Tikrit Journal of Eng.Sciences /Vol.17/No.3/September2010,(13-21)

# *Carbon Dioxide Absorption in Packed Column in Non-Newtonian Fluid*

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## Abstract

Absorption of carbon dioxide into carbonate solution ($Na_2CO_3$) with PAM (non-Newtonian fluid) has been performed in a countercurrent packed column (0.075m i.d. *1.25 m height) packed with glass Raschig rings (1*1cm) to a depth of 1m. The influence of liquid flow rate, gas flow rate, liquid temperature, and polyacrylamind (PAM) concentration on the absorption rate, overall mass transfer coefficient and the reaction kinetics regime are studied at constant carbonate concentration and atmospheric pressure. The results show that the absorption rate and overall mass transfer coefficient increases with increasing liquid flow rate and temperature. The mass transfer coefficient decreases with increasing gas flow rate while the absorption rate of carbon dioxide is virtually independent of gas flow rate. This indicates that carbon dioxide absorption is liquid film controlled. Increasing PAM concentration results of reduction of absorption rate and overall mass transfer coefficient. The reaction kinetics between carbon dioxide and carbonate solution with PAM was obtained as a pseudo first order reaction (Hatta number, Ha >>1).
**Keywords: Carbon dioxide absorption, Non-Newtonian fluid, Mass transfer.**

## امتصاص ثاني اوكسيد الكاربون بواسطة سائل غير نيوتوني في برج امتصاص

**الخلاصة**

تضمن البحث دراسة عملية امتصاص غاز ثاني اوكسيد الكربون في خليط غازي( ثاني اوكسيد الكربون–هواء) باستخدام محلول يسلك سلوك غير نيوتوني "non-Newtonian fluid " والمتكون من كربونات الصوديوم وبولي اكرل امايند"Polyacrylamind (PAM)"بتراكيز مختلفة ليسلك المحلول سلوك غير نيوتوني.

تم دراسة العوامل المؤثرة على عملية الامتصاص لغاز ثاني اوكسيد الكربون وهي معدل جريان الغاز والسائل ، درجة حرارة السائل ،تركيز بولي اكرل امايند .وقد أوضحت النتائج العملية أن معدل الامتصاص لغاز ثاني اوكسيد الكربون و معامل انتقال الكتلة يزداد بزيادة كل من معدل جريان والسائل ، درجة حرارة السائل و تقل بزيادة تركيز بولي اكرل امايند.عند زيادة معدل جريان الغاز لم يظهر تأثير الزيادة على معدل الامتصاص ولكن ظهر تأثير طفيف جداً على معامل انتقال الكتلة. تم تحديد عدد هاتا "Hatta number" وبينت النتائج أن التفاعل تم عند الغشاء والمساحة السطحية هي العامل الأساسي المؤثر.
**الكلمـــات الداله:امتصـــاص ثـــاني اوكســـيد الكـــاربونًٌَُ، ســـائل غيرنيوتـــوني، انتقـــال الكتلـــه**

**Nomenclature**:

$C_B$: Concentration of liquid reactant (B) in the bulk, kmol/m$^3$

$C_{B0}$: Initial concentration of liquid Reactant (B), kmol/m$^3$

$D_{AB}$ : Diffusivity of carbon dioxide in Carbonate solution, m$^2$/s

$D_{AB,L}$ : Diffusivity of carbonate in pure Water, $m^2/s$
d: Column diameter, m
G´: Inert gas mass flux, $kmol/m^2.s$
H: Henry's constant, $atm.m^3/kmol$
$H^o$: Henry's constant in pure water, Atm.m3/kmol
Ha: Hatta number, (-)
I: Ionic strength, $Kmol/m^3$
$J_D$: Correlation factor, (-)
$K_G.a$: Overall mass transfer coefficient, $Kmol/atm.m^2.s$
$k_L^\circ$: Liquid side mass transfer coefficient, m/s
$k_{O\overline{H}}$ : Reaction rate constant of $O\overline{H}$ ion, $m^3/kmol.s$
$k_2$: Reaction rate constant, $m^3/kmol.s$
L: Liquid flow rate: m3/s
$L_m$: Liquid mass flux, $kmol/m^2.s$
N: Absorption rate, Kmol/m
m: Morality, $kmol/m^3$
P: Pressure, atm
T: Temperature, K
u: Superficial velocity, m/s
$u_L$: Superficial liquid velocity, m/s
$Y_1$: Outlet mole ratio, mole $CO_2$/mole air
$Y_2$: Inlet mole ratio, mole $CO_2$/mole air
$y_1$: Outlet mole fraction: mole $CO_2$/ (mole air+mole $CO_2$ )
$y_2$: Inlet mole fraction: mole $CO_2$/ (mole air mole $CO_2$ )
y: Fractional conversion, (-)
Z: Height of packing, m

**Greek symbols**

ε Void fraction, (-)
μ Viscosity, kg/m.s
ρ Density, $kg/m^3$
γ Constant in $eq^n$ (4), (-)

**Dimensionless Groups**

H a Hatta number $\frac{\sqrt{D_{AB}k_{O\overline{H}}C_{B\circ}}}{k_L^\circ}$

$N_{Re}$ Reynolds number $\frac{\rho u d}{\mu}$

$N_{Sc}$ Schmidt number $\frac{\mu}{\rho D_{AB}}$

## Introduction

The conventional gas-liquid contactors employed in acid gas absorption are packed tower, spray column, bubble column and fluidized bed have significant limitations in mass transfer [1], where the mass transfer of gas into liquid is often the rate limiting step [2]. Gas-liquid mass transfer in Newtonian and non-Newtonian fluid is an important issue of gas absorption as in carbon dioxide removal in ammonia production, natural gas treatment hydrogen production and fermentation broth for instance [3-5].The behavior of acid gas absorption (i.e. gas-liquid mass transfer) in non-Newtonian fluid is studied [3-9].

Jiao et al. [6] and Alvarez et al. [7], proved the important effect upon the mass transfer caused by the apparent viscosity of the liquid phase .Furthermore, it is necessary to include the rheological behavior in a serious study concerning the modeling of the absorption process.

Park and co-worker [3-5, 8-9] used different kinds of non-Newtonian fluids such as polyacrylamind (PAM), carboxymethylcellulose (CMC), polyethylene oxide (PEO), polybutene with polyisobutylene(PB,PIB) and xanthan gum for the carbon dioxide absorption with chemical reaction in non-Newtonian fluids. They found a considerable reduction of liquid side mass transfer coefficient ($K_{La}$) and absorption rate is due to the visco-elasticity of the aqueous solution. Also they found that the elasticity effect of the non-Newtonian fluid on the absorption rate is stronger than the effect of viscosity. Where the elasticity of non-Newtonian fluid (polymer kinds and/or concentrations) may reduce or accelerates the rate of absorption of carbon dioxide relative to that of a Newtonian fluid based on the same values of viscosity. After pre-screening of pervious research of carbon dioxide absorption in non-Newtonian fluid were they carried out in an agitated vessel, bubble column and stirred tank.

The present study investigates the effect of non-Newtonian rheological behavior on the absorption rate of carbon dioxide with chemical reaction into aqueous polyacrylamind (PAM) containing sodium carbonate in packed column under different operational conditions; gas (0.9-1.6 liter/min) and liquid (2.5-7.5 liter/min) flow rates, polyacrylamind concentration (0.1, 0.3,1 wt%), and liquid temperature (15, 35,50oC) at constant sodium carbonate concentration (0.1M), 8 mol% carbon dioxide concentration and atmospheric pressure.

## Theoretical Background

The theory of gas-liquid absorption with chemical reaction is sufficiently established and known. In the present work the film theory will be used and this theory assumed that the mass transfer takes place by molecular diffusion through stagnant liquid layer [10]. The chemical reaction occur in this layer between an initially gaseous constituent "$CO_2$" A and a reactant B in an aqueous solution "$Na_2CO_3$" according to the following overall reaction in the ionic term**[11]** :-

$$CO_2 + CO_3^{-2} + H_2O \Leftrightarrow 2HCO_3^{-} \dots (1)$$

The above reaction (1), being tri-molecular, is evidently made up of a sequence of elementary steps. The first reaction is the hydration of carbon dioxide:-

$$CO_2 + H_2O \Leftrightarrow HCO_3^{-} + H^{+} \dots\dots\dots (2a)$$

$$CO_2 + OH^{-} \Leftrightarrow HCO_3^{-} \dots\dots\dots\dots\dots (2b)$$

These reactions are both followed by an instantaneous reaction, leading to the overall results represented by reaction (1):-

$$CO_3^{-2} + H^{+} \Leftrightarrow HCO_3^{-} \dots\dots\dots\dots\dots (3a)$$

$$CO_3^{-2} + H_2O \Leftrightarrow HCO_3^{-} + OH^{-} \dots\dots (3b)$$

The reaction (2) is very slow and may usually be neglected. While the reaction (3) is assumed to be the only one important reaction for the enhancement of mass transfer due to the local rate of reaction of carbon dioxide where it is proportional to the concentration of hydroxyl ion($OH^{-}$) rather than to the concentration of carbonate ion($CO_3^{-2}$)**[11]**.

## Experiment Setup

Carbon dioxide absorption experiments were conducted in a glass packed column with an inside diameter of 0.075m and packed with glass Raschig rings (1*1 cm) The packed height was 1m which give an adequate ratio of column diameter to packing height 8 to 1, to make the flow in a continuous phase and avoid channeling effects in the packed column. A schematic diagram of the experimental apparatus employed throughout this work shown in figure (1). The liquor and gaseous mixture flow rates introduced at the top and bottom of the column respectively through the distributors which were installed at the top and bottom of the packed section. The spent absorbent discharged from bottom of the column through a pipe connected to three ways valve which is manually operated and the samples for the spent absorbent were taken from the column through this sampling device. The feeding tank was equipped with immersion heater and controller for heating the liquor to the desired temperature.

## Physicochemical and Rheological Properties

**1.**Density **(ρ)** of the aqueous solution of PAM solution containing sodium carbonate were measured at 15, 35 and 50$^{o}$C within 0.1-0.115 kg/m$^{3}$ by weighting with a pycnometer (Fisher Scientific Co., USA) and found to be identical within experimental accuracy.

**2.**Apparent viscosity (**μ**) of aqueous solution of PAM containing sodium carbonate and that of water ($\mu_w$) was measured with a Brookfield viscometer (Brookfield Eng. lab. Inc, USA).

To estimate the rheological properties of aqueous PAM solution, power- law model is used.

$$\tau = K\dot{\gamma}^{n} \quad \text{.... (4)}$$

$$\mu = K\dot{\gamma}^{n-1} \quad \text{... (5)}$$

Where n "flow behavior index", K "consistency index" parameters depending on the temperature. These parameters can be obtained from the measurement of shear stress "$\tau$"and shear rate "$\dot{\gamma}$", using a parallel disk type rheometer "0.19mm inner cylinder diameter and 0.25mm outer cylinder diameter". By statistical analysis to equation (4) the parameters n and K can be obtained to estimate the viscosity.

**3.**Diffusivity ($D_{CO2-Na2CO3}$) of carbon dioxide in aqueous sodium carbonate solution was estimated by $CO_2$-$N_2O$ analogy and modified Stonkes-Enstein relation as follows[12]:-

$$\frac{D_{CO_2-Na_2CO_3}}{D_{CO_2,w}} = \frac{D_{N_2O}}{D_{N_2O,w}} \quad \text{... (6)}$$

The diffusion coefficients ($D_{CO_2,w}, D_{N_2O,w}$) of $CO_2$ and $N_2O$ in water are obtained as follows [12-14]:-

$$D_{CO_2,w} = 2.35\times10^{-6}\exp\left(\frac{-2119}{T}\right) \quad \text{... (7)}$$

$$D_{N_2O,w} = 5.07\times10^{-6}\exp\left(\frac{-2371}{T}\right) \quad \text{... (8)}$$

The diffusion coefficient ($D_{N_2O}$) of $N_2O$ in aqueous carbonate solution is obtained using Stonkes-Enstein equation [12]:-

$$D_{N_2O,w}\,\mu_w^{\gamma} = D_{N_2O}\,\mu^{\gamma} \quad \text{... (9)}$$

Where $\gamma$ =0.6 for carbonate solution [12].

The diffusion coefficient of carbonate $D_{Na_2CO_3}$ in aqueous carbonate solution is obtained from the assumption that the ratio of $D_{Na_2CO_3}$ to $D_{CO_2-Na_2CO_3}$ be equal to the ratio in water [3-5, 8-9]:-

$$\frac{D_{Na_2CO_3}}{D_{CO_2-Na_2CO_3}} = \frac{D_{Na_2CO_3,w}}{D_{CO_2,w}} \quad \text{... (10)}$$

Where the diffusivity of carbonate in water ($D_{Na_2CO_3,w}$) can be estimated from wilke and Chang equation [15]:-

$$D_{AB,L} = 7.4*10^{-8}\left(\frac{M_{wt,B}^{0.5}T}{\mu_B \tilde{V}_A^{0.6}}\right) \quad \text{... (11)}$$

**4.** Reaction rate constant (**k2**) In the reaction of carbon dioxide with carbonate, the reaction rate constant is estimated as follows[16,17]:-

$$\log k_2 = 13.635 - \frac{2895}{T} \quad \text{... (12)}$$

Where, $k_2 = k_{\overline{OH}}$ ,when carbon dioxide is absorbed in carbonate solution the rate of reaction of carbon dioxide is proportional to the concentration of $O\overline{H}$ rather than to the concentration of $CO_3^{=}$ [16-18].

**5.**Solubility of carbon dioxide in the liquid phase was calculated using the Henry's law.

Henry's law constant for $CO_2$-$Na_2CO_3$ system was determined as follows [17]:-

$$\log\frac{H}{H^o} = 0.088I \quad \text{... (13)}$$

Where $H^o$ is Henry's law constant for $CO_2$ in water and can be obtained as follows [19]:-

$$\log\left[H^o\right]^{-1} = -4.3856 + \frac{867.4932}{T} \quad \text{... (14)}$$

and I is the ionic strength and is obtained as follows [20]:- I=m (2-y)

**6.** Liquid side mass transfer coefficient $k_L^o$ in physical absorption can be determine by using Wilson and Geomkoplis correlation as follows[21]:-

$$J_D = 0.79 * \frac{0.25}{\varepsilon} * N_{Re}^{-0.31} = \frac{k_L^o}{u_L} N_{Sc}^{2/3} \ldots\ldots (15)$$

## Results and Discussion

Absorption rate of carbon dioxide into carbonate solution with PAM calculated from the material balance with chemical reaction as follows [22]:-

$$G^{/} \frac{y_2}{1-y_2} + L_m C_B = G^{/} \frac{y_1}{1-y_1} + L_m C_{B0} \quad (16)$$

Carbon dioxide absorbed

N=L ($C_B C_{Bo}$)…………………….….... (17)

Carbonate conversion%

$$= \frac{[Na_2CO_3]\ reacted}{[Na_2CO_3]\ input} \ldots\ldots\ldots\ldots (18)$$

Overall mass transfer coefficient based on the gas phase $K_G a$ can be calculated as follows [18]:-

$$K_G.a = \frac{G'}{PZ} Ln \frac{Y_2}{Y_1} \ldots\ldots\ldots\ldots (19)$$

Hatta number [23]:-

$$Ha = \frac{\sqrt{D_{AB} k_{O\bar{H}} C_{B\circ}}}{k_L^\circ} \ldots\ldots\ldots\ldots (20)$$

## Effect of Gas and Liquid Flow Rate

Figures (2-3) illustrates the effect of liquid flow rate at different liquid temperature and a given PAM concentration on the absorption rate of carbon dioxide and overall mass transfer coefficient. It is evident that the absorption rate increases with increasing liquid flow rate. This indicates that carbon dioxide absorption is liquid film controlled. Also, the overall mass transfer coefficient increases with increasing liquid flow rates, this is due to decrease in the effective liquid film thickness, hence the concentration gradient of free carbon dioxide in the liquid will be grater, leading to increase the absorption rate and mass transfer coefficient.

From figures (4-5) it can be noticed that the mass transfer coefficient decreases with increasing gas flow rate. Moreover, the absorption rate of carbon dioxide is virtually independent of gas flow rate. This further confirms that carbon dioxide absorption is liquid film controlled. These results are in agreement with Creyder and Maloney [24], and Yih and Sund [25].

## Effect of Liquid Temperature

The effect of liquid temperature on the absorption rate and mass transfer coefficient of carbon dioxide are shown in figures(2-5) .As can be seen from these figures , that the absorption rate and mass transfer coefficient increases rapidly with increasing temperature. Although an increase in liquid temperature will decrease the equilibrium of carbon dioxide in the absorbent "solvent", it will also decreases the viscosity of the solution and increase the rate of chemical reaction. Thus the increase of absorption rate of carbon dioxide with temperature implies a more predominate increase of reaction rate with temperature and further supports the theory that carbon dioxide absorption is liquid phase mass transfer controlled.

These results are in agreement with findings of Creyder, Maloney [24], Yih, Sund [25] and Farah [20]. Figure (6) shows the effect of liquid feed temperature on the outlet carbon dioxide mole ratio, it can be seen that the outlet mole ratio of carbon dioxide decreased with increasing liquid feed temperature results from increasing absorption rate.

## Effect of PAM Concentration

Figures (7-8) show the effect of PAM concentration on the absorption rate of carbon dioxide and mass transfer coefficient. As can be seen from these figures, that absorption rate and mass transfer coefficient decreases with an increase of PAM concentration. This behavior has been attributed to the effect of elasticity of PAM which reduces the absorption rate of carbon dioxide and mass transfer coefficient. Similar results have been obtained in the previous studies [2-9], which employing different systems and contactors (agitated vessel, stirred tank, and bubble column).

## Reaction Kinetic Type and Regime

In the present work, Hatta number was estimated and found greater than one (Ha >>1), see Table (1.b). This indicates that all the reaction occurs within the film and surface area is the controlling factor [21, 24]. , when Ha >>1, the reaction kinetics of carbon dioxide with carbonate solution undergoes a pseudo first order reaction [23, 26] (i.e. where the concentration of the reactant in the neighborhood of the surface is very little different from that in the bulk of the liquid, and the dissolved gas undergoes a pseudo first order reaction). Further more, the following conditions are also satisfied [17] (see Table 1.b):-

$$\sqrt{D_{AB} k_{\overline{OH}} C_{B\circ}} \geq 5 * k_L^{\circ} \qquad (21)$$

## Conclusions

**1-**Carbon dioxide absorption into carbonate solution with PAM (non-Newtonian solution) can be regarded as a liquid film mass transfer controlled process, in which the absorption rate and mass transfer coefficient increases with increasing liquid flow rate and temperature, the mass transfer coefficient decreases with increasing gas flow rate while the absorption rate of carbon dioxide is independent of gas flow rate.

**2-**The elasticity of PAM reduces the absorption rate and mass transfer coefficient.

**3-**The reaction occurs within the film and surface area is the controlling factor, and the reaction kinetics undergoes a pseudo first order reaction.

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| | |
|---|---|
| 1 | Packed column |
| 2 | Rota meter |
| 3 | $CO_2$ Cylinder |
| 4 | Air Compressor |
| 5 | Liquid Pump |
| 6 | Feeding Tank |
| 7 | Needle valve |
| 8 | Liquid distributor |
| 9 | Gas distributor |
| S | Sample point |

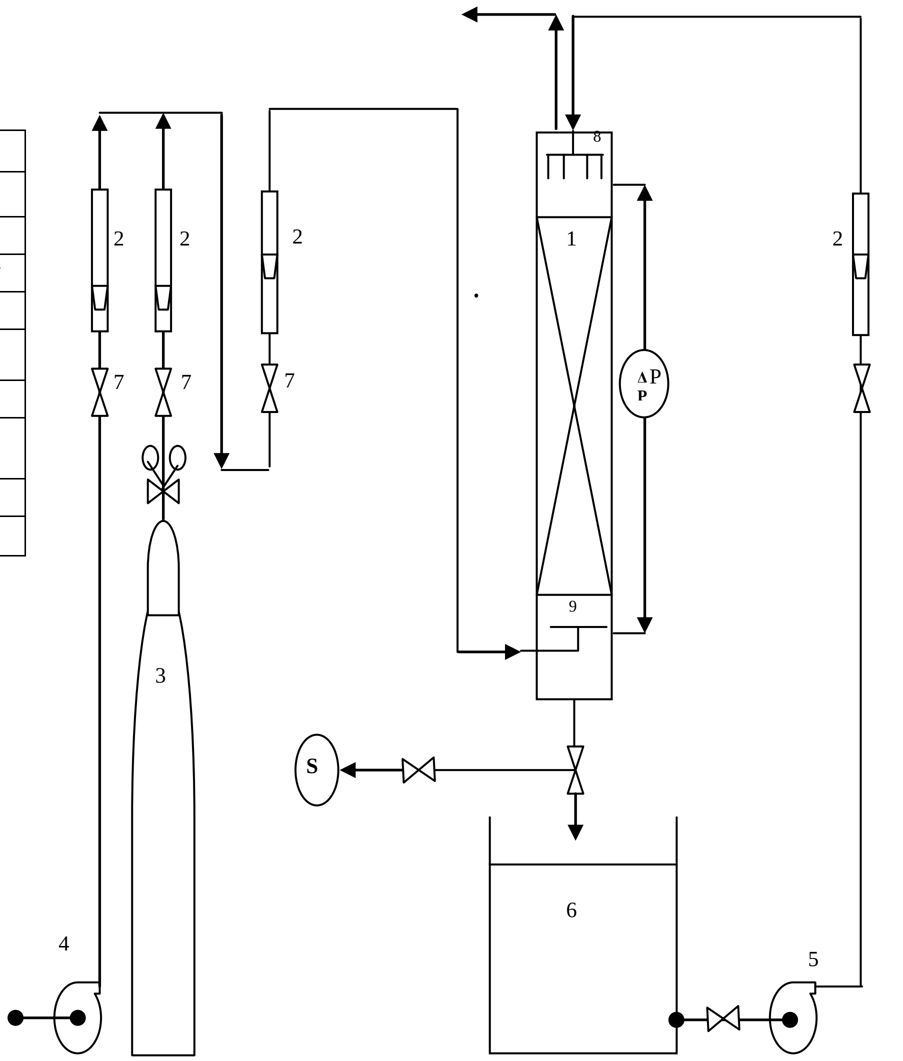


**Figure (1): Schematic diagram of experimental apparatus.**

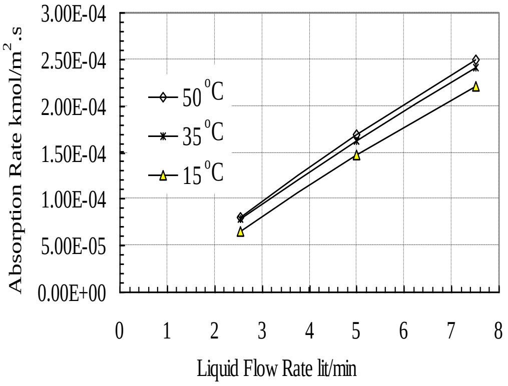


**Figure (2).Absorption rate vs. liquid flow rate at different liquid feed temperature, 30 lit/min gas flow rate and 0.1 wt% PAM concentration.**

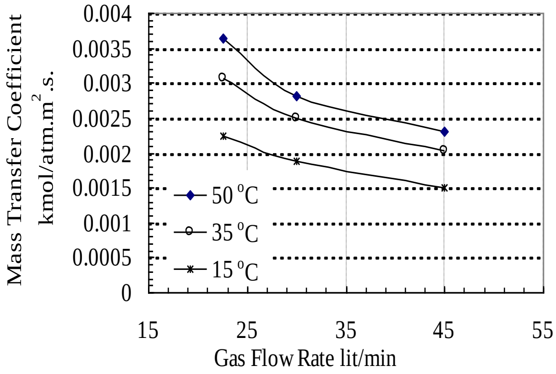


**Figure (5). Mass transfer coefficient vs. Gas flow rate at different liquid feed temperature, 5lit/min liquid flow rate and 0.3 wt% PAM concentration.**

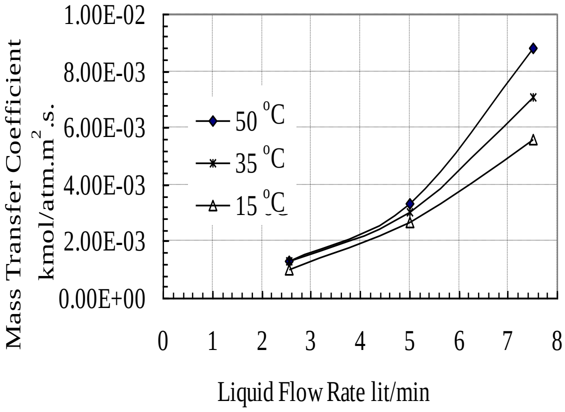


**Figure (3).Mass transfer coefficient vs. liquid flow rate at different liquid feed temperature, 30 lit/min gas flow rate and 0.1 wt% PAM concentration.**

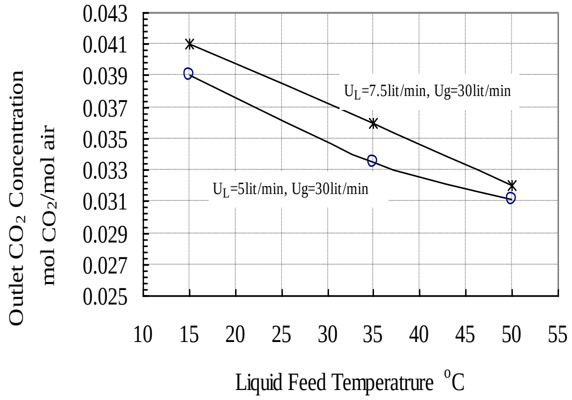


**Figure (6).Outlet $CO_2$ mole ratio vs. liquid feed temperature at different liquid flow rate, and 0.1 wt% PAM concentration.**

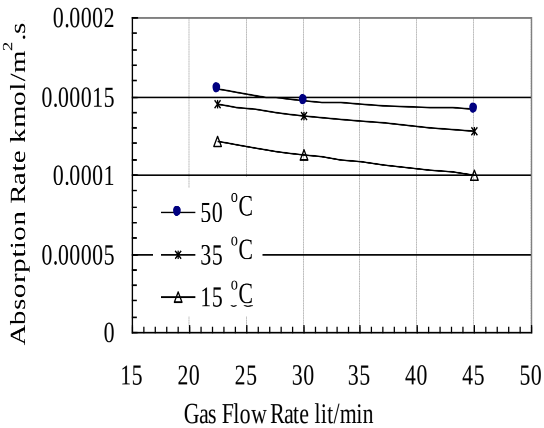


**Figure (4). Absorption rate vs. Gas flow rate at different liquid feed temperature, 5lit/min liquid flow rate and 0.3 wt% PAM concentration.**

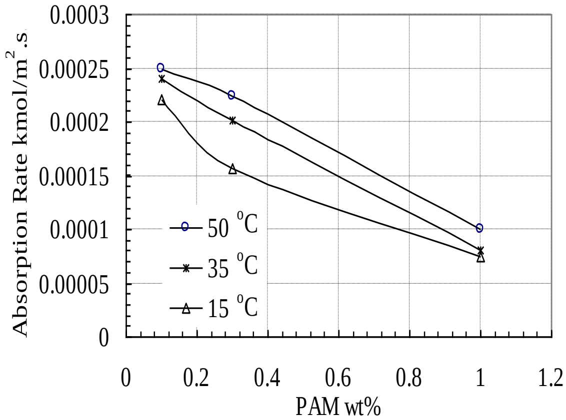


**Figure (7): Absorption rate vs. PAM concentration at different liquid feed temperature, 7.5 lit/min liquid flow rate and 45 lit/min gas flow rate.**

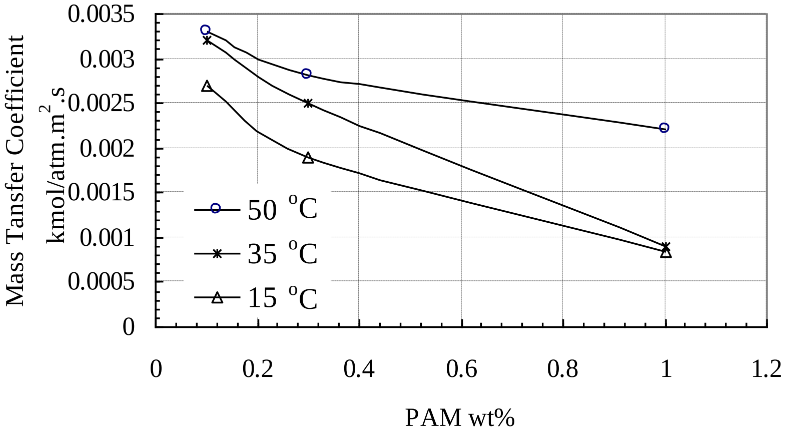


**Figure (8), Mass transfer coefficient vs. PAM concentration at different liquid feed temperature, 5 lit/min liquid flow rate and 30 lit/min gas flow rate.**

**Table (1.a): Physicochemical properties.**

| T $^0$C | $D_{CO_2-Na_2CO_3}$ | $k_{O\overline{H}}$ | H | $\sqrt{D_{AB}k_{O\overline{H}}C_{B\circ}}$ |
|---|---|---|---|---|
| 15 $^0$C | 2.55868E-09 | 3827.512 | 23.766 | 0.000989 |
| 35 $^0$C | 3.54687E-09 | 17204.788 | 37.287 | 0.002470 |
| 50 $^0$C | 4.38031E-09 | 47005.827 | 50.392 | 0.00453 |

**Table (1.b): Results of Hatta number and liquid side mass transfer coefficient.**

| T $^0$C | Ha | $k_L^\circ$ | $\sqrt{D_{AB}k_{O\overline{H}}C_{B\circ}}$ | $5*k_L^\circ$ |
|---|---|---|---|---|
| $u_L$=2.5 lit/min | | | | |
| 15 $^0$C | 25.898 | 3.82108E-05 | 0.000989 | 0.000191 |
| 35 $^0$C | 49.246 | 5.01617E-05 | 0.002470 | 0.000250 |
| 50 $^0$C | 75.647 | 5.99837E-05 | 0.00453 | 0.000299 |
| $u_L$=5 lit/min | | | | |
| 15 $^0$C | 21.677 | 4.56507E-05 | 0.000989 | 0.000228 |
| 35 $^0$C | 41.22 | 5.99286E-05 | 0.002470 | 0.000299 |
| 50 $^0$C | 63.318 | 7.16631E-05 | 0.00453 | 0.000358 |
| $u_L$=5 lit/min | | | | |
| 15 $^0$C | 16.053 | 6.16448E-05 | 0.000989 | 0.000308 |
| 35 $^0$C | 30.525 | 8.09251E-05 | 0.002470 | 0.0004046 |
| 50 $^0$C | 46.890 | 9.67707E-05 | 0.00453 | 0.000483 |