P*.

In some embodiments of the present disclosure, the optimal temperature *T* ranges from about 40 °C to about 151 °C, including all values and ranges therebetween, and is less than the boiling point of the electrolyte at a given pressure *P*, and does not include temperatures below 40 °C.

In some embodiments of the present disclosure, the optimal temperature *T* ranges from about 50 °C to about 151 °C, including all values and ranges therebetween, and is less than the boiling point of the electrolyte at a given pressure *P*, and does not include temperatures below 50 °C.

In some embodiments of the present disclosure, the optimized temperature employed for generating oxygen by electrochemical water splitting is less than the boiling point of the electrolyte at a given pressure *P*. As is known to a person skilled in the art, the boiling point of electrolyte will also vary with change in pressure. Accordingly, the optimized temperature employed in the method of the present disclosure also changes depending on the pressure.

In some embodiments of the present disclosure, the electrolyte comprises water and alkali, and therefore the optimized temperature employed for generating oxygen by electrochemical water splitting is less than the boiling point of the said electrolyte at a given pressure *P*.

In some embodiments of the present disclosure, the optimal pressure *P* ranges from about 1 bar to about 210 bar, including all values and ranges therebetween.

In some embodiments of the present disclosure, the optimal pressure *P* is about 1 bar, about 10 bar, about 20 bar, about 30 bar, about 40 bar, about 50 bar, about 60 bar, about 70 bar, about 80 bar, about 90 bar, about 100 bar, about 110 bar, about 120 bar, about 130 bar, about 140 bar, about 150 bar, about 160 bar, about 170 bar, about 180 bar, about 190 bar, about 200 bar, or about 210 bar, including all values and ranges therebetween.

In some embodiments of the present disclosure, the optimal potential ranges from about 1.45 V to about 1.75 V versus the reversible hydrogen electrode (RHE) at the anode, including all values and ranges therebetween.

In some embodiments of the present disclosure, the optimal potential applied at the anode is about 1.45 V vs. the RHE, about 1.46 V vs. the RHE, about 1.47 V vs. the RHE, about 1.48 V vs. the RHE, about 1.49 V vs. the RHE, about 1.50 V vs. the RHE, about 1.51 V vs. the RHE, about 1.52 V vs. the RHE, about 1.53 V vs. the RHE, about 1.54 V vs. the RHE, about 1.55 V vs. the RHE, about 1.56 V vs. the RHE, about 1.57 V vs. the RHE, about 1.58 V vs. the RHE, about 1.59 V vs. the RHE, about 1.60 V vs. the RHE, about 1.61 V vs. the RHE, about 1.62 V vs. the RHE, about 1.63 V vs. the RHE, about 1.64 V vs. the RHE, about 1.65 V vs. the RHE, about 1.66 V vs. the RHE, about 1.67 V vs. the RHE, about 1.68 V vs. the RHE, about 1.69 V vs. the RHE, about 1.70 V vs. the RHE, about 1.71 V vs. the RHE, about 1.72 V vs. the RHE, about 1.73 V vs. the RHE, about 1.74 V vs. the RHE, or about 1.75 V vs. the RHE, including all values and ranges therebetween.

In some embodiments of the present disclosure, the potential applied at the anode is about 1.5 V versus the RHE and preferably 1.51 V at the RHE.

In some embodiments of the present disclosure, the potential applied at the anode is about 1.6 V versus the RHE.

In some embodiments of the present disclosure, the potential applied at the anode is about 1.7 V versus the RHE.

In some embodiments of the present disclosure, the OER current density ranges from about 10 mA/cm^2 to about 10,000 mA/cm^2, including all values and ranges therebetween.

In some embodiments of the present disclosure, the OER current density ranges from about 400 mA/cm^2 to about 1000 mA/cm^2, including all values and ranges therebetween.

In some embodiments of the present disclosure, the OER current density ranges from about 60 mA/cm^2 to about 1600 mA/cm^2, including all values and ranges therebetween.

In some embodiments of the present disclosure, the OER current density ranges from about 60 mA/cm^2 to about 2500 mA/cm^2, including all values and ranges therebetween.

In some embodiments of the present disclosure, the OER current density is about 10 mA/cm^2, about 20 mA/cm^2, about 30 mA/cm^2, about 40 mA/cm^2, about 50 mA/cm^2, about 100 mA/cm^2, about 200 mA/cm^2, about 300 mA/cm^2, about 400 mA/cm^2, about 500 mA/cm^2, about 600 mA/cm^2, about 700 mA/cm^2, about 800 mA/cm^2, about 900 mA/cm^2, about 1000 mA/cm^2, about 2000 mA/cm^2, about 3000 mA/cm^2, about 4000 mA/cm^2, about 5000 mA/cm^2, about 6000 mA/cm^2, about 7000 mA/cm^2, about 8000 mA/cm^2, about 9000 mA/cm^2, or about 10000 mA/cm^2 including all values and ranges therebetween.

In some embodiments of the present disclosure, the OER current density is about 10 mA/cm^2.

In some embodiments of the present disclosure, the OER current density is about 400 mA/cm^2.

In some embodiments of the present disclosure, the OER current density is about 997 mA/cm^2.

In some embodiments of the present disclosure, the OER current density is about 770 mA/cm^2.

In some embodiments of the present disclosure, the OER current density is about 1241 mA/cm^2.

In some embodiments of the present disclosure, the OER current density is about 1960 mA/cm^2.

In some embodiments of the present disclosure, the OER current density is improved by at least twofold when compared to any method performed outside the stated potential ranging from about 1.45 V to about 1.75 V versus the reversible hydrogen electrode (RHE) at the anode, stated pH range of about 7.4 to about 10.9 and the stated temperature range from about 28 °C to about 151 °C at a given pressure P, wherein the temperature is less than the boiling point of the electrolyte at the given pressure.

In some embodiments of the present disclosure, the OER current density is improved by up to 1000-fold when compared to a method performed outside the stated potential ranging from about 1.45 V to about 1.75 V versus the reversible hydrogen electrode (RHE) at the anode, stated pH range of about 7.4 to about 10.9 and the stated temperature range from about 28 °C to about 151 °C at a given pressure P, wherein the temperature is less than the boiling point of the electrolyte at the given pressure.

In some embodiments of the present disclosure, the OER current density is improved in the range of about twofold to 1000-fold when compared to a method performed outside the stated potential ranging from about 1.45 V to about 1.75 V versus the reversible hydrogen electrode (RHE) at the anode, stated pH range of about 7.4 to about 10.9 and the stated temperature range from about 28 °C to about 151 °C at a given pressure P, wherein the temperature is less than the boiling point of the electrolyte at the given pressure.

In some embodiments of the present disclosure, the OER current density is improved by about twofold, about fivefold, about 10-fold, about 50-fold, about 100-fold, about 200-fold, about 300-fold, about 400-fold, about 500-fold, about 600-fold, about 700-fold, about 800-fold, about 900-fold, or about 1000-fold when compared to any method performed outside the stated potential ranging from about 1.45 V to about 1.75 V versus the reversible hydrogen electrode (RHE) at the anode, stated pH range of about 7.4 to about 10.9 and the stated temperature range from about 28 °C to about 151 °C at a given pressure P, wherein the temperature is less than the boiling point of the electrolyte at the given pressure.

In some embodiments of the present disclosure, the OER current density improves by at least about 80% when the method is carried out at said optimized conditions of pH, temperature, potential, and pressure as described herein. This improvement is in comparison with a similar

method performed under the same conditions of pressure and potential, but at a temperature of 20 °C.

In some embodiments of the present disclosure, the OER current density improves by about 200% when the method is carried out at said optimized conditions of pH, temperature, potential, and pressure as described herein. This improvement is in comparison with a similar method performed under the same conditions of pressure and potential, but at a temperature of 20 °C.

In some embodiments of the present disclosure, the OER current density improves by up to about 21700% when the method is carried out at a pressure of about 1 bar, potential of 1.5 V vs. the RHE at the anode and at optimized temperature and pH conditions.

In some embodiments of the present disclosure, the OER current density improves by up to about 5000% when the method is carried out at a pressure of about 1 bar, potential of 1.6 V vs. the RHE at the anode and optimized temperature and pH conditions.

In some embodiments of the present disclosure, the OER current density improves by up to about 5000% when the method is carried out at a pressure of about 10 bar, potential of 1.6 V vs. the RHE at the anode and optimized temperature and pH conditions.

In some embodiments of the present disclosure, the OER current density improves by up to about 5000% when the method is carried out at a pressure of about 200 bar, potential of 1.6 V vs. the RHE at the anode and optimized temperature and pH conditions.

In some embodiments of the present disclosure, the OER current density improves by up to about 200% when the method is carried out at a pressure of about 1 bar, potential of 1.7 V vs. The RHE at the anode and optimized temperature and pH conditions.

In some embodiments of the present disclosure, the OER current density improves by up to about 200% when the method is carried out at a pressure of about 10 bar, potential of 1.7 V vs. the RHE at the anode and optimized temperature and pH conditions.

In some embodiments of the present disclosure, the OER current density improves by up to about 200% when the method is carried out at a pressure of about 100 bar, potential of 1.6 V vs. the RHE at the anode and optimized temperature and pH conditions.

Accordingly, in some embodiments of the present disclosure, the potential is about 1.5 V vs. the RHE, the pH of the electrolyte ranges from about 7.4 to about 10.7, and the temperature *T* ranges from about 28 °C to about 151 °C and is less than the boiling point of the electrolyte at the given pressure *P*.

In some embodiments of the present disclosure, the potential is about 1.5 V vs. the RHE, the pH of the electrolyte ranges from about 7.4 to about 10.6, and the temperature *T* ranges from about 40 °C to about 151 °C and is less than the boiling point of the electrolyte at the given pressure *P*.

In some embodiments of the present disclosure, the potential is about 1.5 V vs. the RHE, the pH of the electrolyte ranges from about 7.4 to about 10.3, and the temperature *T* ranges from about 50 °C to about 151 °C and is less than the boiling point of the electrolyte at the given pressure *P*.

In some embodiments of the present disclosure, the potential is about 1.6 V vs. the RHE, the pH of the electrolyte ranges from about 8.5 to about 10.9, and the temperature *T* ranges from about 28 °C to about 92 °C and is less than boiling point of the electrolyte at the given pressure *P*.

In some embodiments of the present disclosure, the potential is about 1.6 V vs. the RHE, the pH of the electrolyte ranges from about 8.5 to about 10.7, and the temperature *T* ranges from about 40 °C to about 92 °C and is less than boiling point of the electrolyte at the given pressure *P*.

In some embodiments of the present disclosure, the potential is about 1.6 V vs. the RHE, the pH of the electrolyte ranges from about 8.5 to about 10.3, and the temperature *T* ranges from about 50 °C to about 92 °C and is less than boiling point of the electrolyte at the given pressure *P*.

In some embodiments of the present disclosure, the potential is about 1.7 V vs. the RHE, the pH of the electrolyte ranges from about 10.2 to about 10.9, and the temperature *T* ranges from about 28 °C to about 41 °C and is less than boiling point of the electrolyte at the given pressure *P*.

In some embodiments of the present disclosure, the potential is about 1.7 V vs. the RHE, the pH of the electrolyte ranges from about 10.2 to about 10.6, and the temperature *T* ranges from about 40 °C to about 41 °C and is less than boiling point of the electrolyte at the given pressure *P*.

Accordingly, in some embodiments of the present disclosure, the pressure is about 1 bar, the potential is about 1.5 V vs. the RHE at the anode, and the pH of the electrolyte ranges from about 8.6 to about 10.7, including all values and ranges therebetween, for temperatures ranging from about 28 °C to about 90 °C. Consequently, the OER current density at these conditions ranges from about 10 mA/cm^2 to about 770 mA/cm^2.

In some embodiments of the present disclosure, the pressure is about 10 bar, the potential is about 1.5 V vs. the RHE at the anode, and the pH of the electrolyte ranges from about 7.4 to about 10.7, including all values and ranges therebetween, for temperatures ranging from about 32 °C to about 151 °C. Consequently, the OER current density at these conditions ranges from about 10 mA/cm^2 to about 10000 mA/cm^2.

In some embodiments of the present disclosure, the pressure is about 10 bar, the potential is about 1.5 V vs. the RHE at the anode, and the pH of the electrolyte ranges from about 7.4 to about 10.4, including all values and ranges therebetween, for temperatures ranging from about 40 °C to about 151 °C. Consequently, the OER current density at these conditions ranges from about 25 mA/cm^2 to about 9800 mA/cm^2.

In some embodiments of the present disclosure, the pressure is about 20 bar, the potential is about 1.5 V vs. the RHE at the anode, and the pH of the electrolyte ranges from about 7.5 to about 10.0, including all values and ranges therebetween, for temperatures ranging from about 50 °C to about 151 °C. Consequently, the OER current density at these conditions ranges from about 60 mA/cm^2 to about 9800 mA/cm^2.

In some embodiments of the present disclosure, the pressure is about 50 bar, the potential is about 1.5 V vs. the RHE at the anode, and the pH of the electrolyte ranges from about 7.7 to about 9.4, including all values and ranges therebetween, for temperatures ranging from about 70 °C to about 140 °C. Consequently, the OER current density at these conditions ranges from about 250 mA/cm^2 to about 6800 mA/cm^2.

In some embodiments of the present disclosure, the pressure is about 100 bar, the potential is about 1.5 V vs. the RHE at the anode, and the pH of the electrolyte ranges from about 7.6 to about 9.2, including all values and ranges therebetween, for temperatures ranging from about 80 °C to about 150 °C. Consequently, the OER current density at these conditions ranges from about 435 mA/cm^2 to about 9800 mA/cm^2.

In some embodiments of the present disclosure, the pressure is about 150 bar, the potential is about 1.5 V vs. the RHE at the anode, and the pH of the electrolyte ranges from about 7.6 to about 9.0, including all values and ranges therebetween, for temperatures ranging from about 90 °C to about 150 °C. Consequently, the OER current density at these conditions ranges from about 740 mA/cm^2 to about 9800 mA/cm^2.

In some embodiments of the present disclosure, the pressure ranges from about 1 bar to 210 bar, the potential is about 1.5 V vs. the RHE at the anode, and the pH of the electrolyte ranges from about 7.4 to about 10.9, including all values and ranges therebetween, for temperatures ranging from about 28 °C to about 151 °C. Consequently, the OER current density at these conditions ranges from about 10 mA/cm^2 to about 10000 mA/cm^2

In some embodiments of the present disclosure, the pressure is about 1 bar, the potential is about 1.6 V vs. the RHE at the anode, the pH of the electrolyte ranges from about 8.6 to about 10.6, including all values and ranges therebetween, for temperatures ranging from about 30 °C to about 90 °C. Consequently, the OER current density at these conditions ranges from about 370 mA/cm^2 to about 9300 mA/cm^2.

In some embodiments of the present disclosure, the pressure is about 10 bar, the potential is about 1.6 V vs. the RHE at the anode, the pH of the electrolyte ranges from about 8.6 to about 10.3, including all values and ranges therebetween, for temperatures ranging from about 40 °C

to about 90 °C. Consequently, the OER current density at these conditions ranges from about 700 mA/cm^2 to about 9300 mA/cm^2.

In some embodiments of the present disclosure, the pressure is about 20 bar, the potential is about 1.6 V vs. the RHE at the anode, the pH of the electrolyte ranges from about 8.7 to about 10, including all values and ranges therebetween, for temperatures ranging from about 50 °C to about 90 °C. Consequently, the OER current density at these conditions ranges from about 1200 mA/cm^2 to about 9300 mA/cm^2.

In some embodiments of the present disclosure, the pressure is about 50 bar, the potential is about 1.6 V vs. the RHE at the anode, the pH of the electrolyte ranges from about 8.8 to about 9.8, including all values and ranges therebetween, for temperatures ranging from about 60 °C to about 90 °C. Consequently, the OER current density at these conditions ranges from about 2195 mA/cm^2 to about 9220 mA/cm^2.

In some embodiments of the present disclosure, the pressure is about 150 bar, the potential is about 1.6 V vs. the RHE at the anode, the pH of the electrolyte ranges from about 8.9 to about 9.6, including all values and ranges therebetween, for temperatures ranging from about 70 °C to about 90 °C. Consequently, the OER current density at these conditions ranges from about 3600 mA/cm^2 to about 9200 mA/cm^2.

In some embodiments of the present disclosure, the pressure is about 200 bar, the potential is about 1.6 V vs. the RHE at the anode, the pH of the electrolyte ranges from about 9 to about 10, including all values and ranges therebetween, for temperatures ranging from about 60 °C to about 90 °C. Consequently, the OER current density at these conditions ranges from about 2180 mA/cm^2 to about 9180 mA/cm^2.

In some embodiments of the present disclosure, the pressure ranges from about 1 bar to 210 bar, the potential is about 1.6 V vs. the RHE at the anode, and the pH of the electrolyte ranges from about 8.5 to about 10.9, including all values and ranges therebetween, for temperatures ranging from about 28 °C to about 92 °C. Consequently, the OER current density at these conditions ranges from about 300 mA/cm^2 to about 10000 mA/cm^2.

In some embodiments of the present disclosure, the pressure is about 1 bar, the potential is about 1.7 V vs. the RHE at the anode, the pH of the electrolyte ranges from about 10.2 to about 10.9, including all values and ranges therebetween, for temperatures ranging from about 30 °C to about 40 °C. Consequently, the OER current density at these conditions ranges from about 5700 mA/cm^2 to about 9900 mA/cm^2.

In some embodiments of the present disclosure, the pressure ranges from about 1 bar to 210 bar, the potential is about 1.7 V vs. the RHE at the anode, and the pH of the electrolyte ranges from about 10.2 to about 10.9, including all values and ranges therebetween, for temperatures ranging from about 28 °C to about 41 °C. Consequently, the OER current density at these conditions ranges from about 5500 mA/cm^2 to about 10000 mA/cm^2.

As is apparent from the embodiments above, the criticality of the present disclosure lies in the optimized conditions of pH, temperature, and pressure for a potential range that have been arrived at herein, for generating oxygen by electrochemical water splitting. A person skilled in the art will therefore readily realize that these conditions are unaffected by the type or make of the device or system that employs these conditions for electrochemical water splitting. Hence, any device or system such as an electrolyzer will be able to electrochemically split water equally well and with higher efficiency of the OER, as long as the electrolyzer functions at the optimized conditions of pH, temperature, and pressure for the potential range as described herein.

As mentioned above, since pH is one of the important conditions that has been optimized in the present disclosure, a direct impact of the stated optimization is visible on the concentration or amount of alkali employed in the method for maintaining the stated optimal pH.

Accordingly, in some embodiments of the present disclosure, for maintaining the pH range of about 7.4 to about 10.9 of the present disclosure, the concentrations or amounts of the required alkali in the electrolyte range from about 0.07 mM to about 1.38 mM, including all values and ranges therebetween.

In some embodiments of the present disclosure, the concentration of the alkali in the electrolyte is about 0.07 mM, about 0.08 mM, about 0.09 mM, about 0.1 mM, about 0.2 mM, about 0.3

mM, about 0.4 mM, about 0.5 mM, about 0.6 mM, about 0.7 mM, about 0.8 mM, about 0.9 mM, about 1 mM, about 1.1 mM, about 1.2 mM, about 1.3 mM, about 1.35 mM, or about 1.38 mM, including all values and ranges therebetween.

In some embodiments of the present disclosure, the pH reduces from about 10.7 to about 7.4 with an increase in temperature and the corresponding increase in the OER current density at the potential of about 1.5 V vs. the RHE; and wherein the concentration or amount of the alkali required for maintaining the stated pH reduces from about 1.38 mM to about 0.07 mM at the given pressure *P*.

In some embodiments of the present disclosure, the pH reduces from about 10.9 to about 8.5 with an increase in temperature and the corresponding increase in the OER current density at the potential of about 1.6 V vs. the RHE; and wherein the concentration or amount of the alkali required for maintaining the stated pH reduces from about 1.37 mM to about 0.14 mM at the given pressure *P*.

In some embodiments of the present disclosure, the pH reduces from about 10.9 to about 10.2 with an increase in temperature and the corresponding increase in the OER current density at the potential of about 1.7 V vs. the RHE; and wherein the concentration or amount of the alkali required for maintaining the stated pH reduces from about 1.37 mM to about 0.47 mM at the given pressure *P*.

In some embodiments of the present disclosure, the alkali is selected from a group comprising potassium hydroxide (KOH), sodium hydroxide (NaOH), calcium hydroxide ($Ca(OH)_2$), magnesium hydroxide ($Mg(OH)_2$), and any other alkali or alkaline earth hydroxide, or any combination thereof.

Accordingly, in some embodiments of the present disclosure, for maintaining the stated pH range of about 7.4 to about 10.9 of the present disclosure, the amount/concentration of the required alkali in the electrolyte ranges from about 0.07 mM to about 1.38 mM, including all values and ranges therebetween, and wherein the alkali is potassium hydroxide (KOH).

In some embodiments of the present disclosure, for maintaining the stated pH range of about 7.4 to about 10.9 of the present disclosure, the amount/concentration of the required alkali in

the electrolyte ranges from about 0.07 mM to about 1.38 mM, including all values and ranges therebetween, and wherein the alkali is sodium hydroxide (NaOH).

In some embodiments of the present disclosure, for maintaining the stated pH range of about 7.4 to about 10.9 of the present disclosure, the amount/concentration of the required alkali in the electrolyte ranges from about 0.07 mM to about 1.38 mM, including all values and ranges therebetween, and wherein the alkali is calcium hydroxide ($Ca(OH)_2$).

In some embodiments of the present disclosure, for maintaining the stated pH range of about 7.4 to about 10.9 of the present disclosure, the amount/concentration of the required alkali in the electrolyte ranges from about 0.07 mM to about 1.38 mM, including all values and ranges therebetween, and wherein the alkali is magnesium hydroxide ($Mg(OH)_2$).

In some embodiments of the present disclosure, for maintaining the stated pH range of about 7.4 to about 10.9 of the present disclosure, the amount/concentration of the required alkali in the electrolyte ranges from about 0.07 mM to about 1.38 mM, including all values and ranges therebetween, and wherein the alkali is any alkali or alkaline earth hydroxide.

As mentioned above, in the present disclosure, the anode of the electrolyzer is contacted with an electrolyte comprising water and alkali, in the presence of a metal-doped oxide, hydroxide, or oxyhydroxide catalyst. In some embodiments of the present disclosure, the stated metal-doped oxyhydroxide catalyst is an iron-doped nickel oxide, hydroxide, or oxyhydroxide catalyst, also referred to as a mixed iron/nickel oxide, hydroxide or oxyhydroxide material.

In some embodiments of the present disclosure, the electrolyzer further comprises a cathode within the electrolyte.

In some embodiments of the present disclosure, application of the potential across the electrolyzer generates hydrogen at the cathode.

In some embodiments of the present disclosure, the electrolyzer further comprises a cathode within the electrolyte, and wherein application of the potential across the electrolyzer generates oxygen at anode and hydrogen at cathode.

In some embodiments of the present disclosure, the generated oxygen and hydrogen are further captured.

In some embodiments of the present disclosure, the generated oxygen is further captured in compressed cylinders or tanks.

In some embodiments of the present disclosure, the generated hydrogen is further captured in compressed cylinders or tanks

Accordingly, the present disclosure provides a method of generating oxygen by electrochemical water splitting at optimized conditions of pH, temperature, and pressure for an oxygen evolution reaction (OER) current density ranging from about 10 mA/cm^2 to about 10000 mA/cm^2, said method comprising:

a) contacting an anode of an electrolyzer with an electrolyte comprising water and alkali, in presence of a iron-doped nickel oxide, hydroxide, or oxyhydroxide catalyst, wherein the alkali is selected from potassium hydroxide (KOH), sodium hydroxide (NaOH), calcium hydroxide ($Ca(OH)_2$), magnesium hydroxide ($Mg(OH)_2$), another alkali or alkaline earth hydroxide or any combination thereof; and

b) applying a potential ranging from about 1.45 V to about 1.75 V versus the reversible hydrogen electrode (RHE) at the anode to allow said generation of the oxygen;
wherein the pH of the electrolyte ranges from about 7.4 to about 10.9;
wherein the pH is a function of temperature T at a given pressure P; and
wherein the temperature T ranges from about 28 °C to about 151 °C and is less than the boiling point of the electrolyte at the given pressure P.

In some embodiments of the present disclosure, the method comprises

a) contacting an anode of an electrolyzer with an electrolyte comprising water and 0.61 mM KOH, in the presence of a iron-doped nickel oxyhydroxide catalyst; and

b) application of a potential of about 1.5 V vs. the RHE at the anode to allow generation of the oxygen;
wherein the pH of the electrolyte is about 10.7;
wherein the temperature T is about 28 °C at a pressure P of 1 bar; and
wherein the OER current density is about 10 mA/cm^2.

In some embodiments of the present disclosure, the method comprises

a) contacting an anode of an electrolyzer with an electrolyte comprising water and 0.7 mM KOH, in the presence of a iron-doped nickel oxyhydroxide catalyst; and

b) application of a potential of about 1.5 V vs. the RHE at the anode to allow generation of the oxygen;

wherein the pH of the electrolyte is about 10.4;

wherein the temperature *T* is about 40 °C at a pressure *P* of 10 bar; and

wherein the OER current density is about 28 mA/cm^2.

In some embodiments of the present disclosure, the method comprises

a) contacting an anode of an electrolyzer with an electrolyte comprising water and 0.071 mM NaOH, in the presence of an iron-doped nickel oxyhydroxide catalyst; and

b) application of a potential of about 1.5 V vs. the RHE at the anode to allow generation of the oxygen;

wherein the pH of the electrolyte is about 7.5;

wherein the temperature *T* is about 150 °C at a pressure *P* of 10 bar; and

wherein the OER current density is about 10000 mA/cm^2.

In some embodiments of the present disclosure, the method comprises

a) contacting an anode of an electrolyzer with an electrolyte comprising water and 0.26 mM KOH, in the presence of a mixed iron/nickel hydroxide catalyst; and

b) application of a potential of about 1.5 V vs. the RHE at the anode to allow generation of the oxygen;

wherein the pH of the electrolyte is about 8.8;

wherein the temperature *T* is about 90 °C at a pressure *P* of 50 bar; and

wherein the OER current density is about 761 mA/cm^2.

In some embodiments of the present disclosure, the method comprises

a) contacting an anode of an electrolyzer with an electrolyte comprising water and 0.86 mM KOH, in the presence of a iron-doped nickel oxyhydroxide catalyst; and

b) application of a potential of about 1.5 V vs. the RHE at the anode to allow generation of the oxygen;
wherein the pH of the electrolyte is about 10.1;
wherein the temperature T is about 50 °C at a pressure P of 50 bar; and
wherein the OER current density is about 54 mA/cm^2.

In some embodiments of the present disclosure, the method comprises

a) contacting an anode of an electrolyzer with an electrolyte comprising water and 0.26 mM KOH or NaOH, in the presence of a mixed iron/nickel oxide catalyst; and
b) application of a potential of about 1.5 V vs. the RHE at the anode to allow generation of the oxygen;
wherein the pH of the electrolyte is about 8.6;
wherein the temperature T is about 100 °C at a pressure P of 100 bar; and
wherein the OER current density is about 1241 mA/cm^2.

In some embodiments of the present disclosure, the method comprises

a) contacting an anode of an electrolyzer with an electrolyte comprising water and 0.24 mM NaOH, in the presence of a iron-doped nickel oxyhydroxide catalyst;
b) application of a potential of about 1.5 V vs. the RHE at the anode to allow generation of the oxygen;
wherein the pH of the electrolyte is about 8.4;
wherein the temperature T is about 110 °C at a pressure P of 150 bar; and
wherein the OER current density is about 1966 mA/cm^2.

In some embodiments of the present disclosure, the method comprises

a) contacting an anode of an electrolyzer with an electrolyte comprising water and 0.22 mM KOH, in the presence of an iron-doped nickel oxyhydroxide catalyst; and
b) application of a potential of about 1.5 V vs. the RHE at the anode to allow generation of the oxygen;
wherein the pH of the electrolyte is about 8.2;
wherein the temperature T is about 120 °C at a pressure P of 200 bar; and
wherein the OER current density is about 3032 mA/cm^2.

In some embodiments of the present disclosure, the method comprises

a) contacting an anode of an electrolyzer with an electrolyte comprising water and 0.12 mM NaOH, in the presence of a mixed iron/nickel oxyhydroxide catalyst; and

b) application of a potential of about 1.5 V vs. the RHE at the anode to allow generation of the oxygen;

wherein the pH of the electrolyte is about 7.7;

wherein the temperature *T* is about 150 °C at a pressure *P* of 210 bar; and

wherein the OER current density is about 9691 mA/cm^2.

In some embodiments of the present disclosure, the method comprises

a) contacting an anode of an electrolyzer with an electrolyte comprising water and 0.14 mM $Ca(OH)_2$, in the presence of an iron-doped nickel oxyhydroxide catalyst; and

b) application of a potential of about 1.6 V vs. the RHE the electrolyzer to allow generation of the oxygen;

wherein the pH of the electrolyte is about 8.5;

wherein the temperature *T* is about 92 °C at a pressure *P* of 1 bar; and

wherein the OER current density is about 10000 mA/cm^2.

In some embodiments of the present disclosure, the method comprises

a) contacting an anode of an electrolyzer with an electrolyte comprising water and 0.32 mM KOH, in the presence of a iron-doped nickel oxyhydroxide catalyst; and

b) application of a potential of about 1.6 V vs. the RHE at the anode to allow generation of the oxygen;

wherein the pH of the electrolyte is about 8.9;

wherein the temperature *T* is about 92 °C at a pressure *P* of 100 bar; and

wherein the OER current density is about 10000 mA/cm^2.

In some embodiments of the present disclosure, the method comprises

a) contacting an anode of an electrolyzer with an electrolyte comprising water and 0.70 mM KOH, in the presence of a iron-doped nickel oxyhydroxide catalyst; and

b) application of a potential of about 1.6 V vs. the RHE at the anode to allow generation of the oxygen;

wherein the pH of the electrolyte is about 10.4;

wherein the temperature *T* is about 40 °C at a pressure *P* of 10 bar; and

wherein the OER current density is about 704 mA/cm^2.

In some embodiments of the present disclosure, the method comprises

a) contacting an anode of an electrolyzer with an electrolyte comprising water and 0.52 mM $Mg(OH)_2$, in the presence of a mixed iron/nickel oxyhydroxide catalyst; and

b) application of a potential of about 1.6 V vs. the RHE at the anode to allow generation of the oxygen;

wherein the pH of the electrolyte is about 10;

wherein the temperature *T* is about 50 °C at a pressure *P* of 10 bar; and

wherein the OER current density is about 1271 mA/cm^2.

In some embodiments of the present disclosure, the method comprises

a) contacting an anode of an electrolyzer with an electrolyte comprising water and 0.65 mM KOH, in the presence of a mixed iron/nickel hydroxide catalyst; and

b) application of a potential of about 1.6 V vs. the RHE at the anode to allow generation of the oxygen;

wherein the pH of the electrolyte is about 9.8;

wherein the temperature *T* is about 60 °C at a pressure *P* of 50 bar; and

wherein the OER current density is about 2196 mA/cm^2.

In some embodiments of the present disclosure, the method comprises

a) contacting an anode of an electrolyzer with an electrolyte comprising water and 0.63 mM NaOH, in the presence of a iron-doped nickel oxyhydroxide catalyst; and

b) application of a potential of about 1.6 V vs. the RHE at the anode to allow generation of the oxygen;

wherein the pH of the electrolyte is about 9.6;

wherein the temperature *T* is about 70 °C at a pressure *P* of 100 bar; and

wherein the OER current density is about 3653 mA/cm^2.

In some embodiments of the present disclosure, the method comprises

a) contacting an anode of an electrolyzer with an electrolyte comprising water and 0.56 mM $Ca(OH)_2$, in the presence of an iron-doped nickel oxyhydroxide catalyst; and

b) application of a potential of about 1.6 V vs. the RHE at the anode to allow generation of the oxygen;
wherein the pH of the electrolyte is about 9.3;
wherein the temperature *T* is about 80 °C at a pressure *P* of 150 bar; and
wherein the OER current density is about 5880 mA/cm^2.

In some embodiments of the present disclosure, the method comprises

a) contacting an anode of an electrolyzer with an electrolyte comprising water and 0.47 mM $Mg(OH)_2$, in the presence of a mixed iron/nickel oxyhydroxide catalyst; and
b) application of a potential of about 1.6 V vs. the RHE at the anode to allow generation of the oxygen;
wherein the pH of the electrolyte is about 9.02;
wherein the temperature *T* is about 90 °C at a pressure *P* of 200 bar; and
wherein the OER current density is about 9182 mA/cm^2.

In some embodiments of the present disclosure, the method comprises

a) contacting an anode of an electrolyzer with an electrolyte comprising water and 0.47 mM KOH, in the presence of a iron-doped nickel oxyhydroxide catalyst; and
b) application of a potential of about 1.7 V vs. the RHE at the anode to allow generation of the oxygen;
wherein the pH of the electrolyte is about 10.2;
wherein the temperature *T* is about 40 °C at a pressure *P* of 1 bar; and
wherein the OER current density is about 10000 mA/cm^2.

In some embodiments of the present disclosure, the method comprises

a) contacting an anode of an electrolyzer with an electrolyte comprising water and 0.85 mM NaOH, in the presence of an iron-doped nickel oxyhydroxide catalyst; and
b) application of a potential of about 1.7 V vs. the RHE at the anode to allow generation of the oxygen;
wherein the pH of the electrolyte is about 10.8;
wherein the temperature *T* is about 30 °C at a pressure *P* of 10 bar; and
wherein the OER current density is about 5706 mA/cm^2.

In some embodiments of the present disclosure, the method comprises

a) contacting an anode of an electrolyzer with an electrolyte comprising water and 1.05 mM KOH, in the presence of a iron-doped nickel oxyhydroxide catalyst; and

b) application of a potential of about 1.7 V vs. the RHE at the anode to allow generation of the oxygen;

wherein the pH of the electrolyte is about 10.5;

wherein the temperature *T* is about 40 °C at a pressure *P* of 50 bar; and

wherein the OER current density is about 9848 mA/cm^2.

In some embodiments of the present disclosure, the method comprises

a) contacting an anode of an electrolyzer with an electrolyte comprising water and 1.18 mM NaOH, in the presence of a iron-doped nickel oxyhydroxide catalyst; and

b) application of a potential of about 1.7 V vs. the RHE at the anode to allow generation of the oxygen;

wherein the pH of the electrolyte is about 10.9;

wherein the temperature *T* is about 30 °C at a pressure *P* of 100 bar; and

wherein the OER current density is about 5688 mA/cm^2.

In some embodiments of the present disclosure, the method comprises

a) contacting an anode of an electrolyzer with an electrolyte comprising water and 1.30 mM KOH, in the presence of an iron-doped nickel oxyhydroxide catalyst; and

b) application of a potential of about 1.7 V vs. the RHE at the anode to allow generation of the oxygen;

wherein the pH of the electrolyte is about 10.6;

wherein the temperature *T* is about 40 °C at a pressure *P* of 150 bar; and

wherein the OER current density is about 9818 mA/cm^2.

In some embodiments of the present disclosure, the method comprises

a) contacting an anode of an electrolyzer with an electrolyte comprising water and 1.27 mM $Ca(OH)_2$, in the presence of a mixed iron/nickel oxyhydroxide catalyst; and

b) application of a potential of about 1.7 V vs. the RHE at the anode to allow generation of the oxygen;

wherein the pH of the electrolyte is about 10.9;
wherein the temperature *T* is about 30 °C at a pressure *P* of 200 bar; and
wherein the OER current density is about 5669 mA/cm^2.

In some embodiments of the present disclosure, the method comprises

a) contacting an anode of an electrolyzer with an electrolyte comprising water and 1.38 mM NaOH, in the presence of a iron-doped nickel oxyhydroxide catalyst; and
b) application of a potential of about 1.7 V vs. the RHE at the anode to allow generation of the oxygen;

wherein the pH of the electrolyte is about 10.6;
wherein the temperature *T* is about 40 °C at a pressure *P* of 210 bar; and
wherein the OER current density is about 9801 mA/cm^2.

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In some embodiments of the present disclosure, the method comprises electrolyte comprising water and KOH or NaOH, the catalyst is of a mixed iron/nickel oxide, hydroxide or oxyhydroxide material, the potential applied at the anode is 1.51 V versus the RHE, the temperature is about 90 °C, the pressure is about 1 bar, and the pH of the electrolyte is maintained at 8.6.

In some embodiments of the present disclosure, the method comprises electrolyte comprising water and KOH or NaOH, the catalyst is of a mixed iron/nickel oxide, hydroxide, or oxyhydroxide material, the potential applied at the anode is 1.51 V versus the RHE, the temperature is about 90 °C, the pressure is about 1 bar, the pH of the electrolyte is maintained at 8.6 and the OER current density is about 997 mA/cm^2.

In some embodiments of the present disclosure, the method comprises electrolyte comprising water and KOH or NaOH, the catalyst is of a mixed iron/nickel oxide, hydroxide, or oxyhydroxide material, the potential applied at the anode is 1.51 V versus the RHE, the temperature is about 90 °C, the pressure is about 100 bar, and the pH of the electrolyte is maintained at 8.9.

In some embodiments of the present disclosure, the method comprises electrolyte comprising water and KOH or NaOH, the catalyst is of a mixed iron/nickel oxide, hydroxide, or oxyhydroxide material, the potential applied at the anode is 1.51 V versus the RHE, the

temperature is about 90 °C, the pressure is about 100 bar, the pH of the electrolyte is maintained at 8.9 and the OER current density is about 985 mA/cm^2.

In some embodiments of the present disclosure, the method comprises electrolyte comprising water and KOH or NaOH having a concentration of about 0.15 mM, the catalyst is of a mixed iron/nickel oxide, hydroxide, or oxyhydroxide material, the potential applied at the anode is 1.5 V versus the RHE, the temperature is about 90 °C, the pressure is about 1 bar, and the pH of the electrolyte is maintained at 8.6.

In some embodiments of the present disclosure, the method comprises electrolyte comprising water and KOH or NaOH having a concentration of about 0.15 mM, the catalyst is of a mixed iron/nickel oxide, hydroxide, or oxyhydroxide material, the potential applied at the anode is 1.5 V versus the RHE, the temperature is about 90 °C, the pressure is about 1 bar, the pH of the electrolyte is maintained at 8.6, and the OER current density is about 770 mA/cm^2.

In some embodiments of the present disclosure, the method comprises electrolyte comprising water and alkali (KOH or NaOH) having a concentration of about 0.26 mM, the catalyst is a of a mixed iron/nickel oxide, hydroxide, or oxyhydroxide material, the potential applied at the anode is 1.5 V versus the RHE, the temperature is about 100 °C, the pressure is about 100 bar, and the pH of the electrolyte is maintained at 8.6.

In some embodiments of the present disclosure, the method comprises electrolyte comprising water and alkali (KOH or NaOH) having a concentration of about 0.25 mM, the catalyst is a of a mixed iron/nickel oxide, hydroxide, or oxyhydroxide material, the potential applied at the anode is 1.5 V versus the RHE, the temperature is about 100 °C, the pressure is about 100 bar, the pH of the electrolyte is maintained at 8.6, and the OER current density is about 1241 mA/cm^2.

In some embodiments of the present disclosure, the method comprises electrolyte comprising water and KOH or NaOH having a concentration of about 0.27 mM, the catalyst is of a mixed iron/nickel oxide, hydroxide, or oxyhydroxide material, the potential applied at the anode is 1.5 V versus the RHE, the temperature is about 110 °C, the pressure is about 200 bar, and the pH of the electrolyte is maintained at 8.5.

In some embodiments of the present disclosure, the method comprises electrolyte comprising water and KOH or NaOH having a concentration of about 0.27 mM, the catalyst is of a mixed iron/nickel oxide, hydroxide, or oxyhydroxide material, the potential applied at the anode is 1.5 V versus the RHE, the temperature is about 110 °C, the pressure is about 200 bar, the pH of the electrolyte is maintained at 8.5 and the OER current density is about 1960 mA/cm^2.

The present disclosure also provides a device for generation of oxygen by electrochemical water oxidation. In particular, the device of the present disclosure for generating oxygen by electrochemical water oxidation, comprises

a) a reaction chamber comprising a plurality of walls defining an internal cavity;
b) an anode within the internal cavity comprising a metal-doped oxide, hydroxide, or oxyhydroxide catalyst; and
c) an electrolyte within the internal cavity, in contact with the anode, said electrolyte comprising water and alkali;

wherein the device is configured to carry out the electrochemical water oxidation under optimal temperature and pressure conditions as described above.

In some embodiments of the present disclosure, the device further comprises a cathode within the internal cavity, the cathode being in contact with the electrolyte and a membrane separating the anode from cathode.

In embodiments relating to the device for generation of oxygen, the features and characteristics of the optimal temperature, pressure, or other conditions, and metal-doped oxide, hydroxide, or oxyhydroxide catalyst is as described by any of the embodiments above. For the sake of brevity, and avoiding repetition, each of those embodiments are not being reiterated here again. However, each of the said embodiments, completely fall within the purview of the device for generation of oxygen described herein.

The present disclosure refers to a system for generating oxygen by electrochemical water oxidation. In particular, the system for generating oxygen by electrochemical water oxidation comprises the device as described above, and a power supply configured to receive power from a power source for application of a potential across the device.

In some embodiments of the present disclosure, the system further comprises at least one processor configured to control the temperature, pressure, applied potential, or a combination thereof.

In embodiments relating to the device for generation of oxygen, the features and characteristics of the device, optimal temperature, pressure, or other conditions, and metal-doped oxide, hydroxide, or oxyhydroxide catalyst is as described by any of the embodiments above. For the sake of brevity, and avoiding repetition, each of those embodiments are not being reiterated here again. However, each of the said embodiments, completely fall within the purview of the system for generation of oxygen described herein.

Nonetheless, having said the above, it is important to emphasize that regardless of the device and system described herein, the criticality of the present disclosure lies in the optimized conditions of pH, temperature, potential, and pressure that have been arrived at in the present disclosure, for generating oxygen by electrochemical water splitting. A person skilled in the art will therefore readily realize that these conditions are unaffected by the type or make of the device or system that employs these conditions for electrochemical water splitting. Hence, any device or system such as an electrolyzer will be able to electrochemically split water equally well and with higher efficiency and OER, as long as the electrolyzer functions at the optimized conditions of pH, temperature, and pressure for a range of potentials as described herein.

Overall, the method of the present disclosure possesses at least the following advantages:

a) Gives an overview of the coupled effect and relationship of temperature, pressure, and pH for a range of potentials on the thermodynamics and kinetics of the OER electrocatalyzed by metal-doped oxide, hydroxide, or oxyhydroxide catalyst to understand overall electrochemical water-splitting cycles;

b) Provides optimized operating conditions for electrolyzer that results in high OER current densities (up to 1000-fold higher) and can be used for industrial or large- scale production of pure oxygen;

c) Provides significant reduction in the concentration of alkali to be used in the electrolyte. Accordingly, the operation conditions would not be highly corrosive (at high temperatures) in an alkaline water electrolyzer and its operational life would also increase substantially, as well as providing more favourable scalability and economics;

d) Involves a negligible trade-off in kinetics, thereby leading to cost-savings due to an obviation of downstream hydrogen compression; and

e) Provides an efficient, eco-friendly, cost-effective and industrially scalable method for production of pure oxygen by electrochemical water splitting/oxidation in an electrolyzer.

It is to be understood that the foregoing description is illustrative not a limitation. While considerable emphasis has been placed herein on particular features of this disclosure, it will be appreciated that various modifications can be made, and that many changes can be made in the preferred embodiments without departing from the principles of the disclosure. Those skilled in the art will recognize that the embodiments herein can be practiced with modification within the spirit and scope of the embodiments as described herein. Similarly, additional embodiments and features of the present disclosure will be apparent to one of ordinary skill in art based upon description provided herein.

Descriptions of well-known/conventional methods/steps and techniques are omitted so as to not unnecessarily obscure the embodiments herein. Further, the disclosure herein provides for examples illustrating the above-described embodiments, and in order to illustrate the embodiments of the present disclosure, certain aspects have been employed. The examples used herein for such illustration are intended merely to facilitate an understanding of ways in which the embodiments may be practiced and to further enable those of skill in the art to practice the embodiments. Accordingly, following examples should not be construed as limiting the scope of the embodiments herein.

EXAMPLES

The analysis is based on the standard hydrogen electrode (SHE), whose potential at all temperatures is zero by convention, so that:

$$G^0_{(H^+ + e^-)}(T) = \frac{1}{2} G^0_{H_2(g)}(T) \quad (1)$$

where $G^0_{(H^+ + e^-)}(T)$ and $G^0_{H_2(g)}(T)$ are the standard Gibbs free energies of the proton-electron couple and gaseous hydrogen at temperature T. The superscript ^{0} denotes standard-state

conditions (1 bar pressure and pure water and oxygen). Hydrogen gas is at 1 bar pressure in the reference electrode at the considered temperature. The OER consists of four proton(hydroxide)-coupled electron transfer reactions, presented as follows:

$I_1 \rightleftarrows I_2 + (H^+ + e^-)$ or $I_1 + OH^- \rightleftarrows I_2 + H_2O + e^-$

$I_2 + H_2O \rightleftarrows I_3 + (H^+ + e^-) + O_2$ or $I_2 + OH^- \rightleftarrows I_3 + e^- + O_2$

$I_3 + H_2O \rightleftarrows I_4 + (H^+ + e^-)$ or $I_3 + OH^- \rightleftarrows I_4 + e^-$

$I_4 \rightleftarrows I_1 + (H^+ + e^-)$ or $I_4 + OH^- \rightleftarrows I_1 + H_2O + e^-$

where I_1 = HO*OH + $^{\gamma}$OH, I_2 = HO*O + $^{\gamma}$OH, I_3 = HO* + $^{\gamma}OH_2$, and I_4 = HO*OH_2 + $^{\gamma}$OH, * denotes the Fe active site, and $^{\gamma}$ a Ni atom in the surface layer. Since Fe/Ni are only four-fold-lattice-oxygen-coordinated in the considered surface facet of (Ni,Fe)OOH, two adsorbates can adsorb simultaneously onto the active site, as in I_1, I_2, and I_4.

Example 1: Effect of arbitrary temperature (below the water boiling point) on OER thermodynamics

The standard Gibbs free energies of molecular (H_2, O_2, and H_2O) and ionic ($H^+ + e^-$) species were first obtained at T_r = 25°C at any temperature T lower than the boiling point of water:

$$G_i^0(T) = \mathrm{E}_i^{DFT} + ZPE_i^{DFT} + \left(H_i^0(T) - H_i^0(0\ \mathrm{K}) - TS_i^0(T)\right) \quad (2)$$

where i denotes either H_2(g) or H_2O(g), E_i^{DFT} the species' density functional theory (DFT) total energy at zero Kelvin, ZPE_i^{DFT} its zero-point vibrational energy from DFT, and $H_i^0(T)$ and $S_i^0(T)$ its standard enthalpy and entropy at T, respectively. We determine $H_i^0(T)$ and $S_i^0(T)$ through Shomate equations from the National Institute of Standards and Technology (NIST) database:

$$H_i^0(T) - H_i^0(T_r) = A_i t + \frac{B_i t^2}{2} + \frac{C_i t^3}{3} + \frac{D_i t^4}{4} - \frac{E_i}{t} + F_i - H'_i \quad (3)$$

$$S_i^0(T) = A_i \ln t + B_i t + \frac{C_i t^2}{2} + \frac{D_i t^3}{3} - \frac{E_i}{2t^2} + G_i \quad (4)$$

where $t = T$ [K]$/1000$, and A_i through G_i and H'_i are experimentally derived constants for each molecular species i. $H_i^0(T_r)$ is directly obtained from the same database. We subsequently obtain the standard Gibbs free energy of liquid water at temperature T as:

$$G_{H_2O(l)}^0(T) = G_{H_2O(g)}^0(T) + \Delta G_{c,H_2O}^0(T) \quad (5)$$

where $\Delta G_{c,H_2O}^0(T)$ is the standard Gibbs free energy of condensation of water vapor. The subscript "c" denotes condensation; later, subscripts "r" and "f" will denote reaction and formation, respectively. We calculate $\Delta G_{c,H_2O}^0(T)$ at any temperature T as:

$$\Delta G_{c,H_2O}^0(T) = \Delta G_{c,H_2O}^0(T_r) + \left(\Delta G_{H_2O(l)}^0(T_r \to T) - \Delta G_{H_2O(g)}^0(T_r \to T)\right) \quad (6)$$

where $\Delta G_{c,H_2O}^0(T_r)$ is obtained from the NIST database and $\Delta G_i^0(T_r \to T)$ denotes the standard Gibbs free energy change incurred in changing the temperature of species i from T_r to T; the latter quantity for both liquid water and water vapor is estimated readily using Shomate equations. Finally, we calculate the standard Gibbs free energy of O_2(g) at room temperature as:

$$G_{O_2(g)}^0(T) = \Delta G_{r,H_2O\ split}^0(T) + \left(2G_{H_2O(l)}^0(T) - 2G_{H_2(g)}^0(T)\right) \quad (7)$$

where $\Delta G_{r,H_2O\ split}^0(T)$ denotes the standard Gibbs free energy of reaction for water splitting:

$$\Delta G_{r,H_2O\ split}^0(T) = -2\Delta G_{f,H_2O(l)}^0(T) \quad (8)$$

Here, $\Delta G^0_{f,H_2O(l)}(T)$ denotes the standard free energy of formation of liquid water. Note that a factor of two appears on the right-hand side because $\Delta G^0_{r,H_2O\ split}$ is defined per molecule of O_2 and $\Delta G^0_{f,H_2O(l)}$ is defined per molecule of H_2O; the minus sign appears because water decomposes in the former reaction and forms in the latter one. We estimate $\Delta G^0_{f,H_2O(l)}(T)$ in a manner similar to Eq. (6) above:

$$\Delta G^0_{f,H_2O(l)}(T) = \Delta G^0_{f,H_2O(l)}(T_r) + \left(G^0_{H_2O(l)}(T_r \to T) - G^0_{H_2(g)}(T_r \to T) - \frac{1}{2} G^0_{O_2(g)}(T_r \to T) \right) \quad (9)$$

where $\Delta G^0_{f,H_2O(l)}(T_r)$ is obtained from the NIST database and $\Delta G^0_i(T_r \to T)$ is calculated using the respective Shomate equation. Because the SHE potential at all temperatures and 1 bar pressure is defined to be zero, $\Delta G^0_{r,HER}(T) = 0$, such that the standard Gibbs free energy of the OER equals exactly the standard Gibbs free energy of water splitting, i.e., $\Delta G^0_{r,OER}(T) = \Delta G^0_{r,H_2O\ split}(T)$. At the outset, graphs depicting the free energies of liquid water and gaseous hydrogen and oxygen as a function of temperature from 10 °C to 90 °C were plotted (Figure 1B).

Next, the free energies of adsorbed species were determined as a function of temperature for solids as:

$$G^0_i(T) = \mathrm{E}^{DFT}_i + E_{vib,i} - TS_{vib,i} \quad (10)$$

where G^0_i is the standard Gibbs free energy of the i^{th} adsorbed species, E^{DFT}_i its quantum-mechanical DFT energy at one-quarter per metal site surface coverage $E_{vib,i}$ its vibrational internal energy, and $S_{vib,i}$ its vibrational entropy.

Based on the above equation, the standard Gibbs free energies of each adsorbed species were plotted as a function of temperature in Figure 1C. It was observed that these free energies vary by ~0.15 eV over the temperature range considered. Moreover, the changes in the free energies of the adsorbed species with temperature are comparable to that of the gaseous species.

Further, the standard Gibbs free energy change associated with the j^{th} step in the OER mechanism, $\Delta G^0_{r,j}(T)$, was calculated as:

$$\Delta G^0_{r,j}(T) = \sum_{i=1}^{N_{s,j}} \nu_{ij} G^0_i(T) \quad (11)$$

where $N_{s,j}$ denotes the number of species participating in reaction j and ν_{ij} represents the signed stoichiometric coefficient of species i in reaction j (>0 for products and <0 for reactants).

Next, the standard potentials of each of the four reactions in the OER mechanism as a function of temperature were plotted in Figure 1C. Figure 1C also depicts the standard reduction potential of the OER ($E^0_{OER,RHE}$) measured vs. the RHE as a function of temperature. Further, for both the SHE and RHE, the hydrogen pressure is set to 1 bar; therefore, for the latter, the sole potential variation arises due to its proton activity.

EXAMPLE 2: Effect of arbitrary temperature (below the water boiling point) on OER kinetics

In this example, OER kinetics as a function of temperature at 1 bar pressure was determined. The rate constant for each reaction j, k_j, is given as:

$$k_j(T,U) = \frac{k_B T}{h} \exp\left(-\frac{\lambda_j}{4k_B T}\left(1 + \frac{eE^0_{red,j,RHE}(T,U) + \Delta w_j}{\lambda_j}\right)^2\right) \quad (12)$$

where λ_j and Δw_j are Marcus parameters denoting the reorganization free energy and the *net* (product-reactant) work term, respectively, incurred in reaction j. Further, $E^0_{red,j,RHE}(T,U)$ is the standard reduction potential of the j^{th} step at temperature T and applied potential U, and is obtained as: $E^0_{red,j,RHE}(T,U) = E^0_{red,j,RHE}(T, 0\text{ V}) - eU$. Here, $E^0_{red,j,RHE}(T, 0\text{ V})$ is obtained from the Computational Hydrogen Electrode framework (see Figure 1D for values) and U is referenced to the RHE through the solution pH. Note that $E^0_{red,j,RHE}(T, 0\text{ V})$ is referenced to the RHE through the standard pH (pH^0=0 for protons and pH^0=14 for hydroxide ions). The rate constant, k_j, for each reaction j is converted to a reaction rate, r_j, via multiplying by the reactant

activities, a_i, raised to the absolute value of their respective stoichiometric coefficients, $|\nu_{ij}|$ as $r_j = k_j \prod_{i=1}^{n_r} a_i^{|\nu_{ij}|}$.

The activation free energies of forward ($\Delta G_{a,fj}^{\ddagger}$) and backward ($\Delta G_{a,bj}^{\ddagger}$) reactions are simply related via the free energy of reaction as: $\Delta G_{a,fi}^{\ddagger}(U) - \Delta G_{a,bi}^{\ddagger}(U) = \Delta G_{bulk,i}^{0}(U) + \Delta w_i$, where $\Delta G_{bulk,i}^{0}$ corresponds to the reaction free energy change assuming bulk liquid water, solvated ions, and O_2 gas phases. Δw_i converts the *bulk* reaction free energy to the *surface* reaction free energy by including the work done to transport reactants/products to the catalyst surface, as required for application of Marcus theory. We used the appropriate standard states for H_2O, H^+, OH^-, H_2 and O_2. The species activities (a_i) are defined as follows:

$$a_{H_2O} = \frac{C_{H_2O}^{0} - a_{H^+}}{\left(C_{H_2O}^{0} - a_{H^+}\right) + a_{H^+} + a_{OH^-} + C_{O_2(aq)} + a_{KOH} + a_{K}^{+}} \tag{13}$$

$$a_{O_2} = \frac{C_{O_2(aq)}}{C_{O_2}^{0}} \tag{14}$$

$$a_{H^+} = a_{H_3O^+} = \frac{10^{-pH}\ \mathrm{M}}{1\ \mathrm{M}} \tag{15}$$

$$a_{OH^-} = \frac{K_W a_{H_2O}}{a_{H^+}} \tag{16}$$

$$a_{KOH} = \frac{C_{KOH}}{1\ \mathrm{M}} \tag{17}$$

$$a_{K^+} = \frac{C_{K^+}}{1\ \mathrm{M}} \tag{18}$$

$$a_{I_l} = \theta_l = \frac{C_{I_l}}{C_{sites}} \tag{19}$$

where $C_{H_2O}^{0}$ is the standard-state molarity of pure water, calculated using its molar mass and its density at temperature T. $C_{O_2(aq)}$ is the solubility (mol/L) of O_2 in water at temperature T while $C_{O_2}^{0}$ is the standard solubility (at 1 bar pressure) of O_2 in water at temperature T. K_W is the equilibrium constant for water autoionization, which depends primarily on the temperature of the system. θ_l is the fraction of adsorbed species I_l, and C_{I_l} is the concentration of adsorbed species I_l. C_{sites} is equal to 2.27×10^{12} cm^{-2}, as mentioned above. The effective concentration of water is $(C_{H_2O}^{0} - a_{H^+})$ accounting for the degree of autoionization ($H_2O = H^+ + OH^-$).

To maintain the process at steady state, electrolyte at the appropriate pH must be fed continually into the reactor to make up for water losses due to water splitting and evaporation. Evaporated water can be recovered and reintroduced into the system by condensing out water vapor from the anodic product gas stream. Further, we consider the effects of these phenomena on the vapor-phase composition, i.e., vapor-phase mole fraction of oxygen and water vapor, which does affect the water-splitting rate via the concentration of dissolved oxygen.

The oxygen solubility in water, $C_{O_2(aq)}$, is a function of both temperature and pressure, and is calculated as:

$$C_{O_2(aq)}(T,p) = p y_{O_2(g)} k_{O_2}(T) \tag{20}$$

where p denotes the total pressure of the system, $y_{O_2(g)}$ the vapor phase mole fraction of O_2(g), and $k_{O_2}(T)$ the inverse Henry's law constant for O_2(g). $k_{O_2}(T)$, in units of mol O_2/kg water, is obtained from:

$$k_{O_2}(T) = \exp\left(\frac{0.046T^2 + 203.35T\ln\left(\frac{T}{298}\right) - (299.378 + 0.092T)(T - 298) - 20591}{8.3144T}\right) \tag{21}$$

Here $y_{O_2(g)}$ is determined based on the water vapor saturation pressure and oxygen partial pressure under the relevant reactor conditions. The vapor phase mole fraction of O_2(g) can be calculated as:

$$y_{O_2(g)} = \frac{p_{O_2(g)}}{p} \tag{22}$$

where $p_{O_2(g)}$ is the partial pressure of O_2(g) and is given in terms of the total system pressure and the water saturation pressure, $p_{H_2O(g)}$, as: $p_{O_2(g)} = p - p_{H_2O(g)}$. Further, $p_{H_2O(g)}$ can be calculated using Raoult's law as:

$$p_{H_2O(g)} = p^{sat}_{H_2O(g)} a_{H_2O} = p(1 - y_{O_2}) \tag{23}$$

where $p^{sat}_{H_2O(g)}$ is the saturation pressure of water vapor, which depends on temperature and can be calculated using Antoine's relation:[69]

$$\log_{10} p^{sat}_{H_2O(g)} = A_p - \frac{B_p}{T + C_p} \tag{24}$$

Here, $p^{sat}_{H_2O(g)}$ is expressed in bar, T in K ($255.9 \leq T \leq 373$), and $A_p = 4.6543$, $B_p = 1435.264$, and $C_p = -64.848$, as specified in the NIST database.[70]

Given the system temperature, standard pressure, and the electrolyte pH, $C^0_{H_2O}$, a_{H^+}, $p^{sat}_{H_2O(g)}$, k_{O_2}, and K_W can be determined. To obtain the remaining six species activities, i.e., a_{H_2O}, a_{O_2} (O_2 dissolved in water), y_{O_2} (O_2 in the vapor phase), a_{OH^-}, a_{K^+}, and a_{KOH}, Eqs. (13), (16), (20), and (23) have to be solved in conjunction with the following constraints:

$$a_{KOH} = 0 \tag{25}$$

$$a_{H^+} + a_{K^+} = a_{OH^-} \tag{26}$$

Eq. (25) represents the equilibrium condition for the dissociation of KOH into K^+ and OH^- ions, because KOH, being a strong base, dissociates completely. Eq. (26) enforces electroneutrality in the system. Implicit in the above system of equations is the assumption that water/KOH dissociation and water evaporation are much faster than the OER and can be taken to have reached equilibrium. It follows that all the ionic/gaseous activities are defined solely based on thermodynamics, and the effect of kinetics appears via the adsorbed intermediate activities. Moreover, water lost due to evaporation and oxidation is replenished, as would be in any continuous (flow) reactor system. Finally, the net (forward-backward) rates of the elementary steps are summed over and multiplied by the charge on an electron and the active site density, to obtain the OER current density.

EXAMPLE 3: Effect of pressure on vapor-phase mole fraction

Figure 2A depicts the vapor-phase oxygen mole fraction, y_{O_2}, as a function of pressure at two different temperatures, 25°C and 90°C, and a fixed pH of 12.0. It is observed that y_{O_2} is lower at higher temperature due to the increased evaporation of water. Further, y_{O_2} increases with increasing pressure because the saturation pressure of water can be reached easily, e.g., ~0.03 and 0.7 bar at 25°C and 90°C, respectively, and the head space is expected to be continuously filled with O_2 gas until the desired internal pressure is reached. Further, the aqueous oxygen activity, a_{O_2}, is depicted in Figure 2B at the same temperatures and under the same pressures. It was seen that the aqueous oxygen activity is higher at the lower temperature of 25°C due to the increased solubility of oxygen. Moreover, the aqueous oxygen activity increases with increasing pressure, again due to the higher oxygen solubility in water and higher mole fraction in the vapor phase (Figure 9).

EXAMPLE 4: The activities of other aqueous species as a function of temperature and pH

It is noted that K_W varies from 1.01×10^{-14} at 25°C to 3.83×10^{-13} at 90°C, as given in Figure 3A. The increase in K_W with temperature indicates an increased tendency for water to autoionize. The activities of H_2O, OH^-, and K^+ at 25°C and 90°C as a function of the pH is given in Figures 3B through 3D, respectively. As can be seen from Figures 3A through 3D, as the pH increases, the ionic activity increases, with a corresponding decrease in the water activity. Further, as temperature increases, water activity decreases due to increased dissociation of water. Finally, it is apparent that pH=14, which can be attained readily at room temperature, is physically unattainable at elevated temperatures because, e.g., a_{OH^-} and a_K^+ would need to be as high as ~21 at a temperature of 90°C for the pH to be 14.

Accordingly, moving forward, two pHs were considered (see Figure 4): pH=pH^0 (i.e., the pH where $a_{OH^-} = 1$ M) and pH=pH^{opt}, i.e., the optimal pH for maximizing the OER current density at an applied potential of 1.51 V vs. the RHE. At pH^0 and pH^{opt}, the aqueous species activities adopt reasonable values at all temperatures considered, thereby allowing comparisons between various temperatures. From Figure 4, it is apparent that the optimal pH for water splitting is less than 12 and that $pH^0 > pH^{opt}$ at all temperatures between 10°C and 90°C. Specifically, at 25°C, $pH^0 = 14.0$ and $pH^{opt} = 10.8$, while at 90°C, $pH^0 = 12.4$ and $pH^{opt} = 8.6$. Because the applied potentials (U) vs. the SHE and RHE are related by $U_{SHE} = U_{RHE} - \frac{2.303k_BT}{e}pH$, it follows that the absolute potential required (i.e., vs. the SHE) to obtain a fixed current density would be lower at pH^0 than at pH^{opt}. However, not only is the amount of base required lower at pH^{opt} but also the current density is slightly higher at the same potential.

EXAMPLE 5: Determining the OER current density as a function of the reactor temperature

The OER current density as a function of the reactor temperature is given in Figures 5A and 5B at an applied potential of 1.51 V vs. the RHE and the SHE, respectively. As mentioned above, the results are shown for two pHs: pH=pH^0(T) and pH=pH^{opt}(T). A potential of 1.51 V was chosen vs. the RHE because it leads to an OER current density of ~10 mA/cm^2 at room temperature, which is a benchmark in most experimental studies. A potential of 1.51 V was also considered vs. SHE as the reference electrode to present a contrasting example (however

the $pH^{opt}(T)$ is optimized for 1.51 V vs. RHE, and the two references are only equivalent at pH = 0).

As temperature rises, in both Figure 5A and 5B, the OER current densities increase, as expected. However, unlike in Figure 5B, the current densities at $pH=pH^0$ and $pH=pH^{opt}$ are *qualitatively* comparable at all temperatures in Figure 5A. This follows from the use of the RHE as a reference electrode. Nevertheless, *quantitatively* they differ by around 60 mA/cm^2 at 90°C for a fixed potential of 1.51 V vs. the RHE, indicating a difference of around 6% between the current densities at $pH=pH^0$ and $pH=pH^{opt}$. In contrast, in Figure 5B, one sees that the OER current density is much higher than seen in Figure 5A, and it is greater at pH^0, rather than at pH^{opt}. This is because $pH^0 > pH^{opt}$ *and* due to the use of SHE as a reference electrode (higher the pH, the lower the equilibrium potential for OER, when measured against the SHE). It was noted that the OER current density for a fixed *U* of 1.51 V vs. the RHE(*T*) increases to 595 mA/cm^2 at 80°C, indicating Fe-doped β-NiOOH is a suitable catalyst for commercial electrolyzers that operate at current densities > 400 mA/cm^2 at a temperature of ~80°C. Moreover, the OER current density for the same potential further increases to 997 mA/cm^2 at a temperature of 90 °C, allowing one to achieve a current density as high as ~1 A/cm^2.

EXAMPLE 6: Determination of concentration of alkali as a function of temperature and pH

The operation of the anodic water-splitting half-cell at $pH^{opt} < pH^0$ can enable the use of less alkali but with concomitant production of less oxygen. Figure 6 illustrates the activity of K+ ions as a function of temperature when the solution is maintained at the optimal pH, i.e., pH^{opt}. Because KOH dissociates fully, the activity of K^+ corresponds to the concentration of alkali added initially to the system, assuming a constant volume of the liquid phase. Thus, at pH^{opt}, the concentration of KOH to be introduced is ~0.7 mM at 25°C, which further decreases to ~0.15 mM at 90°C. However, this reduction in the amount of alkali required does not come without a cost. Indeed, operating at pH^{opt} will require a larger absolute potential, if one wants to maintain the same current density as obtained by operating at pH^0. Figure 7A depicts the potential versus the SHE that would be required to obtain a potential of 1.51 V versus the RHE at pH^{opt}. Figure 7B shows the extra potential that would be needed at pH^{opt} to obtain the same current density as at pH^0, which is ~0.28 V at 90°C. At 25°C, operating at pH^{opt}, compared to pH^0, leads to 3 orders of magnitude reduction in the alkali requirement (0.7 mM vs. 1 M). If

one increases the temperature to 90°C, an additional factor of 4-5 reduction in the amount of alkali is possible.

EXAMPLE 7: Effect of temperature-modulated OER thermodynamics on OER kinetics

In this example, the effect of temperature-modulated OER thermodynamics on OER kinetics was quantified, beyond the usual Arrhenius effect, as the temperature varies from 10°C to 90°C.

To this end, the OER current density was calculated at a fixed pH of 13.0, applied potential of 1.51 V vs. the RHE, and pressure of 1 bar, under three different conditions: (i) at 10°C, (ii) at 90°C assuming the OER potential to be unchanged from 10°C, and (iii) at 90°C. The OER current densities in these three cases were found to be ~1.5 mA/cm^2, 197 mA/cm^2, and 808 mA/cm^2, respectively. Thus, by solely accounting for Arrhenius-type effects and not changes in the OER potential, an increase in temperature from 10°C to 90°C would increase the OER current density by about two orders of magnitude. However, the reduction in OER equilibrium potential from 1.24 V (10°C) to 1.18 V (90°C) enables a further increase in the OER current density by nearly an extra order of magnitude, thereby indicating that even minor changes in the OER thermodynamics significantly affect OER kinetics.

EXAMPLE 8: Effect of pressure on OER thermodynamics

The effect of system pressure on OER thermodynamics was tested in this analysis. Although it is possible to operate water electrolyzers under conditions of differential pressure, wherein the anodic (oxygen) headspace is maintained at low pressures and the cathodic (hydrogen) headspace at high pressures, such operation can cause gas flow from one compartment to another. Thus, we consider the case of uniform reactor pressure. The free energy as a function of pressure using data from the NIST database is calculated as:

$$G_i(T,p) = G_i^0(T) + \Delta G_i^{p_0 \to p}(T,p) \tag{27}$$

where $\Delta G_i^{p_0 \to p}(T,p)$ represents the free energy change of pure species $i = \{H_2O(l), H_2(g)\}$ when the system pressure changes from $p_0 = 1$ bar (standard pressure) to any arbitrary pressure p. We calculate $\Delta G_i^{p_0 \to p}(T,p)$ at a fixed temperature T by linearly interpolating between tabulated values with p varying from 1 bar to 100 bar with a linear spacing of 1 bar.

The free energy of pure $O_2(g)$ at arbitrary pressure is calculated as:

$$G_{O_2(g)}(T,p) = \Delta G_{r,H_2O\ split}(T,p) + \left(2G_{H_2O(l)}(T,p) - 2G_{H_2(g)}(T,p)\right) \quad (28)$$

Further,

$$\Delta G_{r,H_2O\ split}(T,p) = -2\Delta G_{f,H_2O(l)}(T,p) \quad (29)$$

However, $\Delta G_{r,H_2O\ split}(T,p) \neq \Delta G_{r,OER}(T,p)$, because $\Delta G_{r,HER}(T,p) \neq 0$ at any arbitrary pressure (these equalities hold only at standard pressure). This is because:

$$\Delta G_{r,HER}(T,p) = G_{H_2(g)}(T,p) - 2G^0_{(H^+ + e^-)}(T) \quad (30)$$

where the two terms on the right-hand side differ because the free energy of the proton-electron couple is always calculated under standard (1 bar) conditions of the SHE. The OER free energy is calculated as follows:

$$\Delta G_{r,OER}(T,p) = G_{O_2(g)}(T,p) + 4G^0_{(H^+ + e^-)}(T) - 2G_{H_2O(l)}(T,p) \quad (31)$$

Using Eqs. (29) through (31), the Gibbs free energies of water splitting, oxygen evolution, and hydrogen evolution at a temperature of 90°C was plotted and provided in Figure 8A. Note that the change in the OER free energy with pressure solely occurs due to the change in the free energy of step 2 (i.e., the one involving O_2(g)) with pressure. Indeed, the free energies of steps 1, 3, and 4 in the OER do not change with pressure (Figure 8B). This is because pressure effects on adsorbed species and the proton-electron couple are respectively negligible (due to adsorbed intermediates being condensed phase entities) and exactly zero (due to the use of 1 bar pressure as a reference).

EXAMPLE 9: Effect of pressure on OER kinetics

The effect of pressure on the kinetics of the OER was determined. The main parameter that changes as a function of pressure is oxygen solubility in water; we assess this quantity self-consistently using Eqs. (20) through (24). Note that the rate constant does not change with pressure; the sole effect of pressure is on the oxygen activity that multiplies the reverse rate constant for reaction 2. The O_2 solubility, $S_{O_2} = k_{O_2}(T)p$, as a function of pressure at two different temperatures, 25°C and 90°C is given in Figure 10. Naturally, the O_2 solubility increases with pressure and decreases with temperature.

The OER current density as a function of the electrolyzer pressure at a fixed temperature of 90°C is given in in Figure 10A, B for two different cases: (i) an applied potential of 1.46 V vs. the RHE (Figure 10A; corresponding to an overpotential of 0.28 V vs. a *fixed* $E^0_{OER,RHE}$(90°C) = 1.18 V) and (ii) an applied potential of 1.51 V vs. the RHE (Figure 11B; corresponding to an overpotential of 0.28 V vs. a *fixed* $E^0_{OER,RHE}$(25°C) = 1.23 V). Although the reactor temperature in both cases is 90°C, we consider $E^0_{OER,RHE}$(25°C) = 1.23 V in case (ii). It is noted that the OER current density decreases mildly in Figure 10A and negligibly in Figure 10B, as pressure changes from 1.013 to 100 bar. This is because the sole effect of pressure is to increase the reverse rate of reaction 2, via an increase in the oxygen activity. As a result, the pressure variation of the OER current density is mild at fixed potentials, with the OER current density decreasing by ~55 mA/cm^2 in case (i) and ~11 mA/cm^2 in case (ii) over the same pressure range at pH = pH^{opt}.

It was also noted that the OER current density is higher at pH = pH^{opt} than at the standard pH = pH^0 (see Figure 10A-B) by construction. Finally, by operating the electrolyzer with an anode potential of 1.51 V vs. the RHE at 90°C and 100 bar pressure, the OER current density would only reduce by ~12 mA/cm^2 to ~985 mA/cm^2 at pH = pH^{opt} (Figure 10B). This suggests that one could obtain compressed hydrogen directly, with no concomitant compromise in the OER kinetics. However, one would need to maintain the pH^{opt} ~8.6 (compared to pH^0 = 12.4) at 90°C, as discussed above. This has the advantage of requiring less amount of alkali.

To summarize the above analysis, the inventors have found that the maximum anodic OER current density is ~997 mA/cm^2 at a temperature of 90°C, as opposed to ~10 mA/cm^2 at room temperature, for an optimal pH = 8.6, standard pressure (1 bar), and an applied potential of 1.51 V vs. the RHE. This finding indicates Fe-doped NiOOH is an appropriate candidate for use in commercial electrolyzers, which typically function under alkaline conditions at current densities of ~400 mA/cm^2 and a temperature of 80 °C. It is also shown that at elevated temperatures (close to 90°C), a pH of 14 corresponds to unphysical species activities, and that one should consider the standard pH^0 ($a_{OH^-} = 1$ M) defined appropriately at each temperature. Furthermore, the role played by the reduction in the OER potential with temperature in accelerating OER kinetics beyond simple Arrhenius effects is also determined.

It was also found that increasing pressure causes the OER potential to increase, thereby making the reaction less favourable thermodynamically. In terms of kinetics, for a fixed applied potential, increasing pressure reduces the OER current density marginally, due to an increased solubility of gaseous oxygen in water. As a result, producing compressed oxygen and hydrogen (~100 bar) involves a negligible trade-off in kinetics, while leading to cost-savings due to an obviation of downstream oxygen and hydrogen compression.

Further, based on the above understanding between the coupled effects of temperature, pressure, and pH (alkali concentration) on the efficiency (OER current density), the present inventors have determined the optimal operating conditions – temperature, pressure and pH, for a range of potentials – for water electrolysis in an electrolyzer and the corresponding OER current density and concentration of alkali required in the method, as given hereinbelow in Table 1 and Table 2. The data generated herein is intended merely to facilitate an understanding of ways in which the embodiments may be practiced and to further enable those of skill in the art to practice the embodiments. Table 1 therefore provides representative examples of optimal combinations of temperature, pressure, pH conditions along with corresponding current density and alkali concentrations required for the said pH at potentials of 1.5 V vs. the RHE (Table 1A), 1.6 V vs. the RHE (Table 1B) and 1.7 V vs. the RHE at the anode (Table 1C). Table 2 further provides representative examples of optimal combinations of temperature, pressure and pH conditions along with alkali concentrations required for the said pH at a potential of 1.5 V vs. the RHE (Table 2A-B) to obtain current density of 10 mA/cm^2 and 10000 mA/cm^2, respectively; at a potential of 1.6 V vs. the RHE (Table 2C) to obtain current density of 10000 mA/cm^2; and at a potential 1.7 V vs. the RHE (Table 2D) at the anode to obtain current density of 10000 mA/cm^2. The data in Table 1 and Table 2 however is not exhaustive, and a person skilled in the art will be able to readily modulate the conditions to arrive at optimum values based on the teachings of the present disclosure. Accordingly, following examples should not be construed as limiting the scope of the disclosure herein.

Table 1: Optimized operating conditions of temperature, pressure, and pH at an applied potential vs. the RHE at the anode and the corresponding OER current density and concentration of alkali for an iron-doped nickel oxide, hydroxide, or oxyhydroxide catalyst.

Table 1A: At an applied potential of 1.5 V vs. the RHE at the anode

1.5 V vs. the RHE at the anode				
Pressure (bar)	**Temperature (°C)**	**pH**	**Current density (mA/cm^2)**	**Alkali concentration (mM)**
1	10	11.373	0.68814	0.69015
1	20	10.996	3.5235	0.67762
1	30	10.604	12.176	0.59039
1	40	10.214	31.856	0.47809
1	50	9.8395	70.214	0.37587
1	60	9.4881	139.39	0.29376
1	70	9.1621	258.21	0.23081
1	80	8.8619	454.58	0.18355
1	90	8.5877	768.34	0.14835
10	10	11.435	0.19051	0.80283
10	20	11.112	1.6055	0.89204
10	30	10.76	8.371	0.85029
10	40	10.374	27.506	0.69599
10	50	9.9817	66.547	0.52514
10	60	9.6071	136.62	0.38895
10	70	9.2597	256.13	0.29102
10	80	8.9418	452.96	0.22216
10	90	8.653	766.98	0.17366
10	100	8.3927	1253.3	0.13933
10	110	8.1605	1986.1	0.11503
10	120	7.9561	3062.6	0.097907
10	130	7.7785	4607.6	0.085913
10	140	7.6256	6778	0.07757
10	150	7.4945	9767.3	0.071642
10	160	7.3816	13810	0.067282
20	10	11.442	0.10546	0.82268
20	20	11.135	0.9996	0.949
20	30	10.811	6.211	0.96575
20	40	10.45	23.878	0.835
20	50	10.065	62.893	0.64078
20	60	9.6864	133.66	0.47056
20	70	9.3311	253.86	0.34573
20	80	9.0045	451.17	0.25867
20	90	8.7072	765.47	0.1983
20	100	8.4389	1251.9	0.15622
20	110	8.1992	1984.7	0.12675
20	120	7.9875	3061	0.10613

20	130	7.8031	4605.6	0.091742
20	140	7.6442	6775.2	0.081708
20	150	7.5081	9763.4	0.074636
20	160	7.3915	13804	0.069508
30	10	11.444	0.072824	0.8329
30	20	11.144	0.72527	0.97658
30	30	10.836	4.9353	1.0288
30	40	10.492	21.091	0.92817
30	50	10.118	59.615	0.72977
30	60	9.7412	130.83	0.538
30	70	9.3832	251.62	0.39274
30	80	9.0521	449.39	0.29085
30	90	8.7498	763.97	0.22046
30	100	8.4764	1250.5	0.17169
30	110	8.2314	1983.3	0.13766
30	120	8.0145	3059.3	0.1139
30	130	7.8249	4603.5	0.097317
30	140	7.6612	6772.5	0.085724
30	150	7.5208	9759.5	0.07758
30	160	7.4007	13799	0.071708
40	10	11.444	0.055562	0.84009
40	20	11.148	0.5688	0.99377
40	30	10.849	4.0931	1.0701
40	40	10.52	18.885	0.9967
40	50	10.156	56.659	0.8024
40	60	9.7826	128.11	0.59629
40	70	9.4239	249.42	0.4346
40	80	9.0903	447.62	0.32007
40	90	8.7849	762.48	0.24086
40	100	8.5079	1249.2	0.18609
40	110	8.2591	1981.8	0.14794
40	120	8.0382	3057.7	0.12129
40	130	7.8444	4601.5	0.10267
40	140	7.6767	6769.7	0.089648
40	150	7.5326	9755.6	0.080463
40	160	7.4094	13793	0.073886
50	10	11.443	0.044882	0.84612
50	20	11.15	0.46768	1.0062
50	30	10.858	3.4956	1.1
50	40	10.539	17.093	1.05
50	50	10.185	53.978	0.86367
50	60	9.8154	125.49	0.648

50	70	9.457	247.26	0.47277
50	80	9.1221	445.87	0.34708
50	90	8.8145	760.99	0.25994
50	100	8.535	1247.8	0.19962
50	110	8.2833	1980.4	0.15768
50	120	8.0592	3056.1	0.1284
50	130	7.862	4599.4	0.10786
50	140	7.6909	6767	0.093477
50	150	7.5437	9751.8	0.083322
50	160	7.4176	13788	0.076026
60	10	11.442	0.037622	0.85113
60	20	11.151	0.39695	1.0162
60	30	10.863	3.0496	1.123
60	40	10.553	15.611	1.0932
60	50	10.207	51.537	0.91672
60	60	9.8423	122.98	0.69468
60	70	9.4849	245.13	0.50796
60	80	9.1492	444.13	0.37227
60	90	8.8401	759.5	0.2779
60	100	8.5586	1246.4	0.21251
60	110	8.3046	1979	0.16703
60	120	8.078	3054.5	0.1352
60	130	7.8781	4597.4	0.1129
60	140	7.7041	6764.2	0.097213
60	150	7.554	9747.9	0.086125
60	160	7.4254	13782	0.078164
70	10	11.441	0.032366	0.85534
70	20	11.151	0.3447	1.0248
70	30	10.867	2.704	1.1417
70	40	10.564	14.363	1.1293
70	50	10.225	49.304	0.96339
70	60	9.8649	120.57	0.73738
70	70	9.5088	243.04	0.5408
70	80	9.1728	442.4	0.39613
70	90	8.8626	758.02	0.29498
70	100	8.5796	1245	0.22484
70	110	8.3239	1977.6	0.176
70	120	8.0951	3052.9	0.1418
70	130	7.8928	4595.3	0.11779
70	140	7.7162	6761.5	0.10088
70	150	7.5637	9744	0.088891
70	160	7.4328	13776	0.080284

80	10	11.439	0.028386	0.85882
80	20	11.151	0.30452	1.0324
80	30	10.87	2.4283	1.1578
80	40	10.572	13.298	1.1602
80	50	10.24	47.254	1.005
80	60	9.8842	118.24	0.77683
80	70	9.5297	240.98	0.57173
80	80	9.1936	440.68	0.41876
80	90	8.8827	756.55	0.31127
80	100	8.5985	1243.7	0.23667
80	110	8.3412	1976.2	0.18467
80	120	8.1106	3051.2	0.14825
80	130	7.9063	4593.3	0.1226
80	140	7.7276	6758.7	0.10447
80	150	7.5728	9740.1	0.091628
80	160	7.4398	13771	0.082366
90	10	11.437	0.025266	0.86297
90	20	11.15	0.27267	1.0394
90	30	10.872	2.2033	1.1716
90	40	10.579	12.379	1.1871
90	50	10.253	45.365	1.0426
90	60	9.901	116	0.81355
90	70	9.5481	238.95	0.60105
90	80	9.2122	438.98	0.44047
90	90	8.9007	755.08	0.32699
90	100	8.6155	1242.3	0.24814
90	110	8.3571	1974.8	0.19308
90	120	8.125	3049.6	0.1545
90	130	7.9189	4591.3	0.12728
90	140	7.7381	6756	0.10803
90	150	7.5814	9736.3	0.094321
90	160	7.4464	13765	0.084452
100	10	11.435	0.022755	0.86607
100	20	11.15	0.2468	1.046
100	30	10.873	2.0162	1.1839
100	40	10.584	11.578	1.2109
100	50	10.264	43.619	1.0769
100	60	9.9157	113.84	0.84783
100	70	9.5645	236.96	0.62896
100	80	9.2289	437.28	0.46127
100	90	8.917	753.61	0.3422
100	100	8.6311	1240.9	0.25922

100	110	8.3716	1973.4	0.2013
100	120	8.1382	3048	0.16061
100	130	7.9306	4589.2	0.13188
100	140	7.7481	6753.2	0.11151
100	150	7.5895	9732.4	0.096996
100	160	7.4528	13760	0.086521
110	10	11.433	0.020691	0.86953
110	20	11.149	0.22537	1.0522
110	30	10.874	1.8581	1.1954
110	40	10.589	10.873	1.2324
110	50	10.273	42	1.1082
110	60	9.9287	111.76	0.88025
110	70	9.5793	234.99	0.65568
110	80	9.2441	435.6	0.48133
110	90	8.9319	752.15	0.35693
110	100	8.6454	1239.6	0.27
110	110	8.385	1972	0.20929
110	120	8.1505	3046.4	0.16659
110	130	7.9415	4587.2	0.1364
110	140	7.7575	6750.5	0.11497
110	150	7.5972	9728.6	0.099641
110	160	7.4588	13754	0.088582
120	10	11.432	0.018964	0.87326
120	20	11.148	0.20732	1.0581
120	30	10.874	1.7228	1.2057
120	40	10.592	10.248	1.2519
120	50	10.281	40.496	1.1373
120	60	9.9403	109.75	0.91093
120	70	9.5927	233.06	0.68135
120	80	9.258	433.93	0.50074
120	90	8.9457	750.69	0.3712
120	100	8.6586	1238.2	0.28056
120	110	8.3974	1970.6	0.21711
120	120	8.162	3044.8	0.17243
120	130	7.9518	4585.2	0.14083
120	140	7.7663	6747.8	0.11836
120	150	7.6045	9724.7	0.10226
120	160	7.4646	13749	0.090639
130	10	11.43	0.017498	0.87689
130	20	11.147	0.19192	1.0641
130	30	10.874	1.6056	1.2153
130	40	10.595	9.6908	1.2695

130	50	10.288	39.094	1.1642
130	60	9.9507	107.81	0.93994
130	70	9.6049	231.15	0.70605
130	80	9.2708	432.26	0.5196
130	90	8.9583	749.23	0.38516
130	100	8.6708	1236.8	0.29085
130	110	8.409	1969.2	0.22474
130	120	8.1726	3043.2	0.17821
130	130	7.9614	4583.2	0.14521
130	140	7.7747	6745.1	0.1217
130	150	7.6115	9720.9	0.10485
130	160	7.4702	13743	0.092666
140	10	11.428	0.016238	0.88048
140	20	11.146	0.17862	1.0695
140	30	10.874	1.5032	1.2245
140	40	10.597	9.19	1.2862
140	50	10.294	37.783	1.1895
140	60	9.9601	105.94	0.96772
140	70	9.6161	229.28	0.72983
140	80	9.2825	430.62	0.53802
140	90	8.9701	747.78	0.39883
140	100	8.6822	1235.5	0.30092
140	110	8.4198	1967.8	0.23226
140	120	8.1826	3041.6	0.18388
140	130	7.9704	4581.1	0.14955
140	140	7.7826	6742.3	0.12504
140	150	7.6181	9717.1	0.10744
140	160	7.4755	13738	0.094704
150	10	11.426	0.015143	0.884
150	20	11.144	0.16702	1.0751
150	30	10.874	1.413	1.2331
150	40	10.599	8.7378	1.3016
150	50	10.299	36.557	1.2131
150	60	9.9686	104.12	0.99416
150	70	9.6263	227.43	0.75296
150	80	9.2934	428.98	0.5559
150	90	8.981	746.34	0.41213
150	100	8.6929	1234.1	0.31081
150	110	8.43	1966.4	0.23967
150	120	8.1922	3040	0.18946
150	130	7.979	4579.1	0.1538
150	140	7.7901	6739.6	0.12834

150	150	7.6245	9713.2	0.11001
150	160	7.4806	13732	0.096722
160	10	11.425	0.014183	0.88864
160	20	11.143	0.15681	1.0807
160	30	10.873	1.3328	1.2413
160	40	10.601	8.3275	1.316
160	50	10.304	35.406	1.2356
160	60	9.9763	102.37	1.0197
160	70	9.6358	225.61	0.77542
160	80	9.3037	427.35	0.5733
160	90	8.9913	744.9	0.42517
160	100	8.7029	1232.8	0.32054
160	110	8.4395	1965	0.24697
160	120	8.201	3038.4	0.195
160	130	7.9871	4577.1	0.15805
160	140	7.7973	6736.9	0.1316
160	150	7.6305	9709.4	0.11257
160	160	7.4855	13726	0.098737
170	10	11.423	0.013335	0.89228
170	20	11.142	0.14776	1.0859
170	30	10.873	1.2611	1.2492
170	40	10.602	7.9534	1.3298
170	50	10.308	34.324	1.2567
170	60	9.9834	100.67	1.044
170	70	9.6446	223.81	0.79711
170	80	9.3131	425.73	0.59038
170	90	9.0009	743.46	0.43805
170	100	8.7123	1231.4	0.33009
170	110	8.4485	1963.6	0.25412
170	120	8.2095	3036.8	0.20042
170	130	7.9948	4575.1	0.16222
170	140	7.804	6734.2	0.13485
170	150	7.6363	9705.6	0.1151
170	160	7.4902	13721	0.10074
180	10	11.422	0.012579	0.8968
180	20	11.141	0.13968	1.0914
180	30	10.872	1.1967	1.2567
180	40	10.603	7.611	1.3427
180	50	10.312	33.304	1.2769
180	60	9.9898	99.023	1.0677
180	70	9.6528	222.05	0.81838
180	80	9.322	424.12	0.60716

180	90	9.0099	742.03	0.45064
180	100	8.7211	1230.1	0.33954
180	110	8.457	1962.2	0.26123
180	120	8.2175	3035.2	0.20581
180	130	8.0022	4573.1	0.16635
180	140	7.8105	6731.5	0.13805
180	150	7.6418	9701.8	0.11762
180	160	7.4947	13715	0.10274
190	10	11.42	0.011902	0.90053
190	20	11.139	0.13243	1.0967
190	30	10.872	1.1384	1.2647
190	40	10.604	7.2964	1.3552
190	50	10.315	32.343	1.2963
190	60	9.9958	97.429	1.0904
190	70	9.6604	220.3	0.83897
190	80	9.3303	422.53	0.62357
190	90	9.0184	740.6	0.46303
190	100	8.7296	1228.7	0.34879
190	110	8.4651	1960.8	0.26821
190	120	8.2251	3033.6	0.21116
190	130	8.0091	4571.1	0.17044
190	140	7.8168	6728.8	0.14125
190	150	7.6472	9698	0.12013
190	160	7.499	13710	0.10475
200	10	11.419	0.011292	0.90481
200	20	11.138	0.12587	1.102
200	30	10.871	1.0854	1.2721
200	40	10.604	7.0064	1.3671
200	50	10.318	31.434	1.3146
200	60	10.001	95.883	1.1123
200	70	9.6674	218.59	0.85924
200	80	9.3382	420.94	0.63978
200	90	9.0264	739.18	0.47525
200	100	8.7375	1227.4	0.35798
200	110	8.4727	1959.4	0.27515
200	120	8.2323	3032	0.21644
200	130	8.0158	4569	0.17452
200	140	7.8227	6726	0.14442
200	150	7.6522	9694.2	0.12264
200	160	7.5032	13705	0.10672
210	10	11.417	0.010739	0.90871
210	20	11.137	0.11992	1.1073

210	30	10.87	1.0371	1.2794
210	40	10.605	6.7382	1.3784
210	50	10.321	30.574	1.3323
210	60	10.006	94.384	1.1335
210	70	9.6741	216.9	0.87892
210	80	9.3456	419.37	0.65553
210	90	9.0341	737.76	0.4873
210	100	8.745	1226	0.36707
210	110	8.48	1958	0.282
210	120	8.2392	3030.4	0.22166
210	130	8.0222	4567	0.17856
210	140	7.8284	6723.3	0.14757
210	150	7.6571	9690.4	0.12513
210	160	7.5072	13699	0.10873

Table 1B: At an applied potential of 1.6 V vs. the RHE at the anode

1.6 V vs. the RHE at the anode				
Pressure (bar)	**Temperature (°C)**	**pH**	**Current density (mA/cm^2)**	**Alkali concentration (mM)**
1	10	11.373	84.938	0.69015
1	20	10.996	184.27	0.67762
1	30	10.604	371.05	0.59039
1	40	10.214	704.03	0.47809
1	50	9.8395	1271.2	0.37587
1	60	9.4881	2199.1	0.29376
1	70	9.1621	3664.1	0.23081
1	80	8.8619	5903.8	0.18355
1	90	8.5877	9230.1	0.14835
10	10	11.435	84.577	0.80283
10	20	11.112	183.94	0.89204
10	30	10.76	370.72	0.85029
10	40	10.374	703.65	0.69599
10	50	9.9817	1270.7	0.52514
10	60	9.6071	2198.4	0.38895
10	70	9.2597	3663.1	0.29102
10	80	8.9418	5902.3	0.22216
10	90	8.653	9227.9	0.17366
10	100	8.3927	14038	0.13933
20	10	11.442	84.179	0.82268
20	20	11.135	183.58	0.949
20	30	10.811	370.35	0.96575

20	40	10.45	703.23	0.835
20	50	10.065	1270.1	0.64078
20	60	9.6864	2197.7	0.47056
20	70	9.3311	3662	0.34573
20	80	9.0045	5900.7	0.25867
20	90	8.7072	9225.4	0.1983
20	100	8.4389	14035	0.15622
30	10	11.444	83.785	0.8329
30	20	11.144	183.22	0.97658
30	30	10.836	369.99	1.0288
30	40	10.492	702.82	0.92817
30	50	10.118	1269.6	0.72977
30	60	9.7412	2197	0.538
30	70	9.3832	3661	0.39274
30	80	9.0521	5899.1	0.29085
30	90	8.7498	9223	0.22046
30	100	8.4764	14031	0.17169
40	10	11.444	83.393	0.84009
40	20	11.148	182.86	0.99377
40	30	10.849	369.63	1.0701
40	40	10.52	702.4	0.9967
40	50	10.156	1269.1	0.8024
40	60	9.7826	2196.2	0.59629
40	70	9.4239	3659.9	0.4346
40	80	9.0903	5897.5	0.32007
40	90	8.7849	9220.6	0.24086
40	100	8.5079	14027	0.18609
50	10	11.443	83.004	0.84612
50	20	11.15	182.5	1.0062
50	30	10.858	369.26	1.1
50	40	10.539	701.99	1.05
50	50	10.185	1268.6	0.86367
50	60	9.8154	2195.5	0.648
50	70	9.457	3658.8	0.47277
50	80	9.1221	5895.9	0.34708
50	90	8.8145	9218.2	0.25994
50	100	8.535	14024	0.19962
60	10	11.442	82.618	0.85113
60	20	11.151	182.14	1.0162
60	30	10.863	368.9	1.123
60	40	10.553	701.58	1.0932
60	50	10.207	1268	0.91672

60	60	9.8423	2194.8	0.69468
60	70	9.4849	3657.7	0.50796
60	80	9.1492	5894.3	0.37227
60	90	8.8401	9215.8	0.2779
60	100	8.5586	14020	0.21251
70	10	11.441	82.235	0.85534
70	20	11.151	181.78	1.0248
70	30	10.867	368.54	1.1417
70	40	10.564	701.17	1.1293
70	50	10.225	1267.5	0.96339
70	60	9.8649	2194	0.73738
70	70	9.5088	3656.7	0.5408
70	80	9.1728	5892.7	0.39613
70	90	8.8626	9213.3	0.29498
70	100	8.5796	14016	0.22484
80	10	11.439	81.854	0.85882
80	20	11.151	181.42	1.0324
80	30	10.87	368.18	1.1578
80	40	10.572	700.76	1.1602
80	50	10.24	1267	1.005
80	60	9.8842	2193.3	0.77683
80	70	9.5297	3655.6	0.57173
80	80	9.1936	5891.1	0.41876
80	90	8.8827	9210.9	0.31127
80	100	8.5985	14013	0.23667
90	10	11.437	81.476	0.86297
90	20	11.15	181.07	1.0394
90	30	10.872	367.82	1.1716
90	40	10.579	700.35	1.1871
90	50	10.253	1266.5	1.0426
90	60	9.901	2192.6	0.81355
90	70	9.5481	3654.6	0.60105
90	80	9.2122	5889.6	0.44047
90	90	8.9007	9208.5	0.32699
90	100	8.6155	14009	0.24814
100	10	11.435	81.101	0.86607
100	20	11.15	180.71	1.046
100	30	10.873	367.46	1.1839
100	40	10.584	699.94	1.2109
100	50	10.264	1265.9	1.0769
100	60	9.9157	2191.8	0.84783
100	70	9.5645	3653.5	0.62896

100	80	9.2289	5888	0.46127
100	90	8.917	9206.1	0.3422
100	100	8.6311	14005	0.25922
110	10	11.433	80.728	0.86953
110	20	11.149	180.36	1.0522
110	30	10.874	367.1	1.1954
110	40	10.589	699.53	1.2324
110	50	10.273	1265.4	1.1082
110	60	9.9287	2191.1	0.88025
110	70	9.5793	3652.4	0.65568
110	80	9.2441	5886.4	0.48133
110	90	8.9319	9203.7	0.35693
110	100	8.6454	14002	0.27
120	10	11.432	80.359	0.87326
120	20	11.148	180.01	1.0581
120	30	10.874	366.74	1.2057
120	40	10.592	699.12	1.2519
120	50	10.281	1264.9	1.1373
120	60	9.9403	2190.4	0.91093
120	70	9.5927	3651.4	0.68135
120	80	9.258	5884.8	0.50074
120	90	8.9457	9201.3	0.3712
120	100	8.6586	13998	0.28056
130	10	11.43	79.991	0.87689
130	20	11.147	179.65	1.0641
130	30	10.874	366.38	1.2153
130	40	10.595	698.71	1.2695
130	50	10.288	1264.4	1.1642
130	60	9.9507	2189.7	0.93994
130	70	9.6049	3650.3	0.70605
130	80	9.2708	5883.2	0.5196
130	90	8.9583	9198.9	0.38516
130	100	8.6708	13995	0.29085
140	10	11.428	79.627	0.88048
140	20	11.146	179.3	1.0695
140	30	10.874	366.02	1.2245
140	40	10.597	698.3	1.2862
140	50	10.294	1263.8	1.1895
140	60	9.9601	2189	0.96772
140	70	9.6161	3649.3	0.72983
140	80	9.2825	5881.6	0.53802
140	90	8.9701	9196.5	0.39883

140	100	8.6822	13991	0.30092
150	10	11.426	79.265	0.884
150	20	11.144	178.95	1.0751
150	30	10.874	365.67	1.2331
150	40	10.599	697.9	1.3016
150	50	10.299	1263.3	1.2131
150	60	9.9686	2188.2	0.99416
150	70	9.6263	3648.2	0.75296
150	80	9.2934	5880.1	0.5559
150	90	8.981	9194.2	0.41213
150	100	8.6929	13987	0.31081
160	10	11.425	78.905	0.88864
160	20	11.143	178.6	1.0807
160	30	10.873	365.31	1.2413
160	40	10.601	697.49	1.316
160	50	10.304	1262.8	1.2356
160	60	9.9763	2187.5	1.0197
160	70	9.6358	3647.2	0.77542
160	80	9.3037	5878.5	0.5733
160	90	8.9913	9191.8	0.42517
160	100	8.7029	13984	0.32054
170	10	11.423	78.548	0.89228
170	20	11.142	178.25	1.0859
170	30	10.873	364.95	1.2492
170	40	10.602	697.08	1.3298
170	50	10.308	1262.3	1.2567
170	60	9.9834	2186.8	1.044
170	70	9.6446	3646.1	0.79711
170	80	9.3131	5876.9	0.59038
170	90	9.0009	9189.4	0.43805
170	100	8.7123	13980	0.33009
180	10	11.422	78.194	0.8968
180	20	11.141	177.9	1.0914
180	30	10.872	364.6	1.2567
180	40	10.603	696.68	1.3427
180	50	10.312	1261.8	1.2769
180	60	9.9898	2186.1	1.0677
180	70	9.6528	3645.1	0.81838
180	80	9.322	5875.3	0.60716
180	90	9.0099	9187	0.45064
180	100	8.7211	13976	0.33954
190	10	11.42	77.842	0.90053

190	20	11.139	177.56	1.0967
190	30	10.872	364.24	1.2647
190	40	10.604	696.27	1.3552
190	50	10.315	1261.3	1.2963
190	60	9.9958	2185.4	1.0904
190	70	9.6604	3644	0.83897
190	80	9.3303	5873.8	0.62357
190	90	9.0184	9184.6	0.46303
190	100	8.7296	13973	0.34879
200	10	11.419	77.492	0.90481
200	20	11.138	177.21	1.102
200	30	10.871	363.88	1.2721
200	40	10.604	695.86	1.3671
200	50	10.318	1260.7	1.3146
200	60	10.001	2184.6	1.1123
200	70	9.6674	3643	0.85924
200	80	9.3382	5872.2	0.63978
200	90	9.0264	9182.3	0.47525
200	100	8.7375	13969	0.35798
210	10	11.417	77.145	0.90871
210	20	11.137	176.86	1.1073
210	30	10.87	363.53	1.2794
210	40	10.605	695.46	1.3784
210	50	10.321	1260.2	1.3323
210	60	10.006	2183.9	1.1335
210	70	9.6741	3641.9	0.87892
210	80	9.3456	5870.6	0.65553
210	90	9.0341	9179.9	0.4873
210	100	8.745	13966	0.36707

Table 1C: At an applied potential of 1.7 V vs. the RHE at the anode

1.7 V vs. the RHE at the anode				
Pressure (bar)	**Temperature (°C)**	**pH**	**Current density (mA/cm^2)**	**Alkali concentration (mM)**
1	10	11.373	1627	0.69015
1	20	10.996	3141.8	0.67762
1	30	10.604	5707.5	0.59039
1	40	10.214	9862.7	0.47809
1	50	9.8395	16337	0.37587
10	10	11.435	1626.2	0.80283
10	20	11.112	3140.7	0.89204

10	30	10.76	5705.7	0.85029
10	40	10.374	9859.9	0.69599
10	50	9.9817	16333	0.52514
20	10	11.442	1625.4	0.82268
20	20	11.135	3139.4	0.949
20	30	10.811	5703.7	0.96575
20	40	10.45	9856.8	0.835
20	50	10.065	16329	0.64078
30	10	11.444	1624.6	0.8329
30	20	11.144	3138.1	0.97658
30	30	10.836	5701.7	1.0288
30	40	10.492	9853.8	0.92817
30	50	10.118	16324	0.72977
40	10	11.444	1623.8	0.84009
40	20	11.148	3136.8	0.99377
40	30	10.849	5699.7	1.0701
40	40	10.52	9850.8	0.9967
40	50	10.156	16319	0.8024
50	10	11.443	1623	0.84612
50	20	11.15	3135.6	1.0062
50	30	10.858	5697.8	1.1
50	40	10.539	9847.8	1.05
50	50	10.185	16315	0.86367
60	10	11.442	1622.1	0.85113
60	20	11.151	3134.3	1.0162
60	30	10.863	5695.8	1.123
60	40	10.553	9844.8	1.0932
60	50	10.207	16310	0.91672
70	10	11.441	1621.3	0.85534
70	20	11.151	3133	1.0248
70	30	10.867	5693.9	1.1417
70	40	10.564	9841.8	1.1293
70	50	10.225	16306	0.96339
80	10	11.439	1620.5	0.85882
80	20	11.151	3131.8	1.0324
80	30	10.87	5692	1.1578
80	40	10.572	9838.8	1.1602
80	50	10.24	16301	1.005
90	10	11.437	1619.7	0.86297
90	20	11.15	3130.5	1.0394
90	30	10.872	5690	1.1716
90	40	10.579	9835.8	1.1871

90	50	10.253	16297	1.0426
100	10	11.435	1618.9	0.86607
100	20	11.15	3129.3	1.046
100	30	10.873	5688.1	1.1839
100	40	10.584	9832.9	1.2109
100	50	10.264	16292	1.0769
110	10	11.433	1618.1	0.86953
110	20	11.149	3128	1.0522
110	30	10.874	5686.1	1.1954
110	40	10.589	9829.9	1.2324
110	50	10.273	16288	1.1082
120	10	11.432	1617.3	0.87326
120	20	11.148	3126.8	1.0581
120	30	10.874	5684.2	1.2057
120	40	10.592	9827	1.2519
120	50	10.281	16283	1.1373
130	10	11.43	1616.5	0.87689
130	20	11.147	3125.5	1.0641
130	30	10.874	5682.3	1.2153
130	40	10.595	9824	1.2695
130	50	10.288	16279	1.1642
140	10	11.428	1615.6	0.88048
140	20	11.146	3124.3	1.0695
140	30	10.874	5680.4	1.2245
140	40	10.597	9821.1	1.2862
140	50	10.294	16274	1.1895
150	10	11.426	1614.8	0.884
150	20	11.144	3123	1.0751
150	30	10.874	5678.4	1.2331
150	40	10.599	9818.1	1.3016
150	50	10.299	16270	1.2131
160	10	11.425	1614	0.88864
160	20	11.143	3121.8	1.0807
160	30	10.873	5676.5	1.2413
160	40	10.601	9815.2	1.316
160	50	10.304	16265	1.2356
170	10	11.423	1613.2	0.89228
170	20	11.142	3120.5	1.0859
170	30	10.873	5674.6	1.2492
170	40	10.602	9812.3	1.3298
170	50	10.308	16261	1.2567
180	10	11.422	1612.4	0.8968

180	20	11.141	3119.3	1.0914
180	30	10.872	5672.7	1.2567
180	40	10.603	9809.3	1.3427
180	50	10.312	16256	1.2769
190	10	11.42	1611.6	0.90053
190	20	11.139	3118	1.0967
190	30	10.872	5670.8	1.2647
190	40	10.604	9806.4	1.3552
190	50	10.315	16252	1.2963
200	10	11.419	1610.8	0.90481
200	20	11.138	3116.8	1.102
200	30	10.871	5668.9	1.2721
200	40	10.604	9803.5	1.3671
200	50	10.318	16248	1.3146
210	10	11.417	1610	0.90871
210	20	11.137	3115.5	1.1073
210	30	10.87	5666.9	1.2794
210	40	10.605	9800.6	1.3784
210	50	10.321	16243	1.3323

Table 2: Optimized operating conditions of temperature, pressure, and pH at an applied potential at the anode along with concentration of alkali for an iron-doped nickel oxide, hydroxide, or oxyhydroxide catalyst to obtain the OER current density of about 10 mA/cm^2 and about 10000 mA/cm^2.

Table 2A: At an applied potential of 1.5 V vs. the RHE at the anode to reach 10 mA/cm^2 current density

1.5 V vs. the RHE at the anode				
Pressure (bar)	**Temperature (°C)**	**pH**	**Current density (mA/cm^2)**	**Alkali concentration (mM)**
1	28.22	10.675	10	0.60871
10	31.297	10.711	10	0.83435
20	33.162	10.702	10	0.93753
30	34.457	10.688	10	1.0019
40	35.46	10.675	10	1.0493
50	36.282	10.662	10	1.087
60	36.981	10.651	10	1.1189

70	37.59	10.64	10	1.1467
80	38.131	10.631	10	1.1715
90	38.618	10.621	10	1.1941
100	39.062	10.613	10	1.2148
110	39.469	10.605	10	1.2341
120	39.846	10.597	10	1.2523
130	40.196	10.589	10	1.2691
140	40.524	10.582	10	1.2852
150	40.833	10.576	10	1.3006
160	41.124	10.569	10	1.3152
170	41.4	10.563	10	1.3295
180	41.661	10.557	10	1.3431
190	41.911	10.551	10	1.3566
200	42.149	10.545	10	1.3696
210	42.377	10.54	10	1.3822

Table 2B: At an applied potential of 1.5 V vs. the RHE at the anode to reach 10 A/cm^2 current density

1.5 V vs. the RHE at the anode				
Pressure (bar)	**Temperature (°C)**	**pH**	**Current density (mA/cm^2)**	**Alkali concentration (mM)**
10	150.66	7.4864	10000	0.07128
20	150.67	7.4997	10000	0.074229
30	150.69	7.5121	10000	0.077119
40	150.7	7.5235	10000	0.079935
50	150.71	7.5342	10000	0.082713
60	150.72	7.5441	10000	0.08545
70	150.73	7.5534	10000	0.088153
80	150.74	7.5622	10000	0.090823
90	150.75	7.5705	10000	0.093448
100	150.76	7.5783	10000	0.09606
110	150.78	7.5858	10000	0.098637
120	150.79	7.5928	10000	0.10119
130	150.8	7.5995	10000	0.10372
140	150.81	7.6059	10000	0.10624
150	150.82	7.6119	10000	0.10874
160	150.83	7.6177	10000	0.11123
170	150.84	7.6233	10000	0.11369

180	150.85	7.6285	10000	0.11615
190	150.86	7.6336	10000	0.11858
200	150.88	7.6384	10000	0.12103
210	150.89	7.643	10000	0.12344

Table 2C: At an applied potential of 1.6 V vs. the RHE at the anode to reach 10 A/cm^2 current density

1.6 V vs. the RHE at the anode				
Pressure (bar)	**Temperature (°C)**	**pH**	**Current density (mA/cm^2)**	**Alkali concentration (mM)**
1	91.862	8.5397	10000	0.14285
10	91.868	8.6024	10000	0.16626
20	91.874	8.6549	10000	0.18916
30	91.88	8.6964	10000	0.20983
40	91.886	8.7306	10000	0.22887
50	91.892	8.7596	10000	0.24669
60	91.898	8.7846	10000	0.26349
70	91.904	8.8067	10000	0.27948
80	91.91	8.8264	10000	0.29475
90	91.917	8.8441	10000	0.30949
100	91.923	8.8601	10000	0.32372
110	91.929	8.8748	10000	0.33753
120	91.935	8.8883	10000	0.35093
130	91.941	8.9007	10000	0.36402
140	91.947	8.9122	10000	0.37682
150	91.953	8.9229	10000	0.3893
160	91.959	8.933	10000	0.40154
170	91.965	8.9423	10000	0.41361
180	91.971	8.9512	10000	0.42543
190	91.977	8.9595	10000	0.43705
200	91.983	8.9674	10000	0.44851
210	91.989	8.9748	10000	0.45983

Table 2D: At an applied potential of 1.7 V vs. the RHE at the anode to reach 10 A/cm^2 current density

1.7 V vs. the RHE at the anode				
Pressure (bar)	**Temperature (°C)**	**pH**	**Current density (mA/cm^2)**	**Alkali concentration (mM)**
1	40.264	10.204	10000	0.47517
10	40.269	10.363	10000	0.69116
20	40.275	10.439	10000	0.82988
30	40.281	10.482	10000	0.92329
40	40.287	10.51	10000	0.99223
50	40.293	10.529	10000	1.046
60	40.298	10.544	10000	1.0897
70	40.304	10.554	10000	1.1262
80	40.31	10.563	10000	1.1575
90	40.316	10.569	10000	1.1848
100	40.321	10.575	10000	1.209
110	40.327	10.579	10000	1.2308
120	40.333	10.582	10000	1.2507
130	40.339	10.585	10000	1.2687
140	40.344	10.588	10000	1.2855
150	40.35	10.589	10000	1.3013
160	40.356	10.591	10000	1.316
170	40.361	10.592	10000	1.33
180	40.367	10.593	10000	1.3432
190	40.373	10.594	10000	1.356
200	40.378	10.594	10000	1.368
210	40.384	10.594	10000	1.3796

The foregoing description of the specific embodiments reveal the general nature of the embodiments herein that others can, by applying current knowledge, readily modify and/or adapt for various applications such specific embodiments without departing from the generic concept, and, therefore, such adaptations and modifications should and are intended to be comprehended within the meaning and range of equivalents of the disclosed embodiments. It is to be understood that the phraseology or terminology employed herein is for the purpose of description and not of limitation. Therefore, while the embodiments in this disclosure have been described in terms of preferred embodiments, those skilled in the art will recognize that

the embodiments herein can be practiced with modification within the spirit and scope of the embodiments as described herein.

Throughout this specification, the term 'combinations thereof' or 'any combination thereof' or 'any combinations thereof' are used interchangeably and are intended to have the same meaning, as regularly known in the field of patent disclosures.

As regards the embodiments characterized in this specification, it is intended that each embodiment be read independently as well as in combination with another embodiment. For example, in case of an embodiment 1 reciting 3 alternatives A, B and C, an embodiment 2 reciting 3 alternatives D, E and F and an embodiment 3 reciting 3 alternatives G, H and I, it is to be understood that the specification unambiguously discloses embodiments corresponding to combinations A, D, G; A, D, H; A, D, I; A, E, G; A, E, H; A, E, I; A, F, G; A, F, H; A, F, I; B, D, G; B, D, H; B, D, I; B, E, G; B, E, H; B, E, I; B, F, G; B, F, H; B, F, I; C, D, G; C, D, H; C, D, I; C, E, G; C, E, H; C, E, I; C, F, G; C, F, H; C, F, I, unless specifically mentioned otherwise.

Any discussion of documents, acts, materials, devices, articles and the like that has been included in this specification is solely for the purpose of providing a context for the disclosure. It is not to be taken as an admission that any or all of these matters form a part of the prior art base or were common general knowledge in the field relevant to the disclosure as it existed anywhere before the priority date of this application.

WE CLAIM:

1. A method of generating oxygen by electrochemical water splitting at optimized conditions of pH, temperature, and pressure for an oxygen evolution reaction (OER) current density ranging from about 10 mA/cm^2 to about 10000 mA/cm^2, said method comprising:
   a) contacting an anode of an electrolyzer with an electrolyte comprising water and alkali, in presence of a metal-doped oxide, hydroxide, or oxyhydroxide catalyst; and
   b) applying a potential ranging from about 1.45 V to about 1.75 V versus the reversible hydrogen electrode (RHE) at the anode to allow said generation of the oxygen;
   wherein the pH of the electrolyte ranges from about 7.4 to about 10.9;
   wherein the pH is a function of temperature T at a given pressure P; and
   wherein the temperature T ranges from about 28 °C to about 151 °C and is less than the boiling point of the electrolyte at the given pressure P.

2. The method as claimed in claim 1, wherein the pH of the electrolyte is determined based on an unconstrained minimization function applied to the negative of an OER current density, and wherein the OER current density is a function of the applied potential.

3. The method as claimed in claim 2, wherein the pH of the electrolyte is maintained in a manner that allows concentration of OH^- ions in the electrolyte to be less than 1 M.

4. The method as claimed in claim 1, wherein when the potential is about 1.5 V vs. the RHE, the pH of the electrolyte ranges from about 7.4 to about 10.7, and the temperature T ranges from about 28 °C to about 151 °C and is less than the boiling point of the electrolyte at the given pressure P.

5. The method as claimed in claim 1, wherein when the potential is about 1.6 V vs. the RHE, the pH of the electrolyte ranges from about 8.5 to about 10.9, and the temperature T ranges from about 28 °C to about 92 °C and is less than boiling point of the electrolyte at the given pressure P.

6. The method as claimed in claim 1, wherein when the potential is about 1.7 V vs. the RHE, the pH of the electrolyte ranges from about 10.2 to about 10.9, and the

temperature *T* ranges from about 28 °C to about 41 °C and is less than boiling point of the electrolyte at the given pressure *P*.

7. The method as claimed in any one of claims 4 to 6, wherein concentration or amount of the alkali required for maintaining the stated pH, ranges from about 0.07 mM to about 1.38 mM.

8. The method as claimed in claim 7, wherein the pH reduces from about 10.7 to about 7.4 with an increase in temperature and the corresponding increase in the OER current density at the potential of about 1.5 V vs. the RHE; and wherein the concentration or amount of the alkali required for maintaining the stated pH reduces from about 1.38 mM to about 0.07 mM at the given pressure *P*.

9. The method as claimed in claim 7, wherein the pH reduces from about 10.9 to about 8.5 with an increase in temperature and the corresponding increase in the OER current density at the potential of about 1.6 V vs. the RHE; and wherein the concentration or amount of the alkali required for maintaining the stated pH reduces from about 1.37 mM to about 0.14 mM at the given pressure *P*.

10. The method as claimed in claim 7, wherein the pH reduces from about 10.9 to about 10.2 with an increase in temperature and the corresponding increase in the OER current density at the potential of about 1.7 V vs. the RHE; and wherein the concentration or amount of the alkali required for maintaining the stated pH reduces from about 1.37 mM to about 0.47 mM at the given pressure *P*.

11. The method as claimed in any one of claims 1 to 10, wherein the pressure *P* ranges from about 1 bar to about 210 bar.

12. The method as claimed in claim 1, wherein the OER current density improves by at least about 80% when the method is carried out at said optimized conditions of pH, temperature, and pressure.

13. The method as claimed in claim 1, wherein the OER current density improves by about 200% when the method is carried out at said optimized conditions of pH, temperature, and pressure.

14. The method as claimed in claim 1, wherein the alkali is selected from a group of highly soluble hydroxides comprising potassium hydroxide (KOH), sodium hydroxide (NaOH), calcium hydroxide ($Ca(OH)_2$), magnesium hydroxide ($Mg(OH)_2$), and an alkali or alkaline earth hydroxide, or any combination thereof.

15. The method as claimed in claim 1, wherein the metal-doped oxide, hydroxide, or oxyhydroxide catalyst is an iron-doped nickel oxide, hydroxide, or oxyhydroxide catalyst, also referred to as a mixed iron/nickel oxide, hydroxide, or oxyhydroxide material.

16. The method as claimed in claim 1, wherein the electrolyzer further comprises a cathode within the electrolyte, and wherein applying a potential across the electrolyzer generates oxygen at the anode and hydrogen at the cathode.

17. The method as claimed in claim 1, further comprising capture of the generated oxygen.

18. The method as claimed in claim 1, further comprising capture of the generated hydrogen.

19. The method as claimed in claim 1, wherein the alkali is KOH or NaOH at a concentration or amount of about 0.15 mM, the catalyst is a mixed iron/nickel oxide, hydroxide, or oxyhydroxide catalyst, the potential applied at the anode is about 1.5 V vs. the RHE, the temperature is about 90 °C, the pressure is about 1 bar, and the pH of the electrolyte is maintained at about 8.6.

20. The method as claimed in claim 1, wherein the alkali is KOH or NaOH at a concentration or amount of about 0.26 mM, the catalyst is a mixed iron/nickel oxide, hydroxide, or oxyhydroxide catalyst, the potential applied at the anode is about 1.5 V vs. the RHE, the temperature is about 100 °C, the pressure is about 100 bar, and the pH of the electrolyte is maintained at about 8.6.

21. The method as claimed in claim 1, wherein the alkali is KOH or NaOH having a concentration or amount of about 0.27 mM, the catalyst is a mixed iron/nickel oxide, hydroxide, or oxyhydroxide catalyst, the potential applied at the anode is about 1.5 V vs. the RHE, the temperature is about 110 °C, the pressure is about 200 bar, and the pH of the electrolyte is maintained at about 8.5.

22. The method as claimed in claim 19, wherein the OER current density is about 770 mA/cm^2.

23. The method as claimed in claim 20, wherein the OER current density is about 1241 mA/cm^2.

24. The method as claimed in claim 21, wherein the OER current density is about 1960 mA/cm^2.

25. The method as claimed in claim 1, wherein when the potential is about 1.5 V vs. the RHE, the pH of the electrolyte ranges from about 7.4 to about 10.6, and the temperature *T* ranges from about 40 °C to about 151 °C or from about 50 °C to about 151 °C and is less than the boiling point of the electrolyte at the given pressure *P*.

26. The method as claimed in claim 1, wherein when the potential is about 1.6 V vs. the RHE, the pH of the electrolyte ranges from about 8.5 to about 10.7, and the temperature *T* ranges from about 40 °C to about 92 °C or from about 50 °C to about 92 °C and is less than boiling point of the electrolyte at the given pressure *P*.

27. The method as claimed in claim 1, wherein when the potential is about 1.7 V vs. the RHE, the pH of the electrolyte ranges from about 10.2 to about 10.6, and the temperature *T* ranges from about 40 °C to about 41 °C and is less than boiling point of the electrolyte at the given pressure *P*.

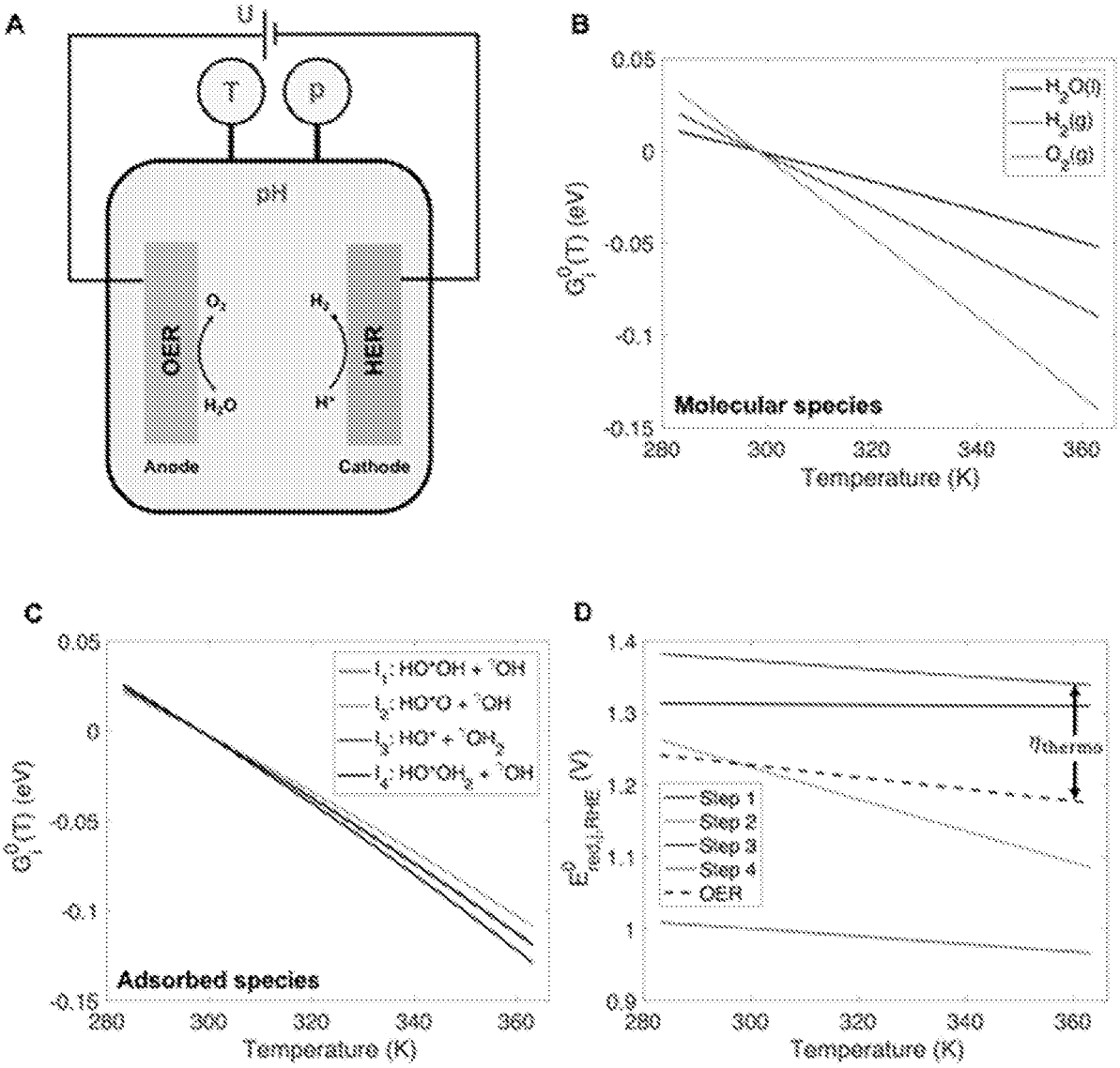

A
U
T
p
pH
OER
HER
O_2
H_2O
H_2
H^+
Anode
Cathode
B
$H_2O(l)$
$H_2(g)$
$O_2(g)$
$G_i^0(T)$ (eV)
Molecular species
Temperature (K)
C
I_1: HO*OH + *OH
I_2: HO*O + *OH
I_3: HO* + *OH_2
I_4: HO*OH_2 + *OH
$G_i^0(T)$ (eV)
Adsorbed species
Temperature (K)
D
η_{therma}
Step 1
Step 2
Step 3
Step 4
OER
$\varepsilon^0_{rev,therm}$ (V)
Temperature (K)

Figure 1

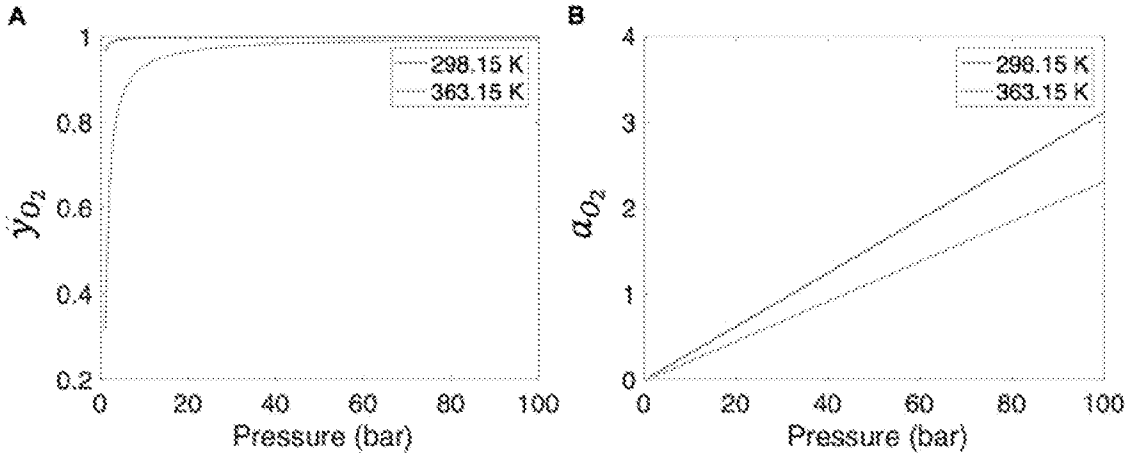


Figure 2

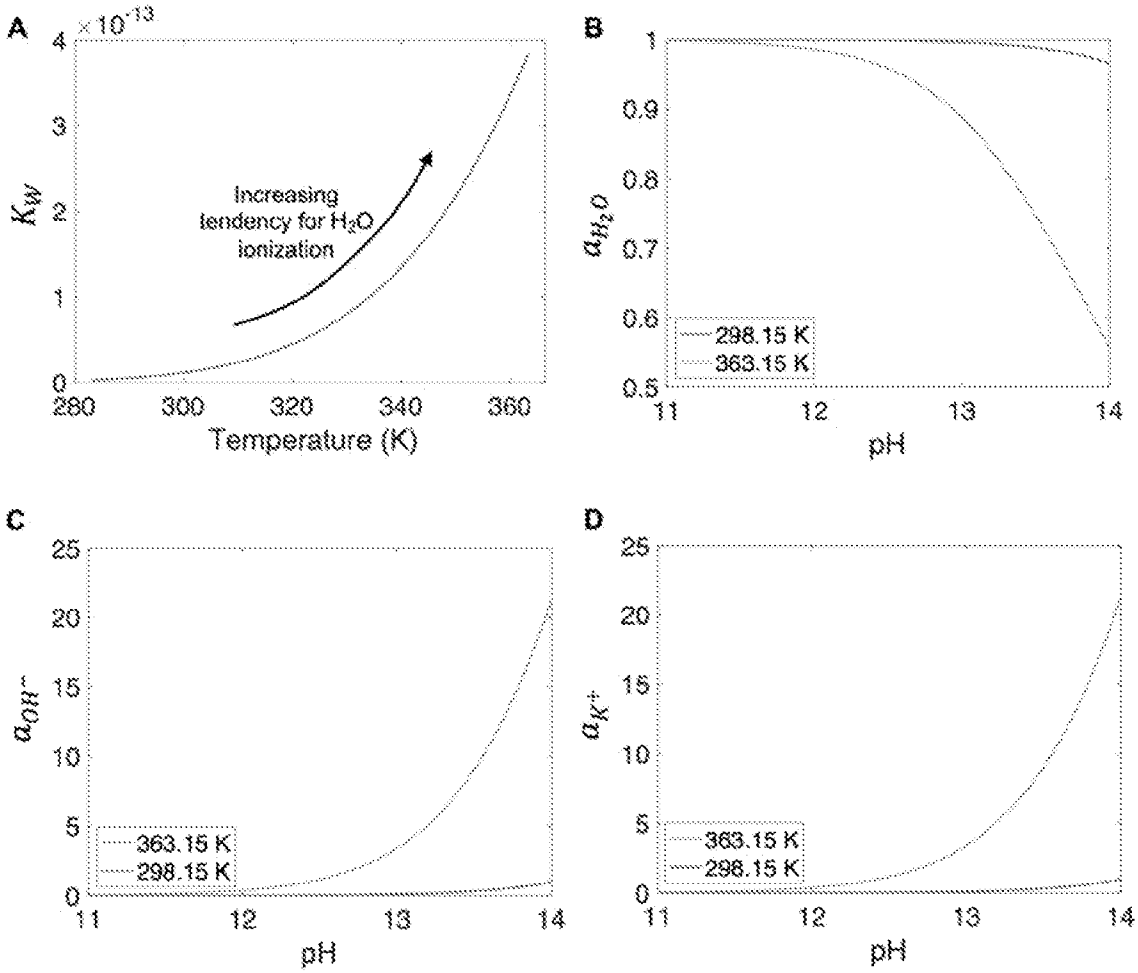


Figure 3

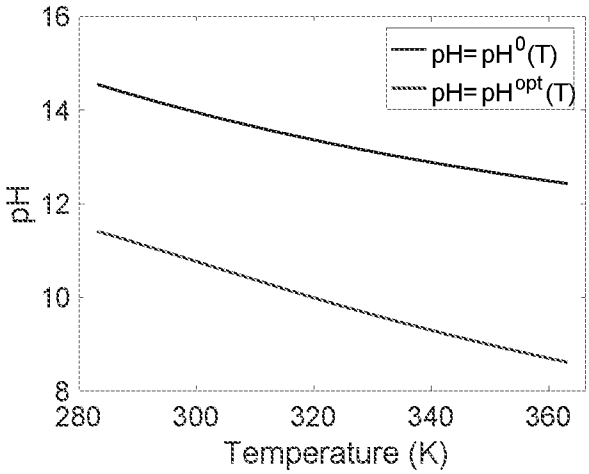


Figure 4

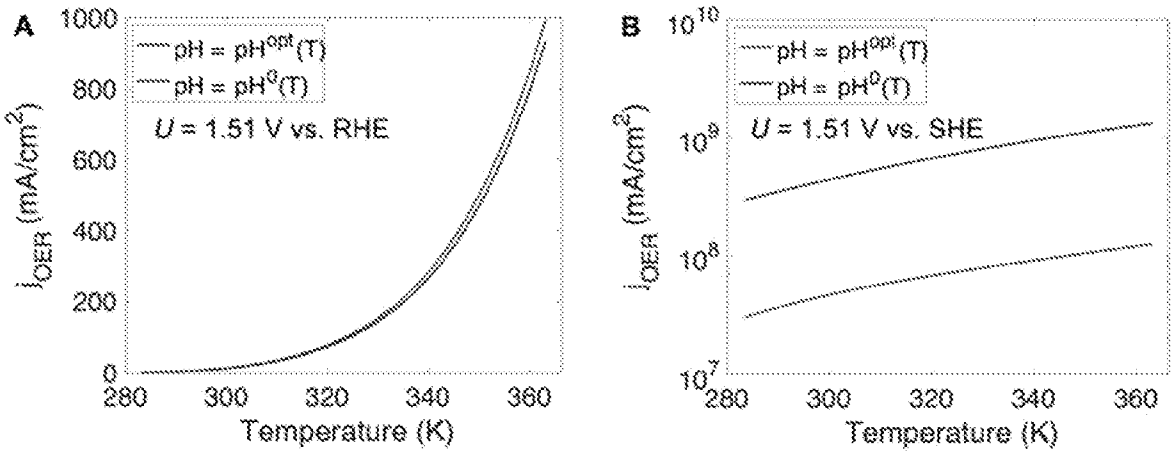


Figure 5

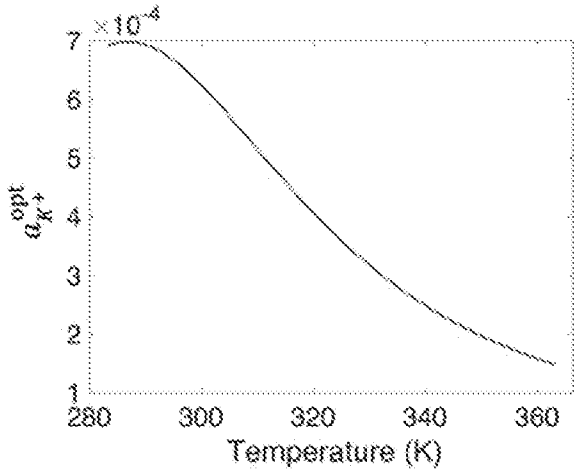


Figure 6

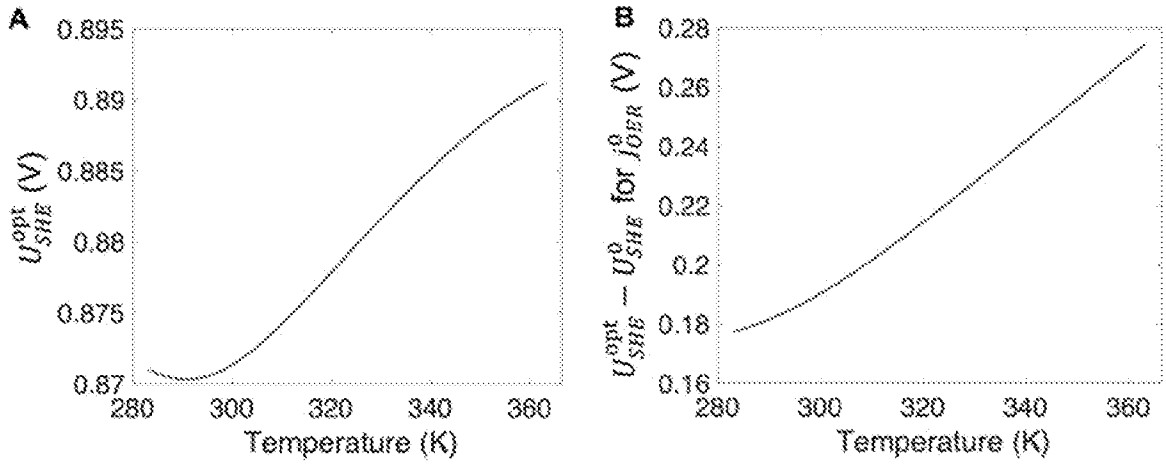


Figure 7

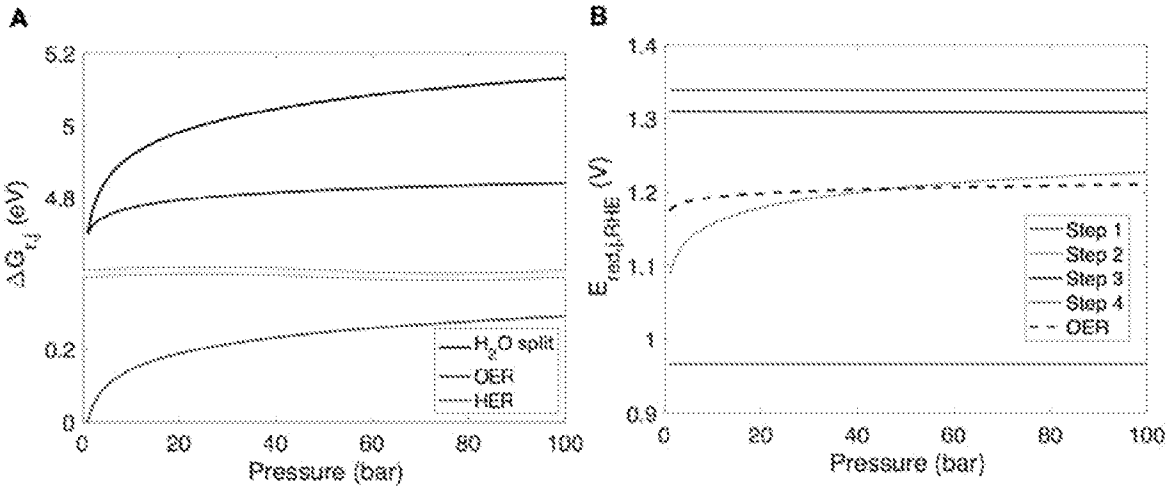


Figure 8

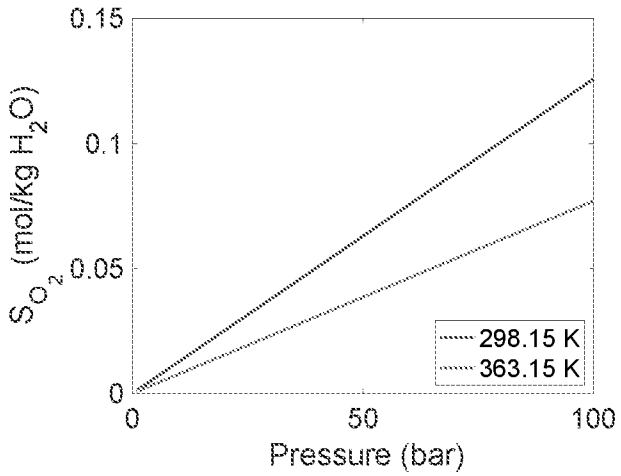


Figure 9

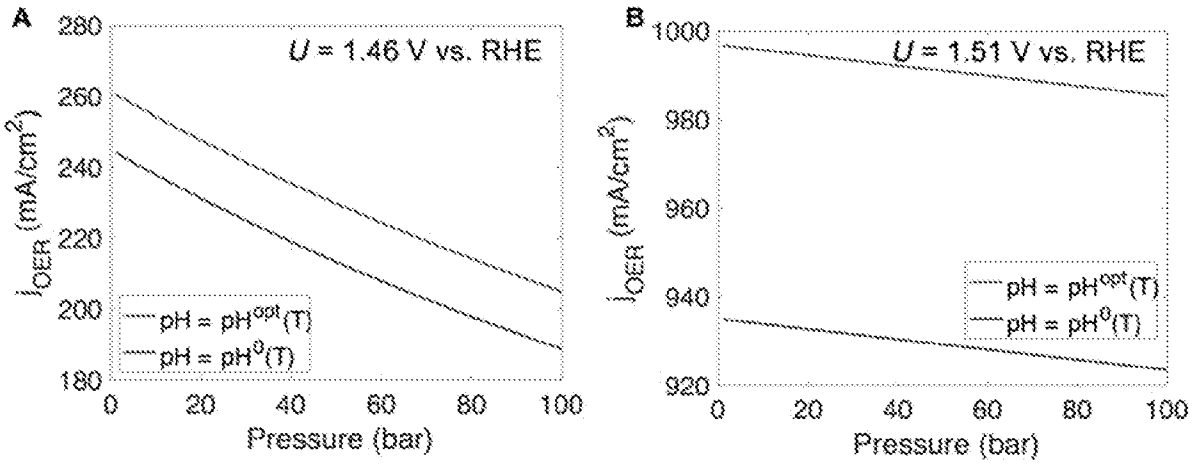

A
U = 1.46 V vs. RHE
j_{OER} (mA/cm^2)
280
260
240
220
200
180
0
20
40
60
80
100
Pressure (bar)
pH = pH^{opt}(T)
pH = pH^0(T)
B
U = 1.51 V vs. RHE
j_{OER} (mA/cm^2)
1000
980
960
940
920
0
20
40
60
80
100
Pressure (bar)
pH = pH^{opt}(T)
pH = pH^0(T)

Figure 10

INTERNATIONAL SEARCH REPORT

International application No.
PCT/IB2022/057592

A. CLASSIFICATION OF SUBJECT MATTER
C01B3/04,C01B3/02 Version=2022.01

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)

C01B

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

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

PatSeer, IPO Internal Database

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	"Microkinetic Model for pH- and Potential-Dependent Oxygen Evolution during Water Splitting on Fe-Doped -NiOOH", Ananth Govind Rajan et al., Energy Environ. Sci., 2020, Published on 13 October 2020, DOI: 10.1039/D0EE02292F. See pages 12-13, Table 1.	1-27
A	"Effect of Temperature on Electrode Kinetic Parameters for Hydrogen and Oxygen Evolutioneactions on Nickel Electrodes in Alkaline Solutions", Miles, M H,J. Electrochem. Soc.; (United States) vol 123:3, N. p., 1976. Web. doi:10.1149/1.2132820. See the whole document.	1-27
A	"Decoupled hydrogen and oxygen evolution by a two-step electrochemical-chemical cycle for efficient overall water splitting", Dotan, H., Landman, A., Sheehan, S.W. et al., Nat Energy 4, 786-795 (2019), Published on 13 September 2019, https://doi.org/10.1038/s41560-019-0462-7. See the whole document.	1-27

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

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26-10-2022	26-10-2022
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INTERNATIONAL SEARCH REPORT

International application No.
PCT/IB2022/057592

C (Continuation). DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	"Electrolyte Engineering towards Efficient Water Splitting at Mild pH", Dr. T. Shinagawa et al., ChemSusChem 2017, 10, 4155 – 4162, DOI: 10.1002/cssc.201701266. See the whole document.	1-27
A	WO 2013160915 A1 (COUNCIL OF SCIENTIFIC AND INDUSTRIAL RESEARCH [IN]) 31 OCTOBER 2013 (31-10-2013) See claim 1.	1-27

INTERNATIONAL SEARCH REPORT
Information on patent family members

International application No.
PCT/IB2022/057592

Citation	Pub.Date	Family	Pub.Date
WO 2013160915 A1	31-10-2013	EP 2841624 A1	04-03-2015
		US 2015136614 A1	21-05-2015