Hydrogen Production from Aluminium-Water Reactions: Thermodynamic Properties Analysis

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Abstract

Hydrogen production from low price materials at moderated temperature should be developed for sustainable commercial production. The aluminium and water reaction at low temperature is very attractive because it can generate hydrogen without a reactor operating at high temperature and pressure required. The hydrogen production process by hydrolysis reactions of aluminium with NaOH in different types of water was studied. It was aimed at better understanding on how the Cl ions in different types of water affect the hydrolysis at the different reactions for the hydrogen production. It was observed that the ionic strength of the solutions strongly increased with the Cl ions and aluminium mass added. However, simultaneous effect resulted in the coefficient of lower hydroxide ions (OH-) activities. The activities of the hydroxide ions at the employed conditions of the reactions were calculated using Aspen Plus™. NaOH additives dissolved in water, produced hydroxyl ions and consequently promoted the hydrolysis reactions between aluminium and water. The maximum hydrogen generated was 583 ml as obtained from the reaction of 1 g aluminium in 30 ml distilled water for 30 min.

Keywords: Hydrogen; Chloride Ions; Aluminum

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Introduction

With the reduction of global crude stockpiles and the results from global warming, current research has developed new alternative fuels to replace crude oil. Hydrogen is a source of pollution-free energy. It is an alternative of sustainable energy that can be applied to the traditional energy use. Most of the hydrogen productions are, nowadays, done via hydrocarbon steam reforming where methane from natural gas is heated with steam to produce a mixture of carbon monoxide and hydrogen. As a result, the methane reforming process results in carbon dioxide emissions. Therefore, the development of hydrogen processes from non-raw materials at moderated temperature should be developed for the sustainable commercial production. The hydrolysis of aluminium and water at low temperature reaction is very attractive because it can generate hydrogen without a reactor operating at high temperature and pressure required [1]. Typically, studies of the aluminium water reaction in experimental systems have been conducted under atmospheric pressure and at the temperature below the boiling point of water [1]. Aluminium and its alloys are considered as suitable metals for the hydrogen production in the future and are likely to be used as alternative energy sources, especially in form of materials. Aluminium is also a superior material for the future hydrogen production [2] since it is abundant in earth with low density, low price and high capacity and excellent reactivity to water. Accordingly, metal utilization has been identified as an effective, easy, and safe way for the hydrogen production and energy storage.

      Several studies on the hydrolysis reaction in neutral water as medium for the hydrogen production from aluminium showed that water quality affected the hydrogen production through the hydrolysis reactions of aluminium powder in water at ambient temperature [3] - [4]. Elitzur, S., Rosenband, V., and Gany, A. [5] studied on the production and storage of hydrogen from aluminium and water reactions to investigate the effects of aluminium on water and hydrogen. The variables used in the experiment were mass ratio of water to aluminium, water temperature and particle size of aluminium. The aluminium used was that with lithium (2.5 wt%) to catalyze the reaction. The results of the experiment were that the lower of water-to-aluminium ratio, the faster the reaction; that from the water temperature was the higher of water temperature; the shorter of the reaction and that from the particle size was the bigger of the particle size; the shorter of the reaction. Rosenband, V. and Gany, A. [6] developed an application of activated aluminium powder for the generation of hydrogen from water to compare the metal alloys that react to water to give a high hydrogen production rate. The experiment was performed under constant and varied temperature conditions. They also experimented with aluminium alloys with other metals such as sodium hydroxide, cobalt oxide and molybdenum oxide, etc., and

the results showed that the aluminium mixed with lithium gave a greater amount of hydrogen than that with other metals. Macanás, J., Soler, L., Candela, A. M., Muñoz, M., and Casado, J. [7] examined the hydrogen production from aluminium corrosion using diverse alkaline solutions. The results confirmed that hydrogen production in water was enhanced by aluminium corrosion. Aluminium corrosion greatly depended on the nature of the dissolved species and pH of the solution.

Thus, better understanding on how the Cl ions affect the hydrolysis reaction at the different types of water reactions for the hydrogen production is required. The objective of this study is to investigate the characteristics of the interaction between water and aluminium powder in different types of water containing different chloride ions. In order to achieve this objective, as follows are to be observed: First, the hydrogen production from aluminium without NaOH in hydrolysis reaction medium for different types of water including pure water, tap water and sea water and the three different ions conditions containing no Cl ion, Cl ions in the tap water and the sea water (containing Cl ions and Na ions) which are considered as similar mass ratios of water to aluminium powder. Next, in order to verify if the observed trends are specifically for the Cl ion effects or not, the experiments are conducted with varied amounts of aluminium powder and NaOH at similar Cl ion condition and aluminium/water mass ratio that are 30, 50, and 100 mL. For all these reactions, the experiments have to be determined at room temperature and atmospheric pressure with the reaction time of 30 min.

Materials and Methods

1. Materials

Tap water with pH = 6.3 - 7.5 and distilled water with pH = 6.5 - 7 were used in this study. Moreover, the sea water collected from Hua Hin beach in Prachuap Khiri Khan province (Thailand) in March 2016, Metallic aluminium powders (Gammaco (Thailand) Co., Ltd) with 99.7% purity and each particle of 45 μm, and NaOH pellets (Gammaco (Thailand) Co., Ltd) with 98.0% purity were also used for all the experiments.

2. Methods

The hydrolysis reactions of aluminium powder with different chloride ion concentrations in the tap water, the distilled water and the sea water were carried out. Batch-type experiments were performed in a glass reactor, see also Figure 1. Each mass of all the individual aluminium in gram was 0.19, 0.38, 0.57, 0.76, and 0.95 in all experiments, respectively. Moreover, 0.1 g of NaOH was added as the promoter. An overview of all the experiments is presented in Table 1. First, different masses of aluminium powder with

0.1 g of NaOH were placed in the reactor, followed by adding amounts of the tap water, the distilled water and sea water of 30, 50, and 100 mL for each experiment respectively and mixed. Then the hydrolysis reaction was started. The hydrogen was produced by the aluminium and water hydrolysis reactions and discharged water from a second glass cylinder into a beaker was placed on an electronic balance as presented in Figure 1. Due to the fact that hydrogen has low solubility in water, the volume of hydrogen evolved in the aluminium and water reactions could be measured by the displaced masses of water. For all these reactions, the experiments were determined at room temperature and atmospheric pressure with the reaction time of 30 min as described in previous section.
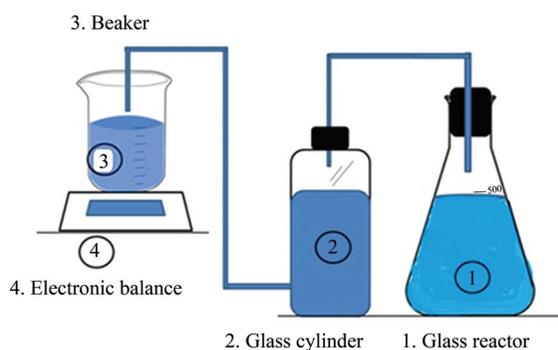

Figure 1 Schematic of the experimental set-up for the hydrogen production from the hydrolysis reactions of Aluminium based compositions (adopted from [6])

Results and Discussion

1. The electrical conductivity and ionic strength

   The major ions for the sea water and the tap water are defined as those ions that have a significant contribution in tap water that is Cl and Na with Cl in sea water, respectively. The concentrations of these major ions for these two hydrolysis solutions were measured by ICP-OES which are presented in Table 1. The results show that the total amounts of major ions were significantly higher in the sea water. Next, electrical conductivity (EC) was measured. These values can be used as indicators for the conduction of current of total dissolved solids in the hydrolysis solutions which are primarily dependent on the concentrations of ionic species. An electrical conductivity meter (EC meter) was used to measure the electrical conductivity in these three solutions. The results showed that Na and Cl ions in the sea water significantly contributed to the electrical conductivity (Table 1).

   Due to the fact that total electrolyte concentration in solution affects the dissociation constant of different ions as observed in previous studies [8]. The ionic strengths (I)

of the three hydrolysis conditions were therefore calculated following the equation (1):

$$I = \frac{1}{2} \sum_{i=1}^{n} m_i z_i^2, \tag{1}$$

where n is the number of ions, m is the molality (mol/kg) of the ion i and z is the charge (-) of ion i

The calculated values are presented in Table 1. It is observed that the ionic strengths of the sea water that are significantly higher than those of the tap water and distilled water.

Table 1 Ionic strength (I) and electrical conductivity in deionized water, tap water and sea water

Water Types	I (mol/kg)	Electrical Conductivity (mS/cm)	Ions (g/kg)	
			Cl$^-$	Na$^+$
Distilled water	-	0.3	-	-
Tap water	0.005	2.4	0.35	-
Sea water	0.571	42.9	19.6	12.71

### 2. Chloride ion on hydrogen production

Based on the data obtained in Figure 2 and Table 2, the reaction using the distilled water with additional of NaOH to aluminium powder at room temperature shows the maximum hydrogen production and therefore results in the maximum hydrogen production yield (hydrogen production per 1 g aluminium).

Again, the maximum amount of hydrogen generated in the distilled water is the highest among others. Using the tap water as a hydrolysis reaction medium produces slightly lower amount as compared to that using the distilled water. These observed trend results are in line with those previously reported [9] - [10]. They are probably due to the fact that there is no chloride ion content in the distilled water whereas chloride ions are present in the tap water and the sea water. Fontana and Staehle reported that the chlorination process to treat the tap water for the control of microbes, its taste and odor will cause significant loss in aluminium and the formation of aluminium chloride [11] which indirectly slows down the hydrolysis reaction [9]. Therefore, the hydrogen production using the tap water and the sea water as reaction media is found to be the minimum.

Chloride ions show significant decreased in the hydrogen production by the hydrolyses of aluminium- based compositions. Two possible explanations are that the reduced activity of water at higher ions concentrations of the sea water decreased the available free water needed for the aluminium hydrolyses and that, as a result, the higher Cl ions concentrations would inhibit the hydrogen liberation.

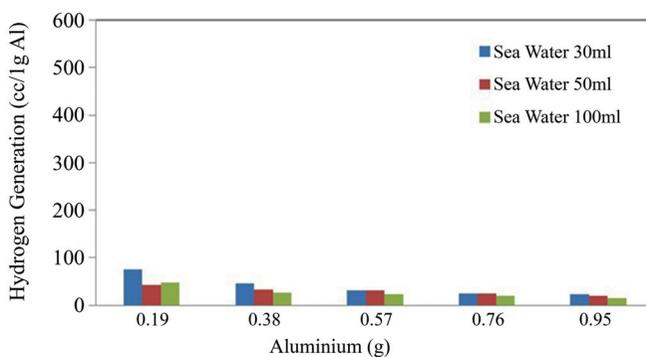**Table 2**   Hydrogen production from aluminium based composition at different types of water conditions

Al (g)	NaOH (g)	Water (mL)	I (mol/L)			Hydrogen Production (mL)		
			Distilled Water	Tap Water	Seawater	Distilled Water	Tap Water	Seawater
0.19	0	30	1.06	1.07	1.67	0	0	0
0.38	0	30	2.14	2.15	2.79	0	0	0
0.57	0	30	3.23	3.24	3.92	0	0	0
0.76	0	30	4.34	4.34	5.07	0	0	0
0.95	0	30	5.45	5.46	6.23	0	0	0
0.19	0.1	30	1.17	1.18	1.72	107.50±1.5	94.50±1.5	14.25±0.8
0.38	0.1	30	2.30	2.31	2.84	110.83±0.8	101.50±0.5	17.50±0.3
0.57	0.1	30	3.36	3.40	3.98	112.83±1.5	106.83±3.5	18.00±1.1
0.76	0.1	30	4.56	4.57	5.12	114.17±1.7	114.50±1.5	19.00±0.0
0.95	0.1	30	5.69	5.70	6.29	115.50±1.2	120.50±1.7	22.50±0.2
0.19	0.1	50	0.68	0.69	1.26	105.00±0.6	85.50±1.2	8.25±0.1
0.38	0.1	50	1.35	1.35	1.92	114.83±1.3	93.00±0.7	12.75±0.2
0.57	0.1	50	2.02	2.02	2.60	118.67±2.0	101.67±0.3	17.50±0.2
0.76	0.1	50	2.69	2.70	3.27	124.83±1.2	106.83±1.2	19.00±1.0
0.95	0.1	50	3.37	3.38	3.95	141.50±2.1	112.83±1.4	19.50±0.5
0.19	0.1	100	0.34	0.35	0.91	99.83±1.2	78.67±1.5	9.00±0.0
0.38	0.1	100	0.67	0.68	1.24	104.67±1.2	90.17±1.2	10.25±0.7
0.57	0.1	100	1.00	1.01	1.58	110.83±2.1	92.50±1.2	13.25±0.8
0.76	0.1	100	1.33	1.34	1.91	112.50±1.1	95.83±0.8	14.75±1.2
0.95	0.1	100	1.67	1.67	2.24	116.33±2.5	101.17±1.1	14.50±0.5
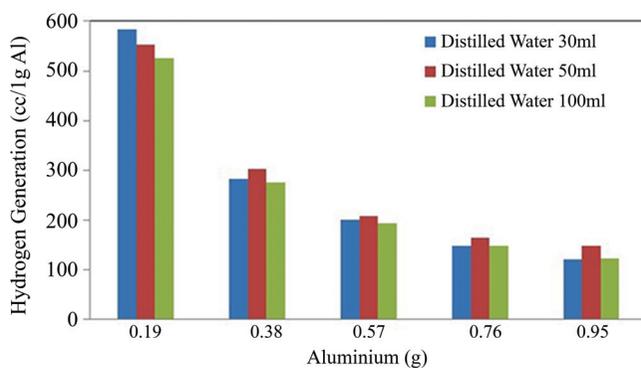
(a) Distilled Water
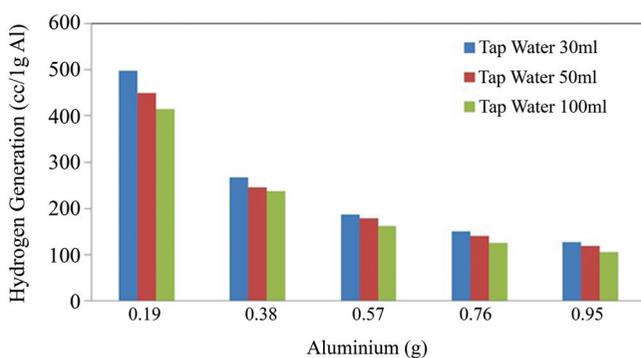
(b) Tap Water

(c) Sea Water

Figure 2   Effects of mass ratios of water to aluminium with 0.1 g NaOH on hydrogen generation from aluminium-based composition at different conditions

It is clear that aluminium exhibits the highest reactivity in the distilled water. However, the mass ratios of water to aluminium showed minor effects on the hydrogen production. The results show that the hydrogen generated in the distilled water, the tap water and the sea water at the difference amounts exhibited similar results. The maximum hydrogen generated is 583 ml as obtained from the reaction of 1-g aluminium in the 30-ml distilled water after reaction for 30 min. It is confirmed that the water quality significantly affects the hydrogen production from aluminium hydrolysis reaction.

In addition, the experimental results show that at the higher amount of aluminium to water, the hydrogen production increased This is because of the very low ratio of water to aluminium where most of the water molecules are adsorbed and thus no molecules remain to assist the water dissociation and too much water led to surface saturation without any space for hydrogen to form the cluster [12].

3. Thermodynamic properties: Hydroxide ions activity

The coefficients of the hydroxide ions activity at the employed reaction conditions have been calculated using the electrolyte Non-Random Two-Liquid (eNRTL) model in the Aspen Plus™ software which was used to analyze the activity coefficients of electrolyte a mixed solvent [13] - [14]. Chen, C.-C., Britt, H. I., Boston, J. I., and Evans, L. B. was originally proposed the Electrolyte NRTL model for aqueous electrolyte systems [15]. It was later extended to mixed solvent electrolyte systems. The expression for the activity coefficient can be derived for the local interactions as including the Pitzer-Debije-Hückel and the Born equation for the excess Gibbs energy. The Pitzer-Debije-Hückel formula is used to represent the long-range interaction contribution. The Born equation is used to account for the Gibbs energy of transfer of ionic species from the infinite dilution state in a mixed-solvent to the infinite dilution state in aqueous phase [16].

The results show that Cl ions in tap water and Na with Cl ions in the sea water significantly contributed to the ionic strength and decrease the coefficients of the hydroxide ions activity as presented in Figure 3. As results shown in the Figure 3, it shows the decreasing of ionic strength when increasing of the coefficients of the hydroxide ions activity as an exponential relationship. This means that, by introducing ions in the reaction mixtures, the hydroxide activity coefficients of the mixed solvent are significantly decreased and are also presents in Figure 3. Thus, the hydroxide ions activity is mainly influenced by the presence of the individual ions.
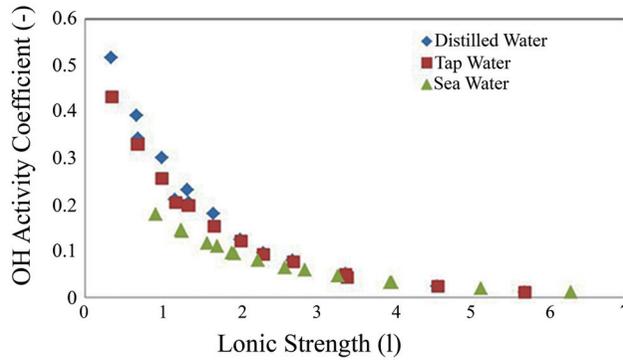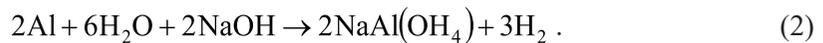
Figure 3   Effects of ions on the coefficients of the hydroxide ions activity

4. Hydrogen conversion efficiency

The coefficients of the hydroxide ions activity at the employed reaction conditions have been calculated using the electrolyte Non-Random Two-Liquid (eNRTL) model in the Aspen Plus™ software which is used to analyze the activity coefficients of electrolyte a mixed solvent [13] - [14]. Basically, the aluminium hydrolysis reaction related to the hydrogen yields can be expressed as follows equation (2):

$$2Al + 6H_2O + 2NaOH \rightarrow 2NaAl(OH_4) + 3H_2 . \tag{2}$$

The hydrogen conversion efficiency of aluminium based composition was calculated as follows equation (3):

$$E = \frac{Y}{(w/mw) \times 1.5 \times 24.45} \times 100, \tag{3}$$

where E is the conversion efficiency of hydrolysis reaction (%); Y is the actual hydrogen yields (unit-L); w is the mass fraction of aluminium in composites (%wt); mw is the molecular weight of aluminium (26.982 g/mol) and 24.45 (unit-L) is the standard volume of 1 mol hydrogen gas at 25 °C and 1 atm.

The results of the coefficients of the hydroxide ions activity on hydrogen conversion efficiency are presented in Figure 4. It is observed that the ionic strength of the solutions strongly increases along with the Cl ions and aluminium mass added, as expected. However, the results also show that the increase in the ionic strength simultaneously effects results in the coefficients of the lower hydroxide ions (OH-) activity (Figure 3). This is probably because the increase ionic strength results in the decrease coefficients of the ion activity (only the hydroxyl ions activity coefficient is shown). As a result, the activity of the hydroxyl

ions is lower and, therefore, decreases the catalytic effect of the NaOH. These corresponding results are according to the literature [6], [17] - [18] as evidenced that it is able to produce hydrogen gas from aluminium corrosion reaction with regeneration of hydroxyl ions.

In general, the hydrogen production efficiency is higher when the higher concentration of NaOH is employed. The hydroxyl ions seem acting as the catalyst for the hydrolysis of aluminium and promoting the hydrogen generation [19] as presented in Figure 4. This is probably because at higher NaOH concentration, there are more hydroxide ions in the solutions to dissolve the alumina film [20] - [21]. Therefore, the hydroxide ions reduce the protective oxide layer on the aluminium surface in an alkaline solutions. As the results, aluminium are readily dissolved in the alkaline solutions even at room temperature [22]. Thus, more NaOH additives in aluminium mixture will lead to the formation of higher amount of hydroxyl ions, consequently promote more hydrogen production (Table 2). These results are in line with the literature [23]
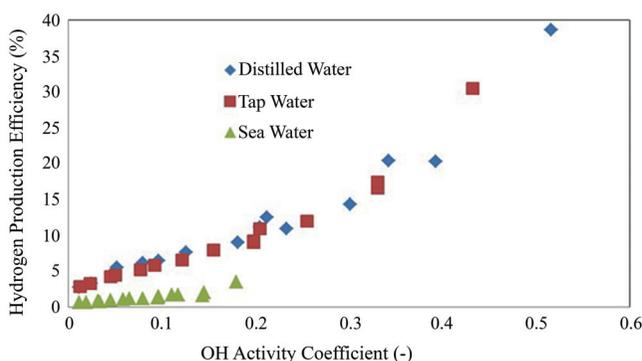

Figure 4   Effects of the coefficients of the hydroxide ions activity on hydrogen conversion efficiency

Conclusion

The hydrogen production process by hydrolysis reactions of aluminium with NaOH in different reaction was studied. Effects of the different Cl ions on the hydrogen production in different types of water were analyzed. It was expected to obtain the more understanding regarding how the Cl ions in different types of water affect the hydrolysis reaction at the different reactions for hydrogen production. The hydroxide ions activity at the employed reaction condition was calculated using the electrolyte Non-Random Two- Liquid (eNRTL) model in the Aspen Plus™ . It was observed that the ionic strength of the solutions strongly increase with the Cl ions and aluminium mass added. However, the simultaneous effect

results in the coefficients of the lower hydroxide ions (OH-) activity. This study demonstrates that the Cl ions in different types of water reduce the hydroxide ions (OH-) activity. However, the hydroxide ions enhance the production of hydrogen from the aluminium powder in water at room temperature. NaOH additives dissolves in water and produces hydroxyl ions and consequently promotes the hydrolysis reaction of aluminium and water. The efficiency of the hydrogen production is higher when the higher hydroxide ion is employed.

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