Hydrogen Production from Waste Aluminum at Different Temperatures, with LCA

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The recycling of waste metallic aluminum with high chemical exergy, which consumes a large quantity of electricity in the refining process, is insufficient. In particular, the so-called dross generated during the remelting process a part of recycling requires expensive treatment, particularly when the metallic concentration is less than 20%, before it can be landfilled. The purpose of this study is to produce hydrogen from waste aluminum sources, such as dross, using an aqueous solution of sodium hydroxide in a beaker and an autoclave. During the study, the effects of temperature of the aqueous solution on the rate of hydrogen generation are to be chiefly examined. The result obtained from an XRD analysis showed that the white product that precipitated during the experiments contained aluminum hydroxide, the rate of hydrogen generation significantly increased with the concentration of sodium hydroxide and temperature of the aqueous solution, and the activation energy was 68.4 kJ mol^{-1}. In the autoclave experiments, hydrogen is released quickly, along with an increase in the inner pressure to a minimum of 1.0 MPa and an increase in the temperature above 473 K. The results suggested a possibility of a new cost effective process of hydrogen production from waste aluminum along with the by prodution of sodium hydroxide. The life cycle assessment (LCA) of the proposed process for producing not only 1 kg of hydrogen but also 26 kg aluminum hydroxide from waste aluminum was carried out to assess the energy requirement and amount of carbon dioxide emissions. Results suggest that the energy requirement of our process is only 2% and the amount of carbon dioxide emissions is 4%, in comparison to a conventional method.

1. Introduction

According to the aluminum data of Japan,[1,2] the annual consumption of aluminum in Japan is 3.6 million ton (Mt), of which 2.4 Mt is imported and 1.2 Mt is domestically recycled from waste aluminum through a remelting treatment. Since the amount of aluminum recycling is insufficient, thus we have still not a small missing, unutilized aluminum.[3–7] The amount of missing reaches 0.80 Mt in processes such as beverage can production and 0.77 Mt in market places. Furthermore, even if we successfully collect the used aluminum products, 0.30 Mt of dross, containing metallic aluminum as a byproduct, is released into the atmosphere during the remelting process. Metallic aluminum has a high potential with a large chemical exergy. Therefore, the purpose of this study is to understand the manner in which aluminum can be utilized in the process with the help of alkaline solutions to chiefly evaluate the effects of temperature of NaOH aqueous solutions on the reaction rate,[8,9] explore the possibility of pressurized hydrogen generation, and perform a life cycle assessment (LCA) of this process.

2. Experimental Procedures

A powdered aluminum reagent (Kojundo Chemical Laboratory Co., Ltd.) was prepared, having a purity mass% of 99% and a particle size of 180–425 µm. Aqueous solutions of NaOH were prepared from a NaOH aqueous solution of 5 mol dm^{-3} (Kishida Chemical Co., Ltd.). The reactor in which the reagent and the diluted solutions were mixed is schematically represented in Fig. 1. Table 1 lists the experimental conditions used in this study W_{Al}: the weight of
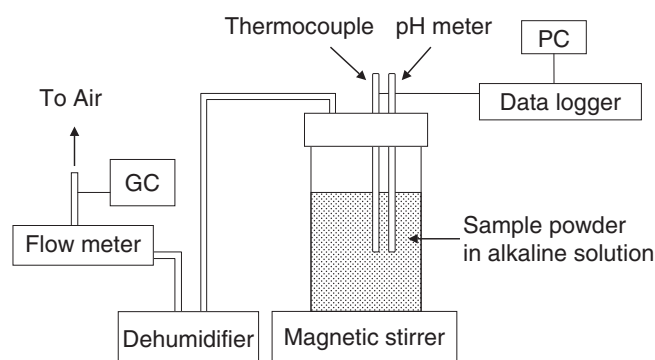

Fig. 1 Schematic diagram of the experimental apparatus.

Table 1 Experimental conditions for validating the reaction between aluminum and NaOH aqueous solutions and evaluating the effect of temperature of NaOH aqueous solutions on the reaction rate.

Experiment	W_{Al}/M_{Al} (mol)	C_{NaOH} (mol·m^3)	V_{NaOH} (m^3)	Molar ratio Al/NaOH	T_0 (K)
a-1	0.25	500	0.0005	1.00	309.5
b-1	0.03	500	0.0005	0.12	291.2
b-2	0.03	500	0.0005	0.12	313.2
b-3	0.03	500	0.0005	0.12	323.2
b-4	0.03	500	0.0005	0.12	333.2

aluminum reagent, M_{Al}: the molar mass of aluminum, C_{NaOH}: the concentration of NaOH aqueous solutions, V_{NaOH}: the volume of NaOH aqueous solutions, T_0: the initial temperature of NaOH aqueous solutions. To promote the chemical reaction, the mixture was continuously agitated using an RCN-3 magnetic stirrer (Tokyo Rikakikai Co., Ltd.). The gas generated during the experiments was passed through a

dehumidifier and then channeled to a flow meter for volume measurement. A fraction of the generated gas was collected and subjected to a composition analysis by gas chromatography using a GL Sciences GC-323 chromatograph. Throughout the experiments, the temperature and pH of the NaOH solution were recorded using a TES-1380 pH/temperature meter (TES Electrical Electronic Corp.) at regular intervals between 10 and 30 s. For the phase identification of the reaction products, X-ray diffractometry (XRD) was carried out using a Rigaku RINT-1500 diffractometer equipped with a rotational Cu anode and a graphite monochromator. To explore the possibility of generating pressurized hydrogen, an autoclave with a safety valve was prepared with the following specifications: an inner volume of $1\,dm^3$ and a maximum pressure of $10\,MPa$. In the experiments, a data acquisition controller monitored the temperature of the aqueous solution and the changes that occurred in the pressure with time.

3. Results and Discussions

3.1 Description of reaction between aluminum and NaOH aqueous solution

In the present investigation, the generation of gas bubbles was detected immediately after the aluminum powder and NaOH aqueous solution were mixed in the reactor. The generated gas was identified as high purity hydrogen, the purity of which was higher than 99.9 mol%. As shown in Fig. 2, the generation rate of H_2 monotonically increased with reaction time up to 540 s, and beyond this time, it started to decrease. With the generation of H_2 gas, the temperature of the NaOH aqueous solution rose quickly with the reaction time and reached its maximum at 338 K within 1200 s after the reaction terminated in the reactor. On the other hand, the pH of the NaOH solution gradually decreased with the reaction, implying the formation of $Al(OH)_4^-$ ions in the solution. When the reaction time exceeded beyond 1200 s, the temperature of the NaOH solution began to decrease. However, the pH of the solution showed a gradual increase beyond this point. The pH increase was found to be preceded by precipitation that occurred at $t \sim 1000\,s$, and the precipitation compound was later identified to contain aluminum
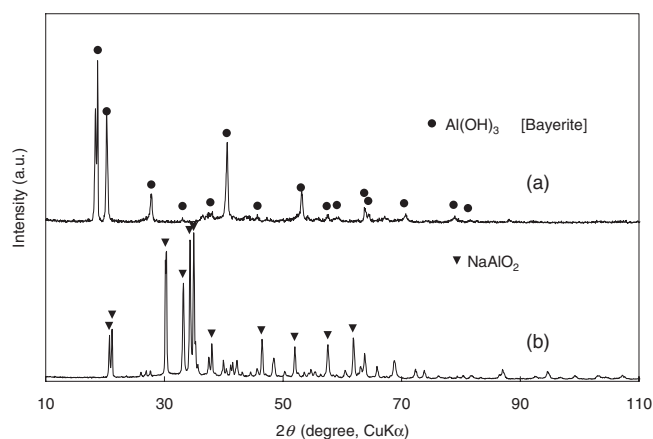
Fig. 3　XRD spectra of the reaction products: (a) the precipitate formed during processing and (b) the crystallite obtained by a rapid dehydration of the filtrate.

hydroxides. Therefore, the variation in the pH after 1200 s is attributable to the decomposition of $Al(OH)_4^-$ ions into aluminum hydroxide and OH^- ions.

3.2 XRD analysis of reaction products

In the present investigation, it was found that precipitation occurred in two stages. The first precipitation was occurred at the time of reaction between aluminum powders and NaOH aqueous solutions, which occurred in the reactor. Upon completion of this reaction, the precipitate was separated by filtration and examined by XRD. The XRD spectrum of the precipitate, shown in Fig. 3(a), was found to correspond essentially to the spectrum of bayerite, an $Al(OH)_3$ polymorph, provided in PDF No. 20-0011. However, a careful examination revealed that the intensity of the observed reflections did not correspond to the relative intensity listed in the PDF datum, and the reflection occurring at $2\theta \sim 18.8°$ was likely to split into two. These results imply that a partial structural modification occurred in the precipitate; thus, a part of the structure might get transformed into another type of polymorphic structure. The second precipitation occurred in the filtered solutions when they were allowed to stand for 86.4 ks under an ambient temperature. The XRD spectrum for the first and second precipitates appeared similar but they differed slightly in their relative intensities of reflections. This observation suggests the possibility that the crystal structure of the second precipitate is similar to that of the first one although the chances of both these precipitates to have identical atomic arrangements are unlikely. Consequently, the precipitates formed in the NaOH aqueous solutions were expected to have a crystal structure close to that of bayerite and to have the composition of $Al(OH)_3$.

In this study, an attempt was also made to obtain a crystalline phase of $NaAlO_2$ by rapid dehydration of the filtered aqueous solution. The filtrate was heated in a muffle furnace set at 473 K until water in the filtrate was completely evaporated; this process usually took 1.8 to 3.6 ks, depending on the volume of the filtrate. An XRD of the residual product was performed and the spectrum shown in Fig. 3(b) was obtained. The major reflections in the spectrum were found to be indexible for $NaAlO_2$ though some other low-intensity
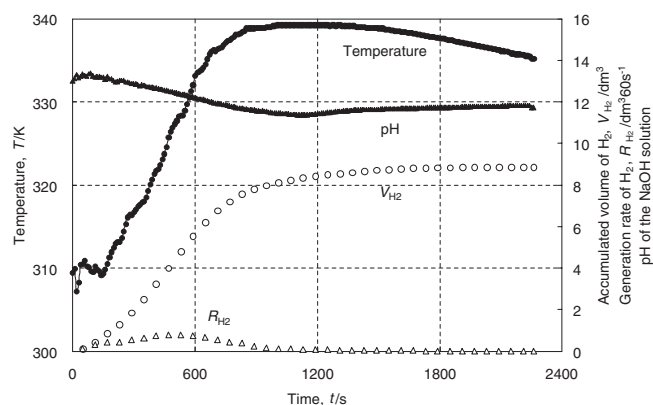

Fig. 2　Time evolution of the temperature and pH of the NaOH aqueous solution, the accumulated volume and the generation rate of hydrogen. (Experiment a-1)
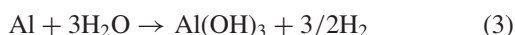
peaks were also included. $NaAlO_2$ is a highly deliquescent compound that reacts easily with moisture present in the air. Therefore, the small peaks might be attributable to a hydrated sodium aluminate that is possibly formed while the specimen was being prepared for XRD measurements.

3.3 Determination of the reaction equations

Based on the results of the gas chromatography and XRD analysis, the possible reactions between aluminum and NaOH aqueous solutions are shown as follows.

$$Al + 3H_2O + NaOH \rightarrow NaAl(OH)_4 + 3/2H_2 \qquad (1)$$

$$NaAl(OH)_4 \rightarrow Al(OH)_3 + NaOH \qquad (2)$$
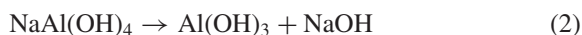
The overall reaction can thus be summed up as follows.

$$Al + 3H_2O \rightarrow Al(OH)_3 + 3/2H_2 \qquad (3)$$

The degree of reaction, f, for the above reaction is accordingly defined in terms of the volume of generated hydrogen, V_{H_2}:

$$f = \frac{V_{H_2}/1.5}{22.4 \times 10^{-3}(T_a/273)} \left(\frac{W_{Al}}{M_{Al}}\right)^{-1} \times 100$$
$$= \frac{8.13 \times 10^5 M_{Al} V_{H_2}}{W_{Al} T_a}, \qquad (4)$$

where W_{Al} and M_{Al} are the weight of aluminum reagents and the molar mass of aluminum, respectively, and T_a is the ambient temperature measured in Kelvin. In the present investigation, it was also revealed that sodium aluminate can be obtained through the dehydration of $NaAl(OH)_4$, which is expressed by the following equation.

$$NaAl(OH)_4 \rightarrow NaAlO_2 + 2H_2O \qquad (5)$$
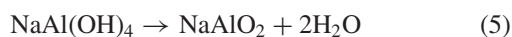
3.4 Effect of temperature

The effect of temperature on the reaction rate was examined by the following procedure. Prior to the experiment, the NaOH aqueous solutions were initially heated to appointed temperatures between 291 and 333 K. Once the temperature was equilibrated, the experiment was terminated by adding aluminum reagent to the preheated NaOH aqueous solution. The results shown in Fig. 4 clearly indicate that the
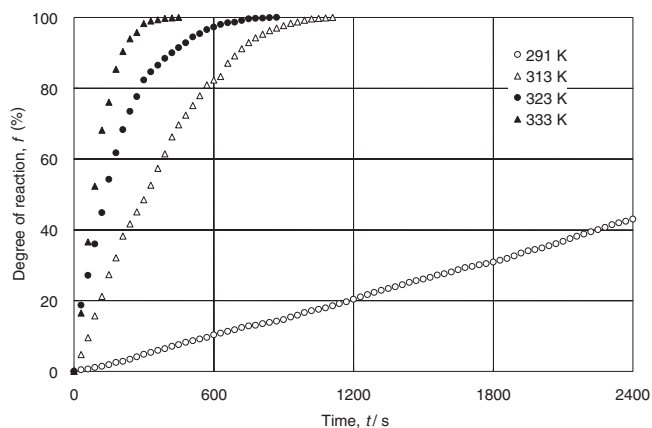

Fig. 4 Time evolution of the degree of the reaction at different temperatures of the NaOH aqueous solutions. (Experiments b-1 through b-4)

reaction rate is significantly influenced by the initial temperature of NaOH solutions. The reaction was completed within 1200 s when the initial temperature was set at 313 K or above, while the generation of H_2 continued at 291 K even after 2400 s. In order to calculate the rate constants for the reaction, the obtained results were analyzed using the shrinking core model.[10] Since the chemical reaction expressed by eq. (3) is a first order reaction, the molar reaction rate of aluminum can be described as follows:

$$-\frac{d}{dt}\left(\frac{4}{3}\pi \frac{\rho_{Al}}{M_{Al}} r_c^3\right) = 4\pi r_c^2 \frac{dn_{Al}}{dt}, \qquad (6)$$

where r_c is the radius of the unreacted core, ρ_{Al} is the density of aluminum, and n_{Al} is the amount of reacted aluminum. The reaction rate, dn_{Al}/dt, is also given by the following equation:

$$\frac{dn_{Al}}{dt} = -\frac{C_{NaOH}}{1/k_c + 1/k_s}, \qquad (7)$$

where C_{NaOH} is the concentration of the NaOH aqueous solution, k_s and k_c are the surface reaction rate constant and the mass transfer constant in laminar boundary layers, respectively. Equation (8), the reaction rate equation in terms of time variation of r_c, can be obtained by combining eqs. (6) and (7).

$$-\frac{dr_c}{dt} = \frac{C_{NaOH}M_{Al}}{\rho_{Al}(1/k_c + 1/k_s)}, \qquad (8)$$

The following relationship is derived from the definition for f.

$$r_c/r_0 = (1-f)^{1/3} \qquad (9a)$$

For a small value of r_c, k_c can be approximated as follows:[11]

$$k_c = \frac{r_0}{r_c} k_{c0}, \qquad (9b)$$

where r_0 is the initial radius of the reacting core and k_{c0} is the initial value of k_c. On substituting eqs. (9a) and (9b) into eq. (8), and on integrating it, eq. (10) is obtained.

$$\frac{1}{k_s} r_0\{1-(1-f)^{1/3}\} + \frac{1}{2k_{c0}} r_0\{1-(1-f)^{2/3}\}$$
$$= \frac{C_{NaOH}M_{Al}}{\rho_{Al}} t \qquad (10)$$

On assuming that the NaOH aqueous solution is sufficiently stirred, the value of k_{c0} can be set as infinite. In this case, eq. (10) can be written as

$$r_0\{1-(1-f)^{1/3}\} = \left(\frac{C_{NaOH}M_{Al}}{\rho_{Al}}t\right)k_s. \qquad (11)$$

The above equation indicates that the slopes of the linear plots of $r_0\{1-(1-f)^{1/3}\}$ against $(C_{NaOH}M_{Al}t/\rho_{Al})$ yield the reaction rate constants k_s. Equation (11) was applied to the experimental data presented in Fig. 4 and k_s at each temperature was obtained as follows: 1.97×10^{-6} m·s^{-1} at 291 K, 2.28×10^{-5} m·s^{-1} at 313 K, 3.49×10^{-5} m·s^{-1} at 323 K, and 6.93×10^{-5} m·s^{-1} at 333 K (see Fig. 5). These results are summarized in an Arrhenius plot, as shown in Fig. 6, and a good fit was obtained. The apparent activation energy for the reaction, E_a, was consequently determined to be 68.4 kJ mol^{-1}.
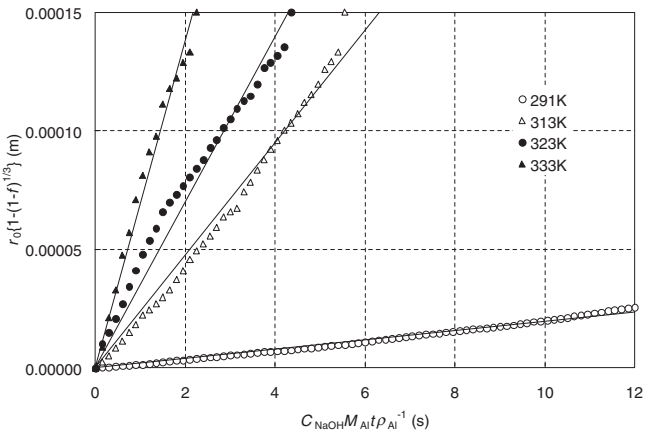
Fig. 5   Linear plot used for the evaluation of the surface reaction rate constants, k_s. The lines in the figure are obtained by a linear regression analysis of the experimental data, and their slope provides the reaction rate constant at each temperature. The details of the plot can be found in eq. (11) in the text.
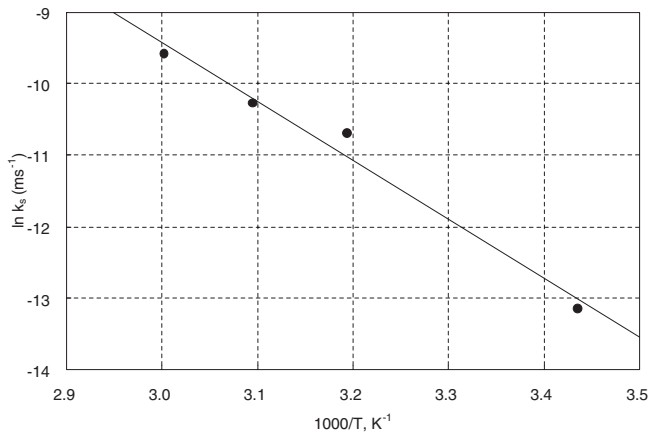

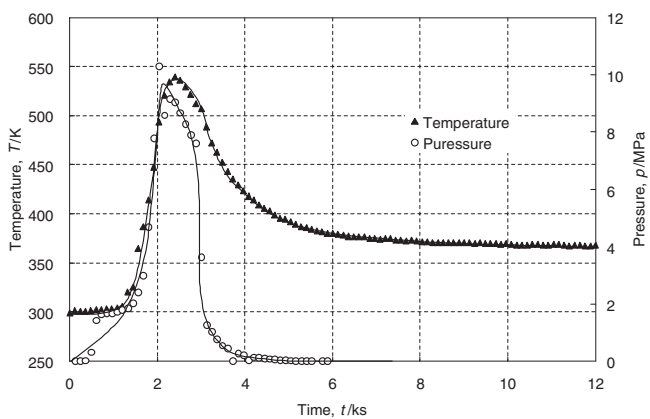Fig. 6   Arrhenius plot of the surface reaction rate constants.

Fig. 7   Time evolution of the temperature and pressure in the closed reactor.

3.5   Generation of pressurized hydrogen

Figure 7 shows the changes in temperature and pressure with time. The experimental conditions were as follows: (1) dross was used as the sample, containing 40 mass% of metallic aluminum, (2) the sample had 1 mole of aluminum, (3) the concentration of NaOH was $600\,\mathrm{mol\cdot m^{-3}}$ and

$0.0003\,\mathrm{m^3}$, (4) the initial temperature was 299 K. The temperature gradually increased with an increase in the pressure. Finally, when the temperature reached 523 K, pressure reached 10 MPa and the safety valve was functional. These results show that the reaction of hydrogen generation is definitely not limited by pressure under the experimental conditions. A very large equilibrium constant of the order of 10^{74} supported this experimental result. The phenomenon of generation of high-pressure hydrogen appears very attractive from the viewpoint of hydrogen purification by placing hydrogen storage alloys, etc., at the down stream.

4.   Systematic Evaluation Based on Life Cycle Assessment (LCA)

To determine the effect of the proposed process on the energy requirement and amount of carbon dioxide emissions, we performed a life cycle assessment (LCA) along with the conventional process. LCA is a well-known method used for assessing the environmental aspects and potential impacts associated with a byproduct. The assessment is performed by (a) compiling a life cycle inventory (LCI) of relevant inputs and outputs of a product system; (b) evaluating the potential environmental impacts associated with the inputs and outputs; (c) interpreting the results of the inventory analysis and impact assessment phases in relation to the objectives of this study. Our proposed process can be expressed by eq. (12), while the conventional process is expressed by eqs. (13a) and (13b).

$$\text{Waste Al} + 3\text{H}_2\text{O} \rightarrow 3/2\text{H}_2 + \text{Al(OH)}_3 + \text{Residue} \quad (12)$$

$$\text{CH}_4 + 2\text{H}_2\text{O} \rightarrow 4\text{H}_2 + \text{CO}_2 \quad (13a)$$

$$\text{Al}_2\text{O}_3 + 3\text{H}_2\text{O} \rightarrow 2\text{Al(OH)}_3 \quad (13b)$$
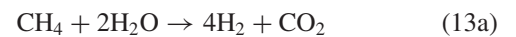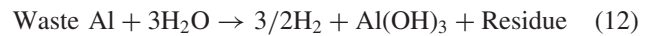
Figure 8 shows the system boundaries of the two methods for LCA. The proposed process (A) shows co-production of hydrogen and aluminum hydroxide from waste aluminum and deionized water, is assumed that produced by ion exchange of water. Here, the residual generated by the proposed process can be landfilled. The conventional hydrogen production (B1) means so-called steam reforming of
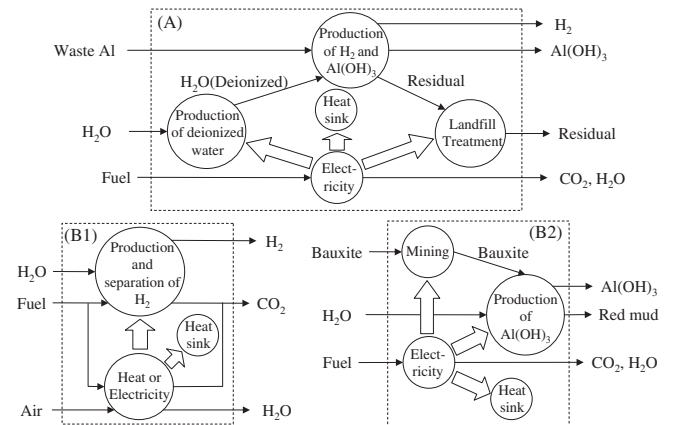

Fig. 8   System boundaries of the two methods for LCA. (A); The proposed process, (B1); The conventional hydrogen production based on steam reforming, (B2); The conventional aluminum hydroxide production by the 1st-stage of Bayer process.

Table 2 Energy requirement and amount of carbon dioxide emissions for producing 1 kg hydrogen and also 26 kg aluminum hydroxide from waste aluminum.

Process	Energy requirement (MJ)	Amount of carbon dioxide emissions (kg-CO$_2$)
Conventional	418	18.2
Proposed	8.3	0.73
Ratio (%) (Proposed/Conventional $\times$ 100)	2.0	4.0

natural gas, which is the most common method of hydrogen production. Steam, at a temperature of 973–1373 K, is mixed with methane gas and introduced into a fixed bed reactor of catalyst.[12,13] The conventional aluminum hydroxide production (B2) corresponds to the 1st-stage of Bayer process. In the process, sodium hydroxide dissolves the aluminum component of bauxite ore to remove impurities from the solution and precipitates aluminum hydroxide.[14]

In the LCA, the following assumptions were made:
(a) Concentration of waste aluminum used was 15 mass%.
(b) Latent environmental burden of waste aluminum was zero.
(c) Deionized water was used for the hydrogen production.
(d) Construction material and scrappings of both buildings and machines were not included in the proposed process because this process was under development.
(e) Waste aluminum and residue was transported to a plant for hydrogen production, and reacted waste aluminum to be used in the landfill was not considered.
(f) In evaluating the conventional process, not only was the production of Al(OH)$_3$ based on the so-called Bayer process considered, but the hydrogen production based on steam reforming of natural gas was also taken into consideration.

Table 2 lists the energy requirement and the amount of carbon dioxide emissions for producing 1.0 kg hydrogen and 26 kg Al(OH)$_3$. The LCA evaluation of the conventional process concluded that the required energy was 418 MJ and the amount of carbon dioxide emissions was 18.2 kg. In contrast, the proposed process showed remarkably small values; the required energy was 8.3 MJ and amount of carbon dioxide emissions was 0.73 kg. These values respectively correspond to only 2.0 and 4.0% of those obtained through the conventional process. The results show that the proposed process has an extremely mild effect on global warming. Appendix gives key information used for LCA.

5. Conclusions

A new technology was proposed for hydrogen production using aluminum and sodium hydroxide solution; this technology was studied both experimentally and theoretically.
(1) In the experiments, the generation of hydrogen and the formation of aluminum hydroxide were observed. We confirmed that the overall reaction of this phenomena, if all the aluminum hydroxide is precipitated, is expressed by: Al + 3H$_2$O → Al(OH)$_3$ + 3/2H$_2$.
(2) Temperature dependence on hydrogen generation was

expressed by the following equation.

$$k_s = 4.29 \times 10^6 \exp\left(\frac{-68.4}{RT}\right) \qquad (14)$$

Using the above equation, the activation energy was calculated as 68.4 kJ mol^{-1}.
(3) The autoclave experiments showed that the proposed process has a possibility of generating hydrogen at a pressure higher than at least 1.0 MPa.
(4) A systematic evaluation by LCA showed that the proposed process was only 2% in terms of energy requirement and 4% in terms of carbon dioxide emission in comparison to the conventional processes used for generation of hydrogen and formation of aluminum hydroxide.

The basic data that we obtained will be effective for designing a new furnace of waste aluminum treatment together with the production of valuable materials such as hydrogen. In fact, such a furnace that produces hydrogen at a scale of 45 Nm^3/day is currently under construction.

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Appendix appear on next page.

Name	Comments	Data Characterization	Process Description	Sources
241I Aluminium hydroxyde (Al(OH)₃): Productio	$A_2O_3 + 3H_2O \rightarrow 2Al(OH)_3$		Production of aluminium hydroxide (Al(OH)₃)	BUWAL 132 (Bundesamt fur Umwelt, Wald und Landschaft) Bern, Februar 1991 Page A22.
241I Aluminium oxide (Al₂O₃): Production	Red mud obtained by the production of aluminium oxide is treated as a substance with soil characteristics and is not included in the balance. Red mud is deposited in immediate proximity of the bauxite mine, covered with soil and then recultivated with flora specific for the site. No information exists about negative impacts of red mud landfills.			Swiss Federal Office of Environment, Forests and Landscape (FOEFL or BUWAL) Environmental Series No. 132. Bern, February 1991. Page A22.
241I Hydrogen (H₂, steam cracking of natural gas): Production	Process route: $CH_4 + 2H_2O \rightarrow CO_2 + 4H_2$	Stoechiometric reaction for mass input and output and energy from bibliography.	Production of Hydrogen by Steam Cracking of Natural Gas	Ullmann's Encyclopedia of Industrial Chemistry 5th edition, Vol A21, VCH Weinheim 1992.
241I Water (De-ionized): Production			Production of Distilled Water by exchange of ion	Laboratorium fur Energiesysteme ETH (Eidgen·sische Technische Hochschule Zurich) Zurich, 1996 Teil 3: Basismaterialieren Page: 83 primary source: Pers·liche Mitteilung von Herrn Kamberger von der Firma SULZER THERMTEC, CH-8401 Winterthur, Februar 1993.
401 Electricity (Japan, 1996): Production. 1	Breakdown of energy sources in Japan: – Coal: 14.39% – Lignite: 0% – Fuel Oil: 20.86% – Natural Gas: 20.07% – Nuclear: 29.86% – Hydro: 8.84% (assumption: hydro + wind + waves + tide) – Process Gas: 3.63% (coke oven gas + blast furnace gas) – Free Electricity: 2.37% (geothermal, solar, biomass and animal products, industrial waste, municipal waste, non-specified assumed being impact free) Import: 0% Distribution losses: 3.64% Efficiencies: – Coal: 39.2975% – Lignite: 32.3% – Oil: 38.5% – Natural Gas: 33% – Nuclear: 33% – Hydro: 90%	Representative of average production in Japan (1996).	Production of Electricity in Japan: production of fuels and combustion in power plants	1) For production and combustion of coal, lignite, heavy fuel oil, natural gas, process gas: Laboratorium fur Energiesysteme ETH, Zurich, 1996 2) for breakdown efficiencies: Energy statistics of OECD countries 1995–1996 International energy agency Paris, 1999
900 Aluminium (Al): Landfill	Net calorific value: 31 MJ/kg		Landfill of Aluminium (Al)	Buwal 250 (Bundesamt fur Umwelt, Wald und Landschaft) Oekoinventare fur Verpackung: Band II Bern, 1996 page 453 primary source: Doka G., Huber F., Labhardt A., Menard M., Zimmermann P., Oekoinventare von Entsorgungprozessen-Grundlagen zur Integration der Entsorgung in Oekobilanzen, ESU-Reihe 1/96; Institut fur Energietecknik, Gruppe Energie-Stoffe-Umwelt, ETH Zurich, 1996.