Scaling Rules and Intensification of Thermal Swing Adsorption

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Thermal swing adsorption (TSA) processes have been used in the industry for a long time, and reviewed by Ruthven (1984), Wankat (1986, 1990), and Yang (1987). Progress in TSA processes has been insignificant compared to pressure swing adsorption (PSA) systems. The causes are twofold: while in TSA systems the cycle time is usually very long (two hours to several days) and there is often a large energy loss, in PSA systems the cycle time is short and the absorbent productivity is higher. To improve TSA processes, one can explore reducing the cycle time and reducing the heat loss. Consider a TSA process with a feed (adsorption) step, a heating (regeneration) step, and a cooling step. The feed is introduced into the column at low (usually ambient) temperature. During regeneration the hot gas flows counterflow to the adsorption step. If solute-free gas is available for cooling, it is passed into the bed in the same direction as for heating. Otherwise, cold gas is passed into the bed in the direction for adsorption and deposits some adsorbate in the bed near the inlet. In some instances, when a pulse of hot effluent does not disrupt the process, the cooling step may be omitted entirely.

Most sorption separators can be decreased in size if the particle diameter, column length and diameter, fluid velocity, and cycle time are scaled appropriately. Such scaling analyses have been carried out for adsorption (Wankat, 1987), chromatography (Wankat and Koo, 1988), isothermal PSA (Kowler and Kadlec, 1972; Suh and Wankat, 1990), and for adiabatic PSA processes (Rota and Wankat, 1990). This note extends these analyses to TSA processes.

For the scaling procedure, assume that there is an existing, satisfactory "old" design based on simulations, laboratory experiments, or a commercial unit. The scaling procedure will not change the absorbent, feed, cooling, and regeneration temperatures, or feed gas concentration. The effects of changes in particle diameter and throughput on the column length, column diameter, inlet gas velocity, pressure drop, adsorbent productivity, and separation will be explored. The goal is to keep pressure drop and separation constant while improving absorbent productivity. A dimensional analysis method (Kowler and Kadlec, 1972; Suh and Wankat, 1990; Rota and Wankat, 1990) will be presented.

Dimensionless Analysis

The mass and energy balance and transfer equations are the same as in the adiabatic PSA. Since the only differences between the scaling procedures for the TSA and PSA lie in the boundary and initial conditions, we will show the boundary and initial conditions for each step of the TSA process and then give the scaling results from dimensionless analyses of TSA systems. Rota and Wankat (1990) and Chen (1990) explain the procedure in detail. The column was assumed to be adiabatic, and storage of energy in the column wall was neglected. Without these assumptions, simple scaling rules are not possible, since one cannot keep heat losses the same in the two designs. The TSA and PSA processes have different extra-column effects, as will be discussed later.

If axial dispersion is neglected, the differential equations for the mass and energy balances are first-order with respect to both dimensionless Z and τ. Only one boundary condition and one initial condition are needed for each variable involved. Let $\tau_I = \tau_{FD}$, $\tau_{II} = \tau_{FD} + \tau_{HT}$, $\tau_{III} = \tau_c$, where τ_I, τ_{II}, and τ_{III} represent the dimensionless time at the end of feed step, the heating step, and the cooling step, respectively, where $\tau = 0$ indicates the initial time for each cycle. The initial conditions at the beginning of each step are the conditions at the end of the previous step. The following boundary and initial conditions are the same for each cycle.

During the adsorption step (*FD*):

$$P(\tau,0)=P_{FD},\quad C_i(\tau,0)=C_{i,f},\quad U(\tau,0)=U_{FD},\quad \Gamma(\tau,0)=\Gamma_{FD}$$

$$C_i(0,Z)=C_i(\tau_c,Z),\quad Q_i(0,Z)=Q_i(\tau_c,Z),\quad \Gamma(0,Z)=\Gamma(\tau_c,Z) \qquad (1)$$

where P, C, U, and T are dimensionless pressure, concentration, velocity, and temperature, respectively. During the heating step (*HT*):

$$P(\tau,1)=P_{HT},\quad C_i(\tau,1)=C_{i,h},\quad U(\tau,1)=U_{HT},\quad \Gamma(\tau,1)=\Gamma_{HT} \qquad (2)$$

$C_{i,h} = C_{i,f}$ when hot feed gas is used to regenerate the absorbent bed during the heating step. During the cooling step (*CL*):

$$P(\tau,1)=P_{CL},\quad C_i(\tau,1)=0,\quad U(\tau,1)=U_{CL},\quad \Gamma(\tau,1)=\Gamma_{CL} \tag{3}$$

The total dimensionless cycle time is

$$\tau_c=\tau_{FD}+\tau_{HT}+\tau_{CL} \tag{4}$$

The scaling procedure consists of defining a scaling operator R_X,

$$R_X=R(X)=\frac{X_{new}}{X_{old}} \tag{5}$$

Assume that one wants to keep recovery and product concentration constant.

$$R_{\text{recovery}}=1,\ R_{\text{prod.conc.}}=1 \tag{6}$$

Equation 6 will be satisfied if the dimensionless groups and boundary conditions for the old and new designs are identical. The requirement that the boundary and initial conditions be kept constant means that

$$R(c_{if})=R(c_{ih})=R(T_{FD})=R(T_{HT})=R(p_o)=1 \tag{7}$$

and

$$R_{\tau_{FD}}=R_{\tau_{HT}}=R_{\tau_{CL}}=1 \tag{8}$$

Equation 7 requires that the old and new designs process the same gas, the same inlet temperatures and pressures. Equation 8 states that the *relative* cycle times are unchanged. The requirement of constant dimensionless groups leads to

$$R(\text{absorbent})=R(P,\text{ all }Z,\tau))=R(\Gamma,\text{ all }Z,\tau)=1 \tag{9}$$

Equation 9 occurs because the Langmuir equilibrium constant is part of a dimensionless group. Equation 9 requires that the dimensionless pressure and temperature profiles are the same, and the same adsorbent must be used. Equation 6 will be satisfied if

$$R(C_i,\text{ all }Z,\tau)=1 \tag{10}$$

and Eq. 8 is satisfied. Thus, the two designs also have identical dimensionless concentration profiles. The remaining scaling rules depend on the controlling transfer mechanism. These rules are given in Table 1 and are the same as for the adiabatic PSA (Rota and Wankat, 1990). The results for film diffusion can also be obtained by using the exact analytical solution, instead of the dimensionless analysis (Chen, 1990).

Table 1. Scaling Rules for Different Mass and Heat Transfer Mechanisms

Controlling Transfer Mechanism		$R_{u_{FD}}$	R_{d_p}	R_D	R_L
Heat	Mass				
Equil.	Equil.	any	$R_{u_{FD}}R_{l_c}^{0.5}$	$R_{N_{FD}}^{0.5}R_{u_{FD}}^{-0.5}$	$R_{u_{FD}}R_{l_c}$
Film	Equil.	$R_{l_c}^{0.446}$	$R_{l_c}^{0.946}$	$R_{N_{FD}}^{0.5}R_{l_c}^{-0.223}$	$R_{l_c}^{1.446}$
Film	Film				
Equil.	Film				
Internal	Equil.	1	$R_{l_c}^{0.5}$	$R_{N_{FD}}^{0.5}$	R_{l_c}
Internal	Internal				
Equil.	Internal				
Equil.	Series	*	$R_{u_{FD}}R_{l_c}^{0.5}$	$R_{N_{FD}}^{0.5}R_{u_{FD}}^{-0.5}$	$R_{u_{FD}}R_{l_c}$
Series	Equil.				

*Numerical calculation required (Rota and Wankat, 1990); Chen, 1990)

Extra-Column Effects

The fluid- and solid-phase equations are related to the fixed-bed section where the adsorption or desorption occurs. There are other sections necessary to complete the column, such as the connector, distributor, valves, and pipes. The effect of these sections on the performance of the process (herein called the extra-column effect) is known to be important although the actual separation does not occur within these spaces (Rota and Wankat, 1990; Wankat, 1990).

Let us consider a column with an extra-column volume of V_0 at each end. This volume is the sum of volumes between the switching valve next to the column entrance (exit) and the top (bottom) of the fixed bed. At the beginning of each new step, this volume is filled with a gas mixture from the previous step. When the new step starts, the entering gas has a different composition and temperature than the gas remaining in V_0 and will mix with this gas. To keep the boundary and initial conditions constant in the old and new designs, the gas streams leaving the extra-column volume and entering the column must have the same composition and temperature in both designs.

The two limiting cases of gas flow within the open volume are a plug flow model and a stirred tank model (Rosner, 1986). Since the latter is more conservative, we will assume that the extra-column volumes are mixed well. The exit stream of this well-mixed volume has composition and the temperature as the gas inside the volume. The overall mass balance and energy balance equations for the stirred tank model are

$$V_0\frac{dc_i}{dt}=V_{\text{in}}c_{i,\text{in}}-V_{\text{out}}c_i \tag{11}$$

$$V_0\rho C_{pm}\frac{dT}{dt}=V_{\text{in}}\rho_{\text{in}}\,C_{pg}(T_{\text{in}}-T)-q_{cp} \tag{12}$$

where q_{cp} is the heat conduction to the cap and the surroundings. Since only physical changes (mixing and heat conduction) occur inside the extra-column volume, the continuity equation is

$$V_0\frac{d\rho}{dt}=V_{\text{in}}\rho_{\text{in}}-V_{\text{out}}\rho \tag{13}$$

Equations 11 and 13 plus the assumption of ideal gas give,

$$\frac{dC_i}{d\tau}=\alpha_m\left[C_{i,\text{in}}-\frac{\Gamma}{\Gamma_{\text{in}}}\frac{P_{\text{in}}}{P}C_i\right]+\left[\frac{1}{P}\frac{dP}{d\tau}-\frac{1}{\Gamma}\frac{d\Gamma}{d\tau}\right]C_i \tag{14}$$

where $\alpha_m = V_{\text{in}}\,t_0/V_0$.

The heat-up of the column cap on the regeneration end of the column causes a unique type of extra-column effect associated with the TSA process. This heat-up can be divided broadly into three periods: first, the heat is provided to increase the temperature of the cap and no energy is released to the surroundings; then, heat is released to the surroundings in addition to the heating the cap; after a certain period of time, all heat loss goes to the surroundings and the cap temperature is constant. Such a heat-up process occurs once per cycle since it always accompanies the heating step. When the TSA process cycles faster, the heat-up will occur more frequently.

The heat conduction to and through the cap in the energy balance is represented by q_{cp}. During the process of heat-up, the temperature of the cap's outside surface ($T_{cp,\mathrm{amb}}$) varies. Approximately, $T_{cp} = (T + T_{cp,\mathrm{amb}})/2$ is the average wall temperature. Neglecting the heat of mixing and kinetic energy effects, one obtains the following overall energy balance

$$V_0\rho C_{pm}\frac{dT}{dt} = V_{\mathrm{in}}\rho_{\mathrm{in}}\, C_{pg}(T_{\mathrm{in}} - T) - V_{cp}\rho_{cp}C_{p,cp}\frac{d(T+T_{cp,\mathrm{amb}})/2}{dt} - h_a A_{cp}(T_{cp,\mathrm{amb}} - T_{\mathrm{amb}}) \quad (15)$$

which is

$$\Gamma_{\mathrm{in}} - \Gamma = \left(\alpha_h\frac{P}{\Gamma} + \alpha_h'\right)\frac{d\Gamma}{d\tau} + \alpha_h'\frac{d\Gamma_{cp,\mathrm{amp}}}{d\tau} + \alpha_h''(\Gamma_{cp,\mathrm{amb}} - \Gamma_{\mathrm{amb}}) \quad (16)$$

where the three dimensionless groups are

$$\alpha_h = \frac{V_0 p_0 C_{pm}}{V_{\mathrm{in}}\rho_{\mathrm{in}}\, C_{pg} t_0 R_g T_0},\quad \alpha_h' = \frac{V_{cp}\rho_{cp}C_{p,cp}}{2V_{\mathrm{in}}\rho_{\mathrm{in}}C_{pg}t_0},\quad \alpha_h'' = \frac{h_a A_{cp}}{V_{\mathrm{in}}\rho_{\mathrm{in}}C_{pg}} \quad (17)$$

and the reference time t_0 is (Rota and Wankat, 1990),

$$t_0 = \frac{L}{u_0} = \frac{L}{\dfrac{p_0 d_p^2\,\epsilon}{150\mu L}\left(\dfrac{\epsilon}{1-\epsilon}\right)^2} = \frac{150\mu L^2}{p_0 d_p^2\,\epsilon}\left(\frac{1-\epsilon}{\epsilon}\right)^2 \quad (18)$$

The initial conditions for Γ and $\Gamma_{cp,\mathrm{amb}}$ should be the same in the old design and the new design. Also, one has the following constraints for the three α's:

$$R_{\alpha_h} = R_{\alpha_h'} = R_{\alpha_h'} = 1 \quad (19)$$

which yields (note that $R_{V_{\mathrm{in}}} = R_{N_{FD}}$)

$$R_{V_0} = R_{(V_{cp}\rho_{cp}C_{p,cp})} = R_{(V_{\mathrm{in}}t_0)} = R_{(N_{FD}t_c)},\quad R_{A_{cp}} = R_{N_{FD}} \quad (20)$$

These scaling rules require that the extra-column volume and the total heat capacity of the cap be scaled in the same way as the product of the cycle time and the throughput. The significance of Eq. 20 is discussed next.

Assume that the caps are hemispheres, the same material is used in both designs and the throughput is the same in both designs. Let D_w and ω be the wall outside diameter and the number of cycles in a given time period. The gas temperature T will be the same in both designs since the operating (regeneration) temperature will not change. Assume that the cycles remain long enough to completely heat the wall. If this assumption is not met in the new design, there will be less heat loss in the new design. The energy accumulation per unit amount of product is

$$HAW + HAC = (V_{cp}\rho_{cp}C_{p,cp} + V_w\rho_w C_{pw})(T - T_{\mathrm{amb}})\omega \quad (21)$$

where

$$V_w = \frac{\pi}{4}L(D_w^2 - D^2),\quad V_{cp} = \frac{\pi}{6}(D_w^3 - D^3) \quad (22)$$

For simplicity, assume that $\rho_{cp}C_{p,cp} = \rho_w C_{pw}$. Then, compare the heat accumulation per unit amount of product in the wall (HAW) in the new design with that in the old design.

$$R_{HAW} = \frac{HAW_{\mathrm{new}}}{HAW_{\mathrm{old}}} = \frac{[L(D_w^2 - D^2)\omega]_{\mathrm{new}}}{[L(D_w^2 - D^2)\omega]_{\mathrm{old}}} = \frac{R_L R_D^2 R(4\delta_w^2 + 4\delta_w)}{R_{t_c}} \quad (23)$$

where δ_w is the ratio of wall thickness to column diameter, $\delta_w = 0.5\,(D_w - D)/D$. The results from Table 1 were used for the last equality in Eq. 23. When internal diffusion controls, $R_D = 1$ and $R_{\delta_w} = 1$. Then from Eq. 23, $R_{HAW} = 1$. If the film diffusion is important, $R_D > 1$ when particle diameter is decreased ($R_{d_p} < 1$). Usually this will result in a column with $R_{\delta_w} < 1$ and there will be less heat loss through the column walls in the new design.

The situation is different for the cap. The comparison of the two designs is:

$$R_{HAC} = \frac{(V_{cp}\omega)_{\mathrm{new}}}{(V_{cp}\,\omega)_{\mathrm{old}}} = \frac{[(D_w^3 - D^3)\omega]_{\mathrm{new}}}{[(D_w^3 - D^3)\omega]_{\mathrm{old}}} = \frac{R_D^3}{R_{t_c}}R(8\delta_w^3 + 12\delta_w^2 + 6\delta_w) \quad (24)$$

If internal diffusion controls, $R_D = 1$ and $R_{\delta_w} = 1$; thus, $R_{HAC} = 1/R_{t_c}$. If external film controls mass transfer, then $R_{HAC} = R_{t_0}^{1.669}R(8\delta_w^3 + 12\delta_w^2 + 6\delta_w)$. When particle diameter is decreased $R_{t_c} < 1$, and in both cases $R_{HAC} > 1$. Thus, heat consumption by the cap increases as one intensifies the operation. If this consumption is small, this effect can be neglected; otherwise, internal insulation can be used to isolate the cap from the gas. Internal insulation makes $HAC \sim 0$, and the value of R_{HAC} becomes irrelevant.

The heat loss to the surroundings is proportional to the surface area. The last of Eqs. 20 states that the cap surface area is required to scale according to the throughput. If the cap geometry (e.g., hemispherical) is unchanged, Eq. 20 is satisfied when internal diffusion controls. If internal diffusion does not control the transfer process, a larger cap surface area will be obtained in the new design and thus there is more heat loss through the cap. Note that the wall area of the packed column is reduced in the new design when one has a shorter

column. Therefore, the cap (top) area and the wall (side) area have different effects on heat loss.

In the following, we will show how the total surface area changes as the cycle time changes. For the internal resistance model, the same column diameter is obtained in the new design if the throughput is constant in both designs. The cap surface area does not change, but the column length is reduced. Therefore, total surface area decreases due to the reduction of wall surface area and the energy loss to the surroundings becomes less in the new design than that in the old design. However, for the external film transfer model, the decrease or increase in the total surface area depends on the ratio of column length to column diameter in the old design, $k = (L/D)_{\text{old}}$, and on R_{t_c}. The total surface area for the old design is,

$$\text{total area} = \text{area 2 caps} + \text{area column walls} \qquad (25a)$$

if heat loss and accumulation from both caps are important, or

$$\text{total area} = \text{area 1 cap} + \text{area column walls} \qquad (25b)$$

if heat loss and accumulation from one cap are important. If one assumes that heat recovery is used for the exiting regeneration gas, Eq. 25a is appropriate. From Table 1,

$R_D = R_{t_c}^{-0.223}$ and $R_L = R_{t_c}^{1.426}$; thus,

$$D_{\text{new}} = D_{\text{old}} R_D = D R_{t_c}^{-0.223},\ L_{\text{new}} = L_{\text{old}} R_L = kD R_{t_c}^{1.446} \qquad (26)$$

Then, the ratio of the total area in the new design to that in the old design is

$$\frac{(\text{total area})_{\text{new}}}{(\text{total area})_{\text{old}}} = \frac{\pi D^2 R_{t_c}^{-0.446} + \pi k D^2 R_{t_c}^{1.223}}{\pi D^2 + \pi k D^2} = \frac{R_{t_c}^{-0.446} + k R_{t_c}^{1.223}}{1+k} \qquad (27)$$

As an example, consider a column where $k = 1$ and one wants $R_{\text{total area}} = 1$. Then, $R_{t_c} = 1.0$ or 0.267. Equation 27 is less than 1 for $0.267 < R_{t_c} < 1$ and larger than 1 for $0 < R_{t_c} < 0.267$. This means that the total area in the new design will decrease for $R_{t_c} = (0.267, 1)$ and the heat loss to the surroundings will be reduced in the new design. The total area in the new design will increase for $R_{t_c} = (0, 0.267)$ and there will be some increase in the heat loss to the surroundings. For any k value, Eq. 27 can be used to find the ratio of areas.

The total heat loss (accumulation and heat loss terms) can be estimated for a particular case from Eqs. 21, 22 and 27, and the appropriate heat transfer coefficients. If the column has external insulation, the accumulation terms will usually dominate. For $R_{d_p} < 1$ ($R_{t_c} < 1$), the new design will have more heat loss (Chen, 1990). If this additional heat loss is significant, internal insulation can be used to reduce heat accumulation and loss in the caps.

Discussion

Generally, the outcome of intensification will be a smaller and lighter, and more efficient system that can perform the same task as the larger ones. Size is critical on barge-mounted units and on offshore platforms. A combination of size and weight are critical for systems in airplanes or on the space shuttle. Reducing the inventory of dangerous chemicals is an important safety principle. The intensified TSA systems should be of interest in all these cases.

It is interesting to compare the scaling of TSA systems with that of PSA systems. In both systems the product recovery, product purity, pressure drop, and throughput can be kept constant, while the adsorbent productivity is increased by using a shorter column packed with smaller particles operated with shorter cycles. The energy consumption during regeneration is the main operating cost for TSA processes. Internal insulation should be used to prevent increased heat loss during the regeneration step. In the TSA systems, the time required to heat, regenerate, and a cool a bed is usually in the range of a few hours to several days while the cycle time for PSA lasts only for a few seconds to minutes. Thus, there is considerably more room for improvement with existing TSA processes than with existing PSA processes. With TSA systems the effect of thermal shocks on absorbent life needs to be considered.

There are some limitations on the analysis presented here. The adsorption or desorption kinetics may become important as the mass and heat transfer rates keep increasing (with decreasing particle size), and the scaling rules developed no longer will work in this case. Most TSA systems are probably far from this limitation. Radial dispersion can become important for short, large-diameter columns; thus, the gas distributor and packing technique should be designed carefully. A long life is necessary for the adsorbent. In the new design, more frequent cycles and the resulting thermal cycling of the adsorbent might cause damage to the internal pore and/or crystal structure due to the frequent expansion and contraction. The adsorbent may deactivate more rapidly with short cycles. In addition, the scaling rules are restricted to the same feed and regeneration gas concentrations, temperatures and pressures.

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Notation

A_{cp} = surface area of the cap, m^2
c = mole concentration, mol/m^3
C = dimensionless concentration, c/c_0
C_{pg} = gas heat capacity at T, J/mol·K
C_{pm} = gas heat capacity at T_{in}, J/mol·K
C_{pw} = wall heat capacity, J/kg·K
d_p = particle diameter, m
D = column diameter, m
D_w = column wall outside diameter, m
h_a = heat transfer coefficient, K
k = ratio of column length to diameter, $(L/D)_{\text{old}}$
L = column length, m
N_{Fd} = total feed rate, mol/s
p = pressure, atm
P = dimensionless pressure, p/p_0
q_{cp} = rate of heat transfer between fluid and cap, W/m^3
R = scaling operator, $R_X = R(X) = X_{\text{new}}/X_{\text{old}}$
R_g = ideal gas constant, atm·m^3/mol·K
t = time, s
t_0 = reference time, Eq. 18
T = temperature, K
u_0 = reference velocity, Eq. 18

U = dimensionless interstitial velocity, u/u_0
V_0 = extra-column volume, m^3
V_{cp} = cap solid volume, m^3
V_{in} = volumetric flow rate, m^3/s
V_{out} = volumetric flow rate, m^3/s
V_w = wall volume, m^3
Z = dimensionless column coordinate, z/L

Greek letters

α_h, α_h', α_h'' = dimensionless groups, Eq. 17
α_m = $V_{in}t_0/V_0$
Γ = dimensionless temperature, T/T_0
δ_w = $(D_w-D)/(2D)$
ϵ = fractional void space available for flow
μ = viscosity, kg/m·s
ρ, ρ_w = density, mol/m^3(gas) or kg/m^3(solid)
τ = dimensionless time, t/t_0
ω = number of cycles

Subscripts

amb = ambient
c = cycle
cp = cap
CL = cooling
f = fluid
FD = feed
HT = heating
i = ith component
i, f = ith component of feed
i, h = ith component of hot purging gas
in = inlet
$INIT$ = initial state
new = new design
old = old design
out = outlet
I, II, III = end of the first, second, and third step of cycle

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