§3. Hydrogen Isotope Separation on Pore-size-controlled Mesoporous Materials

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The recovery of tritium from tritiated water in large volume with low concentration is an important task to reducing tritium emissions into the environment. Selective removal of tritium by using certain adsorbents is an attractive method because of the low cost and the ease of scaling up. Various kinds of adsorbents such as active carbon, amorphous silica, and zeolite have long been investigated, however, the tritium adsorption capacities of these adsorbents are rather low for practical application. Therefore, development of novel adsorbents for tritium removal is still an interesting subject. Mesoporous silicas, e.g., MCM-41 or SBA-15, exhibit several novel properties, as compared to conventional microporous zeolites, and show substantially higher tritium adsorption ability and separation factor.[1)] In this study, we investigated the tritium adsorption on several mesoporous silicas with different pore diameters.

Mesoporous silicas were prepared as follows. For MCM(DTAB), a mixed aqueous solution of dodecyltrimethylammonium bromide, colloidal silica, and NaOH was treated at 130ºC for 24 h for hydrothermal synthesis. For MCM(CTAB), on the other hand, a mixed aqueous solution of cetyltrimethylammonium bromide, tetraethylorhotsilicate (TEOS) and NH_4OH was stirred for 18 h at ambient temperature. Resultant white precipitates were filtered, washed with water, dried at 80ºC, and calcined at 550ºC for 5 h to obtain the final products. SBA-15 was prepared from TEOS, Pluronic-123, and HCl. The mixed aqueous solution was stirred at 40ºC for 2h, and then aged at 60, 80 or 110ºC for 24h. The white precipitates were filtered, washed with water, and dried at 80ºC. Finally, the products were obtained by calcination at 550ºC for 10h. The obtained SBA was denoted as SBA(x), where x represents the aging temperature.

Tritium adsorption experiments were carried out using the adsorbent (ca. 0.5 g) and 10 mL of HTO (ca. 500 Bq/mL). The mixed solution was continuously shaken in a water bath at 5ºC. After 48 h, the solution was filtered and the tritium concentration in the filtrate was measured by a liquid scintillation counter to evaluate the tritium activity. The tritium adsorption ability (W_i; in Bq/g) and separation factor (α) were evaluated from the following equations.

$$W_i = \frac{V_0 \times (C_0 - C_e)}{W_{ads.}} \qquad (1)$$

$$\alpha = \frac{C_0}{C_e} \cdot \frac{V_0}{V_{ads.}} \cdot - \frac{V_e}{V_{ads.}} \qquad (2)$$

Here, V_0 and $W_{ads.}$ represent the volume of tritiated water used and the amount of adsorbent used, respectively. C_0 and C_e represent the initial and equilibrium concentration of tritium in liquid phase, respectively. $V_{ads.}$ and V_e represent the volume of tritiated water adsorbed on adsorbent and that in liquid phase at the equilibrium, respectively.

Fig. 1 shows the pore size distribution curves of the mesoporous silicas prepared. It can be clearly seen that the pore diameter changed depending on the alkyl chain length (MCM) or the synthesis temperature (SBA). The pore diameter and surface area determined are summarized in Table I.

The results of tritium adsorption experiments are summarized in Table I. For W_i, it was found that the maximum value was obtained around the surface area of ca. 950 m^2/g. This suggests the contribution of surface reaction on mesopore surfaces in tritium adsorption. On the other hand, the α value appears to decrease with the increase in pore diameter, indicating the pore size dependency. From these data, it can be concluded that the fine tuning of mesoporosity, including both pore diameter and surface area, of adsorbents is required for the design of novel adsorbents.

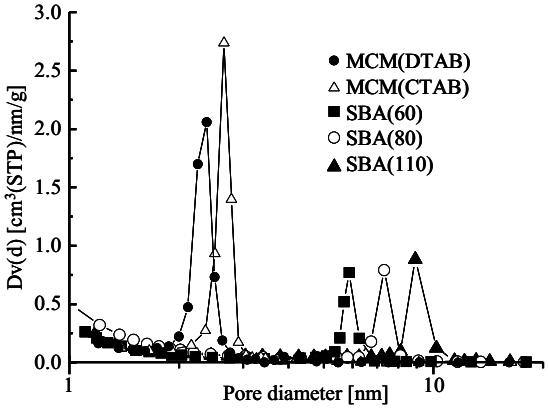

Fig. 1. Barrett-Joyner-Halenda (BJH) pore size distribution curve of mesoporous silicas prepared.

1) Taguchi, A. et al.: Fus. Sci. Technol. **60** (2011) 1395.

Table I Summary of the physicochemical properties and the results of tritium adsorption experiments

	Pore diameter [nm]	Surface area [m^2/g]	Adsorption ability (W_i [Bq/g])	Separation factor (α)
MCM(DTAB)	2.38	1104	68.2 ± 5.7	1.22 ± 0.08
MCM(CTAB)	2.65	1133	52.8 ± 4.9	1.22 ± 0.01
SBA(60)	5.88	624	58.9 ± 6.0	1.16 ± 0.06
SBA(80)	6.55	832	85.2 ±12.3	1.14 ± 0.06
SBA(110)	8.96	951	87.3 ± 8.5	1.18 ± 0.08