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# Hydrophobic platinum–polytetrafluoroethylene catalyst for hydrogen isotope separation

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## ABSTRACT

A composite catalyst, platinum supported on polytetrafluoroethylene (Pt/PTFE), has been successfully prepared by compression moulding forming and used for hydrogen isotope separation by hydrogen–water isotope exchange. The as-prepared Pt/PTFE was characterized by nitrogen adsorption. The results of the catalytic activity for hydrogen–water isotope exchange show that Pt/PTFE has high catalytic activity. The effects of different factors, such as flow rate, temperature, molar flow ratio of hydrogen gas to feed water and time have also been investigated. The present study shows a promising choice of Pt/PTFE as a composite catalyst for hydrogen isotope separation.

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## 1. Introduction

Three hydrogen isotopes, protium, deuterium, tritium, have been widely used in nuclear energy, military affairs and industries. The subject of separation and purification of deuterium and tritium has attracted much interest of scientists in the whole world [1–5]. Various technologies have been developed for the separation of hydrogen isotopes, such as, chemical exchange, liquid hydrogen distillation, cryogenic adsorption [6], palladium or palladium membrane diffusion [7–9], thermal diffusion [10–12], laser separation and electrochemical isotope separation [13] etc. Among them, the chemical exchange has been widely applied in producing and upgrading heavy water, removing tritium from light water or heavy water, and recovering tritium for fusion reactor.

Most of the world's heavy water supplies are currently provided by the Girdler-Sulphide (G-S) process or process based on ammonia–hydrogen catalytic exchange [14]. For the two processes, the ammonia process has size limitations and the G-S process consumes large amounts of energy and

utilizes very hazardous hydrogen sulphide. Accordingly, a number of alternative processes which adopt hydrogen isotope exchange between water and hydrogen using a catalyst have been proposed because of its mild operating conditions and a high separation factor.

One such process is known as Combined Electrolysis and Catalytic Exchange (CECE). The CECE process is a single thermal process that extracts heavy water from normal water by a combination of electrolysis and catalytic exchange between water and hydrogen. Another process to utilize water–hydrogen exchange is the Bithermal Hydrogen Water (BHW) process. The BHW is a process using liquid phase catalytic exchange (LPCE) to generate heavy water.

The CECE and LPCE processes rely upon hydrophobic catalysts to catalyze the exchange reaction between hydrogen gas and liquid water. The mixed catalyst packing contains, both a noncatalytic hydrophilic packing that enhances the exchange between water and water vapor, and a hydrophobic catalyst that accelerates the hydrogen–water isotope exchange. The platinum type catalyst has been widely used,

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however, it becomes inefficient after long contact with liquid water or by capillary condensation of water vapor [15]. Therefore, it is necessary to prepare a hydrophobic catalyst, which allows the transport of the gaseous reactants and reaction products to and from catalytic active centers. More importantly, the catalyst should be suitable for the dual temperature exchange.

In 1972, Stevens published the first patent for a hydrophobic catalyst for water–hydrogen catalytic exchange at low temperature [16]. So that, the liquid phase catalytic exchange can be carried out successfully. By several decades of research and development, there are mainly three types of hydrophobic catalyst used in the liquid phase catalytic exchange process now, including a Pt/C/inert carrier (Pt/C/IC) [17,18], a Pt/C/polytetrafluoroethylene (Pt/C/PTFE) [19,20] and a Pt/styrene-divinylbenzene copolymer (Pt/SDB) [21,22]. The Pt/C/IC has high strength, good chemical stability and strong activity, and at the same time has complex forming technique. The size and shape of the Pt/C/PTFE are easily controlled, whereas the utilization ratio of platinum is low. The Pt/SDB has good activity, yet it shows low strength and small particle size.

Thus, there remains a need for a highly active and very stable catalyst with simple manufacture process. In view of the influence of temperature and pressure in dual temperature exchange, two types of hydrophobic catalysts with different supporting body were investigated at various test conditions. The first catalyst is designed for upper cold tower where the deuterium transfers from hydrogen to water. This modified Pt/SDB has stronger activity and higher strength than conventional Pt/SDB. The second catalyst is designed for a lower hot tower where the deuterium transfers from water to hydrogen gas. This is a new composite catalyst consisting in platinum directly supported on Teflon (Pt/PTFE). This new catalyst has higher strength, lower pressure drop and simpler forming technique than other catalysts besides basic catalytic performance. Due to these properties, this catalyst possesses resistance to heat, resistance to pressure and long life. The present work mainly focuses on the performance evaluation of the catalysts.

## 2. Material and methods

### 2.1. Experimental flow scheme

Fig. 1 shows a flow schematic diagram of the experimental apparatus, which mainly consists of a nitrogen gas source, a hydrogen gas source, a catalyst column and a chromatograph.

### 2.2. The preparation and the process of the experiment

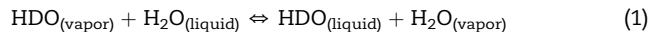
For the isotopic exchange reaction between the hydrogen gas and the liquid water, two types of the hydrophobic catalysts have been obtained, i.e. platinum on Teflon (Pt/PTFE) and platinum on styrene-divinylbenzene copolymer (Pt/SDB). The Pt/SDB catalyst support is fashioned by droplet agglomeration method, using a benzene solution of divinylbenzene to form small spheres. The Pt/PTFE catalyst support is prepared by

compression moulding forming. Both types of catalysts consist of 0.8 wt% Pt deposited on the catalyst supports. Three catalysts are compared mutually in this paper. They are Pt/SDB-1 (1 mm spherical), Pt/SDB-3 (3 mm spherical) and Pt/PTFE (3 mm length, 3 mm width and 1.5 mm thickness).

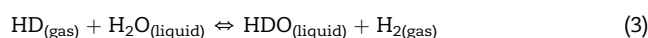
The reactor column is a Pyrex glass tube with an internal diameter of 20 mm and 40 cm length. The column is equipped with a water jacket, through which the thermostated water flows to maintain a constant temperature along the column, and a hydraulic guard installed at the bottom. The column is filled with catalysts. The system is initially purged with nitrogen gas to prevent hydrogen explosion. The hydrogen gas is electrolytic hydrogen with 284 ppm HD which is preheated in the heater, and fed into the bottom of the catalyst column. The deuterated water is distilled water with 300 ppm HDO which is fed in at the top of the column. The hydrogen, flowing in counter-current with the water, passes through the catalyst bed where the isotopic exchange takes place. The hydrogen stream is released from the top of the column and the condensed water vapor returns to the column.

## 3. Theoretical background

The overall catalytic exchange of hydrogen and deuterium between liquid water and hydrogen gas consists of the following two-step reactions [23]:



which totalized becomes the isotopic exchange:



The first equation expresses the vapor–liquid equilibrium, that is, the transfer of deuterium from water vapor to liquid water accompanied by evaporation and condensation. The second equation is the transfer of deuterium between hydrogen and water vapor. The second reaction occurs only on the surface of the catalyst, whereas the first reaction takes place at any gas–liquid interface.

As measures of the degree of separation, the conversion rate  $\eta$  (%) is defined as follows:

$$\eta = (y_b - y_t)/y_t \quad (4)$$

where  $y$  is a molar fraction of deuterium in gas phase. The subscript “b” and “t” mean the bottom of the catalyst column and the top of the catalyst column.

When the molar fraction of deuterium in liquid phase  $x$  is very low, the  $y_e$  is expressed as

$$y_e = x/\alpha \quad (5)$$

where  $y_e$  is a molar fraction of deuterium in gas phase in equilibrium,  $\alpha$  is the separation coefficient which could obtain from a empirical formula as follows:

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