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INCREASING THE EFFICIENCY OF A TEMPERATURE SWING ADSORPTION (TSA) NATURAL GAS DEHYDRATION PLANT

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Abstract

Natural gas is a naturally occurring hydrocarbon gas mixture consisting primarily of methane; but, commonly including small percentage of carbon dioxide, nitrogen, mercaptanes and hydro-gen sulfide. Water causes several problems like corrosion, condensation and decreasing natural gas energy content. Temperature swing adsorption (TSA) is one of the widely used processes in gas plants to remove water from the feedstock, and hence decreasing the dew point of natural gas. In this study, the fixed-bed adsorption of water in a gas dehydration plant on zeolite 3A was investigated. At first, a rigorous modeling methodology is introduced to calculate the breakthrough time of the industrial TSA process using 3A molecular sieve, and then it is applied to study the cycle time of the adsorption process. Based on results obtained by the model, the cycle time for designed and actual conditions are about 12.3 h and 19.5 h, respectively which are close enough to the observed values in the target dehydration plant. Then, to increase the cycle time, a new 3A molecular sieve is proposed. Modeling results show that after replacing existing molecular sieve with the recommended one, breakthrough time increases about 20.8 h, higher enough than the desired value (20 h).

Keywords: Temperature swing adsorption; Molecular sieve 3A; Natural gas dehydration; Adsorption Model.

1. Introduction

The dehydration of natural gas is momentous to prevent problems usually caused by corrosion and hydrate formation. The natural gas industry has recognized that dehydration is necessary to ensure smooth operation of gas transmission lines [1]. One of the methods to do such a task is adsorption process using a suitable zeolite molecular sieve. Zeolites are highly crystalline hydrated alumina silicates from the alkali metals and alkali earth metals groups [2]. Their structural arrangement presents sockets and interconnected channels in which compensation ions, such as Na^+, Ca^{2+}, Mg^{2+}, K^+ and H_2O are present. This microporous structure enables zeolites to have a large surface area in relation to the external one. Applying this approach is common to many industrial processes, such as pipelines, natural gas processing plants, liquefied natural gas (LNG) regasification, and power plants in which natural gas is a feedstock. These systems normally consist of multiple adsorption columns loaded with spherical or cylindrical solid particles. The columns are typically switched between adsorption, regeneration, and cooling.

There are two widely used approaches to estimate the breakthrough time of adsorption processes i.e., experimentation and mathematical modeling. The first method provides a direct and fast answer for a given system; but, it greatly depends upon the experimental conditions, such as ambient temperature and residence time [3]. In contrast, mathematical modeling is simple and easily understood with no experimental set-up required, and there-fore, it has absorbed increasing interest in the industrial applications. With extensive industry applications of adsorption processes, there is significant interest for efficient modeling, simulation and optimization strategies [4]. Due to the complexity of rigorous modeling and simulation of adsorption processes, an accurate and reliable approach to design industrial plants is essentially needed [5].

Up to now, many mathematical models have been presented to describe and calculate the breakthrough time of a column adsorption system in liquid or gaseous phase [6-13]. The main objective of this work is to introduce a simple and accurate method to estimate the breakthrough time of an industrial temperature swing adsorber (TSA) process. According to the mathematical models, the breakthrough time of the target plant is calculated, and it is compared with the actual value. Then, a new adsorbent is recommended for the target TSA plant, and the improvement in the efficiency of the plant is studied.

2. Process description

2.1. Feed and regeneration gas

The target adsorption unit consists of 3 beds (two beds in adsorption mode and one bed in regeneration mode) for continuous operation. Unit is designed to adjust water dew point of sweet natural gas stream before feeding cryogenic ethane recovery facilities. Specification and composition of feed stream are presented in Table 1 and 2, respectively. It can be found that summer case is the worst case due to the higher feed flow rate and consequently higher water loading per beds. For both cases, the specification of the regeneration gas is presented in Table 3.

Table 1. Operating conditions of feed in summer and winter

Variables	Unit	Summer Case	Winter Case
Temperature	°C	49	49
Pressure	barg	74.56	74.56
Volume flow rate	MMSCMD	9.85	9.74
Total mass flow (Wet basis)	kg/h	332327.8	320052.2
Mass flow per bed (Wet basis)	kg/h	166163.9	160026.1
Molecular weight	kg/kmol	19.14	18.64
Mass density	kg/m^3	64.04	61.73
Liquid fraction	-	0	0
Z factor	-	0.84	0.85

Table 2. Composition of feed stream in summer and winter case

component	value (mol. %)		component	value (mol. %)	
	Summer case	Winter case		Summer case	Winter case
N_2	0.13	0.14	n-Octane	0.07	0.10
H_2S	0.0003	0.0003	n-Nonane	0.02	0
CO_2	0.0250	0.0250	M-Mercaptan	0.1551	0.1408
Methane	88.16	89.14	E-Mercaptan	0.0035	0.0028
Ethane	6.03	5.96	2C3Mercaptan	0.0001	0.0001
Propane	2.60	2.48	1Hexanethiol	0.0002	0
i-Butane	0.52	0.47	COS	0.0027	0.0025
n-Butane	0.94	0.66	M-E-Sulfide	0.0001	0.0001
i-Pentane	0.31	0.23	diM-Sulfide	0.0191	0.0137
n-Pentane	0.31	0.22	diMdi-Sulfide	0.0001	0
n-Hexane	0.28	0.14	Water	0.2	0.2
n-Heptane	0.22	0.06			

Table 3. Specification of the regeneration gas

Specification	Unit	Value	
Temperature	^0C	Heating	Cooling
		287.80	53.80
Pressure	Bar	31.64	
Flow Rate	kg/hr	40,145.00	
Component		**Value (%mol)**	
Methane		99.06	
N_2		0.15	
CO_2		0.0183	
Ethane		0.74	
Propane		0.03	
COS		0.0001	

2.2. Bed configuration

The following beds schematic shows existing bed layer configuration of adsorbents for natural gas dehydration:

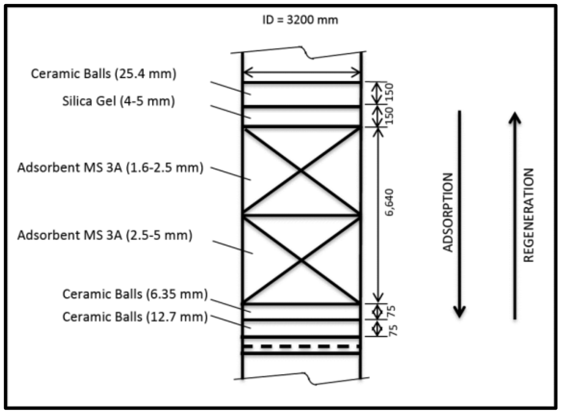

Figure 1. Bed configuration of the target drying plant

2.3. Specifications of the adsorbent (3A molecular sieve)

The specifications of adsorbent for the current case (before improving the TSA process) is presented in Table 4.

Table 4. Specification of the current case adsorbent

Specification		Unit	Value
Shape		--	Spherical
Size, min. 98%		mm	3-5
Bulk Density		g/cm^3	0.65
Equilibrium Water Adsorption Capacity (25 ºC / 24h) max.	10% RH	mass%	5
	50% RH		17
	60% RH		20
Crushing Strength, min.		N	70
Package Moisture Content, 3 hour at 230 ºC, max.		mass%	2.5
Attrition Loss, max.		mass%	0.2
pH		--	9-10.5
Specific Heat, max.		kJ/kg K	1.07
CO_2 Adsorption, max.		mass%	2
N Content, max.		mass%	0.1
C Content, max.		mass%	0.71
Residual Water Content, 3 hour at 550 ºC, max.		mass%	1.5

This molecular sieve (3A) has a pore size 3 angstrom. Any molecule larger than that cannot be adsorbed. It is an alkali metal aluminosilicate, the potassium form of the type-A crystal structure. The required pore size is created when part of the sodium ions of the 4A molecular sieve are replaced by potassium ions. 3A molecular sieve is mainly used for removing moisture from liquefied and gaseous materials.

3. Mathematical modeling

3.1. Theoretical concept

Adsorption is a transient process and the amount of material adsorbed within a bed depends both on position and time. As fluid enters the bed, it comes in contact with the first few layers of absorbent. Solute adsorbs, filling up some of the available sites (see figure 1). Soon, the adsorbent near the entrance is saturated and the fluid penetrates farther into the bed before all solute is removed, thus the active region shifts down through the bed as time goes on. The fluid emerging from the bed will have little or no solute remaining, at least until the bulk of the bed becomes saturated. The break point occurs when the concentration of the fluid leaving the bed spikes as unabsorbed solute begins to emerge. The bed has become ineffective. Usually, a breakpoint composition is set to be the maximum amount of solute that can be acceptably lost, typically something between 1 and 5 percent. As the concentration wave moves through the bed, most of the mass transfer is occurring in a fairly small region. This mass transfer zone moves down the bed until it "breaks through".

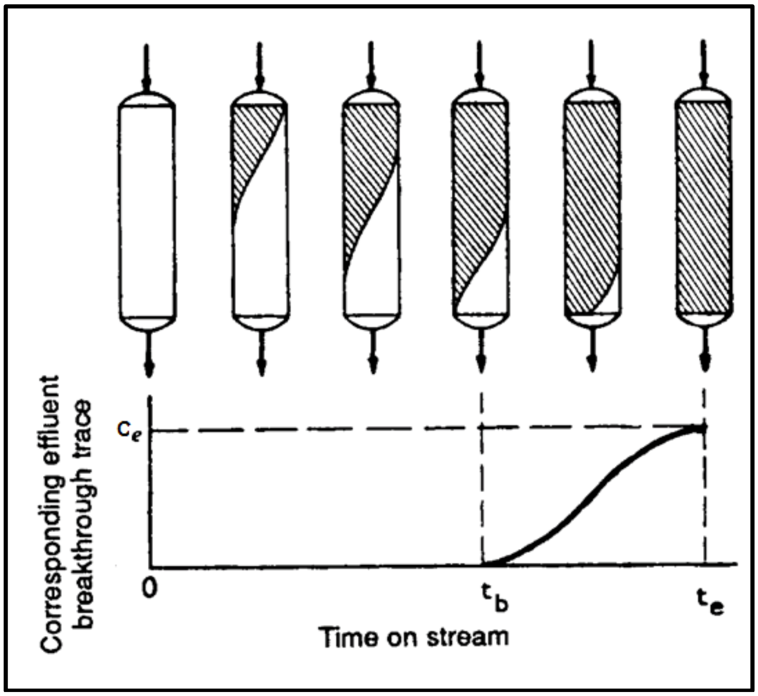

Figure 2. Break through curve of the adsorption bed [14]

The shape of the mass transfer zone depends on the adsorption isotherm (equilibrium expression), flow rate, and the diffusion characteristics. Usually, the shape must be determined experimentally.

Capacity calculations are made based on plots of the composition vs. time (usually near the exit of the bed). Curves are integrated (analytically, numerically, or graphically) to obtain capacities (measured in time units, or how long a bed can run).

The time required for a bed to become totally saturated is obtained by integrating as time goes to infinity:

$$t_{Total} = \int_0^{\infty} \left(1 - {}^{C}\!/_{C_F}\right) dt \qquad (1)$$

where: C is the concentration of gas phase and C_F is the feed concentration. So, to calculate the breakthrough time, the variation of concentration of solute or adsorbate (i.e., water for natural gas dehydration) versus time is needed. Therefore, a mathematical model should be developed to do such a task.

3.2. Model development

The following assumption is used to model the target TSA unit:

- The gas phase is ideal, and is only consisted of methane as the dominant component.
- The bed works in adiabatic and isothermal conditions without any heat transfer between solid and gas phase.
- The bed void is initially filled with methane.
- Only axial mass dispersion is assumed.
- Pressure gradient is related to superficial velocity based on Ergun equation.
- Mass transfer coefficients consist of film resistance and macro pore diffusion coefficient.

Above assumption was widely accepted by numerous adsorption studies [15-17]. Based on these assumptions, the general and particular equations are as follows.

3.2.1. General equations

3.2.1.1. Momentum balance

In this study, bed dimensions and particle diameter are constant; therefore, based on Ergun's equation, the superficial velocity can be related to the total pressure gradient as follows [18]:

$$\frac{\partial p}{\partial z} = -\left(\frac{1.5\times10^{-3}\,(1-\varepsilon)^2}{\left(2r_p\psi\right)^2\,\varepsilon^3}\mu\upsilon_g + 1.75\times10^{-5} M\rho_g \frac{(1-\varepsilon)}{2r_p\psi\,\varepsilon^3}{\upsilon_g}^2\right) \qquad (2)$$

Ergun equation considers laminar and turbulent flow conditions to calculate the pressure drop across the bed.

3.2.1.2 Material balance

The mass balance in the gas phase depends on the effect of axial dispersion, convection term, gas phase accumulation and rate of fluid to the adsorbent as the following [20]:

$$-\varepsilon D_{ax}\frac{\partial^2 c_i}{\partial z^2} + \upsilon_g\frac{\partial(c_i)}{\partial z} + \frac{\partial c_i}{\partial t} + \frac{1-\varepsilon}{\varepsilon}\frac{\partial\overline{q_\imath}}{\partial t} = 0 \qquad (3)$$

The dispersion coefficient in Eq.2 is calculated from the following correlation [19]:

$$D_{ax} = 0.73D_{mi} + \frac{\upsilon_g r_p}{\varepsilon\left(1+9.49\frac{\varepsilon D_{mi}}{2\upsilon_g r_p}\right)} \qquad (4)$$

Moreover, to calculate binary molecular diffusivity (D_{AB}), Fuller, Schettler and Giddings equation is used. This equation includes empirical constants and keeps the form of Chapman- Enskog kinetic theory.

$$D_{AB} = \frac{0.00143T^{1.75}}{PM_{AB}^{1/2}\left[(\Sigma_v)_A^{1/3}+(\Sigma_v)_B^{1/3}\right]^2} \qquad (5)$$

3.2.2 Particular equations

3.2.2.1 Kinetic model

Mass transfer driving force is assumed to be a linear function of solid phase loading with transport mechanism from fluid to solid. Therefore, it consists of two terms as follows [14-19]:

- Extra-particle transport mechanisms
- Intra-particle transport mechanisms

The overall mass transfer coefficient can be calculated as follows:

$$\frac{1}{k_{MTC_i}} = \frac{r_p \bar{K}_{ki}}{3k_{fi}} + \frac{r_p^2 \bar{K}_{ki}}{15\varepsilon_p D_{pi}} \tag{6}$$

In extra-particle transport term the film resistance is estimated from Sherwood number and Wakao-Funazkri correlation as the following [14-19]:

$$k_{fi} = sh_i \frac{D_{mi}}{2r_p} \tag{7}$$

$$\mathrm{sh_i} = 2.0 + 1.1\mathrm{Sc_i^{1/3}}\mathrm{Re}^{0.6} \tag{8}$$

Above correlation is reliable in the Reynolds number between 3 and 10^4 [8].

For the intra-particle transport term, the macro pore diffusion for molecular sieve has both molecular and Knudsen diffusions obtained from the Bosanquet equation as follows:

$$\frac{1}{D_{pi}} = \tau\left(\frac{1}{D_{ki}} + \frac{1}{D_{mi}}\right) \tag{9}$$

$$\mathrm{D_{ki}} = 97\mathrm{r_{pore}}\left(\frac{\mathrm{T}}{\mathrm{M_i}}\right)^{0.5} \tag{10}$$

where τ is the tortuosity factor and is dependent to the porosity as follows [20]:

$$\tau = \varepsilon + 1.5(1 - \epsilon) \tag{11}$$

3.2.2.2 Isotherm model

The relation between loading of molecular sieve and the partial pressure or concentration of adsorbate is known as isotherm curve. The Langmuir-type isotherm is the most relevant model for practical applications. In this research, the Langmuir isotherm for pure component adsorption is used. Details for estimating the isotherm parameters were described in the previous work [7].

3.3 Numerical solution

All of the mentioned general and particular equations are solved using the Matlab software (Mathworks, 2013a) with appropriate initial and boundary conditions to estimate the breakthrough time of the target TSA column.

4. Results and discussions

4.1 Monitoring the performance of the target dehydration plant

Figure 3 shows water content against time for the 1st sample connection (top) of drying vessel. As it is observed, water content has increased after 5 hours (300 minutes) gradually and breakthrough has been occurred. So, it can be concluded that this part of bed has almost been saturated. High water content at start of measurement is due to the initialization of hygrometer sensor.

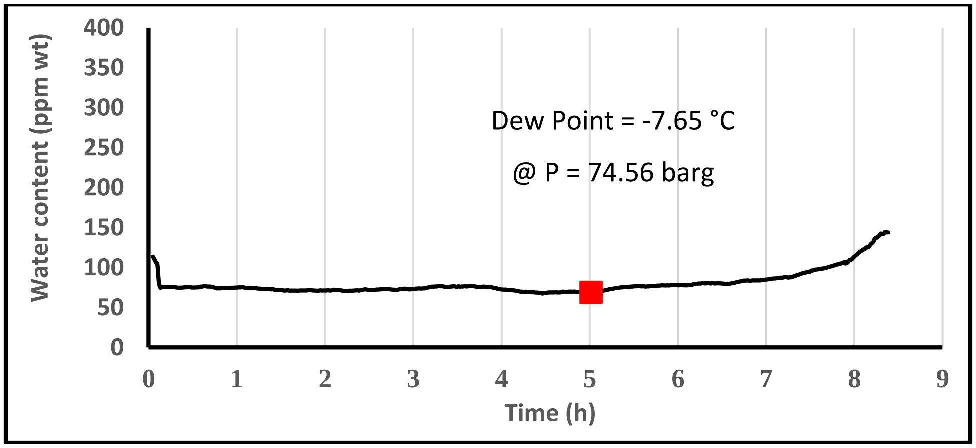

Figure 3. Water content vs. time for 1st sample connection of drying vessel

Figure 4 illustrates water content vs. operation time for 2nd sample connection of vessel A (bottom). As shown in Fig.5, no breakthrough occurs in this sample connection after 12 h from start of operation. As it is obvious, the water content has decreased gradually, and finally it is flatted. It means that breakthrough has not occurred yet during this cycle. It should be noted that the flowrate of the feed is about 40% less than the design value. So, it is concluded that a longer adsorption cycle can be expected due to the higher designed adsorption capacity of the dryer. According to the requirement of the target gas refinery, the dryer should operate at least for 20 h before facing any breakthrough conditions.

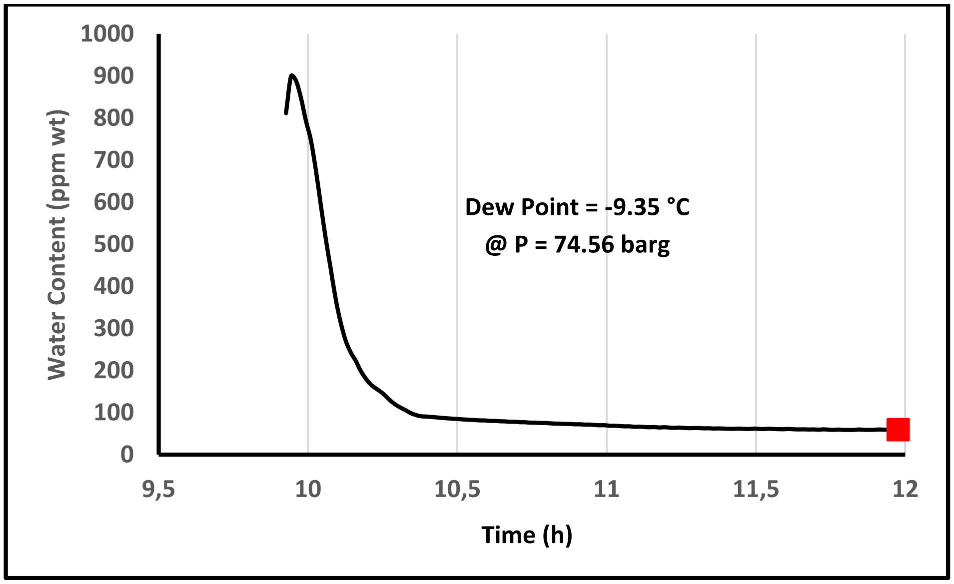

Figure 4. Water content versus time for 2nd sample connection of drying vessel

4.2 Improving the performance of the target dehydration plant

The following operating variables for design case (molecular sieve specifications are presented in Table 4) are calculated based on the modeling methodology introduced in section 3. The static water adsorption in 60% relative humidity is assumed to 22wt%. Accordingly, it is found that the breakthrough time can be expected after 12.3 h.

Table 5. Calculated parameters for the design case

Parameter	Unit	Value
Water loading	kg/h.m^2	40.5
Superficial velocity	m/min	5.3
MTZ length	cm	185.5
Length of unused bed	m	0.151
Breakthrough time	h	12.3 (Note)

In Table 6, the corresponding variables are calculated for the current operating conditions. As mentioned before, in this case, the flowrate of the feed is about 40% lower than the design case. Therefore, the considerable gap between the design (desired values) and the current cases can be justified, and the validity of the model can be concluded.

Table 6. Calculated parameters for the current case

Parameter	Unit	Value
Water loading	kg/h.m^2	24.5
Superficial velocity	m/min	3.21
MTZ length	cm	164.5
Length of unused bed	m	1.35
Breakthrough time	h	19.5

From these results, it is concluded that the target dehydration plant cannot operate more than 20h without facing breakthrough which is essentially required by the target gas refinery. So, to improve the efficiency of the plant, a special type synthetic zeolite of A-type crystal structure in potassium-sodium form with a pore opening of 3 angstrom (0.3 nm) is suggested to replace with the current one. Specifications of the proposed molecular sieve are summarized in Table 7.

Table 7. Specification of the proposed adsorbent to improve the efficiency

Specification		Unit	Value
Shape		--	Spherical
Diameter		mm	3.0 – 5.0
Bulk density		g/L	≥700
Water static adsorption (@25 ºC)	10% RH	wt %	≥17.5
	80% RH		≥21.5
Crushing strength		N	≥70
Attrition		wt %	≤0.1
Moisture content @550 ºC		wt %	≤0.1

In Table 8, the significant variables are recalculated for the proposed molecular sieve based on current operating conditions. It can be concluded that after molecular sieve replacement, breakthrough is happened after 20.8 h which is higher than the required value. But, to meet any operating upsets, a shorter operating cycle should be considered, and therefore the suggested adsorption cycle (about 20 h) is suggested.

Table 8. Calculated parameters for the proposed molecular sieve

Specification	Unit	Value
Water loading	$kg/h.m^2$	24.5
Superficial velocity	m/min	3.21
MTZ length	cm	164.5
Length of unused bed	m	1.95
Breakthrough time	hs.	20.8

5. Conclusions

To design and optimize temperature swing adsorber (TSA) plants, it is essential to develop a simple and accurate rigorous mathematical model. In this study, a method to estimate the breakthrough time of a commercial TSA process, designed to dehydrate natural gas, in consequence with decreasing the product dew point pure was presented.

At first, collected data from the top of the adsorbing vessel showed that water content was gradually increased after 5 hours, and breakthrough phenomenon was occurred. In contrast, observations from the bottom of vessel (water content vs. operation time) confirmed that no breakthrough occurs in this section after 12 h from the start of operation. Because the actual flowrate of the feed was about 40% lower than the nominal value, a longer adsorption cycle could be achievable. Based on the presented modeling approach, the expected cycle time for nominal and actual conditions were about 12.3 h and 19.5 h, respectively. The former was about the same as the observed value (about 12 h), reported by the engineering department of the target plant; therefore, the difference between the nominal and actual cycle times can be justified, and the validity of the model was concluded.

To enhance the cycle time of the gas dehydration plant, and improve the operation, it was required to increase the cycle time to more than 20 h with enough safe margin. Consequently, a special type of 3A molecular sieve (a-type crystal structure in potassium-sodium) was proposed. Based on the results obtained by the introduced model, it was concluded that after replacing molecular sieve, breakthrough was happened after 20.8 h, confidently higher than the desired value. However, to meet any operating upsets, a shorter operating cycle should be considered, and therefore the cycle time of 20 h was suggested.

6. Acknowledgment

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Nomenclature (Units)

C	*Concentration of adsorbate (mol/m^3)*	ρ_g	*Gas density (kg/m^3)*
C_o	*Concentration of adsorbate in effluent at equilibrium (mol/m^3)*	D_{ax}	*Axial dispersion coefficient (cm^2/s)*
C_F	*Feed concentration (mol/m^3)*	ρ_b	*Solid bulk density (kg/m^3)*
P	*Pressure (atm)*	k_{MTC_i}	*Mass transfer coefficient (1/s)*
T	*Temperature (K)*	k_{fi}	*Film resistance coefficient (1/s)*
t	*time (h)*	ε_p	*Particle porosity*
t_b	*Starting of breakthrough time (h)*	D_{pi}	*Macro pore diffusion coefficient (cm^2/s)*
t_e	*End of cycle (h)*	sh	*Sherwood number*
t_{Total}	*Total cycle time (h)*	D_{mi}	*Gas mixtures molecular diffusion (cm^2/s)*
Σ_v	*Summation of atomic diffusion Volume*	Sc	*Schmidt number*
ε	*Bed void fraction*	Re	*Reynolds number*
r_p	*Particle radius (m)*	τ	*Tortuosity factor*
ψ	*Shape factor*	D_{ki}	*Knudsen diffusion (cm^2/s)*
μ	*Dynamic viscosity (Pa.s)*	q_i	*Loading of component i (cc/gr)*
v_g	*Superficial velocity (m/s)*	IP	*Isotherm parameter*
M	*Molecular weight*	p_i	*Partial pressure of component i*

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