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Hydrogen Recovery from Electroplating Wastewater Electrocoagulation Treatment

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Abstract— The electroplating industry is one of the industries that contribute to liquid waste that can pollute the environment. An increase must also follow the rapid development of the electroplating industry in the wastewater treatment system. The presence of heavy metals such as chromium (Cr) and nickel (Ni) in electroplating wastewater can cause problems for humans and the environment; hence, electroplating wastewater treatment needs to be done. One form of electroplating wastewater treatment is that it can be processed as a material to obtain hydrogen gas (H₂) as a new energy source. Electroplating wastewater can be processed into hydrogen gas by the electrocoagulation method using metal electrodes. In this study, the production of hydrogen gas from electroplating wastewater was carried out with a 2 (two)-step process, namely the treatment of electroplating wastewater with an electrocoagulation device and followed by the process of processing electroplating wastewater into hydrogen gas using an oxyhydrogen reactor. In the process of treating electroplating wastewater into hydrogen gas, KOH catalyst is added with varying concentrations, and the electrolysis time is 5 minutes. The purpose of adding a KOH catalyst is to obtain optimum hydrogen gas in the electroplating wastewater treatment process into hydrogen gas. The optimum KOH catalyst concentration for electroplating wastewater treatment into hydrogen gas after 5 minutes is 0.5 M, with a volume of hydrogen gas generated of 2.875 L and H₂ content of 554 mg/m³.

Keywords—Electroplating wastewater; electrocoagulation; hydrogen recovery; treatment.

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I. INTRODUCTION

Electrocoagulation is a wastewater treatment technology that recovers valuable chemicals from wastewater. Electrocoagulation is an electrochemical technique involving the corrosion of anodes (aluminum or iron electrodes) to release active coagulants into solution [1]–[3]. Electrocoagulation has several advantages over other conventional techniques such as chemical coagulation and adsorption, including "in situ" delivery of reactive agents, no secondary pollution generation, and compact equipment [4], [5]. Many studies have reported the efficacy of electrocoagulation in treating a wide range of wastewater, including removing suspended solids, dyes, heavy metals, oil emulsions in water, and complex organics [6]–[8]. Despite a number of studies on electrocoagulation for wastewater treatment, large-scale applications of this technology have been limited [9][10]. One possible explanation is the

electrocoagulation process with high energy demand [11], [12]

Electroplating or plating is a process of coating a solid material with a metal layer using an electric current through an electrolyte solution. Waste from the electroplating process is heavy metal waste which is included in Hazardous Toxic Waste [13], [14]. Some metal elements in electroplating wastewater include iron, chromium, zinc, nickel, manganese, and copper. The quantity of waste generated in the electroplating process is not very large, but the level of toxicity is hazardous, especially chromium, nickel, and zinc [15]–[17]. The characteristics and toxicity of electroplating wastewater vary depending on the operating conditions, the coating process, and the rinsing method [18], [19].

Direct disposal of liquid waste from the electroplating process into the environment without treatment can cause environmental pollution [20]. These contaminants can contaminate microorganisms and their environment through solutions, colloids, or other particle forms. Considering the importance and magnitude of the impact on the environment

if it is disposed of directly, it is necessary to treat or utilize the electroplating liquid waste [21].

One of the uses of electroplating liquid waste is that it can be used as hydrogen gas because oxyhydrogen gas (HHO) in the water has extraordinary potential when maximized [2]. In addition, the metal content of electroplating waste such as iron, chromium, zinc, nickel, manganese, and copper can accelerate the process of forming hydrogen gas because the solution is increasingly electrolytic [22]–[24]. Hydrogen gas is used as one of the new renewable energies because it is projected to be an alternative energy future fuel that is environmentally friendly and more efficient. The energy supply produced is very clean because it only produces water vapor as an emission during the process [25].

The hydrogen production process can be done biologically or chemically. Biologically (biotechnology) is a technique of utilizing living organisms or their parts to make or modify a product and improve/improve the properties of organisms for specific uses and purposes such as for food, pharmaceuticals, and energy [26]. Hydrogen can be produced by photosynthesis *Alga Scenedesmus sp* with variations in irradiation to increase hydrogen gas production [27], [28]. Hydrogen production by adaptation of aerobic bacteria isolates producing hydrogen gas in organic waste medium has also been investigated [29]. The biological hydrogen gas production process can also be carried out by fermentation using biomass as raw material with an anaerobic process [30].

Chemically, hydrogen production can be carried out through methanol decomposition using lanthanum as doping. Nuclear-based hydrogen production can be done through a steam-reforming dimethyl ether (DME) process with a low-temperature nuclear reactor as a heat source. In addition, hydrogen production can be carried out photo-catalytically from pure water on a NaTaO₃ catalyst. Another hydrogen production process is also by non-thermal plasma electrolysis in a glycerol-KOH solution medium. Hydrogen can also be produced by electrolysis using stainless steel as an electrode [31], [32].

This study aims to recover hydrogen gas through water electrolysis using stainless steel electrodes because it is more economical. From the electrolysis of water, hydrogen (H₂) and oxygen (O₂) gases are produced. Processing of electroplating waste into hydrogen gas as new renewable

energy is carried out by processing electroplating waste into clean water first with an electrocoagulation process using an electrocoagulator. The clean water product will then be processed into hydrogen gas using an oxyhydrogen reactor [33].

This research utilizes electroplating waste as raw material to obtain hydrogen gas with various catalyst concentrations [32]. The catalyst used is KOH because the results of the catalyst test in the previous research on hydrogen production from aluminum waste, KOH catalyst is the best catalyst in terms of efficiency in the use of electric power and the flow rate of hydrogen gas production, which results are relatively the same as the NaOH catalyst [34]. Compared with other catalysts such as NaCl and H₂SO₄, because the neutral catalyst does not react in the hydrogen gas recovery process, it is inversely proportional to the alkaline catalyst, which reacts strongly to provide optimum production results [35]. The concentration of KOH catalyst can affect the electrolysis process to produce hydrogen gas where the greater the concentration of KOH catalyst used, it is suspected that the greater the opportunity to produce greater hydrogen gas [36].

II. MATERIAL AND METHOD

In the wastewater treatment process, the electroplating waste used is the result of the chrome and nickel-plating process. The process of treating electroplating waste into clean water uses the electroagulation method with aluminum-type electrodes. The catalyst used was KOH solution with various concentrations.

The integrated liquid waste is accommodated in the feed tank. Then pump 1 is turned on to drain the waste to the carbon and silica filters to the electrocoagulation tank. The electrode is turned on, and set the voltage that would be used for the electrocoagulation process is then, the electric current would automatically be read when setting the voltage. The electrodes are turned off after the electrocoagulation process is complete, and open the tap on the electrocoagulation tank is to accommodate the waste in the settling tank with pump 2 turned on to drain the waste from the settling tank to the granular active carbon (GAS) filter, cartridge block carbon (CTO) and reverse osmosis (RO) until to the product tank. The process flow diagram of the electrocoagulation device is shown in Fig. 1.

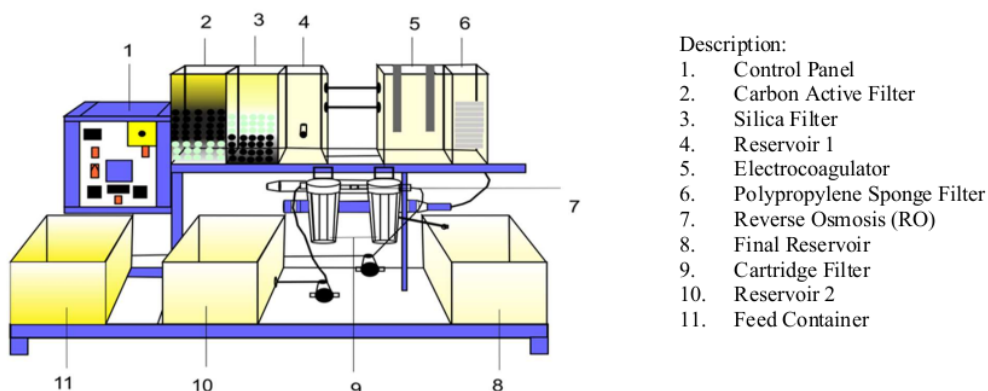


Fig.1 Electrocoagulation Device

Description:

1. Control Panel
2. Carbon Active Filter
3. Silica Filter
4. Reservoir 1
5. Electrocoagulator
6. Polypropylene Sponge Filter
7. Reverse Osmosis (RO)
8. Final Reservoir
9. Cartridge Filter
10. Reservoir 2
11. Feed Container

The clean water obtained from the electrocoagulation process would be used as feed for the oxyhydrogen reactor (HHO). This clean water would be added with a potassium hydroxide (KOH) catalyst before being fed to the device to accelerate the electrolysis reaction process in producing hydrogen gas. In treating hydrogen gas from integrated wastewater using an oxyhydrogen reactor, there is a variable that is varied: variations in catalyst concentration (KOH). A total of 6 liters of feed water resulting from integrated wastewater treatment were added with KOH with each concentration without a catalyst, 0.1 M, 0.2 M, 0.3 M, 0.4 M,

and 0.5 M, and put into the equipment. The oxyhydrogen reactor is through the pipe at the back of the device; previously, the gas flow and pipe taps were opened. The faucet is closed when all the feed is in the reactor, and then attach a urine bag to accommodate the gas produced. The electrodes are turned on at the same time as turning on the stopwatch, record the measured electric current and wait for the pressure measurement to be read and then recorded. Open the valve and let it sit until the urine bag is full of gas. The process flow diagram for the oxyhydrogen (HHO) Cell Reactor is shown in Fig. 2.

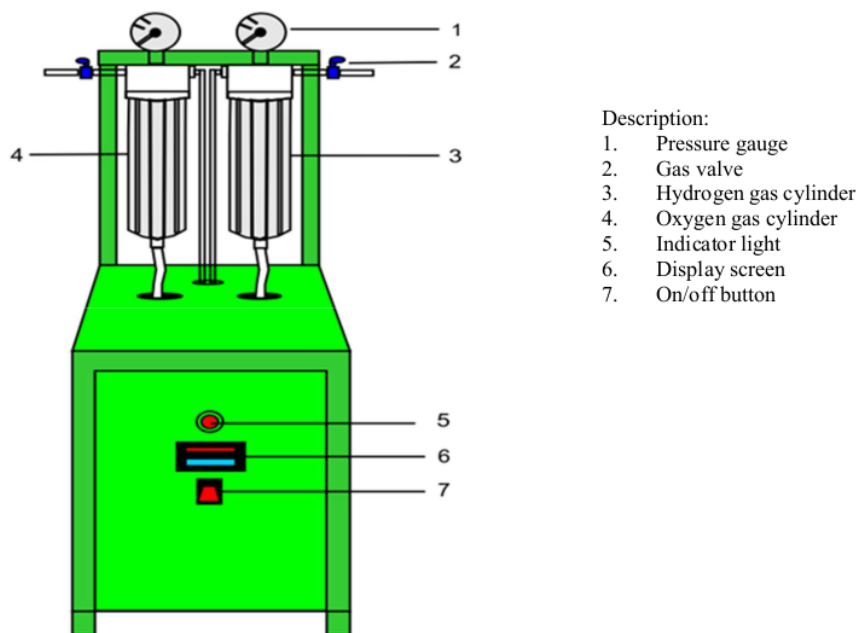


Fig. 2 Design of oxyhydrogen (HHO) Cell Reactor

Description:

1. Pressure gauge
2. Gas valve
3. Hydrogen gas cylinder
4. Oxygen gas cylinder
5. Indicator light
6. Display screen
7. On/off button

III. RESULT AND DISCUSSION

A. Analysis of Preliminary and Final Wastewater from Electroplating Wastewater Treatment

From the results of processing with the electrocoagulation method, it was shown that there was a slight increase in the degree of acidity (pH) from 4.30 to 4.41. The increase in the pH value in the electrocoagulation process was caused by the increase in hydroxide ions (OH⁻) formed. The amount of OH⁻ produced will increase the pH, and the solution is alkaline [37]. This is supported by the statement of Gupta and Ali [38] that the pH of the solution slightly increased during the electrocoagulation process due to the formation of OH⁻ and H₂ gas.

After processing, the total dissolved solids (TDS) and conductivity values were observed to decrease slightly from the initial TDS of 351 mg/L to 347 mg/L, and the initial conductivity of 105.9 S/cm decreased to 100.3 S/cm after treatment with electrocoagulation. This decrease is because, in electrocoagulation, the Al³⁺ ions formed at the anode electrode will bind to OH⁻ ions from the cathode electrode so

that a coagulant will be formed, which will bind the existing contaminants so that they are more easily deposited. The decrease in the TDS value results in a decrease in the conductivity of the solution after electrocoagulation because the conductivity of water is directly related to the concentration of ionized dissolved solids in the water. The conductivity value is influenced by the number of ions contained in the solution. The more amount of dissolved solids, the more the number of ions in a solution because the amount of dissolved solids contains ions that are arranged into compounds in the dissolved solids. This shows that the TDS and conductivity values will have a comparable relationship. This can be proven by the results of research conducted that the conductivity value will decrease if the TDS value also decreases, and the conductivity value will increase if the TDS value also increases.

Chromium in the electroplating industry is used in hard coating processes to be anti-tarnish. The toxic effect of Cr metal from electroplating waste can be bioaccumulative and non-biodegradable. Data on Cr levels obtained through measurements with ICP-OES showed a decrease in Cr levels after treatment with electrocoagulation, with an initial

concentration of 2.21 mg/L decreasing to 1.91 mg/L after processing. This decrease was due to the formation of coagulants. The more coagulant mass, the more easily $\text{Cr}(\text{OH})_3$ precipitates form, and the Cr metal content decrease. The chromium in the waste will interact with OH^- ions which cause Cr to settle in the waste and form $\text{Cr}(\text{OH})_3$. The $\text{Cr}(\text{OH})_3$ precipitate can act as an electropositive floc core and attract excess OH^- in the solution to form floc and other metal cations contained in the waste.

Nickel in the electroplating industry is used in undercoats for decorative chrome plating those coats steel components. Data on Ni content obtained by measuring with atomic absorption spectrophotometry (AAS) showed a decrease in Ni levels, which was 3.514 mg/L, down to 0.746 mg/L, and a decrease in Cr levels. This decrease in Ni levels was due to the formation of many coagulants. The more mass ratio of coagulant causes the formation of $\text{Al}(\text{OH})_3$ precipitate. The more mass of the coagulant, the more OH^- ions will react with Ni metal to form $\text{Ni}(\text{OH})_2$ deposits in the waste.

The purpose of treating electroplating wastewater with this electrocoagulation method is to reduce all the content in electroplating waste. It does not have a high concentration level in the oxyhydrogen reactor feed water because it can disrupt the stability and durability of the oxyhydrogen reactor. Thus, the treated water is feasible to be used as feed for the oxyhydrogen reactor to be processed into hydrogen gas due to the metal. The results also have decreased chromium metal content (Cr), nickel (Ni), TDS, conductivity, and increased acidity (pH).

B. The Effect of Concentration of KOH Catalyst on the Volume of Hydrogen Gas Produced

The volume of hydrogen gas produced from electroplating waste treatment is calculated by calculating the moles of electrons and hydrogen, then the volume of hydrogen gas. The calculation of electron mole is calculated using Faraday's law equation with the required data, namely the electric current data that is read and a predetermined time of 5 minutes, then the data resulting from the calculation of the electron mole is needed for the calculation of moles of hydrogen. The calculation of the volume of hydrogen gas produced from electroplating waste treatment is carried out using the ideal gas law equation with the required data, namely the pressure measured in the oxyhydrogen reactor and moles of hydrogen from the calculation results so that the volume of hydrogen gas produced is as shown in Fig. 3.

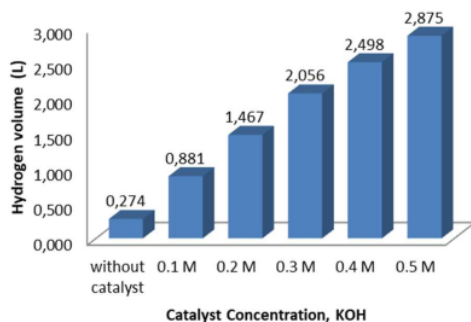


Fig. 3 The effect of the concentration of KOH catalyst on the volume of hydrogen gas (H_2) produced

Fig. 3 shows that there is a relationship between the concentration of the KOH catalyst used and the volume of hydrogen gas produced, with the concentration of the catalyst used at 0.1 M, 0.2 M, 0.3 M, 0.4 M, and 0.5 M above can be seen the volume of hydrogen gas produced. Based on Fig. 3, it can be seen that electroplating wastewater can be processed into hydrogen gas without using a catalyst. This is because electroplating wastewater contains metals that are still carried away after the electrocoagulation process and still have an acidic pH value. The electrolysis process is very dependent on the pH of the solution. The more acidic or wet a solution is, the faster the electrolysis process occurs because the solution will be more electrolytic and have a large conductivity value. The volume of hydrogen gas produced from electroplating waste without using a KOH catalyst is quite small, 0.395 L, so it takes a long time to collect it. In order to get more hydrogen gas and faster time, it is necessary to add a catalyst.

Based on the research results conducted regarding the analysis of the use of various catalysts NaCl, KOH, and NaOH with the observed parameters, namely flow rate and electric power efficiency [38]. NaOH catalyst is the best catalyst to produce 50 mL of hydrogen gas in a relatively faster time, but the time difference is relatively very small compared to the time required to produce 50 mL of hydrogen gas on a KOH catalyst, with a difference of 0.3 seconds. Judging from the electric power efficiency parameters, the KOH catalyst has a much more efficient electric power efficiency than the NaOH catalyst, with a difference of 36 watts. Based on this, KOH catalyst treats electroplating waste into hydrogen gas.

Fig. 3 indicates that of the five variations in the concentration of KOH catalyst used, a concentration of 0.5 M is the largest concentration to produce hydrogen gas, as evidenced by the results of the calculation of the hydrogen gas volume of 2.875 L compared to a concentration of 0.1 M of 0.881 L, a concentration of 0.2 M of 1.467 L, a concentration of 0.3 M 2.056 L, and a concentration of 0.4 M of 2.498 L. The difference in the volume of hydrogen gas produced from each variation of the catalyst concentration is due to the concentration of a solution related to the concentration.

The solution's concentration is the value of the substance or compound dissolved in water. The more substances or compounds that are dissolved in water, the more concentrated the solution will be. The more concentrated the KOH solution, the more ions formed in the solution. Hence, the solution will be more electrolytic, making the electrical resistance in the solution smaller and approaching zero. This phenomenon means that the greater the solution concentration during the electrolysis process, the greater the volume of gas produced. So, producing H_2 and O_2 in large quantities requires increasing the concentration of the solution. Based on the results above, it can be determined that the best concentration of KOH catalyst to produce hydrogen gas (H_2) from electroplating wastewater with a fixed time of 5 minutes is a catalyst concentration of 0.5 M with a volume of 2.875 L.

C. Effect of KOH Catalyst Concentration on Hydrogen and Oxygen Gas Content

The gas parameters measured were hydrogen and oxygen gas. The H_2 parameter is used to determine the amount of hydrogen gas obtained, while O_2 is used to determine the gas's

purity because the less oxygen gas formed, the purer the hydrogen gas obtained. Based on the analysis that has been carried out using a gas measuring device, the results of the measurement of hydrogen gas are obtained, as shown in Fig. 4.

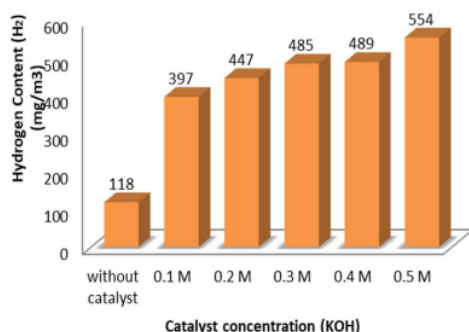


Fig. 4 The effect of the concentration of KOH catalyst on the content of hydrogen gas

From Fig. 4, it can be seen that hydrogen gas produced from electroplating waste with various catalyst concentrations and without catalysts has a fairly high hydrogen gas content. The analysis results show that the waste without a catalyst has the lowest hydrogen gas content and is relatively different from the yield of hydrogen gas using a catalyst, which is 118 mg/m³. From the results of measurements of hydrogen gas with variations in the concentration of KOH catalyst, an increase in hydrogen gas content was produced at each concentration. The concentration of 0.1 M has the lowest hydrogen gas content of 397 mg/m³, the concentration of 0.2 M is 447 mg/m³, a concentration of 0.3 M 485 mg/m³, a concentration of 0.4 M 489 mg/m³, and a concentration of 0.5 M had the highest hydrogen gas content of 554 mg/m³. The analysis results indicate that the concentration of the catalyst also affects the amount of hydrogen gas produced and the volume of hydrogen gas. A graph of the relationship between the content of O₂ gas and the catalyst used can also be made, shown in Fig. 5.

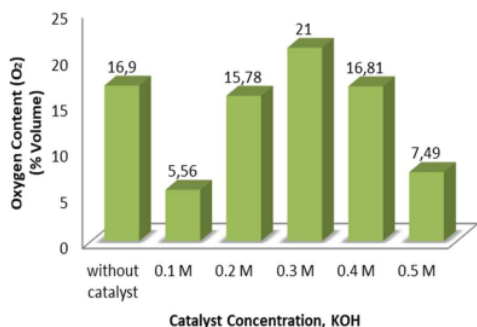


Fig. 5 The effect of the concentration of KOH catalyst on the content of oxygen gas

From Fig. 5, it can be seen that the hydrogen gas produced with various concentrations of catalyst and without a catalyst has no effect on the content of oxygen gas, where the content of oxygen gas without a catalyst has a smaller value of 16.9%

compared to using a catalyst at a concentration of 0.3 M by 21%. This is not under the value of the hydrogen gas content obtained, where the greater the hydrogen gas produced, the smaller the oxygen gas content. This difference is caused by the presence of air that enters the gas gauge during the analysis process. The entry of air is due to the lack of vacuum connecting the hose between the sample container and the sampling pump on the device, causing the detection of oxygen gas in the air. Although the results of the oxygen gas content obtained are not affected by the concentration of the catalyst, the hydrogen gas produced still has a fairly high purity because it contains far less oxygen than the hydrogen gas content. Based on the gas analysis gas results, emission gases are also detected in the air, including CO, NO, NOX, NO₂, SO₂, and CO₂. These gases are also detected the same as O₂ gas which is caused by the presence of air entering the gas gauge.

D. Analysis of Hydrogen Gas Flame Test Results

The flame test analysis process is carried out using soap bubble media, where hydrogen gas that has been accommodated in the urine bag will be blown into a container filled with soapy water so that bubbles will form, then the bubbles containing hydrogen gas will be burned with a lighter. Based on the analysis that has been done, hydrogen gas burns with a fairly large fire, and the results of the flame test produce a yellow-orange color accompanied by an explosion, and the flame disappears quickly.

Under the characteristics of hydrogen gas fire, which is very flammable and the flame tends to disappear quickly in the air so that the damage caused by a hydrogen explosion is lighter than a hydrocarbon explosion and also other characteristics, namely an explosion occurs during the flame test. This explosion indicates that the gas being tested is hydrogen gas caused by the presence of oxygen gas contained in hydrogen gas, indicating that the sample still contains oxygen gas in it.

In the color of the flame, the resulting yellow-orange color structure is called premix flame, where premix flame (premixed flame) is formed when fuel (hydrogen gas) and an oxidizing agent (air) have been mixed before entering the reaction zone. In the premixed flame section, the flame consists of a bright area (yellow color), indicating that the flame is rich in fuel and this premix flame is surrounded by a diffusion flame on the outside (orange color) caused by the combustion flame when carbon monoxide and hydrogen are produced from the fuel-rich interior reacts with the ambient air.

Based on the results of the flame test that has been carried out, it can be seen that the gas produced is actually hydrogen gas because it is flammable, and the characteristics of the flame produced are in accordance with the characteristics of the hydrogen gas flame, namely it disappears quickly, and an explosion occurs. In addition, it produces a bright area (yellow color) in the premix flame, indicating that it is rich in fuel (hydrogen gas).

IV. CONCLUSION

This research analyzed the treatment results of electroplating waste using an oxyhydrogen reactor to recover hydrogen. It can be concluded that the clean water obtained from the electrocoagulation method can be used as feed in the

oxyhydrogen reactor. It is due it has increased pH and decreased TDS value, decreased conductivity, chromium (Cr) content, and decreased conductivity. The optimum KOH catalyst concentration for electroplating waste treatment into hydrogen gas after 5 minutes is 0.5 M, with a volume of hydrogen gas generated of 2.875 L and H₂ content of 554 mg/m³.

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