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

Kotoh, K., Kudo, K. (Kyushu Univ. Eng.)
Nishida, T. (Kinki Univ. Ind. Eng.)
Sugiyama, T., Asakura, Y., Uda, T., Kawano, T.

A process of hydrogen isotope separation is necessary in the environmental safety treatment of exhaust gases from LHD deuterium experiments. We attempt to develop a practical system of pressure swing adsorption (PSA) process for hydrogen isotope separation, applicable to the processing of the LHD exhaust gases. From the result of break-through experiments using adsorption columns with synthetic zeolite adsorbents at 77.4 K, we already confirmed that the PSA process is available for hydrogen isotope separation[2]. At the aim of developing the practical system, however, the PSA process must be improved to operate the separation at a more increased efficiency. The efficiency of separation depends on the characteristics of adsorption equilibrium and mass transfer of hydrogen isotopes on an adsorbent in use. Therefore, we reached the conclusion that the next step of this work should be to investigate or/and develop adsorbents more suitable for the PSA process.

An adsorbent useful to the PSA process should satisfy the following conditions: (1) A stepwise profiling isotherm for hydrogen is exhibited, and its sharp turn appears in a range of practical pressure swing operation. (2) Large numbers of separation factors are obtained in adsorption of a hydrogen isotope mixture. (3) The mass transfer onto/from adsorbent is sufficiently speedy. (4) Its body is tough, mechanically and chemically, and so on.

i) Investigation of Functional Adsorbents

We made a survey of materials having the adsorptive functions advantageous to the PSA operation. The result shows the following complexes promising as a functional adsorbent, which have angstrom-order-diameter channels in their crystal lattice frameworks.

- Cobalt Tris-ethylenediamine
  has one-dimensional regular channels of ϕ 0.6 nm in effective diameter, and shows stepwise isotherms for adsorption of water vapor.
- Terephthalic Acid
  has two-dimensional regular channels of ϕ 0.6 nm in effective diameter.
- Copper Terephthalate Tris-ethylenediamine
  has three-dimensional regular channels of ϕ 0.74 nm in effective diameter, and exhibits stepwise isotherms for adsorption of argon, nitrogen and carbon dioxide.

On cobalt tris-ethylenediamine, adsorption isotherms for H_2O and D_2O were already examined experimentally and analytically in our work. We have developed techniques of finer crystal-powder preparation and its pellet-fabrication. The next work is to develop the techniques for one of the best candidates: copper terephthalate tris-ethylenediamine.

ii) Development of Volumetric Adsorption Apparatus

As shown in Figure 1 an improved volumetric adsorption apparatus was developed for the purpose of observing the isotherms for hydrogen isotopes and these multi-component adsorption behaviors in detail with accuracy. This apparatus can perform the ultimate vacuum in a range of 10^{-7} Pa, and can measure the adsorption equilibrated at a pressure in the order of 10^{-3} Pa within an error of 10 %.

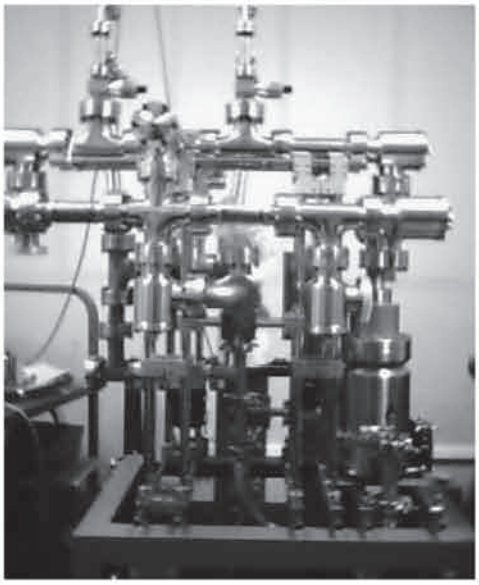

Fig. 1 Volumetric Adsorption Apparatus

iii) Development of Crystalline Adsorption Theory

The adsorption potential theory is most appropriate for describing the adsorption into micro-pores such as zeolite crystalline pores. In this theory, however, there are some problems should be resolved or improved. In this study, we developed a theoretical expression in order to describe the adsorption behavior of hydrogen isotope mixtures in zeolites. This expression assumes that the crystalline void space is identical with the adsorbed-phase volume corresponding to the adsorption potential field, where are molecules trapped on adsorptive inner-surface sites and the others not trapped but attracted over the other active sites[1, 3].

Experimental behaviors of H_2, HD and D_2 adsorbed on several types of zeolite at 77.4 and 87.3 K are interpreted consecutively by this expression. From relationships of its parameters between hydrogen and deuterium, the behaviors of all hydrogen isotopes can be predicted in simple but coherent description[3]. The dependence of parameters on temperatures also is becoming clear in theoretical analysis.

References

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