A Methodology of the Cryogenic Adsorbent Selection for He-3 Separation in Low Tritium Level by Modified Langmuir Equation

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1. Introduction

As regards the research of fusion fuel cycle, the cryogenic adsorption experiment has ever been carried out for the ceramic lithium breeding blanket processing [1]. Through the cryogenic adsorption the tritium can be recovered from helium stream of the breeding blanket. In this study, the cryogenic adsorption is performed for its applicability to the helium-3 collection loop in the ITER SDS (Storage and Delivery System) [2]. In the ITER SDS the residual tritium shall be also separated from helium-3 produced by tritium decay. MS5A, Zeolite, charcoal and Basolite Z1200 are used as adsorbents for the cryogenic adsorption experiments. Langmuir parameters of each adsorbent are obtained from the experiments. A modified Langmuir equation is newly proposed as a selection method of the best adsorbent among them in the range of very low tritium.

2. Experiments and Results

2.1 Experimental Apparatus

A schematic diagram of cryogenic adsorption experiments used in this study is shown in figure 1. The apparatus consists of the gas supply part, the cryogenic adsorption bed, the pressure control and effluent part, and a GC. The cryogenic adsorption bed is submerged in the liquid nitrogen bath.

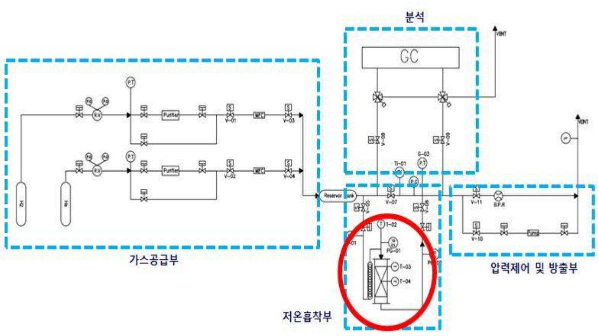

Fig. 1. Schematic diagram of cryogenic adsorption experiments

2.2 Adsorbents and Preconditioning

MS5A, Zeolite, charcoal and Basolite Z1200 are used as adsorbents for the cryogenic adsorption experiments. The characteristics of four adsorbents used in this study are shown in Table I. The Basolite Z1200 is one of nanoporous adsorbents developed recently for the hydrogen storage. Other adsorbents are commonly used in the adsorption processes.

Table I: Characteristics of adsorbents

Adsorbent	Description
Molecular sieve 5A	-Form: Pellets -Particle Size: 1.6 mm
Charcoal	-Formula Weight: 12.01g/mol -Vapor Pressure: <0.1mmHg(20℃) -Granular -Particle Size: 12×20 Mesh
Zeolite	-SiO_2/Al_2O_3 Mole Ratio: 30 -Nominal Cation Form: Ammonium -Na_2O Weight %: 0.05 -Surface Area: 400 ㎡/g -Particle Size: <45 ㎛
Basolite Z1200 (2-Methylimidazole Zinc Salt)	-Particle Size: 4.9 ㎛(D50) -Surface Area: 1300-1800 ㎡/g -Bulk Density: 0.35 g/㎤ -Metal Organic Framework -Zinc Imidazolate Framework -Activated at 100℃(Vacuum)

To improve the reliability and reproducibility of the experiments, the preconditioning for the adsorbents is carried out. During the preconditioning, nitrogen of 2 L/min is supplied to the reactor and the temperature of 200 ^{o}C outside the reactor is set up. As removing impurities the amount of them is measured by GC simultaneously for about 2 hours. After the preconditioning the reactor is cooled down and evacuated. Most impurities in all the adsorbents are removed through this preconditioning procedure before performing the adsorption experiment.

2.3 Analysis

During the experiments, the concentrations of hydrogen and helium in the inlet and outlet streams are measured by GC as a function of time. In Table II, the operating conditions of GC are summarized. TCD as a detector and molecular sieve as column material are used. The length of the GC column is 10 m.

Table II: GC Operating Condition

Carrier Gas	N_2
Flow rate (ml/min)	30
Detector	TCD
Temperature (℃)	
Oven	50
Detector	180
Sampler	
Type	Six ports
Volume	0.5 ml
GC Column	
Material	Molecular Sieve 5A
Length	10 m

2.4 Adsorption Isotherm

The adsorption amount of hydrogen as a function of time is shown in figure 2. Langmuir isotherm is commonly employed to explain the adsorption behavior. Langmuir isotherm can be expressed as equation (1):

$$V/V_m = BP / (1 + BP) \qquad (1)$$

where V_m is an imaginary maximum adsorption amount when pressure in bulk increases infinitely, and B is a ratio of adsorption rate to desorption rate.

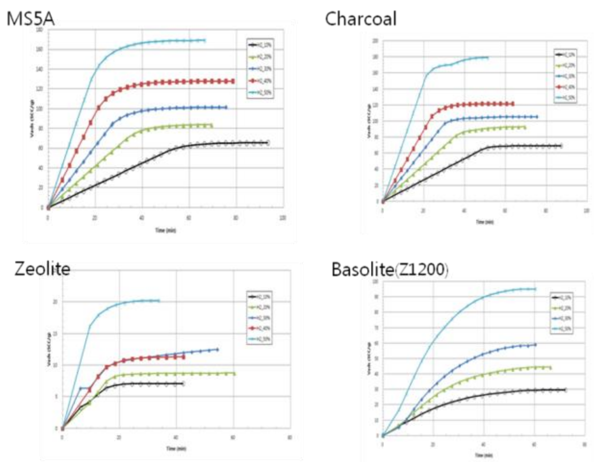

Fig. 2. Adsorption amount as a function of time for each adsorbent

Langmuir parameters of each adsorbent are obtained from the experimental data as in Table III. MS5A and Z1200 have better performance than Zeolite and charcoal.

Langmuir parameters, however, are improper to select the best adsorbent in the low pressure range. So a modified Langmuir equation is newly proposed to correct the shortcomings. The modified Langmuir equation has one parameter which is the product of two parameters of the original Langmuir equation. The modified Langmuir equation is derived in low tritium level and can be expressed as following equation (2):

$$V = V_m BP \qquad (2)$$

The parameter of the modified Langmuir equation is the gradient of adsorption amount versus pressure and indicates an adsorption performance near zero partial pressure. So the modified Langmuir equation has an analogy to Henry's law in the absorption process of gases. According to this analytical method, Zeolite is selected as the best adsorbent for the helium-3 collection loop in the range of low tritium in Table III.

Table III: Parameters of Langmuir Isotherm

	V_m [scc/g]	B [atm^{-1}]	BV_m [scc/g atm]
MS5A	**303.03**	1.91	578.8
Zeolite	67.57	308.33	**20,833.9**
Charcoal	294.12	18.38	5,405.9
Z1200	**2,000.00**	0.1230	246.0

3. Conclusions

The cryogenic adsorption is performed for its applicability to the helium-3 collection loop in the ITER SDS. MS5A, Zeolite, charcoal and Basolite Z1200 are used as adsorbents for the cryogenic adsorption experiments. Langmuir parameters of each adsorbent are obtained from the experiments. A modified Langmuir equation is newly proposed as a selection method of the best adsorbent among them in the range of very low tritium. According to this analytical method, Zeolite is selected as the best adsorbent for the helium-3 collection loop of the ITER SDS.

Acknowledgements

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