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Platinum–ceria–zirconia catalysts for hydrogen production in sulfur-iodine cycle

Yun Chen, Zhihua Wang*, Yanwei Zhang, Junhu Zhou, Kefa Cen

State Key Laboratory of Clean Energy Utilization, Zhejiang University, Hangzhou 310027, China

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ABSTRACT

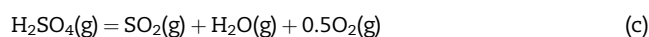
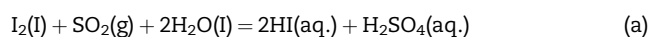
In this study, Pt/Ce_{1-x}Zr_x catalysts with different Zr mole concentration ($x = 0, 0.2, 0.5, 0.8, 1$) have been tested to evaluate their effects on hydrogen iodide (HI) decomposition for hydrogen production in the sulfur-iodine (SI or IS) cycle at various temperatures. The Pt/Ce_{1-x}Zr_x catalysts strongly enhanced the HI conversion to H₂ by comparison with blank test, especially the Pt/Ce_{0.8}Zr_{0.2} catalyst. BET, XRD, TEM, EDS, TPR were performed for catalysts characterization. It was found that, through introducing ZrO₂ into Pt/CeO₂, a synergistic effect between Pt and CeO₂-ZrO₂ solid solution was different from Pt and CeO₂ yield, such as improvement of the thermal stability and increase of Pt-O-Ce reducibility. Among the three samples containing Zr, the one with 20 mol% displayed the best activity for hydrogen production.

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1. Introduction

Hydrogen is an attractive fuel for the future because it is renewable and flexible as an energy carrier. In order to develop a hydrogen energy system, for which huge hydrogen demand is expected, the development of an efficient hydrogen production system is an urgent issue. Thermochemical water-splitting is one of the promising methods for large-scale hydrogen production, in which hydrogen is obtained by water decomposition through a chemical cycle process that consists of several reactions using heat energy from nuclear, solar energy and other kinds renewable energy source. In this process, all reactants are cycled except decomposed water. Further more, every reactions in the chemical cycle process occurs at acceptable relative lower temperature compared to direct thermal decomposition which is around 3000 K for water. Among the large-scale, cost effective and environmentally attractive hydrogen production processes, the sulfur-iodine (SI or IS) thermochemical cycle is considered as

a quite promising one after evaluating 115 thermochemical cycles by Sandia National Labs, General Atomics and University of Kentucky [1–3]. This cycle consists of the following three reactions:



The Bunsen reaction (a), which produces hydrogen iodide (HI) and sulfuric acid (H₂SO₄), is an SO₂ gas-absorbing reaction in aqueous phase. The two acids can be divided into light and heavy phases with a clear boundary by the liquid–liquid phase separation phenomenon in the presence of an excess I₂. The separated HI solution dissolves the excess I₂ and is denoted as the HIX phase. After purification, HI is separated from I₂ by

* Corresponding author. Tel.: +86 571 87953162; fax: +86 571 87951616.

E-mail address: wangzh@zju.edu.cn (Z. Wang).

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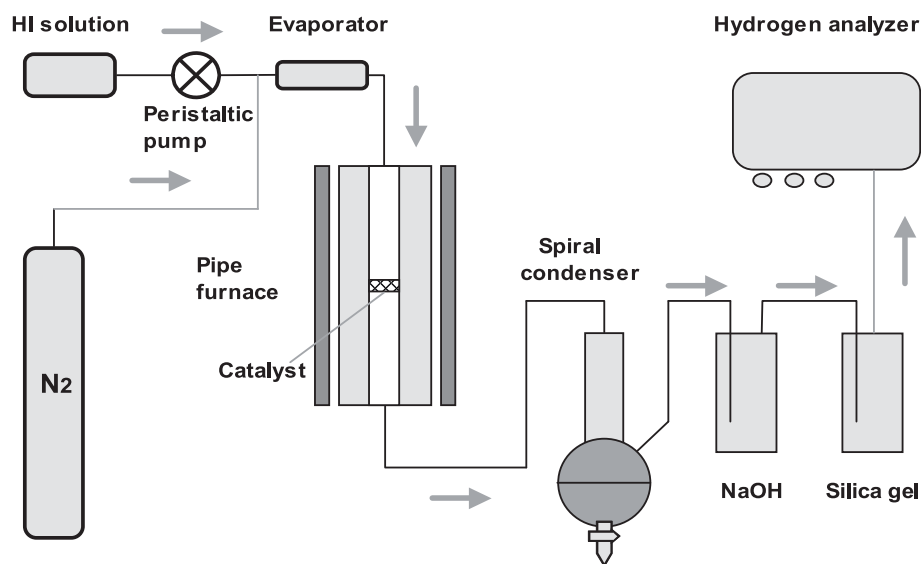


Fig. 1 – Schematic of experimental catalytic activity test system.

distillation, and then decomposed to produce H_2 and I_2 . Similarly, the separated H_2SO_4 solution denoted as the H_2SO_4 phase is purified, concentrated, vaporized, and decomposed to produce O_2 , H_2O and SO_2 . I_2 , H_2O and SO_2 are then recycled back to the Bunsen reaction (a). So only water is decomposed into H_2 and O_2 through the whole cycle. The reaction (b), HI decomposition, is one of the most significant reactions not only in this cycle but also in other thermochemical water-splitting cycles using iodine [4–6]. The homogeneous decomposition of HI could be used directly in a water-splitting scheme; however, to do so would involve relatively high temperatures to obtain workable reaction rate. The use of a catalyst allows a substantial reduction in temperature to achieve the same rate [7]. With regard to heterogeneous catalytic decomposition of HI, many works have been reported with different kinds of catalysts and various catalytic decomposition mechanisms [7–13].

Ceria is abundant, nontoxic and inexpensive material. As a member of rare earth elements, cerium has +1, +3 and +4 oxidation states that make cerium cations to be an attractive species as an electron acceptor or sacrificial agent since they can be oxidized and reduced reversibly. The promoting of ceria was originally believed to include structural aspects and chemical ones, like its participation in the water gas shift and steam reforming reaction or the enhancement of the noble metal dispersion and particular interaction with noble metals [14]. Many researches focused on the interaction between noble metals and pure ceria and found that, electron transfer

from the oxide to the noble metal resulted in a lowering of the effective activation energy [15]. CeO_2 and Pt/CeO_2 catalysts for HI decomposition have been researched by Zhang et al. [16–19]; found that the strong interaction between Pt and CeO_2 and oxygen vacancies could be very important for HI decomposition. It is reported that even small additions of ZrO_2 could decrease the bulk reduction energy of Ce^{4+} comparable to those reported for surface reduction in pure CeO_2 thus more oxygen vacancies are available [20]. Therefore, it is necessary to explore the catalytic activity of $Pt/Ce_{1-x}Zr_x$ catalysts for HI decomposition. Despite the fact that $Pt/Ce_{1-x}Zr_x$ catalysts are well known used as a kind of Three-way catalysts (TWCs) controlling the automotive exhaust pollution [14], its usage for HI decomposition in thermochemical water-splitting cycles has not been widely explored.

The main purpose of this work was to evaluate the performance of platinum–ceria–zirconia catalysts prepared by sol–gel method for hydrogen iodide catalytic decomposition to produce hydrogen. Especially the contribution of zirconium to catalytic activity was investigated in detail.

2. Experimental section

2.1. Catalyst preparation

The 2 wt% $Pt/Ce_{1-x}Zr_x$ catalysts were synthesized by the citric-aided sol–gel method. The nitrate $Ce(NO_3)_3 \cdot 6H_2O$ and

Table 1 – Routine sample characterization.

Catalysts	BET surface area (m^2/g)	Pore volume (cm^3/g)	Average pore radius (nm)	Lattice constants (\AA)	Crystallite size (nm)
Pt/Ce	10.21	2.73×10^{-2}	10.7	5.4082	30.3
Pt/ $Ce_{0.8}Zr_{0.2}$	28.85	7.25×10^{-2}	10.05	5.3382	7.4
Pt/ $Ce_{0.5}Zr_{0.5}$	20.25	3.82×10^{-2}	8.86	5.2596	6.1
Pt/ $Ce_{0.2}Zr_{0.8}$	53.96	5.81×10^{-2}	4.307	5.1545	5.1
Pt/Zr	93.93	1.15×10^{-1}	4.88	Not calculated	Not calculated

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