Calculation of effective interfacial area in a Turbulent Contact Absorber

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Abstract—A Turbulent contact absorber (TCA) column has been installed and operated at Petroleum University of Technology (PUT) to absorb CO2 using caustic solution. In order to survey column efficiency, calculation of mass transfer coefficients (kl, kg) and interfacial area (a) is necessary. Generally because of measurement problems, these parameters are expressed as overall mass transfer coefficient (Koga). CO2 absorption by aqueous solutions (such as caustic) is considered as chemical absorption which takes place in liquid boundary layer and the rate of absorption is a severe function of gas-liquid interfacial area. Through variation of system specifications such as caustic concentration, gas rate, liquid rate and liquid to gas ratio (L/G), which resulted from 70 practical experiments with various operating conditions, subordination of effective interfacial area was investigated; a direct predictive method based on chemical absorption was presented to calculate effective interfacial area; and best operating conditions for TCA column was concluded. The final results from practical experiments illustrated that, at low L/G ratios in absorption processes, using a TCA column whose cross sectional area and packing height is about 0.1 of same parameters in a packed column which operates at the same conditions, five times efficiency can be yielded.

Index Terms—CO₂ absorption, Effective interfacial area, TCA, Three phase fluidized bed

I. INTRODUCTION

Gas absorption using an appropriate solution is one of the most important processes in chemical and petrochemical industries. The process is based on mass transfer through gas-liquid boundary layer. In order to obtain maximum absorption efficiency, it is necessary to utilize proper equipments to maximize gas-liquid contact.

Packed and Tray columns are generally used in absorption processes; nonetheless, when absorption concerns chemical reaction, regarding reaction kinetics and Stoicheiometry, it is necessary to lower L/G ratio. Lowering L/G ratio causes canalization phenomenon which is due to dried spaces in column. Canalization severely reduces mass transfer efficiency [2].

Tray columns are not economical in such conditions. Liquid holdup is high in tray columns and when solution is valuable, increased regeneration costs will affect on total

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absorption cost.

Turbulent contact absorbers (TCA) are new columns which utilize three phase fluidized beds. These columns are operated in fully fluidized state. Packing, having no chemical effect on absorption process, maximizes gas-liquid mixing and renewal. Gas and liquid flow counter currently and packing fill about 20% of total column height. When gas rate is increased, packing start to fluidize and in further rates, fully fluidized state is yield.

TCA columns are used in distillation, absorption and stripping processes. Turbulency and high phase mixing in TCA column cause higher mass transfer efficiency for TCA columns in comparison with packed and tray columns. TCA columns are used in conditions where L/G ratio is to be held low.

A number of models have been developed for prediction of gas-liquid contact area. The widely used Onda et al. correlation assumes the contact area cannot exceed the available packing surface area [3]. The model of Djebbar and Narbaitz [4] is a modification of the model proposed by Onda and coworkers. Bravo and Fair [5], Henriques de Brito et al. [6], Billet and Schultes [7], and Piche et al. [8] have proposed models for the prediction of gas-liquid contact area. Unfortunately, these models have been based on back-calculated areas assuming models for gas and liquid mass transfer coefficients. In addition, no model is presented to predict gas-liquid contact area in TCA columns.

The present work attempts to introduce a method for calculation of effective gas-liquid interfacial area in TCA columns; meanwhile, assuming CO_2 absorption using caustic solution as liquid controlling chemical absorption, thus, using two-film theory, a direct predictive method for effective interfacial area calculation is presented; afterward, system parameters were changed to investigate the effect on effective interfacial area.

II. CALCULATION OF GAS-LIQUID CONTACT AREA

The effective mass transfer area may be estimated using a reactive absorption system such as air- CO_2 -caustic. The absorption system can be described using two-film theory, where the liquid phase mass transfer coefficient is corrected for the chemical reaction [9]:

$$\frac{1}{K_{og}a} = \frac{1}{k_g a} + \frac{H}{\beta k_o^l a}$$
(1)

Where β is enhancement factor and k_o^l is physical mass transfer coefficient.

For the case of an irreversible, pseudo first order reaction, the overall gas phase volumetric coefficient Koga may be

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rewritten [10]:

$$\frac{1}{K_{og}a} = \frac{1}{k_g a} + \frac{H}{\sqrt{D_{AB}C_B k_r}} a$$
(2)

In general, for low concentrations of sodium hydroxide (CB) and high gas velocities, the liquid phase resistance dominates and the gas phase resistance can be neglected. As a result:

$$K_{og}a = \frac{a.\sqrt{D_{AB}.C_B.k_r}}{H}$$
(3)

Therefore, the effective gas-liquid contact area can be deduced using measured Koga values and values of carbon dioxide diffusion coefficients (DAB), sodium hydroxide concentration (CB), the reaction rate constant (kr), and Henry's constant(H).

$$a = \frac{K_{og}a.H}{\sqrt{D_{CO_2}.C_B.k_r}} \tag{4}$$

III. EXPERIMENTAL SYSTEM

A three phase gas-liquid-solid fluidized bed system was designed and constructed to study the mass transfers of the packed and fluidized bed systems under various operating conditions at PUT Research Lab Pilot Plant.

In this section, the apparatus that was employed for mass transfer studies is described, then the methods of operation which were carried throughout the experiments are discussed.

The flow diagram of the experimental equipment is sketched in Fig. 1.



Fig. 1. Flow diagram of the experimental equipment

The test section consists of a Plexiglas column of 100 mm inside diameter with a total height of 180 cm. The glass

column was used for better visual observation of the phenomena occurring in the bed.

Air at room temperature was introduced to the bottom of the column. Liquid was admitted to the bed through a distributor which gave a uniform liquid distribution over the top of the bed.

The reciprocating compressor in this apparatus could push air to the column in once through mode. The solid materials which were used as solid phase were low density (LD) plastic balls with a diameter of about 15mm and a density of 335 kg/m3.These particles were supported by no flooding grids. The experiments were conducted in a single stage column in which the static bed height of packing was 20 cm.

The pressure drops in packed and fluidized beds for the whole range of gas rates were measured by means of a U-tube water manometer. A water manometer was used to measure pressure drop across the column. The liquid and gas flow rates were controlled by globe valves and their flows were measured by Rotameters with stainless steel and aluminum floats, respectively.

The liquid and gas phases were entered the column counter currently. The composition of carbon dioxide in the entering and leaving gas phase was measured by using a precise and calibrated gas analyzer.



Fig. 2. Gas Analyzer

The gas analyzer which was used in this work was PolytectorII gas tester. A picture of the Gas analyzer is also given in Fig. 2. and the general description of this equipment is given below:

A. Gas analyzer

The POLYTECTOR II is a handheld detector for personal protection from gas hazards. The built in pump allows taking gas samples from e.g. rooms, sewers or vessels to check before entering, if confined. With the pump deactivated, the POLYTECTOR II operates permanently in diffusion mode and gives a visual and audible warning, if gas hazards arise.

The POLYTECTOR II has been tested for the use in hazardous areas. The tests were based on the standards DIN EN 50054 "Electrical Devices for Tracing and Measuring of Combustible Gases-General Requirements and Test Method", DIN EN 50057" Electrical Devices for Tracing and Measuring of Combustible Gases-Requirements for the Operational Behavior of Devices of Group II with a Detection Range up to 100% of the Lower Explosion Limit ",DIN EN 50104 Devices for Tracing and Measuring of Combustible Oxygen-Requirements for the Operational Behavior and Test Methods", the guideline T017 of BG Chemie "Warning Devices for Hydrogen Sulfide", the guideline T022 of BG Chemie "Gas Warning Equipment for Landfill Application-Test of Functioning "and DIN EN 50271 "Electrical Devices for Detection and Measurement of Combustible Gases ,Toxic Gases or Oxygen-Requirements and Tests for Warning Equipment using Software and/or Digital Technology".

The Polytector G750 is a handy and compact gas detector for simultaneous monitoring of up to 6 gas hazards which are given below:

- Combustible gases (methane up to 100% LEL)
- Combustible gases (methane up to 100 Vol %)
- Oxygen deficiency or surplus
- Toxic gases (carbon monoxide)
- Toxic gases (hydrogen sulfide)
- Toxic gases (carbon dioxide)

The "Smart Sensor System "allows an easy change of the plug in sensors to adapt the POLYTECTOR II to new measurement tasks or to renew the sensors.

The POLYTECTOR II is easily operated with only a few keys, supported by the clear text in the display. All user functions are affected directly by means of the keys Pump on/off, Alarm Reset, Display illumination on/off. In the detection mode every key stands for one function so you do not need a long training to operate the POLYTECTOR II perfectly.

Depending on the gas to be monitored, the POLYTECTOR II uses different detection principles. Catalytic combustion and thermal conductivity are proven principles to measure combustible gases for explosion protection. Electrochemical sensor with different characteristics is used to measure a wide range of toxic gases and oxygen. The infrared sensor (IR) gives exceptional results for measuring carbon dioxide (CO₂).There fore we use only this analyzer for CO₂ gas and I explain detection principle of this:

The infrared sensor uses the characteristic of gases to absorb light in certain spectral ranges. The light from an infrared source passes the gas in the sensor cell, which reduces the radiation energy by absorption. The absorption of the infrared radiation in a certain wave length range is proportional to the concentration of the gas to be measured. The energy reduction of the infrared radiation is measured by means of detectors. Simultaneously the infrared light is measured in a different wavelength range, in which the gas does not cause any absorption to obtain a reference signal. This provides a high accuracy even if the light source changes or if the mirrors are dirty.

IV. EXPERIMENTAL PROCEDURE

A. Mass transfer

In order to study the principle of mass transfer of carbon dioxide from the gas phase to the liquid of various concentration of NaOH, the overall volumetric mass transfer coefficient, KGa, was measured for absorption of CO_2 from a mixture of air- CO_2 into different concentrations of NaOH.

These experiments commenced according to the following procedure:

NaOH solution of specified concentration was allowed

into the system. Then air was introduced at predetermined flow rate into the column. When the steady state conditions were established in the column carbon dioxide at specified concentration was mixed with the gas phase and introduced to the column. To determine the rate of mass transfer of Carbon dioxide into the liquid phase in the fluidized column, samples from the inlet and outlet of the gas phase at specified time intervals were taken and passed through the gas analyzer to determine the Carbon dioxide concentration at the inlet and outlet of system. To determine the effect of gas and liquid flow rates and state of the bed on mass transfer coefficient, the gas and liquid flow rate were varied over a wide range so that the state of the bed transferred from static condition to fully fluidized state.

To investigate on the effect of caustic concentration on the volumetric overall mass transfer coefficient, KGa, the above experiment was repeated for three different caustic concentrations, i.e. for 0.1, 0.5 and 1 normal (kmoles/m3).

V. MATERIALS AND RANGE OF OPERATION CONDITIONS

The packing material which was used in this investigation was polyethylene spherical. Physical properties of the packing are given in Table 1.

TABLE 1. THE PHYSICAL PROPERTY OF PACKING

Material	Shape	Density
Plastic	Spherical	335Kg/m3
	(Dp=15mm)	

The liquid phase was a mixture of distilled water and NaOH over a wide range of concentration.(0.1N,0.5Nand 1N). The NaOH was merc and with purity of about 98.5%.the physical property of the liquid phase and operation range are given in Table 2.

TABLE 2.	PHYSICAL PROPERTY OF THE LIQUID PHASE AND OPERATION
	RANGE

	ρL (kg/m3)	μL ((kg/(m.hr))	DL*106 (m2/hr)	Surface Tension (kg/hr2)		
NaOH=0.1 Kmole/m3	1002.25	2.945	7.768	71.33		
NaOH=0.5 Kmole/m3	1017.28	3.246	7.133	72.14		
NaOH=1 Kmole/m3	1041.32	3.552	6.498	72.644		
Operation Range	T=300C,P=1atm Gas Velocity 0 to 3(m/s),Liquid Rate 60 to 140 Lit/hr					

VI. EXPERIMENTAL RESULTS

A. Caustic Concentration

In two constant gas flow rates, 0.897 Kmol/hr and 1.496 Kmol/hr, and constant liquid flow rate for each experiment, caustic concentration has been varied between 0.1 and 1 Kmol/m3 and the effect on mass transfer area has been presented in Fig. 3.

As is seen, increase in caustic concentration, decreases mass transfer area. The process is assumed to be pseudo first



order irreversible chemical reaction, thus chemical reaction rate and finally mass transfer rate is expected to increase with increasing one of the reactants, but the results do not satisfy this prospect. The phenomenon can be explained using two-film theory. CO_2 and NaOH reaction takes place in liquid boundary layer. CO_2 has to diffuse through gas layer and enter liquid layer to reach NaOH solution. Increasing caustic concentration causes increased viscosity in liquid phase, thus CO_2 penetration in liquid phase is hardened and finally mass transfer area is decreased.



Fig. 3. Effective interfacial area vs. caustic normality at constant liquid rate for each curve and gas rate of (a) 0.897 and (b) 1.498 Kmol/hr

Diffusivity of dissolved Carbon dioxide in water and aqueous solutions was estimated using (5) [9].

$$D = 5.4 \times 10^{-6} \left(\frac{M^{0.5} L_s^{0.5}}{\mu_L . V^{0.5} . \Gamma^{0.3}} \right)^{0.93}$$
(5)

Referring (5), diffusivity is an inverse function of viscosity. In low gas and liquid flow rates, since there is sufficient residence time, increasing caustic concentration between low (0.1N) and moderate (0.5N), increases chemical reaction rate and causes more mass transfer area. On this situation, an increase in chemical reaction rate overcomes viscosity increase, but when caustic normality is varied to 1N, the drawback of viscosity increasing in the system and decrease of diffusion coefficients is so high that even increase of liquid rate at constant gas rate cannot considerably compensate the effect of viscosity.

B. Gas Rate

At constant caustic concentration, and constant liquid flow rate at each experiment, gas flow rate has been varied





Fig. 4. Effective interfacial area vs. Gas rate at constant liquid rate for each curve and caustic normality of (a)=0.1, (b)=0.5, (c)=1 Kmol/m3

Except in 1Kmol/m3 normality, at all liquid rates and caustic concentrations increase in gas rate results in more mass transfer area. Turbulency and packing movements due to higher gas flow rates cause more phase mixing and better fluidization and results in more gas-liquid interfacial area. Also it is evident that in higher liquid flow rates, because of higher L/G ratio and higher buoyancy force on packing, phase mixing takes place in better manner, therewith higher liquid flow rate results in faster surface renewal in gas-liquid boundary layer, the rapid reaction consumes much of CO_2 very close to the gas-liquid interface, which makes the gradient for CO_2 steeper and enhances the process of mass transfer in the liquid [12].

At 1Kmol/m3 normality, the solution's viscosity is high and chemical absorption is controlling. The viscosity drawback is so high that even increasing phase mixing through more gas rate and increasing L/G ratio can not retrieve mass transfer area decrease. As seen from Fig. 4c., at low liquid rate because of sufficient residence time, gas-liquid interfacial area increases with an increase in phase mixing through gas rate addition. But at higher liquid rates there is no sufficient residence time to conquest the viscosity effect on chemical absorption process.

C. Liquid Rate



Fig. 5. a_e vs. L, at constant G for each curve and caustic normality of (a)=0.1, (b)=0.5, (c)=1 \ Kmol/m3

In order to investigate liquid rate effect on gas-liquid interfacial area, at fixed caustic concentration and constant gas flow rate for each experiment, liquid flow rate has been varied in range of 1.75 to 2.6Kmol/hr and the results are presented in Fig. 5.

Results obtained at both low and moderate caustic concentrations and high caustic concentration at low gas flow rate; agree with the fact; increase in liquid flow rate in an

absorption column, increases gas-liquid interfacial area.

In other words: an increase in liquid flow rate, increases packing buoyancy and column fluidization. It also increases surface renewal at gas-liquid boundary layer and makes CO_2 gradient steeper between phases. At high caustic concentration and high gas flow rate, chemical reaction residence time is limited. Increase in solution viscosity impose a reverse effect on chemical reaction rate that even increase in liquid flow rate, L/G ratio and CO_2 gradient can not compensate.

VII. COMPARISON OF THE EXPERIMENTAL EFFECTIVE INTERFACIAL AREA OF A TCA WITH THAT OF A PACKED COLUMN

Here to show the superiority of a TCA to a packed column, the experimental results of this work, are compared to the data taken from PhD thesis of Dr. Bahram Hashemi Shahraki [1], who has experienced CO_2 absorption using caustic solution in a packed column in the UMSIT pilot plant (1992), for a solution in the normality range of 0.1 to 1. Table 3 and Fig. 5 compare the effective interfacial area values measured in a TCA (this work), with those measured in a packed column by Dr. Bahram Hashemi Shahraki. The operation L/G ratio, caustic normality, column characteristics and packing shape and geometries are also given in the table for these two cases.

TABLE 3. COMPARISON BETWEEN PACKED AND TCA COLUMN

Turbulent Contact Absorber		Packed Column			
NaOH	a(1/m)	L/G	NaOH	a(1/m)	L/G
0.1	1306.27	2.1	0.1	244.781	2.1
0.5	1157.38	2.7	0.5	254.205	2.7
1	665.538	2.9	1	132.293	2.9
Cross sectional area= 0.00875 m ²		Cross sectional area= 0.14657 m ²			
Packing Height= 0.2 m		Packing Height= 2.13m			
Packing size and type:			Packing size and type:		
dP= 15 mm, plastic ball		dP= 38 mm, plastic pall ring			





Fig. 6 Effective interfacial area vs. L/G ratio for TCA and Packed column

It is seen from the Table 3 and Fig. 6 that effective interfacial area in a TCA is approximately five times of the effective interfacial area in a packed bed column for the same L/G ratios and the same caustic concentrations.

VIII. RESULTS AND DISCUSSION

Sizeable parameters to assess TCA column efficiency are effective interfacial area (a) and mass transfer coefficients (kl, kg). Direct method for effective interfacial area calculation in TCA columns and column operating conditions has never been investigated. This work presents a direct effective interfacial area calculation method for TCA columns based on CO_2 absorption using caustic solution system. The process is chemical absorption which takes place in gas-liquid boundary layer. The process is liquid film controlling. Neglecting gas phase resistance and modifying liquid phase resistance for chemical absorption, results in a direct method for effective interfacial area calculations (4).

Assuming the process Isotherm and Isobar and neglecting heat production through the reaction, the sizeable parameters are limited to caustic concentration, gas flow rate and liquid flow rate. CO_2 diffusivity in caustic solution is reverse function of viscosity, reaction constant (kr) is function of caustic concentration, and volumetric mass transfer coefficient (Koga) is function of gas and liquid flow rates. All these parameters were considered in calculations and results from 70 practical experiments were compared.

Positive and negative effects of parameters are:

A. Caustic Concentration Effect

Basically increase in reactant amount in first order irreversible reactions, results in reaction rate increase. Regarding CO_2 absorption process, diffusivity nature of the system and limited residence times, cause evident reverse effect of viscosity increase on chemical absorption process. Therefore, except in low gas and liquid flow rates, it is necessary to use thin solution.

B. Gas Flow rate Effect

Increase in gas flow rate at studied system causes more turbulency, better phase mixing, and more mass transfer area. In the case of high solution concentration, increase in gas flow rate, causes less gas diffusion and less residence time; hence increase in gas flow rate at high solution concentration has negative effect on effective interfacial area.

C. Liquid Flow rate Effect

Increasing the liquid flow rate has two advantages for the system. Firstly it results in better fluidization and phase mixing. Secondly it causes more surface renewal and more new solution is ready for CO_2 to diffuse in.

IX. CONCLUSION

- Fluidized bed columns are best choice when L/G ratio is low.
- 2) Mass transfer rate is a severe function of solution concentration and effective interfacial area.
- 3) In order to increase mass transfer resulted from effective interfacial area, regarding superiority of TCA columns (especially at low L/G ratio) liquid flow rate must be increased to highest possible extent.
- 4) In order to increase mass transfer resulted from chemical absorption, solution concentration should be increased to extent that viscosity has no reverse effect on absorption process.
- 5) In the case of low L/G ratio, through selecting the best concentration for solution, and avoiding negative viscosity effects, using a smaller column than packed columns and less packing, 2 to 5 times of mass transfer in packed columns can be accessed using TCA column.
- 6) Regarding regeneration energy costs, operation at low L/G ratio and solution consumption reduction, utilizing TCA column as a new technology, not only decreases solvent purchase cost, but also optimizes regeneration cost.

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