

CO₂ sorption from a mixture of N₂/CO₂ using an activated carbon and silica gel: equilibrium, breakthrough, column efficiency and mass transfer zone

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Graphical abstract



Abstract

An increased level of CO₂ in the atmosphere has led to global warming and prime contributor of greenhouse gases. The temperature, feed rate, column efficiency, usable bed height, unutilized bed height, length of mass transfer zone, utilization factor and partial pressure are the parameters considered for fixed bed sorption of CO₂ from N₂/CO₂ mixture. The breakthrough time relies strongly on the temperature and feed rate. The prolonged breakthrough and saturation times have been realized for AC. The response curves of AC are vastly steep signifying the maximal utilization of bed capacity at the breakpoint. In general, the length of MTZ increases with raised temperature and feed flow rate. The capacity utilization factor reduces with raised temperature and feed flow rate.

A utilization factor of 0.919 was determined for AC. The maximal capacity for CO₂ reduces significantly with an increased temperature. The maximal capacities of 32.99 gm CO₂/Kg was determined at a temperature of 298 K for AC. The column efficiency, usable bed height and unutilized bed height decrease with increased temperature and flow rate. The maximal column efficiency and usable bed height of 85.13% and 20.43 cm were determined for AC, respectively. The capacity improves considerably with CO₂ partial pressure and AC exhibited higher adsorption capacity compared to SG. The capacity improves considerably with increased feed rates and maximal capacity of 39.14 g CO₂/Kg adsorbent was determined for AC at the feed rate of 8.33 x10⁻⁵ m³/sec. Owing to higher sorption capacity and enhanced characteristic parameters, the AC may be used for economical separation of CO₂ from N₂/CO₂ mixture.

Keywords: Sorption, utilization factor, breakthrough, mass transfer zone, capacity.

1. Introduction

CO₂ is the most profuse greenhouse gas (GHG) contributing significantly to changes in the global climate. Greenhouse gases have become a worldwide issue, and an increasing concentration of GHGs in the atmosphere is detrimental to health and our lifestyle. The combustion of fossil fuels accounts for almost 81% of energy used for commercial purposes, and it releases 3.0x10¹³ Kg of carbon dioxide per year (Hester and Harrison, 2010). The prime environmental issue concerns with the alarming rate at which the carbon dioxide concentration is rising in the atmosphere (Li *et al.*, 2013; Rashid *et al.*, 2016). The power generation sector between the years 2000 to 2030 will have contributed to almost the total increase in global CO₂ emissions (IEA, 2003). As much as 45% of the total global CO₂ emission is attributed to thermoelectric and industrial electricity generating plants (Lee and Park, 2015).

The mean global temperature rise by just 2°C will result in far-reaching consequences. Therefore, it is recommended

that the greenhouse gases be reduced to 50% by the year 2050 (IPCC, 2001). The CO₂ emissions can be reduced by one of the three methods relying on the plant configuration (Alabadiet *et al.*, 2015). The treatment of synthesis gas primarily made up of CO and H₂ gases is performed in the pre-combustion process (Figueroa *et al.*, 2008). Approximately 1.5 times more energy is required in the post-combustion process compared to the pre-combustion method of CO₂ capture (Gibbins and Chalmers, 2008). Primarily, CO₂ and H₂O are the emissions of the oxy-fuel combustion (Blomenet *et al.*, 2009).

There are primarily four technologies available for CO₂ reduction from the emissions of power plants based on fossil fuel. Chemical and physical absorption is realized by a reaction between CO₂ and solvent, primarily the amines solutions (Choi *et al.*, 2009). Amine solutions were utilized for CO₂ absorption from CO₂/air mixture and a capacity of 98.2x10⁻²mol/mol was reported for DEA (Gomes *et al.*, 2015). The streams having CO₂ concentration of more than 50% were normally treated using cryogenic separations (Gupta *et al.*, 2003). In the case of CO₂ concentration being more than 20%, the membrane systems are extremely adaptable (Brunetti *et al.*, 2010). The post-combustion CO₂ diminution is mainly a pragmatic process due to the fact that various suggested technologies can be back fitted to existing plants based on fossil fuel (MacDowell *et al.*, 2010; Cohen *et al.*, 2011; Yang *et al.*, 2008). Adsorption is considered as a well-established technology to capture CO₂ in the post combustion process (Garcia *et al.*, 2011; Valverde *et al.*, 2011). The adsorbent preferentially separates CO₂ and afterwards regenerates to release the CO₂ irrespective of the process configuration (Alonso *et al.*, 2010; Li *et al.*, 2011).

Fine activated carbons have been employed under sound assisted fluidization for CO₂ capture, and the experiments reported enhanced adsorption (Raganati *et al.*, 2015). Fluidized bed column was utilized to analyze the separation of CO₂ from flue gas feed experimentally (Abanadaset *et al.*, 2004; Valverde *et al.*, 2012). In-depth study of breakthrough behavior was conducted utilizing activated carbon beads (Shen *et al.*, 2011). An activated carbon based adsorbent was used for CO₂ capture by temperature swing adsorption (TSA) and result showed that CO₂ adsorption/desorption is influenced by pressure and temperature differently (Raganati *et al.*, 2020). The activated carbons, zeolites and molecular sieve were employed to explore the kinetics and equilibrium (Sarkar *et al.*, 2017). The effect of superficial velocity on breakthrough response utilizing MOF, activated carbons and crystalline pellets was investigated (Al-Janabiet *et al.*, 2018). The beads of activated carbons were utilized to examine the adsorption equilibrium with feed mixture of CO₂ and N₂ (Shen *et al.*, 2010). A MOF (UTSA-16) was utilized for CO₂ separation, and the maximal adsorption capacity of 1.60x10²cm³/cm³ was reported (Xing *et al.*, 2012). The adsorption response of polyaspartamide was predicted using models based on kinetics, and it was concluded that an adsorption is credited to external mass transfer (Yoro *et al.*, 2017). An adsorption of CO₂ employing

various adsorbents has been examined and the maximal capacity was reported for activated carbons (Lu *et al.*, 2008). The parametric study and breakthrough behavior by utilizing two grades of activated carbons employing fixed bed column have been conducted (Al Mesferet *et al.*, 2018; Al Mesfer and Danish, 2018).

A yellow tuff has been utilized for CO₂ capture in fixed bed column and an adsorption capacity of 0.710 mmol/g at 20 °C was obtained with focus on thermodynamics and kinetics of process (Ammendola *et al.*, 2020). Investigators have used Macadamia shell biomass to prepare activated carbons by microwave irradiation technique (Dejanget *et al.*, 2015). Additionally, walnut shell has also been used to synthesize activated carbon and employed for manufacturing the cartridge (Jahangiriet *et al.*, 2012). An activated carbon fiber was prepared, and a capacity of 1.3x10⁻³mol/gm adsorbent was achieved (Calvo-Munoz *et al.*, 2016). The capacity of 9.09x10⁻³mol/gm was reported for synthesized activated adsorbent prepared by treating coal with KOH (Toprak and Kopak, 2017). Activated carbons/N-enriched activated carbons were utilized for CO₂ separation and replicated the breakthrough curve satisfactorily (Dantaset *et al.*, 2010). A synthesized adsorbent of 13x and activated carbon was used for CO₂ separation, and determined a capacity of 2.63 mmol/gm (Regufeet *et al.*, 2018).

The optimal incorporation of carbon nanotubes into the zeolites 13x was carried out, and the increased adsorption capacity was reported (Qasemet *et al.*, 2017). Microporous biochar adsorbent was employed for the capture of adsorbate in a fixed bed for experimental and simulation study (Plaza *et al.*, 2016). The amine adsorbents were synthesized using fly ash, and the increased capacity of PEI/FA type- adsorbents was reported (Zhang *et al.*, 2014). The prolonged breakpoint time equal to 350 sec with a capacity of 0.63 mmol CO₂/gm AC for AC-PKS/CeO₂ has been reported (Hidayu and Muda *et al.*, 2017). Researchers have used walnut and almond shell to synthesize the activated carbons and the internal structure was analyzed using SEM and FTIR (Patilet *et al.*, 2013). Walnut shell-based biomass has been converted into activated carbons, and KOH was reported as the prime favorable activation agent (Xia *et al.*, 2016). The chemical activation method (Mataji and Khoshandam, 2014) was applied to prepare activated carbons from walnut shell biomass, and adsorption capability of the removal of benzene was explored using ZnCl₂/H₃PO₄. A study (Li *et al.*, 2013) focused on adsorption separation of CO₂/CH₄ was carried out at different temperature of 30 °C and 40 °C and capacity of 1.6 mmol/g was reported at 30 °C and 1 atm. The aim of the current work is to study the adsorption response of activated carbon (AC) and silica gel (SG). The sorption response will be analyzed using temperature, feed rate, length of mass transfer zone, utilization factor, column efficiency, usable bed height and partial pressure as the parameters.

2. Experimental

2.1. Chemicals

The activated carbon (AC) with size range 0.4-0.8 cm was procured from Fluka analytical and dried at 110 °C in the drying oven before the experimental works. The silica gel (SG) with average size range 0.2 to 0.4 cm was procured from GCC laboratory reagent (UK) and dried at a temperature of 120 °C before the experimental work. The selected adsorbents were used for experiments as such without any further treatment.

2.2. Setup diagram

The setup sketch of the adsorption column (effectual length- 24 cm) used for experiments has been shown in Figure 1. The column is made up of stainless steel and jacketed to allow the hot water flow to attain the desired temperature. Flow controllers F₁, F₂ and F₃ measure and control the flow of N₂, CO₂ and CO₂ flow to IR sensor respectively. There are 6 thermocouples positioned along the axial direction of the column.

2.3. Procedure

The gaseous mixture of known composition consisting of CO₂ and N₂ enters at the bottom of the column. The flow of CO₂ and N₂ were controlled by the flow controllers F₁ and F₂. The heating jacket was used to control the desired bed temperature and PID controller incorporated in the control console accomplished the task of fixing the desired temperature. The column exit concentration was measured using IR sensor.

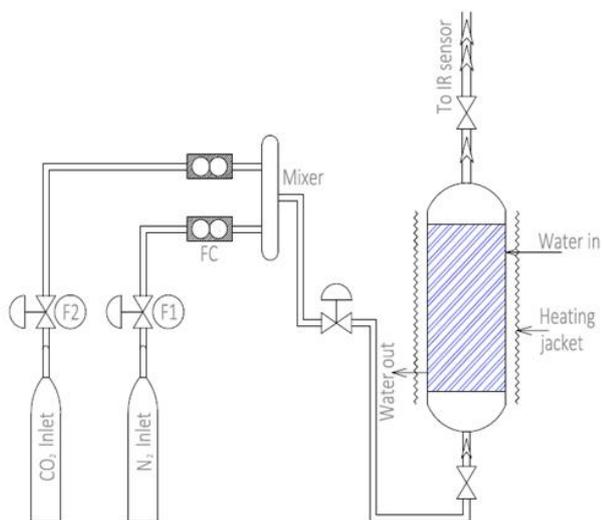


Figure 1. Setup diagram of sorption unit

3. Results and discussion

3.1. Adsorbent surface characterization

Surface area characterization of both the AC and SG was carried out using surface analyzer (Quantachrome NoavaWin-NOVA Instruments). The results obtained are summarized and depicted in Table 1. The single point surface areas of 858 m²/gm and 590 m²/gm were exhibited by activated carbon and silica gel respectively. The AC revealed higher pore volume equal to 0.425 cm³/gm

compared to SG. The nearly same pore radii of 1.838 Å and 1.835 Å have been reported for AC and SG respectively.

Table 1. BET surface characterizations

Characteristics	Activated carbon	Silica gel
Single point surface area (m ² /gm)	858	590
Multipoint surface area (m ² /gm)	862	599
Micropore volume (cm ³ /gm)	0.425	0.273
Pore radius (Å)	1.838	1.835

3.2. Activated carbon (AC)

The reliance of sorption response (in terms of C/C₀) on time at different temperatures using AC (Wt.=180 gm) is depicted in Figure 2. The experiments were performed at a feed rate of 6.67x10⁻⁵m³/sec and carbon dioxide concentration of 0.05 (wt. fraction). The breakthrough time relies strongly on the temperature. The maximal breakthrough and saturation times of 870 sec and 1050 sec were realized at a temperature of 298 K. The breakthrough and saturation time declined to 725 sec and 915 sec on increasing the temperature at 308 K respectively. A reduced breakthrough time of 630 sec was attained at temperature of 318 K. Sorption temperature of 328 K attributed to reduced breakthrough and saturation times of 530 sec and 675 sec respectively. The prolonged breakthrough time at decreased temperature signifies the enhanced capacity.

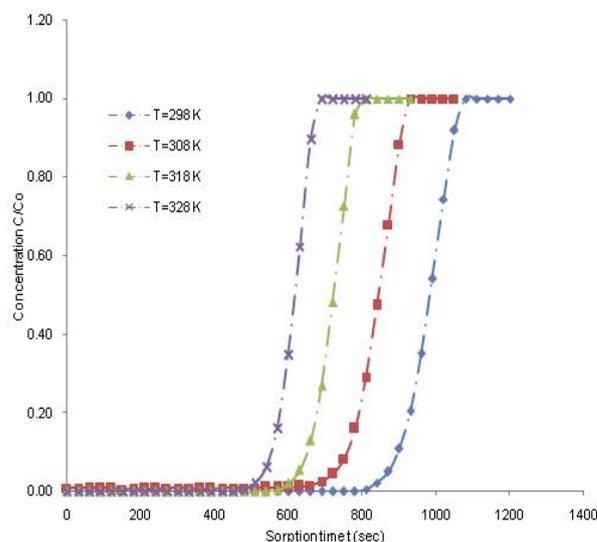


Figure 2. Breakthrough response for activated carbon for AC (F=6.67x10⁻⁵m³/sec, C₀=5%)

On the other hand, the breakthrough curve is vastly steep and it signifies the maximal utilization of the sorbent capacity. The utilization of maximal capacity at breakpoint is desired for economical separation for CO₂. The steepness of the response curve signifies the narrowness of metastable zone (MSZ). Narrow metastable zone attributes to faster adsorption. In all cases, minute differences in the

width of meta-stable zone have been observed which indicate the almost same utilization of bed capacity at the breakthrough point. The lowest layer of the bed is nearly saturated and mostly the adsorption takes place over a comparatively narrow adsorption zone in which the concentration varies quickly. Narrow MTZ characterize the competent use of the adsorbent leads to reduced costs of energy regeneration.

The sorption responses at various feed rates have been depicted in Figure 3. The feed rates of $5.00 \times 10^{-5} \text{ m}^3/\text{s}$, $6.67 \times 10^{-5} \text{ m}^3/\text{s}$ and $8.33 \times 10^{-5} \text{ m}^3/\text{s}$ were selected at fixed temperature of 298 K. The breakthrough and saturation times vary significantly with feed rates. The prolonged breakpoint time of 1145 sec was attained at a feed rate of $5.00 \times 10^{-5} \text{ m}^3/\text{sec}$. The breakpoint time reduced from 1145 sec to 975 sec on raising the feed rates from $5.00 \times 10^{-5} \text{ m}^3/\text{sec}$ to $6.67 \times 10^{-5} \text{ m}^3/\text{sec}$. The minimal breakthrough and saturation times of 815 sec and 985 sec are exhibited by the AC at a feed rate of $8.33 \times 10^{-5} \text{ m}^3/\text{sec}$. The saturation and breakthrough periods reduced considerably with increased feed flow rate. Also, the narrow metastable zone exhibited by AC characterizes the suitability of carbon based adsorbent for carbon dioxide separation from CO_2/N_2 mixture. The steepness of the curve (S-shape) is very significant and highly desirable for economical adsorption. The mass transfer zone moves up the column as feed mixture flow. After the breakpoint time is approached, the concentration C raises very quickly upto the end of curve where the bed is judged unproductive.

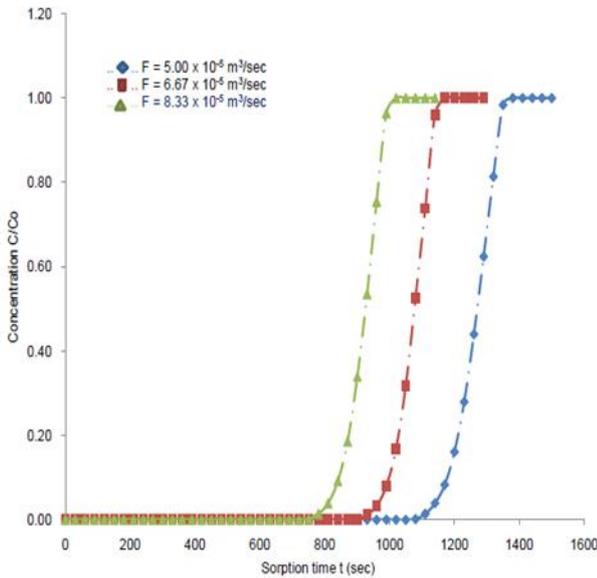


Figure 3. Breakthrough response at various feed rates for AC ($T=298 \text{ K}$, $C_0=5\%$)

The sorption capacity is estimated by dynamic mass balance which needs integration of the adsorption data. Utilizing the curve data, the time (t_s) equal to the total/stoichiometric capacity is normally tabulated by integrating the following equation (Monazamet *et al.*, 2013):

$$t_s = \int_0^{\infty} \left(1 - \frac{C}{C_0}\right) dt \quad (1)$$

where, C is the CO_2 concentration at time t , C_0 is the feed concentration of CO_2 . Knowing t_s , the dynamic capacity q_t of the bed can be calculated as (Serna-Guerrero and Sayari, 2010)

$$q_t = \frac{F t_s C_0}{m_{ad}} \quad (2)$$

where, F is total feed molar flow rate, and m_{ad} is the mass of sorbent used in the bed. The portion of the column where adsorbate is really adsorbed on the sorbent is known as mass transfer zone (MTZ) length. The MTZ normally shifts from the inlet towards exit during the sorption operation. It means that the sorbent adjacent to inlet becomes saturated with the adsorbate and then the sorption zone towards the end-side of the bed. A schematic depiction of the response curve and shifting of the MTZ has been shown in Figure 4. The MTZ length was approximated assuming the constant pattern adsorption (Pota and Mathews, 1999):

$$L_{MTZ} = \frac{2 L (t_s - t_b)}{t_s + t_b} \quad (3)$$

Where L is the bed length; t_b and t_s stand for the breakthrough and saturation time (exhaustion time) respectively. These breakthrough and saturation times are the times corresponding to the outlet concentrations of 5% and 95%. For assumed symmetric breakthrough curve, the bed capacity utilization can be estimated using utilization factor as:

$$f = 1 - \frac{0.5 L_{MTZ}}{L} \quad (4)$$

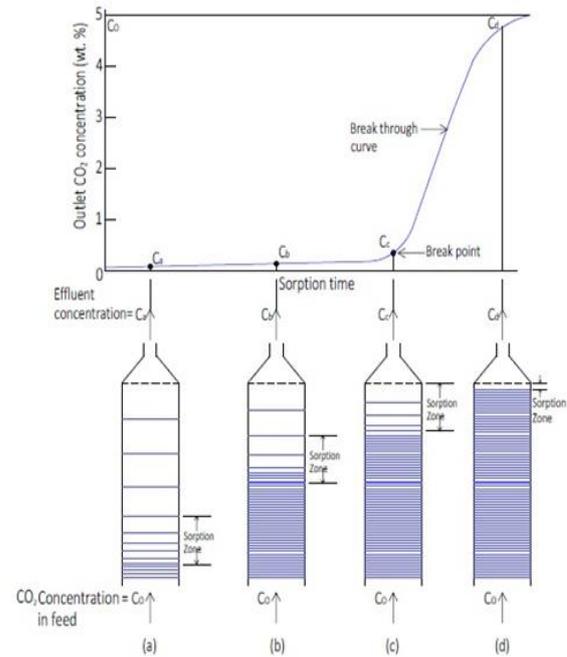


Figure 4. Breakthrough and mass transfer zone

The characteristics parameters i.e. saturation time, breakthrough time, length of the MTZ and capacity utilization factor were determined from the response curves (Figs. 2 and 3) for AC and depicted in Table 2. The breakthrough and saturation times decrease with increased temperature, leading to longer mass transfer zone. Also, breakthrough and saturation times reduce with feed flow rate resulting in increased length of MTZ. In common, the reduced temperature causes shorter MTZ owing to increased nucleation of product species. The length of MTZ increases with increased temperature and flow rate. The maximal length of MTZ equal to 5.78 cm was realized at T=328 K and feed rate of F=6.67 x10⁻⁵ m³/sec and reduced to 4.50 cm at a temperature of 298 K for AC. The utilization factor decreases with increased sorption temperature and feed rate. The maximal value of f=0.919 has been determined at T=298 K and F=5.00x10⁻⁵ m³/sec.

Table 2. Effect of operating conditions on characteristics parameters for AC

T (K)	F, x10 ⁻⁵ (m ³ /sec)	t _b (sec)	t _s (sec)	L _{MTZ} (cm)	f
298	6.67	870	1050	4.50	0.906
308	6.67	725	915	5.56	0.884
318	6.67	630	778	5.05	0.895
328	6.67	530	675	5.78	0.879
298	5.00	1145	1345	3.86	0.919
298	8.33	815	988	4.60	0.904

The CO₂ sorption capacity variation with temperature is predicted below (Figure 5). The data were obtained at a feed rate equal of 6.67x10⁻⁵ m³/sec. The maximal capacity of 32.99 gm CO₂/Kg adsorbent was attained at a temperature of 298 K but reduced to 26.00 gm CO₂/Kg adsorbent at an increased temperature of 308 K. The capacity of 22.44 gm/Kg adsorbent was obtained at a temperature of 318 K. The temperature of 328 K contributed to a CO₂ capacity of 11.22 gm/Kg adsorbent.

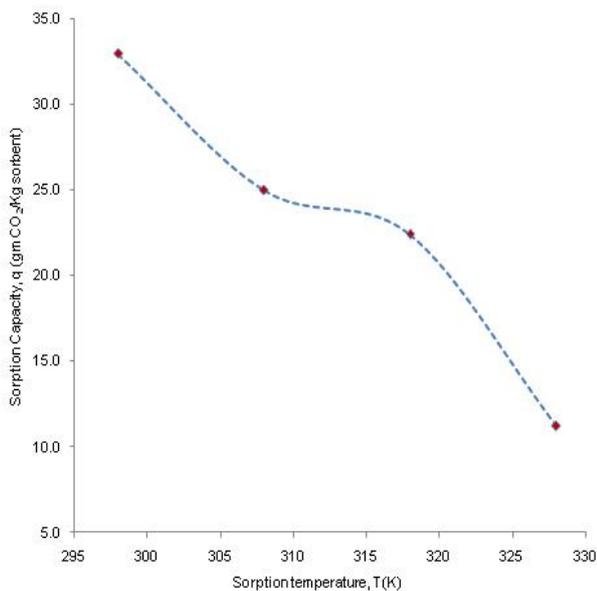


Figure 5. Sorption capacity vs. temperature curve for AC (F=6.67 x10⁻⁵ m³/sec, C₀=5%)

It was found that the maximal selective capacity of the adsorbent for CO₂ was strongly depended on the temperature and significantly reduced with increased temperature. The reduced temperature increases the capacity of CO₂ adsorption from CO₂/N₂ feed mixture. The saturation adsorbent capacity varies significantly with bed temperature. The concentration of adsorbed gas or adsorption capacity lessened with increased temperature at a given equilibrium pressures, as adsorption is an exothermic process.

The adsorption isotherms at a feed rate of 6.67x10⁻⁵ m³/sec using activated carbon (AC) have been exhibited in Figure 6. The impact of different temperatures on sorption response was investigated at a fixed inlet CO₂ concentration of 5%. The capacity increases considerably with increased pressure of the CO₂. At a bed temperature of 298 K, the maximal adsorption capacity of 32.99 gm CO₂/Kg adsorbent was realized at an equilibrium partial pressure of 1.25x10⁵ N/m². The capacity declined from 32.99 gm CO₂/Kg adsorbent to 12.20 gm/Kg adsorbent on reducing the CO₂ partial pressure from 1.25x10⁵ N/m² to 0.50x10⁵ N/m². At the same temperature (T=298 K), the adsorption capacity of 1.42 gm CO₂/Kg adsorbent was determined. For an adsorption isotherm produced at a bed temperature of 308 K, the minimal capacity of 1.09 gm CO₂/Kg adsorbent (P*=0.062 x10⁵ N/m²) was exhibited by the adsorbent bed, and the capacity further enhanced to 20.14 gm/Kg adsorbent at a CO₂ pressure of 1.0x10⁵ N/m². The sorption capacity of 22.441 gm/Kg adsorbent at a partial pressure of 1.25x10⁵ N/m² was realized corresponding to a temperature of 318 K and capacity declined considerably to 10.34 gm CO₂/Kg adsorbent at a CO₂ pressure equal of 0.625 x10⁵ N/m². The maximal and minimal capacities of 11.22 gm/gm adsorbent and 0.47 gm/Kg adsorbent have been exhibited at equilibrium partial pressures of 1.25x10⁵ N/m² and 0.063x10⁵ N/m² respectively at a temperature of 328 K.

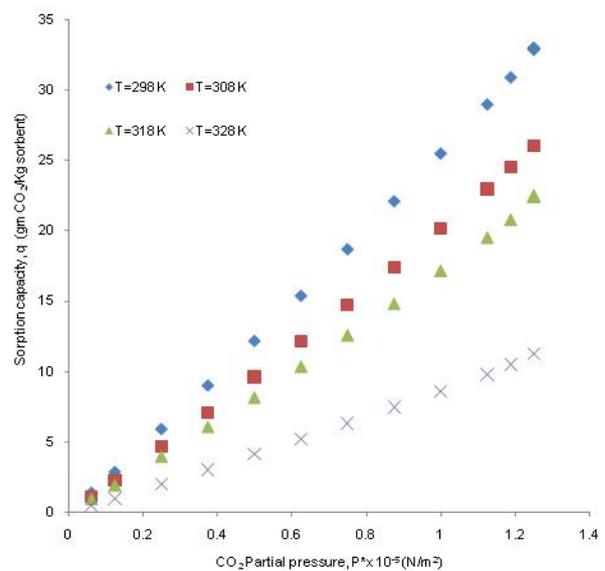


Figure 6. Sorption capacity vs. partial pressure curves for AC (F=6.67 x10⁻⁵ m³/sec)

Different gases are adsorbed to different extents under comparable conditions. It can be concluded that increasing the pressure of carbon dioxide will cause more of the adsorbable gas to be adsorbed as the rising curve indicates. It may be concluded that the adsorption capacity increases remarkably with partial pressure of the CO₂.

The reliance of the feed rate on the capacity utilizing the AC from a CO₂/N₂ mixture has been depicted in Figure 7. The bed temperature was fixed at 298 K and the adsorbable gas concentration in feed (CO₂) was adjusted at 0.05 (vol. %). The sorption capacity increases considerably with increased feed (N₂+CO₂) rate from 5.00x10⁻⁵ m³/sec to 8.33 x10⁻⁵ m³/sec. The capacity of 32.36 gm CO₂/Kg sorbent was realized at a feed rate of 5.00x10⁻⁵ m³/sec and further enhanced to 36.39gm CO₂/Kg sorbent on raising the feed rate at 6.67x10⁻⁵ m³/sec. It was realized that as the increasing amount feed (fluid) are passed through the sorption column, the sorbent adsorbs an increasing amount of solute (CO₂) from the feed gas mixture yielding to increased sorption capacity. Therefore, it can be concluded that the capacity of the sorbent enhances significantly with increasing feed rates. The higher feed rate favors the increased sorption capacity of the adsorbed gas from the feed mixture (N₂+ CO₂).

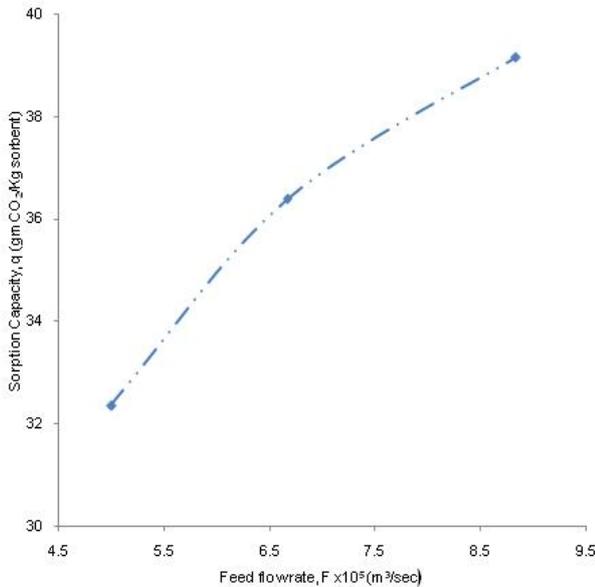


Figure 7. Sorption capacity vs. feed flow rate for AC (T=298 K)

The adsorption isotherms obtained at T=298 K with feed rates ranging 5.00x10⁻⁵ m³/sec to 8.33 x10⁻⁵ m³/sec have been depicted in Figure 8. The capacity enhances with raised partial pressure of the adsorbable gas (CO₂). The capacity equal to 1.43 gm CO₂/Kg sorbent was obtained at a CO₂ pressure of 0.063x10⁵ N/m² and the capacity increased to 25.07 gm CO₂/Kg adsorbent on raising the CO₂ pressure at 1.0x10⁵ N/m² at a feed rate of 5.00x10⁻⁵ m³/sec. The maximal capacity equal to 32.36 gm CO₂/Kg sorbent was exhibited at a CO₂ partial pressure of 1.25x10⁵ N/m². The increased capacity of 28.23 gm /Kg sorbent was determined at a CO₂ partial pressure equal to 1.0 x10⁵ N/m² on fixing the feed rate at 6.67x10⁻⁵ m³/sec. Generally,

capacity improved remarkably with increased feed flow of the CO₂/N₂. The dependence of the capacity is more pronounced with adsorbable gas partial pressures compared to feed rates.

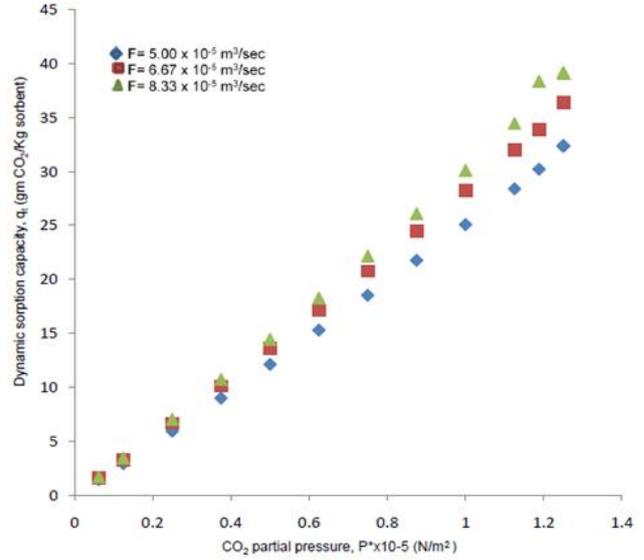


Figure 8. Sorption capacity vs. CO₂ partial pressure curve (T=298 K)

Equation (1) for breakthrough time t_b , which represents the area above the curve between the limit $t=0$ to $t= t_b$, can be written as Equation (5):

$$t_b = \int_0^{t_b} \left(1 - \frac{c}{c_0}\right) dt \quad (5)$$

The efficiency based on column capacity or fraction of total column capacity that is efficiently used can be determined as:

$$\eta = \frac{t_b}{t_s} = \frac{\int_0^{t_b} \left(1 - \frac{c}{c_0}\right) dt}{\int_0^{\infty} \left(1 - \frac{c}{c_0}\right) dt} \quad (6)$$

The length of the adsorption column (H_b) used upto the breakpoint, or usable bed height, is determined using Equation (7):

$$H_b = \frac{t_b}{t_s} H \quad (7)$$

Where, H denotes the total bed height. An unusable bed height can be determined by Equation (8):

$$H_{ub} = \left(1 - \frac{t_b}{t_s}\right) H \quad (8)$$

The characteristic parameters of the CO₂ capture for AC, i.e., effective column efficiency, usable bed and unusable bed height, were evaluated, as depicted in Table 3. The efficiency based on column capacity varies considerably with temperature and feed rate. In general, the efficiency declines with increased temperature and the maximal efficiency of 82.86% was determined for AC at 298 °C with $F= 6.67 \times 10^{-5} \text{ m}^3/\text{s}$. The increased feed rate 5.00 x10⁻⁵ m³/s to 8.33 x10⁻⁵ m³/s contributed to enhanced column efficiency and highest adsorption capacity of 85.13 % was obtained at minimal flow rate of 5.00 x10⁻⁵ m³/s with 298 K

and $C_0=5\%$. The maximum usable bed height of 20.43 cm was determined. The usable bed height or length of bed used upto the breakthrough decreases with increased temperature and feed rate. The maximal usable bed height of 20.43 cm of the effective bed height of 24 cm was observed at 298 K with $F= 5.00 \times 10^{-5} \text{ m}^3/\text{s}$. The highest unutilized bed height of 5.15 cm was evaluated at a temperature of 328 K and $F= 6.67 \times 10^{-5} \text{ m}^3/\text{s}$. Therefore, it can be suggested that bed characteristic parameters vary considerably with operating conditions and may be optimized for better adsorption performance.

Table 3. Characteristics parameters of CO₂ capture for AC

T(K)	F, $\times 10^{-5}$ (m ³ /s)	C ₀ (vol.%)	η (%)	H _b	H _{ub}
298	6.67	5	82.86	19.89	4.11
308	6.67	5	79.24	19.02	4.98
318	6.67	5	80.98	19.44	4.56
328	6.67	5	78.52	18.85	5.15
298	5.00	5	85.13	20.43	3.57
298	8.33	5	82.13	19.79	4.21

The temperature profiles of different temperature sensors (T1-T6) are depicted in Figure 9. Temperature sensors T7 (308 K) represent hot water circulator temperature. The temperature rise above the set point during initial sorption period attributes to the adsorption which is exothermic in nature and accompanied by liberation of heat. The temperatures T1 and T6 represent the top and bottom temperatures respectively, whereas the temperatures T2–T5 correspond to the intermediate column temperature from the top to bottom side. All the temperatures sensors are insulated type-K thermocouples. Due to the exothermic nature of adsorption, the mass transfer front is followed by a rise in temperature that is supported by temperature profiles at different positions. Also, it was observed that as the concentration of CO₂, the heat generated due to adsorption leads to raised temperature inside the adsorption column.

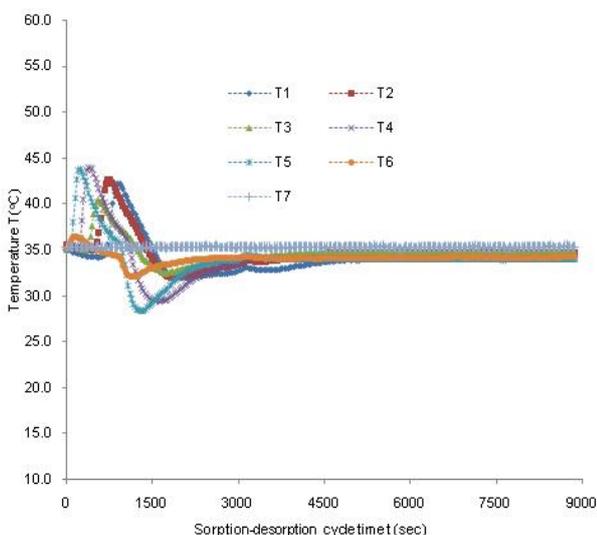


Figure 9. Temperature profiles inside the column of AC ($F=6.67 \times 10^{-5} \text{ m}^3/\text{sec}$)

The accuracy was judged using the repeatability measurement. The repeatability is measured by generating the two sets of data at 298 K and feed rate of $5.00 \times 10^{-5} \text{ m}^3/\text{sec}$. Closeness of the data depicted (Figure 10) indicates the reliability of the data. The R squared (R^2) has been found to be equal to 0.9962 and its value near to 1 signifies the good correlation among the repeated measurements. The data obtained for CO₂ adsorption is of high quality and reliable.

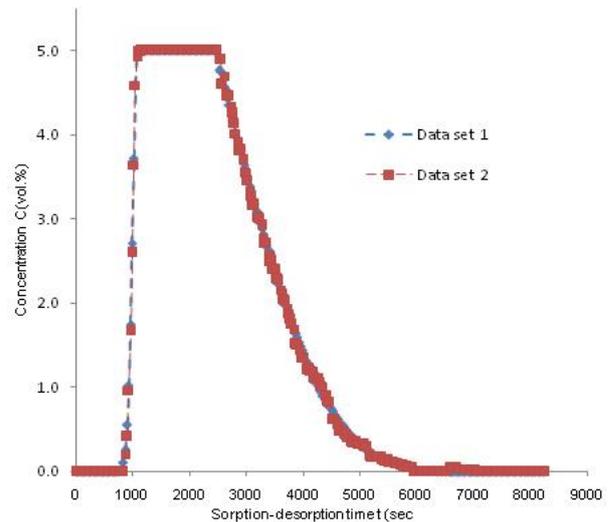


Figure 10. Repeatability measurement for AC ($T=298 \text{ K}$, $F=5.00 \times 10^{-5} \text{ m}^3/\text{sec}$, $C_0=5\%$)

3.3. Silica gel (SG)

The response curves obtained at different temperatures using Silica gel (Wt.=275 gm) are depicted in Figure 11. The feed rate was controlled at $6.67 \times 10^{-3} \text{ m}^3/\text{sec}$ with initial CO₂ concentration fixed at 0.05 (vol.%). The sorption periods alter significantly with bed temperature. The breakthrough and saturation time of 195 sec and 350 sec were estimated as a temperature of 298 K. The breakpoint time reduced to 152 sec on raising the temperature to 308 K with saturation time of 290 sec. The temperature equal to 318 K contributed to a declined breakpoint and saturation times of 122 sec and 240 sec. The prolonged breakthrough and saturation time which characterize the improved capacity was reported for AC compared to SG. Under the same conditions of temperature i.e. 298 K and feed rate ($6.67 \times 10^{-5} \text{ m}^3/\text{sec}$), the breakpoint periods of 870 sec and 195 sec exhibited for AC and SG respectively. The breakpoint time reported for AC is almost more than 4 times of that reported for SG at fixed temperature ($T=298 \text{ K}$) and feed rate ($F=6.67 \times 10^{-3} \text{ m}^3/\text{sec}$).

The influence of feed rates on response of CO₂ separation utilizing SG has been predicted in Figure 12. The prolonged breakthrough and saturation times of 250 sec and 425 sec are exhibited for SG adsorbent at a feed rate of $5.00 \times 10^{-5} \text{ m}^3/\text{sec}$. The breakpoint time declined to 195 sec on increasing the feed rate to $6.67 \times 10^{-5} \text{ m}^3/\text{sec}$. The saturation time of 370 sec was reported at a feed rate of $6.67 \times 10^{-5} \text{ m}^3/\text{s}$. The breakpoint and saturation time of 180 sec and

340 sec were achieved at a maximal feed rate of $8.33 \times 10^{-5} \text{ m}^3/\text{sec}$.

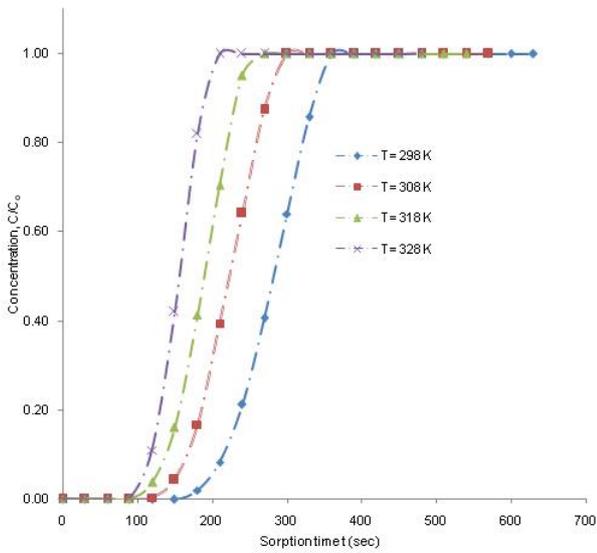


Figure 11. Sorption response at different temperatures for SG ($F=6.67 \times 10^{-3} \text{ m}^3/\text{sec}, C_0=5\%$)

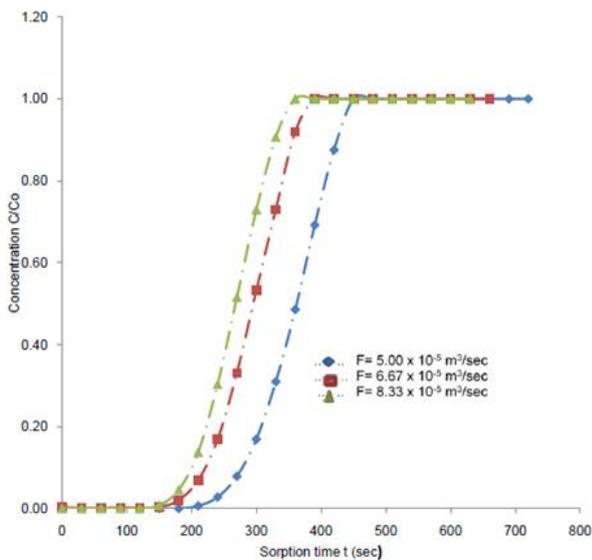


Figure 12 Sorption response at different feed rates for SG ($T=298 \text{ K}, C_0=5\%$)

The characteristics parameters of the CO_2 sorption determined from the breakthrough curves (Figures 11 and 12) for SG are depicted in Table 4. The breakthrough and saturation times reduce with increased temperature resulting in longer MSZ. Also, breakthrough and saturation time declines with increased feed flow rate resulting in increased length of MTZ. In general, the length of mass transfer zone (MTZ) increases with raised temperature and feed rate. The maximal values of $L_{\text{MTZ}}=15.65 \text{ cm}$ was obtained at a temperature of 318 K and feed flow rate of $6.67 \times 10^{-5} \text{ m}^3/\text{sec}$. The utilization factor decreases with increased temperature and feed rate. The maximal utilization factor $f=0.740$ has been determined at $T=298 \text{ K}$ and $F=5.00 \times 10^{-5} \text{ m}^3/\text{sec}$ for SG.

Table 4. Effect of operating conditions on characteristics parameters for SG

T (K)	F, $\times 10^{-5} (\text{m}^3/\text{sec})$	t_b (sec)	t_s (sec)	L_{MTZ} (cm)	f
298	6.67	195	350	13.65	0.706
308	6.67	152	290	14.99	0.688
318	6.67	122	240	15.65	0.674
328	6.67	105	206	15.59	0.675
298	5.00	250	425	12.44	0.740
298	8.33	180	340	14.77	0.692

The dependence of capacity on the temperature for SG is depicted in Figure 13. The data were generated at the superficial velocity of $6.67 \times 10^{-5} \text{ m}^3/\text{sec}$. The capacity adsorption reduced considerably with increased temperature. The maximal sorption capacity of $6.35 \text{ gm CO}_2/\text{Kg sorbent}$ was attained at a temperature of 298 K and it declined to $4.95 \text{ gm CO}_2/\text{Kg sorbent}$ on raising the temperature to 308 K. The temperature of 318 K contributed to a CO_2 capacity of $4.19 \text{ gm CO}_2/\text{Kg}$ at similar operating conditions. The capacity further reduced to $3.30 \text{ gm CO}_2/\text{Kg adsorbent}$ on raising the sorption temperature at 328 K. It was clearly demonstrated that the capacity of the adsorbable gas (CO_2) declined notably with increased temperature. The concentration of adsorbed gas or adsorption capacity lessened with increased temperature at a given equilibrium pressures. The capacity exhibited by the Silica gel (SG) is considerably lower compared to that obtained for activated carbon.

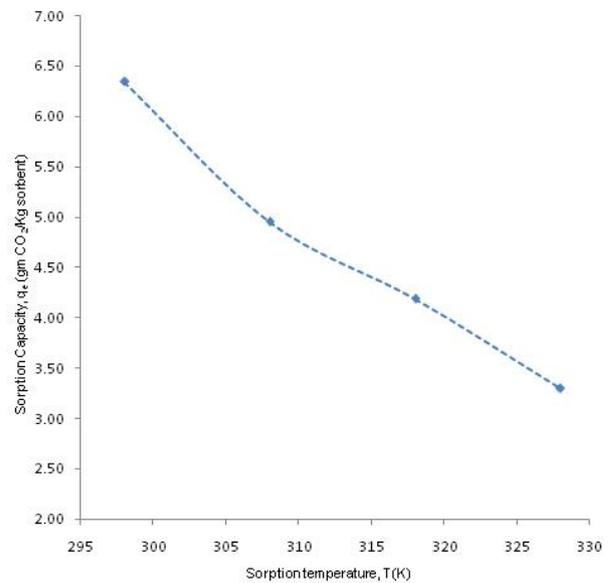


Figure 13. Sorption capacity vs. temperature curve for SG ($Q=6.67 \times 10^{-5} \text{ m}^3/\text{sec}, C_0=5\%$)

The adsorption isotherms produced at different temperatures for SG have been depicted in Figure 14. The capacity increased considerably with increased CO_2 pressure. The capacity of $4.63 \text{ gm /Kg sorbent}$ was determined at a carbon dioxide pressure of $1.0 \times 10^5 \text{ N/m}^2$ for isotherms generated at $T=298 \text{ K}$. At the same sorption temperature of $T=298 \text{ K}$, the minimal and maximal capacity of $0.20 \text{ gm CO}_2/\text{Kg sorbent}$ and $6.35 \text{ g CO}_2/\text{Kg sorbent}$ were attained at CO_2 partial pressures of $0.063 \times 10^5 \text{ N/m}^2$ and

1.250x10⁵ N/m² respectively. The capacity of 4.95 gm CO₂/Kg sorbent was attained at fixed partial pressure of 1.25x10⁵ N/m² with a temperature of 308 K. Similar trends of capacity variation with CO₂ partial pressure has been realized at an increased temperature of 318 K with a feed rate equal of 6.67 x10⁻⁵ m³/sec. The capacity further declined on raising the temperature to 328 K. The adsorption capacity of 1.0 gm CO₂/Kg sorbent was estimated at a CO₂ partial pressure of 0.5x10⁵ N/m² and further increased to 2.76 g CO₂/Kg sorbent on raising the partial pressure at 1.13 x10⁵ N/m².

maximal capacity equal to 5.91 gm CO₂/Kg sorbent was achieved at a CO₂ pressure of 1.25x10⁵ N/m². The capacity increased of 4.85 gm /Kg sorbent was determined at a CO₂ partial pressure of 1.0 x10⁵ N/m² and increased feed rate of 6.67 x10⁻⁵ m³/sec. The maximal capacity improved to 6.78 gm CO₂/Kg sorbent at a partial pressure of 1.25 X10⁵ N/m² under fixed rate of 6.67x10⁻⁵ m³/sec. Generally, the sorption capacity of the sorbent improved remarkably with increased feed flow of the CO₂/N₂. The dependence of the sorption capacity is more pronounced with adsorbables gas partial pressures compared to feed rates.

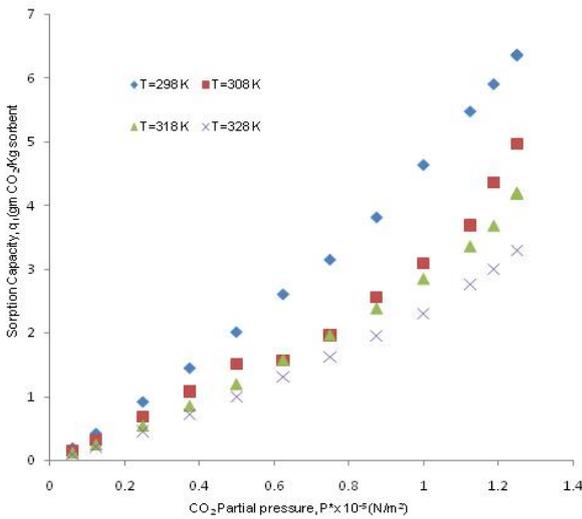


Figure 14. Sorption capacity vs. partial pressure curves for SG (F=6.67x10⁻⁵m³/sec)

The impact of the feed rate on the capacity of CO₂utilizing the SG from CO₂/N₂mixture has been depicted in Figure 15. The temperature was fixed at 298 K and the CO₂concentration was adjusted at 0.05 (vol. %). The capacity increases considerably with increased feed rate (N₂+CO₂).The capacity of adsorption of 5.91gm CO₂/Kg sorbent was realized at the feed rate of 5.00 x10⁻⁵ m³/sec. It was further increased to 6.78 gm CO₂/Kg sorbent on raising the feed rate to 6.67x 10⁻⁵ m³/sec. The maximal capacity equal to 7.64gm CO₂/Kg sorbent was attained at a feed rate of 8.33 x10⁻⁵ m³/sec.It was realized that as the increasing amount of feed (fluid) are passed through the adsorption column, the adsorbent adsorbs an increasing amount of solute (CO₂) from the feed gas mixture resulting in increased adsorption capacity. Therefore, it can be concluded that the adsorption capacity of the sorbent enhanced significantly with feed rate. The higher feed flow favors the increased adsorption capacity of the adsorbed gas from the feed mixture (N₂+ CO₂). It is suggested that higher feed flow is advantageous for achieving the higher adsorption capacity.

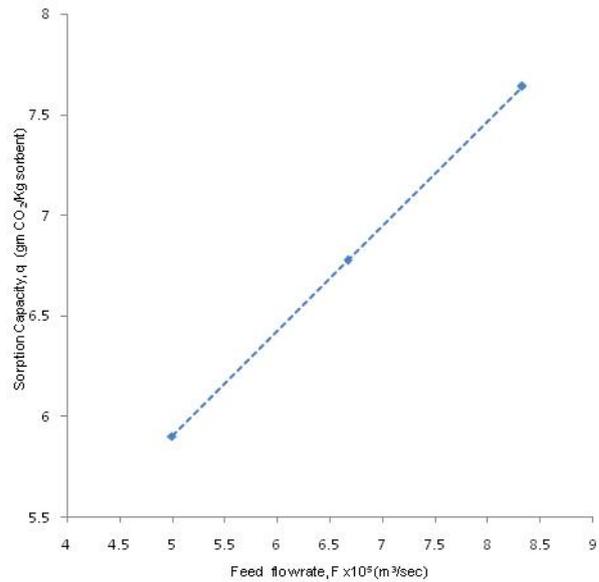


Figure 15 Sorption capacity vs. feed flow rate for SG (T=298 K)

The adsorption isotherms obtained at T=298 K at different feed rates have been depicted in Figure 16. The capacity improves with increased CO₂pressure. The minimal capacity of 0.188 gm CO₂/Kg sorbent was realized at a CO₂ pressure of 0.06 x10⁵ N/m² and the capacity increased to 1.49 gm CO₂/Kg sorbent on raising the pressure at 0.38 x10⁵ N/m²under constant feed rate of 5.00 x10⁻⁵ m³/sec. The

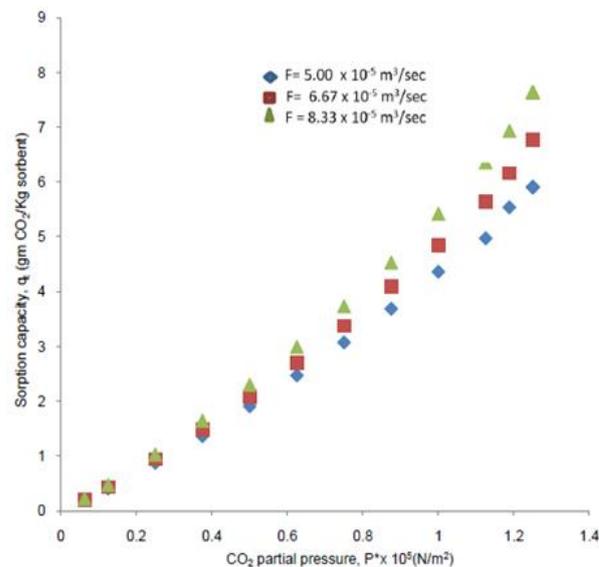


Figure 16. Sorption capacity vs. partial pressure for SG (T=298 K)

The characteristic parameters of the CO₂ capture for SG, i.e., effective column efficiency, usable bed and unusable bed height, were evaluated, as depicted in Table 5. The column efficiency, usable bed height and unutilized bed height depend on temperature and flow rate. The column

efficiency reduces with increased temperature and feed rate and the highest efficiency of 58.82 % was determined for SG at 298 °C and $F=5.00 \times 10^{-5} \text{ m}^3/\text{s}$. The increased feed rate $5.00 \times 10^{-5} \text{ m}^3/\text{s}$ to $8.33 \times 10^{-5} \text{ m}^3/\text{s}$ contributed to enhanced column efficiency and adsorption capacity of 52.94 % was obtained at flow rate of $8.33 \times 10^{-5} \text{ m}^3/\text{s}$ with 298 K and $C_o=5\%$. The efficiency based on column capacity determined for AC is significantly higher than the efficiency obtained for SG.

The usable and unutilized bed heights also depend on the temperature and flow rate of feed mixture. The maximum usable bed height for AC is determined equal to 14.12 cm for total effective column height of 24 cm at 298 K and $5.00 \times 10^{-5} \text{ m}^3/\text{s}$. The usable bed height or length of bed used upto the breakthrough reduces with increased temperature and feed rate. The temperature of 318 K and feed rate $F=6.67 \times 10^{-5} \text{ m}^3/\text{s}$ contributed to minimal usable bed height of 12.19 under different set of conditions. The highest unutilized bed height of 11.81 cm was evaluated at a temperature of 318 K and $F=6.67 \times 10^{-5} \text{ m}^3/\text{s}$. The can be suggested that characteristic performance parameters of AC are better in comparison to that determined for SG under similar operating conditions.

Table 5. Characteristics parameters of CO₂ capture for SG

T(K)	F, $\times 10^{-5}$ (m ³ /s)	C _o (vol.%)	η (%)	H _b	H _{ub}
298	6.67	5	55.71	13.23	10.77
308	6.67	5	52.41	12.58	11.42
318	6.67	5	50.83	12.19	11.81
328	6.67	5	50.97	12.23	11.77
298	5.00	5	58.82	14.12	09.88
298	8.33	5	52.94	12.71	11.29

4. Conclusions

The sorption breakthrough time relies strongly on the sorption temperature and reduces with increased temperature. The maximal breakthrough and saturation periods of 870 sec and 1050 sec were exhibited for AC at a bed temperature of 298 K. The prolonged breakthrough and saturation times have been realized for AC compared to SG under constant operating conditions. The prolonged breakthrough time results in increased adsorption capacity. The breakthrough curves of AC are vastly steep signifying the maximal utilization of bed capacity at the breakpoint. Narrow MSZ characterize efficient use of the sorbent resulting in the reduction of costs of energy regeneration. The breakthrough and saturation times reduce significantly with increased feed rate and the prolonged breakpoint time of 1145 sec was attained at a feed rate of $5.00 \times 10^{-5} \text{ m}^3/\text{sec}$ for AC and these times are considerably higher than the values obtained for SG. In general, the length of MTZ increases with raised temperature and feed flow rate and maximal length of MTZ

equal to 5.78 cm was determined at $T=328 \text{ K}$ and $F=6.67 \times 10^{-5} \text{ m}^3/\text{sec}$ for AC. The utilization factor reduces with increased temperature and feed rate. The maximal utilization factors of 0.919 and 0.740 were determined at $T=298 \text{ K}$ and $F=5.00 \times 10^{-5} \text{ m}^3/\text{sec}$ for AC and SG respectively signifying the relevance of AC for CO₂ separation. The CO₂ capacity strongly depends on the temperature and significantly reduces with an increased in bed temperature. The maximal capacities of 32.99 gm CO₂/Kg sorbent and 6.35 gm CO₂/Kg sorbent were determined at a temperature of 298 K for AC and SG respectively. The capacity improves considerably with raised pressure of the CO₂ and AC exhibited higher sorption capacity compared to SG at a lower adsorption temperature of 298 K and flow rate of $6.67 \times 10^{-5} \text{ m}^3/\text{sec}$. The adsorption capacity increases considerably with increased feed rates ranging from $5.00 \times 10^{-5} \text{ m}^3/\text{sec}$ to $8.33 \times 10^{-3} \text{ m}^3/\text{sec}$ and maximal capacity of 39.14 g CO₂/Kg sorbent was determined for AC. The maximum column efficiency and usable bed height of 85.13% and 20.43 cm were determined for AC, respectively. The AC may be used for economical separation of CO₂ from N₂/CO₂ mixture owing to higher sorption capacity and utilization factor.

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