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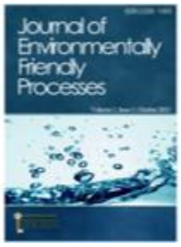

Using RSM method to determine optimum process conditions for flue gas desulfurization through an amine scrubber

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Abstract:

In this study the optimum process conditions for absorption of sulfur dioxide in a mixture of flue gases were determined. Using a selective amine-based absorber, a high amount of SO2 was absorbed in the scrubbing process. Some process conditions such as the temperature, concentration of the SO2, and absorber flow rate in absorption were experimentally researched. The optimized conditions were finally specified after experiments by using response surface method (RSM)experimental design method. It was observed that desulfurization of flue gas for our solvent pilot operates at the maximum performance at absorption temperature 60 C, SO2 concentration of 4000 ppm, desorption temperature of 110 °C, and the gas flow rate of 300((lit)/min)An efficiency of more than 99 % could be obtained by varying the parameters in which all the released SO2 gas was absorbed from the inlet flue gas. An efficiency of more than 99 % could be obtained by varying the parameters in which all the released SO2 gas was absorbed from the inlet flue gas; an achievement that is much favorable for industrial purposes.

Keyword: SO2 , (RSM), Optimization, Sulfur dioxide, Absorption

1. Introduction

Sulfur dioxide is one of the main contaminating gases that is mostly released by fossil power plants, or chemical and petrochemical factories [17][18][19]. Coal burning power plants may also release SO_2 to the atmosphere since most types of coals contain sulfur. [11][10] .The results of these toxic emissions may result in acid rain that has been a great concern during recent years [15][16]. Nowadays, developed countries have passed strict regulations to restrict SO_2 emissions into the atmosphere. Establishment of new methods that introduce regenerative absorbers with low construction costs has brought in new efficient process designs and conditions [9][5][13][20]. Scrubbers are the most applicable equipments for gas absorption in which mass transfer occurs between liquid and gas states in a counter current flow through various packing materials [14]. Packed towers are very useful for removing industrial gaseous pollutants to meet present environmental standards [8]. Many technological absorption methods have been studied among which scrubbing is expected to play a pivotal role in the development of SO_2 capturing process progresses [6][7][21]. Packed scrubbers have mainly played as influencing equipments for removal of flue gas contaminations [4] Researchers are eager to investigate more practical method to attain an allowable rate of SO_2 released to the atmosphere considering global standard limitations [4]. Amine scrubbers purify gaseous airstreams from contaminants [1]. Large wetted surface make extensive mass transfer that is an impressing advantage of the packed scrubbers [12]. Corrosion resistance and low energy consumption are other advantages that make these packed columns applicable. To optimize process conditions of SO_2 absorption in wet flue gas desulfurization, the amount of operation's influencing parameters such as solvent flow rate, SO_2 entering concentration, desorption temperature, and pH value are calculated and set as variables. There is a need of doing experiments to estimate the best process conditions. The experimental design methods like Taguchi bring a certain analysis of process features. This method has proved to be beneficial since it not only reduces the number of experiments but also specifies controllable and uncontrollable factors. It is therefore time efficient and dynamic [2]SO2 absorption performance was investigated by Taguchi's orthogonal array (OA) analysis to obtain appropriate pH value for this pilot plant. This study is focused on the reduction of sulfur dioxide gas with a selective amino compound solvent. Conducting the experiments on the pilot plant in the research center of Abadan Institute of Technology (AIT), the obtained data are discussed in the article. It proved to be a strong solvent; it has brought innovations as SO2 absorber in this research.

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1. Materials and Methods

1.1. Solvent properties

The amine that was used is a selective absorber that absorbs SO2 selectively. CO2 gas, a competitor contamination with SO2 in flue gases, does not interfere with the absorption process. In the process, SO2 is eliminated at ppm levels, while CO2 remains untouched. The physical properties of the absorbent are as the following: viscosity = 0.002 N.s/m2, Absorption density = 1.0643 kg/L, and Boiling point = 105C. The applied absorbent consists of 20 L of distilled water, 2 kg of sulfuric acid, and 2 kg of an ionic-amine additive for selective absorption. Molecular weight of the defined additive was found120 g/mole. The applied absorbent has a great capacity for absorption along with a long durability, as obtained in experimental tests that were done in a 3-month period. During the experiments, no differences were exhibited in the absorbent efficiency. Another important feature of the solvent is that its flow rates as well as concentration in the process are lower than the usual absorbers while the performance is the same, if not better. It needs not to be pure or anhydrous and in industrial scale performs perfectly up to the highest conventional environmental standards. Traditional selective removal processes such as CANSOLV use *20 % (wt/wt) of solvent; however, our solution contains only 8.16 % (wt/wt) of absorber. This reduction in solvent consumption is an innovative aspect that makes the system stronger and more effective. The cost of applied amine (5 $ a pound) is lower than the amine absorbers used in conventional processes (12 $dollars a pound). The production of SO3 during the process results in dilute salt solutions in the process. Later in this paper, Taguchi experimental design method was used to determine absorption parameters and obtain the optimal conditions for removing SO2 from the inlet wet flue gas. All of the working parameters involved in theprocess optimization was investigated in this paper.Soxhlet extraction is a classical extraction technique to remove oil from biomass particle. It is important to study the choice of solvent, extraction time and other process parameters for measuring the % yield of the extracted oil.

1.2. Experimental details

In water solution, dissolved SO_2 undergoes reversible hydration and ionization to produce bisulphite and sulphite according to the following equations.

$$SO_{2(g)} \leftrightarrow SO_{2(aq)} \qquad (1)$$

$$SO_{2(aq)} + H_2O_{(L)} \leftrightarrow H_2SO_{3(L)} \qquad (2)$$

$$H_2SO_{3(L)} \leftrightarrow HSO_3^- + H^+ \leftrightarrow SO_2 + 2H^+ \qquad (3)$$

Adding amine, to the water increases the quantity of SO2 dissolved. According to equation (5-4), the buffer drives the above equilibria to the right by reacting with the hydrogen ions to form ammonium salts. The overall reaction indicates that as the concentration of SO2 in the feed gas increases, the equilibrium moves to the right, i.e. the quantity of SO2 dissolved in the rich solvent increases. Thus, the scrubbing of more concentrated gas streams requires a less than proportional increase in solvent circulation rate. Since the gas volume, and therefore the gas side equipment, remains constant, a relatively small total cost increase is caused by an increase in feed SO2 concentration.

$$NR1R2R3NR4R5 + H^+ \qquad NR1R2R3NR4R5H^+ \qquad (4)$$

1.3. RSM method for design experiment

For optimization we should consider all influential parameters. The parameters affecting SO2 absorption studied in this paper include: the temperature of the desorption column, the temperature of the absorption column, concentration of SO2 in gas inlet of the pilot, and flow rate of the absorbing solution.

Each parameter consider in three level:

- Absorption Temperatures: 40 50, and 60C
- desorption Temperatures: 100, 110, and 120 C

- Absorber flow rate: 100, 250, and 400 mL/min
- Concentration of SO2 inlet: 4000, 6000, and 8000 ppm

This range of absorption temperature is selected because above 60 desorption occurs in absorption Colum and below 40 lack of essential energy lead to low absorption efficiency.

Below the 110 there isn't enough energy for desorption and in temperature above 120 the absorber is boiling and the pressure in desorption column starts increasing and don't allow the so2 realese from absorber.

In selecting absorber rate we consider the evaluable rate in industrial scale, to adapt our process to real form.

concentration the input SO2 was simulated on the basis of this gas concentration in catalytic cracker unit of Abadan oil refinery stack exhaust. Hence, the dealing range is about 6,000 ppm.

Considering all the above mention points we have to done 34 = 81 experiments. There are a lot of experimental method for reduce the number of experiment that we decide to us RSM method for this study. Therefore, 29 experiments are needed to gain optimize SO2 absorption.

Table 1 shows RSM arrays for parameters and levels in the present study. For the designed experiment, we considered four parameters, three levels for each. Therefore, RSM method determines 29 experiments.

Table 1- designed experiment by RSM method

Test #	**L ($\frac{mL}{min}$),**	**Desorption T**	**Absorption tower T (° C)**	**Absorption tower P (Psi)**	**Desorption Tower top P (psig)**	**Desorption Tower bottom P (psig)**	**Solvent concentration % (V/V)**	**Inlet SO2 cocentration(PPM)**	**Outlet SO2 cocentration(PPM)**
1	250	110	50	0.49	2.8	2.8	0.05	2000	15
2	250	110	60	0.49	2.8	2.8	0.05	6000	250
3	400	100	40	0.49	4.2	4.9	0.05	4000	1500
4	100	120	60	0.49	4.2	4.9	0.05	4000	1250
5	400	120	40	0.98	11.2	11.9	0.05	8000	5920
6	100	120	40	0.98	11.2	11.9	0.05	4000	750
7	250	110	30	0.56	11.2	11.9	0.05	6000	310
8	250	110	50	0.56	11.2	1.19	0.05	6000	317
9	100	120	60	0.98	0.7	0.7	0.05	8000	3000
10	400	100	60	0.98	0.7	0.7	0.05	4000	200
11	250	110	50	0.56	11.2	11.9	0.05	6000	287
12	250	100	50	0.56	11.2	11.9	0.05	6000	300
13	250	110	50	0.98	4.2	4.9	0.05	6000	325
14	250	120	50	0.98	4.2	4.9	0.05	6000	1233
15	100	100	40	0.42	11.2	11.9	0.05	8000	1120
16	400	100	40	0.42	10.5	11.2	0.05	8000	2240
17	250	110	50	0.42	2.8	2.8	0.05	6000	287
18	100	100	40	0.42	0.7	0.7	0.05	4000	700
19	100	100	60	0.49	2.8	2.8	0.05	4000	400

20	250	110	50	0.49	2.8	2.8	0.05	10000	800
21	400	110	50	0.49	4.2	4.9	0.05	6000	215
22	400	120	60	0.49	4.2	4.9	0.05	8000	2662
23	400	120	60	0.98	11.2	11.9	0.05	4000	450
24	250	120	50	0.98	11.2	11.9	0.05	6000	325
25	250	110	50	0.56	11.2	11.9	0.05	6000	323
26	100	120	40	0.56	11.2	1.19	0.05	8000	6220
27	250	110	50	0.98	0.7	0.7	0.05	6000	293
28	400	120	40	0.98	0.7	0.7	0.05	4000	1250
29	400	100	60	0.56	11.2	11.9	0.05	8000	1120
30	100	100	60	0.56	11.2	11.9	0.05	8000	1125

To distinguish whether or not our data statistical quality 4 charts should be monitored

1- residuals versus predicted.(fig1)

2- normal plots of residuals(fig2)

3- The predicted versus actual(fig3)

4- Box-Cox(fig4)

If all above Charts will be the necessary conditions, final model would be acceptable.

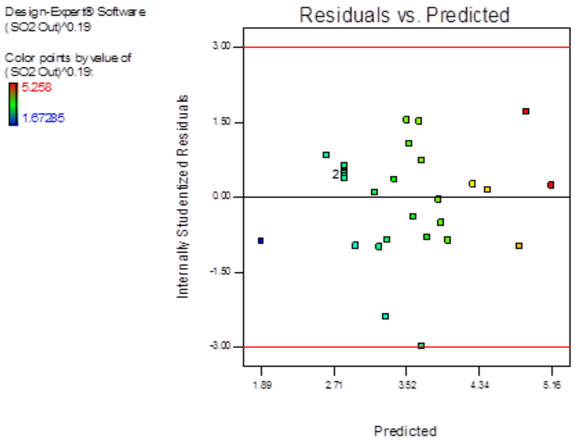


Figure 1- residuals versus predicted

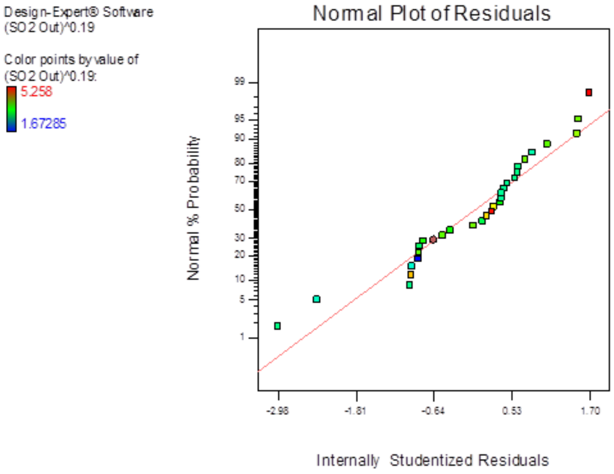


Figure 2- normal plots of residuals

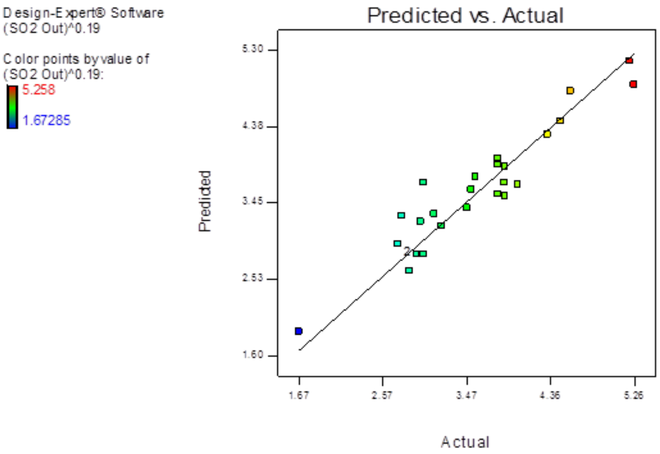


Figure 4- The predicted versus actual

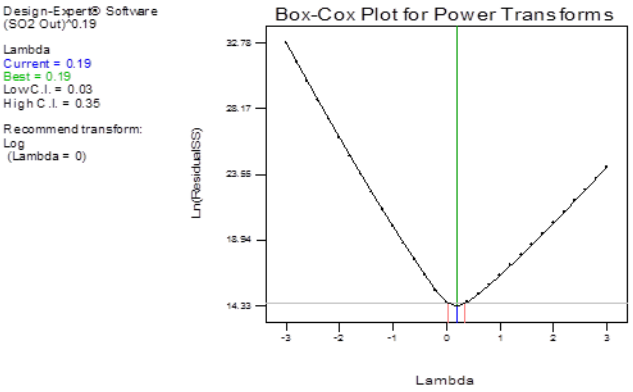


Figure4- Box-Cox

2. Result and discussion

2.1. Three dimentional graphs

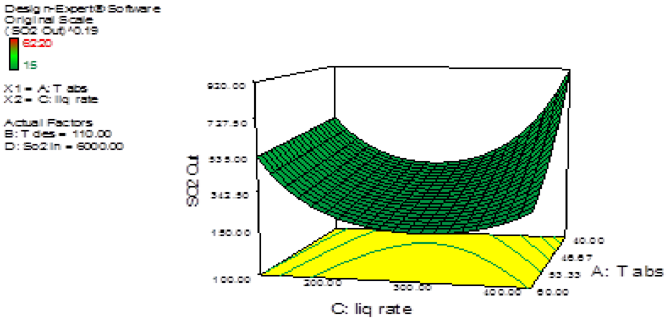


Figure5- So$_2$ out versus absorbtion liquid rate and absorbtion temperature

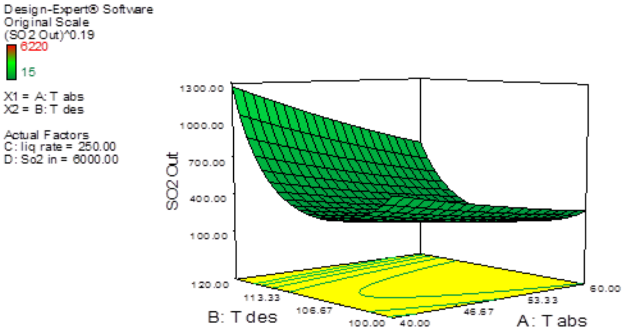


Figure6- So2 out versus desorption temperature and absorbtion temperature

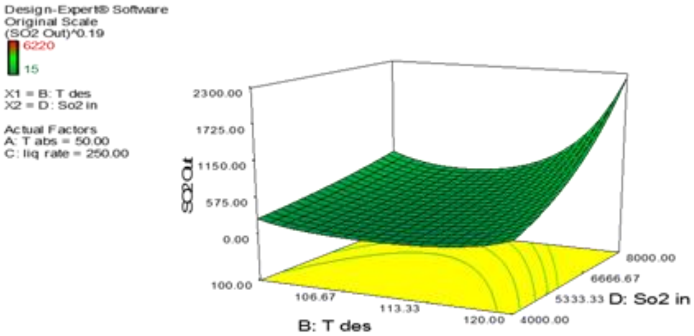


Figure7- So2 out versus desorption temperature and inlet So$_2$ concentration

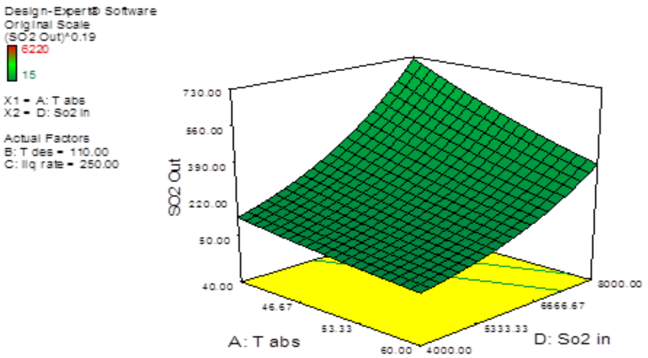


Figure6- So2 out versus inlet So_2 concentration and absorption temperature

2.2. Optimization

After analyzing all datas, software (Design Expert 7)identify 9 points as optimization points

Table2- final optimization

Number	T abs	T des	liq rate	SO2 in	SO2 Out	Desirability
1	60	109.12	277.99	4000.01	54.75	0.87
2	60	109.07	276.58	4000.02	54.75	0.87
3	60	109	274.8	4000	54.78	0.87
4	60	108.85	270.12	4000.01	54.96	0.869
5	60	108.79	268.04	4000.09	55.09	0.869
6	60	109.76	276.64	4000	55.11	0.869
7	60	108.56	289.05	4000.01	55.37	0.869
8	60	109.71	267.1	4000.01	55.38	0.869
9	60	107.93	285.6	4000	56.04	0.867
10	60	110.32	271.41	4000.04	56.08	0.867
11	60	110.55	285.52	4000	56.66	0.866
12	60	110.65	282.4	4000.01	56.77	0.866
13	60	109.76	303.66	4004.83	57.29	0.865
14	60	110.39	296.5	4035.79	58.37	0.863

2.3. Effects of the SO2 concentration in flue gas

As it can be seen from the Figure 3desulfurization depends on the concentration of SO2 in the feed gas. By increasing the SO2 concentration it's clear that the absorbent saturates more quickly and less SO2 can be absorbed. As it is expected by increasing SO2 concentration the absorbent is saturated sooner which cause the outlet concentration of SO2 to increase quickly as shown in Figure 7

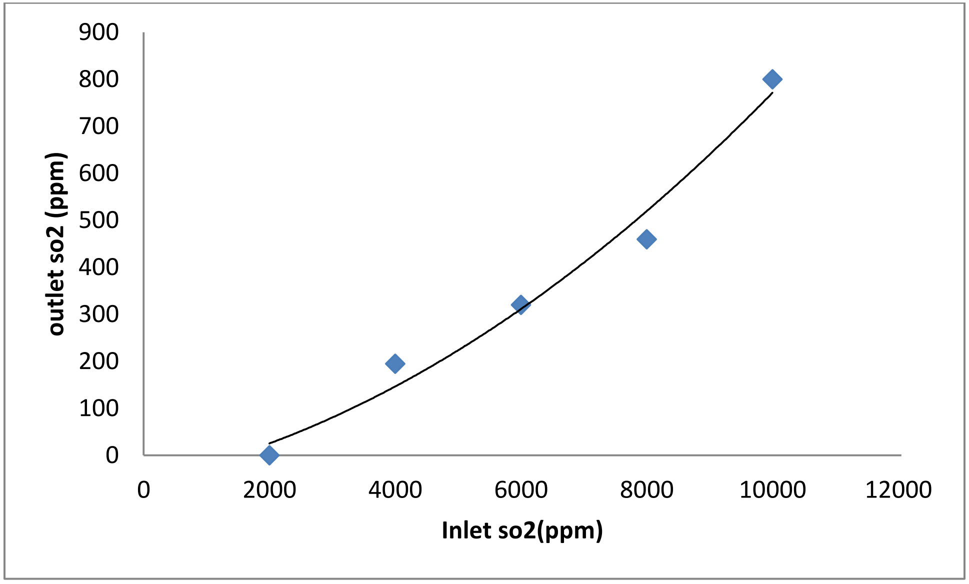


Figure 7-Effects of the SO2 concentration in flue gas

2.4. Effects of the absorber flow rate

To investigate the effect of flue gases flow rate three levels were investigated as shown in Figure 5.4. These three levels are 100 ((lit)/min),250 (lit)/min)and 400(lit)/min). It seems that at 300((lit)/min)the best performance is achieved. As can be seen from Figure 2 in the absorber flow rate change scope, SO2 absorption increased when the absorber flow rate increased. Increasing absorber flow rate is equivalent to reducing the gas-liquid two phase residence time in the absorption column so the reaction rate of SO2 absorption become faster.

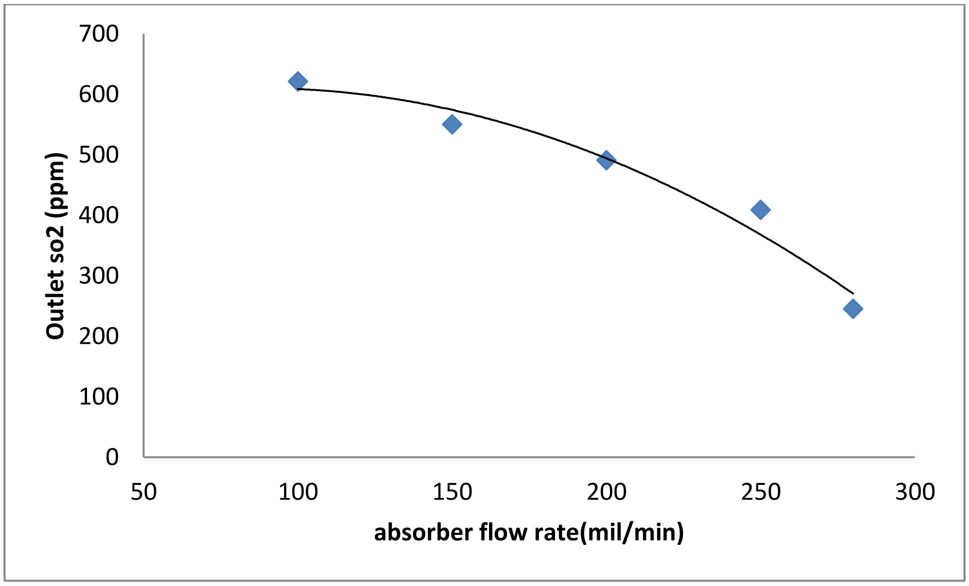


Figure 7- Effect of absorber flow rate

2.5. Effects of the desorption temperature

Figure 4 shows the effect that temperature has on absorption. The results indicate that the desulfurization efficiency will increase when the temperature is increased from 100 °C to 110°C, and will decrease when the temperature is increased from 110°C to 120°C. Because by increasing the temperature to 110 °C better absorption capacity is reached by better absorbent stripping. also by increasing temperature to 120 °C absorption highly decrease by increasing the temperature from 100 °C to 120 °C the desorption pressure increase from 0.3 bar to 0.85 bar which don't permit the absorbent to desorb the dissolved SO2. So The suitable temperature is specified as 110 °C.

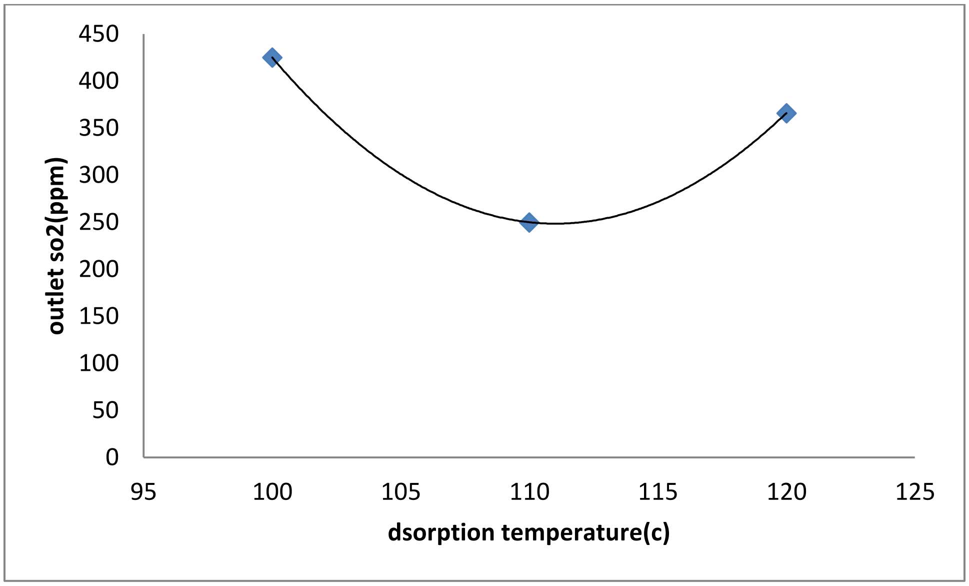
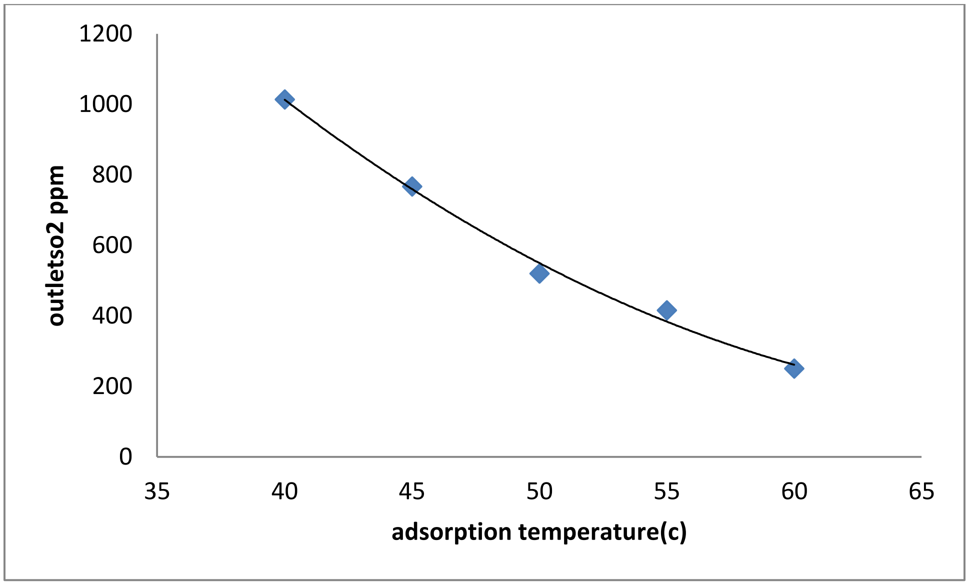


Figure 9-Effects of the desorption temperature

2.6. Effects of the adsorption temperature

Figure 1. shows the effect that temperature has on absorption. The results indicate that the desulfurization efficiency will increase when the temperature is increased from 40 °C to 60°C,. Because by increasing the temperature to 60 °C better sufficient energy is reached by better absorbent capacity.



Firure 10- Effects of the adsorption temperature

3. Conclusion:

Optimization and effect of parameters that affect SO2absorption process are analyzed using RSM method. The selective amine-based absorber was used for absorbing SO2 . Investigations were done to obtain the optimal conditions of the process that yield the least SO2 concentration in the outlet stream of the pilot. Using RSM method in this study, four three-level parameters of inlet SO2 concentration, absorber flow rate, desorption temperature and absorption temperature were considered and analyzed. Analysis of the obtained data determines the optimum conditions of SO2 absorption as: inlet SO2 4,600 ppm, adsorption temperature 60 C, desorption temperature 110C, and the absorber flow rate 3000 mL/min. The effects of varying each parameter on the final concentration of SO2 were also discussed in detail. Among the selected parameters and as illustrated in graphs, increasing SO2 inlet concentration caused decrease of absorption percentage with a linear relation. As obtained in the experiments, increment of solvent flow rate and desorption temperature result in decreasing the outlet SO2 concentration; the second of which is due to better regeneration of absorber. Varying pH amounts also revealed no special relation; it was only obtained that the least outlet SO2 is released in a pH range of 3–4. It was also illustrated that an efficiency of 100 % could be reached in the process, which is a unique achievement for the selective amine-based absorber that was used.

4. References:

[1] Akira M (1986) A method for the removal of sulfur dioxide from exhaust gas utilizing pulsed streamer corona for electron energization. IEEE Trans Ind Appl 22:516–521

[2] Akyalc L, Kaytakog ˘lu S (2010) Flue gas desulfurization by citrate process and optimization of working Parameters. Chem Eng Process 49:199–204

[3] Chris H (2007) Carbon dioxide removal from coal-fired power plants. Kluwer Academic, Dordrecht

[4] Chung WS, Tohno S, Shim SY (2009) An estimation of energy and GHG emission intensity caused by energy consumption in Korea: an energy IO approach. Appl Energy 86:1902–1914

[5] Deo PV (1988) The use of hydrogen peroxide for the control of air pollution. Chem Prot Environ 34:275–292

[6] Gleason GH, Montclair NJ, Loonam AC (1940) Recovery of sulphur dioxide. US Pat 21(106):453

[7] Goalmez A, Fueyo N, TomaA ˚Ls A (2007) Detailed modelling of a flue-gas desulfurization plant. Comput Chem Eng 31:1419–1431

[8] Hao JM, Wang SX, Lu YQ (2009) Handbook on sulfur dioxide pollution control technology in coal combustion. Chemical Industry, Beijing

[9] Hikita H, Asai S, Tsuji T (1977) Absorption of sulfur dioxide into aqueous sodium hydroxide and sodium sulfite solutions. Am Inst Chem Eng J 23:538–544

[10] Karlsson CBaHT (1997) Modeling the absorption of SO2 in a spray scrubberusingthepenetrationtheory.ChemEngSci52:3085–3099

[11] Li JK, Song HL, Geng DM (2008) Causality relationship between coal consumption and GDP: difference of major OECD and nonOECD countries. Appl Energy 85:421–429

[12]Onda K, Takeuchi H, Okumoto Y (1968) Mass transfer coefficientsPROOFbetween gas and liquid phases in packed column.Chem Eng Jpn 1:56–62

[13] Soldavini H, Von W (1991) Epuration des gaz de fume0es par le peroxyde d'hydroge'ne''. Info Chimie 334:181–183

[14] Soren K, Michael L, Kim D (1998) Experimental investigation and modeling of a wet flue gas desulfurization pilot plant. Ind Eng Chem Res 37:2792–2806

[15] Steven JS, Hugh P et al (2001) Global and regional anthropogenic sulfur dioxide emissions. Glob Planet Change 29:99–119

[16]Tang GH (1999) Sulfuric acid Beijing. Chemical Industry, Beijing (in Chinese)solutions containing hydrogen peroxide. Chem Eng Technol 26:497–502

[17] Won JC, Byoung MM, Byung HS, Jong BS, Kwang JO (2009) Characteristics of absorption/regeneration of CO2–SO2 binary systems into aqueous AMP ? ammonia solutions. J Ind Eng Chem 15:635–640

[18] Yang CL, Shaw H (1998) Aqueous absorption of NOx induced by sodium chlorite oxidation in the presence of sulfur dioxide. Environ Prog 17:80–85

[19] Yang CL, Beltran M, Kravets Z, Yamamoto T (1998) Corona-induced chemical scrubber for the control of NOx emissions. Environ Prog 17:183–189

[20]Zhang J, Wang Y, Wu D (2003) Effect investigation of ZnO additive on Mn–Fe/c-Al2O3 sorbents for hot gas desulfurization. Energy Convers Manage 44:357–367

[21] Zhu JL, Wang YH, Zhang JC, Ma RY (2005) Experimental investigation of adsorption of NO and SO2 on modified activated carbon sorbent from flue gases. Energy Convers Manag 46:2173–2184