ELECTROLYTE OPTIMIZATION ON DRY CELL GENERATOR ELECTROLYSIS SYSTEM FOR PRODUCING HYDROGEN GAS USING RSM METHOD (RESPONSE SURFACE METHOD)

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**ABSTRACT**

This study aims to determine the maximum concentration of hydrogen gas produced through the electrolysis process of dry cell generators. This research using experimental generator apparatus with Sensor-MQ-8 Arduino and optimizing using Response Surface Method with Design Expert 6. To produce hydrogen gas is done through the method of water electrolysis by decomposing the molecule H₂O into hydrogen gas and oxygen gas with the help of direct electric current. Hydrogen gas productivity by electrolysis method applied to DC generators using 4/4 plate electrodes (Cu/Al) as cathodes and NaNO₃ solutions as electrolytes. The current and voltage used in this electrolysis process is 0.6 ampere and 2 volts for 1 hour. The optimum condition of hydrogen gas concentration obtained is at NaNO₃ 1 M concentration and 60 minutes with hydrogen concentration produced as much as 143.393 ppm. The verification result value for hydrogen gas concentration is 144 ppm.

**Keywords**: Hydrogen, DC Generator, Electrolysis, Electrolyte, RSM

**INTRODUCTION**

Hydrogen is a clean energy source for the environment.¹⁻² Hydrogen energy is the solution to climate change which is now an obstacle for mankind on earth. Carbon emissions from fossils, trigger the need for strategic solutions for the availability of human energy today.³ One of them is to strive for the production of hydrogen gas through chemical and electrical processes, so that hydrogen energy can be used by humans throughout the world.⁴ Hydrogen is rare on the Earth's surface (0.14%), and it is found only in small amounts (0.07%) in the atmosphere. Although very rare, the presence of hydrogen has been found in larger quantities in some wells containing nitrogen.⁵ Small amounts of hydrogen may exist found in mixtures with natural gas in parts of the Earth's crust. The idea of combining clean renewable energy sources such as solar energy and hydrogen, like energy carriers using almost endless resources such as water have been explored by some researchers before.⁶ Technology of various scales has been developed to produce hydrogen as an alternative energy carrier, widely used as a chemical raw material in the industry. The use of hydrogen on


http://dx.doi.org/10.31788/RJC.2022.1516632

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a large scale as fuel will lead to a multi-fold increase in production capacity. Input energy for hydrogen production is always greater than the energy output of hydrogen. As a result, energy, as well as feedstock, will be required to obtain hydrogen.\textsuperscript{7,8} Given the high level of energy consumption in the world that harms the environment as a result of the effects of fossil-based energy sources, so renewable energy sources are ideal for the production of sustainable hydrogen.\textsuperscript{9}

Compared to the fossil fuels that we commonly use during this time, namely gasoline and diesel, the use of hydrogen as a fuel is much more effective in combustion. In comparison 1 kg of gasoline burned at 25°C and pressure 1 atm will produce heat between 44.5 kJ/kg to 47.5 kJ/kg, while 1 kg of diesel can produce heat between 42.5 kJ/kg to 44.8 kJ/kg. Hydrogen itself is in the same condition (25°C and pressure 1 atm) of the same weight is capable of generate heat of 119.93 kJ/kg to 141.86 kJ/kg, which means almost 3 times the heat that can be generated by gasoline and diesel combustion.\textsuperscript{11-12}

Various previous studies have succeeded in producing hydrogen. Hydrogen can be produced by electrical systems, or by electrolysis.\textsuperscript{13-14} Hydrogen can also be produced biologically, using fermentation, bacterial and microbial methods.\textsuperscript{15} The raw material for hydrogen production can also come from nature and artificially. One method of hydrogen production is by using water electrolysis. Both the implementation of the concentration of anions and cations in water as an integral electrolyte system, as well as a modified generator design. In the dry-cell generator design system, efforts can be made to increase the cathode and anode layers, and the thickness of the cathode and anode plates used. In the electrolyte system, the anions and cations used can also be optimized based on their type and concentration.

Electrolysis cells are the most widely used method for hydrogen gas production, which is the method of electricity use in containers containing water. The breakdown of water molecules to be ionized requires a large amount of energy, which is about 4% of hydrogen can be obtained if using ordinary electrolysis. Therefore, an electrolyte is needed as a break-up of water molecules.\textsuperscript{16-17} The design of dry cell generator reactors uses electric current to disperse hydrogen and oxygen gases in water molecules. This research aims to determine the optimum concentration of hydrogen gas concentration produced using the Design Expert 6 Response Surface Methodology (RSM) program.

**EXPERIMENTAL**

**Material and Methods**
The tools and materials used in this study were Arduino uno, Power Supply, Aluminum (0.7 mm thick), copper (0.4 mm thick), acrylic, socket, bolt 13, saw, drill, tube, gasket (2 mm), hose, MQ-8 sensor, Glassware, NaNO\textsubscript{3}(MERCK), and distilled water.

**Electrode Preparation**
Copper plate (0.4 mm) and aluminum plate (0.7 mm) in the form of sheets cut with a width of 10 cm and a length of 10 cm as much as 8 sheets and arranged sandwiches as shown in Fig.-1. Then the electrodes are limited by gasket and set with a plate count of 4/4 (Cu/Al).
Detection Method
Tools in the form of reactors, electrodes, sensors MQ-8, Arduino Uno, and PC arranged like Fig.-2. Then, the power supply is already paired with plug cables and cables with clamps. Next, prepare a solution of NaNO₃ with various concentrations alternately (0.25 M, 0.5 M, 0.75 M, and 1 M). Prepare the MQ-8 sensor as a measurement of hydrogen gas concentration. Connect the hose from the reactor to the sensor detector.

Furthermore, the tube that already contains electrolyte solution is given DC electric current to react the solution so that water decomposition becomes hydrogen and oxygen gas. Measure for 1 hour, it will be able to final concentration than the reading of Arduino uno software, doing on each variation of concentration and number of plates to be tested. The following Fig.-2 shows Hydrogen gas measurement scheme, and Fig.-3 displays Hydrogen gas measurement flow chart.

RESULTS AND DISCUSSION
When measuring hydrogen concentration (Fig.-2), it must be ensured that the plates are not connected to each other. This is due to the impaired function in producing hydrogen gas as well as the electric current flowed. The plates used in this test are Cu with a thickness of 0.4 mm and Al plate with a thickness of 0.7 mm. Al plate use is thicker than Cu because at the time of electrolysis process occurs Al plate that undergoes oxidation, with the reaction:

\[
\text{Al}_{(s)} \rightarrow \text{Al}^{3+}_{(aq)} + 3e^{-}
\]
This causes a lack of aluminum plate weight during the electrolysis process so that the ability of Al does not decrease, and then the weight is made thicker than Cu. The results of hydrogen gas concentration-response measurement results can be seen in Table-1.

### Table-1: Hydrogen Gas Concentration Response Measurement Results

<table>
<thead>
<tr>
<th>Formulation</th>
<th>Concentration of NaNO₃ (M)</th>
<th>Time (Minutes)</th>
<th>Concentration of H₂ (ppm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>0.625</td>
<td>35</td>
<td>81.0</td>
</tr>
<tr>
<td>2</td>
<td>0.625</td>
<td>35</td>
<td>92.0</td>
</tr>
<tr>
<td>3</td>
<td>0.625</td>
<td>35</td>
<td>90.0</td>
</tr>
<tr>
<td>4</td>
<td>0.625</td>
<td>35</td>
<td>88.0</td>
</tr>
<tr>
<td>5</td>
<td>0.625</td>
<td>35</td>
<td>89.0</td>
</tr>
<tr>
<td>6</td>
<td>0.750</td>
<td>35</td>
<td>83.5</td>
</tr>
<tr>
<td>7</td>
<td>1.000</td>
<td>35</td>
<td>114.5</td>
</tr>
<tr>
<td>8</td>
<td>0.625</td>
<td>60</td>
<td>114.0</td>
</tr>
<tr>
<td>9</td>
<td>0.625</td>
<td>10</td>
<td>99.0</td>
</tr>
<tr>
<td>10</td>
<td>0.250</td>
<td>10</td>
<td>101.0</td>
</tr>
<tr>
<td>11</td>
<td>0.250</td>
<td>10</td>
<td>99.0</td>
</tr>
<tr>
<td>12</td>
<td>1.000</td>
<td>10</td>
<td>85.0</td>
</tr>
<tr>
<td>13</td>
<td>1.000</td>
<td>10</td>
<td>117.0</td>
</tr>
<tr>
<td>14</td>
<td>0.250</td>
<td>60</td>
<td>100.0</td>
</tr>
<tr>
<td>15</td>
<td>0.250</td>
<td>60</td>
<td>114.0</td>
</tr>
<tr>
<td>16</td>
<td>1.000</td>
<td>60</td>
<td>144.0</td>
</tr>
<tr>
<td>17</td>
<td>1.000</td>
<td>60</td>
<td>142.0</td>
</tr>
</tbody>
</table>

Based on Table-1, it can be seen hydrogen gas concentration ranges from 81 ppm to 144 ppm. The variety analysis (ANOVA) results showed that the model selected for the response is quadratic because the R² value is greater than other models, which is 0.8465. In addition, this model is significant with a p-value smaller than 0.05 (0.0009). The ANOVA result also shows that the concentration of NaNO₃, and the time affected the response of hydrogen gas concentrations with an insignificant lack of fit greater than 0.05 (0.5231), as shown by Table-2.

### Table-2: Model Analysis for Hydrogen Gas Concentration

<table>
<thead>
<tr>
<th>Response</th>
<th>Model</th>
<th>Mathematics</th>
<th>Significant (p&lt;0.05)</th>
<th>Lack of fit (p&lt;0.05)</th>
<th>R²</th>
</tr>
</thead>
<tbody>
<tr>
<td>Hydrogen gas concentration</td>
<td>Quadratic</td>
<td>Y= -112.4654A – 1.767007B + 84.05987A² +0.022958B² + 0.96000AB</td>
<td>0.0009</td>
<td>0.5231</td>
<td>0.8465</td>
</tr>
</tbody>
</table>

Table-2 above also shows that the increase of electrolyte concentration and time, the greater the concentration of hydrogen gas produced. It indicates that the concentration of NaNO₃ and time is directly proportional to the concentration of hydrogen gas produced. Adding electrolytes during the electrolysis process can affect the production of hydrogen gas. This is because the electrolyte solution that acts as an electric transmission decomposes into positive ions and negative ions become more so that the distance between ions will be shorter. It impacts the amount of electric current flowing, and the reaction of water molecule breakdown becomes fast, and more hydrogen gas is formed.

In this study, a 4/4 plate (Cu/Al) with an electrode model was arranged by following sandwich model with a gap of 2 mm. The short distances can lead to low resistance to transport ions that can produce hydrogen gas quite well. The shorter the cathode distance to the anode also causes the faster movement of electrons, causing friction between electrons that occur faster. The use of the number of electrodes 4/4 (Cu/Al) refers to research with currents and voltages used respectively 0.6 A and 2 V. That is because if
the current and voltage or a number of plates used are too large or increased, then it can decrease the performance of the generator used so that the hydrogen gas produced is less than the maximum.

RSM equation for optimization of NaNO₃ concentration factor and time to hydrogen gas concentration - response are as follows:

\[ Y = -112.4654A - 1.767007B + 84.05987A^2 + 0.022958B^2 + 0.96000AB \]

Where, A: Concentration of NaNO₃ and B: Time

The equation above shows that the concentration of hydrogen gas will decrease with increasing NaNO₃ concentration and time. The decrease in the concentration value of hydrogen gas is thought to occur because the concentration of NaNO₃ is increasingly saturated. The movement of anions and cations in the electrolyte solution becomes limited, and the electrode is not strong in electrolysis for a long time. This is indicated in the negative constant value. In interactions between NaNO₃ concentrations and time interactions are generally directly proportional to the concentration of hydrogen gas produced indicated in positive constant values. The program recommends optimization solutions in as many as 4 formulas (as shown in Table-3), one of 4 optimization solutions is taken to be validated, namely, formula one.

<table>
<thead>
<tr>
<th>Number</th>
<th>NaNO₃ Concentration</th>
<th>Time</th>
<th>H₂ Concentration</th>
<th>Desirability</th>
</tr>
</thead>
<tbody>
<tr>
<td>1*</td>
<td>1.00</td>
<td>60.00</td>
<td>143.393</td>
<td>0.990</td>
</tr>
<tr>
<td>2</td>
<td>0.25</td>
<td>60.00</td>
<td>105.736</td>
<td>0.627</td>
</tr>
<tr>
<td>3</td>
<td>0.25</td>
<td>10.00</td>
<td>101.736</td>
<td>0.416</td>
</tr>
<tr>
<td>4</td>
<td>1.00</td>
<td>10.00</td>
<td>103.393</td>
<td>0.355</td>
</tr>
</tbody>
</table>

Note *: Selected

Figure-4 shows the surface contour graph as a combination of components that can affect the concentration value of hydrogen gas, and in form of 3D of Hydrogen Gas Concentration Test Results can be seen from Fig.-5. Figure-4 and 5 show the influence of NaNO₃ concentration and time affect the concentration of hydrogen gas produced. Optimization of the highest concentration of hydrogen gas was obtained at a concentration of NaNO₃ 1 M and a time of 60 minutes with a hydrogen concentration of 143.393 ppm. The time and concentration of NaNO₃ are directly proportional to the concentration of hydrogen gas produced. The high concentration of electrolytes can accelerate the reduction of resistance in electrolytes resulting in faster electron transfer of electrolysis. While the increase in temperature during
the electrolysis time also causes the faster movement of electrons to decompose water molecules into forming elements so that the hydrogen gas obtained is more maximal.

**Fig.-5: 3D Form of Hydrogen Gas Concentration Test Results**

**Optimization and Verification Results**

After obtaining the mathematical model of the response, it can be done optimization. The goal of optimization is to get the best combination of models and produce the appropriate response desired. The following Fig.-6 shows contour desirability value of the optimum formula, and Fig.-7 shows 3D form of optimum formula desirability value.

**Fig.-8: Contour Desirability Value of Optimum Formula**

In Table-4, the predicted value of the program in formula one with the resulting hydrogen concentration of 143.393 ppm. The lowest predicted interval (PI) is 138.29 ppm, and the highest is 146.79 ppm. The best optimization value is indicated by a desirability value that is close to one. The range of desirability values is 0 – 1. Based on the optimization process, the Design Expert 6.0.9 program provides 4 optimization solutions with a concentration of NaNO₃ 1 M. A time of 60 minutes is recommended as an optimal formula solution because it has the highest desirability value 0.990. Desirability value approaching one can be concluded that a formula with a concentration of NaNO₃ 1 M and time of 60 minutes will produce a concentration of hydrogen gas that has characteristics that match the optimization target and is predicted to produce a hydrogen gas concentration of 143.393 ppm.
If referring to Fig.-4, the dots on the contour chart show a combination of NaNO₃ concentration and time in different amounts that produce the same specific desirability value. The predicted point in the image shows a combination of NaNO₃ 1 M, and 60 minutes, resulting in a desirability value of 0.990. In the results of verification of the optimum formula recommended design expert program 6.0.9, it obtains hydrogen gas concentration of 144 ppm. If compared to the predicted value (as shown in Table-4), the verification result value is in the range of 95 % low predictive interval and 95 % high predictive interval, the selected formula of the optimum solution recommended by the Design Expert program is quite good.

In this study, a 4/4 plate (Cu/Al) with an electrode model was arranged by following sandwich model, with a gap of 2 mm. The following Fig.-8 shows Sandwich Model Electrodes after electrolysis.

The short distances can lead to low resistance to transport ions that can produce hydrogen gas quite well. The shorter the cathode distance to the anode also causes the faster movement of electrons, causing friction between electrons. The use of the number of electrodes 4/4 (Cu/Al) refers to research with currents and voltages used respectively 0.6 A and 2 V. That is because if the current and voltage or the number of plates used are too large or increased, then it can decrease the performance of the generator used so that the hydrogen gas produced is less than the maximum.

Calculations to determine the weight of Al and Cu are lost or increased when the electrolysis process can be used formula:

\[ Q = I \times t \]

Where, \( Q \) = Electricity (Coulomb), \( I \) = Current (Ampere), \( t \) = Time (second)

The weight of Al lost during the electrolysis process as much as 0.1979 g. In comparison, the weight of Cu electrodes increased by 0.6985 g. Changes in the electrodes' weight indicate that both electrodes experienced oxidation reactions in Al and reduction reactions in Cu.
CONCLUSION
The addition of concentration and time could increase the concentration of hydrogen gas produced in the electrolysis process. Based on the optimization results, it obtained 4 optimization solutions with one verified formula which is formula one. The factors in the selected solution were NaNO$_3$ concentration 1 M and time 60 minutes. The verification value of the selected solution response was hydrogen gas concentration of 144 ppm. As a result, the solution recommended by the Design Expert program was good enough.

ACKNOWLEDGMENT
This study was supported by the Republic of Indonesia Government, PTUPT grant number 414/UN35.13/LT/2021, and Rector of Universitas Negeri Padang for Grants No: 1008/UN35.13/LT/2021.

REFERENCES

[RJC-6632/2021]