§4. Experimental Verification of Hydrogen Isotope Separation and Enrichment 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 for hydrogen isotope separation, using synthetic zeolite adsorbents. From the results of theoretical studies and basic experiments made till now, we estimated that this system could show effectual performance of hydrogen isotope separation in practical use. In this fiscal year, hence, experimental works were planned for verification of effective hydrogen isotope separation and enrichment by the PSA method. Before works, the following improvement of the PSA apparatus of NIFS was made:

- Downsizing of adsorption column's inner diameter for improvement of heat transfer and superficial velocity.
- Use of vacuum thermal-insulation for improvement of a liquid nitrogen cryostat.
- Improvement of vacuum exhaust lines to increase the conductance of a recovery gas stream at low pressure.
- Improvement of sample-gas recovery and gas analyzing system: shortening a sampling line to reduce a response time and upgrading of quantitative measurement using diaphragm-type pressure gages.

Experimental Operations with the PSA Apparatus

Material and geometric conditions:

Adsorption agent: Synthetic Zeolite NaA (5A), pellets of 2 mm spherical, manufactured by MERCK Co. Ltd.

Adsorption column: 21 mm inner diameter Packed-bed of SZ-5A pellets charged at 168 g.

Adsorption Operation

Mass balance of adsorbate hydrogen in the packed bed.

The amount of hydrogen adsorbed in the packed-bed at 77.4 K agrees with that estimated from the isotherm for H_2 on SZ-5A, within 1 % error, which has been measured by a volumetric adsorption equilibrium apparatus.

Displacement Adsorption Operation

Examination of breakthrough behavior of tracer D_2 in a H_2-D_2 mixture flowing through the packed-bed column at 77.4 K.

Operational condition: gas flow rates of 2.0, 3.0, 4.0, 5.0, 6.0, 7.0, 8.0, 10.0 NL/min.

From breakthrough experiments operated at a variety of gas flow rates, we obtained the results that the main process determining the overall mass transfer coefficient was the process of molecular diffusion in the macro-porous media of pellets.

The crystalline transfer in zeolite grains is fast enough to be compared with that in the macro-porous media, because of a-few-micron sizing crystal grains having adequately short diffusion-passes. The mass transfer resistance in the gas-solid boundary film is negligible but the effect of longitudinal dispersion on the overall mass transfer coefficient is significant.

The quantitative ratios of D_2 to H_2 in H_2-D_2 mixtures adsorbed in the packed-bed, obtained from integration of breakthrough curves, are evaluated at 1.8 times of the ratio in the feed gas mixture. The ratio of 1.8 agrees with the separation factor predicted from a theoretical model of multi-component adsorption which has been verified with results of experiment using a volumetric adsorption equilibrium apparatus.

Evacuating Desorption Operation

Operational conditions: evacuating rate of 600 NL/min, ultimate pressure of 100 Pa for an operation period of 20 min.

Repeated operations of adsorption-desorption exhibited reproducible conditions of evacuation. The gas mixture samples recovered by evacuation included the component of D_2 enriched at a ratio of 1.2. This demonstrates that the PSA method is available for hydrogen isotope separation and enrichment.

This ratio 1.2, however, is too low in comparison with the other ratio 1.8 obtained from adsorption breakthrough curve analysis. From this disagreement, we considered that the component of D_2 would be concentrated in the residual volume adsorbed which could not be recovered by evacuation.

Heating and Evacuating Desorption Operation

Operational conditions: evacuating rate of 600 NL/min, heating from 77.4 K up to 273 K, ultimate pressure of 0.1 Pa.

By the heating and evacuating operation following the evacuating desorption process, an amount of gas mixture recovered was about 10 % of the total amount adsorbed. This indicates that about 90 % of the total amount can be recovered by the evacuating operation.

From mass spectrometric analysis, it was proved that the D_2 component enriched at 6.67 times was contained in the residual gas volume recovered by the heating and evacuating operation. With this enrichment ratio 6.67 in the residual volume, the overall mass balance holds in this pressure swing adsorption system.

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