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Modeling of Adsorption in a Packed Bed Tower, the Case Study of Methane Removal and Parametric Calculation

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Abstract

In this work, the modeling of methane adsorption in a tower with fixed bed has been studied. In order to present a mathematical model, the mass balance was written in the tower. The obtained equations with the assumption of no changes in the concentration, temperature, and pressure in the radial direction as well as the axial dispersion of the flow pattern were solved by using numerical methods. Among various numerical methods, an implicit finite difference method was used to solve the equations. Base on the obtained model, the effect of temperature, inlet flow rate, bed length, and pressure on the adsorption tower was investigated. It was observed that with the temperature decreases, the adsorption rate increases. At a specified time, the amount of adsorbate in the gas phase at the outlet of the bed from 306 to 295 decreased by changing the temperature from T=298K to T=308K. also, the effect of pressure, gas velocity, adsorbent size and bed length in separate diagrams was studied and it was determined that with increasing pressure, decreasing gas velocity, increasing bed length and decreasing adsorbent size and adsorption rate increase.

Keywords: Adsorption, Modeling, Methane, Packed bed tower, fixed bed.

1 Introduction

Research on the prevention of air pollution and the environment has increased dramatically over the last few decades, due to increasing social and economic concerns about the environment. As a result environmental organizations have imposed restrictive laws to reduce the emission of pollutants into the atmosphere. Methane is a gaseous pollutant that causes global warming. The main sources of methane from oil and gas production facilities, agricultural activities, such as animal husbandry and meat production. The challenge of removing methane from industrial waste gases is because waste streams are produced at low pressures and have very low concentrations of methane. For example, the release of methane from a coal mine is typically less than 1.5% of methane [1]. The most expensive and commonly used current technique is to remove low concentration of methane from a low-pressure gas flow, return thermal oxidation processes, and catalytic thermal oxidation that converts methane into carbon dioxide and water. Although some oxidation processes have the potential to recover heat, generally these processes typically incur additional costs for production. Therefore, there is a clear incentive to create a methane recovery process that returns methane to the mainstream gas or recycled methane to generate power, which both reduces operational costs and improves environmental performance [2]. Adsorption based processes include swing adsorption (PSA), technologies that have the potential for recovery from methane from released. Flow and methane enrichment to concentrations that can be used in lean- gas turbines and fuel cells [3-7].

Several studies have been carried out on the separation of various materials by adsorption in a fixed bed. In most of these studies, laboratory work and simulations are carried out simultaneously. For modeling a process, it is necessary to simplify the equations and relations governing that process. Given the fact that these equations and simplified relationships can be used to predict the results of a process and thereby improve its performance, the importance of modeling becomes evident.

The adsorption of methane on zeolite 13x has been studied by Alireza Eslami et al. In this study, the adsorption process was investigated at 298, 308, 323 K and at 1 to 5 MPa pressures by Langmuir, Unilan, Sip and Toth isotherms using a genetic algorithm. The results show that the isotherm model of Toth adsorption at 308 K has a very good flexibility to fit experimental data [8]. Delgado et al, also investigated the adsorption of methane and nitrogen on a silica tablet in a fixed bed. The model was used to simulate nitrogen and methane adsorption curves and pressure cycling was proposed to increase methane content from a mixture of methane/nitrogen (85/15) [6].

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Grand et al. examined the adsorption equilibrium of propane and propylene on a honeycomb rock that was blended with 4A zeolite crystals and inert materials in conditions of 423 and 473 K and in the range of 0 to 100 KPa [9]. Ali Akbar Farzaneh et al. reviewed the simulation of absorption towers. In order to solve the model, the dynamic data provided by San and Monire was used and in the equilibrium, the results were compared with the results obtained from the leading approximation method in a non-equilibrium mode with the results obtained from the QUDS method obtained by San and Munire [10]. Khalid Opportunity and colleagues explored the modeling of the torpedo tower used in the sulfiran process. At first, the required experimental data were collected in a laboratory system under different conditions of H2S concentration and gas flow. After performing the desired mass balance, the developed equations were solved using numerical methods, and the results of the modeling were compared with the experimental results. Comparison of modeling and experimental results indicated a sufficient accuracy of the model to predict the behavior of the tower that was used in the sulfiran process [11]. Studies by Shafiyan and his colleagues on absorption modeling in a tower filled up [12-22]. Due to the complexity of the models that have been proposed for surface uptake so far, it seems necessary to provide another model that is compatible with the previous models. Therefore, by studying the different models presented for such systems, considering the conditions of the system under study, suitable model assumptions such as no changes in concentration, temperature and pressure in the radial direction, as well as the axial dispersion of the constant temperature flow pattern have been selected. And the simulation in the MATLAB program is based on it. Finally, the effect of different parameters on the absorption rate and the conformance of this model with the experimental data presented earlier will be investigated.

2 Modeling

The mathematical models used to dynamically simulate the behavior of an adsorption system based on equations. The most important issue in simulating these processes is that during the recovery stage, usually the adsorbent is not completely recovered, and therefore the initial concentration of the absorbed phase will not be zero. In practical systems, there is usually a region at the entrance of the bed in both the adsorption and disintegration stages, where the unpublished material is present. This complexity causes problems in mathematical simulation because the initial distribution of absorbed concentration is not known in advance. In this case, the concentrations remain constant at all points and all times in the cycle and in all parts of the cycle. The number of cycles required to achieve a uniform cyclic state depends on the various parameters of the system, and it may take up to 30 cycles, such calculations are possible only by numerical simulations, and analytical solutions cannot be used to calculate precisely Specifications of operational systems [23, 24]. The performance of a good absorbent is measured in real operating conditions. This can be done experimentally in industrial or semi-industrial conditions, but since this is very costly and time-consuming, using a suitable model can be a good alternative to reducing costs. Hence, in this section, adsorption fixed bed is modeled on the mechanism of diffusion.

1.2 Modeling of adsorption in the fixed bed

The purpose of this study is to find a suitable model for adsorption of methane in the tower. In order to provide the mathematical model of the adsorption tower, we must establish a mass balance for the tower and write the corresponding equations. The exact modeling is necessary for the proper understanding of the physics of the system as well as the phenomena in the process under study. To accomplish this, we consider the differential height of the filled column and write the mass equations for this differential component. The proposed differential equations should be solved to simulate a tower with a specific height and profile.

In an adsorption fixed bed, the gas content of the adsorbate material enters the bed on one side and leaves it out. By passing on the adsorbent solid absorbed and thus the concentration of the absorbing material in the exhaust gases is reduced. With time and saturation of the primary portions of the substrate, the concentration of the absorbing material in the outlet increases and eventually reaches the concentration of the input.In order to mathematical modeling of the adsorption bed, appropriate element of the substrate has been selected. The following figure 1 shows an element of the fixed bed.

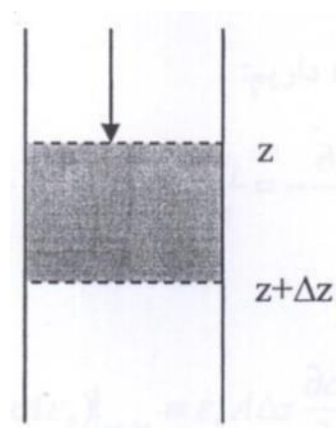


Fig. 1: The element is considered in balance

2.2 Mass balance for each component

The assumptions used in the mass balance are as follows: (a) The thermodynamic behavior of gas is an equation of ideal gas. (b) Changes in Concentrations, temperatures, and pressures in radial and angular directions are not neglected. (c) The mass transfer between the gas phase and the solid phase follows the LDF linear propulsion model. (d) The axial dispersion is assumed to be the flow pattern. (e) It is assumed that the plug flow is negligible with variations in velocity along the bed. With these assumptions, the mass balance equation for component I is written as follows:

Write equations 1:

$$(c_i u A \varepsilon_b) I_z - (c_i u A \varepsilon_b) I_{z+\Delta z} + \left(-D_{ax,i} \frac{\partial c_i}{\partial z} A \varepsilon_b \right) I_z - \left(-D_{ax,i} \frac{\partial c_i}{\partial z} A \varepsilon_b \right) I_{z+\Delta z} = \varepsilon_b A \Delta z \frac{\partial c_i}{\partial z} + (1-\varepsilon_b) A \Delta z \frac{\partial q_i}{\partial t} \quad \text{(Eq. 1)}$$

where εb is substrate porosity, ρp is density of particles (kg/m^3), C_i is concentration of i in the fluid phase (mol/m^3), qi is concentration of i in phosgate (mol/kg), A is cross section of tower (m^2), u is fluid velocity (m/s) and $D_{ax, i}$ is dispersion coefficient (m^2/s). By dividing the sides by the size of the element (AΔz) we have (equation 2):

$$\frac{\partial c_i}{\partial t} + \frac{(1-\varepsilon_b)}{\varepsilon_b}\rho_p \frac{\partial q_i}{\partial t} = -\frac{\partial (uc_i)}{\partial z} + \mathrm{Dax,}\frac{\partial^2 c_i}{\partial z^2}\mathrm{i} \qquad (\mathrm{Eq.\,2})$$

2.3 Pressure drop

Due to the diameter/longitudinal ratio of the substrate studied in this modeling, the pressure variations in the bed are neglected and constant total pressure is considered.

2.4 Equilibrium relationship between two phase concentrations

Due to the type of adsorbent used, which is zeolite and adsorption in zeolites, which is usually single-layered, the Langmuir model is a suitable model for use in simulating this process.

2.5 Mass transfer coefficient between phases

The mass transfer velocity between the solid phase and the gas can be expressed by the approximate linear driving force model. According to the available literature, this model is suitable for long-term adsorption cycles. The most important issue in using this model is to calculate the mass transfer coefficient (K_LDF). The mass transfer coefficient of the film around the adsorbent is obtained according to the following equation 3 [23]:

$$K_{LDF} = \frac{D_M}{2R_p} + \left[2 + 1.1\left(\frac{2uR_p}{\mu}\right)^{0.6}\left(\frac{\nu}{D_M}\right)^{\frac{1}{3}}\right] \qquad (Eq.3)$$

2.6 Boundary and initial conditions and model calculations

The input boundary condition of the adsorption step is, in fact, the input feed condition. Given that the differential equations are two-dimensional relative to the place variable, there is a need for another boundary condition. This condition is provided by assuming zero flux in the output. For an initial condition, it requires a basic assumption. This assumption is assumed to be taken into account when the qi adsorption value is considered to be zero, and the initial concentration of the gas in the bed is also considered to be free from the adsorbate material (the concentration of the adsorbate components is equal to Zero; $t = 0$: $C_{i=}C_i|_{\text{adsorbent free}}$.

In Table 1, the boundary and initial conditions are required. The boundary and initial conditions of the general mass balance are also similar to the partial balance.

Table 1. Boundary and initial conditions

$C_i = C_{i,f}$	$z = 0$	Boundary conditions	Adsorption step
$\frac{\partial c_i}{\partial z} = 0$	$z = L$		
$C_i = 0$	$t = 0$	Initial condition	

2.6.1The numerical form of the equations

Solving the equations of the model involves simultaneous solving NC of the differential equation with partial derivatives of mass balance, NC is the ordinary differential equation of mass transfer between the gas phase and the fluid phase. It is very difficult and even impossible to solve these equations by an analytical method. For this reason, numerical methods are used to solve these equations. Of the various numerical methods, the explicit finite difference method, given the long time of the cycle stages, and the limitation in this method for the length of the time interval, do not seem to be a suitable method for the solution. But in the implicit finite difference method, this limitation does not exist and can be used to solve equations. Based on this method, the numerical form of the terms of the differential equations is as follows:

$$\frac{\partial X}{\partial t} \approx \frac{X_m^j - X_m^{j-1}}{\Delta t} \qquad (Eq.4)$$

$$\frac{\partial X}{\partial z} \approx \frac{X_m^j - X_{m-1}^j}{\Delta z} \qquad (Eq.5)$$

$$\frac{\partial^2 X}{\partial z^2} \approx \frac{X_{m+1}^j - 2X_m^j + X_{m-1}^j}{(\Delta z)^2} \qquad (Eq.6)$$

2.7 The numerical form of the equations in the interior of the substrate

By establishing the partial mass survival law for the detachable component, the following equation is obtained to express the variation in the concentration of the desired component in relative to time and place (equation 4).

$$\frac{\partial c_i}{\partial t} + \frac{(1-\varepsilon_b)}{\varepsilon_b}\rho_p \frac{\partial q_i}{\partial t} = -\frac{\partial (uc_i)}{\partial z} + D_{axi,}\frac{\partial^2 c_i}{\partial z^2} \qquad (Eq.7)$$

By applying these alternatives we have the following in mass balance equation 5:

$$\frac{C_m^j - C_m^{j-1}}{\Delta t} + \left(\frac{1-\varepsilon}{\varepsilon}\right)\left(\frac{q_m^j - q_m^{j-1}}{\Delta t}\right) + \rho\frac{C_m^j - C_{m-1}^j}{\Delta z} = D_{ax}\frac{C_{m+1}^j - 2C_m^j + C_{m-1}^j}{(\Delta z)^2} \qquad (Eq.8)$$

The above equation is solved by the method x = g (x). So we have (equation 6):

$$C_m^j = \frac{(D\frac{\Delta t}{\Delta z^2})C_{m+1}^j + (D\frac{\Delta t}{\Delta z^2} + u\frac{\Delta t}{\Delta z})C_{m-1}^j + C_m^{j-1} - (\frac{\varepsilon_b - 1}{\varepsilon_b})}{2D\frac{\Delta t}{\Delta z^2} + u\frac{\Delta t}{\Delta z} + 1 - \left(\frac{\varepsilon_b - 1}{\varepsilon_b}\right)\frac{q_m^j}{C_m^j}}$$

$(Eq.9)$

2.7.1 The process of solving equations in the isothermal state

In isothermal conditions, the temperature is assumed to be constant at different stages, and various parameters are determined at this constant temperature. Therefore, only the mass balance equations and mass transfer velocity equations will remain. In this case, there is a partial mass balance equation and a mass transfer velocity equation for adsorbate mass and a general mass balance. If we consider the number of divisions over the length of NL, the equations must be solved simultaneously, taking into account the boundary condition NL-1. In the isothermal mode, the form of all relationships related to temperature and energy balance is eliminated, so the convergence time is reduced.

2.8 Describe the computer program

To solve the equations and calculate the values of different variables, the computer program is written in the Matlab 7software environment.

2.8.1 Program capabilities

The provided program has the following capabilities: (1) Calculate and plot the concentration profile of various components along the tower. (2) Calculate the time variation of the concentration of different components at each point of the tower. (3) Investigating the impact of the tower's operational parameters.

2.8.2 Input and output of the program

1. Fixed parameters, including tower and adsorbent specification
2. Operating parameters such as tower's function time
3. Input feed conditions (temperature, pressure, flow intensity and composition percent)
4. Adsorption equilibrium data
5. The concentration of adsorbate component in the gas phase at the outlet of the substrate
6. The concentration of adsorbate component in the solid phase at the outlet of the substrate
7. partial pressure of adsorbate in the gas phase at the outlet of the substrate

2.8.3 Problem-solving

Since the equation is based on time and space, it should start from zero time and solve for the whole region, and after solving it at time zero, it was time one and so on for the whole time. That's mean, at time n, the data is obtained for all points and by that the data is calculated at time n + 1. Therefore, there is a 'for' external loop that actually changes from one step to the next. Inside the 'for' outer loop which time is there, there is a 'for' inner loop for, which indicates location changes from the beginning to the end of the substrate. The inner loop is about the 'while' loop that is solved within this equation loop at a constant time and place. In fact, in the two previous 'for' loops, one time and another determined the location, and in the last loop (while) at this time and place, the amount of adsorbate concentration in the solid phase (q) and the adsorbate concentration in the gas phase (c) comes.

Table 2: Incoming applications

Parameter	**symbol**	**unit**
temperature	T	K
Pressure	P	Pa
number of intervals	m	No
number of spatial distance	n	No
Length of bed	L	m
length ratio to the number of spatial distances	dz	m
time of passage of gas from the whole bed	dt	s
Adsorbate mole fraction at the moment of entering the bed	Y_{in}	No
speed	u	m/s
Adsorbent density	ρ_p	kg/m3
substrate porosity	ε_b	No
Maximum adsorption capacity in dual-Langmuir isotherm	q_{s1}	mol/kg
Maximum adsorption capacity in dual-Langmuir isotherm	q_{s2}	mol/kg
Coagulation coefficient of adsorption in the Langmuir isotherm	b_1	1/Pa
Coagulation coefficient of adsorption in the Langmuir isotherm	b_2	1/Pa
linear force propulsion coefficient	k_{LDF}	1/s
axial dispersion coefficient	D_{ax}	No

3 Results and discussion

Based on the model derived from this study, the effective parameters on adsorption will be analyzed individually in order to provide optimum conditions for adsorption performance in the towers. First of all, you need to ensure the performance of the model. To evaluate the proposed model, experimental data of Habib Mohammadinezhad have been used [25]. In this study, the adsorption curve for carbon dioxide adsorption by zeolite 5A at temperature and pressure of the experiment has been investigated. Comparison of the results of the model and experimental data is shown in Fig. 1. The results of this comparison show that the proposed model can predict the experimental data. As shown in Fig. 1, the model developed by the laboratory data for carbon dioxide adsorption is well-suited to zeolite. Therefore, this model can be used to predict the effect of operating conditions on adsorption and to achieve optimal conditions to improve the performance of an adsorption tower.

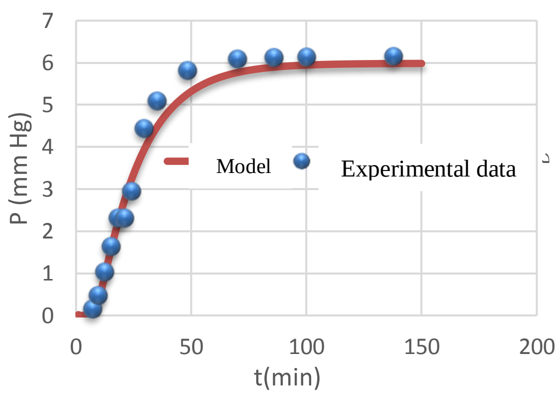


Fig. 2: Comparison of experimental and model adsorption data

3.1 Performance conditions for adsorption

Adsorption of carbon dioxide on zeolite 5A in laboratory conditions. Specifications and test conditions are presented in Tables 3 and 4 [25]. The least squares error method has been used in order to obtain the dual-Langmuir isotherm quantities. Equilibrium quantities of this isotherm are presented in Table 5.

3.2 The effect of changing the input feed conditions of the unit

Feed conditions are one of the main determinants of any chemical process. Therefore, the effect of these conditions on process performance improvement is very effective. In general, adsorption processes have a good flexibility versus changing various feed conditions. In this section, the effect of temperature, flow, and methane concentration of the input has been investigated by using this simulation.

3.2.1 Effect of feed temperature

Since the study is carried out at the same temperature, the temperature of the feed determines the temperature of the adsorption step. In order to investigate the effects of temperature on adsorption, the simulation was carried out at several temperatures and it was found that with increasing temperature, the amount of adsorption decreased. Because the superconducting forces (superficial potential) will be deflected against the temperature (kinetic) forces, although the pressure is too high. As the temperature rises, the adsorption rates are rapidly degraded, so it is best to perform adsorption at lower temperatures if possible [26].

Table 3: Construction characteristics of the filled tower

L =0.254 m	Length of the tower
D=0.0472 m	Tower diameter
ε_b=0.464	Substrate porosity

Table 4: Adsorption Operating Conditions

T=298 K	Temperature
P= 108109 pa	Pressure
U=0.523362 m/s	Speed
K_{LDF}=0.00101 s^{-1}	Linear force
D_{ax}=0.0003	Axial dispersion coefficient
ρ_P= 2220 kg/m^3	The apparent density
Y_{in}=0.0073	Input mole fraction of adsorbate

Table 5: Convergence of the two-site Langmuir isotherm

b_1= 2.2 3446*10^{-5}	pa^{-1}	q_{s1}= 1.599	mol/kg
b_2= 0.048905	pa^{-1}	q_{s2}=0.049	mol/kg

As shown in Fig. 4.1, at 65min the adsorbate amount in the outlet gas phase from the bed at T = 298, T = 308, and T = 318, respectively, is 306.0, 295.0, and 285.0. Since the adsorption step is relatively long and it is considered to avoid temperature changes due to heat exchange with a feed temperature near the ambient temperature.

3.2.2 Effect of feed flow intensity

The flow Intensity in this process is affected by gas velocity in the equations. Increasing gas velocity increases the speed of movement of the activated region, and as the region approaches the outlet, the concentration of the adsorbate material in the product increases. In the lower flow rate due to more contact time, the pollutant has more chance of bonding with the adsorbent particles. Therefore, the adsorption efficiency increases. The amount of air inflow strongly affects adsorption capacity. As the flow rate increases, the adsorption and trapping of methane molecules decrease. The reason for this is that methane remains in the adsorption bed, and some of it leaves the bed before reaching the balance point. Similar results are presented by other researchers [27-31]. As shown in Fig. 4, the adsorbate concentration in the outlet gas phase reaches a value of 0.15, with a velocity of = 0.4, u = 0.52and u = 0.6respectively of 30, 23 and 20 minutes is required.

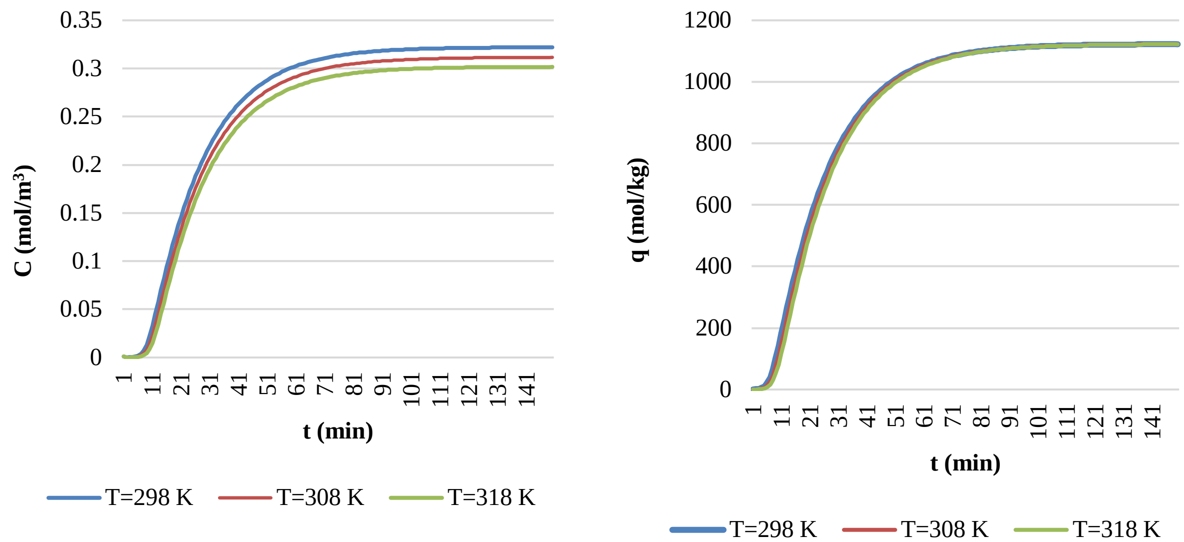


Fig. 3: Effect of feed temperature on output concentration

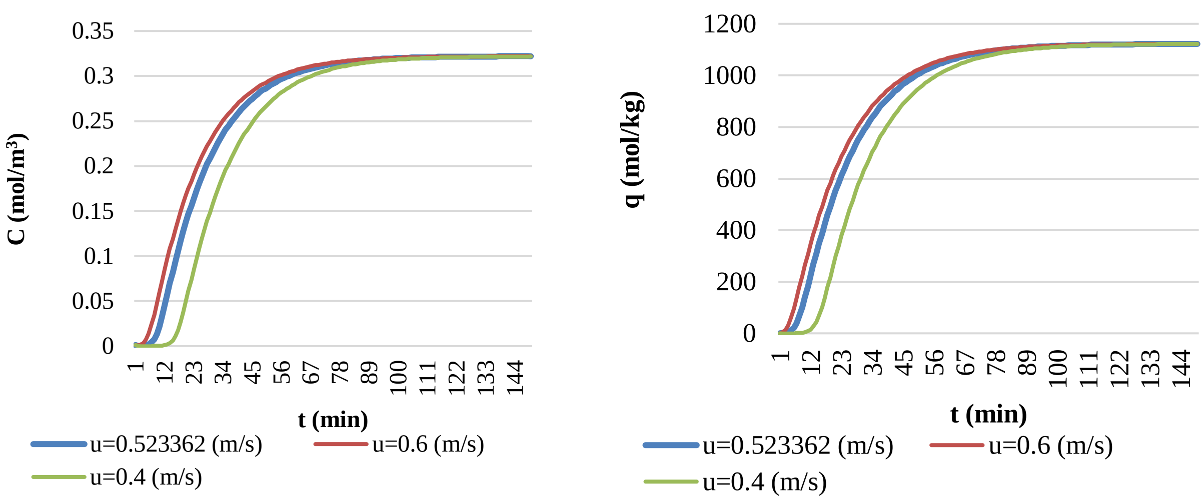


Fig. 4: Effect of speed on output concentration

3.2.3 The effect of pressure

In order to investigate the effect of pressure on the performance of the absorption tower, the crevice curve has been plotted for several different pressures. According to the diagram, with increasing the pressure in the bed, the slope of the curve increases and the time of saturation of the bed specified length decreases. The reason for this is that with increasing pressure the length of the mass transfer region increases. With increasing pressure, adsorbents absorb more pollutant gas. As shown in Fig. 5, at a specified time of 50 minutes, the adsorbate amount in the outlet gas phase from the bed was P = 15.68, P = 18and P = 20= 0.28, 0.33, and 0.37.

3.2.4 Effect of bed length

By increasing the height of the adsorption bed in the column, the contact time of the pollutant is increased with the absorbent and the slope of the deflection curve decreases, which indicates a massive mass transfer region at higher altitudes. These results indicate that adsorbent optimum efficiency is achieved with higher heights. As shown in Fig. 6 the adsorbate concentration in the outlet solid phase from the bed reaches a value of 200, with a bed length of L = 0.254, L = 0.3and L = 0.35, respectively of 13, 20 and 25 minutes are needed.

3.2.5 Effect of adsorbent particle size

The amount of adsorbent in the absorption column determines the number of available and active adsorption sites. According to Equation 3, the mass transfer coefficient with the absorbent diameter has a reverse relation and, with increasing mass transfer coefficient, the absorbent diameter decreases. As shown in Fig. 7, the time to reach the breaking point in the diagram increases with decreasing mass transfer coefficient. By increasing the number of adsorbent particles, the time remaining in the column and the time to reach the breaking point increases, and the saturation of the substrate occurs later, which leads to an increase in adsorption capacity. This is due to an increase in the absorbent special level and absorption sites.

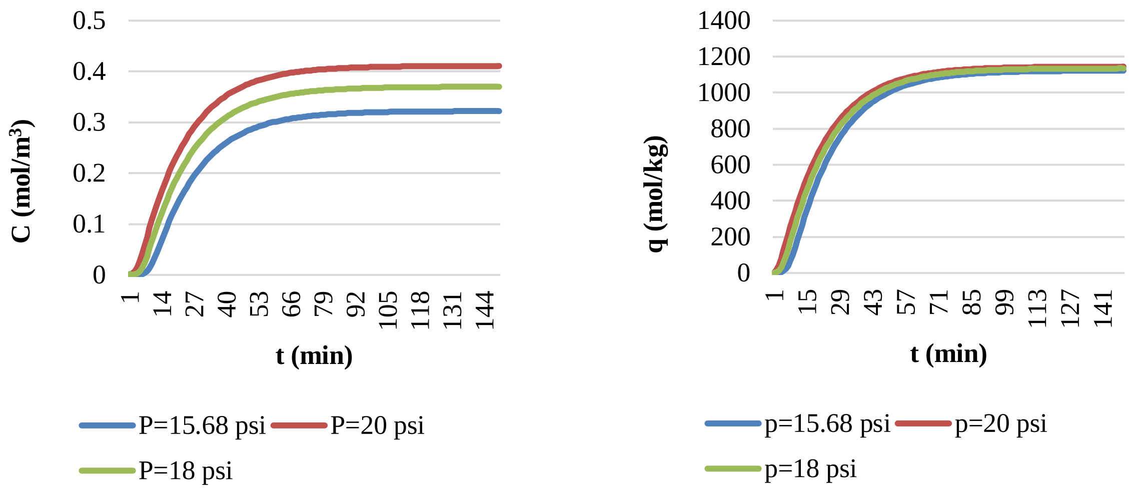


Fig. 5: Effect of pressure on the output concentration

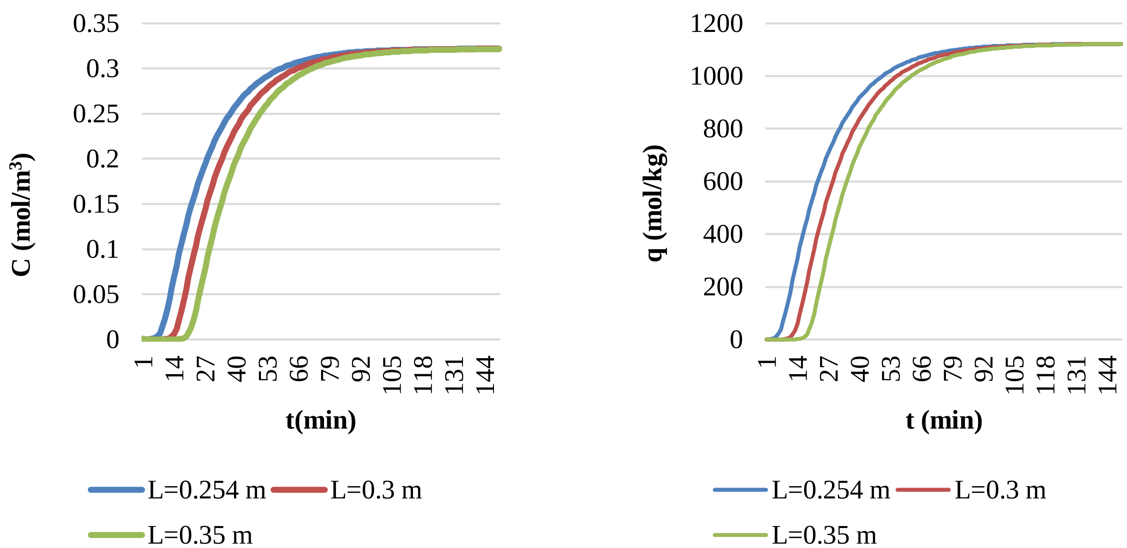
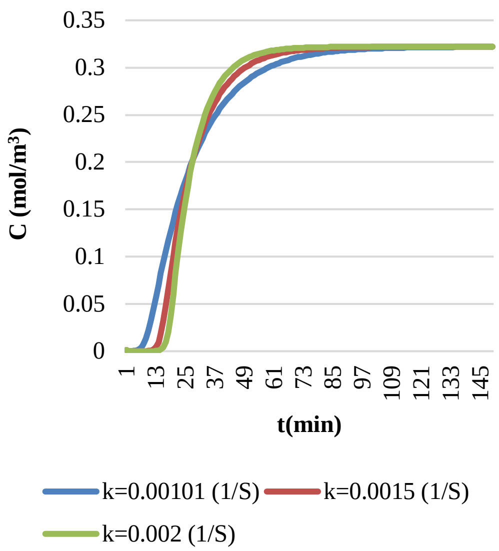


Fig. 6: Effect of bed length on output concentration



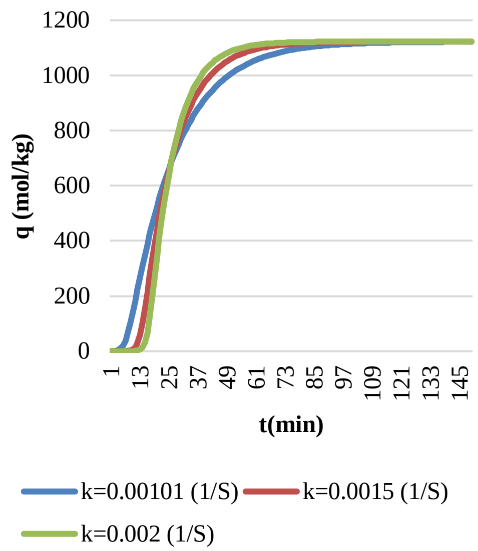


Fig 7: Effect of bed length on output concentration

4 Conclusion

The results of this study indicate that bed length, tower pressure and gas flow rate have a significant effect on the methane separation efficiency. Also, in this study, the parameters of gas velocity and tower temperatures were studied and their effect on the adsorption process was investigated.

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