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CARBON DIOXIDE CAPTURE FROM FLUE GAS BY PRESSURE SWING ADSORPTION USING CARBON MOLECLAR SIEVE SORBENT

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ABSTRACT: - Emissions of carbon dioxide CO_2 from power plant stations have direct effect on global climate change through increasing the average temperature which then threat the human life. Thus it is essential to reduce the CO_2 emissions to the allowed level. Pressure swing adsorption (PSA) process is one of the efficient and economic processes for capturing the CO_2 from flue gases. In the present study two columns 6-step PSA process packed with carbon molecular sieve (CMS) was used to study the effect of adsorption pressure, purge flow rate, and cycle time on the process performance (In term of CO_2 purity). The results showed that the CO_2 purity decreased in the product line and increased in the purge line with increases of the adsorption pressure. Increasing purge flow rate from 0.5 lit/min to 2 lit/min at the range of all cycle time and different adsorption pressures led to decrease in CO_2 emissions. The CO_2 purity decreased with increases of cycle time up to 80 second and then increased slightly at 100 second in spite of increasing of pressure to 4 bar. The CO_2 purity was about 0.7% in the product line, and 38% in the purge out line.
Keywords: Capture of CO_2, Pressure Swing Adsorption, Carbon Molecular Sieve.

1- INTRODUCTION

Carbon dioxide is one of the major combustion products which once released into the air can contribute to the global climate warming effects[1, 2]. With the concern over global warming, CO_2 capture and sequestration has always been the subject of interest in research discussion about global issues [3]. The efficient methods for CO_2 capture is an important target for human society [4]. Several methods are used for CO_2 capture such as; adsorption, membranes, mono ethanolamine chemical absorption, cryogenic distillation. Pressure swing adsorption with two columns is always used for air separation at 1960s. The process is first developed by Skarstrom with four steps and a very important improvement introduced was the equalization step to improve the process performance [3, 5-7]. CO_2 capture by adsorption process is a promising option for many advantages such as; low energy requirement, low capital cost and easy to achieve automated operation [4]. The adsorption process differ from other separation processes, it's based on preferential adsorption of the desired gas (e.g., CO_2) at high pressure, and recovery it at low pressure. The porous sorbent can be reused for subsequent adsorption. Adsorbents of high selectivity, and high adsorption capacity would be reduced the cost of CO_2 separation [8, 9].

Carbon molecular sieves (CMS) is one of the activated carbons family and it can be obtained by various procedures leading to pores narrowing to smaller sizes than 10 Å. Pores narrowing increases diffusion speed for smaller molecules as compared to the larger, which resulting in a fast gas separation caused by the differences in molecules sizes that provide the relatively high adsorption capacity and kinetic selectivity for various molecules [10, 11].

Chue et al. (1995) [12] are used two adsorbent materials zeolite 13X and activated carbon for CO_2 capture by pressure swing adsorption. High purity of CO_2 (over 99%) was

produced by 13X compare to the activated carbon. Chou et al (2004) [13] and Jianghua et al (2012) [14] are studied simple vacuum swing adsorption with two beds and three beds packed with zeolite 13X employed to capture CO_2 from flue gas. The purity of CO_2 was about 46% for two beds. The purity increased to about 85% with increase of feed composition from 20% to %50%. The purity was 63% for three beds and the purity at the product line stream was about 5% with feed composition of 20% CO_2 and 80% N_2. Reynolds, S. P. , et al. (2005) [15] adopt typical Skarstrom cycle (4-ateps), vacuum swing cycle with promoted adsorbent constructed to process a stack gas effluent at 575 K containing 15% CO_2, 75% N_2 and 10% H_2O. The effects of the cycle time purge/feed ratio, and pressure ratio on the process performance explored. The CO_2 purity increased at enrichment stream with increase *cycle time*, pressure ratio, and decreasing purge to feed ratio.

Park et al. (2002) [16] and Grande et.al (2013) [17] are studied the recovery of CO_2 from flue gas used two-stage and two different cycle (4-steps cycle and 5-steps cycle) pressure swing adsorption (PSA) process. At the first stage, CO_2 concentrated to (40−60) % and then concentrated to 99% in the second stage. For 4-steps cycle the purity at rich-stream line never exceeded than 52%, but with 5-steps the purity increased up to 80%.

Xiao et al. (2008) [3]used vacuum swing adsorption to capture of CO_2 from flue gas streams by adsorbent of 13X. A 9-step cycle and a 12-step cycle were both used to investigate the effect of cycle on performance. Longer feed times led to partial breakthrough of the CO_2 front and improve purity. A 12-step cycle was able to increase the purity of CO_2 over 95%. Dantas et al (2012) [18] worked on the experimental setup of pressure swing adsorption consisted of single fixed-bed adsorption and then used to simulated the operation of a unit with several fixed-beds, The commercial activated carbon was used as a adsorbent and the feed included the mixture of CO_2 and N_2 as 15%. There is an increase in the carbon dioxide purity from 25% to 50% (decreases at the product stream) with increasing feed time from 100s to 120s. This indicates that the separation is strongly controlled by the equilibrium.

In this work two column 6-steps pressure swing adsorption unit packed with commercial carbon molecular sieve was used to study the effect of operation conditions such as adsorption pressure, cycle time, and gas purge flow rate on process performance through measuring the purity of the CO_2 at the product stream line and purge stream line.

2- EXPERIMENTAL WORK

Two galvanized column of 2.54 cm in diameter and 100 cm in length packed with carbon molecular sieve (CMS) were used in the experiments. Table (1) presents the physical properties of the carbon molecular sieve used in the experimental work. The feed was prepared at the laboratory using two cylinders one for pure nitrogen and another for pure carbon dioxide. Each gas cylinder was connected to pressure regulator and with gas rotameter to adjust the pressure and the flow rate to prepare a feed contains 15% CO_2 in the feed tank storage. The adsorption pressure was controlled by another pressure regulator (1 to 10 bar, Norgren England) which was installed on the feed stream and then connected to the (PSA) process according to skarstrom cycle (pressurizing, adsorption, depressurizing, and purging) with equalization step. The operation time of the solenoids valve (CASTEL, Italy's manufacture, 220v, 50Hz) was controlled by homemade controller, as shown in table (2). All connections were 12 mm in diameter. Purge and product flow rate adjusted by two gas rotameters (1 – 6 lit/min, Norgren England), and (0.1-1 lit/min, OMEGA, manufactured in Tokyo- Japan), respectively. The purity of the CO_2 was measured by CO_2 infrared analyzer (G110, Geotechnical Company, United Kingdom) after the system reaching steady state. Figure (1) shows the experimental setup of the pressure swing adsorption.

Operation conditions used in the present work were:

- Cycle time = 60, 80, and 100 s.
- Adsorption pressure, P_H = 1 to 4 bar.

- Purge flow rate, Q_{purg}= 0.5 to 2 l/min.
- Product flow rate, Q_{prod}=0.5 l/min.
- Equalization time, t_{eq}= 10 sec.

Experimental procedure was included the following procedure:

- Set the pressure regulator of each cylinder to pressure greater than adsorption pressure.
- Prepare the feed (15% CO_2) by adjusting two gas rotameter and then measure it in the feed storage tank before starting the run.
- Set the feed pressure regulator.
- Switch on the controller
- Set the purge and product flow rates.
- Record the purity of CO_2 after the system reaching steady state.

3- RESULTS AND DISCUSSION

3.1 Effect of adsorption pressure

Figures (2,3 and 4) represent the effect of the adsorption pressure on the PSA process performance to capture of CO_2 at cycle time of 60 ,80 and 100 second, respectively and purge flow rate in the range of 0.5 to 2 lit/min. It can be seen that the performance of the PSA process increased with increase in adsorption pressure up to 4 bar at cycle time of 60 second. This may be due to the increasing adsorption capacity of adsorbent with increasing of the adsorption pressure. The CO_2 purity at the product line decreased to 2%. The purity CO_2 plateaus or no significant effect of adsorption pressure on the performance over the pressure of 3 bar. At the cycle time of 80 second the CO_2 purity dropped to 0.7% at the product line stream. Furthermore, no significant effect of the adsorption pressure over 2 bar at the cycle time of 100 second and the purity was about of 1.6%. This may be due to the inter relationship between the adsorption capacity and the adsorption pressure. The little increasing in the adsorption capacity with increasing adsorption pressure over 2 or 3 bar was unable to adsorbe all CO_2 and then the purity plateaus or increased. The optimum adsorption pressure of 3 bar and cycle time of 80 second was noticed. This may be due to the high adsorption capacity of the carbon molecular sieve to adsorb a lot of CO_2 during pressurizing and producing step. The CO_2purity at the product line stream is better than purity observed by both Chou et al. (2004) [13] and Jianghua et al. (2014) [14]

3.2 Effect of purge flow rate

Figures (5, 6 and 7) show the effect of purge flowrate on CO_2 purity at the product line at different adsorption pressure and cycle time. The CO_2 purity minimized with increase in purge flow rate from 0.5 lit/min to 2 lit/min at all cycle times. This may be attributed to increase the gas requirement to regenerate the carbon molecular sieve and prepare it for new cycle. Figure 6 shows that there is no significant effect of purge flow rate over 1 lit/min at the pressure of 1 bar due to the limited capacity of the adsorbent at the 1 bar. The increasing of purge flow rate led to no improve in the CO_2 purity or slightly decreases noticed. It's approximately the same trend for figure 7 at the pressure of 1 bar. The optimum condition was noticed between (1.5-2) lit/min at all cycle times.

3.3 Effect of cycle time

Figures (8 and 9) show the effect of the cycle time on CO_2 purity at different purge flow rate. It can be observed that the CO_2 purity drops at the product line stream with increasing of cycle time from 60 to 80 second at the adsorption pressure (1 – 2) bar. This may be attributed to increase the time requirement to adsorbe a lot of CO_2 and then the low CO_2 purity noticed. The increasing of cycle time over 80 second led to make the CO_2 purity plateaued, because of increasing amount of feed per cycle time and the adsorbent materials was unable to adsorb all

CO_2 molecular there for the purity was stable or a little increasing observed at the product line. This trend is in agreement with Dantas (2012) [18].

Figure (10, 11) show that the CO_2 purity increased at the product line with increase in cycle time over 80 second at the different purge flow rate, in spite of increasing of adsorption pressure to 3 and 4 bar. This is due to the breakthrough of the CO_2 front and increasing the purity at the product line. The optimum cycle time was 80 second at the purge flowrate of 2 lit/min.

3.4 Effect of pressure on the purge stream purity

Figure (12) represents the effect of the adsorption on the CO_2 purity at the product line and purge stream line purity. The results showed that the CO_2 purity declined at the product line with increase in adsorption pressure up to 4 bar. The CO_2 purity increases at purge out with increasing of pressure to 4 bar, due to the increasing of adsorption capacity led to increase the CO_2 concentrate at the solid phase according to equilibrium theory and release it to bulk during depressurizing step therefor the purity increased to about 38% at the purge line stream. The results are in agreement with Steven et al. (2005) [15], Jong-Ho et al. (2002) [16], and Carlos et.al (2013) [17].

4- CONCLUSION

The results showed that the minimum purity of the CO_2 at the product line was 0.7% at adsorption pressure of 3 bar, cycle time of 80 second, and purge flow rate of 2 lit/min. No significant effect of adsorption pressure over 3 bar on the CO_2 purity at the product line. The optimum pressure was between 3 and 4 bar, and optimum cycle was 80 second. There is significant effect of purge flow rate on the CO_2 purity at all conditions. The maximum purity of the CO_2 obtained at the purge line was about 38.6%.

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Table (1): Physical properties of the carbon molecular sieve

Type	CMS
Company	Pingxiang XingFeng Chemical Packed
Shape	Granular
Particle diameter d_p (mm)	1.7 - 1.8
Bulk density ρ_B (g/L)	680-700
Bed porosity	0.37

Table (2): Status of the Solenoid valves during operation for 6-steps cycle process**.**

Steps	Column steps		Valve Status				
	bed 1	Bed 2	V_1	V_2	V_3	V_4	V_5
1	Pressurizing	Depressurizing	Open	Close	Open	Close	Close
2	Producing	Purging	Open	Close	Open	Close	Close
3	Equalization	Equalization	Close	Close	Close	Close	Open
4	Depressurizing	Pressurizing	Close	Open	Close	Open	Close
5	Purging	Producing	Close	Open	Close	Open	Close
6	Equalization	Equalization	Close	Close	Close	Close	Open

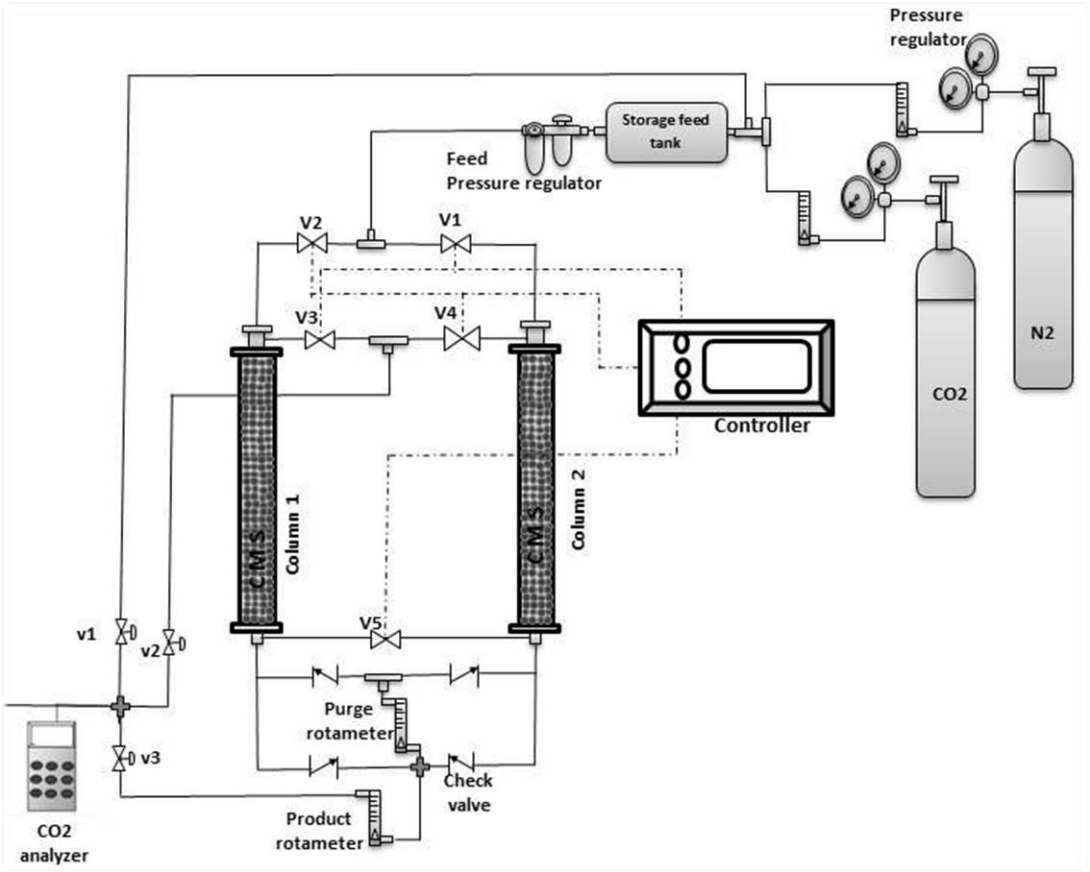

Figure (1): Experimental set up of two beds pressure swing adsorption

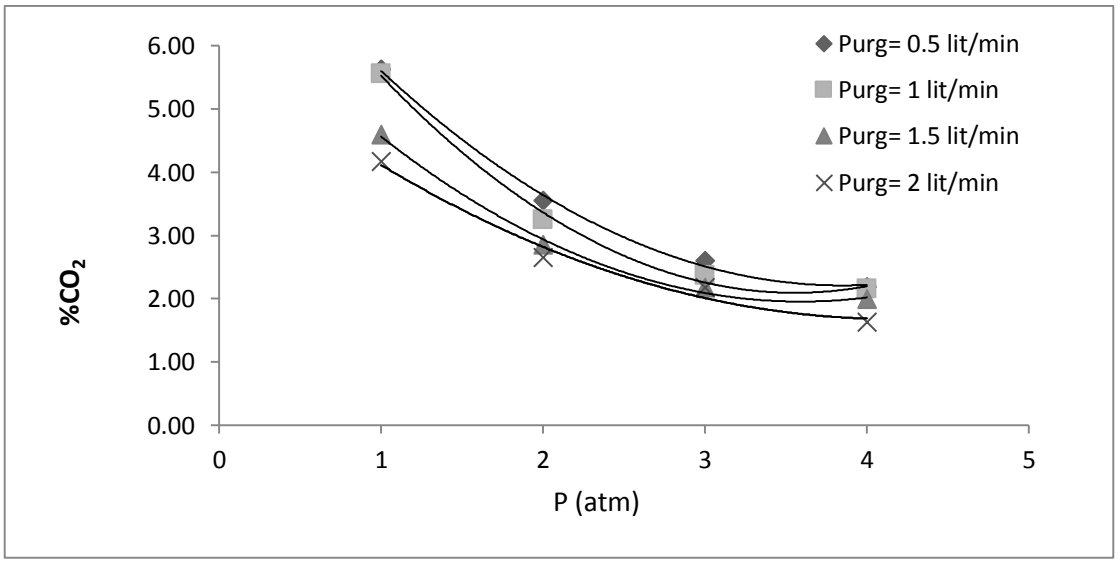

Figure (2): Effect of adsorption pressure on CO_2 purity at cycle time 60 second.

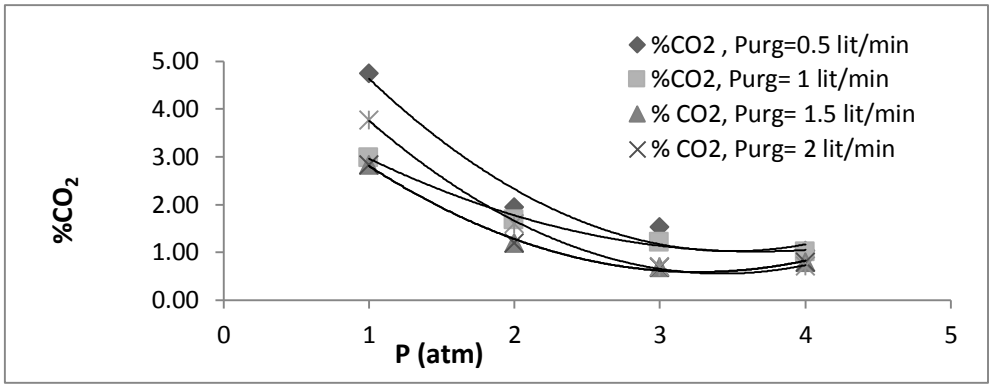

Figure (3): Effect of adsorption pressure on CO_2 purity at cycle time 80 second.

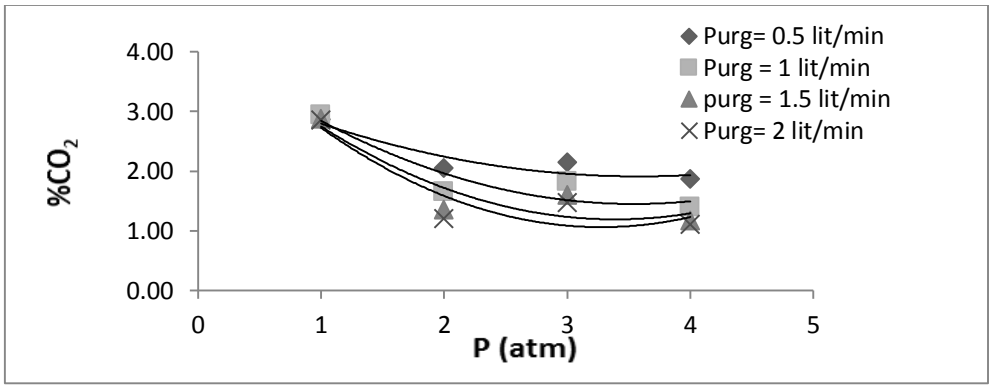

Figure (4): Effect of adsorption pressure on CO_2 purity at cycle time 100 second.

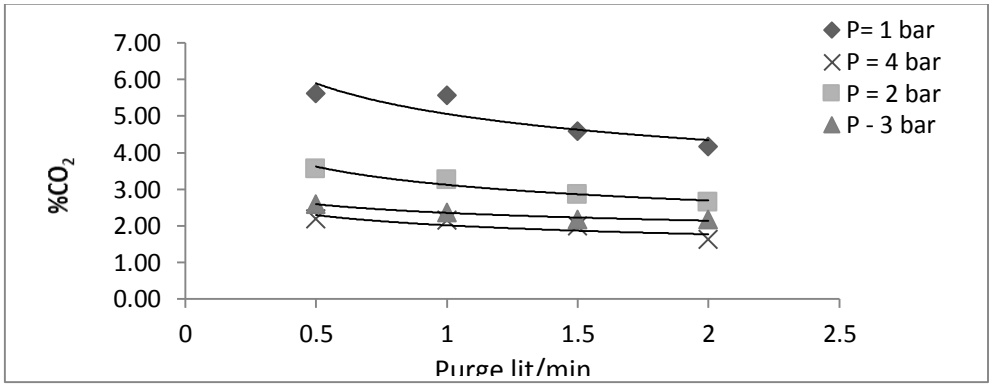

Figure (5): Effect of purge flow rate on CO_2 purity at cycle time of 60 second.

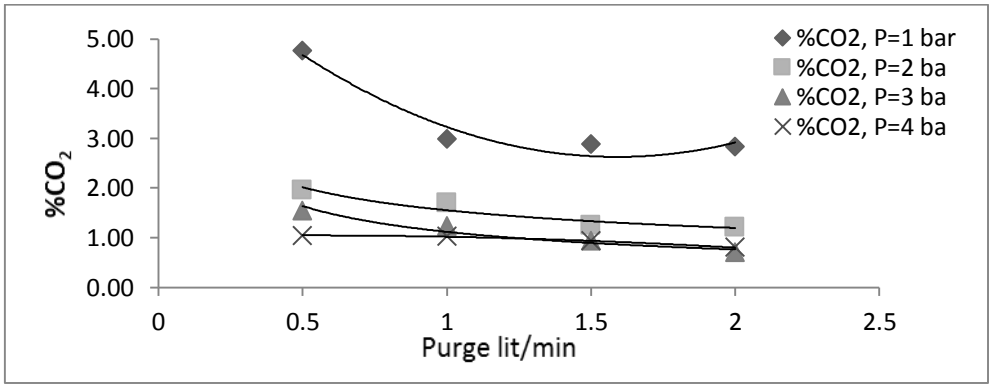

Figure (6): Effect of purge flow rate on CO_2 purity at cycle time of 80 second.

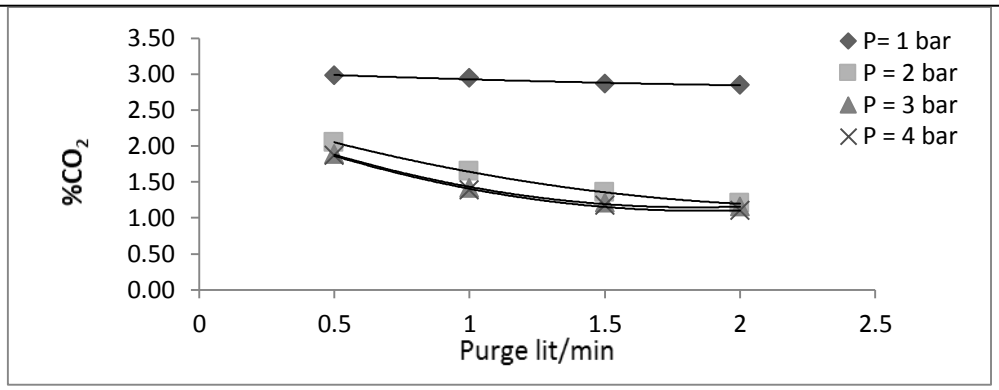

Figure (7): Effect of purge flow rate on CO_2 purity at cycle time of 100 second.

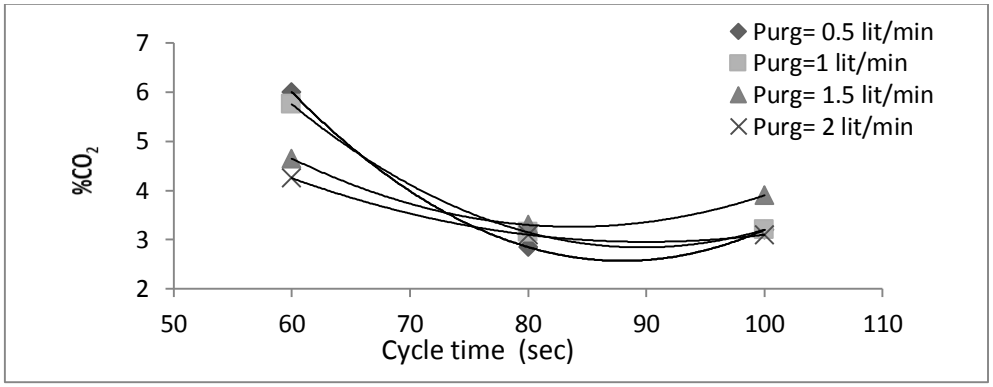

Figure (8): Effect of cycle time at the adsorption of 1 bar.

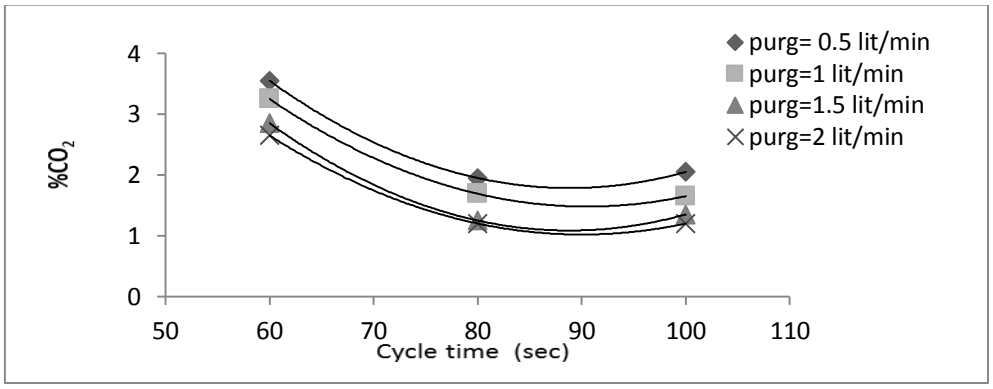

Figure (9): Effect of cycle time on CO_2 purity at the adsorption of 2 bar

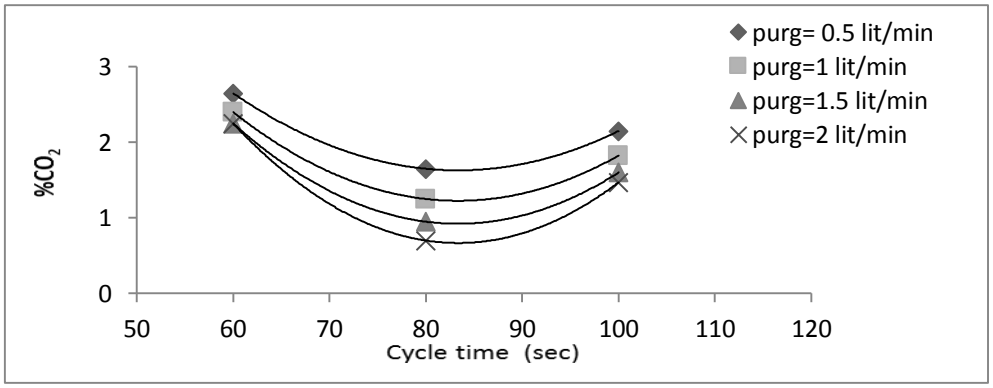

Figure (10): Effect of cycle time on CO_2 purity at the adsorption of 3 bar

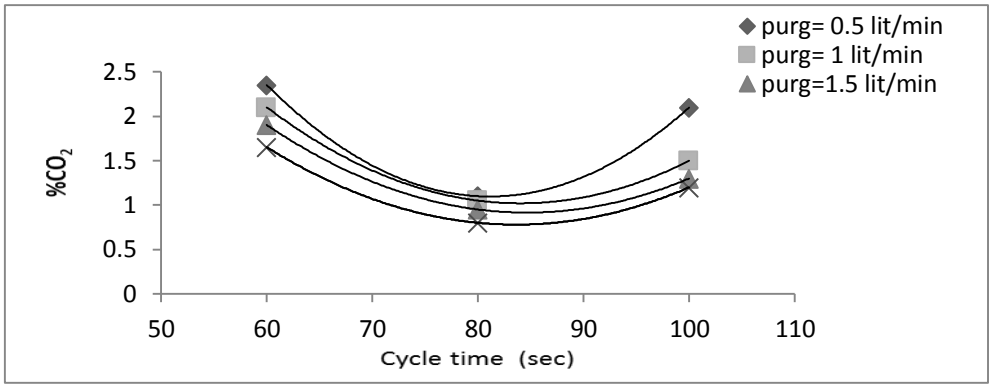

Figure (11): Effect of cycle time on CO_2 purity at the adsorption of 4 bar

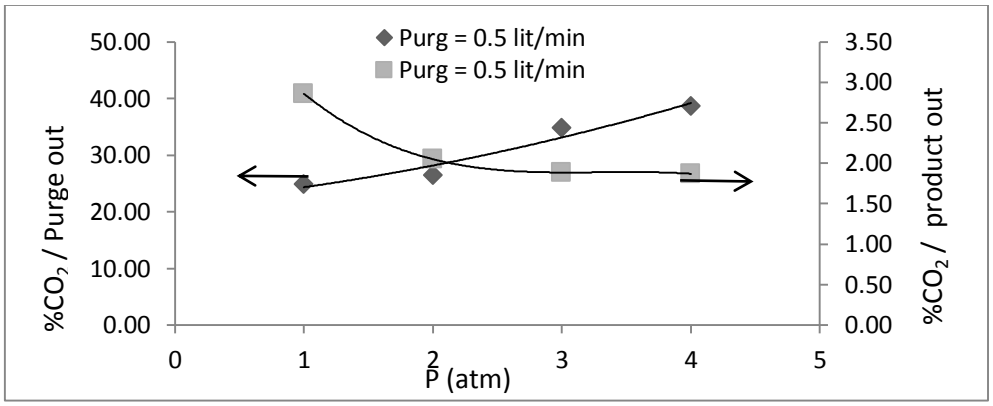

Figure (12): Effect of pressure on the purge out line and product out line CO_2 purity**.**

التقاط غاز ثنائي اوكسيد الكاربون من غاز المداخن بعملية الامتزاز بضغط متأرجح باستخدام جزيئات الكاربون المنخلي كمادة مازة

الخلاصة:

إنبعاثات غاز ثنائي اوكسيد الكاربون من محطات توليد الطاقة لها تأثير في تغيير المناخ العالمي من خلال زيادة معدل درجة حرارة الارض, والتي تهدد حياة البشرية. وبالتالي من الضروري خفض انبعاثات ثنائي اوكسيد الكاربون إلى المستوى المسموح به. عملية الامتزاز بضغط متأرجح هي احدى العمليات الكفوءة والاقتصادية لألتقاط غاز ثنائي اوكسيد الكاربون من غازات المدخنة. الدراسة الحالية استخدمت عمودان للأمتزاز بست خطوات تشغيل مملوءان بـ . جزيئات الكاربون المنخلي لدراسة تأثير ضغط الامتزاز ,معدل تدفق غاز اعادة التنشيط و زمن الدورة على اداء العملية من خلال قياس نقاوة غاز ثنائي اوكسيد الكاربون. النتائج بينت بأن النقاوة تناقصت في خط الانتاج وتزايدت في خط تدفق غاز التنشيط الخارج بزيادة ضغط الامتزاز. زيادة تدفق غاز التنشيط من 0.5 لتر/دقيقة الى 2 لتر/دقيقة على مدى كل الدورات وبضغوط امتزاز مختلفة ادت الى تناقص انبعاث غاز تنائي اوكسيد الكاربون. نقاوة غاز ثنائي اوكسيد الكاربون تناقصت بزيادة زمن الدورة حتى 80 ثانية ومن ثم تزايدت قليلا عند 100 ثانية بالرغم من زيادة الضغط الى 4 بار. نقاوة غاز ثنائي اوكسيد الكاربون كانت بحدود 0.7% في خط الانتاج و 38% في خط تدفق غاز التنشيط الخارج.

الكلمات الدالة : التقاط CO2 , امتزاز بضغط متأرجح, و جزيئات الكاربون المنخلي.