Reducing energy consumption of **advanced** PTSA CO₂ capture process—Experimental and numerical study

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Abstract

Climate change caused by the emission of greenhouse gases (GHGs), in particular carbon dioxide, has become a critical challenge. However, high energy penalty of CO₂ capture processes is still a critical bottleneck that restricts its commercial application. In this study, an advanced pressure-temperature swing adsorption (PTSA) CO₂ capture process by integrating **chemical heat transformer** and pressure recovery is investigated to reduce energy requirement. To evaluate the energy consumption of the proposed adsorption process, experimental and numerical study were both carried out. Initially, the adsorption kinetics (*i.e.* adsorption isotherm, breakthrough curve and isosteric heat) of the sorbent (zeolium[®]F9-HA) were tested using a fixed-bed setup. Based on the obtained experimental data, the energy consumption of the advanced PTSA process was evaluated by a commercial process-simulation software (PRO/II ver. 9.1, Invensys). The simulation results indicate that the energy consumption of the proposed PTSA process decreased to 1.18 MJ/kg CO₂ (approximately 40% that of the conventional PTSA process).

Keywords: CO₂ capture, pressure temperature swing adsorption, heat integration, energy consumption

Nomenclature

H	Enthalpy, J
T	Temperature, °C
ΔT	Temperature difference, K
Q	Heat, J
W	Work, J
c_p	Heat capacity, J/K

Greek letters

v	Flow rate, kmol/hr
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Abbreviations

<i>CCS</i>	CO ₂ capture and storage
<i>GHG</i>	Greenhouse gas
<i>PTSA</i>	Pressure-temperature swing adsorption
<i>CHT</i>	Chemical heat transformer
<i>NGCC</i>	Natural gas combined cycle
<i>PCC</i>	Post-combustion capture
<i>MEA</i>	Monoethanolamine

1. Introduction

CO₂ capture and storage (CCS) is considered to be the most significant strategy to mitigate climate change [1,2]. The whole CCS strategy mainly includes CO₂ capture, transport and storage section. CO₂ capture is considered as the most important and energy-intensive part, and which contributes 75~80% of the total cost of CCS [3,4]. Nowadays, post combustion CO₂ capture is the most dominant strategy (others contain pre-combustion and oxy-fuel combustion CO₂ capture), due to its advantage of retrofitting in existing industrial stations (*i.e.* coal-fired power plant, steel industry and cement factory etc.) [5]. For post combustion CO₂ capture, several technologies (such as absorption, adsorption, membrane and cryogenic) has been widely researched [6]. Amine-based absorption is one of the most common method and has been commercially applied at industrial scale [7,8].

However, the energy requirement for CO₂ capture need to be further optimized to reduce overall energy consumption [9,10]. According to the National Energy Technology Laboratory (NETL) report of 2007, the integration of CCS units (such as MEA absorption) into a retrofitted coal-fired power plant for 90% CO₂ capture can reduce the thermal efficiency from an approximate 35% HHV (high heating value) basis to 24% at a cost of \$80 to capture a ton of CO₂ [11]. Sipöcz and Tobiesen compared the performance of a natural gas combined cycle (NGCC) power plant with and without CCS units, and their results indicated that the energy efficiency decreased from 58.29%

to 49.48% upon retrofitting with a monoethanolamine (MEA) absorption process [12]. Goto *et al.* reviewed the efficiency penalty of coal-fired power plants with CCS [13]. They stated that the efficiency penalty of CCS units was about 10%, and the ratio of the efficiency penalty by CO₂ capture to the total efficiency penalty was around 60–70%. Therefore, the major challenge for existing CO₂ capture technologies is to reduce the energy consumption and thus the capital and operational costs.

As an effective alternative for amine-based absorption processes, adsorption processes using solid sorbents (*i.e.* activated carbonaceous materials, microporous/mesoporous silica or zeolites, carbonates, and polymeric resins etc.) have several advantages including a low energy penalty for regeneration [14-16], easy handling compared with liquid solvents [17], low capital investment [18]. Adsorption-desorption of CO₂ can be achieved by pressure/vacuum, temperature or electrical swing [19]. The energy requirement of regeneration for solid sorbents is significantly reduced compared to alkaline absorption processes, due to avoiding the heat of water in solution [15]. Moreover, the heat capacity of solid sorbent is comparatively lower than that of an aqueous amine solvent.

Pressure temperature swing adsorption (PTSA) is a potential adsorption approach for CO₂ capture from large CO₂ sources [20]. The combination of pressure swing adsorption (PSA) and temperature swing adsorption (TSA) shows significant advantages [21,22]. In the hybrid process, the pressure is required in adsorption stage and temperature increasing is needed for sorbent regeneration in desorption stage.

Compared with the conventional TSA approach, the operating cost of PTSA process can be reduced due to lower regeneration temperature requirement [23]. Meanwhile, the regeneration pressure is normal pressure than that of vacuum conditions required in VSA process [24]. Pugsley *et al.* exploited an advanced CO₂ separation process known as the circulating fluidized bed pressure–temperature swing adsorber [25]. The simulation study performed at various riser operating conditions and flue gas CO₂ concentrations and results indicated that the CO₂ recovery decreases with increasing concentration at a fixed solids circulation flux, but the purity of the recovered product increases. Ghougassian *et al.* proposed an advanced heat-integrated PTSA CO₂ capture process [26]. A vapor compression heat pump is used to circulate adsorption heat between adsorber and desorber. Energetic analysis suggests that an 11.49% increase in net power generation can be attained for 90% CO₂ capture by employing MgO-based CO₂ adsorption instead of the amine process. Recently authors proposed SHRT (self-heat recuperation technology) [27,28]. SHRT recuperate all of process heat and circulated it into the process, leading to significant energy saving of the chemical process. By following SHRT, not only process heat but also absorption heat is recuperated.

The aim of this study was to put forward an advanced PTSA CO₂ capture process based on SHRT with heat circulation and waste pressure recovery. To achieve the target, the adsorption kinetics (*i.e.* adsorption isotherm, breakthrough curve and isosteric heat) of the sorbent (zeolite F-9, TOSO Ltd.) was examined in experimental section. The

adsorption heat was then calculated based on the isotherm curves. Based on the experimental data, the energy consumption and CO₂ recovery of the proposed PTSA CO₂ capture process was numerically studied using a commercial simulation software (PRO II 9.1, Invensys). The influence of CO₂ concentration, pressure and temperature on the energy consumption of the conventional and proposed PTSA CO₂ capture processes was also evaluated in process simulation section.

2. Processes description

2.1 Base case

A typical PTSA CO₂ capture process mainly consists of two or more fixed-bed columns, where each bed goes through a sequence of elementary steps. A schematic two-column configuration is shown in Fig. 1. The flue gas is initially introduced into the adsorption column. At high pressure, the CO₂ in the gas mixture is adsorbed by the sorbent, and any other gas is expelled from the top of the adsorption column with very little adsorption. Since the adsorption of CO₂ is an exothermic process, the temperature in the column increases accordingly. To avoid the adverse influence of adsorption, a cooling water unit is typically used to maintain the temperature of the adsorber in a desired range [29]. The regeneration of the sorbent is an endothermic process, and thus additional heat is required. For the PTSA process, the regeneration of the sorbent is realized by heating the desorption column [30].

2.2 Advanced PTSA CO₂ capture process

To reduce the energy requirement, an advanced PTSA capture process based on adsorption heat circulation and waste pressure recovery was designed. The schematic diagram of the proposed PTSA CO₂ separation process is shown in Fig. 2. As a result all of process heat are recuperated and circulated into the process by following SHRT. The waste pressure of the residual gas at the top of the adsorber is recovered by an expander to generate electricity. The advantage of the proposed PTSA process is that a total capture process can be achieved while only consuming electrical energy (by compressor), and this avoids the requirement for additional heat.

Chemical heat transformer (CHT) mainly consists of a compressor and heat exchangers [31, 32]. In CHT, the liquid working fluid (with a low boiling point) initially exchanges heat with the column of adsorption, and it is heated into a vapor state. The hot working fluid is then further pressurized by a compressor. The obtained high temperature and pressure working fluid then undergo heat exchange with the column for desorption. The exothermic heat of adsorption is recuperated and circulated to the regeneration process. The working fluid (vapor state) after heat exchange with desorption column is continually depressurized by a valve. A liquid working fluid (low temperature and pressure) is then regenerated for the next isosteric heat recuperation cycle.

3. Experimental section

3.1 Apparatus and conditions

To provide a reference for the simulation, the sorbent characteristics (*i.e.* adsorption capacity and adsorption heat) were experimentally tested. The structure of the apparatus for the CO₂ adsorption/desorption cycle experiment is shown in Fig. 3. The fixed-bed absorber was composed of a stainless steel tube (OD×ID×L=13×9×20 mm, Swagelok Ltd., Japan), and filled with Zeolum[®]F-9HA (supplied by TOSO Ltd, Japan). The feed gas was simulated using N₂ and CO₂, and it was prepared as a gas mixture (Fujikin Ltd., Japan). The Zeolum[®]F-9HA was previously treated by passing 1000 mL/min of N₂ at 300 °C over 6 h. After pretreatment, the temperature of the fixed bed was adjusted to the desired value (25, 50, 75 and 100 °C) under different partial pressure (balanced by N₂). The adsorption process is lasted for 60 min in all the experiments. After the adsorption test, the gas flow of binary mixture is turned to pure N₂, and the temperature of fixed bed is heated to 150 °C to regenerate sorbent. The gas flow rate of N₂ and CO₂ was controlled by mass flow controllers (Bronkhorst El-flow) and recorded by flow sensors (Keyence Ltd., Japan). The pressure in the adsorption bed was monitored using an analog pressure indicator (Yokogawa Ltd., Japan). The temperature in the packed bed was measured using three thermocouples (Thermo-Electric K-types) at three axial positions (inlet, center and outlet). The properties of the sorbent are listed in Table 1.

3.2 Adsorption isotherm

The detailed CO₂ adsorption and desorption processes can be described as follows:

The adsorption isotherm of CO₂ by the sorbent can be fit by Langmuir model, which is given as follows:

$$q = \frac{q_{\infty} \cdot k \cdot p}{1 + k \cdot p} \quad (3)$$

where q is the adsorption capacity of the sorbent (mol CO₂/kg_{ads}), q_{∞} is the infinite absorption capacity (mol CO₂/kg_{ads}), k is isotherm parameter, and p is the partial pressure of CO₂ (MPa).

3.3 Isostatic heat

Based on the obtained adsorption isotherm, the exothermic and endothermic heat of the simulation process can be further calculated by the Clausius–Clapeyron equation. The detailed calculation process has been described by Chao, 2011 and written as follows [33]:

$$\frac{-\Delta H}{R} = \left[\frac{\partial \ln P}{\partial (1/T)} \right]_q \quad (4)$$

where P is the pressure, T is the temperature, q is the amount of adsorbed CO_2 , R is the gas constant, and ΔH denotes the exothermic heat of adsorption.

4. Process modeling and calculation

4.1 Mass balance

To better understand the adsorption isotherm, accurately model mass and energy balance in adsorption and desorption process is significant. The mathematical model is based several assumptions to simplify the complexity. 1) The gradient of concentration and temperature in radial direction is negligible. 2) The gas phase (CO_2 and N_2) behaves as an ideal gas. 3) Thermal equilibrium between sorbent and bulk gas is assumed.

The overall mass balance can be calculated by [34,35]:

$$\frac{\partial C}{\partial t} + \frac{1-\varepsilon_b}{\varepsilon_b} \cdot \rho_s \cdot \sum_{j=1}^2 \frac{\partial q_j}{\partial t} + \frac{1}{L} \cdot \frac{\partial (u_g \cdot C)}{\partial z} = 0 \quad (5)$$

where C and q represent the bulk phase and adsorbed phase concentrations. L is the bed length, z is the length in the axial direction, u is the superficial velocity of the gas mixture, ε_b is the bed porosity, t is time, ρ_s is the adsorbent density.

The rate of mass transfer between C_i and q_i is written as:

$$\frac{\partial q_i}{\partial t} = K_{L,i} \cdot (q_i^* - q_i) \quad (6)$$

where K_L is the overall mass transfer coefficient of component i and q_i^* is the amount of component adsorbed at equilibrium.

$$q_i^* = f(C_i, T_g) \quad (7)$$

here, C_i is the concentration of component i in gas phase. T_g is the temperature of bulk gas.

Thus, the mass balance of component i (CO_2 or N_2 in this study) in the bulk phase can be written by:

$$\frac{\partial C_i}{\partial t} + \frac{1 - \varepsilon_b}{\varepsilon_b} \cdot \rho_s \cdot \frac{\partial q_i}{\partial t} - \frac{D_{ax}}{L^2} \cdot \frac{\partial^2 C_i}{\partial z^2} + \frac{1}{L} \cdot \frac{\partial (u_g \cdot C_i)}{\partial z} = 0 \quad (8)$$

here D_{ax} is the mass axial dispersion coefficient.

4.2 Energy balance

The overall energy balance of adsorption process consists of three parts: bulk gas phase, solid sorbent, and wall of adsorption column [36].

- 1) Energy balance of bulk gas phase is depicted in Eq. (9).

$$\begin{aligned} \varepsilon_b \cdot \rho_g \cdot c_g \cdot \frac{\partial T_g}{\partial t} = & k_g \cdot \frac{\partial^2 T_g}{\partial z^2} \cdot \varepsilon_b - (1 - \varepsilon_b) \cdot h_i \cdot \frac{3}{r_s} \cdot (T_g - T_s \Big|_{r=r_s}) \\ & - u_g \cdot \frac{\partial T_g}{\partial z} \cdot \varepsilon_b \cdot \rho_g \cdot c_g - \frac{4}{r_b} \cdot h_i \cdot (T_g - T_w) \end{aligned} \quad (9)$$

where c_g is the heat capacity of gas phase. T_s and T_w are the temperature of sorbent and wall of column, respectively. k_g is the thermal conductivity of gas. r_b and r_s are the radius of bed and sorbent, respectively. h_i is the heat transfer coefficient between gas and wall. ρ_g is the gas density.

2) Energy balance of sorbent can be calculated as the following equation.

$$\begin{aligned} [\varepsilon_s \cdot \rho_s \cdot c_g + (1 - \varepsilon_s) \cdot \rho_s \cdot c_s] \cdot \frac{\partial T_s}{\partial t} = & k_{gs} \cdot \left[\frac{1}{r^2} \cdot \frac{\partial}{\partial r} (r^2 \cdot \frac{\partial T_s}{\partial r}) \right] \\ & + \Delta H_{ads} \cdot \rho_s \cdot \frac{\partial q}{\partial t} \end{aligned} \quad (10)$$

where ε_s is the sorbent porosity. c_s is the heat capacity of sorbent. ΔH_{ads} is the adsorption heat of sorbent.

3) Energy balance of the wall of adsorption column is described in Eq. (11).

$$q_{N_2}(0, z) = q_{N_2}^* \quad (11)$$

where d_i and d_o are the inside and outside diameter of the column. c_w is the heat capacity of column wall. ρ_w is the density of column wall. T_w and T_a are the temperature of column wall and ambient. h_o is the heat transfer coefficient between wall and ambient.

4.3 Boundary condition

Initially, the adsorption column is purged by pure N₂. The boundary conditions are assumed as the following:

$$q_{CO_2}(0, z)=0, \quad q_{N_2}(0, z)=q_{N_2}^*, \quad T(0, z)=298.5 \text{ K}, \quad P(0, z)=101.3 \text{ kPa} \quad (12)$$

where $q_{N_2}^*$ denotes the adsorbed N₂ concentration in equilibrium under the initial temperature and pressure.

4.4 Simulation section

To achieve the target of reducing energy consumption, the performance estimation overall capture processes with and without **CHT** and waste pressure recovery was investigated using the commercial process simulator PRO/II ver. 9.1 (Invensys). The exothermic and endothermic heat of the adsorption process was calculated based on the experimental results. It should be pointed out that pentane was used as the working fluid for the **CHT** in the simulation work. The energy consumption of these processes was calculated based on the following assumptions: 1) The thermodynamic calculation was performed based on the Soave-Redlich-Kwong (SRK) equation. 2) It is assumed that heat loss to the environment is small. 3) The adiabatic efficiency of the compressor and expander was 85%. 4) The minimum approach temperature in all heat exchangers is 10 K.

The conditions of the conventional and proposed PTSA processes for process

simulation are summarized in Table 2. The temperature of sorbent regeneration is set at 150 °C. The pressurization work, feed temperature, CO₂ concentration and recovery are varied to evaluate their influence on the capture performance. It should be noted that the energy consumption in this work contains only CO₂ capture treatment, and compression for transport and storage is not considered here.

5. Results and discussion

5.1 Adsorption kinetics

The adsorption isotherm of Zeolum[®] F-9HA is shown in Fig. 4. To estimate the isosteric heat by Clausius-Clapeyron equation, the adsorption isotherm is tested at four sets of temperatures (25, 50, 75 and 100 °C). The adsorption isotherm of CO₂ is depicted in Fig. 4 (a). At the ambient temperature (25 °C), the adsorption capacity of CO₂ increased with its partial pressure. When the partial pressure varied from 0 kPa to 100 kPa, the adsorption capacity of CO₂ increased from 0 to 5.7 mmol/g_{adsorbent}. Meanwhile, with the increase of adsorption temperature (from 25 to 100 °C), the adsorption capacity of CO₂ decreased. When the adsorption temperature increased to 100 °C, the adsorption capacity of CO₂ decreased to 1.3 mmol/g_{adsorbent}. The high adsorption capacity of CO₂ can be explained by its smaller molecular size and the interaction of quadrupole moment of CO₂ with polar surface of Zeolum[®] F-9HA. Fig. 4 (b) shows the adsorption isotherm of N₂. The adsorption capacity of Zeolum[®] F-9HA

for N₂ is obviously lower compared with that of CO₂. When the adsorption temperature is set 25 °C, the adsorption capacity of N₂ increased with its partial pressure, and reached 0.45 mmol/g_{adsorbent} at 100 kPa. But this value rapidly decreased to 0.03 mmol/g while the adsorption temperature increased to 100 °C. It can be attributed to lower adsorbent affinity toward N₂ compared to CO₂, especially at high temperature.

As a significant characteristic of dynamic adsorption process, breakthrough curve of CO₂ is tested under different adsorption temperatures (as shown in Fig. 5). Initially, most of the mass transfer takes place at the bed inlet where the feed gas first makes contact with the sorbent. As time goes on, the sorbent near the inlet becomes saturated, and adsorption zone moves down towards the bed exit. Meanwhile, the detected CO₂ concentration obviously changed (shown as “S” shape in the curves). At the break point, the sorbent in the fixed bed is completely saturated, and the adsorption zone disappeared. The CO₂ concentration in residual gas becomes equal to that in feed stream. It should be noted that the adsorption temperature adversely affected CO₂ adsorption process. A higher adsorption temperature (*i.e.* 100 °C) leads to a faster breakthrough time. That can be attributed that CO₂ adsorption capacity at low temperature (*i.e.* 25 °C) is higher than that of 100 °C. Thus, CO₂ concentration front takes more time to pass the fixed bed at low temperature. When the adsorption temperature is set at 25 °C, the CO₂ concentration front reaches the bed outlet after 47 min. But this value decreases to 8 min with the adsorption time increased to 100 °C.

The isosteric heat of CO₂ under different adsorption temperatures is shown in Fig. 6. As shown in these curves, the CO₂ adsorption performance weakened with an increase in temperature (from 25 °C to 100 °C). When the adsorption temperature is set at 25 °C, the isosteric heat kept as a constant at the initial 40 min, and then decreased due to saturation adsorption of sorbent. By contrast, the adsorption performance at 100 °C is significantly reduced. After 10 min, the isosteric heat is decreased to 8 kJ/mol. That is due to the sorbent activity decreased with the temperature increasing. Therefore, most of CO₂ in the gas stream can not be adsorbed in the fixed bed at high temperature. The result of isosteric heat indicates that the optimal adsorption temperature is 25 °C.

5.2 Sensitivity analysis by process simulation

5.2.1 Effect of CO₂ concentration on energy consumption

The influence of CO₂ concentration on the capture performance of the conventional and proposed PTSA processes was investigated. The simulated results are shown in Fig. 7. As shown by these results, the energy consumption of the conventional process decreased from 8.89 to 1.83 MJ/kg CO₂ with the CO₂ concentration variation from 5% to 40% (at a CO₂ recovery of 70%). By contrast, the energy consumption of the proposed process that was integrated with isosteric heat and pressure recovery decreased from 3.52 to 0.81 MJ/kg CO₂ when the CO₂ concentration increased to 40% with the same CO₂ recovery. For a typical flue gas from a coal-fired power plant (CO₂

concentration of 15%), an approximate 60% energy penalty is avoided in the advanced PTSA process compared with the conventional process (CO₂ recovery set to 95%). In Fig. 7 (b), when the CO₂ recovery increased from 70 to 95%, around 1 MJ/kg CO₂ reduction is evident for the flue gas with a low CO₂ concentration (5%). However, with an increase of CO₂ concentration (to 40%) in the feed stream, the avoidable energy penalty decreased significantly. That is because that the increase of CO₂ concentration lead to a higher partial pressure in adsorption process, which is favour of enhancing adsorption capacity of adsorbent. In addition, the improvement of CO₂ recovery (from 70% to 95%) is also benefit to reduce energy consumption. That is due to a better CO₂ recovery means more CO₂ could be captured under the same condition. Then, the relative energy consumption of per kg captured CO₂ can be reduced.

The proportion of recovered energy (isosteric heat and waste pressure) varied with CO₂ concentration, as shown in Fig. 8. From these results, for the same CO₂ recovery the recovered energy by pressure recuperation decreased with an increase in CO₂ concentration. For the proposed PTSA process, it varied from 3.50 to 0.28 MJ/kg CO₂ with a CO₂ recovery of 95% while the CO₂ concentrated from 5 to 40%. By contrast, the energy recovered by isosteric heat remained constant (0.53 MJ/kg CO₂). This is because the isosteric heat is dependent on the adsorption of CO₂, and this did not vary with the other conditions. The recovered energy from the residual pressure also decreased with an increase in CO₂ recovery. This can be explained by considering that

with an increase in CO₂ recovery, the waste pressure of the effluent stream decreases accordingly.

5.2.2 Effect of pressurization on energy consumption

The effect of pressurization on the conventional and proposed PTSA CO₂ separation performance is shown in Fig. 9. As shown by these results, the energy consumption increases with an increase in pressurization work, particularly for the case without the adsorption heat and residual pressure recovery. For the conventional PTSA process, the energy consumption increased from 2.75 to 4.04 MJ/kg CO₂ with a variation in pressurization from 404 to 808 kPa (with a CO₂ recovery of 95%). This is because the input energy of the compressor contributed most of the total energy consumption of the process. The waste pressure of the residual gas will increase with inlet pressurization work. Meanwhile, the pressure loss during adsorption process also increased with pressurization work. Therefore, a high pressure leads to high energy consumption in the adsorption process. However, it is obvious that the proposed process can effectively reduce the energy penalty by waste pressure recovery. The energy requirement proposed PTSA process varied from 1.18 to 1.83 MJ/kg CO₂ with gas pressure increased from 404 to 808 kPa (CO₂ recovery set at 95%). When the pressure of the feed gas was compressed to 404 kPa, the energy consumption of the proposed process was 1.18 compared with the 2.75 MJ/kg CO₂ of the conventional method. This means that approximately 57.1% of the input energy can be saved. It should be pointed out that for

the pressurization, lower is not necessarily better. The CO₂ in the feed stream will not be captured effectively if the pressurization work is too low and this would lead to a low CO₂ recovery.

The proportion of recovered energy varied with pressurization, as shown in Fig. 10. From these results, at the same CO₂ recovery the recovered energy by pressure recuperation increased with pressure. With the pressure of feed gas varying from 404 to 808 kPa (with a CO₂ recovery of 70%), the recovery energy of waste pressure increased from 2.01 to 2.89 MJ/kg CO₂. That is due to the partial pressure of the effluent stream (mainly N₂) increasing with the input pressurization work. When the inlet feed gas is compressed to high pressure, more waste pressure can be recovered from the residual gas. Additionally, the recovered energy from the isosteric heat is constant (0.53 MJ/kg CO₂).

5.2.3 Effect of feed gas temperature on energy consumption

The influence of feed gas temperature on the conventional and proposed PTSA adsorption performance is shown in Fig. 11. When the feed gas temperature was set at 25 °C, the energy consumption of the conventional PTSA process was 2.66 MJ/kg CO₂ with a CO₂ recovery of 95%. After introducing **CHT** and expander to the proposed process, the energy penalty was reduced to 1.15 MJ/kg CO₂ under the same conditions. This indicates that around 56.8% of the energy can be saved in the proposed process.

Furthermore, the total energy requirement of the two processes will increase with an increase in temperature (from 25 °C to 100 °C). For the conventional process, the energy consumption increased from 2.66 to 3.04 MJ/kg CO₂ (with a CO₂ recovery of 95%). By contrast, this varied from 1.15 to 1.33 MJ/kg CO₂ for the proposed case. That is attributed that the increasing of feed temperature adversely affected the adsorption capacity of sorbent. In order to keep the same CO₂ recovery, the energy requirement consequently increased. Additionally, the input energy decreased with an increase in CO₂ recovery. For the proposed process, the energy consumption decreased from 1.41 to 1.15 MJ/kg CO₂ (inlet temperature of 25 °C) and the CO₂ recovery increased from 70 to 95%. That can be explained by the fact that a higher CO₂ recovery represents more CO₂ can be captured. Therefore, the average energy consumption of per unit CO₂ is accordingly decreased.

Details about the proportion of recovered energy that varied with gas temperature is shown in Fig. 12. The results show that for the same CO₂ recovery the energy recovered by pressure recuperation increased with feed gas temperature. When the inlet temperature raised from 25 to 100 °C, the recovered energy of waste pressure varied from 1.38 to 1.57 MJ/kg CO₂ with CO₂ recovery of 70%. This is due to under a high temperature the stream can be compressed into a high-pressure state, which means that more waste pressure can be recovered from the outlet of the adsorber in the proposed process. Meanwhile, the energy recovered by the isosteric heat remained constant (0.53 MJ/kg CO₂).

6. Conclusions

An advanced PTSA CO₂ capture process based on isosteric heat circulation and waste pressure recovery was investigated. The adsorption kinetics (*i.e.* adsorption equilibrium, breakthrough curve and isosteric heat) of the sorbent (Zeolum[®]F-9HA) for CO₂ capture was examined using a fixed-bed setup. Based on the experimental results, the exothermic heat of adsorption was calculated. Additionally, the influence of several parameters (*i.e.* CO₂ concentration in the flue gas, pressurization and feed gas temperature) was studied using the commercial simulator PRO/II ver. 9.1 (Invensys). According to the experimental and simulation results, the conclusions can be summarized as follows:

- In the proposed CO₂ capture process, adsorption heat 0.53 MJ/kg CO₂ can be recovered by **chemical heat transformer (CHT)**. Furthermore, the waste pressure of the residual gas can also be recovered by an expander.
- The energy consumption of the proposed PTSA CO₂ capture process decreased with CO₂ concentration in feed stream, and increased with the pressurization work and inlet temperature.
- **When the CO₂ concentration was 15% (for a typical coal-fired power plant), the pressure and temperature of feed gas were 404 kPa and 25 °C, the energy consumption of the proposed CO₂ capture process decreased to 40% that of the conventional process, which was found to be 1.18 MJ/kg CO₂ with a CO₂ recovery of 95%.**

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Figure captions

Fig. 1. Schematic diagram of the conventional PTSA CO₂ capture process.

Fig. 2. Energy balance of the proposed PTSA CO₂ capture process.

Fig. 3. Schematic of the experimental setup.

Fig. 4. Adsorption isotherm of CO₂ (a) and N₂ (b) by Zeolum[®] F-9HA. The adsorption temperature was varied at 25, 50, 75 and 100 °C.

Fig. 5. Breakthrough curves of CO₂ at different adsorption temperatures (25, 50, 75 and 100 °C). C/C_0 represents the CO₂ concentration in the residual gas relative to that of feed gas.

Fig. 6. Isostatic heat of CO₂ generated during the adsorption stage at different adsorption temperatures (25, 50, 75 and 100 °C). (Flow rate is set at 30 mL/min, N₂/85 vol.% and CO₂/15 vol.%)

Fig. 7. Effect of CO₂ concentration on the energy consumption of the conventional process (a) and the proposed (b) TSA process. (The pressure of pressurization and temperature of feed gas are set at 404 kPa and 25 °C).

Fig. 8. Energy recovered by isosteric heat and waste pressure of the proposed PTSA process at different CO₂ concentrations.

Fig. 9. Effect of pressurization on energy consumption in the conventional (a) and the proposed (b) PTSA process. (The CO₂ concentration and gas temperature are set at 15% and 25 °C).

Fig. 10. Recovered energy as a function of isosteric heat and waste pressure of the proposed process at different pressures.

Fig. 11. Effect of feed gas temperature on energy consumption in the conventional (a) and proposed (b) PTSA processes. (The CO₂ concentration and pressure of pressurization are set at 15% and 404 kPa).

Fig. 12. Recovered energy as a function of the pressure and isosteric heat of the proposed process at different feed gas temperatures.

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Table 1

Properties of the sorbent (Zeolum[®]F-9HA, TOSO Ltd, Japan).

Properties	Value
Density of sorbent (ρ_a)	1.4 g/cm ³
Bulk density (ρ_b)	0.62 g/cm ³
Porosity (ϵ_b)	0.348
particle diameter (D_p)	1.5 mm
Thermal conductivity (k)	3.49×10^{-4} J/mm·sec ⁻¹ ·K ⁻¹
viscosity coefficient (μ)	1.639×10^{-5} g/mm·s ⁻¹
heat capacity (C_a)	0.92 J/g·K ⁻¹

Table 2

Simulation conditions for the conventional and proposed PTSA processes*.

Parameters	Value
Flow rate	600 kg/s
Feed temperature	25-100 °C
Feed pressure	101 kPa
Pressurization	404-808 kPa
Feed gas composition	
CO ₂	5-40 wt. %
N ₂	60-95 wt. %
CO ₂ recovery	70-95%

* The sorbent used in the simulation process is defined as Zeolum® F-9HA (supplied by TOSO Ltd, Japan).

Base case

Advanced PTSA process

Graphic abstract