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Thermogravimetric analysis of hydrogen production of Al–Mg–Li particles and water

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ABSTRACT

Hydrogen is a renewable and environmentally friendly fuel with high calorific value. Aluminum is a good option for working as hydrolyzing metal, and aluminum–water reactions at medium–high temperature serve as a good energy supply in the use of both hydrogen and released heat, which leads to high energy efficiency. This study focused on hydrogen production characteristics and the chemical kinetics of aluminum particles with the addition of magnesium and lithium below 1030 °C. The experiments were conducted with THERMO CAHN's Thermax500 pressurized thermogravimetric analyzer. The Al content was settled at 85%, and the Mg and Al contents were adjusted from 0% to 15%. The addition of Li and Mg into Al resulted in good hydrolysis performance, and the ratio of the reacted Al to the total Al was over 50% and even reached up to 89%. The reaction process showed an obvious three-stage feature, and the three stages primarily corresponded to the reactions of Li, Mg, and Al. The products of hydrolysis in our tests were LiAlO₂, Li₂Al₄O₇, Al₂O₃, and MgO. The different promotion mechanism of Li and Mg on the reaction of Al–H₂O resulted in a hydrolysis performance that did not change monotonously with Li content.

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Introduction

Energy is an essential requirement for the development of human life and modern society, and the energy used today is mostly based on fossil fuels. The combustion of fossil fuels leads to emissions of pollutants and greenhouse gases. With the serious depletion of fossil fuels and their increasing pollutant emissions in recent years, renewable resources such as solar radiation, winds, waves, and tides have been identified, developed, and applied [1]. Hydrogen is source-independent and has higher energy content per mass than any other conventional fuels [2]; it could thus become a clean

energy carrier if generated with renewable energy sources [1,3]. However, hydrogen is mostly produced from methane and coal reforming, with only a small percentage produced from renewable sources [4]. As a result of the increasing attention to the production, application, and storage of hydrogen [3], the technologies and theory of hydrogen production are developing rapidly.

Conventional hydrogen production technology is mainly based on water electrolysis, reforming of nature gas, coal gasification, or other new methods [1,3,5]. However, these technologies have some weaknesses, such as CO₂ emission and unsatisfied efficiency [6]. In recent years it has been

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received increasing concerns that hydrogen generation from the reactions of active metals (Li, Be, B, Mg, Al, Zn, et al.) and water. The superiority of metal-water reaction is that it can be applied in both hydrogen production and energy storage, and metal oxides can be deoxidized and reused with renewable energy [5,7]. Metals such as Li, Zn, Mg, and Al are studied to produce hydrogen by different researchers. It is found that Zn, Mg and Al could react with water to produce hydrogen, which indicates that metal-water reaction prospect to be a revolutionary method in hydrogen industry [8–10]. Among these metals, Be is poisonous, Zn has a lower energy density than others, and B is hard to react continually. Aluminum is admitted to be the most promising metal for hydrogen generation.

Aluminum is the most abundant metal element in the earth's crust, and theoretically, aluminum can be completely recycled. With a high theoretical hydrogen yield of 0.11 g(H₂)/g(Al), aluminum is a good choice for hydrolyzing metals, thus making aluminum-based hydrogen production potentially inexpensive but with high H₂ capacity [11]. A passive oxide layer that covers the aluminum surface hinders the continuous reaction of aluminum and water. This passive oxide layer is a primary resistance that should be overcome to generate hydrogen efficiently. Some studies have concentrated on this problem and proposed several effective methods, such as mechanical polishing or cutting, doping some metals to form aluminum alloys, elevating temperature, using fine particles, and adding additives [10–13]. Additives include alkali, metal oxide, salt, hydride, and so on. Given that micronized Al particles are more difficult to be reacted to than nanopowder in water [14], nano Al particles or nano additives are considered beneficial [9]. C, MgO, TiO, In, Zn, Bi, NaOH, NaCl, and KCl with certain contents could more or less promote hydrogen generation and increase hydrogen yield [11,12,15].

Magnesium is also considered a suitable candidate for hydrogen generation. Uda et al. found that Mg nanopowder, with an average diameter of 265 nm, vigorously reacts with water at room temperature to produce 110 mL of hydrogen per 1 g of powder [16]. The hydrolysis reaction of Mg and Mg–Ni composite is quick and extensive when they are immersed in a borate buffer solution with 1 mol KCl; the addition of Ni has no effect on hydrolysis reaction in nonconductive media [17]. The Mg–H₂O reaction is accelerated when magnesium is mixed and milled with salts, and AlCl₃ shows the best performance among KCl, NaCl, LiCl, and MgCl₂ [18]. A Pt-coated Ti net is used to catalyze the hydrolysis of a secondary Mg sample in NaCl aqueous solution, and the grinding of the catalyst against the surface of Mg causes the removal of the Mg(OH)₂ passive layer and generate a large volume of H₂ [19]. Magnesium presents hydrolysis characteristics that are similar to those of aluminum, and a number of promotion methods work well for both metals. Aluminum alloys with magnesium and other hydrolytic metals could achieve good manifestation. Aluminum alloys, such as Mg–Al–Fe, exhibit good hydrolysis reactivity. The Mg₆₀–Al₃₀–Fe₁₀ (wt%) alloy in 0.6 mol L⁻¹ NaCl solution at 25 °C produces 1013 mL g⁻¹ hydrogen, and the maximum rate reaches up to 499 mL min⁻¹ g⁻¹ [20]. Al–Li alloys produced via ball milling [21] and melting [22] achieve a hydrogen yield of 100%, and a

high Li content improves hydrolysis properties because excess LiOH accelerates aluminum hydrolysis in water [21]. The addition of NaCl enhances both the hydrogen generation rate and the hydrogen yield of Al–Li alloys and reduces Li contents [23]. Solution composition has some effect on the hydrogen production of Al–Li alloys with water, and Mg²⁺ or NO₃⁻ shows a negative influence on the ultimate hydrogen yield [22]. The hydrolytic performance of Al–Li–In–Zn alloys improves via the synergistic effects of composition, milling time, and hydrolytic temperature. In–Li alloys function as an initial active center and produce LiOH in water, which further stimulates the hydrolysis of Al–In–Zn alloys [13]. As for Al–Ga–In–Sn alloys, Wang and his colleagues [24] proposed a mechanism of Al corrosion in water based on the partial coverage of the Al grain surface with Ga–In–Sn.

As indicated in the discussion above, most investigations into the hydrolysis of Al alloys were conducted in liquid solutions below 100 °C. In these studies, the massive amounts of excess water reduced the solution temperature, which resulted in a low H₂ yield and waste of reaction heat. Moreover, chemical kinetics was seldom a concern, with apparent parameters such as H₂ yield and generating rate being the primary focus. The objective of the present study is to describe the chemical kinetics and process features of H₂ generation via Al–Mg–Li below 1030 °C. The results of the reactivity of Al–Mg–Li would provide a reference for power supply via both hydrogen fuel cell and reaction heat utilization.

Experimental

All metal particles in the experiments originated from three kinds of powders obtained from commercial sources. These powders were aluminum powder with an average size of 10 μm and a purity of 99.9%, magnesium powder with a mean size of 100 μm and a purity of 99.9%, and aluminum–lithium alloy with a mean size of 45 μm and a purity of 99.99%. The alloy comprised 80% aluminum and 20% lithium in weight. Before the tests, the three powders in certain proportions were mixed and stirred by hand sufficiently in atmosphere of inert gas. The proportion of aluminum was settled at 85% in all samples, and the contents of magnesium and lithium were adjusted from 0% to 15%. The Li contents of the seven samples were 0%, 2%, 5%, 7.5%, 10%, 13%, and 15%.

The experiments were conducted with THERMO CAHN's Thermax500 pressurized thermogravimetric analyzer (PTGA, accuracy 0.02%), as shown in Fig. 1. The electronic balance with an accuracy of 0.1% was used to measure the weight of samples at room ambient temperature. The stainless steel reactor had a cylindrical shell with an inner diameter of 5 mm and a height of 10 mm. Argon was chosen as the carrier gas to carry water vapor into the reactor because nitrogen might react with aluminum at high temperatures. Nitrogen gas was used as the balance gas. A glass crucible with 10 mg of sample was placed in the reactor chamber first. Before the test, argon was fed into the reactor to drain air out and avoid powder oxidation by oxygen in air. The temperature was increased from room temperature to 1030 °C with a heating rate of 25 °C/min. The temperature was then maintained at 1030 °C for 10 min to obtain complete hydrolysis. As the temperature

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