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Experimental investigation on hydrogen adsorption performance of composite adsorbent in the tank with high vacuum multilayer insulation

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ABSTRACT

Hydrogen is one of the main residual gases in the high vacuum multilayer insulated tank. H_2 adsorption performance of the composite adsorbent was investigated by using a test bench, an X-ray diffraction and a scanning electron microscopy.

The composite adsorbent is composed of molecular sieve 5A and getter that includes the different proportion of PdO and Ag_2O. The getter shall is laid as flat as possible when fed into the tank. Leakage and outgassing rate can be decreased by 64% after placing getter. The crystal phase structure of PdO and Ag_2O in getter is unchanged by adsorption performance. Experimental results showed that the H_2 adsorption rate is high at the initial stage, and then it starts to become slow during a relatively long period. The type IV isotherms were obtained with these samples, and the H_2 adsorption principle is also discussed. The optimum percentage content of Ag_2O in the getter is 22%. Under the allowed highest pressure, the composite adsorbent that includes 15% Ag_2O, 85% PdO and molecular sieve 5A is preferentially used in the tank interlayer. The experimental results and performance analyses can be used in the design of the high vacuum multilayer insulated tank.

1. Introduction

It has been a haunting concern for designers as to how to obtain and keep the required interlayer pressure in the high vacuum multilayer insulated tank. The adsorbents in the interlayer play a very important role in the process. The interlayer pressure depends largely on the characteristics of the adsorbents. In addition, how to achieve its adsorption adequately is also important. The adsorption performance of adsorbents can be illustrated effectively by the adsorption isotherm. Under the vacuum and thermal environment conditions, the metal materials and multilayer insulated materials will deflate. Based on much research on the compositions of the gases generated by leakage and outgassing, the results show that the amount of H_2 exceeds 70% [1–3]. At temperature of liquid oxygen and liquid nitrogen, H_2 adsorption capacity of molecular sieve 5A and activated carbon is very small. Therefore, the major problem is the residual hydrogen in the tank. An effective H_2 getter is necessary in vacuum technology. Zr-based non-evaporable getters are widely used in high vacuum applications. However, these getters cannot be used in the high vacuum multilayer insulated tank due to their limited adsorption capacity and the need to be activated at relatively high temperatures (>300 °C) [4]. The use of transition metal oxides is promising for these purposes. It has been reported that PdO is an outstanding hydrogen getter for a vacuum tank [5–7]. So far, little work has been performed to study the effect of H_2 adsorption performance of getters on the interlayer pressure. The experiential and semiempirical design methods are used to select the related adsorbents, amounts of PdO, etc. For instance, it is found in the field survey that Zhangjiagang CIMC Sanctum Cryogenic Equipment Co. Ltd and Chart Cryogenic Engineering Systems (Changzhou) Co. Ltd used, respectively, 1 g and 0.75 g getters in the 175 l cylinder. Although the amount is different, the same effect is achieved. Expenditure is greatly reduced in the latter case. Therefore, it is urgent to grasp accurately H_2 adsorption performance of adsorbents.

The main objective of this work is to study H_2 adsorption performance of the composite adsorbent in the tank. Firstly, leakage and outgassing rate of the tank before placing the getter is compared with the rate after placing the getter. Secondly, H_2 adsorption of composite adsorbent is studied. The quantitative analysis and computation are carried out by using H_2 adsorption isotherm, which is obtained at room temperature. Thirdly, morphologies and crystalline phases of the getter are analyzed by

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Nomenclature

2θ	diffraction angle, °
α	H_2 adsorption capacity, ml (STP)/g
a, b, c	lattice constant, nm
g	leakage and outgassing rate, $Pa\cdot m^3/s$
H	H atom of surface adsorption
$H_{2,(p)}$	H_2 molecule of physical adsorption
P	equilibrium pressure, Pa
ΔP	variation of the interlayer pressure during Δt, Pa
S	adsorption site of Pd surface
STP	standard temperature and pressure, 273 K and 1.013×10^5 Pa
Δt	time interval ($= 8.64 \times 10^{-4}$), s
V	interlayer volume ($= 41.07 \times 10^{-3}$), m^3
x^{α}_{max}	maximum amount of hydrogen adsorbed in Pd, forming a dilute solid solution, αH/Pd
x^{β}_{min}	minimum amount of hydrogen adsorbed in Pd, forming β-hydride phase, H/Pd

using scanning electron microscopy (SEM) and X-ray diffractometry (XRD) respectively. The intention of the research is to obtain the principle for H_2 adsorption.

2. Experimental procedure

2.1. Samples

The composite adsorbent is composed of molecular sieve 5A and getter. Molecular sieve 5A is selected due to its unique adsorption characteristics and ability to trap contaminated molecules. The getter includes different proportions of PdO and Ag_2O. More details about the composition and condition of samples are shown in Table 1.

2.2. Experimental setup

The experimental apparatus is shown in Fig. 1. H_2 adsorption is studied by means of the pressure drop in a vacuum tank, whose volume is known. H_2 consumption is determined by measuring the decreased H_2 pressure. The apparatus is mainly composed of the following parts.

(1) The high vacuum multilayer insulated tank: The tank is a cylindrical stainless-steel column, with internal and external diameters of 200 and 300 mm respectively, and internal length of 750 mm. Not only the experiment of the leakage and outgassing rate can be carried out in the vacuum tank, but also H_2 adsorption kinetics and isotherm can be measured.

Table 1
Composition and condition of samples.

Sample	Composite adsorbent		Mass of getter (g)	Placement method of getter	Manufacturer
	Getter	Adsorbent			
1#A	100% PdO	Molecular sieve 5A	0.9813	Pack	Shanghai Elegant Molecular Sieve Co. Ltd
1#B	100% PdO	Molecular sieve 5A	0.9984	Flat	Shanghai Elegant Molecular Sieve Co. Ltd
2#	85% PdO, 15% Ag_2O	Molecular sieve 5A	1.0281	Flat	Shanghai Elegant Molecular Sieve Co. Ltd
3#	70% PdO, 30% Ag_2O	Molecular sieve 5A	1.0375	Flat	Shanghai Elegant Molecular Sieve Co. Ltd

(2) H_2 cylinder: Hydrogen used in H_2 adsorption is a highly pure gas (99.999%). The quality of H_2 is in accordance with the relevant regulations of GB/T7455-1995.
(3) Calibration container: This has three ports that are connected with the vacuum unit, H_2 cylinder and the tank respectively. The decreased amount of H_2 can be measured by the calibration container.
(4) Vacuum unit: This includes a mechanical pump and a molecular pump. The tank and the calibration container are evacuated by vacuum unit. In addition, the pressure of the calibration container is also controlled by it.
(5) Measurement system: This composed of an intelligent manometer and two sets of compound vacuum gauges (ZDF-5227).

2.3. Experiments on H_2 adsorption

The first step is the preliminary stage of H_2 adsorption. Approximately 1 g of getter was first fed into the tray in the tank interlayer. The volume of the interlayer was carefully calibrated. Molecular sieve 5A of 800 g was then divided evenly into two groups, which were placed on the upper and lower heads of inner cylinder in the tank interlayer respectively. The tank was evacuated by vacuum unit to 1×10^{-2} Pa at room temperature. The pretreatment of composite adsorbent was conducted, because there might be some contamination on their. At the same time, molecular sieve 5A was activated. So the sample was preheated continuously under vacuum for 1 day at a higher maximum temperature of no more than 150 °C. Afterwards, the tank was cooled down to room temperature.

The second step is the measurement of the leakage and outgassing rate. Firstly, leakage detection was carried out on the tank with a helium mass spectrometer. Then the experiment of the leakage and outgassing rate couldbe performed in the tank within permissive leakage.

At the third step, H_2 adsorption was carried out in the vacuum interlayer. The detailed procedure of H_2 adsorption is as follows: Firstly, the room temperature and atmospheric pressure were recorded by using the intelligent manometer. Secondly, the calibration container was filled with H_2, when it was evacuated to a suitable pressure. After the pressure of calibration container attained a certain value, valve 14 was closed. Meanwhile, the value was recorded. Thirdly, valve 11 was opened. The background pressure of the tank was recorded before valve 11 was opened. Fourthly, when the pressure of the tank reached a certain value, valve 11 was

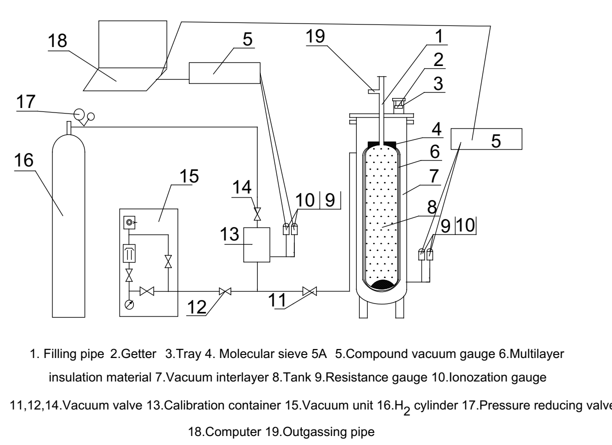

Fig. 1. Experimental apparatus of H_2 adsorption performance of composite adsorbent.