§4. Development of High-Power Adsorbents for Hydrogen Isotope Separation by Pressure Swing Adsorption Method

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A process of hydrogen isotope separation is necessary in the environmental safety treatment of exhaust gases from LHD deuterium experiments. We have attempted to develop a pressure swing adsorption (PSA) process, using a general adsorbent such as synthetic zeolite 5A, for hydrogen isotope separation. In the last fiscal year, we began to research and develop adsorbents suitable to the PSA process for hydrogen isotope separation, aiming at improving its performance. In this work program, a volumetric adsorption apparatus was made to investigate the characteristics of adsorptive function of candidate materials. On the other hand, we proposed theoretical model equations for describing multi-component adsorption behavior of hydrogen isotopes on zeolites, based on chemical thermo-dynamics. The theoretical expression was verified with experimental results.

In this fiscal year a work was to calibrate the volumetric adsorption test device for determining correct values from observed pressures affected by a system effect. In this test device, thermal transpiration occurs. Pressures measured by the gage attached to one gas container at high temperature are not equivalent to those in the other gas container at low temperature. The effect of thermal transpiration depends on the ratio of the mean-free-path of gas molecules λ resulting in gas density to the inner diameter of a pipe connecting two containers d_p. This device has a connection pipe with an inner diameter of ϕ 15.6 mm. From the experiment with He, H_2 and D_2, we derived an equation to calibrate the system effect of thermal transpiration, as follows:

$$\Phi = \frac{1-\left(P_{cold}/P_{hot}\right)}{1-\sqrt{T_{cold}/T_{hot}}} \qquad (1)$$

$$\Phi = c\left[1-\int_{-\infty}^{\mu_X}\exp\left\{-\frac{(\mu-\overline{\mu})^2}{2\sigma^2}\right\}d\mu\right] \qquad (2)$$

where $\mu = \log\left(P_{hot}d_p\right)$.

The thermal transpiration factor Φ can be shown by an error-functional expression. In Eq.(2), c is a coefficient indicating the non-ideality of molecular reflection on a pipe wall surface, and $\overline{\mu}$ and σ are respectively the mean and the deviation of the Gaussian distribution.

Experimental results and characteristic curves are shown in Fig. 1. Coefficient values and deviation values among He, H_2 and D_2 are respectively agreeable, those are c = 0.9005, σ =0.5583. Values of the mean are also agreeable between hydrogen isotopes H_2 and D_2: $\overline{\mu}_{H2}, \overline{\mu}_{D2}$ = -1.533. The mean for He $\overline{\mu}_{He}$ is -1.336. The difference $\Delta\overline{\mu}_{He/H2} = \overline{\mu}_{He} - \overline{\mu}_{H2}$ can be related to the logarithm of the molecular mean-free-path ratio, $\log\left(\lambda_{He}/\lambda_{H2}\right)$. A value of 1.574 was obtained for $\lambda_{He}/\lambda_{H2}$ and $\lambda_{He}/\lambda_{D2}$. This value may be adoptable for the other hydrogen isotopes. Hence, the system effect on tritium became predictable. The characteristic curves of Φ shown here are also available for another system using a different size pipe.

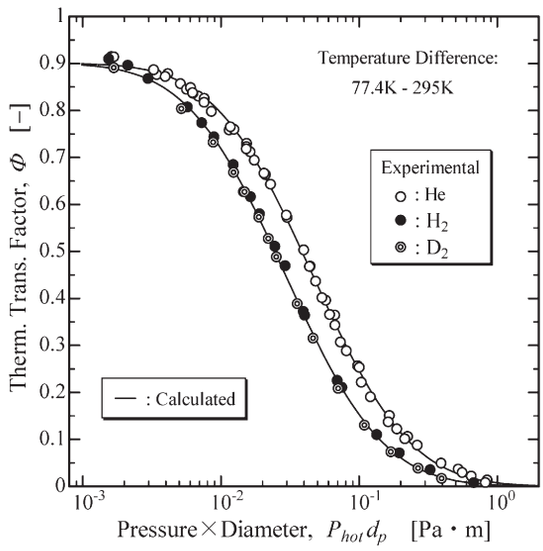

Fig. 1 Thermal Transpiration Factor with respect to Hot Pressure times Pipe inner Diameter.

The other work in this fiscal year was to develop materials having the adsorptive functions advantageous to the PSA operation, such as to exhibit a stepwise profiling isotherm for hydrogen. We have taken note of the crystal lattice structure of cobalt tris-ethylenediamine $[Co(en)_3]Cl_3$ having homogeneous pore channels of ϕ 0.6 nm, because this type pores may show stepwise-shaped adsorption isotherms. In this work program, a fine crystal sample of $[Co(en)_3]Cl_3$ was prepared, and then, the amount of hydrogen isotopes able to be adsorbed on this sample was measured by the method using the volumetric adsorption apparatus. A result so far is that the amounts adsorbed at 77.4 K around a pressure of 17 kPa are indicated as 1/3 times of those on synthetic zeolites. Treatment of the candidate sample is not easy because this complex compound is decayed by heating at a temperature higher than around 400 K. The investigation of functional materials is to be continued.

References

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