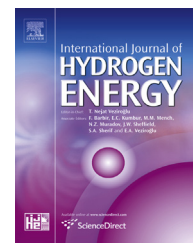


Available online at www.sciencedirect.com

ScienceDirect

journal homepage: www.elsevier.com/locate/he

Catalytic membrane reactors for SO₃ decomposition in Iodine–Sulfur thermochemical cycle: A simulation study

Lie Meng, Masakoto Kanezashi, Toshinori Tsuru*

Department of Chemical Engineering, Graduate School of Engineering, Hiroshima University, 1-4-1 Kagamiyama, Higashi-Hiroshima 739-8527, Japan

ARTICLE INFO

Article history:

Received 3 February 2015

Received in revised form

17 July 2015

Accepted 24 July 2015

Available online 15 August 2015

Keywords:

Catalytic membrane reactor

SO₃ decomposition

IS process

Simulation

ABSTRACT

The possibility of applying a catalytic membrane reactor (CMR) to SO₃ decomposition in a low-temperature range was theoretically evaluated with the purpose of producing CO₂-free hydrogen in an Iodine–Sulfur thermochemical cycle. A one-dimensional, isothermal and plug-flow model was developed for a cocurrent membrane reactor with selective permeation from the reactant stream to the permeate stream. Simulation results have revealed that CMRs can greatly reduce the reaction temperature for SO₃ decomposition from the conventional 1200–1400 K to about 900 K. We predicted that porous inorganic membranes with a high O₂ permeability and with selectivities of more than 50 for O₂/SO₃ and less than 10 for O₂/SO₂ had the potential to effectively improve SO₃ conversion. CMRs were simulated to carry out SO₃ decomposition at different catalyst weights, reaction temperatures, and SO₃ feed flow rates as well as pressures in feed and permeate streams. SO₃ conversion at 900 K was increased to 0.93 beyond the equilibrium conversion of 0.28 due to a shift in thermodynamic equilibrium.

Copyright © 2015, Hydrogen Energy Publications, LLC. Published by Elsevier Ltd. All rights reserved.

Introduction

With the simultaneous growth of the demand of global energy and of urgent concerns for reduction of greenhouse gas emissions, hydrogen has become a promising energy carrier [1]. Currently, hydrogen is derived from nonrenewable natural gas and petroleum, but could in principle be generated from renewable resources such as biomass or water. Splitting water via a thermochemical cycle presents a viable option for large-scale hydrogen production [2]. Among the large number of thermochemical cycles that can be used to split a molecule of

water, the Iodine–Sulfur thermochemical water-splitting cycle (IS process), which was initially proposed by General Atomics, is considered an efficient, massive and CO₂-free approach [3,4]. As indicated in Fig. 1, the IS process primarily consists of three chemical reactions that involve the decomposition of sulfuric acid (H₂SO₄) and hydrogen iodide (HI), followed by the regeneration of these reagents using the Bunsen reaction [5]. High temperatures equal to, or greater than, 1100 K are required to decompose H₂SO₄. The exothermic Bunsen reaction is performed at temperatures ranging from 300 to 400 K. Hydrogen is generated during the HI decomposition, using a heat source higher than 600 K.

* Corresponding author.

E-mail address: tsuru@hiroshima-u.ac.jp (T. Tsuru).

<http://dx.doi.org/10.1016/j.ijhydene.2015.07.124>

0360-3199/Copyright © 2015, Hydrogen Energy Publications, LLC. Published by Elsevier Ltd. All rights reserved.

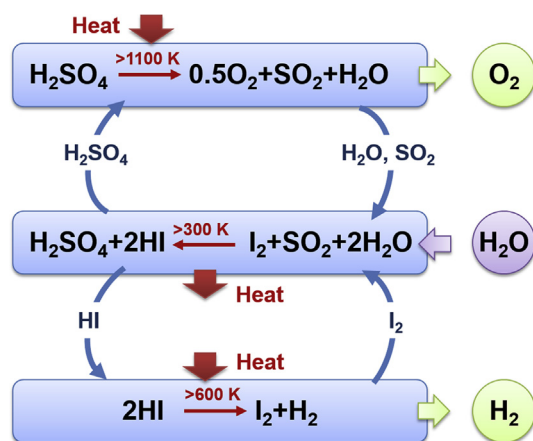
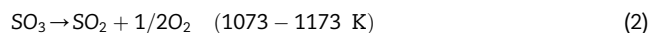
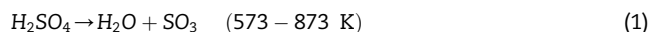


Fig. 1 – Schematic of the iodine–sulfur thermochemical water-splitting cycle.

The thermal decomposition of H_2SO_4 is actually divided into two sub-reactions:



Both of those reactions are highly endothermic and proceed smoothly without side reactions and with a high equilibrium conversion ratio at the temperature range indicated. Fig. 2 shows the effect of temperature on computed equilibrium concentrations of components obtained from the decomposition of 1 mol of pure H_2SO_4 [6]. The above two processes (1 and 2) were compared for dependence on temperature: the decomposition of H_2SO_4 to H_2O and SO_3 was predominant between 400 and 700 K while the reduction of SO_3 to SO_2 reached high conversions at temperatures higher than 1200 K [7]. Therefore, one key challenge for the IS process is the extremely high temperatures that are required for the decomposition of SO_3 to SO_2 and O_2 , which requires a substantial expenditure of energy. Recent reports have cited great potential for nuclear heat to power hydrogen production plants that utilize the IS process without producing

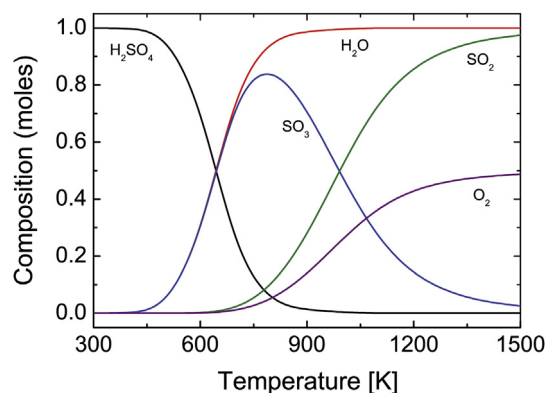


Fig. 2 – Temperature dependence of equilibrium composition for the decomposition of 1 mol of pure H_2SO_4 at 1 atm.

greenhouse gas emissions [8]. Under normal operating conditions, the output of a nuclear reactor can achieve temperatures of more than 1100 K [9]. A laboratory-scale, fixed-bed reactor with supported Pt/Pd catalysts was constructed and experimentally evaluated for SO_3 decomposition. At 1103 K, SO_3 conversions were equivalent to 0.60, which reached 80% of the equilibrium value [10]. Moreover, the development of catalysts with high catalytic activity and stability is also an important issue for the enhancement of SO_3 decomposition [11,12]. If SO_3 decomposition could attain a higher degree of conversion at medium temperatures (900–1000 K), which extend the catalyst lifetime, this coupled system could result in hydrogen production that is both highly efficient and economic.

A catalytic membrane reactor (CMR) is a combination of a permselective membrane and heterogeneous catalysts [13]. CMRs have the inherent capability and advantage of combining both a reaction and a separation in a single unit. The membrane provides selective removal of products in parallel with a reversible reaction, which shifts the equilibrium towards the product side and thus results in a higher reaction conversion even at lower temperatures [14]. Because equilibrium-limited reactions usually take place at high temperatures, the membranes used for CMRs are either metal or ceramic: palladium [15], dense perovskite [16], zeolite [17], silica [18], and alumina [19]. Considerable research effort has been devoted to the utilization of CMRs for catalytic reactions such as methane steam reforming [20] and water gas shift reactions [21]. Tsuru et al. [22] developed a bimodal catalytic membrane reactor for H_2 production via the steam reforming of methane. With H_2 extraction through the membrane, methane conversion was highly enhanced from 0.44 to 0.70 at 500 °C because of the equilibrium shift effect.

The modeling and simulation of CMRs has attracted increasing interest [23,24], with objectives that include modeling the flow as well as mixing and dispersion processes to develop appropriate guidelines for reactor design [25,26] and simulating the performance of an experimental reactor to validate a mathematical model [27,28]. Gallucci et al. [23] proposed a mathematical model for the simulation of a CMR in methane steam reforming, and the effect of parameters, such as reaction pressure, temperature, reactor length, and membrane thickness on methane conversion were theoretically investigated. Recently, a one-dimensional, isothermal, and plug-flow model for catalytic membrane reactors operated under cocurrent flow conditions [29] was applied to simulate several catalytic reactions, which included methane steam reforming [30], NH_3 decomposition [31], and the dehydrogenation of methylcyclohexane [32]. With a given value for catalyst weight, which was the single fitting parameter, the simulation results obtained from the CMRs with/without membrane extraction were in good agreement with the experimental investigation, which verified that the mathematical model was effective in predicting the performance of the CMRs. This kind of simple model is beneficial to an understanding of the mechanism of CMRs and to the development of an optimal design, including membrane performance (e.g., permeance and selectivity) and operation conditions such as temperature, pressure and feed flow rate.

Previous theoretical studies [33,34] have confirmed that a dense O_2 removal membrane could significantly increase the

Download English Version:

<https://daneshyari.com/en/article/1278786>

Download Persian Version:

<https://daneshyari.com/article/1278786>

[Daneshyari.com](https://daneshyari.com)