



The apparent counter diffusion coefficient of water absorbing on zeolite particles in the case of the water moving through macro-size pores in a column packed with zeolite particles



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HIGHLIGHTS

- Water reduces the adsorption performance of nitrogen in Li-X type zeolite particles (molecular sieve).
- Concentration distributions of water absorbing on zeolite particles were measured by MRI system.
- One-dimensional diffusion equation of water in the column under transient conditions was analyzed.
- The apparent diffusion coefficient of water absorbing on the zeolite particles was determined.

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ABSTRACT

The oxygen concentration discharged from a compact adsorption-type oxygen concentrator decreases when water attaches to the zeolite particles used as a nitrogen adsorbent. In order to estimate the expansion of the water adsorption region in the zeolite column, it is necessary to measure the water diffusion occurring in a zeolite column after stopping PSA (Pressure Swing Adsorption) operations. Li-X type zeolite particles (molecular sieve OXYSIV-700) with particle diameters in the range of approximately 0.4–0.5 mm were used as a nitrogen adsorbent column of length 157 mm and of inner diameter 33 mm. By comparing the water concentration distributions obtained from MR (Magnetic Resonance) images and the distributions calculated by analytical solution of a one-dimensional diffusion equation under transient conditions, the counter diffusion coefficient of water absorbing on zeolite particles in the column was determined. The apparent diffusion coefficient of water absorbing on the zeolite particles moving into the zeolite column with macroscale pores was determined to be $1.25 \pm 0.15 \times 10^{-11} \text{ m}^2/\text{s}$.

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1. Introduction

Compact adsorption-type oxygen concentrators are widely used as a medical device for supplying oxygen in Japanese homes and as an oxygen supply device for fish farming. A compact adsorption-type oxygen concentrator has two columns packed with zeolite particles, and the length of the column is 157 mm. By supplying air to the column, the pressure in the column rises, and more nitrogen is adsorbed on the zeolite particles compared with oxygen. A high oxygen concentration gas from the outlet of the column is then discharged. When the pressure in the column is decreased by opening a stop valve, the nitrogen is discharged by desorbing from the zeolite particles. A compact adsorption-type oxygen concentrator can continuously discharge a high oxygen concentration

gas by repeatedly raising and lowering the pressure of the zeolite column [1–3]. This process is called PSA (Pressure Swing Adsorption) operation.

A compact adsorption-type oxygen concentrator does not have a dryer to keep the cost down. Water is also absorbed in the air supply side of the zeolite columns when the supply air contains water vapor during PSA operations. In contrast, water desorbs from the zeolite during the decompression stage of PSA operation. Since adsorption and desorption of water on the zeolite particles are repeated during the PSA operation, the expansion of the water adsorption region in the zeolite column is limited to about 15 mm. However, when the PSA operation is suspended, the size of the water adsorption region expands by water diffusion. Since the expansion of the water adsorption region reduces the nitrogen adsorption performance of the zeolite particles, the discharged oxygen concentration is thereby reduced. In order to predict the

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reduction in the discharged oxygen concentration, it is necessary to measure the diffusion coefficient of water in the zeolite column.

There are reports of self-diffusion of the water adhering to a zeolite crystal having nanoscale pores [4,5]. The values of the self-diffusion of water described in two papers are about $2 \times 10^{-9} \text{ m}^2/\text{s}$ between 40 and 45 °C. Since this value is almost equal to that of the bulk water, it is presumed to represent the self-diffusion coefficient of water in the nanoscale pores of the zeolite. When zeolite particles having a diameter of about 0.4–0.5 mm are used to fill a column having an inner diameter of 33 mm, a large number of macro-scale pores having a size of about 0.15 mm are formed. The above value of the self-diffusion coefficient of water in nanoscale pores is not appropriate for the counter diffusion coefficient of water in such a column. The value needed for our study is the counter diffusion coefficient of water moving in the zeolite column containing macro-scale pores. In addition, there is also reported that the intra-particle diffusion coefficient of water in zeolite particles in the case of liquid-phase adsorption for the dehydration of ethanol is $5 \sim 10 \times 10^{-12} \text{ m}^2/\text{s}$, and its value depend on the average pore diameter of zeolite [6]. However, this value is not the diffusion coefficient of water which occurs in a column filled with zeolite particles and the air as gas phase. Techniques for measuring porous media with short T_2 relaxation time by MRI (Magnetic Resonance Imaging) have been developed, and there are papers describing the results of measuring the spatial concentration distribution of the water or oil in porous media using these MRI measurement techniques [7–12]. However, the counter diffusion coefficient of water moving in a zeolite column containing macro-scale pores has not been calculated in these papers.

In this study, in order to obtain the diffusion coefficient of water occurring in a zeolite column after stopping PSA operations, we measured the concentration distribution of water attached to the air supply side of a zeolite column by MRI. Spherical zeolite particles with a diameter of approximately 0.4–0.5 mm were filled in the zeolite column which is of length 157 mm and of inner diameter 33 mm. We obtained MR images during the time in which the region containing adsorbed water expanded after stopping the PSA operation. The water concentration distributions obtained from MR images were compared with those obtained by conducting a numerical analysis of the one-dimensional diffusion equation. The apparent diffusion coefficient of water absorbing on the zeolite particles moving into the zeolite column with macroscale pores was determined for the first time by this comparison.

2. Experimental equipment and MRI apparatus

2.1. Li-X type zeolite particles

A photograph of the Li-X type zeolite particles (molecular sieve OXYSIV-700 manufactured by Union Showa K. K.) used is shown in Fig. 1. Their diameter was in the range of approximately 0.4–0.5 mm.

The average diameter of the micro-size pores on the zeolite particle surface was measured by mercury porosimetry (AutoPore IV 9500, Shimadzu Corporation). The obtained result is shown in Fig. 2. From the peak position in the figure, it was found out that the average diameter of micro-size pores into which mercury can enter was 0.41 μm . Gas enters into the inside of zeolite particles by passing along these micro-size pores.

On the other hand, the zeolite crystal has many nano-size pores which adsorb nitrogen. An average diameter of the nano-size pores in the zeolite crystal was measured by argon adsorption isotherms at the temperature of 77.241 K (3Flex, Micromeritics). The obtained result is shown in Fig. 3. From the peak position in

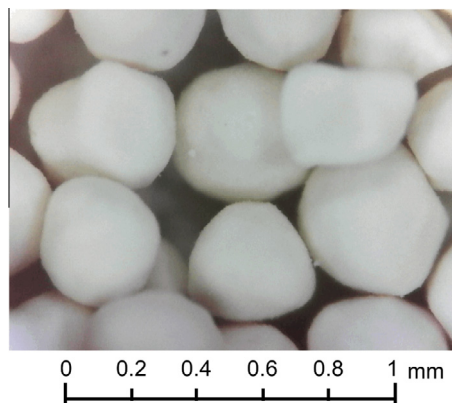


Fig. 1. Photograph of the Li-X zeolite particles used in this work.

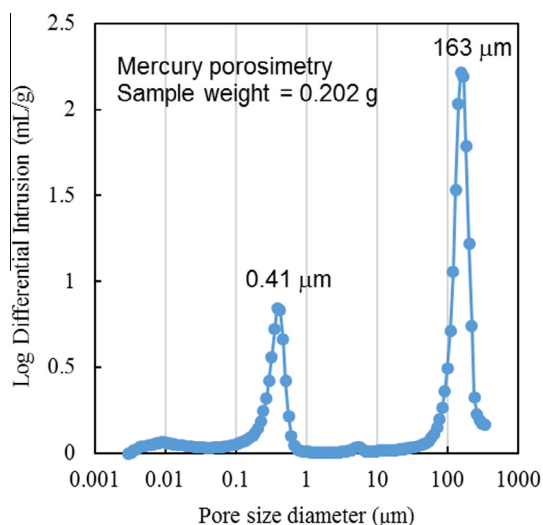


Fig. 2. The micro-size pore distribution on the zeolite particle surface measured by mercury porosimetry.

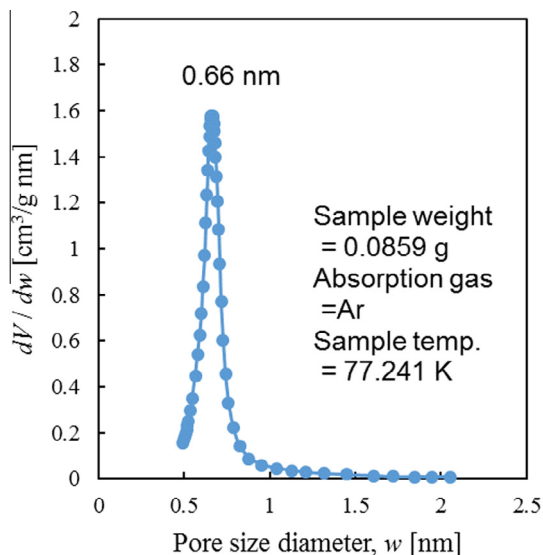


Fig. 3. The nano-size pore distribution in the zeolite crystals measured by Argon adsorption isotherms at the temperature of 77.241 K.

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