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REMOVAL OF SINGLE AND BINARY ORGANIC VAPORS FROM AIR BY ADSORPTION METHOD

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ABSTRACT

Removal of isopropyl alcohol and isobutyl alcohol vapors from air by activated carbon was carried out. The equilibrium data for single component systen1s were obtained at different temperatures 301, 308, 3 J9 and 327 K. The equilibrium data for binary system was found at temperature 319 K. The equilibrium isotherm results for single and binary components fit very well with Langmuir's isotherm. The effect of the inlet concentration on the dynamic capacity, MTZ velocity, MTZ length and thermal wave velocity was studied for single and binary systems. Empirical correlations describing these variables were determined. A mathematical model was used to predict the breakthrough curves and temperature profiles. These results were in good agreement with the experimental results.

KEY WORDS: adsorption isopropyl alcohol, isobutyl alcohol

INTRODUCTION

Adsorption is a process by ,which the solid surface attracts the molecules from a stream of gas or liquid. These attractive forces vary according to the nature of the gas or liquid molecules and the solid surface (Lund, 1971). The simplicity of the physical adsorption process reduces capital costs, operating costs and affords design flexibility and high reliability (Othmer, 1978).

Systems used for gaseous adsorption are fixed bed, moving bed and fluidized bed adsorbers. Fixed beds are most frequently used for gas purification and dehydration (Riesenfeld and Kohl, 1974; Nouhebel, 1972). In industry there must be at least two fixed bed adsorbers in each system, one bed is in operation while the other one is being regenerated. Regeneration can be achieved by one of the following: steam-stripping, thermal desorption, vacuum desorption, thermal swing cycle, pressure swing cycle, purge gas stripping, oxidation and displacement cycle (Bethen, 1978).

The most widely used adsorbents are activated carbon, activated alumina, activated bauxite and silica gel (Hassler, 1974). Activated carbon is unique in its ability to remove materials at relatively low operating cost with minimal variation in efficiency due to concentration (Lund,١٩٧١ ; Othmer, 1978; Riesenfeld and Kohl, 1974; Nouhebel, 1972; Bethen, 1978; Hassler, 1974).

Isopropyl and isobutyl alcol1ols were chosen as gaseous pollutants because of their industrial importance each of them is used as a food additive (Sax,1975).

EXPERIMENTAL ARRANGEMENTS AND PROCEDURE

Materials

Two liquids (Perry and Chilton,1984) were used as adsorbate namely isopropyl alcohol (I.P.A) (99.7% purity) and isobutyl alcohol (I.B.A) (99.7% purity).

Air was used for the preparation of polluted air mixture. Nitrogen (99.99% purity) was used for gas chromatography. Hydrogen was generated by means of hydrogen generator and supplied to the flame ionization detector in the gas chromatography.

Granular activated carbon (supplied by Norit Verkoop Gentral, Amsterdam, Holland) was used as adsorbent (0.86 mm diameter and 3.34 mm length). The physical properties are: ash content 7.5%, bulk density 400 kg/m^3, grain density 1400 kg/m^3 and surface area 95 x 10^4 m^2/kg.

Adsorption Equipment

Three different sets of experiments were carried out. IPA was used in the first set of experiments, IBA was used in the second set while in the third set of experiments a mixture of IPA and IBA liquid were used. The equipment used in the present research consisted of three sections as follows: 1- air mixture preparation section consisted of two Q.V.F spherical vessels (each of capacity 0.005 m^3 Raschig rings were placed at the bottom of the saturator to generate high number of bubbles which insured gas saturation. 2- adsorption section consisted a carbon steel column (0.1 m I.D. and 0.615 m lengtl1). Five thermocouples (copper constantan) were placed in column. All pipe connections were Q.V.F. glass and Teflon tubes. The entire apparatus is shown in Fig. 1 for the first and the second experimental sets, while Fig. 2 was for the third experimental set. 3- gas analysis section consisted of a chromatographic system placed on line to analyze the inlet and outlet gas from the adsorption column.

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RESULTS AND DISCUSSION

Equilibrium Isotherms for IPA and IBA as Single Contents

Equilibrium isotherms were obtained at temperatures 310, 308, 319 and 327 K respectively for both IPA and IBA separately. The breakthrough curves were plotted as the effluent concentration against time Figs. 3, 4 for IPA and IBA respectively.

The capacity as a function of concentration were plotted in Figs. 5, 6 for IPA and IBA respectively. These figures showed that the adsorption capacity at a given temperature increased with the increased inlet cocentration. At low inlet concentration it increased very rapidly and at high inlet concentration it reached nearly to a steady value. This means that the equilibrium is of the favorable type. The adsorption capacity decreased at a given inlet concentration with increasing temperatures. This was an indication that the behavior of the system undergoing physicaI adsorption was normal. IBA was preferentially adsorbed compared with IPA when the two vapors were as single components (the adsorbed phase concentration for IBA was more than IPA).

$$q = \frac{Q_o\ b\ C}{1 + b\ C}$$

The values of (C_0/q) were plotted versus (C_0) for each temperature Figs. 7, 8 for IPA and IBA respectively. These figures showed straight lines which means that the two systems fit very well with Langmuir's equation.

Jh

Hg

The constants Q_0 and **b** in eq. (1) were tabulated in Table 1.

Table1 Langmuir' s Constant tor IPA and IBA as single Constants

Temperature	IPA System		IBA System	
(K)	Q_0	b	Q_0	B
303	0.878	42.46	0.834	70.47
308	0.668	50.00	0.825	54.35
319	0.661	40.89	0.810	45.87
327	0.592	32.03	0.767	45.12

Heat of Adsorption

The heat of adsorption for each system was calculated 1,y using Clausis-Clapeyron's equation (Lund, 1971). This was found to be equal to 690.24 kJ/kg for IPA and 599.78 kJ/kg for IBA. These values were in very good agreement with the values of the latent heat of condensation (662.23 kJ/kg for IPA and 577.14 kJ/kg for IBA). This is an indication that the two systems undergo physisorption.

Equilibrium Isotherm for Binary Components

For constant inlet concentration IPA (C_{po}) and different IBA inlet concentration (C_p), the breakthrough curves (C_p/C_{po} against time) where shown in Fig. 9. While for constant IBA inlet concentration (C_{bo}) and different inlet concentration (C_b) for IPA, the breakthrough curves (C_b/C_{bo} versus time) were plotted in Fig. 10.

Figure 10 indicated that when the IPA inIet concentration was varied, an inverse variation of the peak height was observed. This was a result of the relative competition for adsorption of activated carbon surface between IPA and IBA. In another word the peak heights were increased with decrease of IPA inlet concentration. This means that the adsorbility of the IPA component was decreased. The breakthrough curves in Fig. 9 showed that the peak height indicated that the IBA inlet concentration was increased as the peaks of IPA component curves was increased. This means that adsobility of the IPA component was decreased.

The values of (C_{po}/q_p) were plotted against (C_{po}) at constant IBA inlet concentration (C_{bo} =43.4x10^{-3} kg/m^3) at temperature 319 K. values of (C_{bo}/q_b) were also plotted versus (C_{bo}) at constant IPA inlet concentration) C_{po}=28.93x10^{-3} kg/m^3 at temperature 319 K Fig. 11. This figure illustrated two straight lines which means that the binary components system (IPA-IBA- activated carbon) fits very well with Langmuir's equation for multi component materials (Lund, 1971).

$$q_i = \frac{Q_{oi} b_i C_i}{1+\sum_{i=1}^{n} b_i C_i}$$

The results showed that the constant Q_{op} and Q_{ob} were approximately the same (Q_{op}=0.742 kg/kg and Q_{ob}=0.749 kg/kg), the two other Langmuir constants were b_p=34.557 m^3/kg and b_b=59.398 m^3/kg for IPA and IBA in binary system respectively.

Fitting the experimental results was aclueved by incorporating the Langmuir constants as single component from eq. (1) into eq. (2) for IPA and IBA as binary mixture. There were an excellent agreement

between the experimental data (binary mixture) and Langmuir equation (Othmer, 1978) for IPA or IBA when IBA or IPA at constant inlet concentration respectively.

Non-Isothermal Adsorption

Single Component System

The breakthrough curves were obtained for IPA and IBA by plotting the effluent concentration against time Figs. 12, 13 respectively.

Effect of Concentration on Dynamic Capacity

The dynamic capacity was plotted against inlet concentration by both systems. These figures showed that the dynamic capacity was increased with increasing inlet concentration for both systems. But at high inlet concentration (for IPA and IBA were 20x10-3 kg/m^3 and 30 x10^{-3} kg/m^3 respectively) the curves reached gradually to steady state. This was the result of the heat generation and a high temperature due to high concentration (adsorption is an exothermic process).

Effect of Concentration on Breakthrough and Equilibrium Time

The following correlations were obtained for the breakthrough time and the equilibrium time as follows:

IPA system	**IBA system**
$t_b = 8.346 \times 10^2 C_{po}^{i-p92}$	$t_b = 15.208 \times 10^3 C_{bo}^{-0.2}$
$t_c = 16.819 \times 10^2 C_{po}^{-0.75}$	$t_e = 20.692 \times 10^3 C_{bo}^{-0.14}$

Effect of Concentration on the MTZ Length

The mass transfer zone (MTZ) length (Lund, 1971) for IPA and IBA systems were calculated using eq. (3)

$$\bar{Z} = \frac{Z(t_e - t_b)}{t_e - (1-f)(t_e - t_b)}$$

Where f= $\frac{A_1}{A_2}$, A_1= area under the curve between t_b and t_e, A_2= area above the curve between t_b Two correlations were obtained.

IPA system	IBA system
$\bar{Z} = 0.586\ C_{po}^{-0.76}$	$\bar{Z} = 0.356\ C_{bo}^{-0.67}$

Effect of Concentration on the MTZ Velocity

The following correlations between MTZ velocity and inlet concentration were obtained:

IPA system

$\bar{v} = 3.102 \times 10^{-4}\ C_{po}^{-0.84}$

IBA system

$\bar{v} = 1.752 \times 10^{-5}\ C_{bo}^{-0.15}$

The IBA system showed that the MTZ velocity increased sharply up to 5×10^{-3} kg/m^3 concentration then it slightly increased with concentration. While the IPA system showed slight increasing with concentration. This was due to the high adsorbed capacity for IBA by the adsorbent than for IPA.

Temperature Profiles

The temperature profiles (temperature versus time) at various axial locations were plotted as shown in Figs. 14, 15 for IPA and IBA system respectively (for bed height 0.35 m).

These profiles showed that for most inlet concentration a sharp temperature rise occurred in the first thermocouple compared with the others. This was due to the high adsorption rate occurred at the first part of the bed.

The temperature profiles increased sharply with increasing in inlet concentration. This was due to the high rate of the adsobility when concentration increased followed by more heat generated.

Effect of concentration on the Thermal Wave (T_w) Velocity

The following correlation were obtained between the T_w velocity and inlet concentration for the two systems:

IPA system

$\bar{T} = 3.118 \times 10^{-4}\ C_{po}^{-0.86}$

IBA system

$\bar{T} = 2.488 \times 10^{-5}\ C_{po}^{-0.23}$

Binary Component System

Dimensionless effluent concentration groups C_p/C_{po} and C_b/C_{bo} were plotted versus time (breakthrough curves) as shown in Fig. 16 at constant C_{bo} and Fig. 17 at constant C_{po}. These curves showed that each component advanced at a different speed i.e. IPA component propagated with the faster moving front at first. After that the situation changed when IBA component became adsorbed within the bed and then by displaced part of IPA component, this procedure resulting a higher IPA outlet concentration. As IBA component breakthrough a little less sharply than IPA, the displacement of IPA component was reduced. Then the

concentration of the two components tends to reach their inlet values. It was clear from the breakthrough curves of each component of the binary system (IP A and IBA) that IBA displaced IPA from the adsorber because IBA was more desirable to be adsorbed from the vapor mixture. IBA component was adsorbed more prominently near the inlet of the column, forcing downstream an essentially pure IPA component.

Effect of Concentration on the Dynamic Cauacity

The binary system with constant C_{po} indicated that the dynamic capacity was more than for the binary system with constant C_{bo}. This was due to that IBA was preferentially adsorbed from the mixture.

The dynamic capacity of IPA and IBA in binary component system (IPA-IBA- activated carbon) were less than their values as single component. This could be explained by the effect of adsorption of the component on another in binary system, i.e., the capacity of component in mixture was less compared witl1 the capacity as single component.

The dynamic capacity for IPA at constant C_{po} or fpr IBA at constant C_{bo} decreased with increasing their inlet concentration.

Effect of Concentration on Breakthrough and Equilibrium Time

The experimental results showed that the breakthrough time and equilibrium time decreased with increasing the inlet concentration. The following correlations were obtained.

IPA component at constant C_{po}

$$t_b = 2.52\ C_{po}^{-2.34}$$

$$t_e = 1.985 \times 10^2\ C_{po}^{-1.31}$$

IBA component at constant C_{bo}

$$t_b = 1.367 \times 10^2\ C_{bo}^{-1.1}$$

$$t_e = 6.607 \times 10^2\ C_{bo}^{-0.75}$$

Effect of Concentration on the MTZ Velocity

The velocity of MTZ were calculated (Hasso, 1987) by using eqs. (4, 5).

$$\nabla = \frac{z}{t_s}$$

where t_s is the stoichiometric time

$$t_s = t_m - \frac{\int_o^{im} C(t)dt}{C_o}$$

where t_e and t_m are the time when the concentration profiles are flattened out into plateau zones.

It was found that the MTZ velocity increased with the increasing the inlet concentration. The following correlations were obtained to represent the experimental data.

IPA component at constant C_{po}

$$\bar{v} = 2.493 \times 10^{-3}\ C_{po}^{1.19}$$

IBA component at constant C_{bo}

$$\bar{v} = 1.025 \times 10^{-3}\ C_{bo}^{-089}$$

Effect of Concentration on the MTZ Length

The length of MTZ was calculated by using eq. (3). It was found that the length of MTZ was increased with increasing the inlet concentration. These results indicated also that the MTZ length for IPA at constant C_{po} was increased with the increasing of C_{bo}. The same behavior was observed for IBA at constant C_{bo}. The following correlations were found from the experimental data.

IPA component at constant C_{bo}

$$\bar{Z} = 1.996 \times 10^{-2}\ C_{po}^{0.02}$$

IBA component at constant C_{po}

$$\bar{Z} = 1.188 \times 10^{-2}\ C_{bo}^{-03}$$

Temperature Profiles

The temperature profiles (temperature versus time) were plotted for IPA and IBA at constant C_{po} and constant C_{bo} at various axial positions (bed length equal to 0.35m).

The temperature profiles for the binary component system gave the same shape as temperature profiles for the single component system; i.e. the same conclusion for the binary system could be adopted as the single component system as far as the pattern behavior of the curves were the same.

Effect of Concentration on the Thermal wave (T_w) Velocity

It was found that T_w velocity were increased with the increasing inlet concentration for both systems. To represent the experimental data the following correlations were obtained.

Binary system at constant C_{bo}

$$\bar{T} = 4.859 \times 10^{-2}\ C_{t}^{1.82}$$

Binary system at constant C_{po}

$$\bar{T} = 0.955 \times 10^{-2}\ C_{t}^{1.23}$$

Predication of the breakthrough curves and temperature profiles.

The theoretical model described by Ruthven, et al., 1975 was used to predicts the concentration as well as the temperature profiles of a non-isothermal single component adsorption. This was carried out by using two dimensionless differential equations.

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$$\frac{dy}{d\Gamma} = \frac{y(1-y)}{y + \left(\frac{1-\lambda}{\lambda}\right)e^{x}}$$

$$\frac{dx}{dt} = \frac{1}{\beta}\frac{dy}{d\tau} - \alpha x$$

Solutions of the two differential equations mentioned above were obtained numerically for various values of the parameters λ , β and α by using a fourth order Runge-Kutta Scheme (Hasso, 1987; Sulaymon et al., 1986; Kasser and Sulaymon, 1999).

The predicted concentration gave a good representation compared with the experimental results at high inlet concentration for IPA (average absolute error 14%-20.5%) except for tl1e experiments with inlet concentration equal to 23.46 x 10^{-3} kg/m^3 (average absolute error 39.2%). While the average absolute error for IBA were found to be 20.4% to 35% for all the experiments except for inlet concentration equal to 6.128 x 10^{-3} kg/m^3 was found to be 44%.

A similar behavior between the experimental and the predicted temperature were noticed. The reasons for the diversion are due to the random packing and the assumption of a constant behavior in the model which leads to the inevitable degree of unreliantly when applied to the systems IPA and IBA at low concentration.

CONCLUSIONS

1- The equilibriun1 isotherms, for the systems (IPA-activated carbon and IBA-activated carbon) were well represented by Langmuir's equation for single component (eq. 1). The equilibrium isotherm for binary system (IPA-IBA-activated carbon) were well represented by multi-component Langmuir's equation (eq. 2).

2- The heat of adsorption for IPA and IBA were in good agreement with their latent heat of condensation, i.e., adsorption of both systems are physisorption.

3- For a given bed height, the dynamic capacity, MTZ velocity, MTZ length and the thermal wave velocity increased with increasing inlet concentration for both non-isothermal single or binary systems.

4- Correlations were obtained to represent the experimental data (t_b, t_c, $\bar{V}$, $\bar{Z}$, $\bar{T}$) for single and binary systems. For binary system, T w velocity correlation was found.

5- The experimental breakthrough values and temperature profiles for a single component system (IPA or IBA) were in good agreement with the predicted model.

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NOMENCLATURE

A_1	Area under the breakthrough curve between t_b and t_e	
A_2	Area above the breakthrough curve between t_b and t_e	
a_p	Effective mass transfer area	M^2
b	Constant in Langmuir equation	M^3/kg
c	Fluid concentration of component within the particle	Kg/m^3
C_o	Inlet concentration of gas	Kg/m^3
Cp	Heat capacity of gas	J/mole K
Cp_s	Heat capacity of adsorbent	J/mole K
Cp_o	Isopropanol inlet concentration	Kg/m^3
Cb_o	Isobutanol inlet concentration	Kg/m^3
d	Diameter of column	m

h	Overall heat transfer coefficient	J/m s K
K	Overall mass transfer coefficient	m/s
Q_o	Weight of solute adsorbed per unit weight at concentration C (in Langmuir equation)	Kg/kg
q_o	Particle adsorbate concentration in equilibrium with gas feed concentration	Kg/m^3
q	Adsorbed phase concentration	Kg/kg
R	Gas constant	J/mole K
$\bar{T}$	Thermal wave velocity	m/s
T_w	Well temperature	K
t	Time	s
t_e	Time at which transfer zone flatten out to plateau zone (equilibrium time)	s
t_b	Breakthrough time	s
t_s	Stoichiometric time	s
t_m	Time at which transfer zone flatten out to plateau zone	s
v	Linear gas velocity in bed	m/s
v'	Linear velocity of constant pattern front	m/s
$\bar{v}$	Mass transfer zone velocity	m/s
x	Dimensionless temperature	
y	Dimensionless concentration	
$\bar{z}$	Mass transfer zone Iength	m
z	Bed height	m
p	Density of gas phase	Kg/m^3
p_s	Density of particle	Kg/m^3
ε	Porosity of particles bed	
ι	Dimensionless time	

$$\alpha = \frac{4h}{q^*_{o}\, K\, a_p\, d(1-\varepsilon)R\left[\frac{-\Delta H}{RT_w}\right]^2}$$

$$\beta = \frac{\left[1-\frac{v}{v\prime}\right]\varepsilon pCp+(1-\varepsilon)Cp_s}{q_o^*\,(1-\varepsilon)R\left[\frac{-\Delta H}{RT_w}\right]^2}$$

$$\lambda = \frac{bC_o}{1+bC_o}$$

ΔH	Heat of adsorption	kJ/kg

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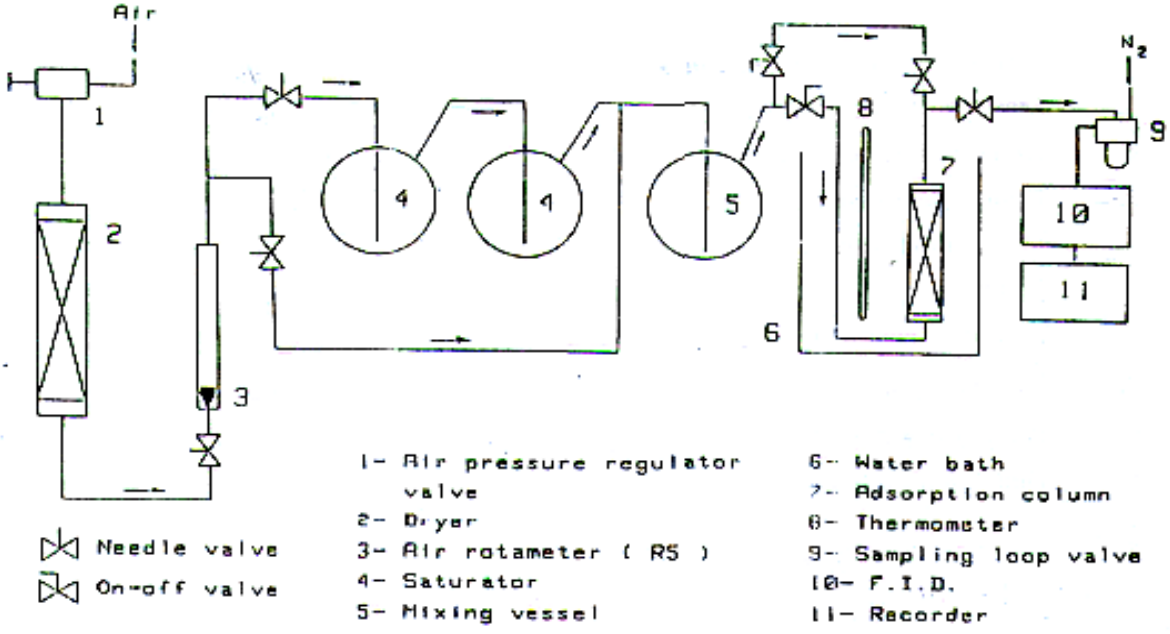


Fig.(1) Apparatus for Determination of Equilibrium Data for Single Component

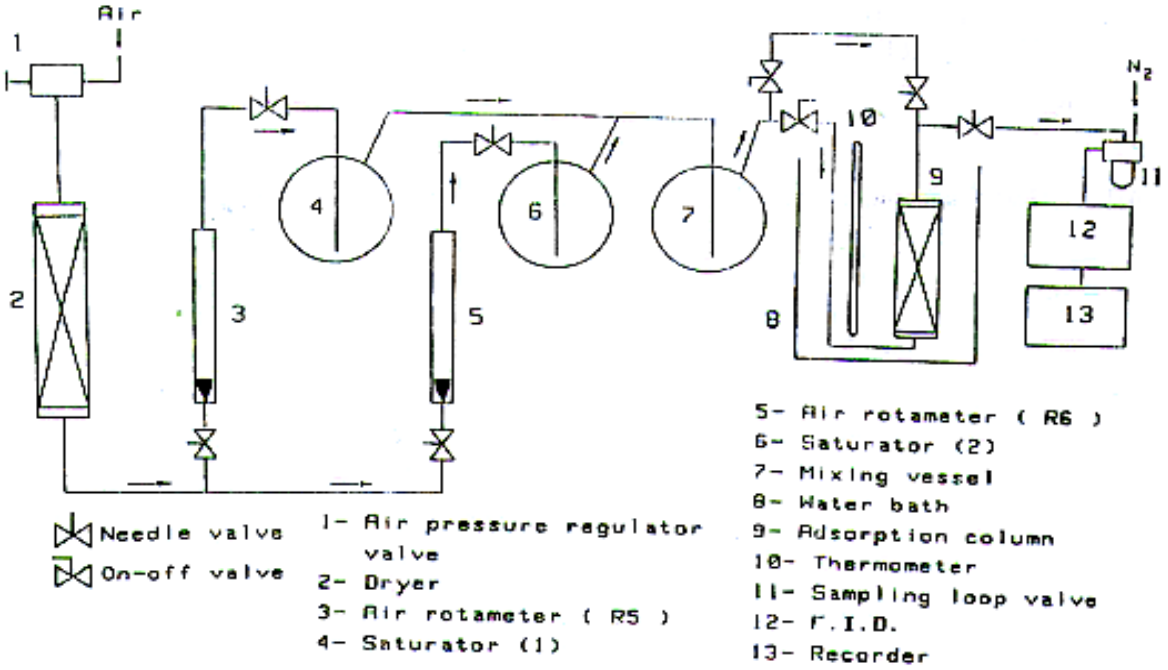


Fig.(2) Apparatus for Determination of Equilibrium Data for Binary Component

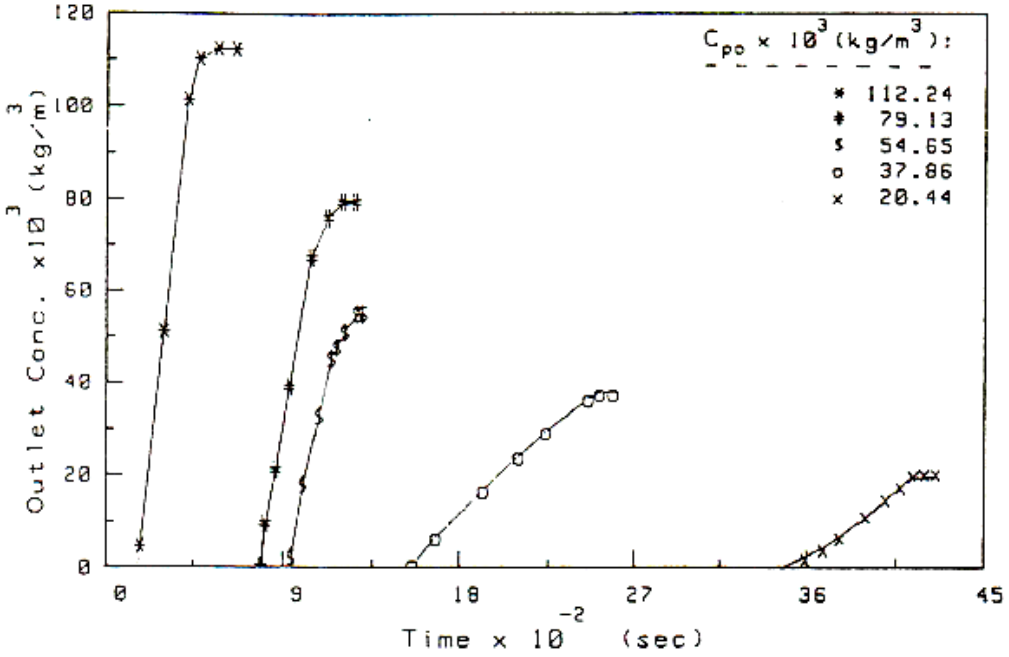


Fig.(3) Breakthrough Curve for Equilibrium Isotherm at Temperature =319 K for Isopropyl alcohol

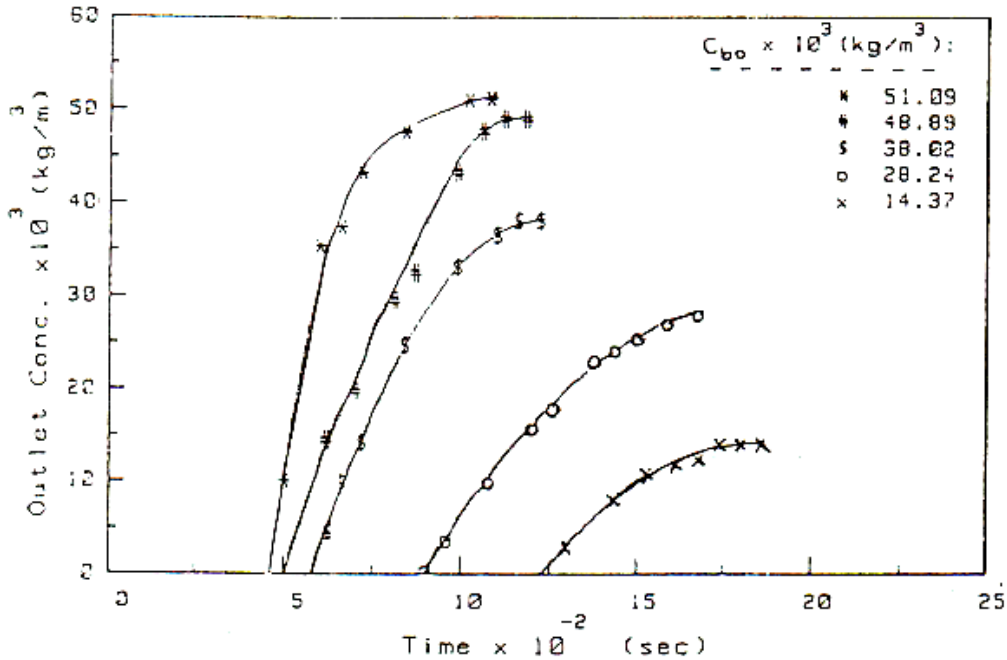
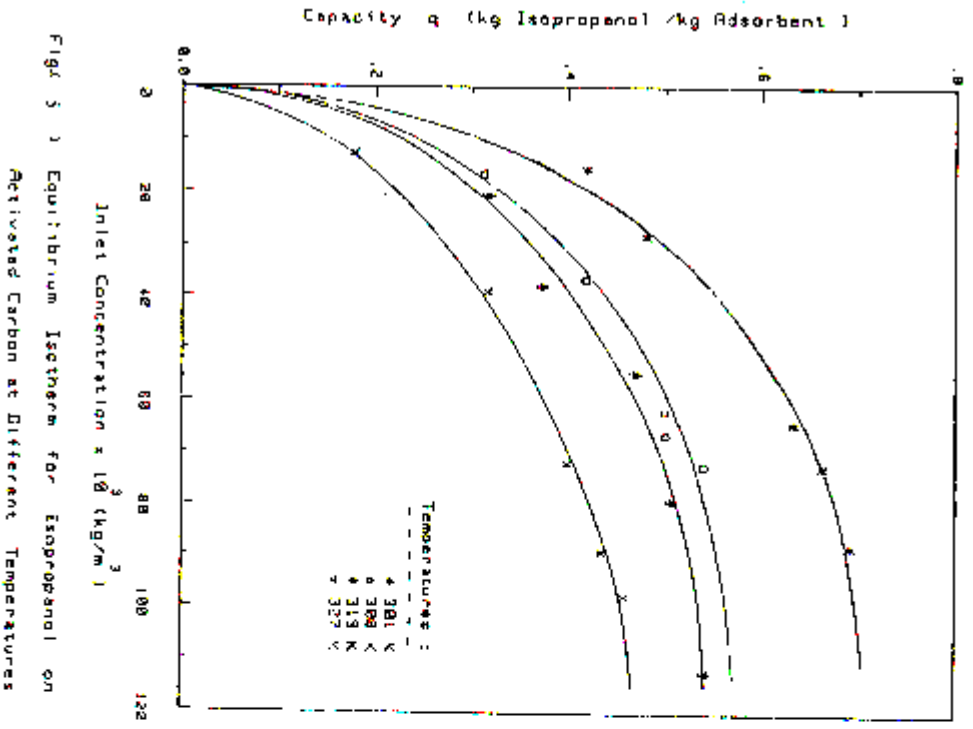
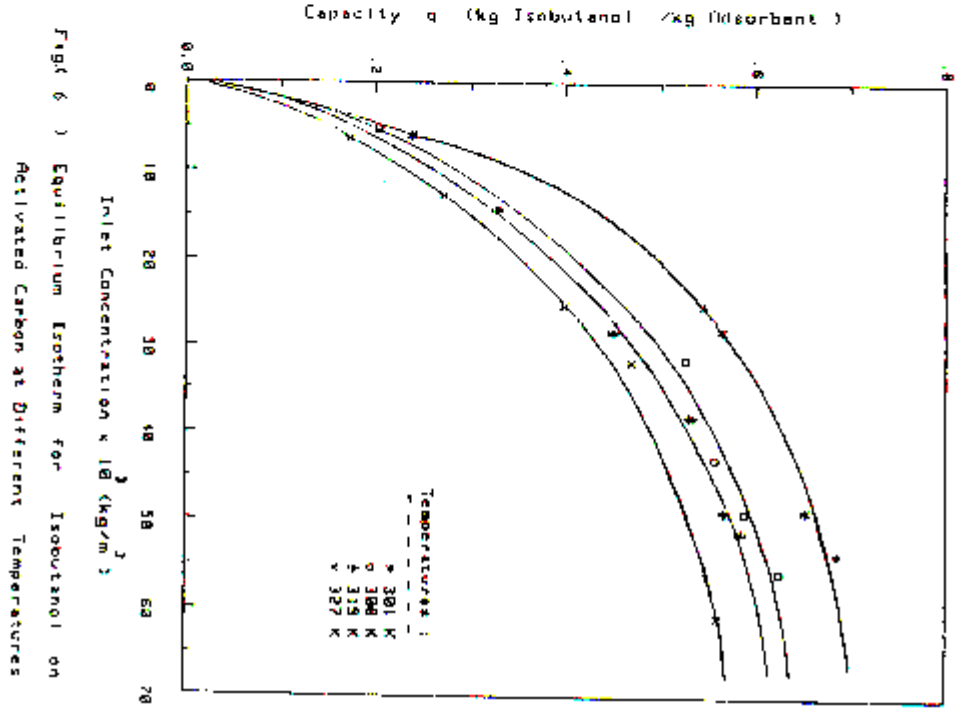


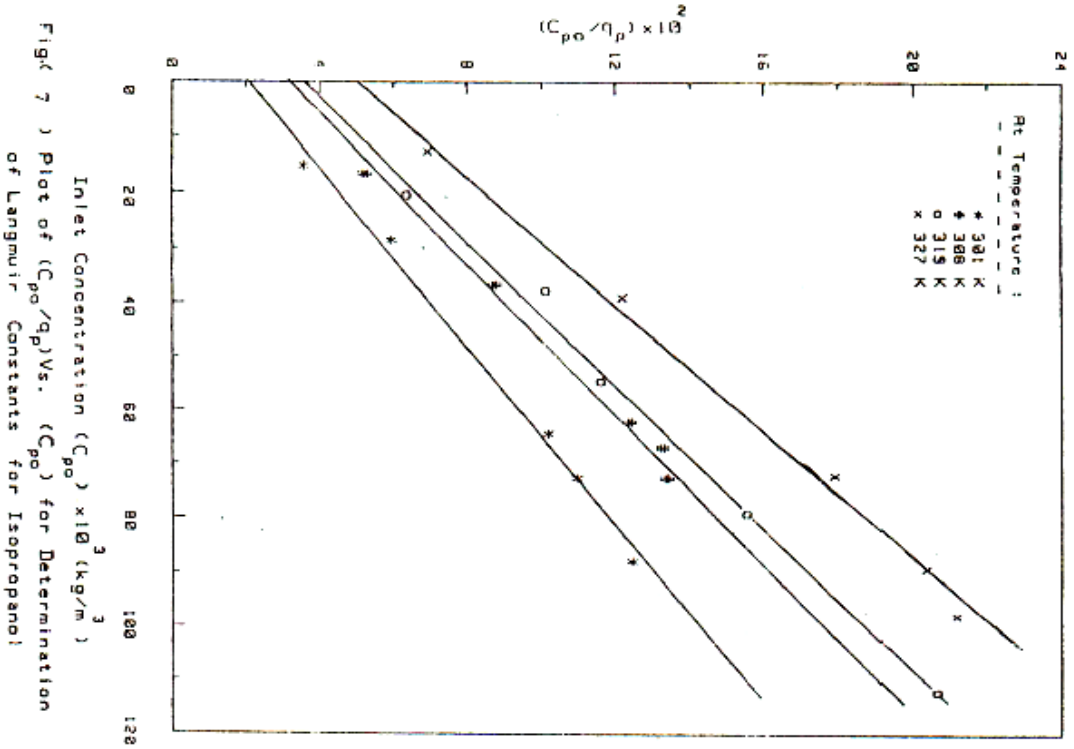
Fig.(4) Breakthrough Curve for Equilibrium Isotherm at Temperature = 319 K for Isobutyl alcohol



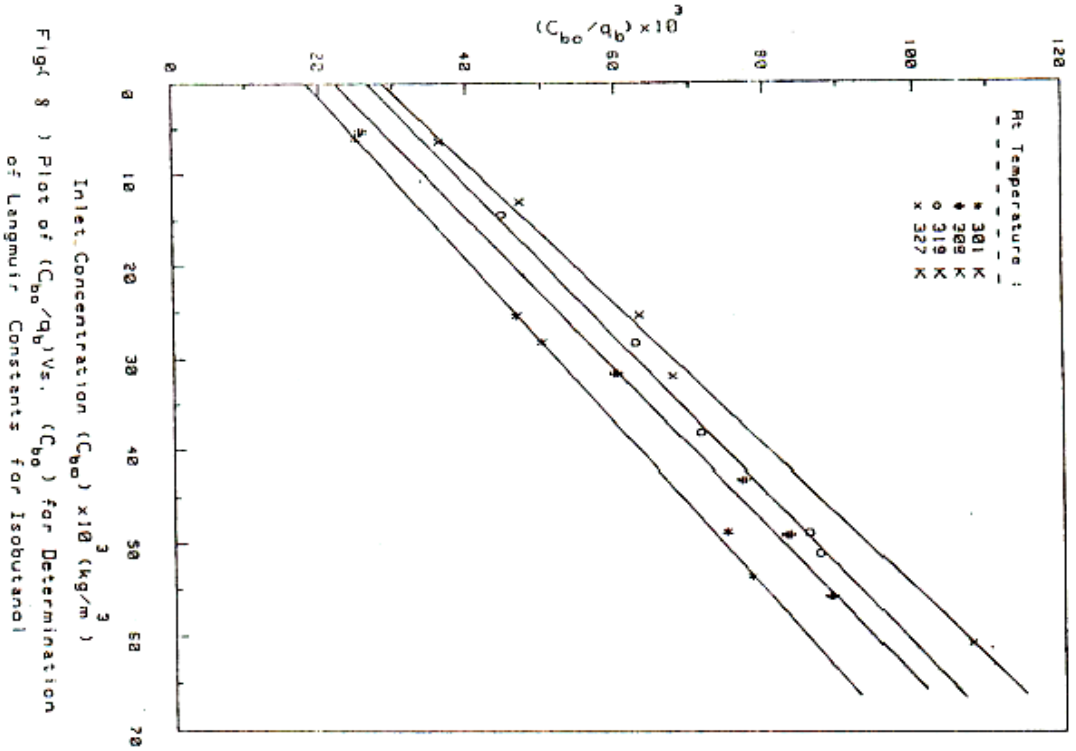
Fig(5) Equilibrium Isotherm for Isopropanol on Activated Carbon at Different Temperatures



Fig(6) Equilibrium Isotherm for Isobutanol on Activated Carbon at Different Temperatures



Fig(7) Plot of (C_{po}/q_p)Vs. (C_{po}) for Determination of Langmuir Constants for Isopropanol



Fig(8) Plot of (C_{bo}/q_b)Vs. (C_{bo}) for Determination of Langmuir Constants for Isobutanol

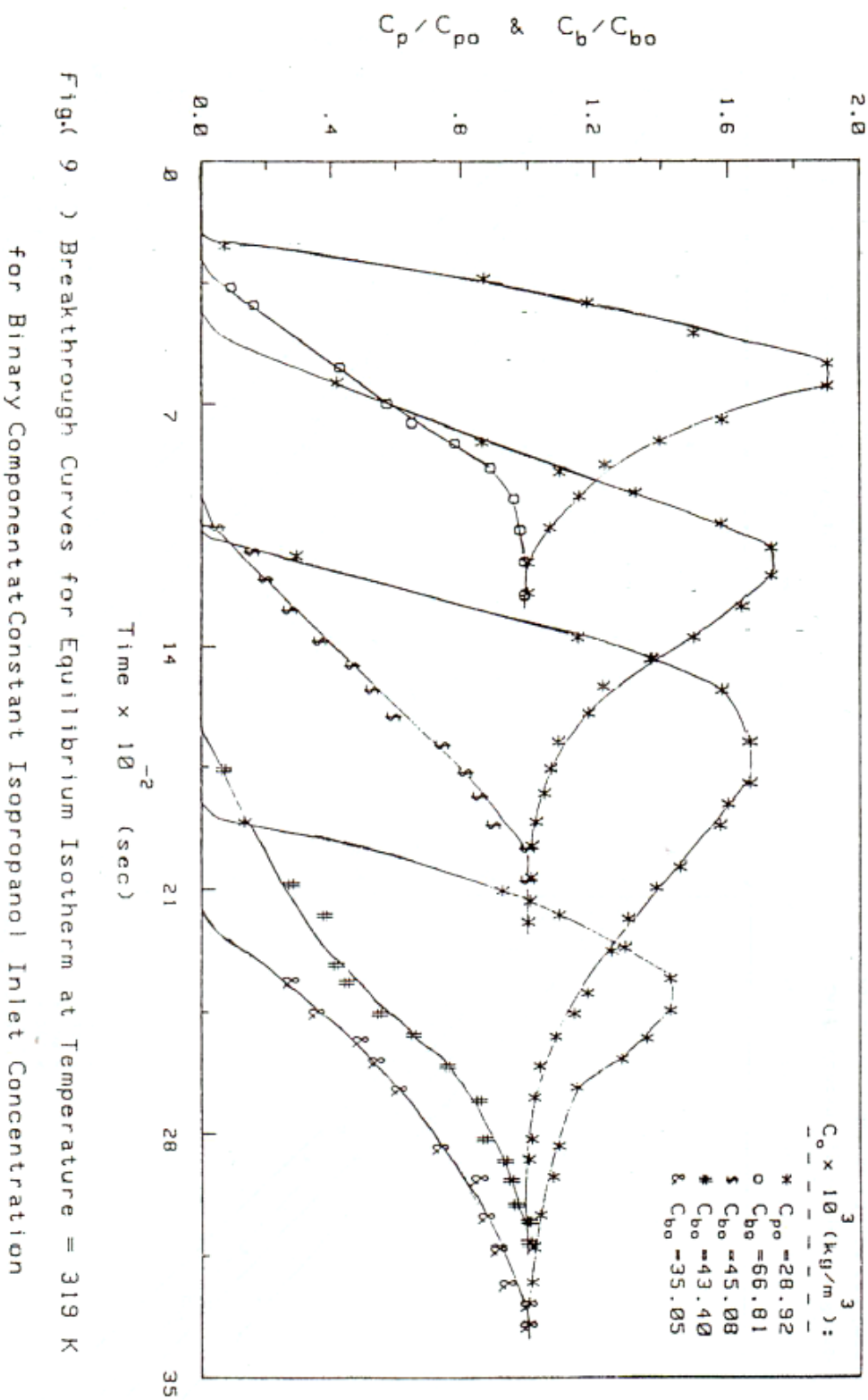


Fig.(9) Breakthrough Curves for Equilibrium Isotherm at Temperature = 319 K for Binary Component at Constant Isopropanol Inlet Concentration

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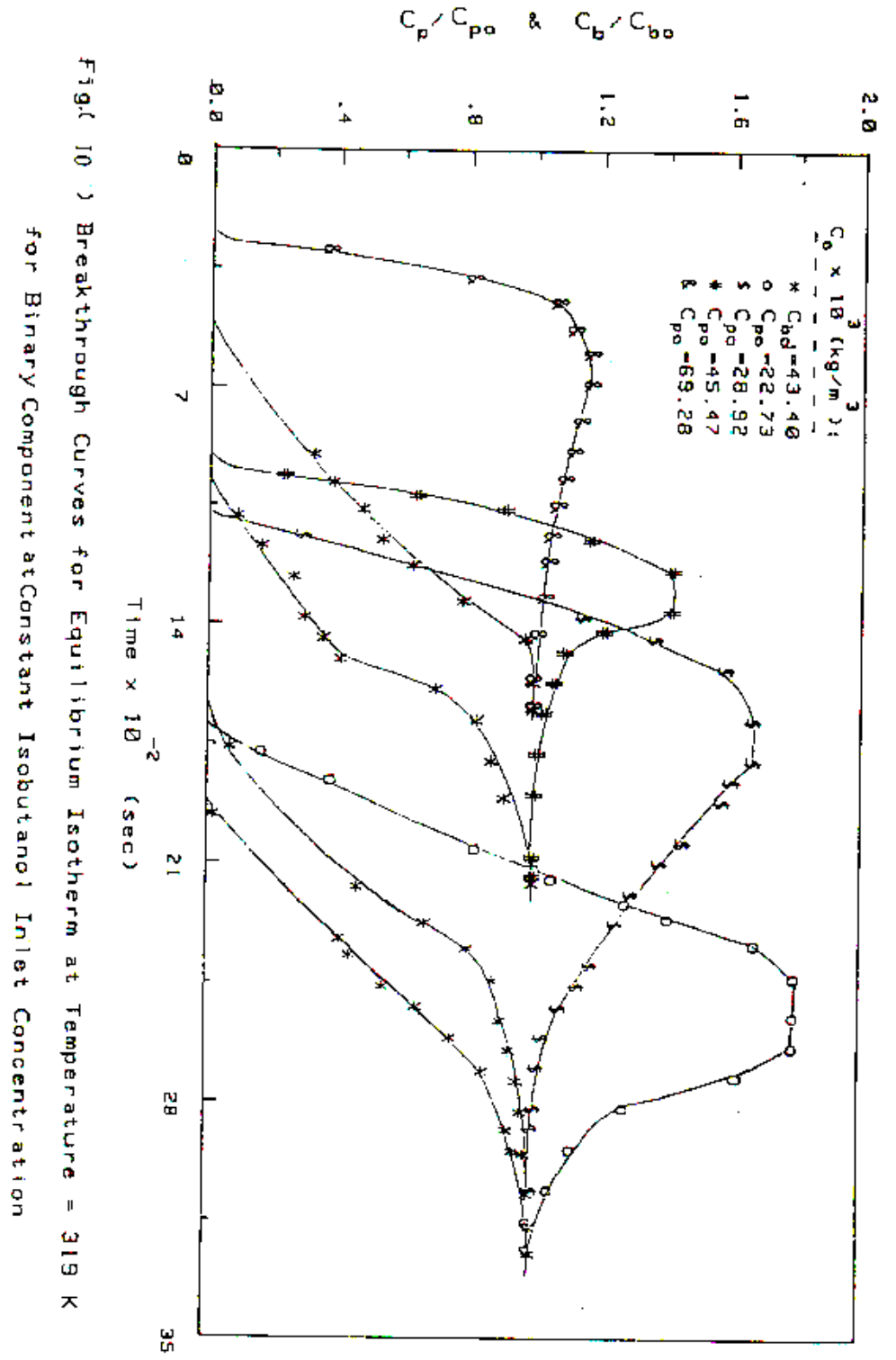

Fig.(10) Breakthrough Curves for Equilibrium Isotherm at Temperature = 319 K for Binary Component atConstant Isobutanol Inlet Concentration

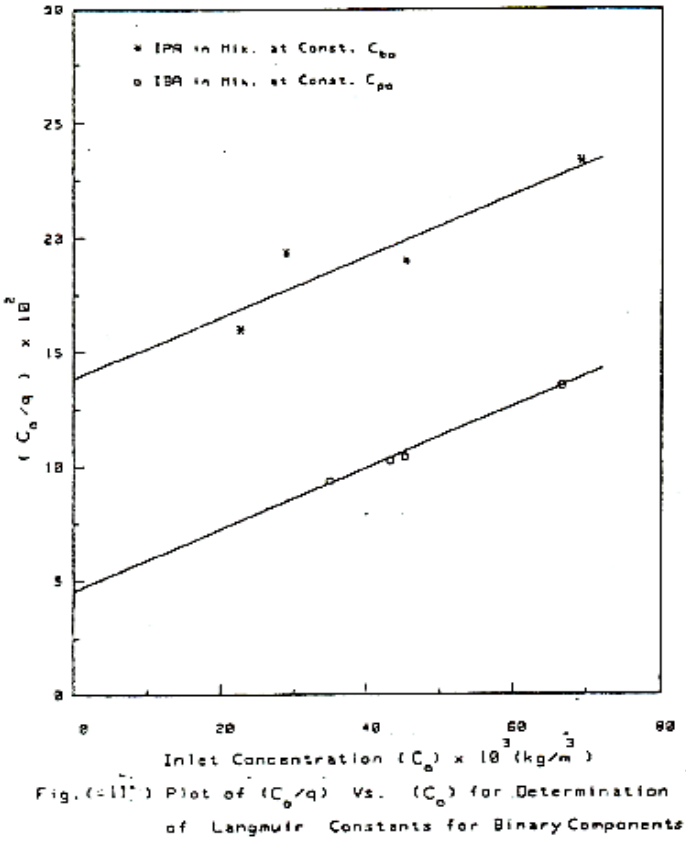

Fig.(11) Plot of (C_o/q) Vs. (C_o) for Determination of Langmuir Constants for Binary Components

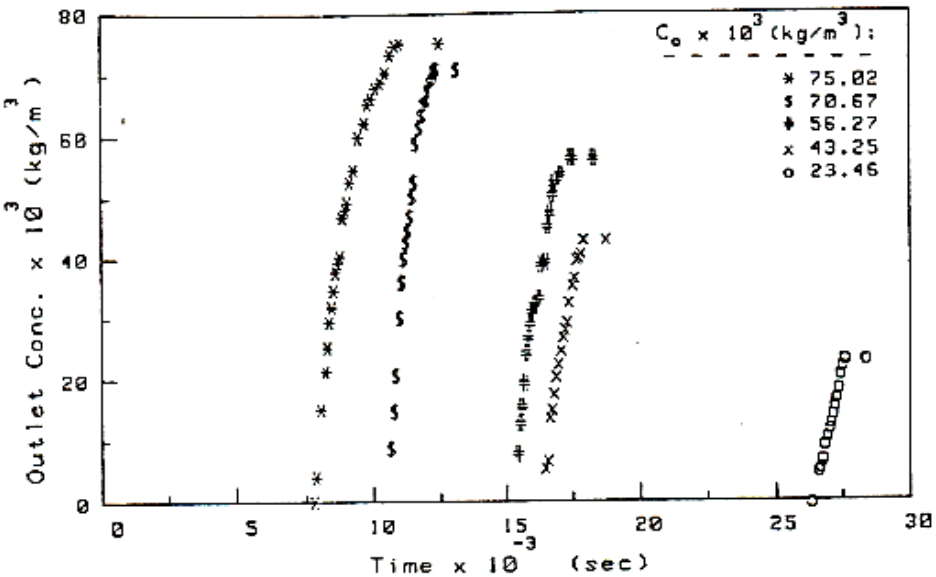

Fig.(12) Breakthrough Curves for Non-Isothermal Adsorption of Isopropanol on Activated Carbon at Bed Height = 0.35 m

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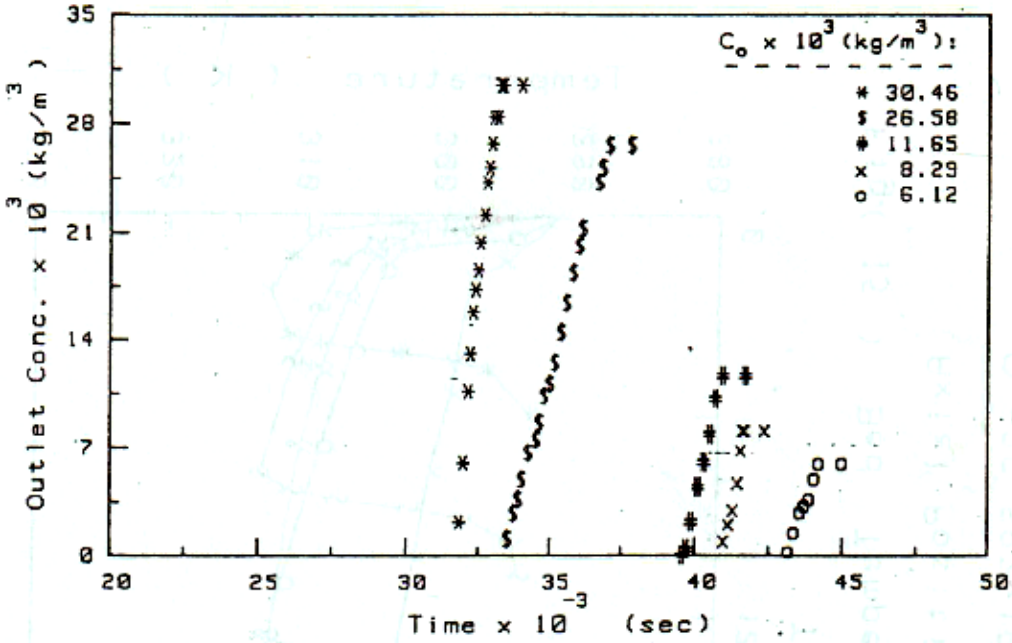

Fig.(13) Breakthrough Curves for Non-Isothermal Adsorption of Isobutanol on Activated Carbon at Bed Height = 0.35 m

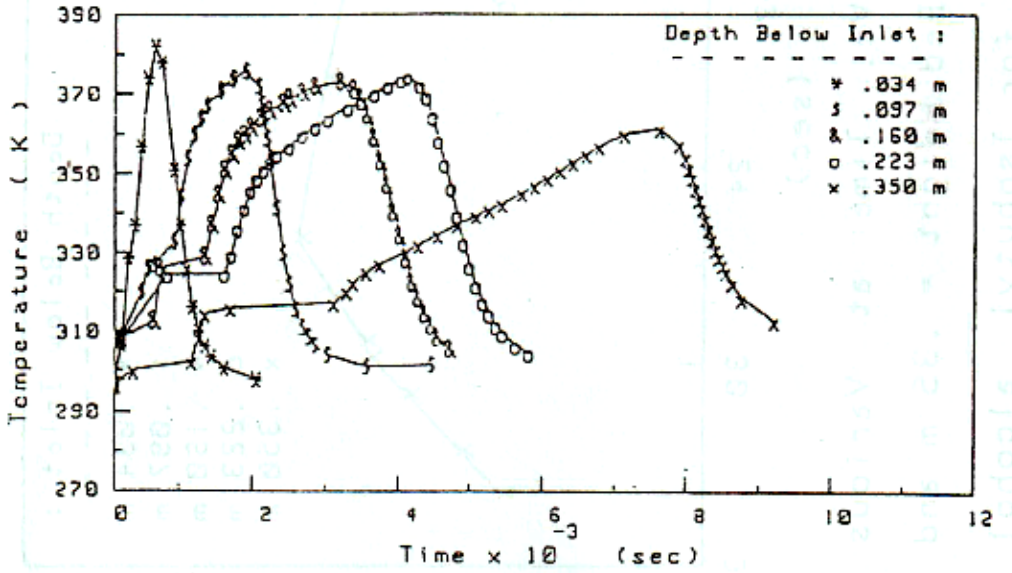

Fig.(14) Bed Temperature Vs. Time at Various Axial Positions for Bed Height = .35 m and C_o =75.021x10^{-3} (kg/m^3) for Isopropyl alcohol

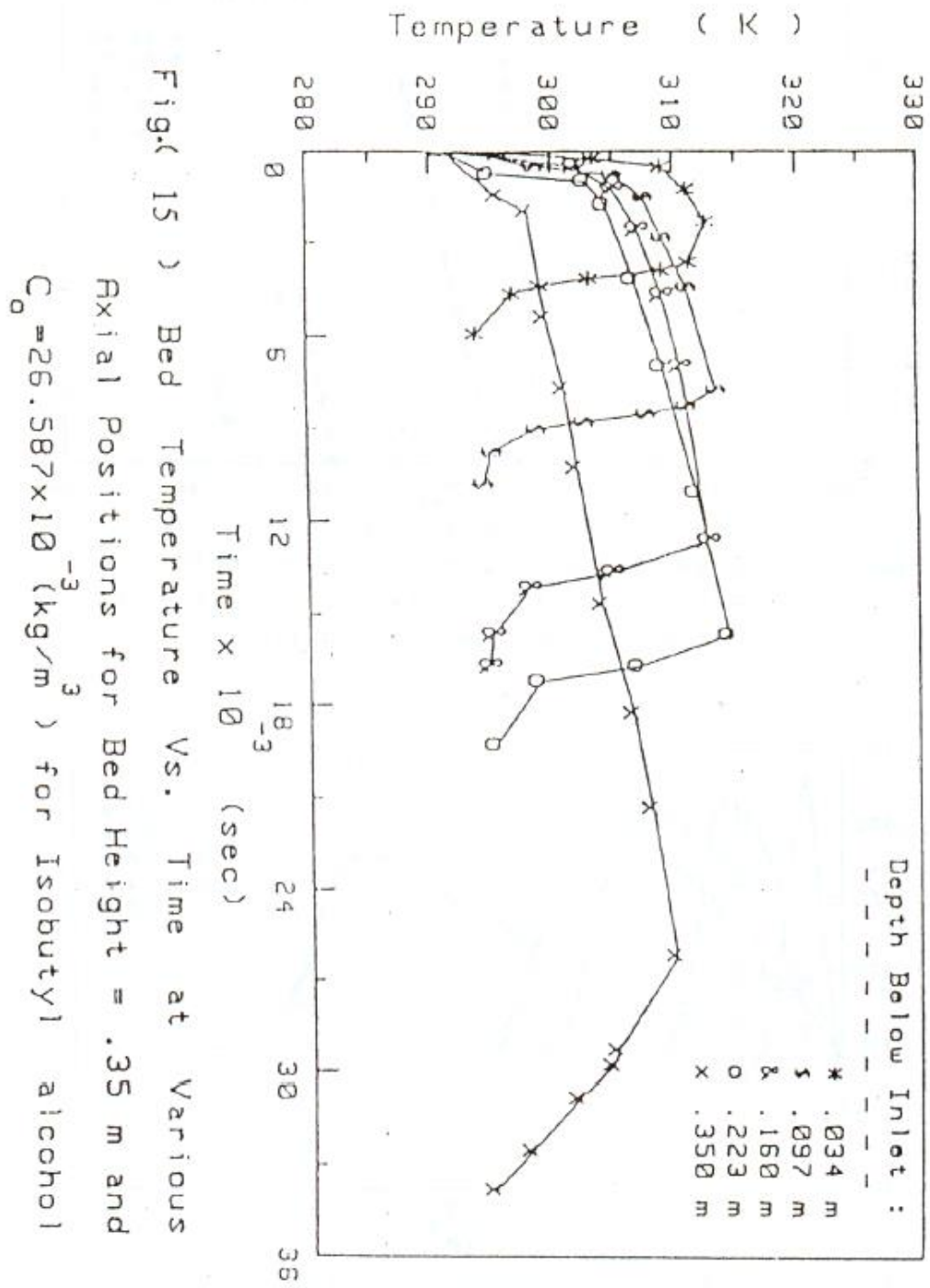

Fig.(15) Bed Temperature Vs. Time at Various Axial Positions for Bed Height = .35 m and C_o =26.587x10^{-3} (kg/m^3) for Isobutyl alcohol

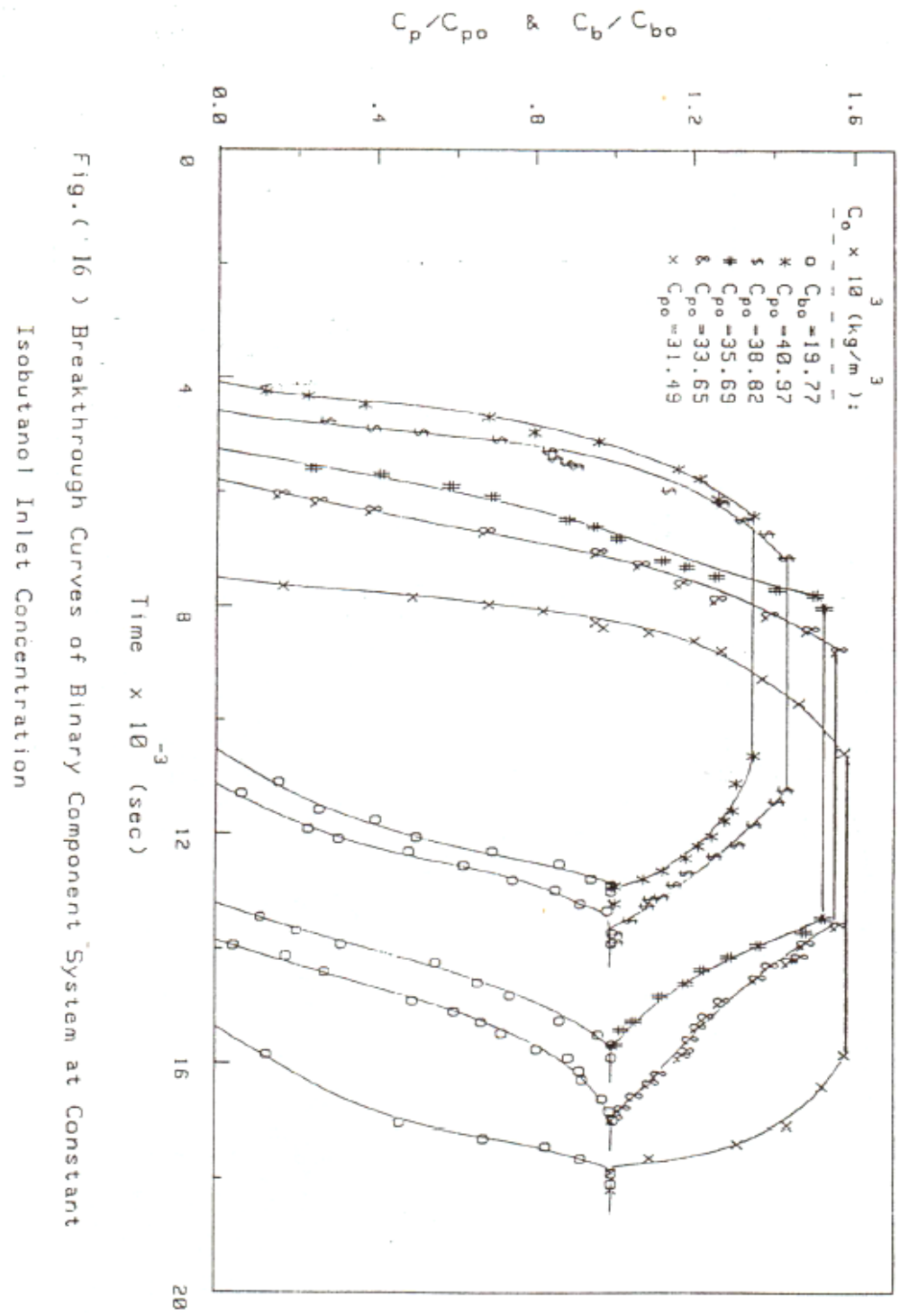

Fig.(16) Breakthrough Curves of Binary Component System at Constant Isobutanol Inlet Concentration

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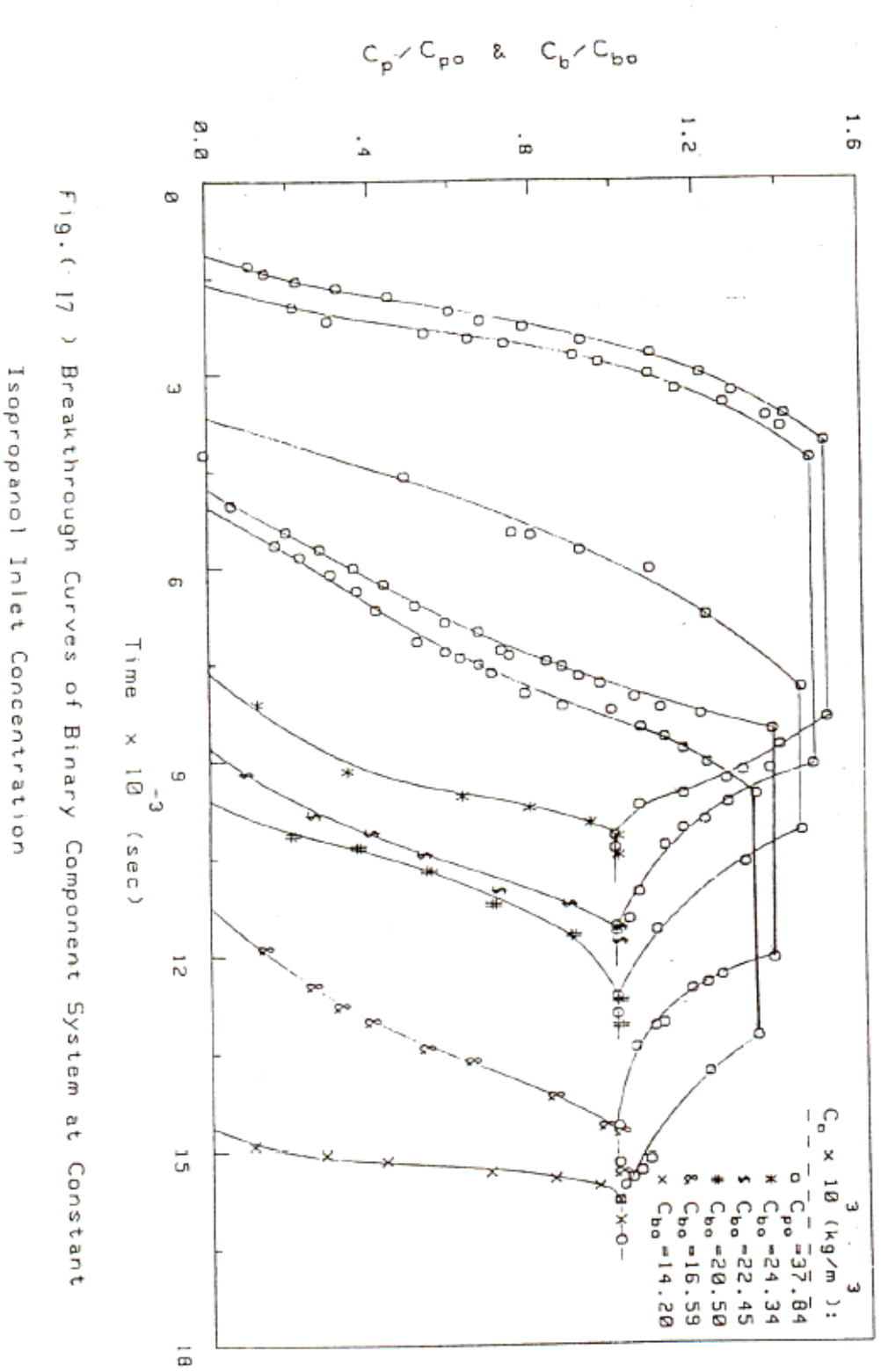

Fig.(17) Breakthrough Curves of Binary Component System at Constant Isopropanol Inlet Concentration