Removal of SO₂ in Dry Fluidized and Fixed Bed Reactors using Granular Activated Carbon

Dr. Neran K. Ibrahim^{*} & Omar A. Jabbar^{*} Received on: 8/12/2010

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Abstract

Flue gas desulfurization process has been studied using granular activated carbon in bubbling fluidized bed reactor. For the sake of comparison, fixed bed configuration has been also studied at the same operating conditions. The effect of temperature $30 \leq$ $T \le 80$ °C, inlet SO₂ concentration $500 \le C_o \le 2000$ ppm, and flue gas flow rate $2.5 \le$ $Q \leq 30 \ \ell/\text{min}$ were investigated. The results showed that the SO₂ removal efficiency increases with increasing reaction temperature up to 80 °C. Also, it was noted that the removal efficiency decreases with increasing the inlet SO_2 concentration within the range of temperatures studied. The effect of flue gas flow rate on the desulfurization activity was in two ways; an increase in the removal efficiency with increasing gas flow rate was observed below flow rate = 7.5 ℓ/\min , while a decrease in the efficiency was observed upon any increment in the gas flow rate beyond the 7.5 ℓ /min. The results of fluidized bed reactor were used to obtain an empirical correlation and the experimental results were well correlated with the proposed form with a correlation coefficient, (R) = 0.989. A reaction rate equation was proposed for the oxidative desulfurization and the activation energy was obtained using differential analysis of integral reactor technique. The resulted value of apparent activation energy was 2.981 kJ/mol.

Keywords: SO₂ Removal, Activated Carbon, Fluidized & Fixed Bed Reactors

ازالة ثنائي اوكسيد الكبريت في مفاعلي الطبقة المميعة والثابتة باستخدام الكاربون المنشط

الخلاصة

تم في هذا البحث در اسة عملية از الة غاز ثنائي اوكسيد الكبريت من الغاز الناتج من حرق الوقود الحاوي على الكبريت (الغاز العادم) باستخدام الكاربون المنشط في مفاعل الطبقة المميعة ذو الفقاعات. لغرض المقارنة، تمت در اسة العملية في مفاعل الطبقة الثابتة و عند نفس الظروف التشغيلية. تم در اسة تاثيرات درجة حراره المفاعل (30-80) درجه مئوية، تركيز غاز ثنائي اوكسيد الكبريت الأبتدائي (200-500) جزء من المليون, و سرعة تدفق الغاز (2.5-30) لتر/دقيقة. اظهرت النتائج ان كفاءة الاز الة لغاز ثنائي اوكسيد الكبريت تزداد بزيادة درجة حرارة المفاعل بحيث الى حد 80 درجة مئوية. دلت النتائج على أن التراكيز الابتدائية العالية لغاز ثنائي وكسيد الكبريت تقلل من كفاءة از الته عند جميع درجات المرارة التي درست. اثبتت النتائج ان تاثير معدل تدفق الغاز على كفاءة الاز الية هو ذو نوعين؛ عند معدلات التدفق الحجمي الاقل من 7.5 لتر/دقيقة، زيادة التدفق الغاز على كفاءة الاز الية هو ذو نوعين؛ عند معدلات التدفق الحجمي الاقل من 7.5 لتر/دقيقة، زيادة التدفق الحمي للغاز تؤدي الى زيادة في كفاءة الاز الة، بينما عند معدلات التدفق الحجمي الاقل من 7.5 لتر/دقيقة، ويادة الذ العاز على نوريد الذرائية من كناءة الاز الة مند معدلات المواتي المولية التو من 5.5 لتر/دقيقة، ويادة التدفق الغاز على كفاءة الاز الية مو ذو نوعين؛ عند معدلات الموات التدفق الحجمي الاقل من 5.5 لتر/دقيقة، ويادة التدفق الحمي للغاز تؤدي الى زيادة في كفاءة الاز الة، بينما عند معدلات التدفق الحجمي الاقل من 5.5 لتر/دقيقة، وياضير

*Chemical Engineering Department, University of Technology/Baghdad

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2412-0758/University of Technology-Iraq, Baghdad, Iraq This is an open access article under the CC BY 4.0 license <u>http://creativecommons.org/licenses/by/4.0</u> وان النتائج العملية توصف جيدا على اساس الصيغة المفروضة وكانت قيمة معامل الارتباط (R) تساوي 0.989. تم فرض معادلة لمعدل تفاعل اكسدة ثنائي اوكسيد الكبريت وطاقة التنشيط حسبت باستخدام طريقة التحليل التفاضلي لنتائج المفاعل التكاملي. وجدت طاقة التنشيط تساوي 2.981 كيلو جول/مول.

1. Introduction

ulfur dioxide (SO₂) is considered among the most toxic gases emitted to the atmosphere during the combustion of fossil fuels, for example, coal and diesel oil [1]. Sulfur dioxide can cause acid rain, which has acidified soils and streams, accelerated corrosion of buildings and reduced visibility. Long-term exposure to SO₂ also results in various respiratory diseases [2]. Among the famous emissions reducing methods is flue gas desulfurization (FGD). A variety of flue gas desulfurization techniques are available for SO₂ removal from flue gases. These can be broadly classified into two general categories: non-regenerable regenerable and processes. These can be subdivided into wet and dry processes [2].

It has been shown in the literature that activated carbon act simultaneously as adsorbent and as catalyst for SO₂ oxidation even at low temperatures [3]. The removal of SO₂ over activated carbon in the presence of oxygen and water vapor at relatively low temperatures (20-150 °C) involves a series of reactions that leads to the formation of sulfuric acid as the final product. The overall reaction is [4]:

For flue gas desulfurization, the fluidized bed reactor has the following advantages over fixed bed reactor:

1. It can prevent the plugging phenomenon that can occur in a fixed-bed reactor as fly ash passes through the reactor.

- 2. High thermal /mass transfer coefficient, high gas/solid, and solid/solid contact areas are continuous.
- 3. It does not require a reactor shut down for the replacement of the adsorbent [5].

Lee [6] studied SO_2 removal with char from flash carbonization process as a potential adsorbent. A fluidized bed was employed. He found that increasing the velocity reduces the mass transfer resistance (both pore and external film resistance) by compression forces. He also reported that removal efficiency decreased when the velocity of gas increased.

Rau et al. [5] studied the removal of acid SO₂ gas by oxidation on activated carbon in fluidized bed reactor. They used activated carbon with a diameter of 1 mm classified as Geldart B powders, with constant flue gas velocity of $u/u_{\rm mf} = 1.47$. The results of their work indicated that for activated carbon treated with HNO₃ removal efficiency reaches over 84%. They concluded that the fluidized bed adsorbent/catalyst reactor have a high potential for simultaneous removal of high concentrations of SO₂ and fly ash from flue gases. Cho and Miller [7] worked on the desulfurization by activated coal and found the temperature to influence the average removal efficiency by increasing it upon increasing the temperature. The profiles of curves of average removal efficiency against reaction temperature were exponential in shape in their work. Liu et al. [8] used waste semicoke as the raw material to prepare carbon catalyst for SO_2 removal. They found the dependence of sulfur dioxide retention on space velocity was of two types. Sulfur dioxide retention increased from 7.4 to 9.6 g SO_2 /100 g C with increasing space velocity from 540 to 830 h⁻¹, however, further increasing the space velocity decreased the sulfur retention. Thus, they

concluded that the kinetic behavior varied with space velocity and the desulfurizing property was controlled by diffusion at space velocities below 830 h⁻¹, and controlled by the oxidation step (kinetic step) at space velocities above 830 h⁻¹. Most recently Li et al. [9] have confirmed the results of Liu et al. [8], where similar results were obtained and desulfurization activity was controlled by diffusion below 800 h⁻¹ and controlled by the oxidation reaction above 800 h⁻¹. Mochida et al. [10] conducted a kinetic study of the removal of SO₂ using polyacrylonitrilebased activated carbon fibers. They developed a power-law model to describe their data and obtained the exponents of concentration terms in the model. The exponent of sulfur dioxide was between 0 and 1. Gaur *et al.* [11] examined the effect of concentration on the breakthrough curves. It was found that the outlet concentration of the SO₂ expectedly increased with increase in the inlet SO₂ concentrations, indicating relatively earlier saturation of the AC at higher inlet SO₂ concentrations.

Raymundo-Pinero *et al.* [12] Lizzio and DeBarr [13], and Otake *et al.* [14] have proposed the SO_2 oxidation to be the rate-limiting step. Following the assumption that the ratedetermining step in the overall reaction is catalytic oxidation of SO_2 to SO_3 , an overall rate expression was proposed by Lizzio and DeBarr [13]:

$$-\mathbf{r}_{\mathrm{SO}_2} = \mathbf{k} \left[\mathbf{C}_{\mathrm{SO}_2} \right]^n [\mathbf{C}_{\mathrm{A}}] \tag{2}$$

Where $[C_A]$ denotes the concentration of the free active sites, k is rate constant, and *n* is the order of reaction with respect to SO₂. The rate is then only a function of the concentration of free sites and SO₂. For concentrations of SO₂ <1500 ppm, it was found that values of *n* between 0.5 and 1, and for concentrations of SO₂ >1500 ppm, the value of *n* approached 0. Up to concentration of SO₂ of approximately 500 ppm the value of *n* was 1. For higher concentrations, the value of *n* decreased [13].

The overall objective of this research is to investigate the performance of granular activated carbon for SO₂ removal from simulated flue gas in fluidized bed reactor and comparison with that of fixed bed reactor at the same operating conditions. The operating parameters are: flue gas flow rate [(2.5-12.5)] /min for fixed bed and (13.5-30) \mathbf{l} /min for fluidized bed], reaction temperature (30-80) °C, and inlet SO₂ concentration (500-2000) ppm.

2. Experimental Work

The experimental runs were performed in a laboratory-scale apparatus. The general layout of the experimental apparatus is displayed in Figure (1). The experimental rig consists of three sections; SO_2 generation section, reaction section, and analysis section. The reaction section is a QVF column of 1 in diameter and 50 cm long and contains activated carbon bed to a height of 12 cm and weight of 30 gm with particle diameter dp = 1.09 mm. The physical properties of the activated carbon were measured using surface area analyzer and are presented in Table (1). Heat is supplied to the reaction section via electric heater of 60 Watt power. A Ttype thermocouple fitted along the axis of the reactor reaching the middle of the packing measured the temperature of the bed. The thermocouple sends its signal to a PID temperature controller. The power consumed by the heater was controlled manually by varying the voltage of the current by means of Variac transformer.

2.1 Experimental Procedure

The reaction section was charged with 30 gram of activated carbon with an average particle diameter of

(1.09) mm which gave a bed height of 12 cm above the distributor plate. This charge was replaced with fresh one after each test to ensure the bed activity maintained constant. is In the generation vessel, 300 gram of Sodium Sulfite powder was placed for each run. The analysis section was filled with 400 ml of standard Iodine solution (0.1 N) which was replaced at the end of each run. The controller was set to the desired temperature and the variac transformer was adjusted to control the amount of the power supplied depending on the gas flow rate and temperature required. Sulfur dioxide gas was synthesized by dropping the desired concentration of sulfuric acid solution into the SO₂ generation vessel

that contains sodium sulfite powder according to the chemical reaction:

$$Na_2SO_3 + H_2SO_4 \rightarrow SO_2 + Na_2SO_4 + H_2O$$
(3)

Air stream at the desired flow rate carries the SO_2 to the reaction section. The outlet gas stream (from the top of the bed) was introduced into iodine solution to absorb the residual sulfur dioxide gas according to the reaction:

$$SO_2 + I_2 + H_2O \rightarrow 2HI + H_2SO_4$$
 (4)

In order to measure the concentration of SO_2 in the effluent gas stream, (10) ml of the iodine sample was taken from the absorption trap, titrated with standard sodium thiosulfate solution (0.1 N), in the presence of starch indicator. This step was repeated every 5 minutes till the end of experimental run time (1 hr). Each experiment was repeated twice to validate the experimental results. The removal efficiency of SO₂ was calculated as the ratio of SO_2 concentration that was removed by activated carbon to the initial

concentration of SO_2 gas fed to the bed:

$$\eta (\%) = \frac{C_0 - C}{C_0} * 100$$
 (5)

The average removal efficiency of SO_2 was calculated as the average of the values of removal efficiencies obtained at each 5 min throughout a run.

$$\eta_{\rm av} = \frac{\Sigma \eta}{12} \tag{6}$$

3. Results and Discussion

The variation of SO_2 removal efficiency with flow rate is shown in Figure (2) at different operating temperatures and constant initial concentration of SO_2 of 500 ppm, for one hour duration time for fluidized bed operation.

The purpose of these experiments was to determine the best flue gas flow rate value in fluidized bed that gives higher removal efficiency in the range of flow rates studied. Although higher average removal efficiency of 96% was obtained at Q= 13.5 I/min, this value was slightly above the minimum fluidizing velocity. Bubbling fluidization was clearly visualized at Q = 15 I/min. This value was used throughout the ongoing sets of experiments of the effect of reaction SO₂ temperature and initial concentration as bubbling fluidization is the regime understudy in the present work.

As shown in Figure (2), on increasing the flue gas flow rate the removal efficiency decreases, this can be explained by the decrement of SO_2 residence time in bed thus decreasing the amount of SO_2 retained by the surface of carbon particles which depends mainly on the exposure time in the vicinity of the inner carbon surface that decreases with high velocities. These results agree with that of Lee [6].

Figure (3) shows the dependence of average removal efficiency of sulfur dioxide on reaction temperature for different SO₂ inlet concentrations at the constant flue gas flow rate of 15 1/min in fluidized bed. Sulfur dioxide removal efficiency significantly increased with increasing temperature and reached highest value at $T = 80 \ ^{\circ}C$ for each of the four inlet SO_2 concentrations examined. The slowest step in the surface steps is the SO_2 catalytic oxidation performed by surface groups [12,13,14]. Thus SO₂ interacts with surface groups due to the

strong attractive forces exerted by these groups that capture SO₂ and catalyze its oxidation reaction to SO₃. Increasing the temperature will increase the rate of formation of SO₃ due to the faster oxidation of SO₂ as a consequence of rapid activation of the SO₂ molecule that will react in a shorter period of time at the increased temperatures. SO₃ formed is less volatile, strongly adsorbed on carbon surface [15], so that SO₃ do not require the attractive forces of the high energy adsorption sites to hold it on surface. After oxidizing an SO₂ molecule, the functional groups are freed to attract successive SO₂ molecule and oxidizing it, which increases the amount of SO₂ uptaked. Any increase in temperature would have its effect on the reaction rate thus increases the rate constant k, as it is evidenced by the likeness of profiles of the curves, which show that the average removal efficiency has an exponential-like dependence on reaction temperature emerging from obeying Arrhenius equation in its variation with temperature. These results agree with those of Rau et al. [5] who reached maximum value of removal efficiency of 84% at optimum reaction temperature of 200 °C, and with the results of Cho and Miller [7].

Effect of inlet SO_2 concentrations on SO_2 removal efficiencies for various reaction temperatures is shown in Figures (4) to (6) for fluidized bed operation. These figures show that SO_2 removal efficiency decreased as the concentration of SO_2 increased at different operating temperatures. This is attributed to the order of oxidation reaction (*n*), which is the rate determining step, being less than 1 with

respect to SO_2 (when the initial concentration roughly exceeds 500 ppm according to Lizzio and DeBarr [13] so that the gas removal efficiency is decreased with increased concentration. Thus it can be concluded from these results along with the findings of Lizzio and DeBarr [13] that the desulfurization efficiency as well as the order of dependence of reaction rate $(-\mathbf{r}_{SO_2})$ on SO₂ concentration, are both functions of inlet SO₂ concentration. The removal efficiency is inversely proportional to the inlet SO₂ concentration, but directly proportional to the weight of carbon bed [6].

The effect of flue gas flow rate on SO₂ removal efficiency at constant inlet concentration of 500 ppm and different temperatures for fixed bed reactor (combined with that of fluidized bed reactor) is shown in Figure (7). The results show that as the gas flow rate increases the removal efficiency increases up to a value of Q = 7.5 \mathbf{I} /min at which maximum removal efficiency is reached, thus flow rate of 7.5 \mathbf{I} /min is the optimum flue gas flow rate. Beyond this value any increase in gas flow rate will lower the removal efficiency.

At lower flue gas flow rates (below the optimum flow rate), the desulfurizing behavior is controlled by diffusion of reactant SO_2 into the inner

surface of the sorbent. In this way, increasing the gas flow rate will increase the diffusion rate as a result of the increased forces of convection. The retention times at these flow rates in the vicinity of active surfaces are long enough to convert adsorbed SO_2 to H_2SO_4 , due to the minimized diffusion resistance. Hence, sulfur dioxide retention increases with flow rate up to the optimum flow rate (7.5 1/min), where starting from this value the desulfurizing behavior is possibly controlled by oxidation step (i.e. kinetic step) because of completely minimized diffusion resistance. Due to the reduction of retention time at higher velocities (at or above the optimum flow rate), reactants cannot have the sufficient time for oxidation of large amount of SO₂ to form SO₃ to take place, which leads to the decrease of sulfur dioxide retention [8].

Referring to Figure (7), it can be recognized two regions of effect of gas flow rate on removal efficiency suggesting that there are two different ways by which gas flow rate can change the efficiency. The experimental runs can be divided according to either of the two regions of Figure (7) they belong to, either into the region below the optimum flue gas flow rate of 7.5 \mathbf{l} /min, which is the case of fixed bed reactor, or the region at or above the optimum flow rate in which all mass transfer resistance due to the accessibility obstacles to the porous structure is well overcomed (because of high velocities) and gas molecules is flushed through the carbon, which is the case of the highest velocities in fixed bed reactor, and fluidized bed reactor. According to Gauthier [16] the elimination of diffusional limitations (internal diffusion limits inside particles), gives

rise to an effectiveness factor very close to unity. This makes the oxidation step controls at or above the 7.5 \mathbf{I} /min, where the global rate will be the same as reaction rate. These results are consistent with those of Li et al. [9], Liu et al. [8].

Figure (8) shows the effect of temperature on the average removal efficiency of SO₂ at the optimum flue gas flow rate of 7.5 \mathbf{I} /min for different inlet SO₂ concentrations in fixed bed conditions. As shown in the figure, increasing temperature increased the removal efficiency and approached 100% removal at 80 °C and 500 ppm. Thus temperature has positive effect on the desulfurization process in that it increases the reaction rate. An increase in removal efficiency on increasing reaction temperature was also observed by many investigators [7,8,9,14].

The trend of the results shows a similar pattern as that obtained in fluidized bed, thus temperature affects in the same manner in both reactors emerging from same reaction steps in both reactors. As in fluidized bed results, the exponential-like behavior is also observed in Figure (8) for fixed bed operation.

Figures (9) to (11) illustrate the effect of inlet SO₂ concentrations on its removal efficiencies at various reaction temperatures for fixed bed operation, with the gas flow rate kept constant at its optimum value of 7.5 \mathbf{l} /min. The removal of SO₂ decreased with increasing the inlet SO₂ concentrations, indicating the suitability of the current amount of sorbent to raise the value of SO₂ conversion with rising inlet concentrations only for low levels of inlet concentration [11]. At 500 ppm inlet concentration, gas nearly

complete removal was observed, the removal efficiency decreased to 97% at

the inlet concentration of 1000 ppm both at the best oxidative reaction temperature obtained for this work of 80 °C (Figure (11)). Removal efficiency was observed to decrease to its lowest values at all temperatures tested corresponding to the inlet gas concentration of 2000 ppm.

Gaur et al. [11] showed that the exit gas concentration of SO₂ gradually increased at higher inlet SO_2 concentration suggesting inadequacy of bed for high concentrations. The amount of sorbent existing is fixed while the inlet SO₂ concentration is continuously increasing. This is the reason of why the reaction order is less than 1 at high concentrations. This idea of the change of the order of dependence of reaction rate on SO_2 concentration, with increased initial concentration of SO_2 due to consumption or decaying of free sites was first suggested by Lizzio and DeBarr [13]. A decrease in removal capacity upon increasing SO₂ concentration observed was by Mochida et al. [10].

3.1 Empirical Correlation

Based on the experimental results obtained in the fluidized bed reactor, an empirical correlation that includes the effect of operating variables on the removal efficiency was obtained in this study. The correlation incorporates the effect of temperature in terms of exponential-like dependence, and the gas velocity which is the crucial variable in fluidized beds. Gas velocity has effects on the removal process through affecting residence time in the bed as well as the efficiency of contact between particles and gas during the residence time. The first term is well described by a dimensionless velocity. It is well proven that the particle Reynolds number (Re_p) is a good characteristic dimensionless group for

fluidized beds representing gas velocity. The second effect is due to formation of gas voids in different forms such as channels, bubbles, and slugs by the excess gas $(u-u_{\rm mf})$. Hence gas holdup or bed voidage, a critical hydrodynamic parameter, is used as the convenient dimensionless expression of the inefficient contact component of the gas velocity effect on η . At first, bed viodage was correlated with the velocity using a form similar to that first suggested by Richardson and Zaki [17] for liquid fluidized beds, and used by Davies and Richardson [18] for gassolid systems:

$$\frac{u}{u} = \mathbf{a}(\varepsilon)^{n} \tag{7}$$

The STATISTICA software is applied to evaluate the empirical constants, a, and, n, and were found to be 0.4607 and 2.3827 respectively with correlation coefficient (*R*) = 0.973.

The second step a correlation form is suggested for the dependence of removal efficiency on the operating variables as mentioned earlier and as follows:

$$\eta_{av} = 1 - Y = 1 - \frac{c}{c_0} = A (exp\{\frac{B}{T}\})(\epsilon)^{N1} (Re_p)^{N2}$$
(8)

Where A, B, N1, and N2 are constants. The constants were calculated using STATISTICA software. The final empirical formula is:

$$\eta_{av} = 1 - Y = 1 - \frac{c}{co} = 179.7090 \ (exp - \frac{3.3804}{T})$$

(ϵ)^{-0.0691} (**Re**_p)^{-0.1864} (9)

with correlation coefficient (*R*) = 0.989. Figure (12) gives the experimental η_{av} against that predicted by the correlation Eq. (9).

4. Kinetic Analysis

Kinetic analysis of the experimental results was conducted to obtain the kinetic parameters of the catalytic oxidation of SO_2 , since oxidation reaction is the rate limiting step for the range of flue gas flow rates at or above

the optimum flow rate of 7.5 ℓ/\min as shown in the region of decaying efficiency in Figure (7). The suggested rate expression assumes first order reaction with respect to SO₂ as reported by Otake et al. [14], and facilitating the findings of Lizzio and DeBarr [13] that the reaction order = 1up to approximately 500 ppm inlet concentration of SO₂, which is the concentration used in the experiments of varying W/F_{SO2,0} values in this work. The rate expression given in Eq. (10) is based on the rate expression of Lizzio and Debbar [13] (Eq. (2)) with simplification made by dropping the term of active sites decaying, since η decreased a little during the run (1 hour), thus:

Differential method of kinetic analysis stated by Levenspiel [19] was applied.

 $-\mathbf{r}_{SO_2} = \mathbf{k} \mathbf{C}_{SO_2}$

At first the conversion (which is taken as the average removal efficiency η_{av}) Vs. W/F_{SO2,o} curves are plotted as shown in Figure (13) for C_o = 500 ppm = 0.0203 mol/m³. Then the slopes ($\frac{dx}{d(\frac{W}{F_{SO2,o}})}$) at selected points of W/ F_{SO2,o}

on the conversion vs. $W/F_{SO2,o}$ curve which equal to the reaction rate $-r_{SO_2}$,

were plotted against C_{SO_2} . The linear relation between $-r_{SO_2}$ and C_{SO_2} (as shown in Figure (14) for T = 80 °C as an example) gives the rate equation:

$$-\mathbf{r}_{SO_2} = \left(\mathbf{0.882} \ \frac{\mathbf{m}^3}{\text{Kg cat.min}}\right) \left(\mathbf{C}_{SO_2}, \frac{\text{mol}}{\mathbf{m}^3}\right)$$
(11)

The results of the rate constant at the three tested temperatures are given in Table (2). The apparent activation energy for the oxidative desulfurization reaction is calculated by using the Arrhenius equation

The plot of $(\ln k)$ vs. (1/T) shown in Figure (15) gives a straight line with

 $\label{eq:slope} \begin{array}{ll} \text{slope} = - \ E/R \ \text{and intersection} = ln \ k_o \\ \text{so that} \ E=2.981 \ kJ/mol & k_o = 2.2 \\ m^3/Kg \ cat.min \end{array}$

Thus the reaction rate constant will be: $k= 2.2 * \exp(-2981/8.314*T)$, T in K

(12)

And the oxidation reaction rate equation is

$$-\mathbf{r}_{SO_2} = 2.2 \, \exp\left(-\frac{2981}{8.314*T}\right) \mathbf{C}_{SO_2}, \, \text{T in K}$$
(13)

5. Conclusions

For both fixed and fluidized bed reactors an increase in the SO₂ removal efficiency was observed with an increase in the reactor temperature and а decrease of the SO₂ initial concentration in the flue gas stream. Curves of η_{av} Vs. T showed an exponential like behavior for both reactors. The bed voidage in fluidized bed reactor was correlated with u/u_t with (R) = 0.973. The effect of fluidized bed hydrodynamics on the removal efficiency was correlated with (R) = 0.989. A rate expression was suggested for the oxidation reaction. This rate equation was found to be accurately describes the reaction kinetics and the activation energy of the reaction was found to be 2.981 kJ/mole.

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Nomenclature

Constant in Eq.(8)	[-]
Constant in Eq.(9)	[-]
Constant in Eq.(9)	[-]
Inlet concentration	ppm
of Sulfur Dioxide	
Concentration of	mol/m ³
Sulfur Dioxide in	or
the reactor	mg/m ³
Effluent	ppm
concentration of	
Sulfur Dioxide	
Activation energy	kJ/mol
Molar Sulfur	mole/m
Dioxide flow rate	in
Reaction rate	m ³ /min
constant	. kg cat
Pre-exponential	m ³ /min
(frequency) factor	. kg cat
Constant in Eq.(8)	[-]
	Constant in Eq.(8)Constant in Eq.(9)Constant in Eq.(9)Inlet concentration of Sulfur DioxideConcentrationof Sulfur Dioxide in the reactorEffluent concentration of Sulfur DioxideSulfur DioxideActivation energyMolarSulfur Dioxide flow rateReactionrate constantPre-exponential (frequency) factor

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		1
N2	Constant in Eq.(8)	[-]
$-\mathbf{r}_{SO_2}$	Oxidation reaction	mol/kg
	rate	cat .
		min
Rep	Particle Reynolds	[-]
	number = $\frac{\rho_g dp u}{\mu_{\sigma}}$	
	'8	-
Т	Bed temperature	°C
t	Time	min
и	Superficial velocity	m/s
	of gas	
$u_{\rm mf}$	Minimum	m/s
	fluidization velocity	
ut	Terminal	m/s
	(maximum) velocity	
W	Weight of activated	kg
	carbon in the bed	
х	Sulfur dioxide	[-]
	conversion to sulfur	
	trioxide	
Y	Dimensionless SO ₂	[-]
	concentration = $\frac{C}{C_0}$	
	Co	
ε	Voidage of the	[-]
C	expanded bed	1
η	Removal efficiency	[-]
.,	of SO_2	LJ
η_{av}	Average removal	[-]
·lav	efficiency of SO_2	11
AC	Activated Carbon	[-]
AU	Activated Carboll	[1]

Table (1) Physical Properties of Activated Carbon

Bulk density, (kg/m ³)	500
Surface area, (m ² /g)	804.32
Bed voidage, ε_m , (-)	0.48
Particle density, (kg/m ³)	965

 Table (2) The Values of Rate Constant for the 1st Order Reaction

 Rate at Different Temperatures.

Reaction temperature (K)	k (m ³ /min. Kg cat)
303	0.76
323	0.8
353	0.882

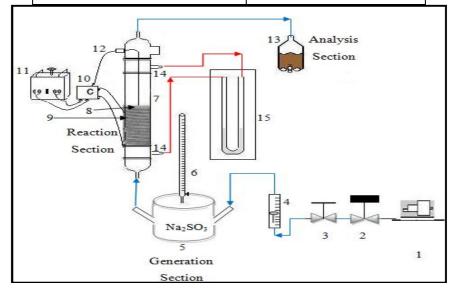


Figure (1) Schematic Diagram of the Experimental Setup. (1) Compressor (2) Pressure regulator (3) Needle Valve (4) Rotameter (5) SO₂ generation vessel (6) Burette (7) Reactor (8) Activated Carbon (9) Heater (10) Temperature controller (11) Variac (12) Thermocouple (13) Absorption trap (14) Pressure tap (15) U-tube manometer.

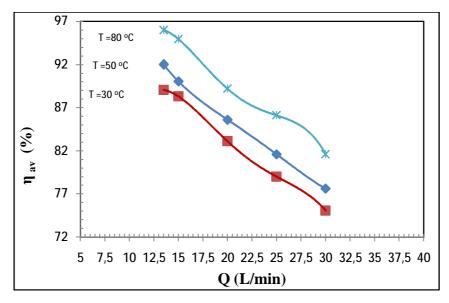


Figure (2): Effect of flue gas flow rate on the average removal efficiency of SO_2 at $C_0=500$ and different temperatures in fluidized bed for one hour duration.

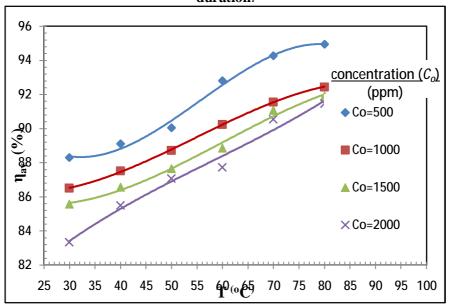


Figure (3) the effect of temperature on the average removal efficiency of sulfur dioxide at Q=15 1/min and different inlet SO₂ concentrations in fluidized bed.

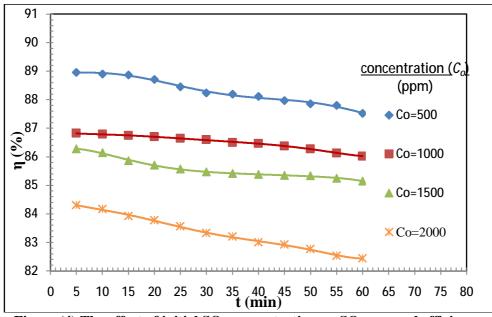


Figure (4) The effect of initial SO_2 concentration on SO_2 removal efficiency $(T=30 \ ^0C)$ in fluidized bed.

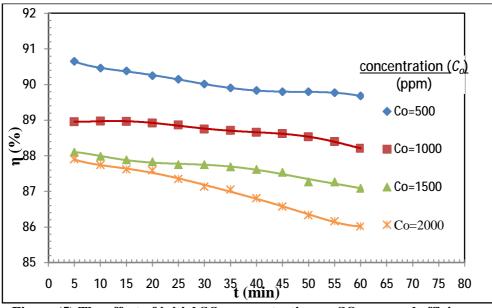


Figure (5) The effect of initial SO₂ concentration on SO₂ removal efficiency $(T=50 \ ^{0}C)$ in fluidized bed.

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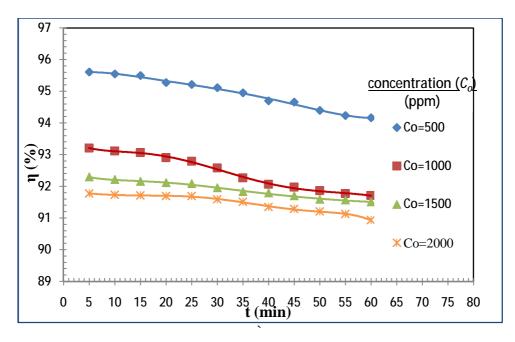


Figure (6) The effect of initial SO₂ concentration on SO₂ removal efficiency $(T=80 \ ^{0}C)$ in fluidized bed.

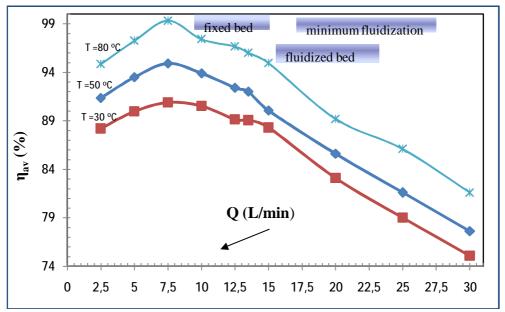


Figure (7) The effect of flow gas flow rate on the average removal efficiency of SO_2 at different temperatures in fixed and fluidized bed for one hour duration

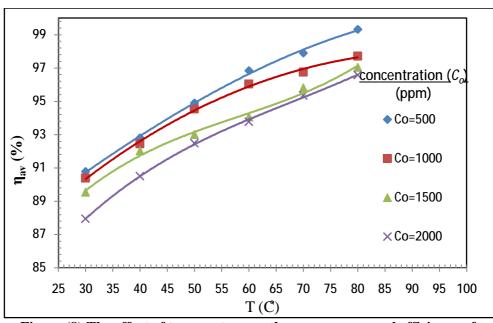
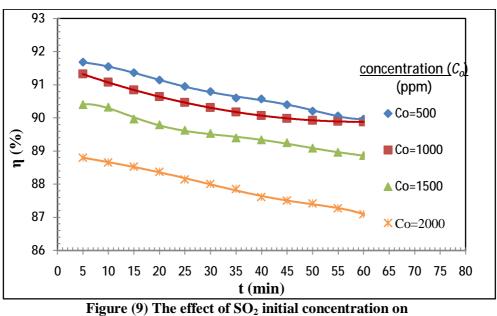


Figure (8) The effect of temperature on the average removal efficiency of sulfur dioxide at Q=7.5 ¹/min and different inlet SO₂ concentrations in fixed bed.



removal efficiency (T = 30 ^oC).

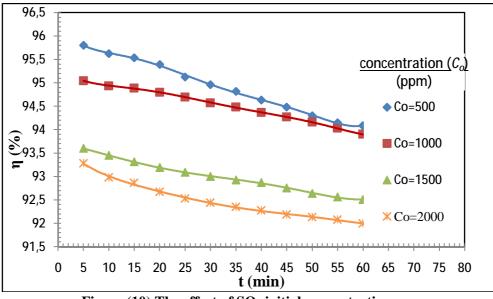
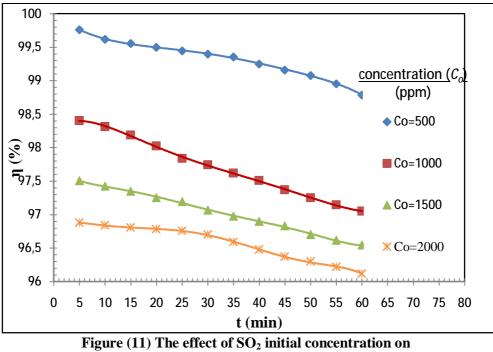


Figure (10) The effect of SO_2 initial concentration on removal efficiency (T= 50 0 C).



removal efficiency ($T = 80^{\circ}C$).

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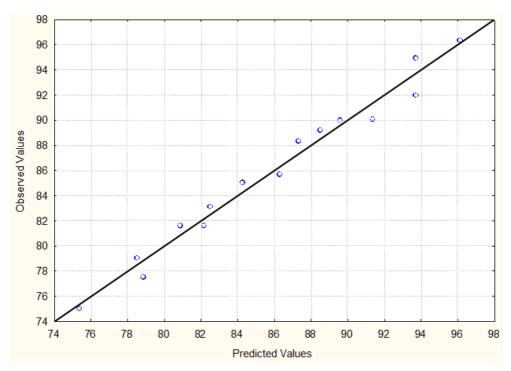


Figure (12) The observed Vs. predicted values of average removal efficiency. $C_0 = 500$ ppm, T=(30-80) °C, and Q = (13.5-30) ℓ /min for one hour duration.

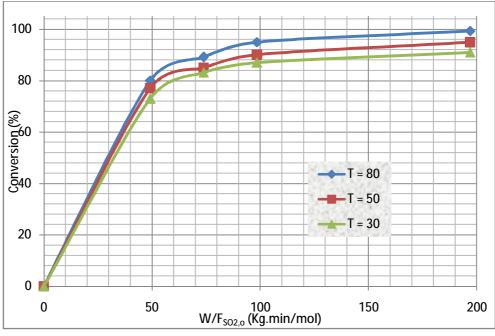


Figure (13) Conversion of SO_2 as a function of $W/F_{SO2,O}$ at different reaction temperature.

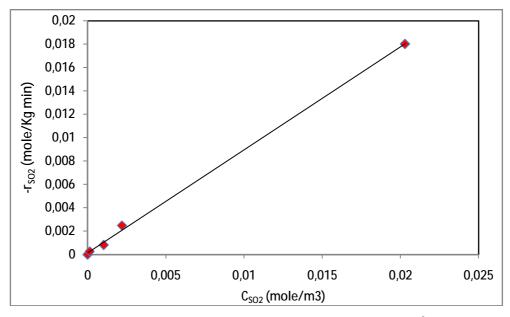


Figure (14) Relation between $-r_{SO_2}$ and C_{SO_2} for $T=80\,^{o}C$

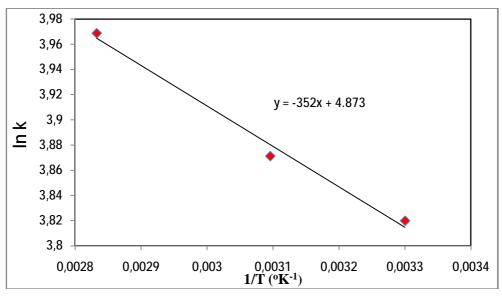


Figure (15) Plot of ln k Vs. 1/T.