§4. Experimental Analysis of Hydrogen Adsorption for Isotope Separation/ Purification by Pressure Swing Adsorption

Kotoh, K., Kudo, K. (Kyushu Univ. Eng.) Nishida, T. (Kinki Univ. Eng. Kyushu) Sugiyama, T., Asakura, Y., Uda, T., Kawano, T., Yamada, H., Miyazawa, J.

In order to perform the LHD deuterium experiments, a practical system of hydrogen isotope separation is necessary for the environmental safety treatment of exhaust gases. We are scheming to apply the pressure swing adsorption (PSA) to the isotope separation. In the previous study, we have clarified the equilibrium characteristics of multi-component adsorption of hydrogen isotopes on a candidate adsorbent, synthetic zeolite 5A-type and have carried out fundamental experiments for examining the breakthrough characteristics of tracer D_2 in a H_2 - D_2 mixture with a zeolite 5A-type packed-bed column at a cryogenic temperature of 77.4 K.

In the present study, we investigate overall effective mass transfer coefficients which are practical and useful in an engineering design of a PSA process. A zeolite 13X-type is also examined as a candidate adsorbent.

i) Breakthrough experiments with adsorption column

Breakthrough behavior of D_2 in a hydrogen gas flowing through a synthetic zeolite packed-bed column is examined by using a cryogenic PSA apparatus in the NIFS. The test column is used of $\phi 40$ mm in inner diameter, where adsorbent particles of $\phi 2$ mm in mean diameter are charged at an amount of 700 g on a dry basis. The column is initially filled with pure hydrogen at 77.4 K after activated by increasing in temperature to 573 K with helium purge. Breakthrough curves are obtained by measuring the concentration of D_2 at the outlet of the column while the hydrogen mixture including D_2 at a concentration of 10,000 ppm is introduced from the inlet. The hydrogen isotope gas analyzing system developed in the previous study is used in this experimental study.

ii) Breakthrough curve analysis

Since this experimental system is of an isotope molecular exchange reaction which exhibits the Henry type adsorption, breakthrough curves of tracer D_2 carried on bulk H_2 are crossly simulated by theoretical curves calculated with the following equation.

$$\frac{c}{c_0} = e^{-x-r} I_0 \left(2\sqrt{XT} \right) + \int_0^T e^{-x-s} I_0 \left(2\sqrt{XS} \right) dS$$
$$X = \frac{K_F a_r Z}{u}, \quad T = \frac{K_F a_r t}{\beta \gamma}$$

c: concentration, t: time, u: superficial velocity,

 $K_F a_v$: overall effective mass transfer coefficient,

 β : adsorpsion coefficient, Z: height of packed bed,

 γ : packing dencity

Overall effective mass transfer coefficients $K_{r}a_{v}$ are obtained for the zeolite 5A-type and 13X-type from the comparison of experimental curves with analytical ones. The values of $K_{r}a_{v}$ are, then, plotted against superficial velocities in Fig. 1.



Fig. 1 Overall effective mass transfer coefficients

As shown in Fig. 1, the values of $K_F a_v$ for the zeolite 5A-type are slightly larger than those for 13X-type. This fact suggests that the rate determining process is inter crystalline diffusion rather than inner crystalline diffusion. The SEM pictures for the crystal structure of the zeolites may support the inference (Fig. 2).



Fig. 2 Crystal structure of the zeolites

It is also clear from Fig. 1 that the values of $K_r a_v$ are increasing monotonously with superficial velocity. This fact indicates that the axial dispersion is dominant in the whole mass transfer process for the present experimental condition.

In this study, valuable knowledge and data are obtained for considering the design and operation of a PSA process for hydrogen isotope separation.

References

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