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Preparation of alveolate hydrophobic catalyst for tritium waste gas treatment

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HIGHLIGHTS

- The catalyst is hydrophobic, it will not be poisoned by steam in room air at room temperature which is better than Pt-Al_2O_3.
- At room temperature, the conversion of low concentration of H2 and tritium gas in room air over the catalyst is high.
- The air resistance of catalyst is much lower than graininess Pt-Al_2O_3.
- It is inorganic and will not burn.

GRAPHICAL ABSTRACT

The alveolate hydrophobic catalyst was essentially waterproof and not easily poisoned by moisture. At room temperature, the conversion of 1% H_2 in humid air over the catalyst was higher than 95% at 0–16000 h^{-1} and 0–100% relative humidities. The reaction rate constant of the oxidation of tritium in humid room air over alveolate hydrophobic catalyst at room temperature is much higher than normal honeycomb catalyst.

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ABSTRACT

To prepare a catalyst for the detritiation of waste gases at high flow rates, a heat-resistant hydrophobic zeolitic molecular sieve coating was synthesized on the surface of alveolate cordierite by hydrothermal processing. The alveolate hydrophobic catalyst prepared from the support was essentially waterproof and not easily poisoned by moisture. At room temperature, the conversion of low concentrations of H_2 in humid air over the catalyst was higher than 95% at different space velocities (0–16,000 h^{-1}) and different relative humidities. The reaction rate constant of the oxidation of tritium over alveolate hydrophobic catalyst is 0.182 s^{-1} at 293.3 K–293.7 K and 59%–60% RH, it is much higher than the catalyst of reference honeycomb catalyst.

0. Introduction

Tritium is an important fusion fuel, but its use presents the problem of handling huge volumes and low concentrations of tritium

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waste gas exhaust. However, because tritium is a radioactive gas, waste gases that contain it must not be exhausted without proper disposal. The oxidation of tritium gas into tritiated water over a catalyst and the recycling of the tritiated water are generally used detritiation techniques in tritium waste gas disposal; the catalyst is key to the success of the approach. Excluding high efficiency catalysts, the pressure difference should also be considered. High air resistance in the catalyst will make air circulation difficult, so the air resistance of catalyst should be low. In recent years, the Japan National Institute for Fusion Science [1,2] has used a hydrophilic honeycomb-shaped catalyst manufactured by Tanaka Kikinzoku Co. Ltd for high-flow tritium waste gas disposal; the pressure drop of the catalyst is obviously lower than the pellet catalyst, which is beneficial for the safe operation of the detritiation system.

Volumes are another factor to consider: because of huge volume of catalyst (from 10^0 m^3 to 10^1 m^3) and high air flow (from 10^3 m^3/h to 10^5 m^3/h) which will carry off heat, the heating of a large catalyst bed will be much more difficult than a small catalyst bed. The tritium waste gas will be heated to higher than 373.2 K in catalyst bed, then the gas must be cooled to lower than normal temperature in order to absorb the HTO in absorbent bed. Before the tritium waste gas can be exhausted, the gas must be detritiated repeatedly, the repeatedly heating and cooling will consume a lot of energy. Thus, the catalyst should have good efficiency at normal temperature. This requires the catalyst to be hydrophobic, because moisture will block the active sites of the catalyst at normal temperatures [1–4]. For example, the hydrophilic honeycomb-shaped catalyst at the Japan National Fusion Science Institute must be used at temperatures higher than 373.2 K [5,6].

In addition, a small amount of hydrogen is often added in the air flow to elevate the catalyzed tritium oxidation efficiency, but adding hydrogen raises the temperature of the catalyst surface. Because organic hydrophobic catalysts are fairly combustible, a hydrophobic inorganic supported catalyst which is able to endure high temperature is needed.

Based on these considerations and with reference to related literature [7–9], this paper describes the transformation of honeycomb-shaped cordierite ($Mg_2Al_4Si_5O_{18}$, which is hydrophilic and has low air resistance) into a hydrophobic support, from which an inorganic hydrophobic catalyst which can endure high temperature was prepared.

1. Experimental

1.1. Modification of the support

To prepare the honeycombed inorganic hydrophobic support for use as a high flow detritiation catalyst, silicon source and the structure-directing agent were used to synthesize a hydrophobic zeolite film on honeycomb cordierite by the hydrothermal synthesis method. The substrates were cordierite ($2MgO{\cdot}2Al_2O_3{\cdot}5SiO_2$) honeycombs having cell densities of 100 cells per square inch (cpsi) and wall thicknesses of 480 μm. To prepare the support, silicon source, additive agent and NaOH were added into a reaction kettle and thoroughly mixed until uniform. Then, water and the structure-directing agent were added with stirring. The honeycomb cordierite was added into the kettle, and the reaction vessel was sealed and heated in an oven for 48 h for the hydrothermal synthesis at 453.2 K. The resulting material was submerged in distilled water and ultrasonicated for 30 min, and was then dried for 2 h at 393.2 K. This was followed by calcination at 873.2 K to remove the structure-directing agent residues, which afforded the hydrophobic honeycomb inorganic support.

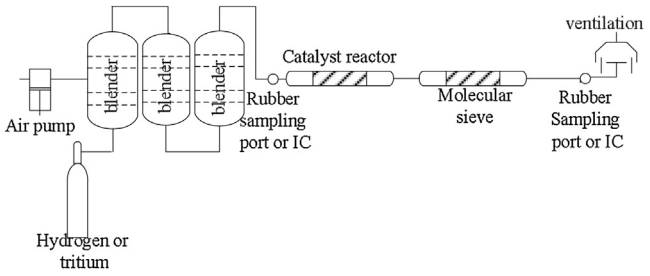

Fig. 1. Flow diagram for the catalyzed oxidation of hydrogen(tritium) over the hydrophobic catalyst.

1.2. Preparation of the catalyst

The hydrophobic honeycomb inorganic support was mixed with a solution of $HPtCl_4$ at a 1.37% mass ratio of Pt to the support. The material was soaked for ~3 h while ultrasonicated at room temperature; then, the temperature was increased to 333.2 K, and the acetone was moisturized by ultrasonication. The sample was deoxidized under a flow of 10% H_2/N_2 mixed gas at 433.2 K for 20 h to product the catalyst.

1.3. Support and catalyst characterization

The support and catalyst were characterized by scanning electron microscopy (SEM) and X-ray diffraction (XRD). SEM images of the support were observed using a JEOL JSM-5900LV scanning electron microscope at 20 kV. Detailed structures of the catalyst were examined using an FEI Inspect F SEM instrument at 20 kV. The XRD spectra were recorded using a Philips X'PERT PRO MP instrument with Cu Kα radiation, 40 kV tube voltage, and scan step of 0.03° at a rate of 0.2 s/step.

The surface contact angle of support and catalyst was observed using a Krüss G2 instrument.

The static absorbed water of the hydrophobic honeycomb catalyst was tested with the method of Chinese standard: *GB 6287-86: Determination of static absorbed water for molecular sieve.*

1.4. Activity of the catalyst

The catalytic activity was tested in the oxidation of hydrogen or hydrogen mixed with tritium gas or tritium mixed with Ar; a schematic diagram of the process flows is shown in Fig. 1. The concentration of hydrogen was measured by gas chromatograph (GC) (Varian cp-3800 gas chromatograph), and the concentration of tritium was determined in an ionization chamber (IC) (home-made).

The test honeycomb catalyst (the Pt content is 1.37%) is Φ4 cm*2.5 cm, 6 catalysts (188.4 cm^3) is put into the catalyst bed when the space velocity of gas is from 0 to 16000 h^{-1}, 4 catalysts (125.6 cm^3) is put into the catalyst bed when the space velocity of gas is 24000 h^{-1}, 3 catalysts (94.2 cm^3) is put into the catalyst bed when the space velocity of gas is 32000 h^{-1}, The gas flow is 0–3 m^3/h. The average particle size of Pt-Al_2O_3 (Shanxi Kaida Chemical Engineering Co., Ltd) is Φ0.25 cm–Φ0.35 cm, the Pt content is 1.4%, Pt-Al_2O_3 is filled in the catalyst bed uniformly, the bulk density is 0.62 g/ml, the volume of the Pt-Al_2O_3 bed is Φ4 cm*15 cm (188.4 cm^3), the gas flow is 0–0.6 m^3/h, the space velocity of gas is from 0 to 3370 h^{-1}. The size of the 3 blenders is Φ10 cm*50 cm. The humidity is the room air humidity, the room air humidity is range from 30% to 84% which is measure by a humidometer (RS 408-6109 hygrometer. in/out thermo), if higher humidity is needed, a water bubbler is added before the gas enter the reactor.

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