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Hydrogen Generation by Electrical Discharge Through Metal / Water System

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Abstract — Reactive metals like aluminum generate hydrogen gas when it reacts with water. Aluminum, despite its high chemical affinity with water, cannot continue the reaction due to the passive oxide layers formed on its surface. When the reaction is assisted by electrical energy dissipation in the form of discharge, the reaction is more likely to be sustained. In this report, some preliminary experimental results are presented regarding the hydrogen generation based on this scheme.

1. Introduction

Generation or storage of hydrogen molecules for the use in vehicular applications, as seen in fuel cells, imposes a very important but formidable technological challenge to solve. Traditional methods include a reformer which utilizes a hydrocarbon fuel, storage in the form of liquid or high pressured gas or of metal hydride. Technologically sound as each method may appear there are still many technical problems to be solved, particularly reducing size or weight of the generation or storage device, fast utilization, purification of the product gas.

The hydrogen generation based on electrical discharge through reactive metal/water system has been reported for such applications as a fast hydrogen generator[1)] or a vehicular fuel source[2)]. Most commonly used metal is aluminum. It is true that the cost of the reactive metals is formidably high, about 5 times higher than that of fossil fuel on the basis of equal energy output. However, methods such as recycling of the metal oxide the reaction product or utilization of the reaction heat could reduce the overall hydrogen production cost improving the possibility of practical applications of the method.

These methods are in principle very simple. It is well known that although aluminum has high oxidation potential, the passive oxide film on the surface prevents a further oxidation. By raising temperature or adding chemicals such as NaOH the oxidation reaction proceeds. The electrical discharge is, in a sense, a replacement of the chemical effect. The method described in reference 1 (from now called method I) induces a very fast chemical reaction, less than 1 msec, in a premixed aluminum particle/water mixture by sending a very high power electrical pulse, larger than 1 MW, through it. The electrical discharge in the pulse form provides a "hot" reaction zone which, in the beginning, is confined to a local region but spreads into the entire reaction zone with time. The method proposed in reference 2 (from now called method II) induces a steady state electrical discharge between two electrodes placed in water. One of the metal electrodes is made of aluminum wire which can be easily consumed during the discharge to engage the combustion with water. The major technical problem to be solved with these electrical methods is to improve the electrical energy efficiency defined as the ratio of the metal consumed (measured in total enthalpy change in the metal/water reaction) to the electrical energy input. For the fast generation method[1)] in which the aluminum fuel is premixed with water, the energy efficiency already exceeds 100. Conceptually attractive as it appears, the method II is still in its infancy in terms of optimization. Although the basic concept is presented in the reference, no quantitative results have been published.

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The difficulty with the method II is ascribed to that a stable reaction zone is not easily established. The steady reaction is materialized when the combustion reaction between the reactive metal and water is sustained, the major driving force for the reaction coming from the chemical exothermicity. Here the electrical discharge should play a minor role in helping sustain the reaction. Thus the energy efficiency should be very low when the discharge alone becomes the sole source of driving the metal/water reaction. The results of our recent study[1] showed that a critical condition for igniting the reaction is the focusing of the electrical energy dissipation at the tip of the wire electrode. This means that the side surface of the wire should not be exposed to water so that electrical energy is not wasted through electrolysis. Under this condition the tip portion is activated and dispersed into water engaging the reaction with it. The initiation of the reaction, besides the insulation of the wire surface, also necessitates the adequate voltage shape. The voltage in the beginning of the discharge may have to rise to a few kV to jump the dielectric gap between the electrodes if the cathode wire keeps a certain distance from the anode. Once the conduction channel is established, the voltage has to switch to a low value, typically under 500V, while the current mode switches to a high value, in the range of a few amperes. Then the joule heat dissipation would be large enough to activate the tip and to start the reaction. After the initiation of the reaction, in order for the reaction to be sustained there should be a timely supply of the fuel (the metal wire) and the oxidant (water). The metal wire needs to be fed by an electric motor to maintain a small gap (typically 2~3 mm). To supply enough amount of water into the combustion zone a vigorous agitation of the medium around the reaction zone is necessary.

Optimizing all these variables and establishing the sustained reaction zone requires systematic investigations on the effects and mutual correlations of the involved variables. Among these variables we only attempted to adjust the voltage (or current) shape and the insulation of the metal wire surface. With these conditions adjusted we measured the volume of the generated hydrogen as the function of the dissipated electrical energy. In this report we present some preliminary results regarding the hydrogen generation.

2. Experimental Methods

The experimental setup shown in Fig. 1 basically consists of three parts; the DC power source, the reaction chamber and the measuring device. The DC power source provides two modes of power: high voltage/low current (0~6.5 kV, 0~100 mA) and low voltage/high current (0~750 V, 0~10 A). The high voltage source causes the electrical breakdown between the electrodes, which is about 2~3 mm apart in low conductivity water (>18 MΩ-cm). An extra RLC components (R≈10 Ω, L=7 mH, C=20 nF) were added to the discharge circuit. As the cathode, aluminum wire (AWG, ER4043 and ER5336 type, 0.8, 1.2, 2.0 mm dia.) was used whereas a plane electrode made of aluminum, stainless steel or nickel was used as the anode. The breakdown establishes a high electrical conduction path so that the electrical energy from the low voltage/high current mode activates the fuel metal electrode. The tip of the cathode wire was encapsulated by a ceramic housing as shown in Fig. 2. Initially the inside of the housing is filled with water. But a small amount of gas generated by a short electrolysis replaces the water making the side of the wire electrically insulated.

After the electrical discharge the product gas was determined by pressure measurement, gas chromatography analysis (HP 5890 II) or volume measurements based on water replacement. The voltage

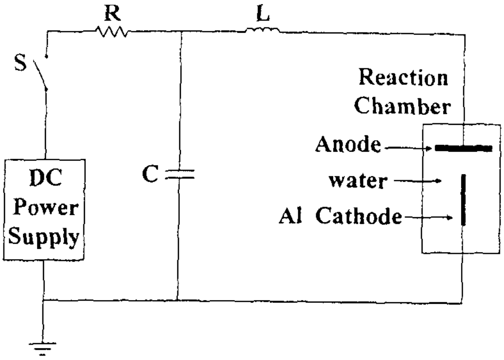

Fig. 1. The electrical circuit for inducing reactive metal/water reaction.

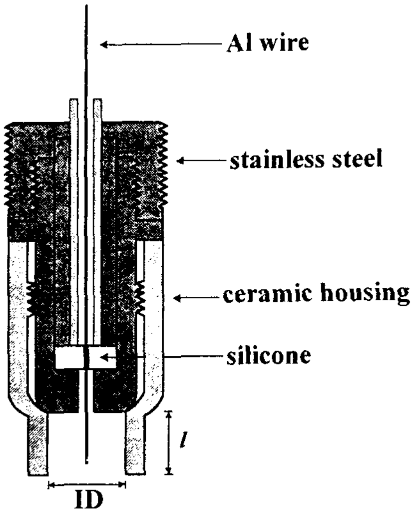

Fig. 2. The details of the cathode tip which is encapsulated by the ceramic housing. The dimensions of the ceramic housing are represented by the inside diameter ID and the length *l* (see Table 1).

across the electrodes and the discharge current were measured using the usual voltage divider and hall effect current probe, respectively. A DC motor was attached to the aluminum wire feeder to control the feeding rate. The water was deionized by three-step water purifier incorporating ion-exchange and microfilter columns.

3. Results and Discussions

3-1. Voltage-current Profile

Over-all effects of RLC addition to the circuit and encapsulating the aluminum wire by the ceramic housing helped stabilize the high current discharge. The duration of the high current mode usually lasts about 5 seconds (Fig. 3), indicating the dispersed metal particles somewhat increases the conductivity between the gap. In this experiment the cathode wire actually hits the anode fairly fast in order to start the discharge, there is no initial surge of the voltage to jump the dielectric gap. It is not certain yet that the metal instantly engage the reaction right after being dispersed. Since the wire feeding is controlled by a DC motor whose speed is preset independent of the discharge current, the discharge eventually comes to halt due to the mismatch of the feeding rate to the maintenance of the adequate conductivity between the electrodes. The power and the total dissipated electrical energy is shown in Fig. 4, reflecting the step increase of the total energy due to the alternation of the two kinds of the power modes.

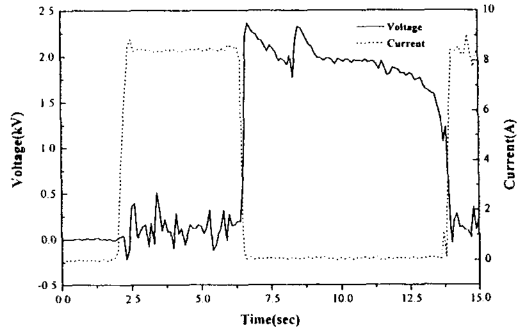

Fig. 3. The voltage and current profiles of the electrical discharge through aluminum wire and anode placed in water (Test 2 of Table 1). The time zero does not necessarily coincide with the closing time of the discharge circuit. The first two seconds during which the voltage trace shows a plateau indicates the cathode wire feeding time.

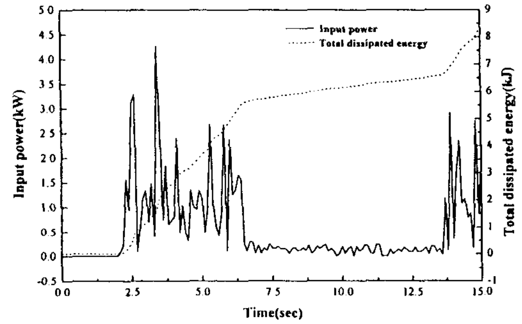

Fig. 4. The power and the total dissipated energy of the electrical discharge shown in Fig. 3.

3-2. Hydrogen Measurements

The total volume of the generated hydrogen was measured using water replacement method. The results showed that the efficiency of the generation depends on the shape of the ceramic housing (see Fig. 2). The results of hydrogen measurements and the dimension of the two kinds of ceramic housing used are summarized in Table 1. The housing with smaller ID and longer height *l* showed the efficiency more than two times higher than the case of larger ID and shorter height. Comparing the amount of the gen-

Table 1. Hydrogen gas produced by the discharge method. The amount depends on the shape of the ceramic housing.

	Test 1	Test 2
Dimensions of the ceramic housing (ID in mm/*l* in mm)	8/7.7	6.3/21
The weight of Al wire consumed (mg)	102	90.6
The electrical energy input (kJ)	18.9	13.4
The amount of the hydrogen gas generated (m*l*)	30.7	66.7

erated hydrogen to the weight of aluminum proves that more than 50% of the consumed metal underwent the chemical reaction with water.

$$2Al + 6H_2O \longrightarrow 3H_2 + 2Al(OH)_3$$

The results of the gas analysis using the gas chromatography method showed that the product gas does not contain any significant amount of oxygen gas. Hydrogen was the only gas product detected. If the electrical discharge induces any kind of water dissociation through electrolysis or plasmolysis[4], oxygen should be generated. Or there is some possibility existing that the generated oxygen could recombine with hydrogen molecule nearby to reform water. But this possibility may be remote due to the presence of abundant water molecules along the discharge channel.

The effect of the shape of the ceramic housing on the efficiency could be ascribed to the difference in the portion of the wire activated by the high current discharge. The electrical power curve seen in Fig. 4 shows that each high current discharge delivers the power over 2 kW which could easily melt or vaporize a few hundred milligram of the metal. The wire of 1.2 mm dia and 10 cm long weighs 320 mg. Therefore, the electrical power in one discharge can activate all the aluminum inside the housing, making them readily react with water. The longer housing happens to contain more aluminum. This fact also implies that the feeding rate should be increased in the such a speed as to maintain the narrow gap. The speed of the DC motor was far below the consumption rate of the wire. Besides the role of insulating the wire, the ceramic housing also provides a better combustion region which prevents the large mass of water (heat sink) from entering the region.

However, the fact that over 10 kJ electrical energy is used for generating less than 100 m*l* of hydrogen implies that the activation of the wire material solely depended on the electrical energy. It also implies that no sustained reaction is ever established along the discharge path. The chemical heat from the metal/water reaction was not utilized for further activation of the wire.

The reason for the failure of forming the stabilized reaction zone is also partly due to the inadequate provision of both the fuels, Al wire and the oxidant, water molecules. When the metal/water reaction is initiated, the generation of hydrogen molecules and aluminum oxide could prevent a ready access of water molecules into the reaction zone. A vigorous agitation of the media along the discharge path will help supply of water molecules.

4. Conclusions

The dissociation of water employing reactive metals and electrical discharge is a viable method for generating hydrogen fuel, particularly for on-demand applications such as vehicular fuel source. The method based on feeding the aluminum fuel as a consumable electrode has yet to optimize many experimental variables. Thus far the values of electrical components like L, R, C in the discharge circuit and insulation of the consumable electrode surface turn out to be crucial to lengthen the reaction. With these conditions adjusted the initiation of the electrical discharge was able to consume about 100 mg of Al wire before the termination of the discharge. That amount of the metal fuel yielded about 50% of the theoretical value for the generation of hydrogen gas. The dissipated electrical energy is about 15 kJ indicating that the activation of the metal fuel for the reaction with water totally depends on electrical energy.

Acknowledgment

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