The efficiency of hydrogen production by water electrolysis with bitter melon as a media using stainless steel/Fe-Co-Ni electrodes

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Abstract. This study aims to determine the production efficiency of hydrogen gas with the addition of bitter melon using stainless steel/Fe-Co-Ni electrodes on water electrolysis process and also to obtain the smallest required energy of hydrogen gas production. Stainless steel/Fe-Co-Ni was made from electrodeposition stainless steel by using e-DAQ E-Chem instrument at a rate of 50 mV/s for 10 minutes. Electrodeposition result was characterized using linear voltammetry, SEM-EDX (Scanning Electron Microscope-Energy Dispersive X-Ray) and XRD (X-Ray Diffraction). Stainless steel/Fe-Co-Ni electrodes was used as working electrodes on water electrolysis. The process of water electrolysis used 0.5 % NaHCO₃ solution and an additional amount of bitter melon (0.1-1 %w/v). Each of the samples was electrolyzed in a cyclic voltammetry by using e-DAQ E-Chem instrument at a rate of 50 mV/s. The study results showed that the addition of bitter melon in electrolyte solution is less efficient to produce the hydrogen gas. But, related to the energy, the smallest required energy of hydrogen gas production by water electrolysis with bitter melon as a media using stainless steel/Fe-Co-Ni electrodes occurs at the addition of 0.6 % (w/v) bitter melon flour, which is 0.001 Volt with the efficiency of hydrogen gas generation was 51.06 %. Thus, we can state that the optimum efficiency of hydrogen gas production occurs at the addition of 0.6 % bitter melon flour in 0.5 % NaHCO₃ solution.

Keywords: bitter melon, electrolysis, hydrogen production, stainless steel/Fe-Co-Ni electrode.

1. Introduction
Electrodeposition is the process of metal deposition on the others metal (electrode) through electrolysis process using electrochemical principle by applying an electric current. The electrodeposition process is divided into two methods [1], i.e. electroplating and electrolysis. Electroplating process uses electrode as a coating material, while electrolysis process uses electrolyte as a coating material. Electroplating system consists of an external circuit, a negative electrode (cathode), electrolyte solution, and a positive electrode (anode).

\[ M_\text{(s)} \rightarrow Mn^{n+} + ne \]  \hspace{1cm} (1)

The metal will be oxidized into metal ions and release some of the electrons. The positive ions in the electrolyte solution will move to the cathode, while the negative ions will move to the anode. The positive ions will join with the electrons to form metal deposits at the cathode [2]. The equation of the reduction reaction will be as follow [3]:

\[ Mn^{n+} + ne \rightarrow M_\text{(s)} \]  \hspace{1cm} (2)
According to Salehi et al. [4], overall deposition of metal is influenced by hydrogen evolution reactions. Cations will react with hydroxyl ions to form mono-hydroxides. The mono-hydroxide is absorbed by the cathode surface and forms a neutral metal and also releases the hydroxyl ions. The deposited metal will decrease protons but increase the hydroxyl ions. The overall reactions are followed by equations (3), equation (4), equation (5) and equation (6).

\[
2\text{H}_2\text{O}_\text{aq} + 2e^- \rightarrow \text{H}_2 + 2\text{OH}_\text{aq} \quad (3)
\]

\[
\text{M}_\text{aq}^+ + \text{OH}_\text{aq}^- \rightarrow \text{M(OH)}_\text{aq} \quad (4)
\]

\[
\text{M(OH)}_\text{aq}^- \rightarrow \text{M(OH)}_\text{aq}^\text{+} \quad (5)
\]

\[
\text{M(OH)}_\text{aq}^+ + 2e^- \rightarrow \text{M} + \text{OH}_\text{aq}^- \quad (6)
\]

Hydrogen gas can chemically be produced by water electrolysis method. According to Chang [5], electrolysis is a process using electrical energy in order to make nonspontaneous chemical reactions. Electrolysis method is a simple method so that the production of hydrogen gas can be conducted easily. The process of electrolysis to produce pure H and O gas by utilizing electrical energy in the system is called as water electrolysis. The gas produced from the water electrolysis process is called HHO gas or oxyhydrogen or also called Brown’s Gas. Electrolysis of H\text{O} is a breakdown of the water element (H\text{O}) by applies DC electric current to breakdown water into HHO gas (Brown’s gas) [6]. Since the atoms of water release their electrons while the oxygen atoms get the electrons, the oxygen atom becomes negative (O) and the hydrogen atom becomes positive (H). These hydrogen atoms combine into H\text{g} gas through the cathode to form bubbles. It also occurs in the O ion which then converges into O.

The reaction occurred in the electrode on electrolysis process of water is as equations (7) and equation (8) [7]:

\[
\text{Cathode : } 2\text{H}_2\text{O}_\text{aq} + 2e^- \rightarrow 2\text{OH}_\text{aq}^- + \text{H}_2 \quad (7)
\]

\[
\text{Anode : } 4\text{OH}_\text{aq}^- \rightarrow \text{O}_2 + 2\text{H}_2\text{O}_\text{aq} + 4e^- \quad (8)
\]

The process of electrolysis depends on the type of electrolyte, the type of electrode, current, voltage, and reaction time [9]. Variations of current and more addition of catalysts may effects on the production of hydrogen gas in the water electrolysis process. The bigger of the current occupied, the production of hydrogen gas will also get bigger. According to the first Faraday’s Laws, it states that a number of the substances formed on each electrode are directly proportional to the amount of electrical current flowing of electrolysis [2]. In the room temperature, water separation is very small approximately 10 mol/L. This is due to pure water is a very poor conductor of electricity. Therefore, acid or base is used to improve the conductivity [8]. A substance that a change the rate of a chemical reaction but it is still on the previous form at the end of the reaction is called a catalyst. Thus, the catalyst can accelerate or slow down a chemical reaction. The addition of a catalyst in the electrolyte solution will decrease the resistant of the electrolysis solution. So that, it uses low power consumption [7]. The amount of energy consumption in the electrolysis process causes the uneconomic production of hydrogen. Therefore, extra treatment is required to increase the production of hydrogen gas. Related to this, a media of bitter melon is added in the process of water electrolysis. Bitter melon is an abundant fruit and its price is affordable. According to the research journals by Maysarrah and Isana [10], the addition of 1 g of cornstarch can provide energy efficiency in water electrolysis with stainless steel/Fe-Co-Ni electrodes. Alkene cornstarch, the use of bitter melon in the electrolyte solution is expected to increase the efficiency of hydrogen gas production by water electrolysis.

2. Materials and methods

2.1. Materials and instruments
Stainless steel electrode type S-430 with a thickness 1.2 mm, width 3 mm, and long of 110 mm, FeSO\text{.7H}_2\text{O}, Co(NO\text{.)}_\text{6H}_\text{2O}, NiSO\text{.6H}_\text{20}, HBO\text{., saccharin, NaCl, NH\text{Cl, NaHCO}_3, platinum, Ag/AgCl, bitter melon, distilled water. Chemicals and solvents were obtained commercially. The grade of them is pure analysis (p.a.) and used without further purification. Linear voltammetry (e-DAQ E-Chem), SEM-EDX, and XRD, and glassware laboratory.
Table 1. The concentration variable of bitter melon media in 0.5% NaHCO₃ solution

| Concentration of Media % (w/v) | Sample Number | Concentration of Media % (w/v) | Sample Number |
|-------------------------------|---------------|-------------------------------|---------------|
| 0                             | 1             | 0.6                           | 7             |
| 0.1                           | 2             | 0.7                           | 8             |
| 0.2                           | 3             | 0.8                           | 9             |
| 0.3                           | 4             | 0.9                           | 10            |
| 0.4                           | 5             | 1                             | 11            |
| 0.5                           | 6             |                               |               |

2.2. Electrode preparation
The stainless steel electrode was coated with Fe, Co, and Ni ions (ratio 1:1:1) by electrodeposition using e-DAQ E-Chem at a rate of 50 mV/s for 10 minutes. The electrolytic deposition process uses stainless steel as the active electrode, platinum as a passive electrode, and Ag/AgCl as a reference electrode. In the electrolyte solution, it was added 1.5 g H₃BO₃, 0.1 g saccharin, 2.0 g NaCl, 2.0 g NH₄Cl per liter of distilled water to facilitate Fe, Ni, Co deposited on stainless steel. The stainless steel and stainless steel/Fe-Co-Ni electrodes are then characterized by linear voltammetry, XRD (X-Ray Diffraction), and SEM-EDX (Scanning Electron Microscope-Energy Dispersive X-Ray).

2.3. Electrolyte solution preparation
To make bitter melon flour, bitter melon is peeled, removed the seeds, washed, sliced and dried in the sun to dry, then milled and sifted. Further, the electrolyte solution was prepared by adding 5 g of NaHCO₃ and bitter melon flour as a mediator with a mass variation (1, 2, 3, 4, 5, 6, 7, 8, 9, 10 g) in 1 L of distilled water.

2.4. Electrolysis process
Electrolysis takes place with 5 g of NaHCO₃ per liter of distilled water (see table 1). The electrolysis of water was carried out by cyclic voltammetry at a rate of 50 mV/S with an initial potential of -1000 mV and a final potential of 1000 mV. The stainless steel/Fe-Co-Ni electrode was used as an active electrode, platinum as a passive electrode, and Ag/AgCl as a reference electrode.

2.5. Voltammetry characterization

2.5.1. Determination of the production efficiency of hydrogen gas. The production efficiency of hydrogen gas (ε) can be determined by comparing the cathodic current peak (Ic) of the voltammogram between the sample using and without the bitter melon flour.

2.5.2. Determination of overpotential value. The overpotential value (ΔE) is determined by reducing the potential value when the reaction takes place with a potential value of half the hydrogen reaction at the standard state (E° H₂O/H₂ = -0.828 V). The optimum condition of hydrogen gas production can be known by the optimum efficiency value of hydrogen gas production and the smallest required energy (over potential).

3. Results and discussion
Electrolysis used for metal deposition on electrode surfaces is known as electrodeposition or electroplating [2]. Electrodeposition is a process of deposition using electrochemical principle, i.e. ions can be decomposed by giving potential to the electrode [11]. According to Faraday's Law, the amount of metal attached to the electrode is directly proportional to the current flowing through the cell. The success of the electrodeposition process using cyclic voltammetry can be seen by the reduction current associated with the reduction process of metal ions to be neutral metals [12]. The amount of the reduction is directly proportional to the concentration of the reduced compounds by assuming that the electrode surface area of the diffusion and non-diffusion coefficients constant.
Deposited metal is based on the standard potential value. The more positive standard potential value of a metal, the more unreactive the metal [13]. The less reactive metal means that it is difficult to release the electrons so that it will have strong oxidizing agents and have reduction reaction. There are two competitions of metal ions in the electrodeposition process to form a binary metal. Metals with a more positive standard potential value will be deposited first [14]. The electrode plays as a conductor of the electric current, flowing from a voltage source into the electrolyte solution. In the electrolysis cell, the cation flow into the cathode to be reduced, while the anions flow into the anode to be oxidized [15]. During the electrodeposition process, the metal was oxidized to form a cations metal and release electrons. Further, metal cations will be pushed towards the cathode, which is then deposited on the surface of the cathode.

In linear voltamograms of stainless steel before coating (figure 1a) and linear voltamograms of stainless steel after coating (figure 1b), it can be seen differences. At linear voltammogram of stainless steel after coating appears the peak. It shows that a current increase or that a metal oxidation reaction has occurred. Moreover, the oxidized metal will be deposited on the cathode. Stainless steel/Fe-Co-Ni produced by electrodeposition was characterized using linear voltammetry, XRD, and SEM-EDX.

XRD profile of stainless steel electrode and stainless steel/Fe-Co-Ni electrode in figure 2 showed the difference of a number of peaks. XRD profile of stainless steel appears three peaks, while XRD profile of stainless steel/Fe-Co-Ni appears four peaks. It indicates that the stainless steel electrode has been successfully coated. The stainless steel and stainless steel/Fe-Co-Ni electrodes are further
**Table 2.** The composition of stainless steel and stainless steel/Fe-Co-Ni by SEM-EDX.

| Metal | Stainless steel | Stainless Steel/Fe-Co-Ni |
|-------|----------------|--------------------------|
| Fe    | 80.11          | 50.34                    |
| Co    | 0.05           | 0.14                     |
| Ni    | 0              | 0.17                     |

**Figure 3.** EDX profile of (a) stainless steel and (b) stainless steel/Fe-Co-Ni.

Characterized using SEM-EDX. SEM equipped EDX (SEM-EDX) is used to analyze the surface morphology and composition of the compound [6]. EDX profile of the stainless steel and stainless steel/Fe-Co-Ni electrodes can be seen in figure 3 and figure 4. Clearly, composition of stainless steel and stainless steel/Fe-Co-Ni can be seen in table 2. It shows that the coating on the stainless steel electrode is successfully performed which is marked by the appearance of nickel metal with a content of 0.17%.

Based on figure 4, the highest efficiency of hydrogen production in the water electrolysis process using stainless steel/Fe-Co-Ni electrodes is sample 1, i.e. no addition bitter melon flour with a current of -0.398 mA. The amount of current produced by the water electrolysis is directly proportional to the amount of hydrogen gas produced. According with the first Faraday’s Laws, it states that a number of the substances formed on each electrode is directly proportional to the amount of current strength/electrical current flowing during electrolysis. However, the process of water electrolysis without media addition in sample 1 requires some energy because the result of overpotential is 0.08 Volt. Overpotential is directly proportional to the energy required. The smaller overpotential value formed in the process of producing hydrogen gas, the less energy required in the process of producing the hydrogen gas. Refers to observing aspects of energy needed, the addition of 0.6 % of media bitter melon requires the smallest energy, which is 0.001 Volt with the efficiency of hydrogen gas generation was 51.06 %. Thus, we can state that it is the optimum condition especially in terms of efficiency of hydrogen production in water electrolysis (see table 3).
Table 3. The optimum condition of electrolysis process using stainless steel/Fe-Co-Ni electrode

| Matter                        | optimum condition stainless steel/Fe-Co-Ni |
|-------------------------------|--------------------------------------------|
| Concentration of media (%)   | 0.6                                        |
| H₂ production efficiency (%) | 51.06                                      |
| Over-potential (V)            | 0.001                                      |

Figure 4. The efficiency and overpotential of electrolysis using stainless steel/Fe-Co-Ni electrode.

The addition of media bitter melon actually inhibits the water electrolysis process. This is shown by the production of hydrogen gas without the addition of media bitter melon which is bigger when compared to the production of hydrogen gas. The less production of hydrogen gas was 38.32 % with a current of -0.1525 mA. It is shown on sample 3 with the addition of 0.2 % of media bitter melon. The process of water electrolysis becomes obstructed due to covering the electrode surface. It makes the activity of the electrode surface is reduced so that the production of hydrogen gas is also reduced.

4. Conclusions
Water electrolysis using stainless steel/Fe-Co-Ni electrode is relative high efficient than stainless steel, but the addition of bitter melon in electrolyte solution is less efficient to produce the hydrogen gas. By observing aspects of energy needed, the addition of 0.6 % of media bitter melon requires the smallest energy, which is 1 mV with the efficiency of hydrogen gas generation, was 51.06 %. Thus, we can state that the optimum efficiency of hydrogen gas production occurs at the addition of 0.6 % bitter melon flour in 0.5 % NaHCO₃ solution.

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