



Hydrogen generation by reaction of Al–M (M = Fe, Co, Ni) with water



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ABSTRACT

The addition of Fe, Co and Ni to Al significantly improves the production of hydrogen in Al–H₂O reaction. The total mass of Fe and Ni is less than the mass of Co. However, the hydrogen evolution induced by Co reaches ~1000 ml g^{−1} and the induction time shortens to 1.4 h at 35 °C. After the initial induced reaction, Al rapidly reacts with water even at 25 °C and the yield reached 90.0%. The additional Fe, Co and Ni combined with Al forms the galvanic cell and induce Al–H₂O reaction. The Al(OH)₃ hydrate formed in the reaction accelerates the removal of alumina and the reaction rate of Al–H₂O reaction, along with a decrease in the corrosion potential and an increase in the corrosion current. Beside that, both the pH and released heat in the local domain favor the Al–H₂O reaction.

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1. Introduction

Hydrogen energy as one of the clean energies has been attracted much attention, especially; hydrogen is used as the energy source in portable applications where it reacts with oxygen to produce electricity. Several methods have been used to generate hydrogen [1,2], such as in hydrocarbon fuel, water electrolysis and metal hydride. However, these methods do not satisfy the requirements of practical application completely. Therefore, the sustainable, simple and inexpensive hydrogen generation method should be developed.

Most of the hydrogen in the earth remains in water; hence the reaction of Al with water can be utilized to continually supply a high amount of pure hydrogen in an inexpensive and simple manner [3–5]. However, this method suffers from the spontaneous formation of an inert, dense and stable alumina film on the Al surface, thus hindering Al–H₂O reaction. This problem can be efficiently solved by breaking this film by ball milling Al metal [6–8], increasing the temperature and varying the pH [9] and composition of the solution [10]. The particle size of Al and other metal decreases by ball milling, thus increasing the reaction surface area and various defects and accelerating the Al–H₂O reaction. The

Al–H₂O reaction is exothermic, thus spontaneously proceeds at a certain rate. Moreover, when the initial test temperature is set to 70 °C, the oxide film is easily destroyed and the Al rapidly reacts with water [11]. Al is oxidized to form Al(OH)₄[−] at a high alkali concentration (pH > 12) and, it becomes Al³⁺ at a less acidic medium. The addition of salts (NaCl, MgCl₂, NiCl₂, CoCl₂, and FeCl₃) partially favors Al–H₂O reaction by pitting or forming the microgalvanic cell [10,12,13].

Undoubtedly, the mixing of Al with Ga, Bi, In, Sn, Li, Mg and Ca by ball milling effectively breaks the alumina film and promotes the corrosion of Al because of the formation of a microgalvanic cell or the production of a soluble hydroxide [7,14–16]. However, these small particles are easily oxidized in the air and lose the reaction activity. Recently, the hand-mixing of Al and TiO₂ followed by immersing the mixture in an Al(OH)₃ solution has also been effective in increase the production of hydrogen [16,17]. Previously, we reported that the mixing of Al and Fe–B using a mortar for less than 5 min significantly accelerated the corrosion of Al and hydrogen evolution [18]. However, it was inevitable for these particles to agglomerate in the air. In this study, the commercial Fe, Co and Ni without any treatment was directly used as the catalyst, meanwhile, the effect of these metals on Al–H₂O reaction was investigated.

2. Experimental

The commercial Co, Fe and Ni (analytical reagent, 99.9%)

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purchased from Sinopharm Chemical Reagent Co. Ltd was directly used as the raw material without any treatment. The different mass of Co, Fe and Ni were manually mixed with 0.2 g Al (Tianjin Damao Chemical Reagent Factory, 99.0% purity) for < 5 min. The mixture was rapidly transferred into the deionized water (30 mL, pH = 6.7–6.8) in a sealed 50 mL flask under stirring. The initial reaction of the mixture was allowed to proceed for 400 min, and then consecutive batches of pristine 0.2 g Al were added to the same flask individually in succession. The flask was heated on a water bath to maintain a constant temperature during the entire reaction, as shown in Fig. 1. The hydrogen produced was collected in an inverted burette completely filled with tap water. The volume of hydrogen produced within 400 min was recorded at 10 min intervals from the change in the water level in the inverted burette. Each reaction repeated more than three times to get the parallel data.

The Tafel curves of Al in $\text{Al}(\text{OH})_3$ solution were recorded using a CHI 660E electrochemical workstation. A mixture of 0.2 g Al, acetylene black and polyvinylidene fluoride (PVDF) in a weight ratio of 9:0.5:0.5 was dispersed in *N*-methylpyrrolidinone (NMP). The slurry was cast on a polished nickel plate ($1 \times 2 \text{ cm}^2$). The test was carried out using this electrode as the working electrode, a Pt working electrode as the counter work electrode, and a saturated calomel electrode (SCE) as the reference electrode. The measurements ranged from -1 V to 1 V at 0.01 mV s^{-1} .

The structure of the byproduct was determined using a Bruker D 8 Advanced X-ray diffractometer (XRD) equipped with Cu $K\alpha$ radiation at a voltage and current of 40 kV and 40 mA, respectively. The morphology of the products was investigated using an S-4800 field-emission scanning electron microscope (FE-SEM).

3. Results and discussion

3.1. Effects of Fe, Co and Ni contents on hydrogen generation

Al almost does not react with water at low temperature ($< 50^\circ\text{C}$). When Fe, Co or Ni and 0.2 g Al were immersed in deionized water, the reaction rate and the production of hydrogen at 35°C increased as shown in Fig. 2 and Table 1. When the amount of Fe was increased from 0.01 to 0.07 g, the production of hydrogen reached $\sim 875 \text{ mL g}^{-1}$ ($x = 0.02 \text{ g}$) and the induction time was $\sim 2 \text{ h}$ at 35°C . The hydrogen evolution induced by Co was $\sim 970 \text{ mL g}^{-1}$ ($x = 0.2 \text{ g}$) and the induction time shortened to $\sim 1.4 \text{ h}$. When 0.05 g of Ni was added, the production of hydrogen reached 1075 mL g^{-1} ($x = 0.05 \text{ g}$) and the induction time was $\sim 2.7 \text{ h}$. By comparing the effect of Fe, Co and Ni, it can be concluded that the induction time of

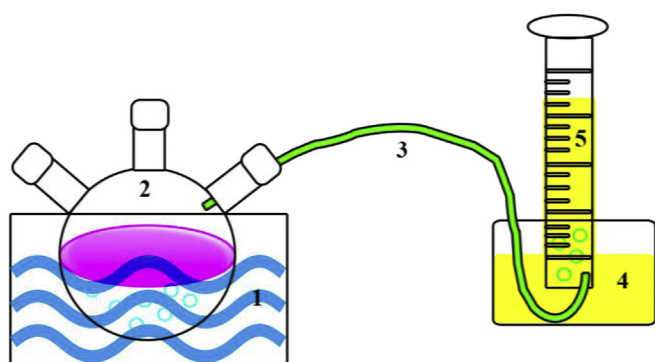


Fig. 1. Scheme of the experimental apparatus. 1. Water bath, 2. Three-necked flask, 3. Silicon rubber tube, 4. Water container, 5. Inverted burette.

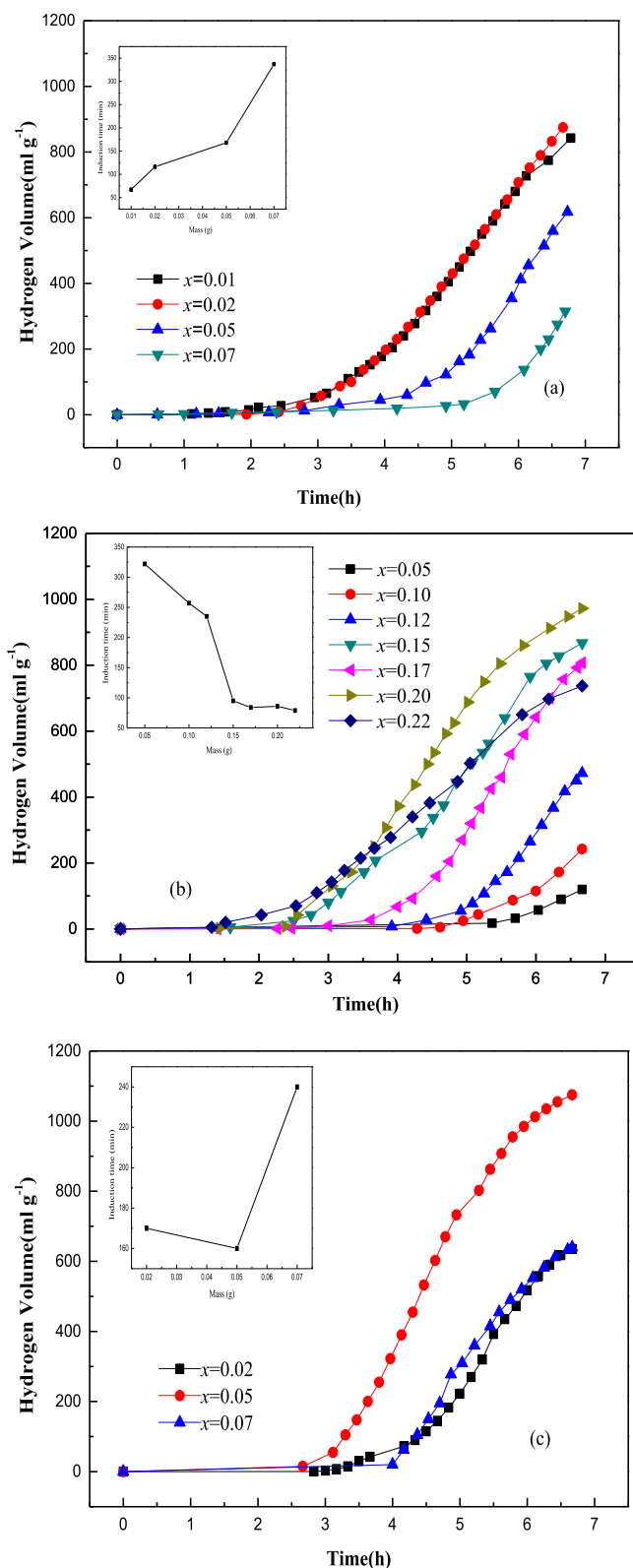


Fig. 2. Effect of Fe (a), Co (b) and Ni (c) on Al-H₂O reaction at 35°C , the inset plot is the effect of mass on the induction time.

Ni-promoted reaction was longer; however, the yield was higher at 35°C . When the initial temperature was increased to 45°C (S-1) as expected, the hydrogen generation rate of Al-H₂O reaction clearly increased for A and B. The maximum reaction rate for A increased

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