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Methane reforming and water splitting using zirconia-supported cerium oxide in a volumetric receiver–reactor with different types of foam devices

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Abstracts

Syngas and hydrogen production from CeO_2/ZrO_2-coated foam devices was investigated under simulated solar radiation. Three different types of foam devices were employed in order to find the effect of the physical properties of foam material on the cyclic reaction of methane reforming and water splitting. CeO_2/ZrO_2-coated SiC, Ni, and Cu foam were prepared using drop-coating method. The methane reforming was performed at 900 °C for 15–25 min, and the water splitting was performed at the same temperature for 45 min. In addition, the methane reforming and water splitting were repeated ten cycles to compare the CO and H_2 yields in the methane reforming, and the H_2 yield in the water splitting. Metallic foam such as Ni and Cu showed better production yield than SiC foam and ten cycles average conversion of CeO_2 on the SiC, Ni and Cu foam were 54%, 74% and 65%, respectively. It is believed that the increased production yield of metallic foam was due to higher thermal conductivity of metallic foam and the partial reduction and re-oxidation of Ni and Cu foam.
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1. Introduction

The thermochemical conversion of concentrated solar heat into chemical fuels has an advantage of enabling the transport of solar energy and long term storage (Gokon et al., 2009; Kodama et al., 2003). Several solar thermochemical energy conversion technologies have been studied, such as steam methane reforming, two-step water splitting using metal oxide, coal gasification, and natural gas reforming (Kang et al., 2008). In solar two-step water splitting, when a metal oxide is reduced into metal and oxygen using high-temperature solar heat, the reduced metal can react with water to produce hydrogen without CO_2 emission (Jang et al., 2011). However, the thermal reduction of a metal oxide requires very high temperature (>1400 °C). Instead of high-temperature thermal reduction and steam reforming, if methane is used as a reducing agent, the reduction of a metal oxide can be conducted at relatively lower temperature (800–1000 °C) than in direct thermal reduction.

Recently, two-step processes for syngas/hydrogen production by methane reforming/water splitting with a metal oxide have been proposed (Steinfeld et al., 1995, 1998; Kodama et al 2000; Kang et al., 2010; Jeong et al., 2011). The syngas and hydrogen production process (reduction and oxidation step) can be written as:

$$CH_4 + \text{metal oxide} \rightarrow \text{reduced metal oxide} + CO + 2H_2$$

$$\text{Reduced metal or metal oxide} + H_2O \rightarrow \text{metal oxide} + H_2$$

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This redox reaction is a promising chemical pathway for the storage and transportation of solar heat by converting solar energy into chemical energy. In the first reaction, reduced metal oxide is produced by a solar endothermic step, and syngas is produced with an H_2/CO ratio of 2.0, which is suitable for methanol synthesis or Fischer–Tropsch processes. Efficient utilization of the reduced metal oxide involves water splitting to generate hydrogen in the second reaction. The second reaction produces hydrogen which is uncontaminated with CO and CO_2 gases, and the hydrogen gas can be directly utilized for PEM fuel cells.

This redox system for syngas and hydrogen production has been studied for many researchers. Steinfeld et al. (1993) showed that the production of iron and syngas by Fe_3O_4-reduction (Fe_3O_4 to Fe) with methane. The yield of Fe_3O_4 to FeO during the reduction with 10% CH_4/Ar flow at 1000 °C was 32%, and the FeC was formed due to deposited carbon by the methane decomposition. As a result, the H_2/CO ratio was higher than 2.0, and the CO gas from FeC was released during the H_2 production step of oxidation of reduced Fe with water.

Kodama et al. (2000, 2002, 2003) studied Fe_3O_4, ZnO, SnO_2, In_2O_3, V_2O_5, MoO_2 and WO_3 as the metal oxides for syngas production using methane at temperatures around 900 °C, and WO_3 was found to be the most reactive and selective metal oxide phase. The reactivity of WO_3 was much improved by supporting WO_3 on micron-sized ZrO_2 powder. Ni(II)-, Co(II)- and Zn(II)-ferrites were also studied for the production of syngas and hydrogen via the solar redox process, and it was found that the methane conversion and H_2 yield of the Ni(II)-ferrite system was higher than those of the other ferrite systems and the Fe_3O_4 system (Kodama et al., 2002).

Recently, Abanades and Flamant (2006) studied a solar two-step water splitting system based on CeO_2/Ce_2O_3 oxides. In this CeO_2/Ce_2O_3 redox system, heat is the only energy input, and no reducing agent is used during the reduction. Hence, the produced hydrogen is not contaminated by carbon oxides, and this process may be a promising sustainable option for utilizing solar thermal energy. However, the thermal reduction was operated at a high temperature of 2000 °C. Nonetheless, because CeO_2 and Ce_2O_3 have sufficiently high melting points (>1950 °C) and carbide phase has not been reported during methane reforming, the CeO_2/Ce_2O_3 system is a promising candidate for redox systems with syngas and hydrogen production at lower temperature. In addition, it has been reported that CeO_2 can decrease carbon deposition during the methane reforming when used as the support. Hence, Kang et al. (2010) investigated the redox cycling of $CuFe_2O_4$ supported on ZrO_2 and CeO_2 for a two-step syngas and hydrogen production process. They reported that using the supports improved the reactivity, and CeO_2 was revealed to be a better support than ZrO_2 in terms of reactivity and CO selectivity during repeated cyclic reactions. Also, Jeong et al. (2011) found that the CeO_2/ZrO_2 exhibited good performance and reproducibility for repeated cyclic reaction throughout syngas and hydrogen production in the tube furnace. Since in case of the CeO_2/ZrO_2 system, some Ce atoms in its crystal lattice were replaced by other cations such as Zr and this replacement leads to structural defects which should enhance the oxygen migration in the lattice and consequently the thermal stability and oxygen storage capacity (Jeong et al., 2011).

A volumetric solar chemical reactor equipped with a ceramic honeycomb receiver/absorber coated with an active ferrite powder has been proposed and validated by Roeb et al. (2006). The ferrite-loaded ceramic honeycomb, utilized as a solar receiver/absorber was employed in a volumetric solar reactor. Kodama and Gokon studied a solar two-step water splitting cycle with metal-oxide-coated ceramic foam devices (Gokon et al., 2008, 2011; Kodama et al., 2009). The advantage of using a ceramic foam device is that it makes possible to effectively absorb solar radiation due to a large specific surface area.

In this study, the cyclic production of syngas and hydrogen was performed at fixed temperature (900 °C) with different types of SiC, Ni, and Cu foam devices under simulated solar radiation. The idea of using the ZrO_2 support in a fine powder form was adapted from the previous studies in order to well disperse the metal oxide, to suppress sintering of the metal oxide, and to improve its reactivity (Kodama et al., 2000, 2003; Jeong et al., 2011). In addition, ZrO_2 is known to be inactive in these previous studies. The idea of using the SiC foam was also adapted from a previous study (Gokon et al., 2009, 2010) and from the experience in our laboratory in order to effectively absorb the solar radiation (Jang et al., 2011). The employed metallic foam of Cu and Ni have higher thermal conductivity than of ceramic foams such as alumina and SiC, expecting a more uniform radial temperature distribution on the illuminated surface of the foam device. Also, CeO_2/ZrO_2-coated Ni may act as an active catalyst for methane partial oxidation, and CeO_2/ZrO_2-coated Cu showed no activity for carbon deposition from aliphatic hydrocarbon (Pengpanich et al., 2004; Kang et al., 2008). The reduction–reoxidation cycle was repeated ten cycles to compare the gas yields in the methane reforming step (MR), and water splitting step (WS). Before and after the cyclic reactions, physical and chemical change in the ZrO_2-supported CeO_2 and foam devices were investigated using SEM, XRD and EDS analyses.

2. Experimental

2.1. Synthesis of ZrO_2-supported CeO_2 powder

ZrO_2 powder (Alfa Aesar, 99.5% purity) with a particle size smaller than 1 μm was used as a support for CeO_2. A ZrO_2-supported CeO_2 sample was prepared by a precipitation method. The ZrO_2 powder was previously suspended in distilled water, and an aqueous solution of cerium nitrate was prepared by dissolving $Ce(NO_3)_3 \cdot 6H_2O$ (Kanto chemical, 99.99%) in the ZrO_2 suspension using the stirrer

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