Study the Behavior of Long Spiral Tube Adsorber for Oxygen Separation from Air

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Received on: 27 /9/2012 & Accepted on: 11/6/2013

ABSTRACT

Single long spiral tube column (25 mm diameter, and 4 m bed length) had been constructed to study the separation of oxygen from air using commercial 13X zeolite. The effect of adsorption pressure on the system breakthrough curves was studied. Single column with initial air pressurizing simulates the work of 2-columns, 4-steps PSA process, whereas single column with initial intermediate pure oxygen pressurizing simulates the work of 2-columns, 6-steps PSA process with pressure equalization steps of the two columns. No significant effect of pressure on the product oxygen purity is noticed when pressure increased from 2 to 5 bar in both cases.

For initial air pressurizing case, the average maximum effluent oxygen purity of 88% is obtained. The range of zeolite loading capacity is q=0.25-0.35 mole N_2/kg zeolite, and only 40% of the range has been utilized before breakthrough time. Whereas for initial oxygen pressurizing case, the maximum oxygen purity of 95% is obtained. The range of zeolite loading capacity is q=0.39-0.87 mole N_2/kg zeolite, and 95% of the range has been utilized before breakthrough time, which agree well with the equilibrium data of multicomponent Langmuir adsorption equation.

Keywords: Spiral Tube Adsorber, Breakthrough, Oxygen Separation, PSA.

دراسة تصرف انبوب الامتزاز الملفوف الطويل لفصل الاوكسجين من الهواء

الخلاصة

تم تصنيع عمود ملفوف طويل (قطر ٢٥ ملم وطول ٤ م) لدراسة فصل الاوكسجين من الهواء باستخدام الزيولايت التجاري 13X . ودرس تاثير ضغط الامتزاز ومعدل الجريان من خلال منحنى

https://doi.org/10.30684/etj.31.17A.13
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الاختـراق. يحـاكي العمـود المفـرد فـي حالـة بـدء الضـغط بـالهواء ، عمليـة الامتـزاز بتغيـر الضـغط لعمودين واربع خطوات ، بينمـا يحـاكي فـي حالـة بـدء الضـغط بالاوكسـجين ، عمليـة الامتـزاز بتغيـر الضغط لعمودين وست خطوات مـن ضـمنها خطـوتي تسـاوي الضـغط فـي العمـودين. لـم يلاحـظ اي تاثير لضغط الامتزاز من ٢ الى ٥ بار على نقاوة الاوكسجين في الحالتين.

فـي حالـة بـدء الضـغط بـالهواء ، كانـت اقصـى نقـاوة للاوكسـجين هـي ٨٨%. وكـان حـدود سـعة الزيولايت ($q=0.25-0.35$ mole N_2/kg zeolite) وان ٤٠% فقط يستفاد منها قبل زمن الاختـراق. بينما في حالة بدء الضغط بالاوكسجين ، كانت اقصـى نقاوة للاوكسجين هي ٩٥%. وكان حدود سعة الزيولايـت ($q=0.39-0.87$ mole N_2/kg zeolite) وان ٩٥% يسـتفاد منهـا قبـل زمـن الاختـراق، وهي متوافقة بصورة جيدة مع معلومات الاتزان في معادلة لانكمور للامتزاز.

INTRODUCTION

Design of single fixed bed adsorption column often relies on the concept of mass transfer zone (MTZ). Figure (1) shows the progress of the zone through the bed [1].

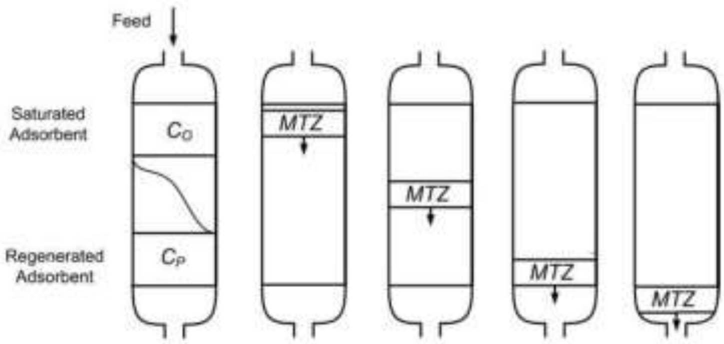

Figure (1) Progress of the mass transfer zone (MTZ) Through a fixed bed adsorption column [1].

The breakthrough time t_b is always less than the ideal adsorption time t_s, and the actual amount of solute adsorbed at the break point can be determined by integrating the breakthrough curve up to the time t_b, as shown in Figure (2). If the mass transfer zone is narrow relative to the bed length, the breakthrough curve will be rather steep as in Figure (2a), and most of the capacity of the solid will be utilized at the break point. When the mass transfer zone is almost as long as the bed, the breakthrough curve is greatly extended, as Figure (2b), and less than half of the bed capacity is utilized. A narrow mass transfer zone is desirable to make efficient use of the adsorbent and to reduce the energy costs in regeneration. In the ideal case of no mass transfer resistance and no axial dispersion, the mass transfer zone would be of infinitesimal width and the breakthrough curve would be a vertical line from 0 to 1.0 when the entire solid was saturated [2].

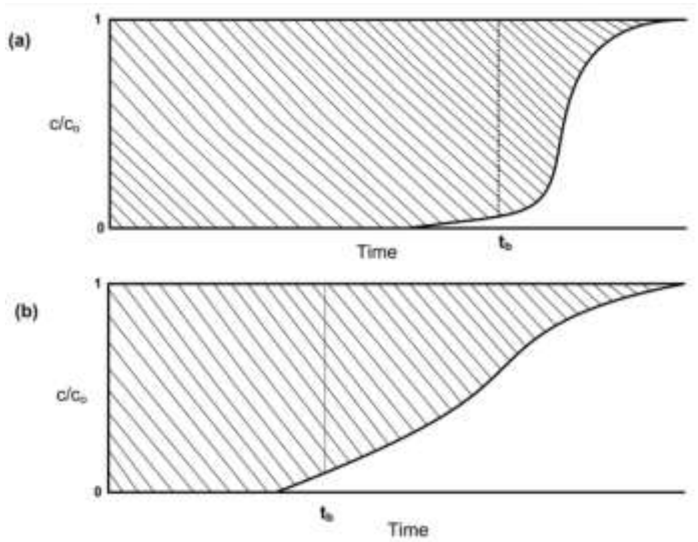

Figure (2) Breakthrough curve for; (a) narrow and (b) wide Mass transfer zone (MTZ) [2].

Single component Langmuir adsorption equilibrium equation for oxygen separation from air is written as follows [3]:

$$\frac{q_i}{q_{si}} = \frac{b_i P_i}{1 + b_i P_i} \qquad \text{… (1)}$$

Whereas the multi-component Langmuir adsorption equilibrium equation is written as follows [3]:

$$\frac{q_i}{q_{si}} = \frac{b_i P_i}{1 + \sum b_j P_j} \qquad \text{… (2)}$$

Pressure swing adsorption (PSA) with dual columns is usually used for oxygen separation from air [4, 5]. The process is first developed by Skarstrom [6]. Four steps were used in dual bed unit, pressurization with air feed, producing with continuous air feed through one end with product withdrawal through the other, depressurization countercurrent to the feed, and purging performed by product flow countercurrent to feed [3].

A very important improvement was the introduction of the pressure equalization step of the two columns [7-9]. The pressure equalization, using a product-enriched current, leads to a significant economy in energy consumption, since less mechanical energy is required to re-pressurize the column at low pressure, after the purge stage. The product recovery is, in this way, also increased because less feed gas is necessary to re-pressurize the column [10].

In the present work, a single column with air initial pressurizing simulates the work of 2-column, 4-steps PSA process, whereas a single column with initial intermediate pure oxygen pressurizing simulates the work of 2-columns, 6-steps PSA process with pressure equalization steps of the two columns.

The aim of the present work is to construct a small scale single long spiral bed column, packed with commercial 13X zeolite, and to investigate the characteristic of the single column, through the analysis of the breakthrough curves at different adsorption pressures.

EXPERIMENTAL WORK

Figure (3) shows the experimental set-up of the single column characteristics. The spiral column of Copper of 2.54 cm in diameter and 4 m in length, with fittings and connections 12mm in diameter, pressure regulator (1 to 10 bar, max. temp. 120 ºC, Norgren England), the solenoid valves of 6 mm size. A rotameter is used for the effluent stream (1 l/min, Oxygen). Each column contains 1300 g of zeloite 13X, the characteristic of the adsorbent is shown in Table 1. Oxygen analyzer type is GOX 100 Greasing Electronic GmbH. Table 2 shows the operating conditions of the single column characteristics.

The experimental procedure was:

1. Preparation of the system shown in Figure (3), using vacuum and O_2 purging to ensure the zeolite activity.
2. Adjust the air feed pressure by pressure regulator.
3. Adjust the system condition with or without initial pure O_2 intermediate pressure of the column to the desired value.
4. Open the air feed valve and open the output valve and adjust the flow rate to the desired value (1 liter/min), using a gas rotameter and a regulating valve.
5. Recording the product purity (O_2 %) measured by the analyzer with time.
6. Calculation of q and q_B as the area under the curve of O_2 purity versus time according the following equation [11]:

$$q = (Q_{eff} / 22.4w)(79/21)\sum(y_{O2} - 0.21)\Delta t \quad (3)$$

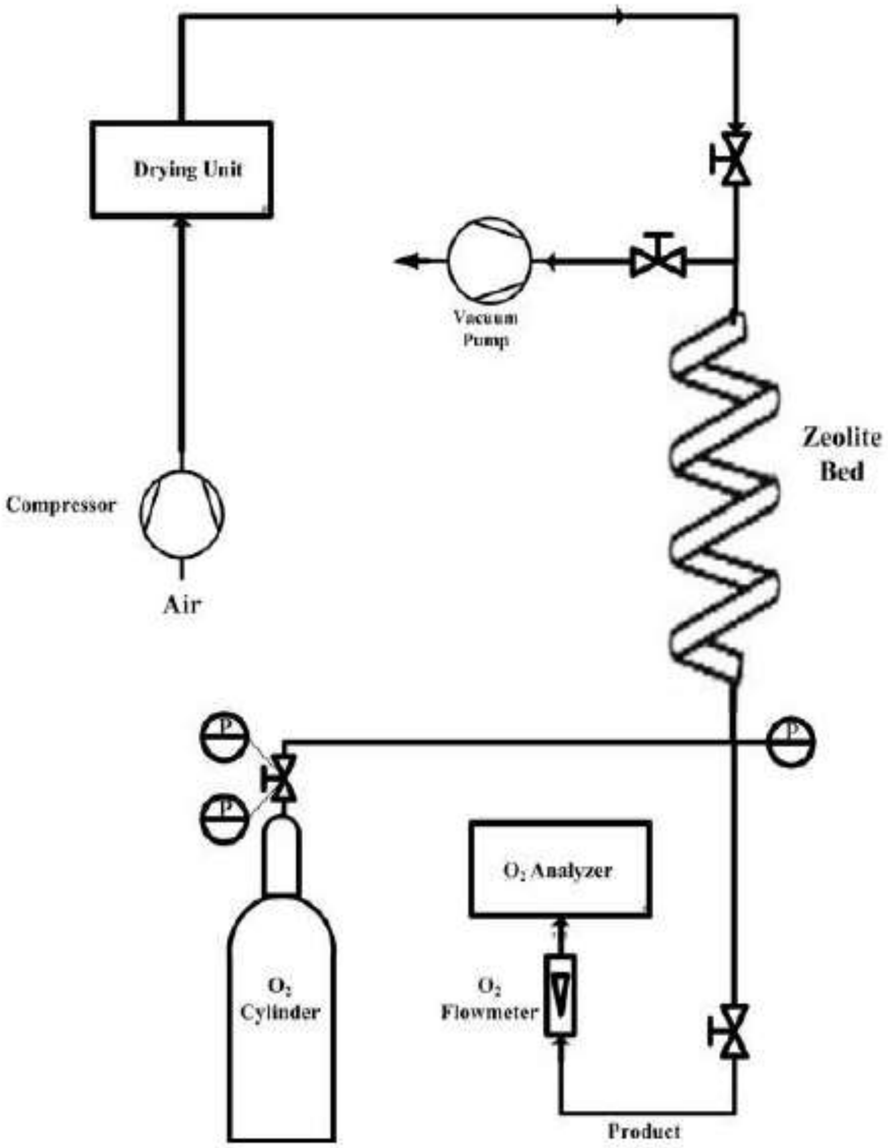

Figure (3) Experimental setup.

Table (1) Details of column and adsorbent.

Column Length	L	4 m
Column diameter	D	25 mm
Adsorbent Type		13X zeolite
Shape		Sphere
Particle diameter	d_p	1.7-2.6 mm
Particle density	ρ_p	1070 kg/m^3
Bulk density	ρ_B	670 kg/m^3
Bed porosity	ε	0.4
Adsorbent weight	w	1.3 kg
Langmuir isotherm parameters[4]		
Oxygen	q_{sO2}	3.091 mole/kg
	b_{O2}	0.0367 bar^{-1}
Adsorption heat of O_2	ΔH_{O2}	12.8 kJ/mole
Nitrogen	q_{sN2}	3.091 mole/kg
	b_{N2}	0.1006 bar^{-1}
Adsorption heat of N_2	ΔH_{N2}	17 kJ/mole

Table (2) Experiments of single column performance.

NO.	P bar	Feed of pressurizing
1	2	Air
2	3	Air
3	4	Air
4	5	Air
5	2	Intermediate initial pure oxygen pressure of 1.5 bar +feed air to 2 bar
6	3	Intermediate initial pure oxygen pressure of 2 bar +feed air to 3 bar
7	4	Intermediate initial pure oxygen pressure of 2.5 bar+feed air to 4 bar
8	5	Intermediate initial pure oxygen pressure of 3 bar+feed air to 5 bar

RESULTS AND DISCUSSION

The experimental results for the single column characteristics are analyzed using breakthrough curves.

Figure (4) represents breakthrough curve for air feed pressurizing, the purity of oxygen produced by the single column as a function of time at different values of pressure. The effluent flow rate from the column is 1 liter/min. The effluent product oxygen purity is the range of about 87 % to 89 % up to about 60 seconds, and after this point the effluent oxygen purity decreases, due to the penetration of the bed by pressurized air feed. This point represents the breakthrough point of the single bed with air pressurizing. The figure also shows the effect of pressure on the maximum effluent oxygen purity before breakthrough point. No significant change was noticed between the pressure range studied of 2 and 5 bars.

The average maximum effluent product oxygen purity is of about 88 % in the present work which is less than that of the published literature of higher than 90% [5], in spite of using long column. This can be attributed to the large particle size (1.7-2.6 mm compared to 1mm) used in the present study. Air pressurizing increases the axial dispersion of nitrogen and the shape of purity curve represented by very wide mass transfer zone (MTZ).

Figure 5 presents breakthrough curve for the column filled with pure oxygen to desired intermediate pressure and thence followed by air feed to the column to adsorption pressure. The performance of the column with intermediate pure oxygen pressurizing, is better than the pressurizing with air, because the breakthrough time is higher than the process of air pressurizing (from 9 to 18 minutes compared to one minute). In addition, the average maximum effluent oxygen purity is of about 95 %. After the air feeding for long time (minutes), mass transfer zone (MTZ) is steeper or represents shock wave, and high ratio of adsorbent is depleted before the breakthrough by nitrogen. Pressurizing with pure oxygen minimizes axial dispersion.

Same trends were found by Auob [11] and Abdel-Rahman et al. [12], when studying single vertical bed characteristics, with air initial pressurizing, and with initial intermediate pure oxygen pressurizing, to simulate the work of 2-column (D=50 mm, and L=570 mm), 4-steps, and 6-steps PSA processes. The present work shows higher maximum oxygen purity for the two cases (eighties and nineties maximum oxygen purity in the present work compared to seventies and eighties oxygen purity for the previous work, for the two cases respectively).

Figure (6) shows the total amount of nitrogen adsorbed (q) with and without pure oxygen pressurizing and it increased as pressure increases, and the total amount of nitrogen adsorbed, with initially pure oxygen pressurizing is higher than that with air feed pressurizing, confirming the above results. The range of zeolite loading capacity is (q = 0.25-0.35 mole N_2/kg zeolite) for initial air pressurizing case. Whereas the range of zeolite loading capacity is (q = 0.39-0.87 mole N_2/kg zeolite) for initial intermediate oxygen pressurizing case, which is agree well with the equilibrium data of multi-component Langmuir adsorption equation using 79% N_2 feed.

The average of 40% of the range of zeolite loading capacity has been utilized before breakthrough time for initial air pressurizing case. Whereas the average of 95% of the range of zeolite loading capacity, has been utilized before breakthrough time for initial intermediate oxygen pressurizing case, as shown in Figure (7).

The objective from the single column characteristics is to predict the maximum product oxygen purity, productivity, breakthrough point, adsorption time, adsorption pressure, and product flow rates, because all these characteristics are the basic information for PSA unit design. So the expected characteristics are the maximum product oxygen purity of about 88%, and the breakthrough time of about 60 seconds, for 2-columns, 4-steps PSA process. Whereas for 2-columns 6-steps PSA process the expected characteristics are the maximum product oxygen purity of higher than 90%, and the breakthrough time of about 9 to 18 minutes, depending on pressure. These results have been verified using dual columns PSA process by Mhdi [13], and Abdel-Rahman et al.[14].

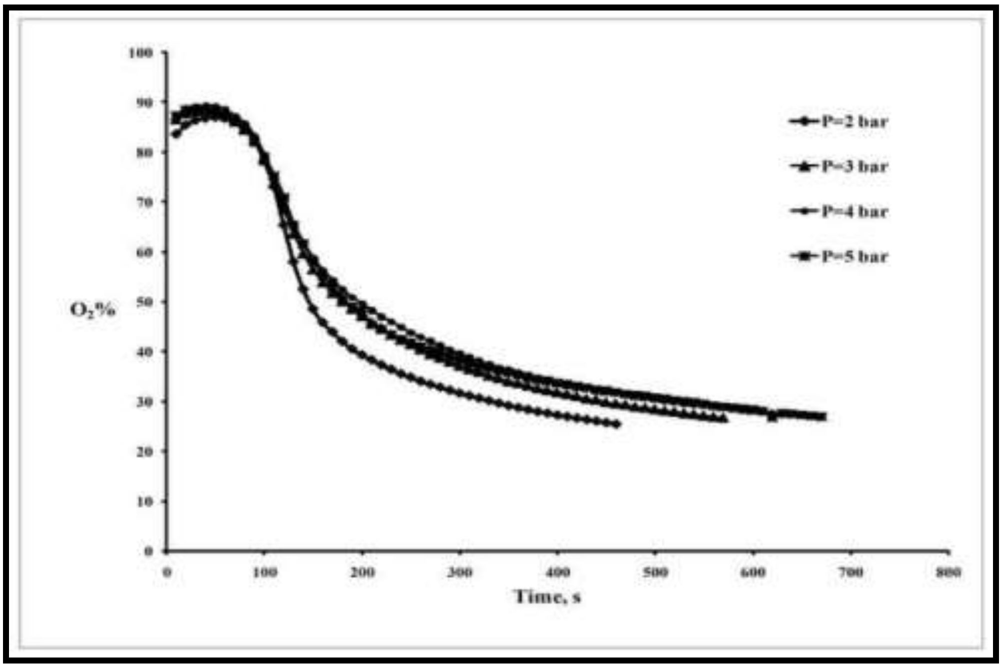

Figure (4) Breakthrough curve, using air feed pressurizing at different adsorption pressures and Q_{eff}=1 lit/min.

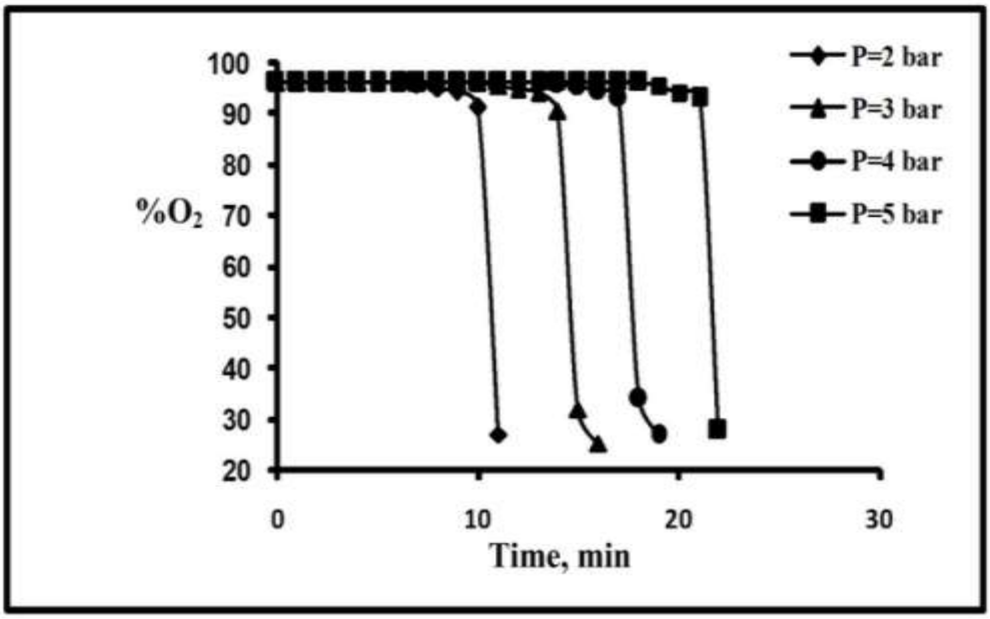

Figure (5) Breakthrough curve, by intermediate initial pure oxygen Pressurizing at Q_{eff}= 1 lit/min.

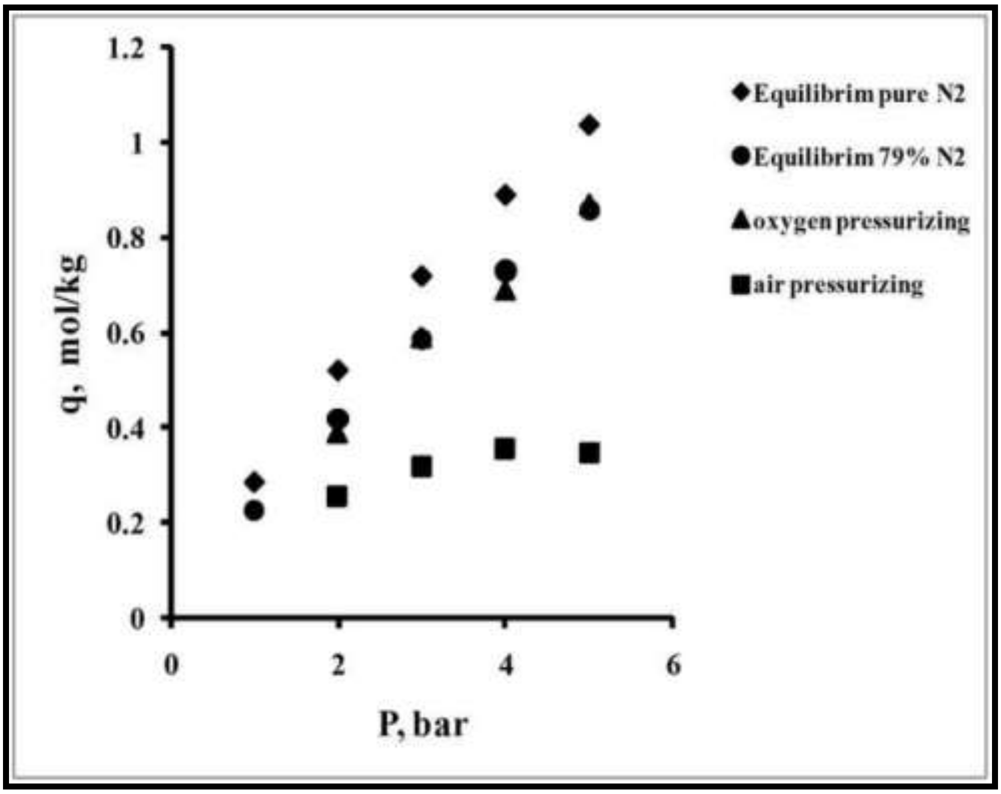

Figure (6) Total single column capacity for nitrogen adsorption With and without oxygen pressurizing.

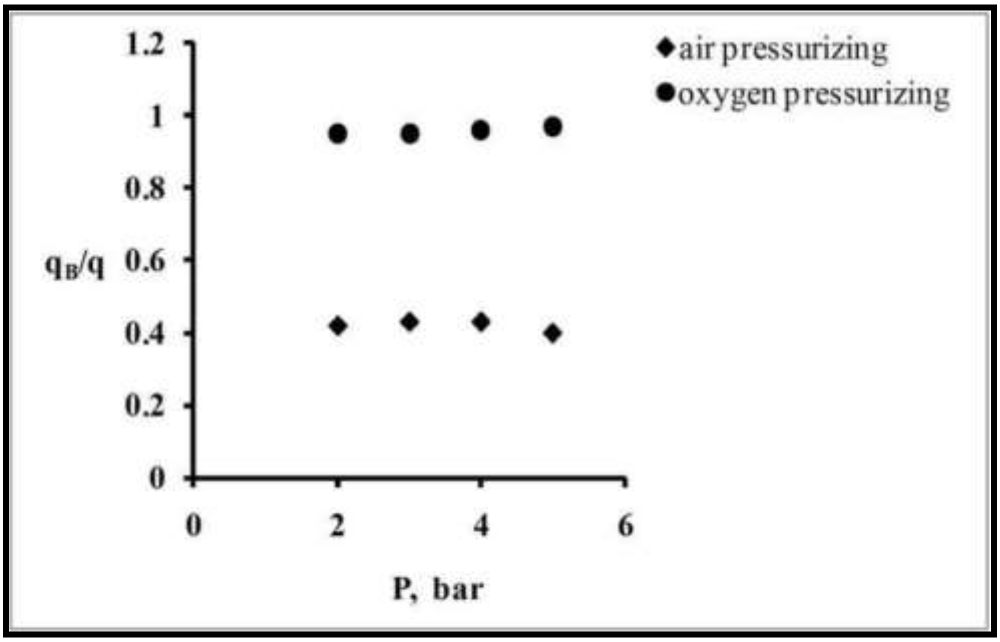

Figure (7) the ratio of the capacity of the column before breakthrough time to the total capacity with and without oxygen pressurizing.

CONCLUSIONS

1. No significant effect of pressure on the product oxygen purity is noticed when pressure increased from 2 to 5 bar
2. The average maximum oxygen purity of 88% is obtained for initial air pressurizing case, and about 95% is obtained for initial intermediate pure oxygen pressurizing case.
3. The range of zeolite loading capacity (q) is 0.25-0.35 mole N_2/kg zeolite for initial air pressurizing case, and average of 40% of the range has been utilized before breakthrough time.
4. The range of zeolite loading capacity (q) is 0.39-0.87 mole N_2/kg zeolite for initial intermediate oxygen pressurizing case, and average of 95% of the range has been utilized before breakthrough time, which is agree well with the equilibrium data of multi-component Langmuir adsorption equation.

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NOMENCLATURE

B Langmuir isotherm constant, bar^{-1}
C Solute concentration, mole/l
C_o Solute feed concentration, mole/l
C_p Solute product concentration, mole/l
D Column diameter
d_p Zeolite particle diameter, mm
L Column length, m
P Adsorption pressure, bar
P_i Pressure of component (i), bar
Q_{eff} Effluent flow rate, l/min
Q Adsorbent capacity, mole/kg
q_i Adsorbent capacity of component (i), mole/kg
q_s Maximum adsorbent capacity of Langmuir isotherm, mole/kg
q_B Adsorbent capacity before breakthrough time, mole/kg
t Time, s
t_b Breakthrough time, s
t_s Ideal adsorption time or saturation time, s
W Adsorbent zeolite weight, kg
YO2 Oxygen mole fraction

GREEK SYMBOLS

ρ_B Bulk density, kg/m^3
ρ_P Particle density, kg/m^3
ε Bed porosity
ΔH_{O2} Adsorption heat of O_2, kJ/mole
ΔH_{N2} Adsorption heat of N_2, kJ/mole
Δt time increment , s