Technology

Utilization of digital technology for PSA process development

AIBA Keisuke*   NAKAGAWA Shoichi**

1.   Introduction

Pressure Swing Adsorption (PSA) is a process for separating and purifying gas mixtures by periodically switching operational steps, such as adsorption and regeneration, through pressurization and depressurization, exploiting the pressure dependence of gas uptake on the adsorbent. In recent years, PSA has increasingly been applied to carbon-neutral technologies, including CO_2 separation and recovery as well as biogas processing. Our company has developed a multi-column PSA[1] for purifying hydrogen from ammonia decomposition gas for fuel cells and a CO_2 recovery PSA unit[2] with a capacity of 10 tons/day.

Historically, our PSA process development has relied primarily on experimental evaluation using test facilities, which was both labor-intensive and time-consuming. Although simulations were available, their scope was limited to specific operating conditions or localized sections of the adsorption column, making it difficult to capture the behavior of the entire PSA process or to reuse an individual model across different configurations and operating conditions. Whenever the process configuration or operating conditions changed, model revisions were required.

To support future PSA process development, where more diverse gas systems and operating conditions are expected, establishing a simulation-based environment prior to experimentation is essential for improving development efficiency. By constructing a model that can consistently handle the entire PSA process and a highly versatile adsorption column model as its core component and applying parameters identified through experimental data, it is expected that the efficiency of the process development can be improved and accumulated knowledge can be effectively reused.

This paper presents our simulation model targeting multi-column PSA and Vacuum-PSA (VPSA), outlining the overall configuration and the adsorption column as its key component. Application examples to a two-column PSA and a four-column VPSA for hydrogen purification from ammonia decomposition gas demonstrate that the model reproduces pressure and temperature behavior and impurity levels in the product gas, thereby illustrating its utility.

2.   Simulation Model for PSA Processes

2.1 Overview

In this model, targeting multi-column PSA and VPSA, it computes the transient evolution of pressure, temperature, and composition along the adsorption column, as well as impurity concentrations in the product gas, by inputting raw gas conditions, operation steps, and step switching times.

As an example, Fig.1 shows a two-column PSA process, comprising an adsorption column model and valve models. The adsorption column model solves the mass and energy balances within the column packed with adsorbent, calculating the time-dependent changes in axial profiles of pressure, temperature, composition, and adsorbed amount. The valve models define the boundary conditions for flow rate and pressure at the column inlet and outlet through their open/close states. In the case of VPSA, a regeneration step under vacuum using a vacuum pump is added; therefore, the inlet/outlet flowrates and pressures of the column during evacuation are calculated by reflecting pressure-flow characteristics of the vacuum pump. Through the operational sequences using each model, the simulation calculates the behavior of product gas extraction while impurities in the feed gas are adsorbed and removed within the column.

Table1 summarizes the typical operating steps in a two-column PSA process. Each step is defined by a specific configuration of valve open/closed states. In the simulation, the overall behavior of the process is determined by solving a system of simultaneous equations derived from the adsorption column and valve models for each step. This same framework can be

* R&D Administration Department, R&D Planning Division, R&D Unit

** Gas Separation Development Department, Yamanashi Technology Solution Center, R&D Unit

extended to simulate processes other than two-column PSA by accounting for variations in system configuration and operating conditions.
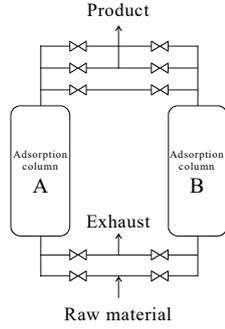

Fig. 1 Schematic flow of two-column PSA

Table 1 Operation steps of two-column PSA

	Step 1	Step 2	Step 3	Step 4
Column A	Adsorption	Pressure equalization	Regeneration	Pressure equalization
Column B	Regeneration	Pressure equalization	Adsorption	Pressure equalization

The following sections provide an overview of the adsorption column model, which is the core component of the PSA process, the adsorption rate model and equilibrium adsorbed amount model, which are the constituent elements of the adsorption column model, and the pressure drop.

2.2 Adsorption Column Model

The adsorption column is modeled as a porous packed bed assuming one-dimensional flow, neglecting radial distributions within the column. This model simultaneously solves a system of equations, including mass and energy balances for both gas and solid phases, as well as equations for adsorption rate, equilibrium adsorbed amount, and pressure drop. The energy balance accounts for the heat of adsorption and heat exchange with the exterior through the column wall. These governing equations are spatially discretized along the axial direction and formulated as a system of ordinary differential equations. For the calculation of time-dependent changes, an implicit time integration method is employed to ensure numerical stability even during rapid pressure changes at the time of step switching.

2.3 Adsorption Rate Model

The adsorption rate is represented by the Linear Driving Force (LDF) model, which assumes that the rate is proportional to the difference between the current adsorbed amount in the solid phase and the local equilibrium adsorbed amount. The solid-phase adsorbed amount q_i for component i is expressed by the following equation:

$$\frac{\partial q_i}{\partial t} = k_i \big(q_i^{eq} - q_i\big) \qquad (1)$$

Where q_i^{eq} is the local equilibrium adsorbed amount of component i, and k_i is the LDF rate coefficient. The temperature dependence is expressed by $k_i = k_{0,i}\exp(-E_{a,i}/RT)$, which includes a parameter $k_{0,i}$ and the activation energy $E_{a,i}$. $k_{0,i}$ and $E_{a,i}$ are identified based on experimental data.

2.4 Equilibrium Adsorbed Amount Model

For the equilibrium adsorbed amount of a multi-component system q_i^{eq}, the Dual Site Langmuir (DSL) equation (Eq. (2)) is employed, assuming that there are two types of adsorption sites on the solid surface with different properties, such as adsorption energy.

$$q_i^{eq} = \sum_\alpha \frac{m_\alpha K_{i,\alpha} p_i}{1 + \sum_j K_{j,\alpha} p_j} \qquad (2)$$

Here m_α is the saturation capacity and p_i is the partial pressure of component i. Also, $K_{i,\alpha}$ represents the adsorption equilibrium constant, which accounts for temperature dependence via $K_{i,\alpha} = K_{0,i,\alpha}\exp(Q_{i,\alpha}/RT)$, where the factor K_0 and the heat of adsorption $Q_{i,\alpha}$ are used. Each parameter is identified based on adsorption isotherm data. The value of q_i^{eq} obtained from Eq. (2) is applied to Eq. (1).

2.5 Pressure Drop

The pressure distribution within the adsorption column is calculated by accounting for the pressure drop in the packed bed. The pressure drop is determined using the Ergun equation, which incorporates both viscous and inertial resistance, as shown in Eq. (3). The pressure drop Δp within a spatial element is expressed by the following equation:

$$\Delta p = \frac{150 \mu u \Delta l (1-\varepsilon)^2}{d_p^2 \varepsilon^3} + \frac{1.75 \rho_g u^2 \Delta l (1-\varepsilon)}{d_p \varepsilon^3} \qquad (3)$$

Where Δl is the element length of the adsorption column, ρ_g is the gas density, ε is the bed porosity, d_p is the particle diameter of the adsorbent, u is the superficial velocity, and μ is the gas viscosity. For each element

discretized along the axial direction, the pressure drop Δp is calculated using Eq. (3), and the pressure profile over the entire column length is obtained by integrating these values from the inlet pressure. This pressure, along with the temperature, provides the total molar concentration C via the equation of state, while the partial pressures p_i are subsequently defined based on the gas composition.

3. Application Examples for PSA Processes

3.1 Application to Two-Column PSA

First, an application to the two-column hydrogen purification PSA shown in Fig. 1 is described. The impurity concentrations in the product gas at two different hydrogen recovery rates, as well as the pressure and temperature within the column, were calculated and compared with experimental results. In this process, the feed gas is an ammonia decomposition gas consisting of 75% hydrogen and 25% nitrogen. The operation cycle is configured such that feed gas is introduced into either Column A or Column B according to the step shown in Table1, while the other column simultaneously undergoes depressurization-regeneration or pressure equalization step. In the simulation, key operating conditions, such as column dimensions, feed gas flow rate, and column inlet pressure, were matched to the experimental conditions. The parameters for the equilibrium adsorbed amount and the adsorption rate model were identified using obtained experimental data related to the two-column PSA.

Fig. 2(a) and (b) show the pressure and temperature within the adsorption column during a cycle that has reached a cyclic steady state. The solid and dotted lines represent the experimental and simulation result for Column A and Column B, respectively. Both pressure and temperature values are normalized based on representative values. As shown in Fig. 2, the simulation and experimental results for the internal column pressure show good agreement throughout the process. In particular, although the column pressure changes rapidly at the start and end of the pressure equalization step and during the transition to the regeneration step due to valve operations, the simulation results stably reproduce the behaviors without numerical divergence. Regarding the column temperature, the model captures behaviors such as the temperature rise during adsorption and the temperature drop during desorption in the regeneration

step to the same experimental results. This indicates that the model appropriately accounts for the major thermal phenomena, including the heat of adsorption and heat exchange with the column wall.

(a) Pressure in column [-]
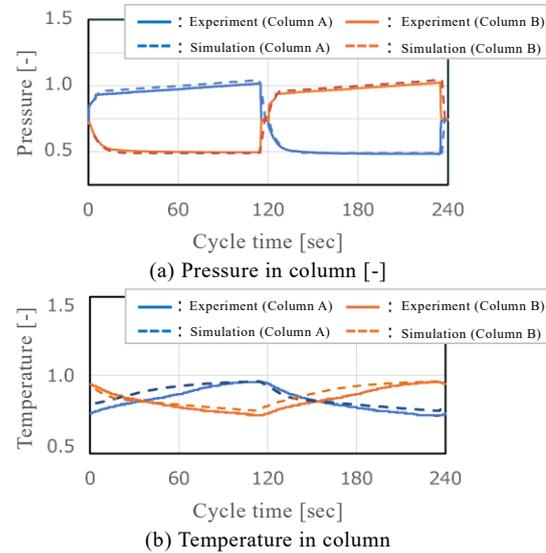

(b) Temperature in column

Fig. 2 Comparison of pressure and temperature in the two-column PSA (The vertical axes are normalized based on representative values)
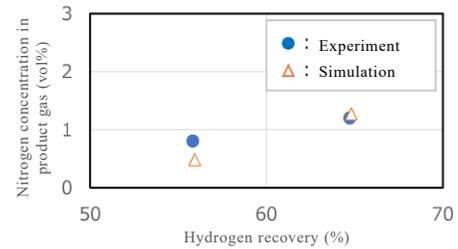

Fig. 3 Comparison of nitrogen concentration in the product gas extracted from the two-column PSA

In this study, the hydrogen recovery is defined as the ratio of the hydrogen amount in the product gas to the total amount of hydrogen in the feed gas. In a two-column PSA, a portion of the product gas is utilized as purge gas for the regeneration step; therefore, increasing the hydrogen recovery relatively reduces the purge amount, leading to an accumulation of residual nitrogen in the column. Consequently, a trade-off exists where an improvement in the hydrogen recovery results in an increase in nitrogen concentration in the product gas and a corresponding decrease in hydrogen purity. Fig. 3 shows a comparison between the experimental and simulation results for the relationship between the hydrogen

recovery and the nitrogen concentration (vol%) in the product gas for each operating condition. These results are generally in agreement. Since the simulation results fall within a practically acceptable range, this confirms that the proposed simulation is an effective means for the quantitative evaluation of two-column PSA processes.

3.2 Application to Four-Column VPSA

Following the two-column PSA, this section describes an application to a four-column hydrogen purification VPSA with a different number of columns and operating steps. Fig. 4 shows the schematic flow of the four-column VPSA. In this simulation, the same adsorption column model as that for the two-column PSA is employed. The parameters for equilibrium adsorbed amount and adsorption rate were identified based on the existing experimental data for the four-column VPSA, assuming that the same adsorbent as in the two-column PSA is used. Additionally, a vacuum pump model was newly incorporated to represent the evacuation process. Using this model, transient simulations were performed under the same column dimensions, feed gas conditions, and key operating conditions as those used in the experiments. The four adsorption columns are operated cyclically according to the steps shown in Table 2.

Fig. 5 shows a comparison between the experimental and simulation results for the relationship between the hydrogen recovery and the nitrogen concentration in the product gas for each operating condition. Although the nitrogen concentration (vol. ppm) in the product gas from the four-column VPSA is in a significantly lower range than that of the two-column PSA, the results are generally in agreement. This demonstrates that the same simulation framework is applicable even to processes where the number of columns and operating steps have been changed from those of the two-column PSA.
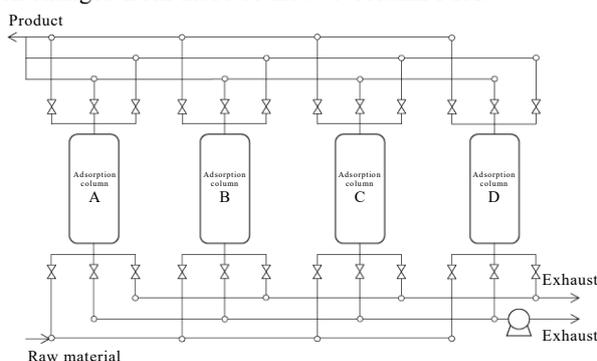

Fig. 4 Schematic flow of four-column VPSA

Table 2 Operation steps of four-column VPSA

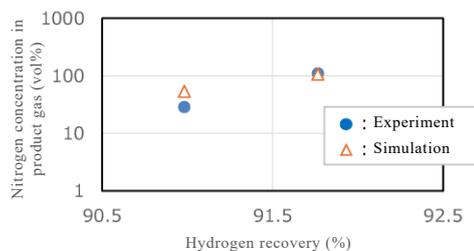|	Step 1	Step 2	Step 3	Step 4
Column A	Adsorption	Depressurization	Evacuation	Pressurization
Column B	Pressurization	Adsorption	Depressurization	Evacuation
Column C	Evacuation	Pressurization	Adsorption	Depressurization
Column D	Depressurization	Evacuation	Pressurization	Adsorption

Fig. 5 Comparison of nitrogen concentration in the product gas extracted from the four-column VPSA

4. Conclusion

We constructed a simulation model for the purpose of improving the efficiency of PSA process development and the reuse of technical knowledge. This paper has described the configuration of the model and provided an overview of the adsorption column model, which serves as a core component of the simulation model. Through applications to a two-column PSA and a four-column VPSA for hydrogen purification, it was confirmed that the simulated internal column behaviors and impurity concentrations in the product gas were in general agreement with the experimental results. Furthermore, it was demonstrated that stable calculations are possible even when increasing the number of columns, changing operating steps, or incorporating vacuum evacuation.

In this simulation model, process configurations and operating conditions can be arbitrarily set. By applying parameters identified from experimental data to the simulation model, it can be extended to PSA processes with different gas systems or configurations. Furthermore, by implementing this simulation model into a computational platform and developing an environment for its use, it is possible to conduct the screening of design conditions, equipment scale-up, and cycle studies prior to experimentation. This will reduce rework in test planning and shorten development lead times, and the effects of

digital technology are expected in terms of both improving development efficiency and the leveraging of technical knowledge.

In the future, we will extend this simulation model to other carbon-neutral PSA processes, including CO_2 capture/separation and biogas processing, to further contribute to the advancement and efficiency of PSA development.

References
1) T. Adachi, Hydrogen Purification for FCV from Ammonia Cracking Gas, Taiyo Nippon Sanso Technical Report, No. 37, 2018.
2) K. Yamamoto, CO_2 Recovery PSA Apparatus with Capacity of 10 Tons Per Day, Taiyo Nippon Sanso Technical Report, No. 42, 2023.