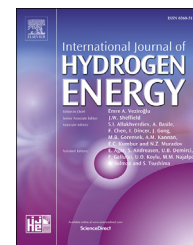


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Solar thermochemical ZnO/ZnSO₄ water splitting cycle for hydrogen production

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ARTICLE INFO

Article history:

Received 16 December 2016

Received in revised form

24 February 2017

Accepted 25 February 2017

Available online 18 March 2017

Keywords:

Solar fuels

Solar reactor

Thermodynamics

Hydrogen

Water splitting

Zinc oxide

ABSTRACT

In this paper, solar reactor efficiency analysis of the solar thermochemical two-step zinc oxide–zinc sulfate (ZnO–ZnSO₄) water splitting cycle. In step-1, the ZnSO₄ is thermally decomposed into ZnO, SO₂, and O₂ using solar energy input. In step-2, the ZnO is re-oxidized into ZnSO₄ via water splitting reaction producing H₂. The ZnSO₄ is recycled back to the solar reactor and hence can be re-used in multiple cycles. The equilibrium compositions associated with the thermal reduction and water-splitting steps are identified by performing HSC simulations. The effect of Ar towards decreasing the required thermal reduction temperature is also explored. The total solar energy input and the re-radiation losses from the ZnO–ZnSO₄ water splitting cycle are estimated. Likewise, the amount of heat energy released by different coolers and water splitting reactor is also determined. Thermodynamic calculations indicate that the cycle (η_{cycle}) and solar-to-fuel energy conversion efficiency ($\eta_{\text{solar-to-fuel}}$) of the ZnO–ZnSO₄ water splitting cycle are equal to 40.6% and 48.9% (without heat recuperation). These efficiency values are higher than previously investigated thermochemical water splitting cycles and can be increased further by employing heat recuperation.

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Introduction

The sulfur–iodine cycle [1,2] and its variation the hybrid sulfur cycle [3] are considered as more promising towards H₂ production via water splitting reaction as the temperatures required to run these cycles are considerably lower than other thermochemical water splitting cycles i.e. zinc oxide cycle, iron oxide cycle, tin oxide cycle, ceria cycle, ferrite cycle, and perovskite based cycle [4–24] and comparable to hybrid photo-thermal sulfur–ammonia water splitting cycle [25,26]. Although these cycles are advantageous in terms of lower

operating temperatures, the most challenging part is the decomposition of SO₃. The reduction of SO₃ into SO₂ and O₂ requires high operating temperatures and it is possible only under catalytic conditions. In past, various transition metal based catalysts such as nickel, manganese, etc. were tested towards the SO₃ decomposition reaction. However, it was observed that at higher temperatures these metal catalysts get converted into metal sulfates and their catalytic activity is hindered due to the sulfation poisoning [27]. Noble metal based catalysts (supported on different metal oxides) were also tested and observed to be attractive in terms of their catalytic activity towards decomposition of SO₃. However, the

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<http://dx.doi.org/10.1016/j.ijhydene.2017.02.190>

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extremely high cost of the noble metals pose limitations towards utilization of such catalysts for commercial scale application.

Converting the sulfur–iodine and the hybrid sulfur cycle into solar driven metal oxide–metal sulfate (MO–MS) thermochemical water splitting can be considered as an alternative for the production of H_2 at lower operating temperatures. It is a two-step process in which the first step belongs to the solar thermal decomposition of metal sulfate (MSO_4) into metal oxide (MO), sulfur dioxide (SO_2) and oxygen (O_2). The metal oxide and sulfur dioxide produced in step-1 can be re-oxidized into metal sulfate in step-2 with the help of water splitting reaction which further yields into H_2 production. The reaction chemistry associated with the MO–MS water splitting cycle is shown below.



In one of our recent studies [28], the iron oxide–iron sulfate (IO–IS) water splitting cycle has been thermodynamically investigated. The results obtained in this study indicate that the solar-to-fuel energy conversion efficiency in case of iron oxide–iron sulfate cycle is significantly higher than the previously studied metal oxide based water splitting cycles. In past, several metal oxides were examined towards water splitting reaction and in addition to the iron oxide, zinc oxide is also considered as one of the promising one. Therefore, in this paper, the thermodynamic analysis of the zinc oxide–zinc sulfate (ZnO – $ZnSO_4$) water splitting cycle is carried out by using HSC Chemistry software and its thermodynamic database. The results obtained are compared with the previously studied thermochemical water splitting cycles. The typical two-step solar thermochemical ZnO – $ZnSO_4$ water splitting cycle is depicted in Fig. 1.

Thermodynamics equilibrium compositions

The specific reaction chemistry associated with the ZnO – $ZnSO_4$ water splitting cycle is as follows:

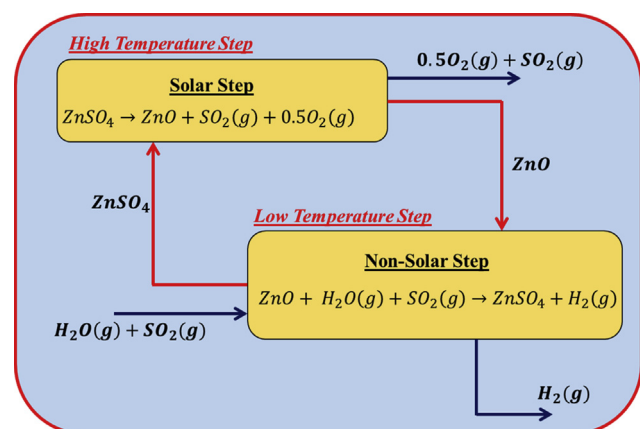
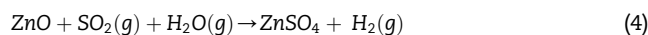
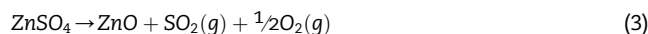


Fig. 1 – Typical two-step solar thermochemical ZnO – $ZnSO_4$ water splitting cycle.



The equilibrium compositions of the thermal reduction and water splitting steps are determined by performing HSC simulations and by assuming the continuous operation of the solar reactor with molar feed rate of $ZnSO_4$ equal to 1 mol/s. HSC simulations are performed to study the effects of various operating parameters towards variations in the equilibrium compositions and solar reactor efficiency parameters.

At first, the feasibility of the thermal reduction and water splitting reactions is analyzed by calculating the change in the Gibbs free energy (ΔG) as a function of thermal reduction temperature (T_H) and water splitting temperature (T_L) in absence of inert Ar. The results reported in Fig. 2a and b indicate that the ΔG in case of thermal reduction of $ZnSO_4$ decreases with the increase in the T_H , whereas the ΔG in case of water splitting reaction increases with the increase in the T_L . For instance, in case of thermal reduction of $ZnSO_4$, ΔG decreases from 194.3 to -97.6 kJ/mol as the T_H increases from 500 to 1700 K. The ΔG associated with the water splitting reaction increases from -21.5 to 110.2 kJ/mol as the T_L increases from 300 to 900 K. The obtained results show that the complete thermal reduction of $ZnSO_4$ is feasible above 1270 K (in

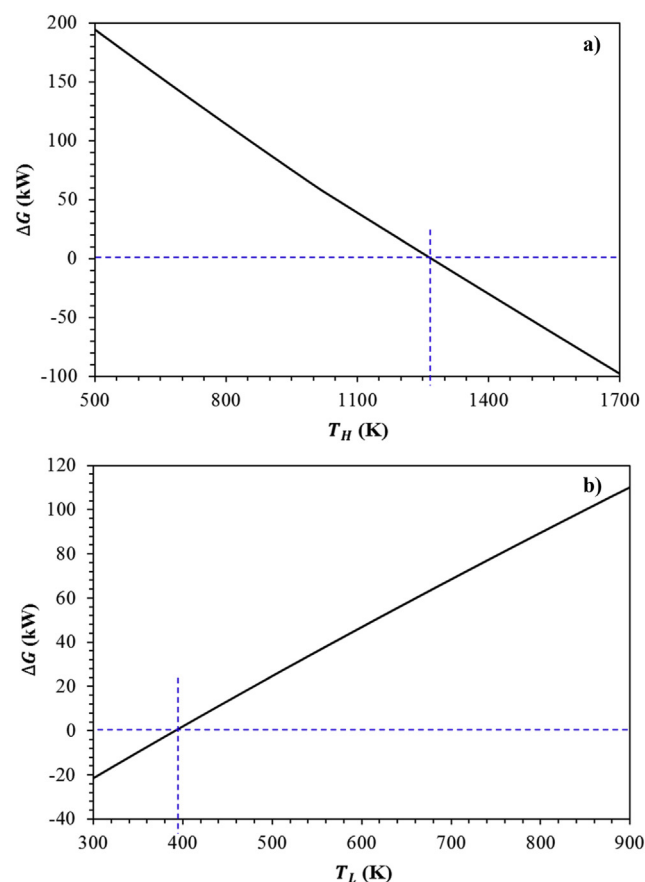


Fig. 2 – Effect of (a) T_H and (b) T_L on ΔG associated with the thermal reduction and water splitting step associated to the solar thermochemical ZnO – $ZnSO_4$ water splitting cycle.

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