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

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The hydrogen isotope separation process is necessary for the environmental safety management of exhausted gas from LHD deuterium experiments. Several methods are available for the isotope separation. The method of pressure swing adsorption (PSA), operating rapid desorption in a short period, has become applied to medium-massive separation of air-components. In this study, aiming at applying the PSA to the isotope separation process, we examine the multi-component adsorption behavior of hydrogen isotopes on an adsorbent.

In this work, we investigate the adsorption isotherms for H_2 and D_2 on synthetic zeolite 5A-type at the liquid nitrogen temperature 77.4 K and the isotope separation factors in H_2-D_2 or H_2-HD-D_2 systems, using a volumetric adsorption apparatus.

Isotherms are necessary for theoretical prediction of the multi-component adsorption behavior. Figure 1 shows experimental isotherms for H_2 and D_2 on the zeolite in a wide range of equilibrium pressure from 10^{-2} to 10^5 Pa, which can be closely approximated by an error-functional expression with respect to adsorption potential, proposed here:

$$\frac{Q}{Q_s} = \frac{1}{\sigma\sqrt{2\pi}} \int_{-\infty}^{\varepsilon} \exp\left\{-\frac{(\varepsilon-\bar{\varepsilon})^2}{2\sigma^2}\right\} d\varepsilon , \quad (1)$$

$$\varepsilon - \bar{\varepsilon} = RT\ln(P/\bar{P}) \quad (2)$$

where Q is the amount adsorbed on a basis of evacuated adsorbent, and Q_s the amount adsorbed in saturation. The adsorption potential is expressed by Eq.(2), where P is the equilibrium pressure, and $\bar{P}$ the representative pressure corresponding to the mean potential $\bar{\varepsilon}$.

The isotope separation factor in an adsorption system is defined as

$$SF_{ads} = \frac{(x_{heavy}/x_{light})_{ads}}{(y_{heavy}/y_{light})_{gas}}, \quad (3)$$

where x is the mole fraction of a heavy or light component in the adsorbed phase, and y the mole fraction in the gas phase. Figure 2 shows hydrogen isotope separation factors with respect to the amount of each isotopic mixture adsorbed in a wide range roughly between 0.002 and 4 mol/kg, where experimental values are plotted. A solid line represents the values of SF_{D_2/H_2} estimated from the ideal adsorbed solution (IAS) theory using the isotherms for H_2 and D_2. This solid line curve simulating the plot lineup of SF_{D_2/H_2} suggests that the behavior of hydrogen isotopes on the zeolite can be interpreted by the IAS theory. Broken

lines empirically approximate the experimental $SF_{D_2/HD}$ and SF_{DH/H_2}. The experimental values of $SF_{D_2/HD}$, SF_{DH/H_2}, and SF_{D_2/H_2} in each of H_2-HD-D_2 mixtures are consistent with accuracy of a 0.4-2.3% error in the relationship of

$$SF_{D_2/H_2} = SF_{D_2/HD}SF_{DH/H_2} . \quad (4)$$

Figure 2 demonstrates that SF_{D_2/H_2} values from about 2 to more than 3, $SF_{D_2/HD}$ values from about 1.6 near to 2, and SF_{DH/H_2} values from about 1.3 to around 1.6 are expectable in the cryo-sorption system of hydrogen on the zeolite 5A-type at 77.4 K.

In prediction of the adsorption behavior of hydrogen isotopes including tritium, it is inferable from the zero-point-energy mass-effect that the separation factors SF_{T_2/H_2} and SF_{DT/H_2} are larger than SF_{D_2/H_2} while the factor SF_{HT/H_2} is near but less than SF_{D_2/H_2}.

The fundamental characteristics of adsorption, important in the consideration of the isotope separation process using the PSA method, are revealed in this work.
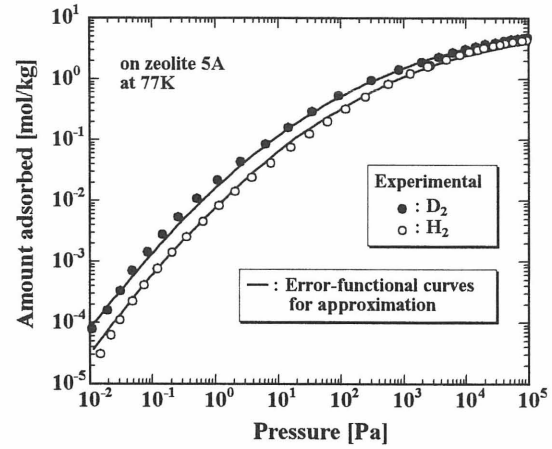

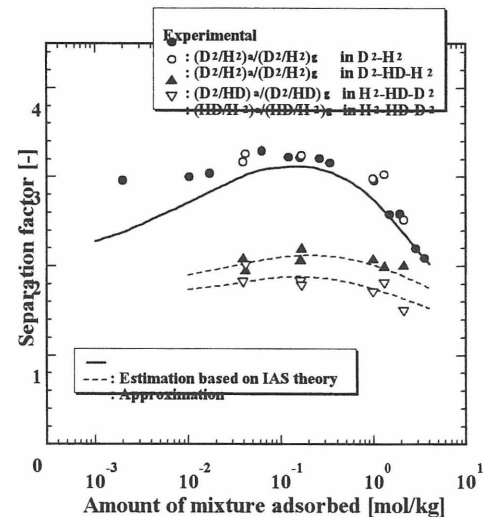Fig. 1   Isotherms for H_2 and D_2 on zeolite 5A-type at 77.4 K

Fig. 2   Separation factors of hydrogen isotopes on zeolite 5A-type at 77.4 K