§1. Experimental Study on Performance of Multi-column Pressure Swing Adsorption System for Hydrogen Isotope Separation

Kotoh, K. (Kyushu Univ., Eng.),
Takashima, S., Moriyama, S. (Kyushu Univ., Eng.),
Sugiyama, T. (Nagoya Univ., Eng.),
Tanaka, M., Asakura, Y., Uda, T.

Pressure Swing Adsorption (PSA) process[1-4] is usable for the successive hydrogen isotope separation necessary in an environmental tritium safety system such as for removal of tritium from LHD exhaust gases during D-D experiments or of volume reduction of tritiated-hydrogen waste gas storage by extraction/release of tritium-purified hydrogen volume. We have been clarifying the static and kinetic characteristics of adsorption of hydrogen isotopes onto synthetic zeolites packed in columns at 77.4 K, for the purpose of considering the design and operation of a tritium processing PSA system.

In this fiscal year, the breakthrough curves of trace HD or/and D_2 were examined with a column packed with SZ-5A or SZ-13X pellets in diameter of 2 mm. In the column, the replacing adsorption of tracer in a hydrogen mixture for bulk H_2 adsorbed occurs. The isotopic replacing adsorption of a tracer exhibits Henry's type behavior. The step response breakthrough curves of adsorption in Henry's type can be described as follows:

$$\frac{c}{c_0} = I_0\left(2\sqrt{XT}\right)\exp\{-(X+T)\}$$
$$+ \int_0^T I_0\left(2\sqrt{XT}\right)\exp\{-(X+T)\}dT, \qquad (1)$$

where I_0 is the first-kind modified Bessel function of zero order, and

$$X = \left(K_F a_v/u\right)Z, \qquad T = \left(K_F a_v/\beta\gamma\right)t. \qquad (2)$$

c/c_0 is the breakthrough tracer concentration normalized by its concentration fed to the packed column. t is time, and Z and u are the height of a packed-bed and the superficial total velocity of fluid. γ is the packing density of particles, and $\beta\gamma$ is the adsorption coefficient defined as $Q_0\gamma/C_0$, where Q_0 is the amount adsorbed in equilibrium with C_0: the concentration in gas phase. The adsorption coefficient of trace D_2 in H_2 is obtained in the following relation with the coefficient of bulk H_2:

$$\gamma\beta_{D2/H2} = \gamma\beta_{H2}SF_{D2/H2}, \qquad (3)$$

where $SF_{D2/H2}$ is the separation factor of D_2 for H_2 adsorbed in equilibrium defined by

$$SF_{D2/H2} = \frac{x}{1-x}\bigg/\frac{y}{1-y}, \qquad (4)$$

where x and y are the fractions of D_2 in the adsorbed phase and the gas phase, respectively. K_F and a_v are the overall mass transfer coefficient of trace component in an adsorbent packed-bed and the specific surface aria of particles in the packed-bed.

$K_F a_v$ is the volumetric transfer coefficient. Its reciprocal denotes the comprehensive resistance of mass transfer from fluid into porous adsorbent pellets as follows:

$$\frac{1}{K_F a_v} = \frac{1}{k_s a_v \gamma\beta} + \frac{1}{k_F a_v} + \frac{d_p}{u \cdot Pe}\left(1+\frac{\varepsilon}{\gamma\beta}\right)^2. \qquad (5)$$

On the right side, the first term is of the resistance in the porous media, the second term is of the gas-solid film resistance, and the third term is of the apparent resistance due to the longitudinal dispersion, where d_p and ε are the representative diameter of packing particles and the packed-bed void, and Pe is Peclet's number defined by

$$Pe = d_p u/\varepsilon D_L. \qquad (6)$$

D_L is the longitudinal dispersion coefficient depending on the superficial velocity as a correlation by Edwards $et\ al.$:

$$D_L = 0.73 D_M + \frac{0.5(u d_p)^2}{u d_p + 9.7 D_M}, \qquad (7)$$

under the condition:

$$0.008 < Re = \rho u d_p/\mu < 50, \quad 0.377 < d_p < 6.0 \ [\text{mm}], \quad (8)$$

where D_M is the molecular diffusion coefficient, and Re is Reynold's number.

The experiments were conducted at superficial velocities between 0.026 and 0.142 m/s. From the curve-fitting analysis with Eq. (1) for the experimental curves, optimum values were obtained of $K_F a_v$ affected by the superficial velocities. The analysis according to Eq. (5) shows the values of D_L resulting from the mixing behavior of H_2 as a bulk fluid flowing through packed-bed at the cryogenic temperature. The dependence of D_L on u, however, can be interpreted never by Eq. (7) but by the following relation:

$$D_L = 6.0\times 10^{-6} \cdot Re \quad [\text{m}^2\text{s}^{-1}]., \qquad (9)$$

This also can accurately characterize the dependency of D_L obtained from similar experiments with SZ-4A.

The result obtained here makes it possible to estimate the breakthrough curves with the packed columns operated at various flow rates of a hydrogen gas including tracer isotope.

1) Kotoh, K., $et\ al.$, $Fusion\ Eng.\ Des.$, **86**, 2799-2804 (2011).
2) Kotoh, K., $et\ al.$, $Fusion\ Sci.\ Technol.$, **60**, 771-775 (2011).
3) Kotoh, K., $et\ al.$, $Fusion\ Sci.\ Technol.$, **60**, 1355-1358 (2011).
4) Takashima, S., $et\ al.$, $Fusion\ Sci.\ Technol.$, **60**, 1436-1439 (2011).