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Effects of various factors on the hydrogen isotope separation efficiency of a novel hydrophobic platinum-polytetrafluoroethylene catalyst

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ABSTRACT

The application of platinum supported on polytetrafluoroethylene (Pt/PTFE) as a composite catalyst for the separation of hydrogen isotopes holds much promise but warrants further refinement for improved performance. The objective of the present study was to examine the performance of a new hydrophobic Pt/PTFE catalyst during hydrogen-water exchange-based deuterium separation. The influence of diverse factors such as flow rate, column height, temperature, the volume ratio of filler to catalyst, and flow mode (co-current or counter-current), and so on, on catalytic performance was investigated. The deuterium conversion rate from co-current exchange was superior to that from counter-current exchange. Decreasing the hydrogen flow rate, increasing the feed water flow rate, and decreasing the molar flow ratio of hydrogen to water improved the deuterium conversion rate. In terms of layered filling of the catalyst column, adding more hydrophilic fillers improved the deuterium conversion rate. The characterization results highlight the high catalytic activity of the Pt/PTFE catalyst for hydrogen-water exchange, as well as its high stability in water.

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Introduction

Various techniques are applied for hydrogen isotope (protium, deuterium, and tritium) separation for a diverse range of applications [1–5]. Among hydrogen isotope separation technologies [6–14], CECE (combined electrolysis catalytic exchange) is widely favored and has been broadly applied in heavy water production, detritiation of nuclear fuel reprocessing wastewater [15], and the recovery of tritium from tritium system fusion reactor cooling water or waste water [16–18]. For successful CECE, selection of the catalyst is extremely important. Conventional hydrophilic catalysts lose their activity and become inefficient the more they come directly in contact with

liquid water or condensed steam. Ideally, to deter such inefficiency, the catalyst should be hydrophobic and be capable of transporting gas reactants and products to and from the catalytic active center.

Methods for the preparation of hydrophobic and hydrophilic catalysts are similar, but the nature of the support, the waterproofing approach, and the active metal deposition methods differ [19,20]. The first hydrophobic catalyst patent for water-hydrogen isotope exchange at low temperature was published in 1972 [21], after which the CECE process could be successfully accomplished.

The preferred hydrophobic catalyst employed for CECE is a Group VIII metal (platinum preferably) with a waterproofing organic polymer or resin coating (polytetrafluoroethylene

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preferably) that is permeable to water vapor and hydrogen gas [22–30].

Platinum (Pt) is broadly recognized as the most active metal used in catalysts, at either low or high temperatures. There are three main types of hydrophobic Pt catalyst used in the GECE process, a Pt/C/inert carrier (Pt/C/IC) [24,25], Pt/styrene divinylbenzene copolymer (Pt/SDB) [26,27], and Pt/C/polytetrafluoroethylene (Pt/C/PTFE) [28–30]. The Pt/C/IC catalyst is superior to the others in terms of chemical stability, strength, and catalytic activity, but its fabrication is complex. It is easy to control the size and shape of the Pt/C/PTFE catalyst, yet the utilization ratio of Pt is low, leading to a less efficient process. The catalytic activity of Pt/SDB is good, but the particle size is usually small and it is easily broken.

Fluorine covers the carbon backbone of the fluorocarbon PTFE, preventing unwanted reactions between the backbone bonds and aggressive media. The replacement of fluorine is improbable, as the bond between carbon and fluorine is very strong. PTFE is not soluble in most solvents, and chemicals that can attack PTFE are rare. Moreover, the thermal stability of PTFE is high, as it is highly crystalline [31,32]. With these particular properties, PTFE is a very good waterproofing agent.

Considering the favorable potential of Pt/C/PTFE catalyst constituents, improving the performance of Pt/C/PTFE catalysts is highly desirable. We previously fabricated a hydrophobic catalyst comprised of Pt directly supported on porous PTFE (Pt/PTFE) [33]. Compared with a Pt/SDB catalyst, the Pt/PTFE catalyst had basic catalytic activity, exhibited high mechanical strength, a low pressure drop, and was prepared through a simple molding compression method. It also showed good resistance to heat and pressure, as well as a long productive life. The performance of a catalyst in different environments is key to evaluate its applicability. Based on our previous work, the presented study aimed to fabricate a Pt/PTFE catalyst, evaluate its performance under various conditions, and to optimize process parameters for maximum catalyst performance. The effects of diverse factors such as gas and water flow rates, column height, temperature, time, the volume ratio of filler to catalyst, and flow mode on the deuterium conversion rate were examined in detail.

Experimental

Materials

Chloroplatinic acid (H_2PtCl_6) (Shanghai Shiyi Chemical Reagent Co., Ltd., China), PTFE (Zhejiang Jusheng Fluorochemical Co., Ltd., China), and Naphthalene (C_{10}H_8) (Tianjin Kermel Chemical Reagent Co., Ltd., China) were used as received. Hydrogen gas and nitrogen gas were sourced from Dalian Guangming Special Gas Chemical Research Institute. Distilled deionized water was used in all experiments.

Catalyst support preparation

Compression molding was used to prepare the Pt/PTFE catalyst support. Seventy percent PTFE suspension resin (40 mesh) was uniformly mixed with 30% C_{10}H_8 (40 mesh). Then, the mixtures were molded to 2–3 mm thick and 25 mm diameter

dimensions using a hydraulic molding press (Tianjin Gaoke New Technology Development Co., Ltd., China). The formed product was dried in an oven set to between 323 K and 373 K for 24 h to remove the pore-forming agent, followed by sintering at high temperature in the range of 603 K–653 K for 14 h. The resulting catalyst support was sheared into small samples (about 3 mm wide, 3 mm long, and 1.5 mm thick).

Platinum deposition

Platinum was deposited onto the catalyst support by impregnation with acetone. The catalyst contained 0.8 wt% Pt. Then, the semi-finished products were reduced by H_2 , at 523 K–553 K for 3 h.

Catalytic activity of the hydrophobic catalyst

A pilot device was set up to test the performance of the catalyst. The pilot device facilitated the testing of hydrogen-water isotopic exchange in co-current or counter-current conditions.

The flow diagram of the pilot device is shown in Fig. 1. The main part of the device, similar to that previously described [33], was the catalyst column (20 mm diameter, 600 mm length) with an outer heating jacket and a hydraulic guard at the bottom. The catalyst column was filled with catalyst and filler (Dixon ring, 2×2 mm) layers. Nitrogen gas was used to purge the system in order to prevent build-up of explosive hydrogen gas.

Hydrogen with 284 ppm hydrogen deuteride (HD) was preheated to the same temperature as the water and was then fed in at the bottom or the top of the catalyst column. The hydrogen, co-current or counter-current to water, went through the catalyst bed to enable isotope exchange. The water with 300 ppm semi-heavy water (HDO) entered in through the top of the catalyst column. The hydrogen was released and the condensed steam was collected.

The catalytic performance was evaluated by the conversion rate η (%):

$$\eta = (y_{\text{in}} - y_{\text{out}}) / y_{\text{in}} \quad (1)$$

where y is the molar fraction of deuterium in the gas phase, and in and out refer to the inlet and the outlet, respectively.

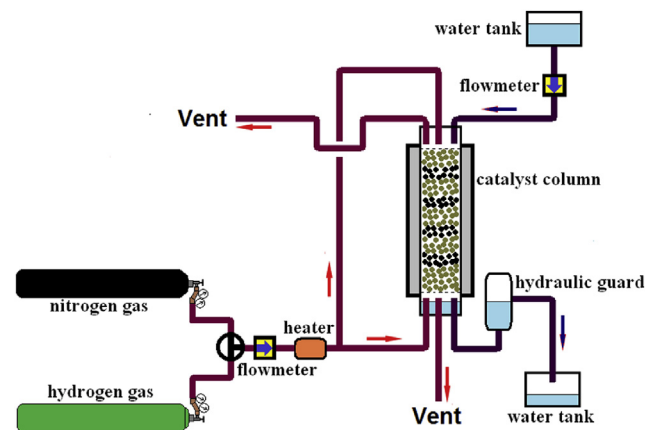


Fig. 1 – Experimental devices.

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