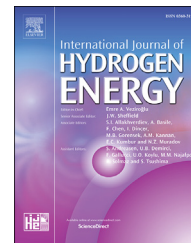


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Analysis of H₂ and CO production via solar thermochemical reacting system of NiFe₂O₄ redox cycles combined with CH₄ partial oxidation

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ABSTRACT

Thermal reduction of the partial oxidation of CH₄–NiFe₂O₄ followed by oxidation with H₂O and CO₂ was numerically investigated for H₂ and CO production. P1 radiation model was used to account for radiative heat transfer. The synergistic effect of the reactivity of Fe/Ni exhibited a very promising strategy for producing 45% of syngas with 2.54 ratios of H₂:CO at the first step and 55% of syngas with 2.34 ratios of H₂:CO at the second step. The increase in incident radiation heat flux to 437.69 kW/m² resulted in higher reduction kinetics of species conversion until the formation of oxygen carriers consisting of 65% of FeO, 35% of Ni–Fe and 2.6% of carbon deposition. However, during the reduction process, the decrease in total pressure to 0.05 MPa enhanced the species reactivity and the production of H₂ and CO while minimizing carbon deposition. Moreover, the oxidation temperature, operating pressure and the concentration of oxidizing species have strong impacts on the oxidation kinetics. Unlike high thermal reduction process, increasing the total pressure to 1 MPa has favorable effects on syngas production at oxidation step.

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Introduction

Energy is considered as the heart of industrial development and can be used to boost economic growth. The ever-increasing global demand for clean transportation fuels, chemicals and power electric associated with developing of

CO₂-neutral processes gave rise to a great interest in hydrogen and syngas production research and development [1–3]. Syngas is finding increased usage in power and electricity generation with minimum impacts on the environment while decoupling the link between energy utilization and greenhouse gas emissions [4]. Moreover, the mixture of H₂ and CO is utilized as a building block material for the production of

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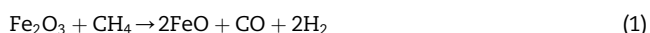
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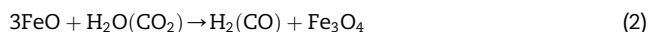
synthetic liquid fuels such as Fischer-Tropsch diesel, methanol and dimethyl ether via industrially available processes [5–7]. Therefore, hydrogen and syngas production technologies development have great potential in addressing major challenges towards sustainable development in the energy area.

Thermochemical solar energy-to-fuel conversion technology using concentrated solar energy as a heat source to drive a high-temperature chemical reaction is one of the most attractive research areas for CO₂ depletion and turning CO₂ emission into valuable products such as syngas (H₂ and CO) and solar fuels [1,8]. The mechanism is essentially based on the solar thermochemical cycles consisting of cyclical series of endothermic and slightly exothermic chemical reactions [5]. Among solar thermochemical cycles that have been proposed, two-step processes based on metal oxides have been considered well suited to concentrated solar radiation applications [6,9,10]. In this way, the raw materials are reduced under high-temperature at the first step and oxidized at the second step with CO₂ and H₂O [11,12]. In the recent years, great efforts have been developed on a wide variety of transition metal oxides: Cr₂O₃, CoO, CeO₂, Fe₃O₄, MnO₂, and ZnO used as oxygen carriers in the solar thermochemical reacting system for hydrogen and syngas production [13,14]. However, Fe-based oxygen carrier exhibited enough high reactivity in a wide operating temperature range of 900–2000 K [15]. Besides, Fe-based oxygen carrier is considered as the most appropriate candidate from its abundant, cost standpoint and environmentally benign. The use of mixed metal oxides (e.g. Me/Fe, Me: Mn, Ni, Co, and Cu) as oxygen carrier candidates has improved the reactivity of Fe-based oxygen carrier thereby exhibiting better reactivity and selectivity towards H₂ and CO formation [16–18]. Thus, a deep understanding of the synergistic effects of mixed materials with the reaction temperature changes could highly improve the process of solar fuels production. Moreover, combining the partial oxidation of CH₄ to Fe-based oxygen carrier redox cycles have gained significant attention in syngas research development [16,19–21]. For example, the solar thermochemical process using Fe₂O₃ oxygen carrier combined with CH₄ partial oxidation can be described by the following reactions [22].

Highly endothermic reduction step



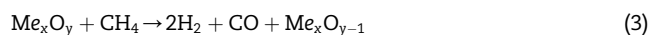
The reduced metal oxide thus produced can be used for splitting H₂O or CO₂ in the second step.



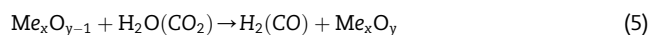
As well-known from Ni catalysts in steam/CO₂-reforming and the partial oxidation of CH₄, among oxygen carriers, Ni carrier materials have excellent reactivity and a higher selectivity for CH₄ partial oxidation [22,23]. Most of the literature reported that adding Ni to the raw material will result in the great amount of CO production, that is, ten times higher than Ce–Fe oxygen carrier without Ni catalyst [13] and four times H₂ production from NiFe₂O₄ higher than those produced from Fe₂O₃ [24]. Moreover, NiFe₂O₄ is considered as catalyst precursor from its redox ability which is obviously good than single Fe-based oxygen carrier since it is a variety of Ni–Fe

bimetallic oxide [16,24,25]. Thus, NiFe₂O₄ can be considered as the most appropriate candidates for solar thermochemical applications due to the spinel structure, thermal stability, high reactivity and the flexibility of oxygen carrier [26–30]. Besides, important efforts have been developed to gain a deep insight into the reaction mechanisms and kinetics of NiFe₂O₄ redox behavior. Huang et al. [19] have studied thermodynamics analysis of NiFe₂O₄ redox ability and the species reactivity was tested by H₂-Temperature-Programming Reduction and CO₂-Temperature-Programming Oxidation experiments to verify the redox performance of NiFe₂O₄. They reported that four phases including two species of NiO and Fe₂O₃ can be formed during the first decomposition of NiFe₂O₄. The thermal reduction observed from X-ray diffractometer (XRD patterns) of different oxygen carriers in CH₄ atmosphere indicated that NiFe₂O₄ can be reduced to Fe(Ni) alloy state. The reduction of NiO–Fe₂O₃ mixed oxides can yield FeO, Ni, and Fe(Ni) while NiO and Fe₂O₃ can be reduced to Ni and FeO, respectively [19,31]. The partial oxidation of CH₄ with reduction of NiFe₂O₄ compounds (Me_xO_y) followed by oxidation of oxygen carriers (Me_xO_{y-1}) with H₂O and CO₂ can be described as follows.

Thermal reduction step:



Oxidation with H₂O and CO₂.



Reaction (4) is the possible CH₄ cracking while Reactions (6) and (7) are carbon gasification and Boudouard reaction, respectively. Steam/Dry reforming of CH₄ can occur during oxidation due to the concentration of unreacted CH₄ as follows.



Lorentzou et al. [32], Kostoglou et al. [33], Kaneko et al. [34] and GO et al. [35] have also investigated the reaction kinetics of ferrites reduction and oxidation for H₂ production via the solar thermochemical process. Moreover, the reduction of NiFe₂O₄ has been a subject of intensive researches. The literature is often related to the carbothermic reduction of NiFe₂O₄ and the reduction of NiFe₂O₄ with CH₄ [36–38], as well as NiFe₂O₄ direct reduction via solar thermal energy [39–42]. The recent studies suggested that the reduction of NiFe₂O₃ in the CH₄ atmosphere can result in higher reducibility with additional production of H₂ and CO due to the activity of different oxygen carriers [19,37]. However, most of the studies were limited to the thermal reduction of NiFe₂O₃ while a few studies were extended to the oxidation step. Also, part of metallic Fe and Ni could divorce from the spinel structure resulting in the

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