

ON THE EFFICIENCY OF PROTON EXCHANGE MEMBRANE ELECTROLYZER: NUMERICAL ANALYSIS

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ABSTRACT. Researchers have often turned to the search for clean and stable energy systems in recent years. The most promising energy carrier is hydrogen as the best solution for alternative energy. It can be stored in more weight than other fuels in the same volume. Hydrogen is produced in different ways, but the simplest and most promising is water electrolysis. The chemical reaction process that occurs in water electrolysis splits water into oxygen and hydrogen molecules. A proton exchange membrane water electrolysis (PEMWE) system is environmentally friendly and relatively easy to integrate with renewable energy sources such as photovoltaic and wind. The present work focuses on analyzing the effect of different anode nanocatalysts on the performance of PEMWE. For this purpose, the temperature regime, membrane thickness, current carrying length and molar fraction distribution of substances in the gas channels are investigated to observe and compare the effects of different nanocatalysts. The performance of the electrolysis cell is modeled using a point and distributed numerical scheme based on Control Metaphysics software. The ways to increase the hydrogen yield with changes in temperature and membrane thickness are investigated. The results show that the use of thinner membrane is important for the Ni-NiO₂ nanocatalyst compared to the Pt catalyst. The Ni-NiO₂ catalyst reduces the cell voltage and provides higher current density. The maximum value of the molar fraction of hydrogen in the gas channel of the cathode increases by more than 50%. The theoretical and experimental results obtained here can find application in the implementation of industrial technologies in the Tajik Aluminum Company (TALCO) and Yavan Electrochemical Plant (Tajikistan).

Introduction

Over the past decades, there has been a continuous increase in energy consumption due to population growth and improvement in the quality of life of people. Moreover, with the increasing global warming and environmental pollution, the development of renewable energy sources is becoming more and more important. Hydrogen is one of the most promising and stable energy carriers and emits only water as a by-product, without any carbon emissions [1]. Hydrogen with many attractive properties as an energy carrier with high energy density (140 MJ/kg),

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which is more than 2 times higher than typical solid fuels (50 MJ/kg) [2]. Currently, the total global production of hydrogen exceeds 500 billion cubic meters (m³) per year [3]. The produced hydrogen is mainly used in many industrial applications such as fertilizers, oil refining processes, petrochemicals, fuel cells and chemical industry [4]. Hydrogen is produced from various non-renewable and renewable energy sources such as fossil fuels, especially, steam conversion of methane [5], oil/naphtha conversion [6], coal gasification [7], biomass [8], biological sources [9] and water electrolysis (WE) [10].

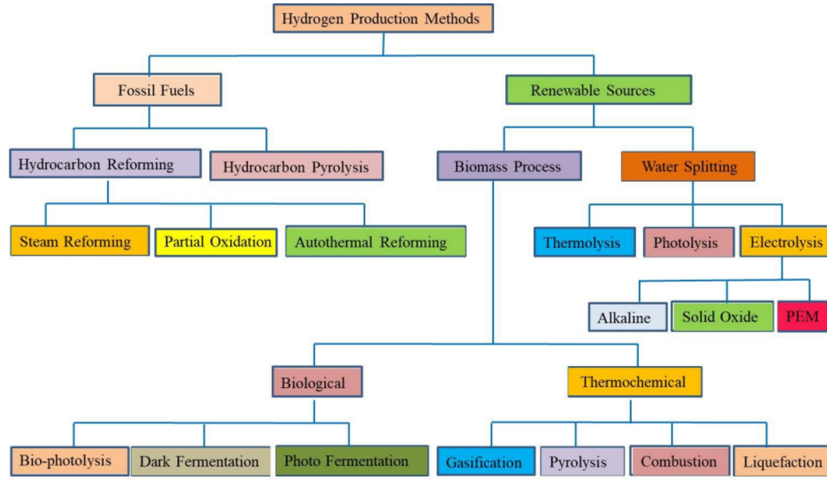


Fig. 1. Hydrogen production methods

The different methods of hydrogen production are summarized in Figure 1. As of January 1, 2025, 96% of global hydrogen production is attributed to non-renewable fuel sources (types), mainly steam conversion of methane. However, the use of fossil fuels produces lower frequency hydrogen with high concentrations of harmful greenhouse gases [11]. At the same time, steadily increasing global energy demands and limited fossil fuel reserves together with detrimental environmental impacts dictate the development of new energy approaches without any carbon emissions. Currently, much attention is being paid to clean energy strategies that could replace the current fossil fuel-based energy production [12]. This can be achieved when hydrogen is produced from water. It is through electrolysis of water that high purity carbon (99.999%) can be produced. The basic reaction of the electrolytic process of hydrogen production is as follows:



However, the efficiency of hydrogen production by water electrolysis remains low to be economically competitive, due to high energy consumption and low hydrogen release rate.

The present work focuses on improving the efficiency of water electrolysis and reducing energy consumption by selecting alternative and inexpensive electrocatalysts. Nickel and nickel oxide nanoscale electrocatalysts will be proposed. The

features of the kinetics and catalysis of the electrolytic reaction will be revealed using numerical analysis methods. The theoretical and experimental results obtained here can find application in the implementation of industrial technologies in Tajik Aluminum Company (TALCO) and Yavan Electrochemical Plant (Tajikistan).

1. Water Electrolysis Technologies

Water electrolysis is a promising method for hydrogen production because it utilizes renewable water (H_2O) and produces only pure oxygen as a byproduct. In addition, the electrolysis process uses direct current (DC) from sustainable energy resources such as solar, wind and biomass power. However, currently, only 4% of hydrogen is produced by water electrolysis due to economic issues [12]. Therefore, further development of renewable energy sources and involvement of new high-tech materials as electrocatalysts is a crucial issue. Water electrolysis has serious advantages such as high efficiency of the electrolytic cell and high rate of hydrogen production with high purity. These advantages further enable the conversion of hydrogen into electrical energy using low temperature fuel cells [13]. In the electrolysis process, the reactant is a water molecule that dissociates into hydrogen H_2 and oxygen O under the influence of electricity. Water electrolysis can be classified according to its electrolyte, operating conditions, and ionic agents.

Thus, there are 4 methods of electrolysis:

1. Electrolysis of alkaline water (AWE) [14];
2. Electrolysis of solid oxides (SOE) [15];
3. Microbial electrolysis cells (MEC) [16];
4. Proton exchange membrane electrolysis (PEM) [12].

We are mainly interested in the fourth method in this paper.

2. Electrocatalysts In PEM Electrolysis Of Water

A review paper [17] found that PEM electrolysis of water utilizes noble metal based electrocatalysts. For example, based on Pt/Pd at the cathode for the hydrogen evolution reaction (HER) and RuO_2/IrO_2 at the anode for the oxygen evolution reaction OER. Water in PEM electrolysis is electrochemically split into hydrogen and oxygen at the respective electrodes, hydrogen at the cathode and oxygen at the anode. PEM electrolysis of water [9] is accomplished by pumping water to the anode where it is split into oxygen (O_2), protons (H^+) and electrons (e^-). The protons move through the proton conducting membrane to the cathode side. The electrons leave the anode through an external force circuit, which provides the driving force (cell voltage) for the reaction. On the cathode side, the protons and electrons recombine to form hydrogen. The corresponding mechanism is shown in Figure 2.

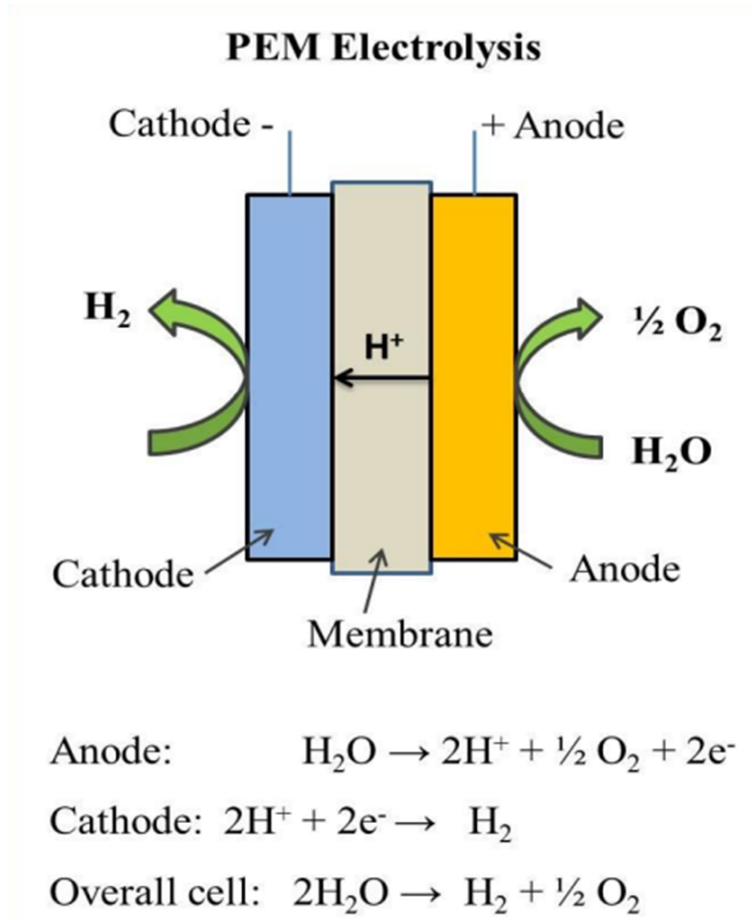


Fig. 2. Schematic illustration of PEM electrolysis of water

Recall that the first article on PEM electrolysis of water was published in 1973 by employees of General Electric (USA), in which the rationale for the use of PEM electrolysis cell. This paper experimentally proved the significant efficiency of the electrochemical reaction and obtained a performance of 1.88 Vat at an operating current of 1A.cm⁻² and 2.44 Vat at 2Acm⁻² with a cell lifetime of 15000 hours, without any deterioration in performance [18]. However, the cost of PEM electrolysis remained very high due to the use of expensive metals. Despite these disadvantages, PEM electrolysis has an advantage over other types of water electrolysis. These are high operating current densities, high gas frequency and high process efficiency. Further, to improve the cost effectiveness of PEM electrolysis, researchers have proposed inexpensive methods and developed various alternative electrocatalysts based on improving the efficiency and stability of inexpensive multimetallic oxides, on reducing the use of expensive metals (Pt, Ir). These electrocatalysts are presented in Figure 3.

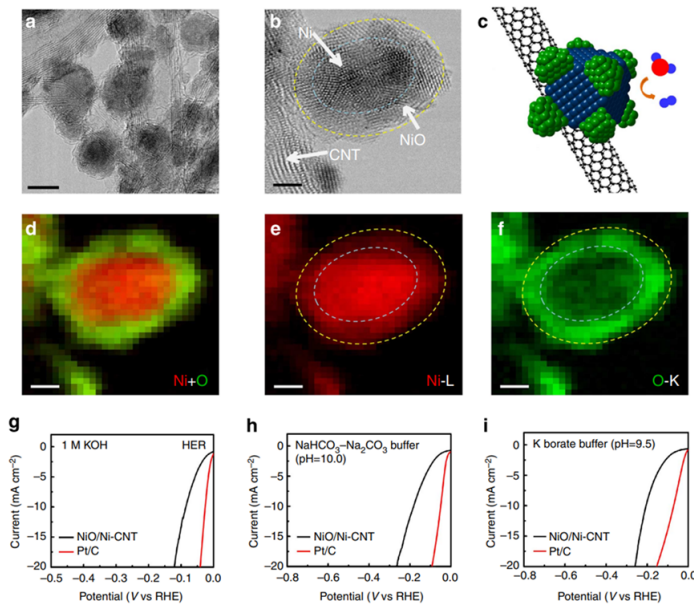


Fig. 3. Structure and performance of a highly active NiO/Ni-CNT nano-hybrid. (a) Low-magnification STEM bright-field image showing the typical morphology of the NiO/Ni-CNT sample, with 8–15 nm particles attached to CNTs. Scale bar, 2 nm. (b) Atomic resolution STEM bright-field image showing the structure of a typical NiO/Ni particle on a CNT (small NiO nanoparticles over a larger Ni nanoparticle core). Scale bar, 2 nm. (c) A schematic illustration of the NiO/Ni-CNT structure. Scale bar, 2 nm. (d–f) Chemical maps for the spatial distribution of Ni and O and their overlay, from the whole area shown in b. The two dashed circles highlight the NiO particle layer on the surface of the Ni core. Scale bar, 2 nm. (g–i) Linear sweep voltammetry of NiO/Ni-CNT and Pt/C in g) 1 M KOH (h) NaHCO₃-Na₂CO₃ buffer (pH=10.0) and (i) potassium borate buffer (pH=9.5) at a scan rate of 1 mVs 1 under the loading of 0.28 mg cm² on RDE showing high HER catalytic activity of NiO/Ni-CNT [18]

2.1. Electrocatalysts for hydrogen separation reaction (HER). The problems associated with the reaction of HER hydrogen evolution in electrolysis have focused on the development of electrocatalysts for the cathode. In most cases, platinum oxides have been used as the standard catalyst because Pt provides excellent HER activity and demonstrates exceptional stability in acidic media. However, such catalysts are very expensive, so most researchers focus on reducing the cost of electrocatalysts, as well as reducing operating costs by increasing the specific productivity and durability of catalysts. In [19], it was noted that along with the search for an alternative to Pt-based electrocatalysts, the formation of dispersed carbon nanoparticles to expand the surface area of the catalyst and thereby reduce the Pt loading is also important. To this end, the electrocatalyst was placed in gas diffusion electrodes based on cost-effective electronic carriers with large surface area, such as carbon nanotubes/soot. Later, other researchers initiated the use of different types of soot (CB) as a substrate material for Pt-based catalyst as a standard electrocatalyst towards HER [18]. In addition, Badwal et al. in [20] reported the development of a PEM water electrolyzer system for high frequency hydrogen and oxygen production. In theirs, the electrolyzer was operated at a pressure of 2 bar at 75–85 °C, and Nafion 112 and 115 membranes were used as Pt/C electrolyte with a loading of 0.4 m²cm⁻². Several researchers studied MoS₂ electrocatalyst for HER [22] Mo₂S mixed with commercial, conductive carbon showed superior catalytic performance towards HER, but only compared to other Mo₂S electrocatalysts, which are still not comparable to Pt-based catalyst.

To further reduce the cost of HER catalysts obtained using other carbon-based electrocatalysts, especially those consisting of only earth-widespread materials and low cost, such as A-Ni-C, MoS₂/CNT doped CoFeS₂/CNT WO₂/C nanofibers and CoFe nanofibers encapsulated in N-doped graphene, etc., have been thoroughly investigated as potential HER electrocatalysts alternative to Pt [21, 22].

In the next section, we will focus on another class of HER electrocatalysts, which we believe to be the most promising. We will talk about nanoscale nickel/nickel oxide heterostructures, which were first introduced in Ref. [23].

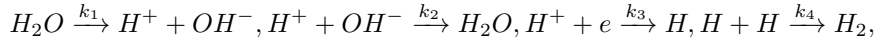
2.2. Nanoscale nickel oxide/nickel heterostructures for HER. We have already noted that an efficient way to produce high-purity hydrogen is the electrochemical splitting of water into hydrogen and oxygen in an electrolyzer. In [25], a group of scientists from Stanford University proposed a new breakthrough idea concerning the problem of electrocatalysts. They reported a heterojunction structure of nickel-nickel oxide (NiO/Ni) type attached to a weakly oxidized carbon nanotube (NiO/Ni-CNT) showing high HER catalytic activity close to commercial Pt/C catalysts in some basic solutions (pH=9.5 /4). NiO/Ni nanohybrids were prepared randomly in a low-pressure thermal annealing experiment, providing partial reduction of nickel hydroxide (Ni(OH)₂) coated with oxidized CNT. They act as a substrate to prevent complete aggregation of nickel. The high catalytic activity of NiO/Ni CNTs towards HER enables a high performance electrolyzer with a current of 20mA cm⁻² at 1.5V.

The synthesis of NiO/Ni-CNTs was carried out by low-temperature hydrolysis of Ni salts in nickel hydroxide on the side walls of weakly oxidized [Ni(OH)₂/ox-CNTs] followed by annealing at low pressure (1.5 Torr) at 300°C in Ar flow. The synthesis is simple and could be easily scaled up for practical applications. Scanning and transmission electron microscopy (STEM) images showed the presence of 10nm nanoparticles attached to the CNT multilayers (Figure 3 a and b). Chemical mapping using electron energy loss spectroscopy (EELS) allowed us to determine the distribution of O, Ni and C with sub-nanometer spatial resolution, identifying a core-shell-like structure with O and Ni in the shell and only Ni in the core (Fig. 3 e, f) above the CNT network. High-resolution STEM imaging (Fig. 3b) and the corresponding chemical mapping (Fig. 3d) showed that the NiO shells are heterogeneous. It contains small NiO grains with different orientations and also contains small gaps in the shell to expose some NiO/Ni nanointerfaces (Fig. 3c).

The catalytic property of HER for NiO/Ni-CNT electrocatalyst was measured in three-electrode configuration with rotating disk electrode (RDE) at 1600 rpm to remove the formed bubbles. H₂. Commercial Pt/C was studied side-by-side at the same loading of 0.28 mgcm⁻². In 1M potassium hydroxide (KOH), the NiO/Ni-CNT catalyst gave a small overvoltage at low current density with only a slightly lower current increase compared to Pt/C and a Tafel slope of 82mV per tenth. (Figure 3g). Impressively, the NiO/Ni-CNT catalyst achieved a current density of 10mAcm⁻² at an overvoltage of <100mV. Moreover, the highly efficient HER catalyst was observed in carbonate/bicarbonate buffer (pH=10.0) and borate buffer (pH=9.5) (Figure 3h, i)

3. Numerical Modeling of Near-Electrode Processes

In [24] it is noted that despite the large number of works devoted to modeling of electrolysis processes, the mathematical models existing at present need to be adapted for each specific electrolyte and electrode composition, since the processes taking place in different melts and solutions with different hydrogen exponent are different. Here, we are interested in the current yield of a substance when analyzing the efficiency of a PEM water electrolyzer. In this case, it seems rational to consider the inverse problem arising in modeling of near-electrode processes. Consider the electrolysis of potassium hydroxide (KOH) solution, which is a strong base, in which K^+ ions are not oxidized at the electrodes, they accumulate in the cathodic space and their concentration decreases in the anodic space over time. The following stage of electrochemical reactions takes place at the cathode



where k_1, k_2, k_3, k_4 – rate constants of the corresponding processes. Water dissociation reaction $H_2O \rightleftharpoons H^+ + OH^-$ is heterogeneous, and hence depends on the water concentration surfaces defined by the electrode area.

The electrochemical processes occurring during PEM electrolysis of water can be described by a system of kinetic equations in point form. Let us introduce the following notations

$$\theta_1 = C_{H_2O}, \theta_2 = C_{OH^-}, \theta_3 = C_{H^+}, \theta_4 = C_H, \theta_5 = C_{H_2},$$

where $C_{H_2O}, C_{H^+}, C_{OH^-}, C_H, C_{H_2}$ – concentrations of water, hydrogen ions, hydroxyl group, hydrogen atom and hydrogen molecules, respectively. Taking into account the introduced notations, we obtain the following kinetic system

$$\begin{cases} \frac{\partial \theta_1}{\partial t} = -k_1 \theta_1 + k_2 \theta_2 \theta_3, \\ \frac{\partial \theta_2}{\partial t} = k_1 \theta_1 - k_2 \theta_2 \theta_3, \\ \frac{\partial \theta_3}{\partial t} = k_1 \theta_1 - k_2 \theta_2 \theta_3 - k_3 \theta_3, \\ \frac{\partial \theta_4}{\partial t} = k_3 \theta_3 - k_4 \theta_4^2, \\ \frac{\partial \theta_5}{\partial t} = k_4 \theta_4^2, \end{cases} \quad (2)$$

where by the rate constants k_i , $i = 1, 2, 3, 4, 5$ and concentration values are subject to the following restrictions:

$$k_1 \geq 0, k_2 \geq 0, k_3 \geq 0, k_4 \geq 0, k_5 \geq 0; \quad (3)$$

$$\theta_1 \geq 0, \theta_2 \geq 0, \theta_3 \geq 0, \theta_4 \geq 0, \theta_5 \geq 0. \quad (4)$$

For the system (2) we set the Cauchy problem with the following initial conditions

$$\theta_1(0) = \theta_{10}, \theta_2(0) = \theta_{20}, \theta_3(0) = \theta_{30}, \theta_4(0) = \theta_{40}, \theta_5(0) = \theta_{50}. \quad (5)$$

Here $Q_{10}, Q_{20}, Q_{30}, Q_{40}, Q_{50}$ – constants corresponding to the initial data of the simulated experiment and calculated from the percentage electrolyte concentration, ionic product of water, and hydrogen solubility coefficient.

At known values of rate constants of cathode processes k_1, k_2, k_3, k_4, k_5 the hydrogen prediction problem is not particularly difficult, and it is solved by the Runge-Kutta method in [24]. In addition, in [24] the inverse problem is solved when the rate constants are unknown and the concentration of molecular hydrogen at the outlet at certain time moments is given. To solve the problem, the functional is minimized

$$M(\theta_{5,1}, \dots, \theta_{5,n}) = \sum_{i=1}^n (\theta_{5 \text{ exper.}, i} - \theta_{5,i}),$$

where $\theta_{5 \text{ exper.}, i}$ – experimental value of hydrogen concentration at the moment of time t_i , $i = 1, \dots, n$, $\theta_{5,i}$ – calculated hydrogen concentration, at the same moment of time given as a function of reaction rates k_1, k_2, k_3, k_4, k_5 .

In [24], the maximum of the functional is searched using a modified Hooke-Jeeves method with constraint checking (3), (4). In this case, the calculation of the concentration $\theta_{5,1}$ as functions depending on the rates of stage reactions, is obtained by solving system (2) with initial condition (5) by Runge-Kutta method for the system of fourth-order equations.

To study different kinds of spatiotemporal structures, a diffusion term describing the diffusion of adsorbed particles over the catalyst surface should be added to the point model (2). Surface diffusion differs from ordinary diffusion in that it strongly depends on the presence of empty adsorption sites, since it is carried out by "jumping" of an adsorbed molecule or atom to a neighboring free site.

The distributed mathematical model of the reaction under consideration is a system of five quasilinear parabolic equations describing changes in surface concentrations

$$[H_2O], [OH], [H], [H_2], [H+]$$

and has the form

$$\frac{\partial \theta_i(x, t)}{\partial t} = D_i \left(z \frac{\partial^2 \theta_i}{\partial x^2} + \theta_i \sum_{j=1}^5 \frac{\partial^2 \theta_j}{\partial x^2} \right) + f_i, \quad i = \overline{1, 5}, \quad (5)$$

where f_i right-hand sides of the system (4), D_i , $i = \overline{1, 5}$ - diffusion coefficients of adsorption particles

$$[H_2O], [OH], [H], [H_2], [H+], z = 1 - \theta_1 - \theta_2 - \theta_3 - \theta_4.$$

If the catalyst surface is not strongly filled with adsorbed particles, the usual diagonal diffusion matrix can be used to describe diffusion processes over the surface:

$$\frac{\partial \theta_i(x, t)}{\partial t} = D_i \left(z \frac{\partial^2 \theta_i}{\partial x^2} \right) + f_i, \quad i = \overline{1, 5}. \quad (6)$$

In this case models (5) and (6) give close results.

The solutions of the system (5) are considered either on the segment $0 \leq x \leq 1$, or, on an infinite line, or in some region in the plane, for example, in a rectangle, then x is a vector, and differentiation is given for each component (x_1, x_2) . The system of equations (5) is supplemented with boundary conditions and initial distributions of concentrations of adsorbed substances. At the ends of the segment either periodic conditions or Neumann conditions are set.

The mixed problem thus obtained represents a mathematical model of the mesolevel and can describe the formation, evolution, and interaction of various spatial and temporal structures arising on the catalyst surface during the reaction.

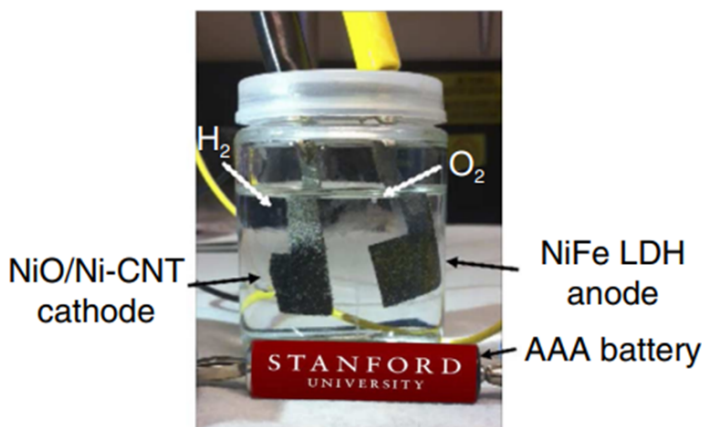


Fig. 2. Demonstration of a water splitting device operating on AAA batteries with a nominal voltage of 1.5 volts

4. Conclusion

This paper analyzes the development of PEM electrolysis of water, including the hydrogen generation reaction and the advantage of $Ni/NiO_2 - CNT$ -type nanoelectrocatalysts over other materials.

A method of numerical optimization of chemical kinetics problems has been implemented, which allows to find the rates of constants of electrode processes following the given experimental data on yield, as well as to calculate the concentration of substances participating in the electrode processes at specific moments.

Future research directions will be related to the improvement of PEM water electrolyzers based on renewable energy sources. This approach, in our opinion, will be cost-effective.

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