

§1. Design Problems of Cryogenic Pressure Swing Adsorption System for Hydrogen Isotope Separation in Fusion Fuel Cycle

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Pressure Swing Adsorption (PSA) process is applicable to an isotopic decontamination system for DT fuel gases contaminated with H atoms in the DT fusion fuel cycle. Such a system has to treat an isotope mixture of T₂, DT, D₂, HT, DH and H₂, where the former three are major with the other trace. Therefore, for considering this application, we should know the dynamic behaviors of hydrogen isotopes not only in the trace adsorption system but also in the non-trace adsorption system.

During flowing through a packed column, the partial flow rate of an adsorbate component varies with its adsorption into adsorbent. The total flow-rate variation effects a change of the breakthrough curve which reflects the moving distribution of concentration in the packed column. This change hence appears with increase in the concentration. Thus, in consideration of the design and operation of a non-trace adsorption system, it is required to elucidate the breakthrough-curve concentration dependency.

The breakthrough curves were examined of non-trace D₂ or H₂ in the H₂-D₂ adsorption process with a synthetic zeolite SZ-13X packed column at 77.4 K, using the H₂-D₂ mixtures containing D₂ at 2, 4, 6, 8, 10 and 12 mol% or H₂ at the same concentrations. Difference appeared in the curves between two types of isotopic gas mixtures, where the effect of concentration is slightly but surely observed on each group of breakthrough curves.

A mathematical model was considered here of the mass balance of a non-trace H₂-D₂ mixture in adsorbent packed-bed with assumption of fixed total-pressure ignoring the pressure drop causing the mass flow. In an infinitesimal section, the total mass-flow balance is described as follows:

$$C \frac{\partial U}{\partial z} + \gamma \frac{\partial \bar{Q}}{\partial t} = 0, \quad (1)$$

where z and t are distance and time, and C the total density (concentration) of the two-component mixture, which is constant since ignoring the total-pressure drop. U is the superficial total-velocity of fluid in this packed column. γ is the packing density of adsorbent, and \bar{Q} denotes the total amount adsorbed into adsorbent. That is,

$$C = c_{H_2} + c_{D_2} \quad : \text{constant}, \quad (2)$$

$$\bar{Q} = \bar{q}_{H_2} + \bar{q}_{D_2}, \quad (3)$$

$$U = u_{H_2} + u_{D_2}, \quad (4)$$

where c and \bar{q} are the partial concentration of H₂ or D₂ in the fluid mixture and the partial amount of H₂ or D₂ adsorbed, and u the superficial partial-velocity of H₂ or D₂ in the column. Each component mass-balance in the packed-bed can be assumed as follows:

$$\frac{\partial(c_{D_2}U)}{\partial z} + \varepsilon_B \frac{\partial c_{D_2}}{\partial t} + K_{FD_2} a_v (c_{D_2} - c_{D_2}^*) = 0, \quad (5)$$

$$\frac{\partial(c_{H_2}U)}{\partial z} + \varepsilon_B \frac{\partial c_{H_2}}{\partial t} + K_{FH_2} a_v (c_{H_2} - c_{H_2}^*) = 0. \quad (6)$$

The term $c - c^*$ in brackets means the driving force, where c^* indicates the corresponding concentration in equilibrium with the partial amount adsorbed. K_F and a_v are the overall mass-transfer coefficient of H₂ or D₂ and the specific surface area of particles packed, respectively. The product $K_F a_v$ is the volumetric transfer coefficient. Same values as $K_{FD_2} a_v$ are given to $K_{FH_2} a_v$ in Eqs. (5) and (6).

As shown in Fig. 1, the numerical curves are fitted with experimental non-trace breakthrough plot-lineups, perfectly, where $K_F a_v$ employs 3.0 s⁻¹ for the curves from adsorption of H₂ replacing D₂ while selecting 4.5 s⁻¹ for the curves of D₂ replacing H₂. This difference suggests that the mass-transfer rate-determining process is the desorption of H₂ or D₂ adsorbed in advance, in addition to the isotopic effect of longitudinal dispersion in packed-bed on the corresponding mass-transfer resistance.¹⁾

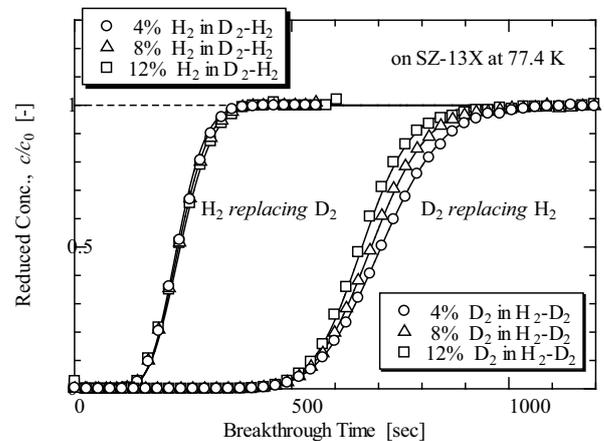


Fig. 1 Breakthrough curves of non-trace H₂-D₂ mixtures replacing H₂ or D₂ adsorbed in advance with SZ-13X packed column.

1) Kotoh, K., Moriyama, S., Takashima, S., Takahashi, K., "Breakthrough curves of non-trace H₂-D₂ mixture replacement adsorption with SZ-13X packed column at 77.4 K", Proceedings of SOFT 2012, Liege, Belgium, Sept. 24-28, 2012; in press, *Fusion Eng. Des.* (2013).