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Preparation and catalytic properties of honeycomb catalyst for hydrogen isotope oxidation

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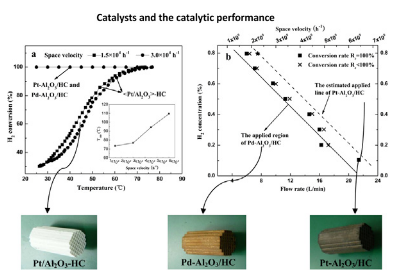
HIGHLIGHTS

- Honeycomb catalysts with good physical properties were prepared for detritiation.
- The catalysts increase gas flow rate significantly without decreasing the conversion rate.
- The catalysts were used at room temperature with high H_2 conversion rate.
- The confines of H_2 concentration and flow rate for catalyst application were tested.

GRAPHICAL ABSTRACT



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ABSTRACT

The method of catalytic oxidation and adsorption is widely used for atmosphere detritiation. But traditional particle catalyst has large gas resistance, which limited the space velocity for detritiation. Honeycomb catalyst can enormously increase the gas handling capacity due to its low pressure drop and high dispersity of active ingredients, but has not been used in detritiation so far. A coating of alumina was deposited on the honeycomb substrate of cordierite using ultrasonic technology. By the method of excessive impregnating, noble metal (Pt or Pd) supported catalysts were prepared. The catalysts were characterized by X-ray diffraction (XRD), N_2-adsorption/desorption (Brunauer–Emmet–Teller – BET method), scanning electron microscope (SEM) and laser particles sizer. The result shows that the alumina coatings are well distributed, well knitted and the specific surface area of honeycomb catalyst rises to about 20 m^2/g. Catalytic activities were evaluated by H_2 conversion rate in gas mixture (with different H_2 concentration and various flow rates). The results indicated that all catalysts exhibited excellent catalytic performance for H_2 oxidation; the conversions of hydrogen were 100% at room temperature when the gas space velocity was up to 6×10^5 h^{-1}.

1. Introduction

In nuclear fusion reactor facilities, multi-confinement system is applied to prevent tritium leaking to the environment, because tritium is permeable and radioactive. The last barrier, a building containing all equipment and facilities, must typically involve a secure air cleanup system (ACS) or an atmosphere detritiation system (ADS) in order to prevent tritium leaking to the environment when a severe accident takes place [1–5]. In ACS, the gaseous tritium in air is oxidized by catalysts, and then tritiated water is collected by adsorbents [6–12]. This method can remove tritium effectively, but high pressure loss in catalyst bed becomes a big problem when the gas velocity is large. Traditional packed beds filled with particle catalyst have large gas resistance, which

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results in significant pressure loss and low gas cleanup capability. Whereas, honeycomb catalysts can substantially decrease the pressure loss by 1–2 orders of magnitude [14,15], thus reduce the volume of catalytic bed. Moreover, honeycomb catalyst is a combination of catalyst, carrier and reactor, so it is easy to maintain, replace and exchange.

Uda, Tanaka, Munakata, etc. from National Institute of Fusion Science (NIFS) of Japan and Akita University respectively have carried out much R&D for honeycomb catalysts used in ACS or ADS [13–17]. The results indicated that honeycomb catalyst can significantly increase the gas flow rate without decreasing the conversion rate of hydrogen and its isotopes, while the system is easy to scale up, which is satisfied for applications. We prepared catalysts of Pt/Al_2O_3-HC (Pt loaded on the honeycomb substrates coated with γ-Al_2O_3) and Pd/Al_2O_3-HC with different methods and the catalytic performance is tested with hydrogen at very high space velocity, which is the highest in the reference. And the region at which the catalyst can be used with 100 percent conversion rate without heating is obtained. The region is of importance for application.

2. Materials and methods

2.1. Catalyst preparation

2.1.1. Coating of substrates

Before the process of coating, honeycomb substrates of cordierite are first dipped into the solution of nitric acid (10 wt%) and hold for 24 h. After washing with distilled water for three times and dried at 120 °C for 3 h, the pretreated substrates are weighed and immersed in alumina sol or slurry. The alumina sol is made from hydrolyzing of aluminum isopropoxide and the viscosity maintains 11 mm^2/s. The alumina slurry is obtained from milling the mixture of aluminum powder and distilled water (or alumina sol) for 10 h, and the rev is 350 r/min. The substrates are immersed in the sol or slurry with or without the assistant of ultrasound for 15 min, and then fished out, blew off the superfluous slurry by compressed gas. Dried at 120 °C for 3 h, weighed and then the substrates are calcined at 500 °C for 3 h. The process of coating repeats for three times.

2.1.2. Loading of noble metal

The solution of $PdCl_2$ and $H_2PtCl_6{\cdot}6H_2O$ with mass concentration of 0.5% is confected, and the PH value is regulated to 2.0 by dripping diluted nitric acid. Appropriate quantity of solution is used according to the volume of substrates (planed loading rate of noble metal is 2 g/L). Coated substrates are impregnated into the solution and hold for 15 min, and then fished out, blew off the superfluous solution and dried (120 °C) for 2 h. Repeat the impregnation until the solution is used up, and after the last time, the loaded substrates are calcined at 500 °C for 3 h.

2.2. Test of physical properties

XRD is used to identify disperse of noble metal, and SEM to observe the surface of substrates with or without alumina coatings. AUTOSORB-1-C Physic/Chemisorption Analyzer manufactured by Quantachrome Co. Ltd. is used to measure the specific surface area of substrates and catalysts. The laser particle sizer is employed to analyze the average particle size of alumina sol and slurry.

The adhesion between alumina coatings and cordierite substrates are evaluated by the loss rate after the ultrasound test. Coated substrates are dipped into distilled water and vibrated by ultrasound for 15 min, the power and frequency is set 100 W and 40 kHz respectively. After thoroughly dried the coated substrates

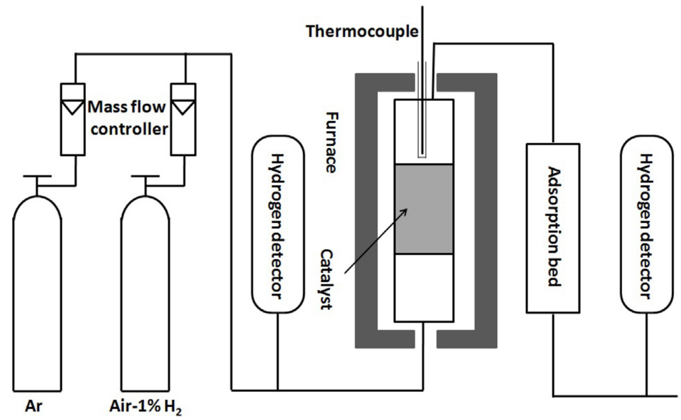

Fig. 1. Schematic diagram of catalytic activities test system.

are weighed and the loss rate Δm (%) of coating is defined in the following equation:

$$\Delta m = \frac{m_3 - m_1}{m_2 - m_1} \times 100$$

where m_1 and m_2 are weights of uncoated and coated substrates, respectively. And m_3 is the weight of coated substrate after the test.

2.3. Catalytic performance test

Before the test, the catalysts are heated up to 350 °C under pure hydrogen for reduction. Fig. 1 shows the experiment apparatus used in this study. Deuterium is used as a reactant, and dry air as a carrier gas. The deuterium concentration of the inlet gas can be controlled by the mass flow controllers. Two hydrogen detectors are used to measure the deuterium concentrations of the inlet and outlet of the reactor. The conversion rate R_c (%) and space velocity S_v (h^{-1}) are defined as:

$$R_c = \frac{C_{in} - C_{out}}{C_{in}} \times 100$$

$$S_v = \frac{Q}{V}$$

where C_{in}, C_{out}, Q and V are hydrogen concentration of inlet and outlet of the reactor, gas velocity to the reactor (m^3/h) and apparent volume of the catalyst (m^3), respectively. C_{in} is set to 0.8% in the test.

3. Results and discussion

3.1. Coating

Honeycomb substrates of cordierite are coated by alumina sol or slurry, coating rate and the specific surface area of coated substrates are shown in Table 1. The specific surface area of substrates is significantly increased by coating with γ-Al_2O_3, which is propitious to load the noble metal. The coating rate indicates that coating with slurry is easier than sol, for the coating rate is over

Table 1
The coating rate and specific surface area of substrates coated by different methods.

Samples	Coating rate (%)	Specific surface area (m^2/g)
Uncoated substrate	0	0.75
Coated with sol	6.37	18.66
Coated with slurry	14.08	20.71