

A MODIFIED CORRELATION TO EVALUATE LIQUID MASS TRANSFER COEFFICIENT IN AN IMPINGING-JET TALL BUBBLE COLUMN

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ABSTRACT

An impinging-jet bubble column has been installed in Al-Mansoor Co.-Baghdad for measurements of the ozone mass transfer applications in water treatment. Two injectors were used to produce turbulent gas-liquid jets in the working fluid by placing them at an intersecting angle of 120° . The impact of the two jets increased the gas-liquid mass transfer rates. Experiments are conducted at different ranges of Reynolds number for gas and liquid such that $12.8 < Re_G < 78$ and $804.24 < Re_L < 2928$. A correlation to predict volumetric liquid mass transfer coefficient is developed based on CSTRM applied on each section of the tall bubble column. Using the minimization technique, the following correlation was obtained:

$$k_L a = 21.09 u_G^{1.14} u_L^{0.078}$$

KEY WORDS

Mass transfer, impinging-jet bubble column, water treatment

NOMENCLATURE

A_C = column cross sectional area (m^2)

c_G = concentration of ozone in gas ($mg L^{-1}$)

c_L = concentration of dissolved ozone in liquid ($mg L^{-1}$)

C_{Gin} = percentage weight fraction of ozone in feed gas (%mg ozone/mg feed gas)

C_{Gout} =percentage weight fraction of ozone in off-gas (%mg ozone/mg off-gas)

C_L =concentration of dissolved ozone in liquid ($mg L^{-1}$)

C_L^* =equilibrium concentration of dissolved ozone in liquid ($mg L^{-1}$)

D = column diameter (m)

g =gravitational accelaration= $9.81 ms^{-2}$

h_{i-j} =axial distance between two pressure taps (m)

H =Henry law constant ($kpa L mg^{-1}$)

k_w =specific ozone utilization constant (s^{-1})

$k_L a$ =overall mass transfer coefficient (s^{-1})

l = axial distance along the column (m)

L = total length of the column (m)

M_{O_3} = molecular weight of ozone ($mg/mmol$)

n = number of a column section

p_{O_3} =partial pressure of ozone (kpa)

P = atmospheric pressure (kpa)

P_T = total hydrostatic head in the column (kpa)

Q_G = volumetric flow rate of feed gas ($L hr^{-1}$)

Q_L = volumetric flow rate of feed liquid ($L\ hr^{-1}$)

R = ideal gas constant ($kpa.m^3/mol.K$)

s = specific gravity

t = time (s)

T = liquid temperature (K)

u_G = gas superficial velocity ($m\ s^{-1}$)

u_L = liquid superficial velocity ($m\ s^{-1}$)

Z = dimensionless axial length (l/L)

Greek letters

α = fitting parameter of equation (10)

β = fitting parameter of equation (10)

ε = hold-up (-)

γ = fitting parameter of equation (10)

ρ = density

Subscripts and super subscripts

d = dispersed phase

G = gas

L = liquid

o = initial

INTRODUCTION

The volumetric mass transfer coefficient $k_L a$ in bubble columns normally evaluated based on one of the following assumptions for the flow pattern of the liquid phase:

- (a) complete mixing stirred-tank reactor model (CSTRM)
- (b) plug-flow model (PFM)
- (c) axial dispersion model (ADM)

Although estimations of $k_L a$ based on CSTRM give accurate results, the applicability of the CSTRM is only restricted to large diameter, say $D_C > 0.3\text{m}$ or small length to diameter ratio (between 1 and 3) ^[1,2].

Deckwer et al. ^[1,2,3] Chang ^[4], , Fan et al. ^[5], Schugerl et al. ^[6], Shioya and Dunn ^[7], Ying et al. ^[8], and Zhou et al. ^[9] have reported the mass transfer coefficient based on the axial dispersion model (ADM).

Voyer and Miller ^[10], and Alvarez-Cuenca et al. ^[11] have used the plug-flow model (PFM).

Wachsmann et al. ^[12], Huynh et al. ^[13], and Thalasso et al. ^[14] have reported that bubble columns using impinging-jet mixer give higher mass transfer rates than conventional bubble columns, this is due to the generation of smaller bubbles that had a larger specific interfacial area and also producing larger liquid-phase shear stresses.

In the present work a CSTR technique is applied on a tall bubble column to evaluate experimentally and numerically the

volumetric liquid phase mass transfer coefficient using impinging-jet mixer for an ozone/distilled water system without violating the CSTRM criteria settled by Shah et al.[1] and Deckwer et al.^[2].

EXPERIMENTAL METHOD AND DATA ANALYSIS

Pilot-Scale Experiments

The ozone mass transfer experiments were conducted in a pilot-scale impinging jet bubble column (see Figs.1,2). The bubble column was made from plexiglass and had inside diameter of 100 mm. and a total height of 2250 mm. A schematic of the configuration of the impinging-jet mixer is shown in Fig.3. Two injectors were placed 30 mm. above the bottom of the column at an intersecting angle of 120 deg. The distance between the centers of the nozzles was set equal to 60 mm. The bottom part of the bubble column was made of stainless steel to withstand the total weight of the column and the effects of jet backmixing.

The axial gas hold-up for each run is calculated from the axial pressure drop measurements along the column using ΔP transmitter type Rosemount calibrated to 0-250 mbar input signal and an equivalent output signal of 4-20 mA which is directed to a data acquisition system.

During ozonation tests, water samples were withdrawn from nine sampling taps that were inserted inside the reactor at a

distance equal to one third of the column diameter and at equal intervals along the column height. Outlet gas was analyzed for ozone concentration using on-line ozone monitor type.

Ozone concentration in the feed gas ranged from 3.125 to 6.25%w/w . A wide range of superficial liquid and feed-gas velocities was investigated to study their effects on the performance of the impinging-jet ozone bubble column. The operating conditions of the pilot-scale ozonation experiments are presented in Table-1. Ozone was supplied by an ozonizer at Al-Mansoor Co., where our pilot-scale apparatus was installed and operated.

DATA ANALYSIS FOR THE KINETIC STUDY

Ozonation is one of the treatment processes used in treating water and wastewater and it takes place in gas-liquid reactors. In water treatment applications, ozonation is limited by the mass transfer process as it is considered the controlling process that dictates the overall performance of ozone contactors [22] . A number of processes occur simultaneously during the ozonation treatment. The processes are: convection and backmixing processes of the liquid and gas phases flowing through the contacting chamber, ozone gas mass transfer process, ozone auto-decomposition process, and competitive reactive processes of dissolved ozone with the constituents present in water [9]. The overall ozone auto-decomposition rate is influenced

by the instantaneous dissolved ozone concentration and the amount of the utilized ozone during the ozonation process ^[15]. Ozone auto-decomposition process could be represented by a modified pseudo-first-order rate expression when ozonation is used to treat waters that do not exhibit significant initial ozone demand ^[16,17]. This modified pseudo-first-order rate expression can be written as follows:

$$\frac{dC_L}{dt} = -k_w C_L \dots\dots\dots(1)$$

Equation (1) was considered valid if the plotted data of $\ln[O_3]$ vs. t could be represented by a straight line. The slope of that line would represent specific ozone utilization rate constant (k_w), while the intercept would represent $\ln[O_3]$. Gamal El-Din and Smith ^[18] showed that the effect of T_L on k_w could be described by the well-known Van t Hoff-Arrhenius relationship:

$$\ln(k_w |_{T_L}) = \ln(k_w |_{20C}) + (T_L - 20) \ln \theta \dots\dots\dots(2)$$

Where θ is the dimensionless temperature correction factor, and T_L is the water temperature. Fig.5 is adapted from Gamal El-Din and Smith ^[18].

Correlations for Mass Transfer Coefficient

In conventional bubble columns, $k_L a$ was affected by u_G only^[1,2]. Deckwer^[4] proposed an empirical correlation to be as follows:

$$k_L a = \alpha (u_G)^\beta \quad (3)$$

where α and β are the empirical coefficients that are obtained through a nonlinear regression analysis.

The combined venturi-bubble columns studied by Huynh et al. [13] emerged as a promising new bubble column design. According to the study conducted by Huynh et al.^[13], $k_L a$ was found to be dependent on u_G and u_L as in the following relationship:

$$k_L a = \alpha (u_G)^\beta (u_L)^\gamma \quad (4)$$

where α , β , and γ are the empirical coefficients that are obtained through a nonlinear regression analysis.

Estimation of Liquid Mass Transfer Coefficient:

The column was divided hypothetically into ten sections, each section has a height equal to twice the column diameter. The following principles assumptions are stated before starting the method of estimation:

- a- Plug flow is assumed for gas phase, caused by relatively large buoyancy of the gas bubbles ^[19,20,21].
- b- At each section, an intense mixing condition (i.e., complete mixing) existed in the liquid phase caused by small reactor aspect ratio ($L/D=2$), ^[2,21].
- c- The gas density depends linearly on the column height, Owing to the hydrostatic head.
- d- Interfacial area and gas holdup are constant along the height of the column.
- e- Henry's law applies.
- f- Ozone decay rate is pseudo-first-order in the liquid phase, whereas it is negligible in the gas phase ^[15,16].
- g- Samples taken from taps during each run do not affect the volumetric flow rate of liquid through the column.
- h- The experiments are performed under isothermal conditions.

Mathematical Equations Used

- Due to ozone absorption in the liquid phase along the column, the superficial gas velocity is variable according to the following equation:

$$u_G = \frac{(y_{N_2} + y_{O_3})Q_G}{A_C} \dots\dots\dots(5)$$

- Gamal El-Din and Smith ^[22] assumed an exponential function to describe the variation of gaseous ozone molar fraction along the column height.

$$y_{O_3} = (y^0_{O_3}) e^{-\Omega Z} \dots\dots\dots(6)$$

Where Ω is the dimensionless coefficient of the gas-phase ozone molar fraction decrease along the column height, and Ω was estimated using equation (6) in which y_{O_3} at $Z=0$ and $Z=1$ were obtained experimentally through the monitoring of the ozone concentrations in the feed and off-gas lines. The mathematical representation for liquid phase mass transfer coefficient calculation is shown in Fig.4 .

- Ozone overall mass balance around the column:

$$Q_G c_{Gin} + Q_L c_{Lin} = Q_G c_{Gout} + Q_L c_{Lout} \dots\dots\dots(7)$$

- Ozone mass balance around section (1):

$$Q_G c_{Gin} + Q_L c_{Lin} = c_{G1} A_C u_G \epsilon_G + Q_L c_{L1} \dots\dots\dots(8)$$

- Ozone mass balance around section (n):

$$c_{G_{n-1}}A_C u_{G_{n-1}} \varepsilon_G + Q_L c_{L_{n-1}} = c_{G_n}A_C u_{G_n} \varepsilon_G + Q_L c_{L_n} \dots \dots \dots (9)$$

It is of importance to remember that each section is considered as a CSTR, and this leads to the fact that c_{L_n} is the same at any point inside section n, also all the physical properties are the same at any point in section n.

Equations (1) to (9) are used to solve for c_{G_n} , u_{G_n} at each run..

Rewriting equation (4) for each section:

$$(k_L a)_n = \alpha(u_{G_n})^\beta (u_L)^\gamma \dots \dots \dots (10)$$

where $(k_L a)_n$ is the volumetric mass transfer coefficient for the liquid phase in section n, where $n=1,2,\dots,10$.

- The equation used to estimate axial and average hold-up values from pressure differential measurements is developed as follows:

The gas hold-up in a gas-liquid system can be expressed in terms of the liquid density, and the density of gas-liquid dispersion ρ_d (i.e., density of the expanded liquid) as,

$$\varepsilon_G = \frac{\rho_L - \rho_d}{\rho_L - \rho_G} = 1 - \frac{\rho_d}{\rho_L} \dots \dots \dots (11)$$

(65-77) The density of the expanded liquid between any two pressure taps, i and j can be calculated from the measured pressure drop $(\Delta p)_{i-j}$ and the known distance between the pressure taps, h_{i-j} ,

$(s_d)_{i-j} = \frac{(\Delta p)_{i-j}}{h_{i-j}}$ and $(\rho_d)_{i-j} = (s_d)_{i-j} \rho_{H_2O}$(12)
 Where $(s_d)_{i-j}$ is the specific gravity of the dispersion between pressure taps i and j, by substituting this expression into equation(11), to obtain:

$$(\varepsilon)_{i-j} = 1 - \frac{(\Delta p)_{i-j}}{s_L h_{i-j}} \dots\dots\dots(13)$$

The major source of error in calculating the average gas hold-up within a segment h_{i-j} , is in the measurements of $(\Delta p)_{i-j}$ and s_L . The pressure drop is a rapidly fluctuating quantity, particularly at higher gas flow rates due to the passage of slugs. In calculations, the arithmetic average of the maximum and the minimum observed values was employed.

- The liquid phase mass transfer coefficient can be represented by [23].

$$N_{O_3} = k_L a (C_L^* - C_L) \dots\dots\dots(14)$$

where N_{O_3} is in milligram of solute absorbed per unit time per unit volume of the section under consideration. Also, we have:

$$p_{O_3} = P_T y_{O_3} = HC_L^* \dots\dots\dots(15)$$

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(66-77)

The hydrostatic head P_T in the column decreases linearly with the

$$C_L^* = \frac{P_T y_{O_3}}{H} \dots\dots\dots(16)$$

depth as long as ϵ_L is constant, and hence:

$$P_T = P_t + \rho g \epsilon_L (L - l) \dots\dots\dots(17)$$

Substitute (16) into (14) to obtain:

$$N_{O_3} = k_L a \left(\frac{P_T y_{O_3}}{H} - C_L \right) \dots\dots\dots(18)$$

$$k_L a = \frac{N_{O_3}}{\left(\frac{P_T y_{O_3}}{H} - C_L \right)} \dots\dots\dots(19)$$

From the definition of N_{O_3} , it can be expressed by:

$$(N_{O_3})_n = \frac{(y_{O_3}^{n-1} - y_{O_3}^n) Q_G P_T M_{O_3}}{V_n RT} \dots\dots\dots(20)$$

Where :

(67-77)

$(N_{O_3})_n$: milligram of ozone absorbed per unit time per unit volume of section n.

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V_n : volume of section n in cubic meter.

$(y_{O_3}^{n-1} - y_{O_3}^n)$: mole fraction of ozone in gas at inlet and outlet of section n respectively.

Substitute equation (20) into (19) to obtain:

$$(k_L a)_n = \frac{(y_{O_3}^{n-1} - y_{O_3}^n) Q_G P_T M_{O_3}}{V_n \left(\frac{P_T y_{O_3}^n}{H} - C_L \right) RT} \dots\dots\dots(21)$$

$k_L a$ can be estimated from experimental data using equation (21).

- Following the minimization technique used by Zhu et al.^[25], the parameters of the model in equation (10) can be evaluated using the following criteria:

$$\Phi = \sum_{i=1}^n ((k_L a)_{EXP.} - (k_L a)_{MOD.})^2 \dots\dots\dots(22)$$

where a set of values for α , β , and γ which give a minimum for the function Φ was searched.

RESULTS AND DISCUSSION

68 The operating conditions used in the current study and the results of the minimization techniques for the pilot-scale ozone mass experiments are shown in Tables 1. Henry law constant (H) of 0.187 kpa L mg⁻¹ was obtained from Roth and Sullivan [24]. (68-77) Each ozone concentration was based on a number of two replicates. The absolute error between each pair of dissolved ozone concentration was < 4.6% with a mean value of 1.7%. The convergence criteria was satisfied when the minimization function (Φ) was minimized with a relative error of < 10⁻⁴.

The liquid temperatures were measured at the bottom and at the top of the bubble column and were found to be approximately the same with negligible difference of 0.4-0.7 °C.

Volumetric flow rate of gas and liquid were arranged such that, experiments were carried out at different ranges of Reynolds number for gas and liquid respectively, from Table.1 it is seen that reynold number are in the ranges of $12.8 < Re_G < 78$ and $804.24 < Re_L < 2928$.

For investigating the effect of u_G on the dissolved ozone concentration profiles and using the minimization technique, a comparison was done between Rno. 15 and 4 that were conducted at the same u_L . As u_G increased from 2.5E-03 ms⁻¹ to

7.5E-03 ms⁻¹, the estimated k_La increased from 1.63E-02 s⁻¹ to 5.77E-02 s⁻¹. Similar phenomena were observed when comparing the results of Rno. 6 and 3.

(69-77) For investigating the effect of u_L on the dissolved ozone concentration profiles and using the minimization technique, a comparison was done between Rno. 15 and 13 that were conducted at the same u_G. As u_L increased from 7.7E-03 ms⁻¹ to 1.4E-02 ms⁻¹, the estimated k_La increased from 1.63E-02 s⁻¹ to 1.79E-02 s⁻¹. Similar phenomena were observed when comparing the results of Rno. 10 and 14.

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The k_La was found to be dependent on u_G and u_L. Using the minimization technique, the nonlinear regression parameters (α, β, and γ) were estimated to be 21.90, 1.14, and 0.078, respectively with a correlation r=0.975.

As u_L increased, k_La increased. The same observation was reported by Huynh et al.^[13]. In the impinging-jet bubble column, by increasing u_L, the turbulence in the liquid phase increases and this will cause high shear stresses leading the large gas bubbles to break into smaller bubbles. As a result, the gas bubble specific interfacial area (a) will increase leading to an increase in the overall mass transfer coefficient (k_La). Also as u_G increased, k_La increased considerably as observed before by several researchers [4],[20],[13], and [15]. This could be explained by the following : as u_G increases, the gas bubbles mean specific interfacial area, a, increases due to the increase in Sauter diameter(d_s).

Fig.(6) describes a comparison between the k_{La} correlation developed in the present work using the minimization technique, with those developed in previous studies. As shown in Fig.6, the impinging-jet bubble column has proven to cause a significant increase in the overall ozone mass transfer coefficient (k_{La}) compared to the conventional bubble column design.

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CONCLUSIONS

(70-77)

With regard to the measurements of the ozone transfer rates it can be concluded:

- (1) For the tested distilled water, it was found that k_{La} was dependent on the liquid and gas superficial velocities.
- (2) Placing the injectors at an intersecting angle of 120° at the bottom of the bubble column caused impinging of the gas-liquid jets. This arrangement has led to a significant increase in the turbulence intensities in the ambient fluid and consequently, a significant increase was observed in the mass transfer rates compared to conventional bubble column designs.
- (3) The isothermal condition already assumed in section 3.1 for the experiments is verified by measuring the temperature difference of the liquid at the bottom and at the top of the column indicating that heat of the physical absorption of ozone in distilled water can be neglected.

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Table.1 Operating conditions of the ozone mass transfer experiments In the pilot-scale apparatus installed in AL-Mansoor Co.

RUN NO.	Q_G (LH^{-1})	Q_L (LH^{-1})	$C_{G IN}$ (%W/W)	$C_{G OUT}$ (%W/W)	K_{LA} (S^{-1})$\times 10^2$
1	107.5	212	3.1	1.79	2.60
2	310.8	650	3.11	1.77	9.57
3	206.4	395.6	3.12	1.82	5.75
4	212	217.6	3.125	2.2	5.77
5	73.5	223	3.125	1.5	1.69
6	67.8	395.6	3.15	1.2	1.62
7	82	619	3.15	0.925	2.08
8	101.7	395.6	3.16	1.425	2.57
9	180.8	619	3.16	1.433	5.15
10	50.8	619	3.17	0.683	1.20
11	282.6	790	3.17	1.683	8.69
12	107.5	217.6	6.14	3.4	2.61

13	70.6	395.6	6.2	2.25	1.79
14	50.8	678	6.22	1.25	1.25
15	70.6	217.6	6.25	2.925	1.63
16	248.6	619	1.84	1.0	7.37



Fig 1. View of experimental setup layout.

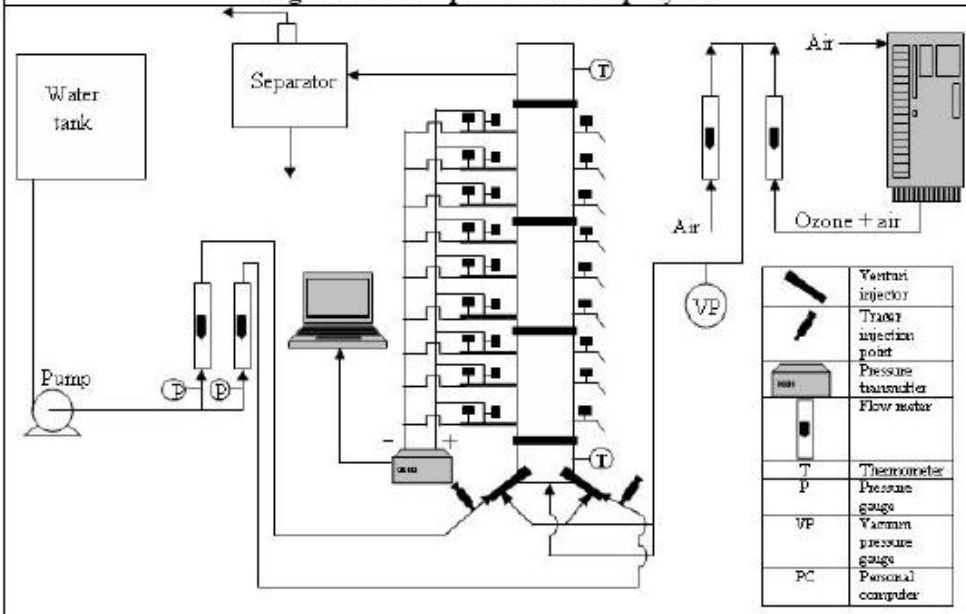


Fig 2. Experimental setup for measurement of the liquid phase transfer coefficients ($K_L a$)

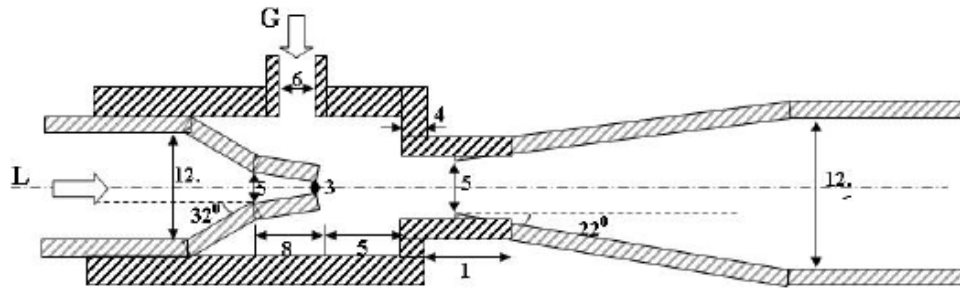


Fig 3. Design of the impinging jet mixer

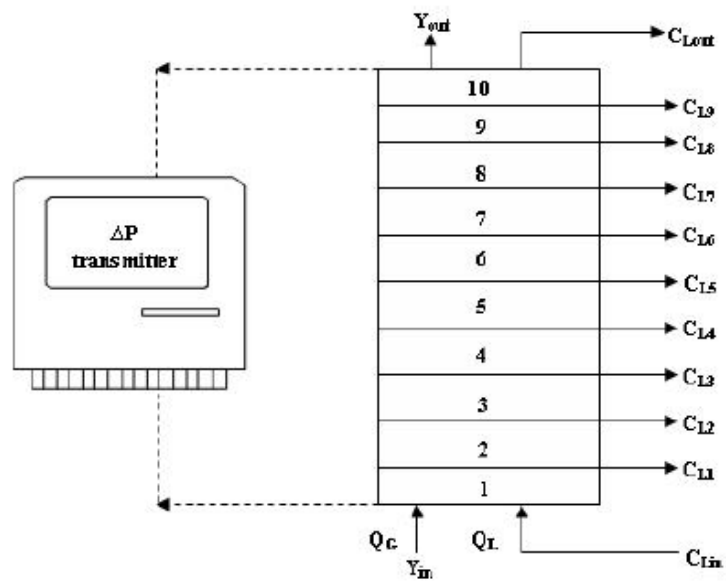


Fig 4. Mathematical representation of the bubble column

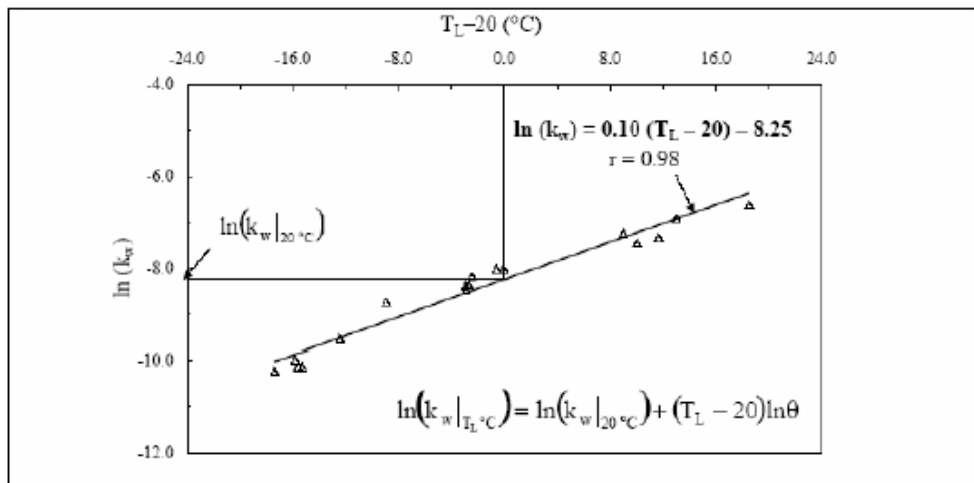


Fig 5. The effect of water temperature on the ozone auto-decomposition rate constant

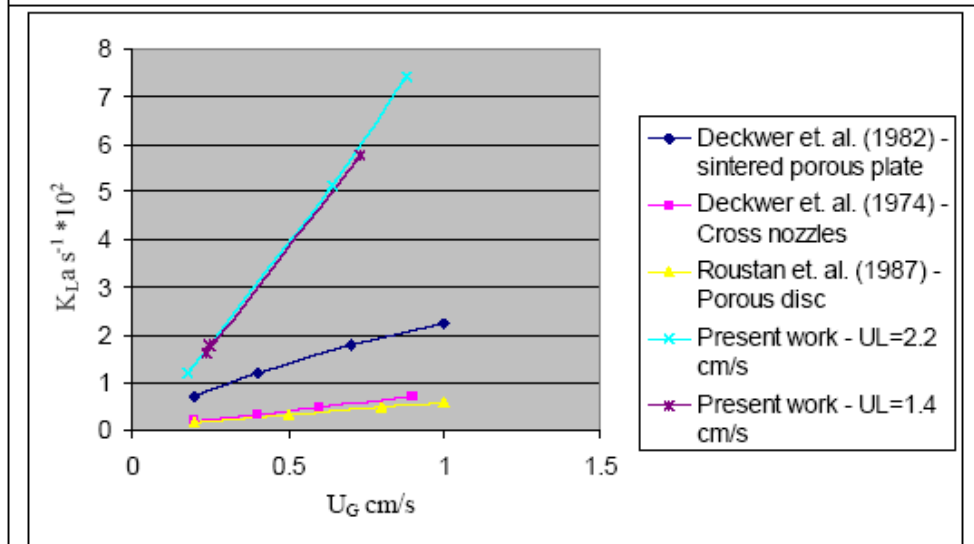


Fig 6. A comparison between the $K_L a$ in various types of bubble columns.

تحسين تصحيح لحساب معامل انتقال المادة في الأعمدة الفقاعية

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مدرس

قسم الهندسة الكيمياءوية - الجامعة التكنولوجية

الخلاصة

عند شركة المنصور في بغداد تم تنصيب منظومة أساسها عمود فقاعي يعمل بالتيارات التصادمية لأغراض دراسة معدلات انتقال الكتلة لغاز الأوزون في الماء المقطر و هذه الدراسة يمكن استخدامها لتحسين تصاميم الأعمدة الفقاعية المستعملة في أنظمة معالجة المياه 0 عند قاعدة العمود الفقاعي تم تنصيب قاذفتين لتيارات السائل-غاز بزواوية تقاطع مقدارها 120 درجة وذلك لإحداث تيارات اضطرابية في العمود 0 و قد أدت هذه التيارات إلى زيادة معدلات انتقال الكتلة مقارنة بالأعمدة التقليدية المستخدمة في دراسات وبحوث سابقة 0 كما تم استنتاج علاقة رياضية تبين اعتما دية معامل انتقال الكتلة في السائل على سرعة كل من الغاز و السائل

$$k_La=21.09u_G^{1.14}u_L^{0.078}$$

إن الأعمدة الفقاعية و التي تستخدم التيارات التصادمية يمكن استخدامها بكفاءة أعلى من الأعمدة الفقاعية التقليدية 0

الكلمات الدالة

انتقال الكتلة- الأعمدة الفقاعية ذات التيارات التصادمية- معالجة المياه