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A novel water-splitting electrochemical cycle for hydrogen production using an intermediate electrode



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HIGHLIGHTS

- A novel water electrolysis system using an intermediate electrode is proposed.
- Manganese dioxide was selected for the intermediate electrode.
- Oxygen and hydrogen evolution reactions take place in different steps.
- Average voltage for water electrolysis was below 1.6 V at 60 °C.

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ABSTRACT

A novel water electrolysis system containing an intermediate electrode is proposed, which can generate oxygen and hydrogen gases separately through a two-step electrochemical cycle. This multi-step water electrolysis system can achieve a high energy conversion efficiency during hydrogen production by reducing the ohmic overpotential between the electrodes through use of a thinner separator than that used in conventional systems. In the present study, manganese dioxide (MnO₂) was selected as the intermediate electrode because its oxidation-reduction potential is located between the potentials of the hydrogen and oxygen evolution reactions. In addition, the electrolysis cell contained using metal hydride and nickel hydroxide as negative and positive electrodes, respectively, to split the gas evolution reactions at the solid/liquid/gas three-phase boundary into reactions at the solid/gas and solid/liquid two-phase boundaries. The electrochemical performance of the fabricated electrolyzer cell was tested at 25, 40, and 60 °C and the results show that oxygen and hydrogen can be easily separated by producing them in different steps. Furthermore, a low average voltage for water electrolysis, below 1.6 V, was obtained at 60 °C. Thus, the proposed water electrolysis system has potential as an electrochemical reactor for highly efficient, high-purity hydrogen production.

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1. Introduction

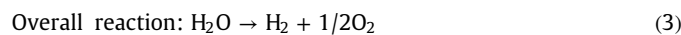
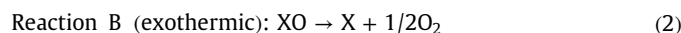
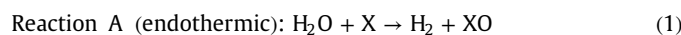
With increasing energy demands, renewable energy sources such as solar and wind power have begun to play important roles in alternative electric power generation because they are environmentally friendly and inexhaustible. However, solar and wind power are intermittent and variable energy sources that depend strongly on weather conditions. To compensate for variable power and improve power quality, energy storage systems (ESSs) such as secondary batteries and hydrogen production technologies are

required (Beaudin et al., 2010; Turner, 1999; Yekini Suberu et al., 2014). In the case of secondary batteries, a number of research and development projects on lithium ion, sodium sulfur, and redox-flow batteries are currently underway as ESSs (Yekini Suberu et al., 2014). However, it is difficult to store all of the energy generated by renewable energy sources in secondary batteries because their storage capacities are limited. Therefore, power-to-gas, which converts electricity into hydrogen by water electrolysis, has attracted a great deal of attention for large-scale energy storage from renewable energy sources (Gahleitner, 2013). In addition to water electrolysis, there are several technologies for hydrogen production, such as thermolysis and photoelectrolysis (Holladay et al., 2009). For thermochemical water splitting (water thermolysis), which uses heat (especially solar energy in recent research) alone

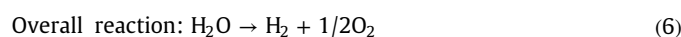
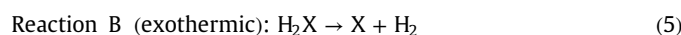
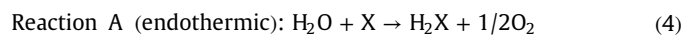
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to decompose water to hydrogen and oxygen, several two-step water-splitting cycles based on metal oxide (intermediate compound) redox reactions have been suggested, as follows:



or



where X is an intermediate compound. By splitting the single-step thermolysis reaction into two endothermic and exothermic reaction steps, water decomposition takes place at much lower temperature than the single-step water decomposition (approximately 4300 K). Oxygen and hydrogen gases are generated during the reduction (Eqs. (2) and (4)) and oxidation (Eqs. (1) and (5)) reactions of X, respectively. Several processes have been tested, such as ZnO/Zn, Fe₃O₄/FeO redox pairs (Charvin et al., 2007; Steinfeld, 2005, 2002; Steinfeld et al., 1999). Furthermore, four-step UT-3 and SI thermochemical cycles for hydrogen production have been proposed (Aihara et al., 1992, 1990; Sakurai et al., 1996a, 1996b, 1996c, 1995, 1992). The maximum temperature for water decomposition by the four-step thermochemical cycle is 1033 K, which is less than the two-step thermochemical cycle and leads to higher energy conversion efficiency for hydrogen production.

Water electrolysis for power-to-gas conversion has several advantages, such as high energy conversion efficiency, simple technology, and the ability to produce high-purity hydrogen for ESSs (Barbir, 2005; Chisholm et al., 2014; Holladay et al., 2009). A conventional alkaline water electrolysis is described by the following reactions (Holladay et al., 2009):

Positive electrode:



Negative electrode:



Overall reaction of water electrolysis:



The conventional alkaline water electrolysis system is based on a single electrochemical cycle in which simultaneous evolution of oxygen and hydrogen takes place at the positive and negative electrodes, respectively, as shown in Fig. 1(a). To obtain high-purity hydrogen gas by preventing its mixing with oxygen gas, relatively thick membrane gas separators are necessary, which results in a high ohmic overpotential (Holladay et al., 2009; Vermeiren, 1998; Zeng and Zhang, 2010).

Therefore, in the present study, we propose a novel electrochemical water-splitting cycle for hydrogen production, which consists of two-step electrochemical cycles for oxygen and hydrogen evolution reactions by introducing an intermediate electrode. Fig. 1(b) shows a diagram of the reaction mechanism in the proposed novel water-splitting electrochemical cycle for hydrogen

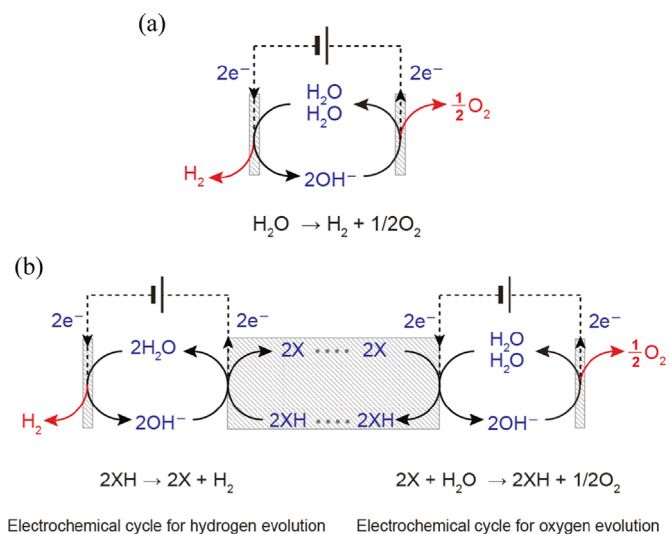


Fig. 1. Schematic diagram of the reaction mechanisms for (a) conventional alkaline water electrolysis; (b) the proposed alkaline water electrolysis (where X is an intermediate electrode).

production. The proposed alkaline water electrolysis consists of a two-step electrochemical cycle: (i) an electrochemical cycle for oxygen evolution between the positive and intermediate electrodes and (ii) an electrochemical cycle for hydrogen evolution between the intermediate and negative electrodes, as follows:

(1) 1st step: Electrochemical cycle for oxygen evolution reaction
Intermediate electrode:



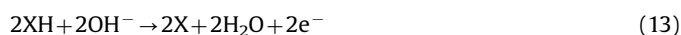
Positive electrode:



Overall reaction for oxygen generation:



(2) 2nd step: Electrochemical cycle for hydrogen evolution reaction
Intermediate electrode:



Negative electrode:



Overall reaction for oxygen generation:



where X is an intermediate electrode material. Multi-step water electrolysis offers the following advantages: (i) higher-purity hydrogen can be obtained because hydrogen and oxygen gases are generated in different steps and (ii) high energy conversion efficiency for hydrogen production can be achieved by reducing the ohmic overpotential between the electrodes through the use of a thinner separator than a conventional one. We fabricated an alkaline electrolyzer cell that consisted of metal hydride, manganese dioxide, and nickel hydroxide as negative, intermediate, and positive electrodes, respectively. We then examined the electrolyzer cell to validate the water-splitting electrochemical cycle for hydrogen production using an intermediate electrode.

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