

## Validation of a model for non-equimolar counter diffusion of binary gas mixture in a micropores

Katsutoshi Hamano, Takami Kai, Tsutomu Nakazato

### Abstract

In 1833 Graham<sup>2)</sup> reported that the gas exchange rate is proportional to the square root of the reciprocal of molecular density from the results of the isobaric gas exchange of binary gas system. Hoogschagen<sup>3)</sup> found the similar results after 120 years. This phenomenon was reconfirmed by many researches, and the relation has called Graham's law. This phenomenon has not been sufficiently understood, probably because many researchers have proposed mechanisms using the equimolar diffusion coefficient. Kai *et al.*<sup>1)</sup> defined an intrinsic gas diffusion coefficient for each component and proposed a model coupled with bulk flow caused by pressure difference due to non-equimolar diffusion. In this study, binary gas diffusion was performed in isovolumetric system by varying the pore size and gas combination.

A stainless tube was used for the packed bed column and for the storing device of a bundle of capillaries. Nonporous glass beads and capillary tubes were filled in the column. Four kinds of glass beads were used, and the diameter was 16.7, 25.3, 40.7 and 58.7  $\mu\text{m}$ . The assembly composed of 400 capillary tubes were also used for diffusion tests. The inner diameter was 20  $\mu\text{m}$  and 30  $\mu\text{m}$ . Six types of gases were used.

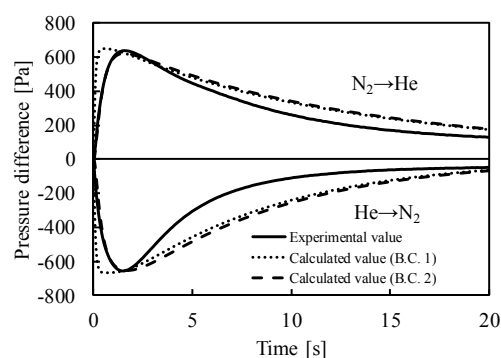
**Fig. 1** shows the comparison of experimental data with calculations by the model for the temporal change in the pressure difference. In this case, diffusion experiments were carried out in the packed bed, the particle diameter was 16.7  $\mu\text{m}$ . The gas combination system was helium/nitrogen. Both the experimental results and the calculation results showed a similar tendency that the pressure difference rapidly reached the maximum or minimum value and thereafter the pressure difference gradually recovered. However, there is a difference in the initial change rate of pressure and the pressure change during the recovery process. In the case of the calculated value (B.C. 1) in this figure, the boundary condition of the surface of packed bed is assumed that the mole fraction of Gas B is 100% after gas switching. However, in actuality Gas B does not instantly replace Gas A. Therefore, the boundary condition was changed as Gas B replace Gas A exponentially after gas switching. The calculation result in this case is shown by the dashed line (B.C. 2) in Fig. 1. In this case, the time until 50% of Gas A is replaced by Gas B was set to 250 ms. The calculation results is close to the experiment results by considering the gradual change of the boundary conditions. The restoring rate of pressure offset depends on the viscous flow due to pressure gradient. In this model, Kozeny-Carman equation is used. Even though any adjustable parameters are not used. The temporal change in pressure can be sufficiently estimated.

**Fig. 2** compares the experimental and calculated values for the maximum or minimum values of pressure when gas combination and capillary diameter were changed. For the viscous flow in the case of the capillary, Hagen-Poiseuille equation is used, and there are no experimentally determined constants like the Kozeny-Carman constant. As can be seen from this figure, a good agreement is shown by comparing the experimental and the calculated values. At least for the initial pressure change, it is considered that the model can take into consideration the influence of gas properties and pore diameters.

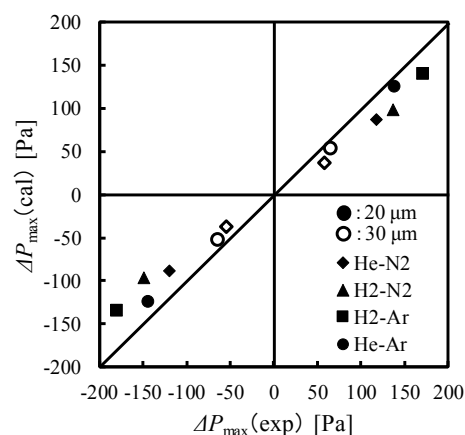
### Reference

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- 2) Graham, T., *Philos. Mag.*, **2**, 175–190 (1833)
- 3) Hoogschagen, J., *J. Chem. Phys.*, **21**, 2096–2097 (1953)

Department of Chemical Engineering, Kagoshima University, Kagoshima 890-0065, Japan



**Fig. 1** Comparison of experimental and calculated values the proposed model for He / N<sub>2</sub> system in packed bed.



**Fig. 2** Comparison between experimental maximum pressure differences with calculated values (Capillary 20 and 30  $\mu\text{m}$ )