

Volumetric Mass Transfer Coefficient of SO₂ Gas Absorption into Aqueous Sodium Sulphite Solution in Plate Column

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Abstract

The volumetric mass transfer coefficient ($K_{G,a}$) for SO₂ removal from gas mixture into aqueous Na₂SO₃ solution was studied in a plate column at constant temperature (± 25 °C), and liquid holdup. The $K_{G,a}$ values were evaluated over ranges of operating independent variables: gas flow rate (Q_G), SO₂ concentration in inlet gas ($C_{SO_2, in}$), and concentration of aqueous Na₂SO₃ solution ($C_{Na_2SO_3}$). The experimental results showed that $K_{G,a}$ decreased with increasing of $C_{SO_2, in}$, increased with increase of Q_G , and $C_{Na_2SO_3}$. The influence of gas flow rate on $K_{G,a}$ is more than the influence of SO₂ concentration in inlet gas, and concentration of aqueous Na₂SO₃ solution respectively. Computer program Statgraphics/Experimental design was used to find the linear fitted models of the $K_{G,a}$ in terms of the dimensional and dimensionless of independent operating variables. The Q_G , $C_{SO_2, in}$, and $C_{Na_2SO_3}$ have significant effects on $K_{G,a}$, while the interactions of them have no significant effects on it, and could be neglected. The R-squared statistic indicates that the model as fitted explains 90.4949% of the variability in $K_{G,a}$.

Keywords: Plate column, SO₂ absorption, Na₂SO₃ solution, $K_{G,a}$

Nomenclature:

$C_{SO_2, in}$ = SO₂ concentration in inlet gas v/v %

$C_{SO_2, out}$ = SO₂ concentration in outlet gas v/v %

$C_{Na_2SO_3}$ = concentration of Na₂SO₃ aqueous solution mol/l

$K_{G,a}$ = volumetric mass transfer coefficient s⁻¹

Q_G = gas flow rate m³/s

V_L = liquid holdup in plate column m³

Subscripts:

G = gas mixture

L = liquid

Na₂SO₃ = aqueous sodium sulphite solution

SO_{2, in} = sulphur dioxide inlet gas

SO_{2, out} = sulphur dioxide outlet gas

1. Introduction

Flue gas desulfurization (FGD) is presently receiving much attention in many countries. Removal of sulphur dioxide gas from flue gas is very important in the controlling of atmospheric pollution. The wide spread processes used for this purpose are the wet process absorption where the flue gas absorbed in lime slurries, the additives sometimes are used with slurries to increase the absorption rate, and dry process absorption when limestone and lime injected into the hot flue gas. These processes, besides being non regenerative, create the problem of disposal of large quantities of waste (Dutta, Basu, Pandit, & Ray, 1987; Duric, Omerovic, Brankov, Dzaferovic, & Stanojevic, 2011). Several regenerable processes of sulphur dioxide removal involving absorption into aqueous sodium citrate solution (Erga, 1980; Skribic, Cvejanov, & Paunovic, 1991) which have been

received most attention in recent times due to regenerative viability of absorbent solution (Bravo, Camacho, Moya, & Aguado, 1993). Process developed by several investigators and has been used for various scales and even in commercial scale. The leading SO₂ recovery process is presently the Wellman-Lord (WL) process shall here be used in this study (Erga, 1988). In the WL process SO₂ is absorbed in an aqueous sodium sulphite solution producing sodium bisulphate:



The SO₂ is a very toxic gas, has hard effects on the health and environment. The current OSHA standard for SO₂ is (5 ppm.) of air average over an eight-hour work shift. Several investigators studied absorption into different absorbents such as: aqueous reactive slurries of calcium and magnesium hydroxide (Sada, Kumazawa, Sawada, & Hashi-Zume, 1981; Dagaonkar, Beenackers, & Pangarkar, 2001), aqueous sodium hydroxide and sodium sulphite solution (Hikita, Asal, & Tsuji, 1977), aqueous solution of sodium carbonate (Witte & Kind, 1986), Urea solution (Barbooti, Ibraheem, & Ankosh, 2011), and aqueous sodium citrate solution (Skribic, Cvejaanov, & Paunovic, 1993).

The aim of present work to estimate the volumetric mass transfer coefficient ($K_{G,a}$) of SO₂ gas into aqueous sodium sulphite solution using the following equation:

$$K_{G,a} = \frac{Q_G}{V_L} \ln \frac{C_{\text{SO}_2, \text{in}}}{C_{\text{SO}_2, \text{out}}} \quad (2)$$

2. Experimental

Experiments were performed on absorption of SO₂ from SO₂/N₂ gas mixture into aqueous sodium sulphite solution, at constant liquid holdup, and temperature (± 25 °C) in plate column, according to experimental plan as shown in Table 1.

Table 1. Experimental plan for SO₂ gas absorption into aqueous Na₂SO₃ solution in plate column

Experiment No.	Gas flow Rate (Q_G) (m ³ /s)	concentration of aqueous Na ₂ SO ₃ solution ($C_{\text{Na}_2\text{SO}_3}$) (mol/l)	SO ₂ concentration in inlet gas mixture ($C_{\text{SO}_2, \text{in}}$) (v/v %)
1	0.0025	0.5	0.15
2	0.0025	1.5	0.15
3	0.0015	1.5	0.15
4	0.0015	0.5	0.15
5	0.0015	1.5	0.55
6	0.0015	0.5	0.55
7	0.0020	1.0	0.35
8	0.0025	1.5	0.55
9	0.0020	1.0	0.35
10	0.0025	0.5	0.55
11	0.0020	1.0	0.35

The equipments used in present work are shown in Figure 1. The main equipment is plate column (1), and complementary equipments are as follows:

N₂-gas cylinder (2), SO₂-gas cylinder (3), Mixing chamber (4), SO₂-gas rotameter (5), N₂-gas rotameter (6), condenser (7), SO₂-gas analyzer (8), Evaporator (9), mixing tank for preparing Na₂SO₃ solution (10), and Na₂SO₃ solution feed pump (11).

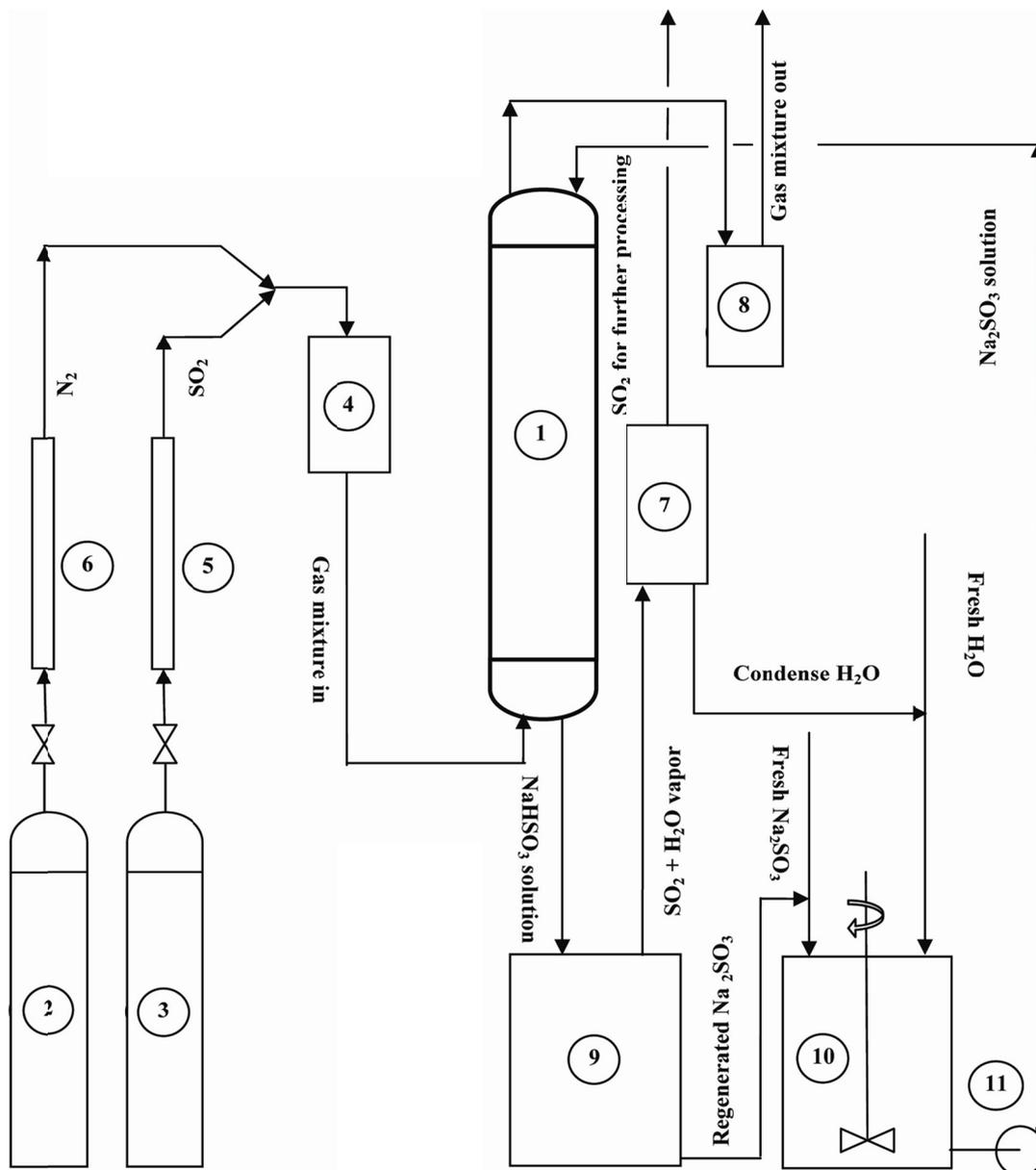


Figure 1. Experimental apparatus of SO₂ absorption into Na₂SO₃ aqueous solution in plate column

During the normal operation, the main reaction takes place in plate column (1) of 150 mm inside diameter, and 1000 mm height. The gas mixture enter the column from lower part while, the aqueous sodium sulphite solution enter the upper part of the column. The sulphur dioxide from gas mixture absorbed into aqueous sodium sulphite solution, producing sodium bisulphate solution, which discharge from downer part of the plate column into evaporator (9). The loaded aqueous solution is thermally regenerated by evaporation of H₂O + SO₂, thereby reversing reaction (1). After condensing the water vapor, nearly pure gaseous SO₂ is left as an overhead product, ready for further processing. Solid Na₂SO₃ which precipitates in the evaporator is redissolved in condensate/water in the mixing tank (10), to proper concentration for a new absorption-regeneration cycle.

3. Results and Discussion

The estimated values of volumetric mass transfer coefficients ($K_{G,a}$) and the experimental design plan are shown in Table 2.

Table 2. Data base for experimental design and results of $K_{G,a}$ for SO_2 gas absorption into aqueous Na_2SO_3 solution in plate column

Experiment No.	Gas flow Rate (Q_G) (m^3/s)	Concentration of aqueous Na_2SO_3 solution ($C_{Na_2SO_3}$) (mol/l)	SO_2 concentration in inlet gas mixture ($C_{SO_2, in}$) (v/v %)	Volumetric mass transfer coefficient ($K_{G,a}$) (s^{-1})
1	+ 0.0025	- 0.5	- 0.15	2.715
2	+ 0.0025	+ 1.5	- 0.15	3.581
3	- 0.0015	- 1.5	- 0.15	2.149
4	- 0.0015	- 0.5	- 0.15	2.062
5	- 0.0015	+ 1.5	+ 0.55	1.963
6	- 0.0015	- 0.5	+ 0.55	1.491
7	0 0.0020	0 1.0	0 0.35	2.424
8	+ 0.0025	+ 1.5	+ 0.55	2.609
9	0 0.0020	0 1.0	0 0.35	2.273
10	+ 0.0025	- 0.5	+ 0.55	2.029
11	0 0.0020	0 1.0	0 0.35	2.323

By using computer program (Statgraphics/experimental design) to find the linear model of $K_{G,a}$ in terms of independent variables: gas flow rate (Q_G), SO_2 concentration in inlet gas mixture ($C_{SO_2, in}$), and concentration of aqueous Na_2SO_3 solution ($C_{Na_2SO_3}$). The fitted model with dimensional independent variables as follows:

$$K_{G,a} = 0.722 + 817.250 Q_G - 1.509 C_{SO_2, in} + 0.501 C_{Na_2SO_3} \quad (3)$$

The fitted model with dimensionless independent variables is presented in the following form:

$$K_{G,a} = 2.329 + 0.409 Q_G - 0.302 C_{SO_2, in} + 0.251 C_{Na_2SO_3} \quad (4)$$

The validity of the linear models is as follows:

Model in Equation (3)	Model in Equation (4)
$0.0015 \leq Q_G \leq 0.0025 \quad m^3/s$	$-1 \leq Q_G \leq +1$
$0.15 \leq C_{SO_2, in} \leq 0.55 \quad v/v \%$	$-1 \leq C_{SO_2, in} \leq +1$
$0.5 \leq C_{Na_2SO_3} \leq 1.5 \quad mol/l$	$-1 \leq C_{Na_2SO_3} \leq +1$

The independent variables: gas flow rate, SO_2 concentration in inlet gas mixture, and concentration of aqueous Na_2SO_3 solution have significant effects on volumetric mass transfer coefficient ($K_{G,a}$), while the interaction of independent variables have no significant effects on $K_{G,a}$, and could be neglected. The influence of gas flow rate on $K_{G,a}$ is higher than that of SO_2 concentration in inlet gas mixture, and concentration of aqueous Na_2SO_3 solution as shown in following Figure 2:

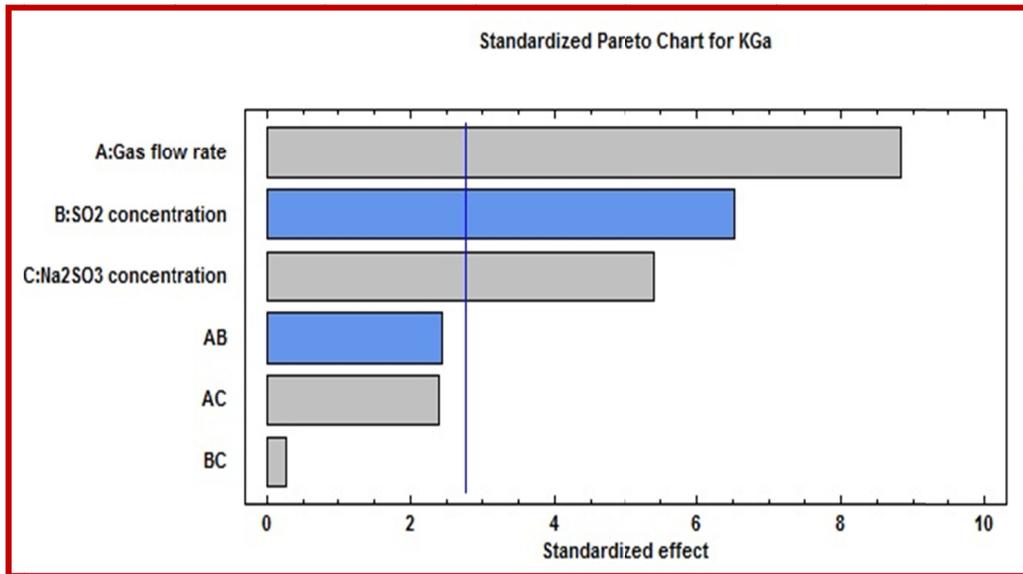


Figure 2. The standard effect of the independent variables on volumetric mass transfer coefficient ($K_{G,a}$)

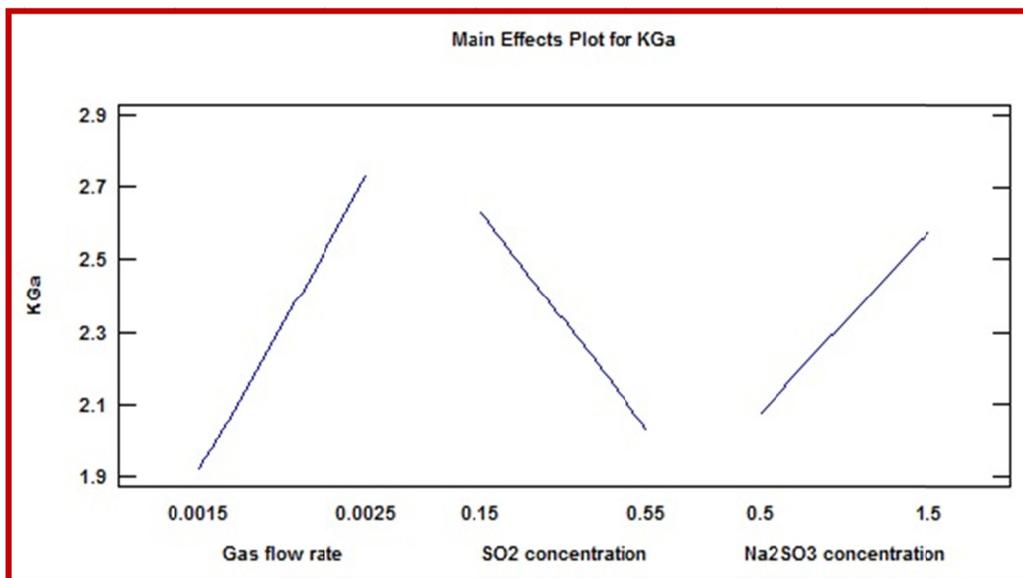


Figure 3. The main effects of independent variables on $K_{G,a}$ for dimensional model in Equation (3)

The main effect of independent variables: gas flow rate, SO₂ concentration in inlet gas mixture, and concentration of aqueous Na₂SO₃ solution on $K_{G,a}$ could be seen in Figure 3 and Figure 4, for dimensional and dimensionless models respectively. The $K_{G,a}$ decreased by increasing in SO₂ concentration in inlet gas mixture, increased with increasing gas flow rate, and concentration of aqueous Na₂SO₃ solution.

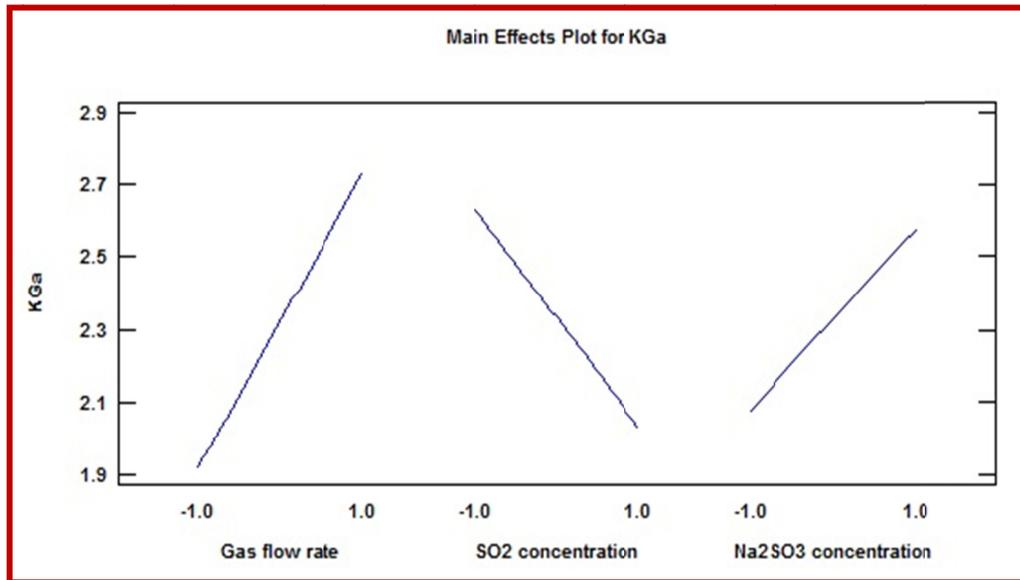


Figure 4. The main effects of independent variables on $K_{G,a}$ for dimensionless model in equation (4)

The observed and predicted $K_{G,a}$ from dimensional and dimensionless models in Equations (3) and (4) are shown in Figure 5. The R-Squared statistic indicates that the models as fitted explains 90.4949% of the variability in $K_{G,a}$. The adjusted R-squared statistic, which is more suitable for comparing models with different numbers of independent variables, is 86.4212%. The standard error of the estimate shows the standard deviation of the residuals to be 0.19627.

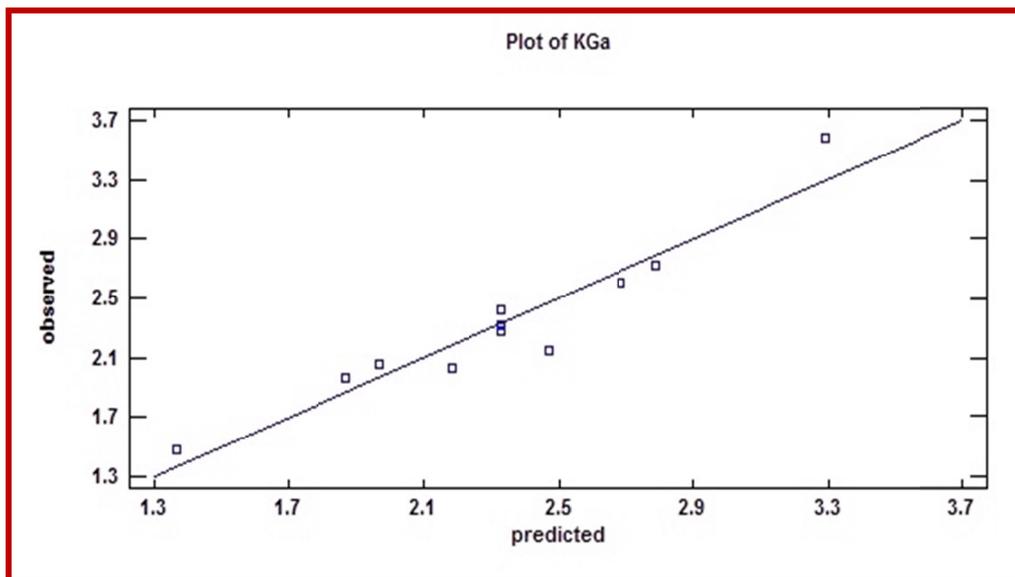


Figure 5. The observed and predicted $K_{G,a}$ from dimensional and dimensionless models in Equations (3) and (4)

Figure 6. Represents the 3 D surface plot of volumetric mass transfer coefficient ($K_{G,a}$) of SO_2 absorption into aqueous Na_2SO_3 solution, Gas flow rate (Q_G), and concentration of aqueous Na_2SO_3 solution ($C_{\text{Na}_2\text{SO}_3}$). $K_{G,a}$ increases with increasing in Q_G , and $C_{\text{Na}_2\text{SO}_3}$.

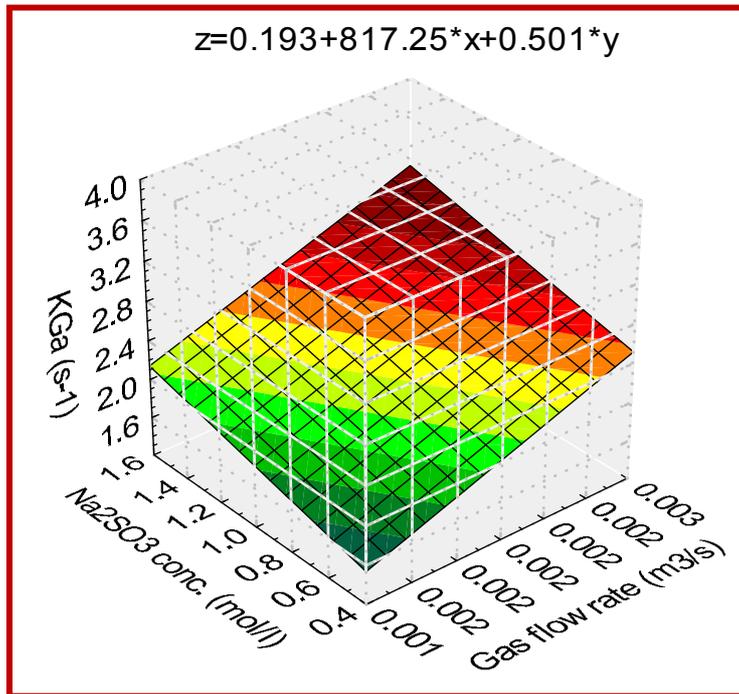


Figure 6. Variation of $K_{G,a}$ for SO_2 absorption with gas flow rate (Q_G), and concentration of aqueous Na_2SO_3 solution ($C_{\text{Na}_2\text{SO}_3}$)

The relationship between $K_{G,a}$ for SO_2 gas absorption into aqueous Na_2SO_3 solution and independent variables: gas flow rate (Q_G), and SO_2 concentration in inlet gas mixture ($C_{\text{SO}_2, \text{in}}$) could be seen at Figure 7. The $K_{G,a}$ increases with increasing in Q_G , and decreases with increasing of $C_{\text{SO}_2, \text{in}}$.

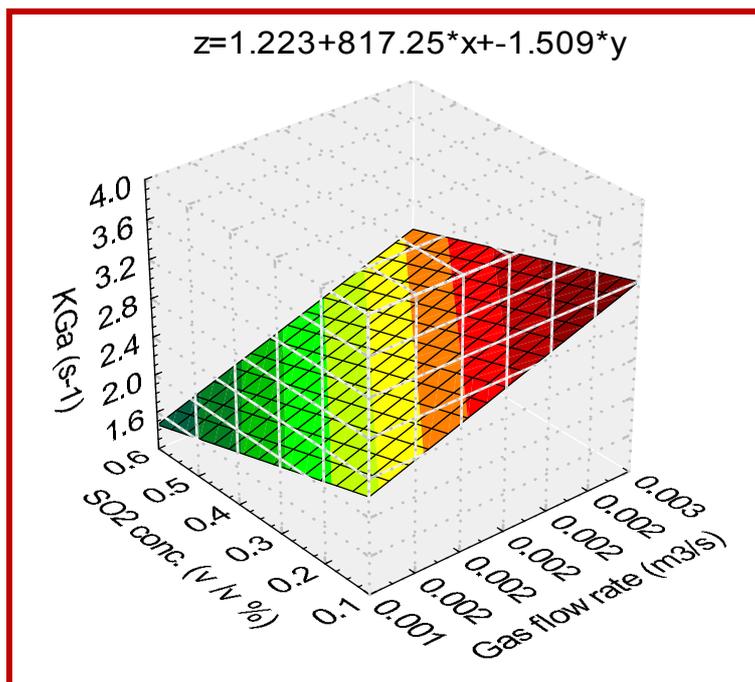


Figure 7. Variation of $K_{G,a}$ with SO_2 concentration in inlet gas mixture ($C_{\text{SO}_2, \text{in}}$) and gas flow rate (Q_G)

The volumetric mass transfer coefficient ($K_{G,a}$) increases with increasing of concentration of aqueous Na_2SO_3 solution ($C_{\text{Na}_2\text{SO}_3}$), and decreasing with increasing the SO_2 concentration in gas mixture ($C_{\text{SO}_2, \text{in}}$) as shown in the following Figure 8.

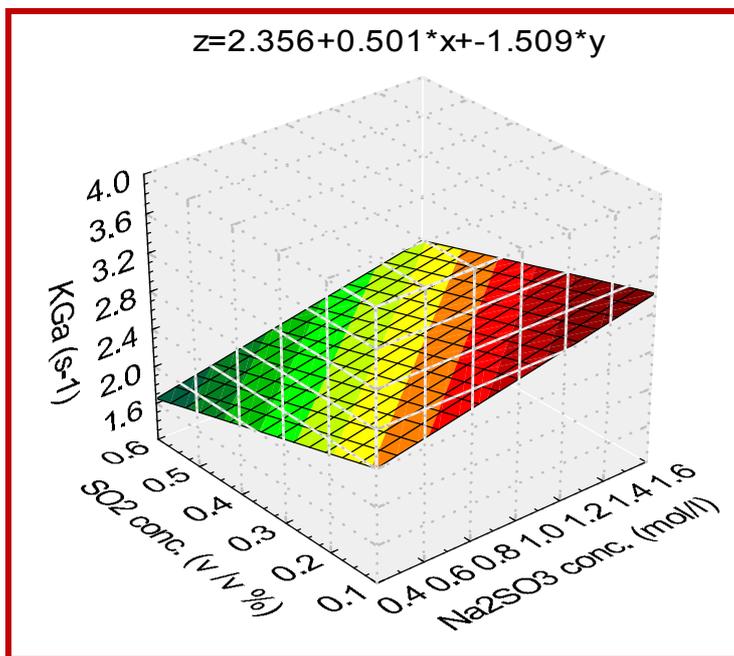


Figure 8. Variation of $K_{G,a}$ of SO_2 absorption with $C_{\text{Na}_2\text{SO}_3}$ and SO_2 concentration of inlet gas mixture ($C_{\text{SO}_2, \text{in}}$)

The $K_{G,a}$ for SO_2 gas removal from flue gas into aqueous Na_2SO_3 solution was investigated in packed column with various operating conditions of independent variables (Wang, Yang, & Zhang, 2010). The $K_{G,a}$ values for SO_2 in flue gas of coal fired power plant were measured and calculated by aqueous ammonia in packed column as well (Qiu, Zhang, Guo, Li, Zheng, & Gong, 2010). The measured results of both investigators showed that the independent variables: gas flow rate, SO_2 concentration in flue gas, and absorbents concentrations have significant effects on $K_{G,a}$ of SO_2 absorption. These results are practically similar to the present results showing significant effects of independent variables on $K_{G,a}$ of SO_2 gas removal by different of aqueous solutions.

4. Conclusions

- Absorption of SO_2 gas from gas mixture (SO_2/N_2) into aqueous Na_2SO_3 solution were performed using different operating conditions of independent variables: gas flow rate, SO_2 concentration in inlet gas mixture, and concentration of aqueous Na_2SO_3 solution.
- The $K_{G,a}$ of SO_2 gas absorption into aqueous Na_2SO_3 solution was calculated. The results showed that $K_{G,a}$ increases with increasing gas flow rate and concentration of aqueous Na_2SO_3 solution, and by decreases with increasing in the SO_2 concentration in inlet gas mixture.
- Using computer program (Statgraphics/Experimental design) to find the fitted linear models for dimensional and dimensionless of independent variables: Q_G , $C_{\text{Na}_2\text{SO}_3}$, and $C_{\text{SO}_2, \text{in}}$. The two fitted models for $K_{G,a}$ reveal that Q_G , $C_{\text{Na}_2\text{SO}_3}$, and $C_{\text{SO}_2, \text{in}}$ have significant effects on $K_{G,a}$, while the interaction of them have no significant effects on it, and could be ignored as shown in Figure 2.
- The R-squared statistic indicates that the models as fitted explains 90.4949% of the variability in $K_{G,a}$. The adjusted R-squared statistic, which is more suitable for comparing models with different numbers of independent variables, is 86.4212%. The standard error of the estimate shows the standard deviation of the residuals to be 0.19627.
- The $K_{G,a}$ of SO_2 absorption could be improved by increasing of gas flow rate, and concentration of aqueous Na_2SO_3 solution, and by decreasing the SO_2 concentration in inlet gas mixture.
- Pilot plant designed based on the results of present work, and erected at sulphuric acid production factory in Baghdad to reduce the emission of SO_2 to atmosphere for preventing the air pollution.

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