

# Production of H<sub>2</sub> from aluminium/water reaction and its potential for CO<sub>2</sub> methanation

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**Abstract.** Carbon dioxide (CO<sub>2</sub>) is a natural gas that presents in excess in the atmosphere. Owing to its ability to cause global warming, capturing and conversion of CO<sub>2</sub> have attracted much attention worldwide. CO<sub>2</sub> methanation using hydrogen (H<sub>2</sub>) is believed to be a promising route for CO<sub>2</sub> removal. In the present work, H<sub>2</sub> is produced using aluminum-water reaction and tested for its ability to convert CO<sub>2</sub> to methane (CH<sub>4</sub>). Different type of water i.e. tap water, distilled water, deionized water and ultrapure water, concentration of sodium hydroxide (NaOH) (0.2 M to 1.0 M) and particle size of aluminum (45 m to 500 μm) were varied as parameter study. It was found that the highest yield of H<sub>2</sub> was obtained using distilled water, 1.0 M of NaOH and 45 μm particle size of aluminium. However, the highest yield of methane was achieved using a moderate and progressive H<sub>2</sub> production (distilled water, 0.6 M of NaOH and 45 μm particle size of aluminium) which allowed sufficient time for H<sub>2</sub> to react with CO<sub>2</sub>. It was concluded that 1130 ml of H<sub>2</sub> can produce about 560 ppm of CH<sub>4</sub> within 25 min of batch reaction using nickel catalyst.

## 1. Introduction

The development of industrial revolution requires significant increase in energy supply. Fossil fuels play a vital role in supplying the demanded energy. However, huge consumption of fossil fuels has instigated serious environmental impact such as global warming due to greenhouse gases emissions i.e. carbon dioxide (CO<sub>2</sub>), nitrous oxide, and methane (CH<sub>4</sub>) [1]. Therefore, many scientists are currently focusing on the development of carbon-clean and alternate energy. According to Zhou et al. (2016), CO<sub>2</sub> capture and utilization is believed to be one of the reliable energy sources [2]. Among the methods for CO<sub>2</sub> utilization are CO<sub>2</sub> hydrogenation to methanol and CO<sub>2</sub> reforming to methane.

Methanation of CO<sub>2</sub>, which is commonly known as Sabatier reaction is a reversible and extremely exothermic reaction [3]. The product of reaction, CH<sub>4</sub> is the major component of conventional natural gas which can be acquired from associated gas, stranded gas and shale gas [4]. Together with the renewable H<sub>2</sub>, CH<sub>4</sub> can be the key for future energy turnaround strategy in countries such as Switzerland and Germany as it reduces CO<sub>2</sub> emission [5].

Hydrogen (H<sub>2</sub>) is an essential molecule in CO<sub>2</sub> methanation reaction and has drawn much attentions over the decades [6, 7]. H<sub>2</sub> produces three times larger reaction energy than hydrocarbon fuels when it reacts with air [8]. Additionally, the combustion of H<sub>2</sub> produces zero carbon emission and is likely made through water electrolysis [9]. Nonetheless, the production cost is always the issue due to the



conventional method to produce H<sub>2</sub> (mainly water electrolysis) is neither economical nor sustainable [10]. Next, storage and transport of H<sub>2</sub> pose safety concern and has always been a challenge [9, 11]. Thus, a more simple, inexpensive and safe method to generate H<sub>2</sub> is preferred.

Aluminium-water reaction is considered as an alternative reaction method to produce H<sub>2</sub> as aluminium metal is abundant in Earth's crust and the by-product of reaction can be recycled [6, 12]. As reported by Teng et al., 1 g of aluminum can generate 1360mL of H<sub>2</sub> gas in complete reaction with water under ambient condition [13]. However, the restrictive properties of the protective alumina layer lower the reaction rate and H<sub>2</sub> production yield [12]. Therefore, aqueous alkaline solution is applied to remove the passive alumina layer to increase the yield of H<sub>2</sub> gas production [13]. Alkaline solutions such as sodium hydroxide (NaOH), potassium hydroxide (KOH), and calcium hydroxide (Ca(OH)<sub>2</sub>) have been tested [7]. As reported by Huang et al., NaOH and KOH have been proven to improve the H<sub>2</sub> production rate [14].

Thus, in this study, a cheaper source of H<sub>2</sub> gas via aluminum-water reaction is tested for methanation process. The process will be utilized to replace the commercial H<sub>2</sub> gas. So far in the literature the usage of H<sub>2</sub> gas produced from aluminum-water reaction is yet to be tested for methanation. Moreover, a potential new route of CO<sub>2</sub> methanation to generate CH<sub>4</sub> will be explored. Through this way, an alternative and low cost H<sub>2</sub> gas can be generated and used as a source of energy production via CO<sub>2</sub> methanation to CH<sub>4</sub>.

## 2. Materials and Method

### 2.1. Chemical reagents

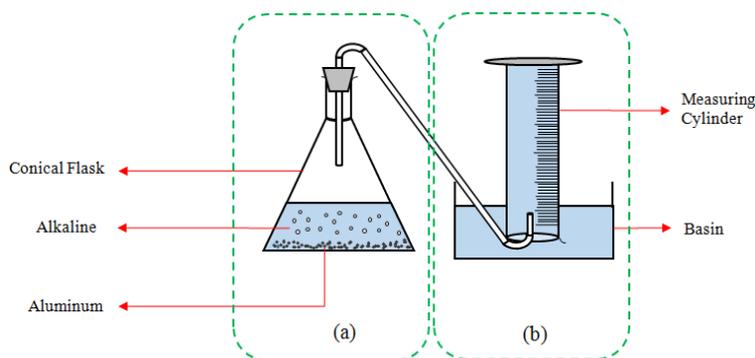
*Favorit* distillation system (*W4L WaterStill*, Malaysia), Elga water purification system (*Micra*, Republic of Korea) and Human Corporation water purification system (*New Human UP 900*, United Kingdom) were used to produce distilled water (resistivity of 0.3 MΩ·cm), deionized water (resistivity of 10 MΩ·cm) and ultrapure water (resistivity of 18.3 MΩ·cm) respectively. Nickel (II) nitrate hexahydrate (Ni(NO<sub>3</sub>)<sub>2</sub>·6H<sub>2</sub>O), aluminium powder and sodium hydroxide (NaOH) were purchased from Sigma Aldrich, USA. Palm shell activated carbon (PSAC) were purchased from Nikom Global Marketing (M) Sdn. Bhd. Malaysia. Pure lab grade CO<sub>2</sub> and N<sub>2</sub> gas cylinders were purchased from Linde Sdn. Bhd. Malaysia. All these materials were used as received.

### 2.2. Preparation of catalyst

Catalyst for the methanation was prepared according to Younas *et al.* [15]. Palm shell activated carbon (PSAC) was used as a support to prepare nickel based catalyst. 15wt% of Ni on PSAC was prepared using incipient wetness impregnation method. Stoichiometric amount of nickel nitrate hexahydrate (Ni(NO<sub>3</sub>)<sub>2</sub>·6H<sub>2</sub>O) was dissolved in distilled water and PSAC was added into the solution. The mixture was sonicated at 60 °C for 5 hours. Then, the mixture was evaporated in a rotary evaporator under reduced pressure. After evaporation, samples was dried in the oven at 110 °C overnight. The dried sample was then calcined in N<sub>2</sub> at 500 °C for 3 hours in a tubular furnace.

### 2.3. Hydrogen Production via Water Displacement Apparatus

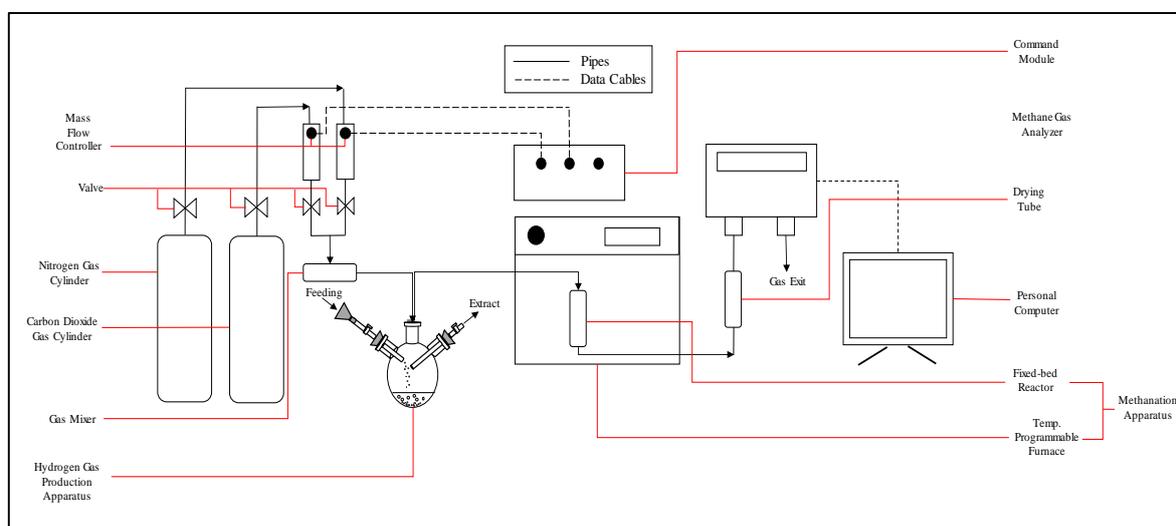
Figure 1 shows the experimental setup. A defined amount of NaOH solution and aluminum powder were added into a conical flask. A rubber stopper with plastic tubing at the neck was used to prevent the gas leakage and channel the gas produced to the measuring cylinder. A measuring cylinder was fully filled with water, inverted and was immersed into a basin of water. The generated H<sub>2</sub> gas occupied the space in the measuring cylinder, lowering the water level in the measuring cylinder. The occupied level in the measuring cylinder was recorded based on time. A range of parameter study i.e. type of water used for reaction (tap water, distilled water, ultrapure water and deionized water), NaOH concentration (0.2 M, 0.4 M, 0.6 M, 0.8 M, 1.0 M) and different particle sizes of aluminum powder (45 μm, 63 μm, 300 μm, 355 μm, 500 μm) were tested for H<sub>2</sub> production. Each test was repeated twice and an average value was reported.



**Figure 1.** Schematic diagram of hydrogen production apparatus (a) and water displacement apparatus (b).

**2.4. CO<sub>2</sub> methanation**

0.5 g of nickel catalyst was used to initiate CO<sub>2</sub> methanation reaction at temperature 220 °C [4]. The produced H<sub>2</sub> gas and CO<sub>2</sub> were flowed through the CO<sub>2</sub> methanation apparatus until a stoichiometric ratio of H<sub>2</sub>/CO<sub>2</sub> = 4 was achieved [16]. To achieve the ratio 20ml of CO<sub>2</sub> was purged every minute into the system. The outlet gases (CH<sub>4</sub> and CO<sub>2</sub>) produced from the methanation reaction were collected into a gas sampling air bag every 5 min. The outlet gases were analysed using *MRU Air Emission Monitoring System (VARIOplus, Germany)*. Figure 2 shows the schematic diagram of CO<sub>2</sub> methanation apparatus. Methanation test was repeated five times for accuracy and an average value was reported.



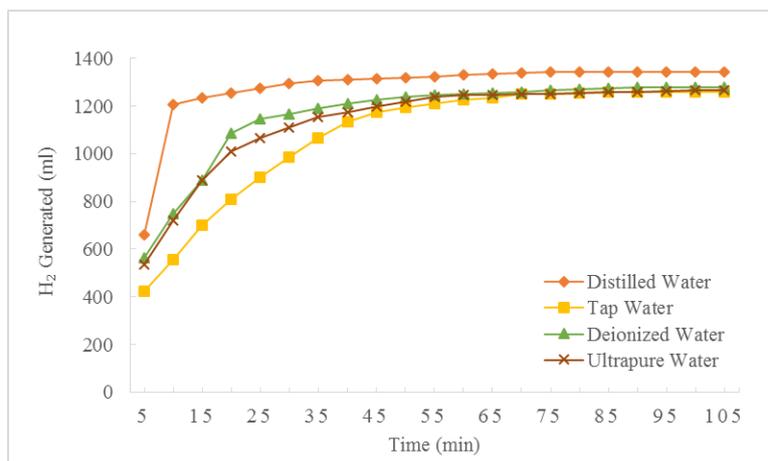
**Figure 2.** Schematic diagram of CO<sub>2</sub> methanation apparatus.

**3. Results and Discussion**

The effects of different type of water, concentration of NaOH and aluminium particle size were studied. H<sub>2</sub> solubility in water is very low, thus the volume of H<sub>2</sub> generated from the aluminium-water reaction was measured by the volume of water displaced with gas. The cumulative amount of H<sub>2</sub> produced from aluminium-water was measured and compared with the maximum theoretical yield of H<sub>2</sub> *i.e.* 1360 mL from 1 g of aluminium powder [13].

### 3.1. Effect of Water Type

200 mL of 1.0 M NaOH was prepared using distilled water, tap water, deionized water and ultrapure water respectively to examine the effect of type water for the H<sub>2</sub> generation. 1 g of 45 μm particle sized aluminum powder was used in this test. Figure 3 represents the amount of H<sub>2</sub> generated from aluminum-water reaction using different type of water.



**Figure 3.** Hydrogen generated (ml) using different type of water.

Based on the data obtained, reaction using distilled water developed the fastest reaction rate. The reaction rate increased rapidly from the 1<sup>st</sup> min to the 7<sup>th</sup> min and reached stable condition after the 9<sup>th</sup> min. The maximum amount of H<sub>2</sub> generated was 1343 ml and was the highest amongst the others. Reaction using deionized and ultrapure water, produced slightly lower amount compared to distilled water. Moreover, the reaction rate was much slower compared to distilled water. It took about 20 min for deionized and ultrapure water to achieve about 1085 ml and 1010 ml respectively. Similar results was reported by Wang et al. [11]. It was known that salt content could help to activate aluminum surface and indirectly speed up the reaction of H<sub>2</sub> production. The more concentrated the salt content in the solution, the shorter the induction period of the reaction, the faster the reaction to reach production yield [17, 18].

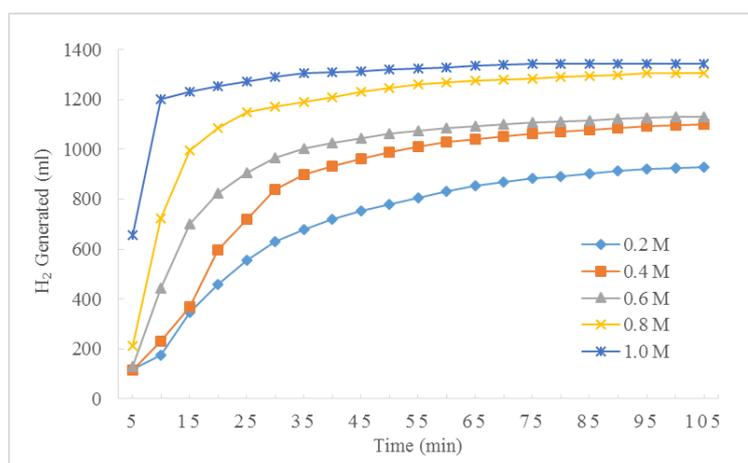
Resistivity is a measurement of water quality as function of salt content (or ion) in water [19]. The resistivity of water is inversely proportional to the concentration of salt content. The resistivity of tap water, distilled water, deionized water and ultrapure water in this study were found to be as 0.0073, 0.3, 10.0 and 18.3 MΩ·cm respectively. It is clearly seen that the ascending order of resistivity of the water was in line with the generation of H<sub>2</sub>. However, it was noticed that even though the resistivity of tap water was very low, the production of H<sub>2</sub> was still the least with a maximum yield of 1260 ml after 105 min of reaction. This was probably due to the impurities and chlorine content in the tap water. It is known that chlorination will cause significant loss in metallic aluminum and formation of aluminum chloride [20] which indirectly slows down the reaction. Therefore, the reaction rate of tap water was found to be the slowest.

### 3.2. Effect of NaOH concentration

To evaluate the effect of NaOH concentration on H<sub>2</sub> generation, a series of experiments were performed with different concentration of NaOH from 0.2 M to 1.0 M. 1 g of 45 μm aluminum powder was employed to react with 200 mL of NaOH solution in distilled water for this parameter study. Figure 4 describes the cumulative amount of H<sub>2</sub> generated from aluminum-water reaction at different concentration of NaOH.

As shown in figure 4, the higher the concentration of NaOH, the faster the rate of reaction for H<sub>2</sub> production. When the concentration of NaOH was 0.2 M, the rate of reaction was the slowest,

compared to reaction using higher concentration of NaOH. The fastest reaction was observed at 1.0 M, whereby within 10 min about 1200 ml of H<sub>2</sub> has been produced. At the same time, 0.2, 0.4, 0.6 and 0.8 M produced only about 175, 230, 445 and 725 ml of H<sub>2</sub> respectively. The highest yield was also obtained from 1.0 M i.e. 1342.5 ml. It was noticed that 0.6 and 0.4 M showed a trend of progressive and moderate reaction with almost a similar yield after 105 min of reaction. Similar results was reported other researchers [21, 22].



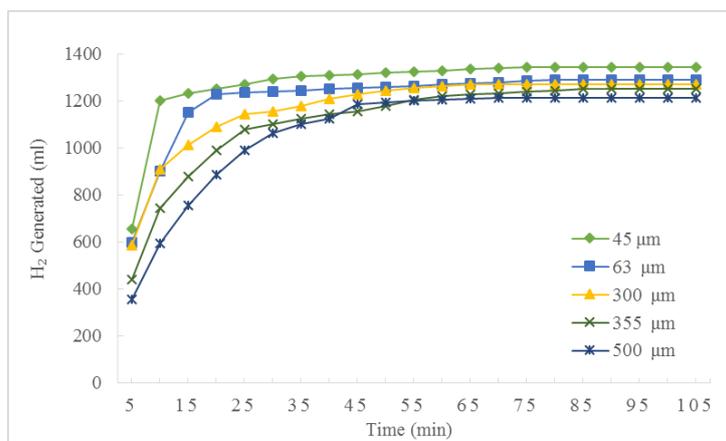
**Figure 4.** Hydrogen generated (ml) using different concentration of NaOH.

In general, the rate of reaction is faster when high concentration of alkaline is employed. This is because when the alkaline concentration is high, there are more hydroxide ion in the solution to dissolve the alumina film [14, 23]. The chemical attack progress results in more fresh aluminum surface which enhances the reaction with water to generate H<sub>2</sub>. Therefore, the initial rate of aluminum-water reaction was fast. As the hydroxide ion was consumed along the reaction, there were lesser hydroxide ion in the solution to dissolve the alumina film, leading to less reaction between fresh aluminum surface and water. Hence, the reaction rate dropped and the amount of H<sub>2</sub> generated correspondingly reduced.

### 3.3. Effect of aluminium particle size

Different particles sizes of aluminum, i.e. 45 μm, 63 μm, 300 μm, 355 μm and 500 μm were investigated with 200 mL of 1.0 M NaOH solution on H<sub>2</sub> production using distilled water. The experimental results were shown in figure 5. Figure 5 clearly shows that the smaller the particle size of aluminum, the more rapid the reaction rate and a shorter time is needed to achieve the maximum yield of H<sub>2</sub>. 45 μm exhibited the most rapid reaction rate with 1200 mL of H<sub>2</sub> generated within 10 min. After that, the reaction started to slow down until the 75<sup>th</sup> min. On the other hand, reaction with 300 μm showed a moderate reaction rate. The reaction was initiated with a fast rate within the first 10 min. Then, the reaction became stable and reached maximum yield of H<sub>2</sub> at the 90<sup>th</sup> min. For the reaction with 500 μm, only 355 mL of H<sub>2</sub> was generated within the first 5 min.

The rate of reaction increased gently after that until it reached a yield a maximum yield at the 75<sup>th</sup> min. A similar work was reported by Ilyukhina, Ilyukhin and Shkolnikov [24]. It is known that smaller size of particle has more surface area for reaction to happen. Smaller size also enhances the dissolution of alumina film [8, 25]. Thus, reaction with smaller aluminum particle size produces more amount of H<sub>2</sub> at a rapid rate [12]. At a point where hydroxide ion is used up in dissolving the alumina outer layer, there is no fresh aluminum to react hence, production is stopped. This shows that the system is stable and working well for H<sub>2</sub> production.

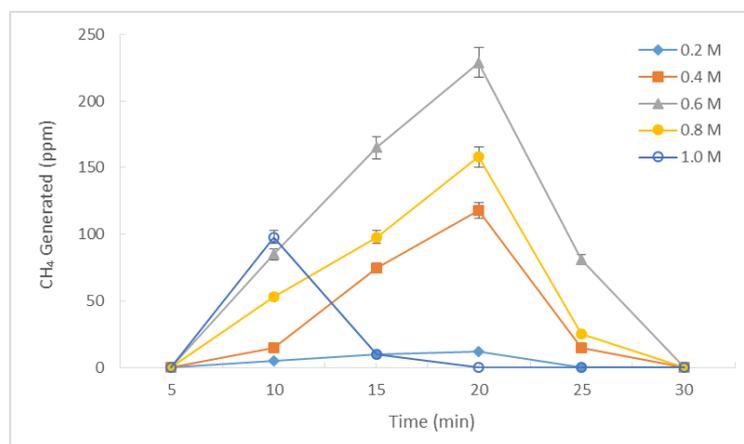


**Figure 5.** Hydrogen generated (ml) using different particle size of aluminum.

### 3.4. CO<sub>2</sub> methanation

CO<sub>2</sub> methanation was conducted using the optimized results of water type and aluminium particle size. The concentration of NaOH was varied to understand the difference in methanation if the production rate of H<sub>2</sub> is fast, moderate or slow. The reacted gas was collected every 5 min for a 35 min of continuous reaction. Figure 6 shows the methanation results. It was observed that a maximum of 25 min of reaction time was needed to produce CH<sub>4</sub> from a batch system. Reaction using 0.2 M and 1.0 M of NaOH obtained the lowest amount of CH<sub>4</sub> after 25 min of reaction as shown in Table 1. Interestingly 0.6 M showed the highest amount of methanation with about 229 ppm of CH<sub>4</sub> produced at the 20<sup>th</sup> min and a total of 560 ppm for a complete 25 min of reaction.

The rate of methanation in 1.0 M was low due to insufficient time for the reaction to happen between CO<sub>2</sub> and H<sub>2</sub> for CH<sub>4</sub> conversion. It was observed that at 1.0 M, H<sub>2</sub> was generated too fast with maximum yield within 10 min. It was suspected that most of the generated H<sub>2</sub> passed through the reactor without reaction as the analyser detected high concentrations of unused CO<sub>2</sub> in the outlet. A moderate and progressive reaction using 0.6 M of NaOH has shown the best conversion. Progressive or moderate production rate of H<sub>2</sub> allowed sufficient time for the reaction between H<sub>2</sub> and CO<sub>2</sub> and maintained the stoichiometric ratio between H<sub>2</sub> and CO<sub>2</sub>. Moreover, there was no CH<sub>4</sub> produced in the first 5 min of the reaction for all concentrations of NaOH. This was probably due to insufficient amount of H<sub>2</sub> produced which indirectly hindered the conversion. Thus, CH<sub>4</sub> was not produced. After 10 min of reaction, CH<sub>4</sub> was detected and the amount of CH<sub>4</sub> increased rapidly until the 20<sup>th</sup> min for 1.0 M. This was because there was sufficient amount of H<sub>2</sub> produced to convert CO<sub>2</sub> to CH<sub>4</sub>.



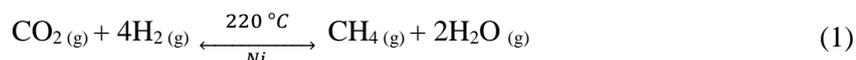
**Figure 6.** Methane generated (ppm) using different concentration of NaOH.

**Table 1.** Total amount of CH<sub>4</sub> produced after 25 min of reaction at different concentration of NaOH.

NaOH concentration (M)	CH <sub>4</sub> produced (ppm)
0.2	27
0.4	223
0.6	560
0.8	334
1.0	108

After the 20<sup>th</sup> min, the reaction declined and stopped at the 30<sup>th</sup> min which identified the completion of the aluminum-water reaction. Hence, there was no H<sub>2</sub> produced for CO<sub>2</sub> methanation to take place.

Besides, reaction using 0.2 M of NaOH could not produce high amount of CH<sub>4</sub> due to low production rate and yield of H<sub>2</sub>. To date, literature only focuses on catalyst performance of CO<sub>2</sub> methanation. For that reason, there is no similar data or report from literature to be compared. However, the batch CO<sub>2</sub> methanation reaction was expected to be similar to Sabatier reaction as shown in equation 1 below, where CO<sub>2</sub> reacts with H<sub>2</sub> and produces CH<sub>4</sub> at 220 °C in the presence of nickel catalyst.



#### 4. Conclusion

In this study, H<sub>2</sub> gas was produced from aluminum-water reaction. The type of water, concentration of NaOH and particle size of aluminum powder were used as experimental parameter and has been optimized. In short, H<sub>2</sub> yield was directly proportional to the concentration of NaOH and was inversely proportional to resistivity of water and particle size of aluminum. Additionally, the optimum conditions of aluminum-water reaction investigated from these studies were 1.0 M of NaOH solution prepared by distilled water and 45 μm particle size of aluminum powder could yield a maximum of 1343 mL of H<sub>2</sub>.

CO<sub>2</sub> methanation using H<sub>2</sub> produced from aluminium-water reaction could only last for 25 min of reaction. The maximum total yield of CH<sub>4</sub> for 25 min of reaction was 560 ppm and this amount could be only achieved using a moderate reaction rate of H<sub>2</sub> production using 0.6 M of NaOH. The results have proven that H<sub>2</sub> generated from aluminium-water reaction could be successfully used for CO<sub>2</sub> methanation in a batch study.

#### Acknowledgments

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