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Effect of pulse potential on alkaline water electrolysis performance

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ABSTRACT

In this study, the effect of pulse potential on alkaline water electrolysis energy consumption is investigated. A specially designed electrical circuit is used to observe the effect of different duty cycles and frequency values on water electrolysis energy consumption in different concentration values of alkaline solution. The results show that using pulse potential enhances the mass transport of oxygen and hydrogen bubbles due to the pumping effect. This provides less contact with oxygen bubbles to improve corrosion resistance of anode electrodes. Moreover, decreasing mass transfer losses on the electrode surface resulted in a 20–25% lower energy consumption to produce 1 mol of hydrogen in the cell. The optimum frequency for 10% and 50% duty cycle and 10% and 15% concentration are investigated. For 10% duty cycle, the optimum frequency is specified around 140–200 kHz and for 50% duty cycle, it is around 380–400 kHz for all concentration values.

Introduction

Increasing demand of fossil fuels and their finite amount on Earth attract researchers' attention to hydrogen energy. Hydrogen has a great potential as an energy carrier but is widely found in compound form within nature. For this reason, these compounds need some energy to separate them into hydrogen and other molecules. Thermodynamically, energy consumption for this separation process is always higher than obtained energy from hydrogen molecules. Thus, it is crucial to produce hydrogen efficiently for the sustainable hydrogen economy [1,2].

Water electrolysis is one of the easiest method to produce hydrogen, especially since the demand for clean and renewable energy systems is increasing [3–5]. Extensive use of water electrolysis has some challenges regarding minimization of energy consumption, electrode stability and reliability. Particularly, energy consumption costs due to inefficient hydrogen production is one of the biggest problems to overcome. Water electrolysis efficiency is around 56–73% which is not ideal for the economical production of hydrogen [6,7]. Electrolysis reactions take place when DC voltage is higher than almost 1.6 V which is sum of theoretical voltage 1.23 and electrode overpotentials to continue reactions. In DC electrolysis, current flow in aqueous solution is limited by the diffusion coefficient of the species. Especially bubble coverage of electrode is a big problem for the mass transfer losses. In water electrolysis, hydrogen molecules produce in cathode side and the oxygen molecules produce at the anode side. These molecules in the bubble form, stick to the electrode surface until the equilibrium forces that cause surface separation and movement in the electrolyte. High saturation of dissolved gas in the electrolyte implies that bubbles are less likely to form autogenous within the aggregation. Equilibrium condition between electrode surface and the movement of species in the electrolyte cause mass transfer limitations. For this reason, DC electrolysis is reaction rate limited by the

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diffusion of ions via electrolyte and the diffusion layer near the surface of each electrode [8,9]. For this reason, improving electrolysis efficiency is difficult without increasing input power to obtain the same volume of hydrogen [10]. Several researchers used different external methods to improve electrolysis performance, such as magnetic fields [11–15], super gravity fields [16] and ultrasonic waves [17]. For these methods, an external DC source is used as the current source. On the other hand, changing the nature of the DC source with different pulse voltage waveforms is an alternative effective method. Because of related mass transfer limits in the electrochemical systems, lowering these losses due to the species transport in the electrolyte may be improved with the help of pulsed hydrogen production method. It is akin to a dielectric relaxation on the electrode- electrolyte interface that impacts charge double layer dynamics [18]. Relaxation of electrode surface improves the reactions rate on the electrodes due to the acceleration of gas bubbles.

Several researchers are focused on the effect of fluctuation of DC sources on different water electrolysis systems with modeling and experimental studies. Ursua et al. [19] investigated different predicted electrical input response to the water electrolysis with modeling approach. They used different source of capacitance and impedance between two electrodes. They predicted the response of electrolyser to an electrical profile input. Similar experiments conducted by Mazloomi and Sulaiman [20] with a representation of electrolysis cell by parallel positioned capacitance to a resistance. They used a Pulse Width Modulation (PWM) control circuit to deliver complicated current waveforms response of the electrolysis cell. They concluded that this circuit reached resonance frequency and lower impedance value rely on the distance of the electrodes and molar concentration of the electrolyte. As an experimental study, Shimizu et al. [2] firstly claimed the application of short pulses to the water electrolysis. They used 300 ns pulse from 2.2 V to 12.6 V potential range. Their aim was the solution of diffusion limitations on the cell with several pulse periods. Similarly, Vanags et al. [21] increased pulse time to around 500 ns until 600 V. They mentioned that higher voltage pulse provides more electron pumping into the electrolyte. They defined the electrolyte as a capacitance, and the performance improvement is a result of this capacitance behaviour. Dobo et al. [22] investigated voltage fluctuation of the power supply to observe performance change of an alkaline water electrolysis. In the 50 kHz and ±10 V with 8A maximum current value of power, the supply hydrogen production and energy consumption may be improved. However, according to our knowledge, there is no detailed study for different concentration and 0–1200 kHz frequency in the hydrogen production and energy consumption approach.

In this study, investigation on hydrogen production in an electrolytic cell is made. It was found that the hydrogen production in the cell decreased remarkably by specific optimal pulse potential and frequency of water electrolysis. In DC and different pulse potential electrolysis cell, the same amount of hydrogen is accumulated, and their energy consumption values are measured separately. When pulse potential is applied in different duty cycles, a higher current value is obtained for the same voltage. This study shows that less power is needed when water electrolysis is performed under pulse potential. In pulse potential applied cell, the energy consumption amount is achieved is up to 25% less with the optimal duty cycle and frequency.

Experimental system

A schematic view of experimental setup is shown in Fig. 1, 200 ml volume of KOH solution bath is used as electrolysis cell. Similar experimental setup is used by Kaya et al. [15] to investigate magnetic field effects on water electrolysis.

In alkaline water electrolysis, oxidation reaction (OR) takes place at the anode side to produce oxygen gas:

$$2OH^- \rightarrow \frac{1}{2}O_2 + H_2O + 2e^- \quad E_a^0 = +0.401(V) \tag{1}$$

Reduction reaction (RR) takes place at the cathode side:

$$2H_2O + 2e^- \rightarrow H_2 + 2OH^- \quad E_c^0 = -0.82806(V) \tag{2}$$

Total reaction:

$$H_2O \rightarrow H_2 + \frac{1}{2}O_2 \quad E^0 = -1.229(V) \tag{3}$$

Cylindrical Pt electrodes are used as cathode and anode electrodes. The distance between electrodes is fixed to 10 mm. Electrode surface areas for the Pt electrodes are 31.4 mm^2 and 200 ml volume, handmade, polymeric based and alkali resistant solution bath is used as electrochemical cell. In DC mode, the current density values for 10% and 15% concentrations reach up to 3.7 A/cm^2. However, in the pulsed mode the current density value for the same concentrations can reach around 4.04 A/cm^2. For all experiments, electrolyte concentration is selected as 10 wt% and 15 wt%. The temperature is selected as room temperature for all experiments. Produced hydrogen gases are accumulated in accumulation glass tubes to measure the hydrogen flow rate. In Fig. 2, a specially designed electrical circuit can be seen for the square type pulses voltage generation.

In this circuit, a MOSFET driver is used with an external function generator to adjust the duty cycle from 10% to 100%. The output frequency of the signal generator is changed between 0.1 Hz and 1.2 MHz. Lower side switching topology [9,23] is used to apply pulsed power to the electrolyser. In Fig. 3, the desired characteristic of the square type electrical pulse can be seen. A power supply is used as the current source. For DC experiments, voltammeter and ammeter are used to measure the voltage and current, respectively.

Because of the high frequency, calculating energy consumption is not possible only with voltammeter and ammeter. Thus, an oscilloscope is used to measure different duty cycle condition, consumed energy is calculated by Eqs. (4) and (5) [24].

$$V_{RMS} = \sqrt{\frac{1}{T}\int_0^T V(t)^2 dt} \tag{4}$$

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