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Optimum Operating Condition of a Multistage Gas-Solid Fluidized Bed Reactor without Downcomer for Control of Harmful Gaseous Pollutants from Flue Gas

K. Mahalik[1], G.K. Roy[2], Y.K. Mohanty[2]

Associate professor, Department of Chemical Engineering, Gandhi Institute of Engineering and Technology, Gunupur, Odisha, India[1]

Professor, Department of Chemical Engineering, Gandhi Institute of Engineering and Technology, Gunupur, Odisha, India[2]

ABSTRACT: In this paper adsorption of sulfur dioxide on lime have been undertaken in a three stage gas solid fluidized bed reactor without downcomer. The effect of various operating variables such as particle size, superficial gas velocity and static bed height on percentage removal efficiency of sulfur dioxide have been studied. Model equations for prediction of removal efficiency of sulfur dioxide have been developed and the predicted values have been compared with the experimental ones. It is observed that the predicted values are found to be agree well with the corresponding experimental counterpart with very good correlation coefficients. Further, the developed models were optimized using quadratic programming to maximize the removal efficiency of sulfur dioxide. The maximum total removal efficiency of sulfur dioxide in three stages at optimum condition is found to be 56% at normal temperature.

KEYWORDS: ANOVA, RSM, optimum parameters, Removal efficiency

I.INTRODUCTION

In developing countries like India pollution of environment has become a major problem due to emission of host of substances such as sulfur dioxide and other harmful hazardous compounds. The main source of emission of these compounds include pulverized coal-fired thermal power plants, roaster and smelter for copper, zink and lead, petroleum refinery, fluidized bed catalytic cracking unit (FCC) and sulfuric acid plants etc. The emission of sulfur dioxide has greater effect on human beings, vegetation, animals and material [1-5]. It is a well known fact that destruction of forest in northern Europe is due to acid rain which in turn is due to the emission of sulfur dioxide in to the atmosphere. Therefore the harmful effect of this pollutant demands a viable control option for sulfur dioxide.

Literature review reveals that both dry and wet methods have been practiced for the control of sulfur dioxide. These methods include oxidation, absorption, condensation and adsorption. Absorption is a well known wet method which involves dissolution of gaseous components in a absorbing liquor. A few studies on flue gas desulfurization have been reported by [6-10].

On the other hand control of sulfur dioxide by dry method includes condensation, oxidation and physical adsorption. Removal of sulfur dioxide by condensation have been reported by liptak [11]. Higher concentration of sulfur dioxide is required for removal of sulfur dioxide by the method of oxidation. But in most of the coal fired thermal power plant, sulfuric acid plants and oil refineries the concentration of sulfur dioxide is less than 1000 ppm which is too low for profitably recovery of sulfur dioxide. On the other hand a substantial amount of works have been reported for removal of SO_2 by adsorption from flue gas [12-14]. Work had also been reported on SO_2 removal based on Zeolite, silica jel, ion exchange resins, molecular sieve and styrene polymer [15]. Kato et. al.[16] studied removal of SO_2 by limestone

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particles in a single stage fluidized bed column. Recovery of SO_2 by adsorption have been studied by various researchers in a single stage fluidized bed column [17-21]

II. RELATED WORK

Critical appraisal of literature survey reveals that to achieve high removal efficiency of SO_2 use of multistage reactor seems to be attractive to meet the stringent regulation. Multistage fluidized beds are used extensively in a variety of industries because of their large capacity, low construction cost, easy operability, and high thermal efficiency. To separate and concentrate solvents such as acetone, methylene chloride, ethanol, and ethyl acetate, and to remove harmful pollutants from flue gas, continuous multistage fluidized adsorption can be applied. This is superior to the conventional fixed bed process in which the components are periodically adsorbed on to activated carbon particles and then stripped by steam [22]. In the case of drying, when the particles require similar drying times and are only surface wetted, the multi-staging of flowing solids reduces their residence time considerably and eliminates bypassing [22]. Further, some delicate pharmaceutical materials whose particles require an identical drying time can be dried using a multistage fluidizer, in which the distributors rotate on schedule to drop the particles from bed to bed [22].

Review of the relevant literature reveals a number of investigations into the use of multistage fluidizers for drying [23] and the removal of hazardous pollutants from flue gas [24-26]. Additionally, Martin-Gullon et. al.[12] studied the stable operating velocity range of a multistage fluidized bed reactor. However, in each of these cases, down-comers have been used to move the particles from bed to bed. In the case of drying, the use of down-comers is highly desirable, as the perfect mixing of solids in a single-stage fluidizer reduces the drying efficiency [27,28].This drawback can be overcome by designing a multistage fluidizer in which the flow pattern of solids changes from being well-mixed to a plug-flow [29-31]. However, for the adsorption of gaseous pollutants from flue gas, the use of a multistage fluidizer with down-comers reduces the removal efficiency of adsorption, because the same adsorbent has to pass through the down-comer in each bed.

Therefore, the present investigation aims to alleviate this reduced removal efficiency by designing a multistage fluidizer that does not use a down-comer. Our design ensures there is no flow of solids from bed to bed, and that each bed is separated by a distributor plate. The present design enhances the removal efficiency of harmful pollutants, such as sulfur oxides and nitrogen oxides, from flue gas by increasing the surface area of the adsorbent in subsequent stages. Thus, in this study, we examine the percent removal of SO_2 by using a novel adsorbent in a multistage fluidizer without down comers.

III.MATERIALS AND METHODS

The experimental setup (Fig. 1) consists of a three-stage fluidized bed column made of Perspex having same diameter and length for each stage. The bottom of the column is fixed to a Perspex flange. Air distributors made of perforated stainless steel plate, and having an 8% open area with respect to the column cross-section, are placed between the columns to distribute air uniformly through the entire cross-section of the bed. The holes on the distributor plates are laid out in a triangular pitch manner. Two pressure tapings are provided in each stage measure the pressure drop through a differential monometer, in which carbon tetrachloride (density 1.59 kg/m^3) is used as the manometric fluid. A calming section is placed just below the lower stage, and is filled with glass beads to ensure a uniform distribution of air. Two short windows, one at the bottom and one at the top, are cut into the column walls to load and unload materials to and from the column. After loading and unloading, the windows are closed by tightening a butterfly bolt.

A multistage air compressor with a capacity of 1297 kg_f/cm^2 is used to supply air to the fluidizer through a silica gel column and rotameter. The silica gel column is used to arrest moisture from the air. The calibrated rotameter (capacity 120 m^3/h) is used to measure the flow rate of air from the silica gel column.

To fluidize the entire bed material, air from the compressor is allowed to flow into the lower column through the silica gel column. The air flow rate is regulated by the calibrated rotameter. SO_2 gas from the SO_2 cylinder is introduced between silica jel column and calming section which mixes with the compressed air and moves up in the beds. Here compressed air is used as a motive fluid which carries SO_2 gas up in the beds and helps in fluidizing the beds .Hydrated

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lime of different particle sizes are taken as bed material. Experiments are conducted by varying superficial gas velocity, particle size and static bed height.

Samples at the inlet of the column and outlet of each stage were drawn. The SO_2 gas samples were collected at each point with the help of midget impinger and aspirator bottles. The gas samples were analyzed for sulfur dioxide by the "Tetrachloro-Mercurate method" [IS: 5182(Part-VI].Then by comparing the inlet and outlet concentration of SO_2 in each bed the percent removal of SO_2 were calculated.

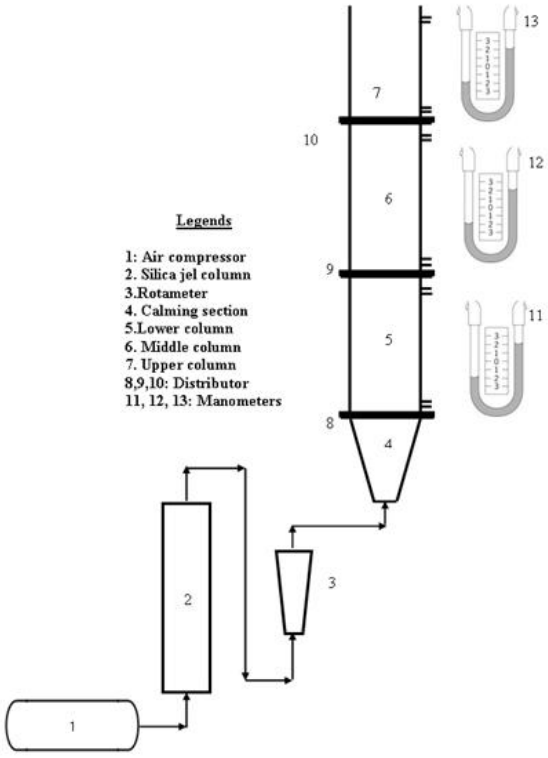

Fig. 1. Schematic diagram of experimetal set-up

IV. RESULTS AND DISCUSSIONS

Development of models: The statistical software package "Design Expert" has been used for regression analysis of experimental data and to draw response surface plot. ANOVA has been used to estimate the statistical parameters. The complete experimental range and level of variables are given in the Table 2 and Table 3 which shows the design of experiments together with the experimental results. As suggested by the software, the quadratic model has been selected which was not aliased. The final empirical model in terms of coded factor for percentage removal of SO_2 (Y) is shown in Eqs.1- 3.

$$Y_1=9.89-0.71A+2.22B+0.033C-0.19AB-0.062AC+0.94BC+0.043A^2-0.22B^2+0.021C^2 \quad (1)$$

$$Y_{2=}13.86-1.01A+2.94B-0.20C-0.14AB+0.012AC+0.99BC+0.018A^2-0.16B^2-0.19C^2 \quad (2)$$

$$Y_{3=}15.84-1.11A+3.38B-0.18C-0.22AB+0.00054AC+0.95BC+0.13A^2-0.25B^2-0.20C^2 \quad (3)$$

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Table 1

Actual value and corresponding coded value of the variables

Code	Actual level of variable
$-\alpha$	A_{min}
-1	$[(A_{max}+A_{min})/2]-[(A_{max}-A_{min})/2\beta]$
0	$(A_{max}+A_{min})/2$
+1	$[(A_{max}+A_{min})/2]+[(A_{max}-A_{min})/2\beta]$
$+\alpha$	A_{max}

Where A_{max} and A_{min} are maximum and minimum values of A, respectively,β is $2^n/4$.

Table 2
Level of independent variable

Response	Variables	Sumbol	$-\alpha$	-1	0	+1	$+\alpha$
% removal	Particle size, mm	A	0.55	0.89	1.23	1.56	1.9
Of sulfur dioxide	Static bed height, cm	B	3	4.75	6.5	8.25	10
	Superficial gas velocity, m/s	C	0.2386	0.6	1.13	1.66	2.19

The model F values of 72.895, 23.688, 30.162 of lower, middle and upper bed respectively implies that the models are significant. Further Values of "Prob > F" less than 0.0500 indicate that the model terms are significant. In this case A, B, BC and C^2 are significant model terms for all the three beds. Adequacy Precision measures the signal to noise ratio and it compares the range of the predicted values at the design points to the average prediction error. A ratio greater than 4 is desirable. The ratios of 11.847, 16.548 and 18.325 in lower, middle and upper bed respectively in our study, indicates an adequate signal. Hence this model can be used to navigate the design space. One of the most important parts of the data analysis is to check the adequacy of the developed model. This is done by examining the residual plots which have been shown in the Figs. 2 and 3. It is observed from the Fig. 2 that there was neither response transformation nor any apparent problem with normality. Fig.3 shows the internally studentised residual vs predicted

Table 3
Experimental design matrix and response of % removal sulpher dioxide

	Codded level varable			Response(% removal of Sulpher dioxide		
Run	A(mm)	B(cm	C(m/	Y1	Y2	Y3
1	0.82	4.42	0.6	8.5	11.9	13.6
2	1.63	4.42	0.6	7.5	10.5	12
3	0.82	8.58	0.6	11.5	16.1	18.4
4	1.63	8.58	0.6	10	14	16
5	0.82	4.42	1.66	7	10	11.8
6	1.63	4.42	1.66	6	8.5	10.3
7	0.82	8.58	1.66	14	18	20.5
8	1.63	8.58	1.66	12	16	18
9	0.543	6.5	1.13	11.5	17	18.95
10	1.906	6.5	1.13	9	13	14.7
11	1.225	3.001	1.13	6	8.5	9.5
12	1.225	9.998	1.13	13	19.7	22
13	1.225	6.5	0.238	10.5	14.7	16.8
14	1.225	6.5	2.021	9.875	13.825	15.8
15	1.225	6.5	1.13	9.875	13.825	15.8
16	1.225	6.5	1.13	9.875	13.825	15.8
17	1.225	6.5	1.13	9.875	13.825	15.8
18	1.225	6.5	1.13	9.875	13.825	15.8
19	1.225	6.5	1.13	9.875	13.825	15.8
20	1.225	6.5	1.13	9.875	13.825	15.8

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percentage of removal of SO_2 of all the three stages which represents a random scatter plot indicating that the variance of original observation is constant for all values of the response. This is also an indication that there was no need for transformation of response variables. However, this plot would exhibit a funnel shaped pattern if the variance of the response depended on the mean level of response [39].

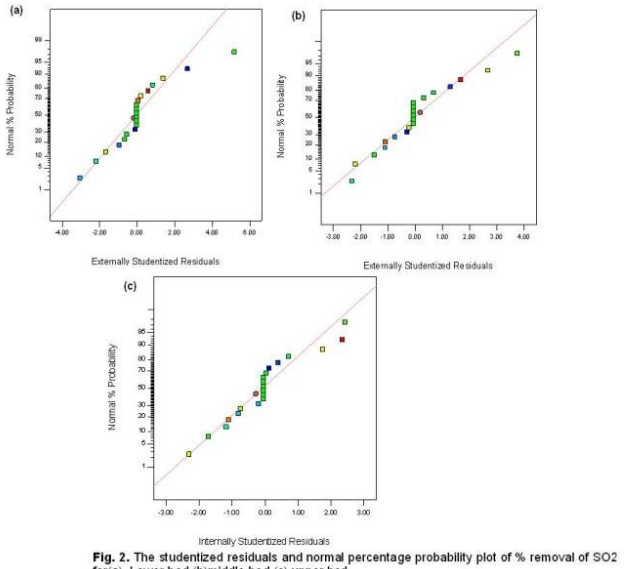

Fig. 2. The studentized residuals and normal percentage probability plot of % removal of SO2 for(a) Lower bed (b)middle bed (c) upper bed

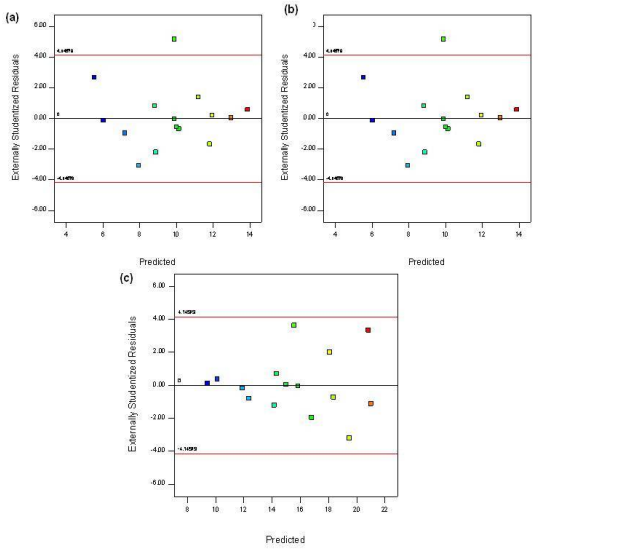

Fig. 3. The predicted % removal of SO_2 and studentized residuals plot for(a) Lower bed (b)middle bed (c) upper bed

The actual and the predicted percentage removal of SO_2 of three beds have been shown in Fig. 4 and it is observed from Tables 4-6 that the values of R^2 and R^2_{adj} and R^2_{pred} have been found to be 98%, 97% and 88% respectively for lower bed, 95%, 91% and 82% for middle bed and 96%, 93% and 86% for upper bed. The fair correlation coefficients 82% 86% and 88% might be due to the insignificant terms in the models as well as the non-linear influence of the investigated variables on the response.

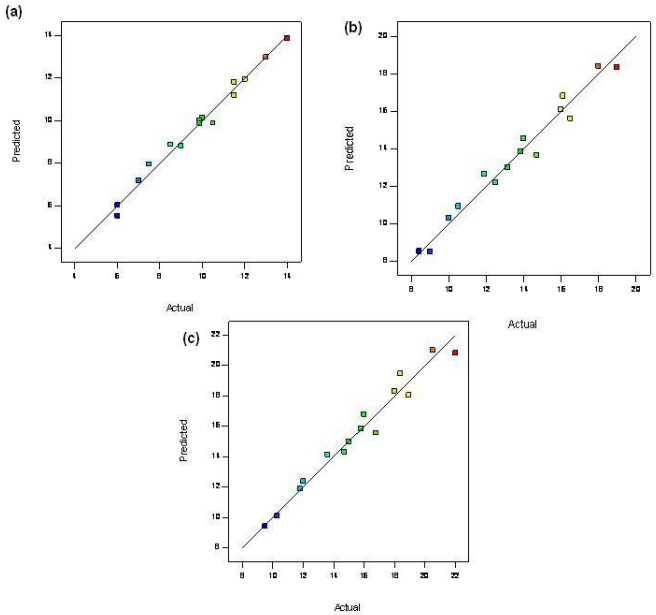

Fig.4. Comparison of actual and predicted values of % removal of SO_2 for (a) Lower bed (b)middle bed (c) upper bed

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Combined effect of system parameters on percentage removal of SO_2 : Both individual and combined effect of operating variable such as particle size, static bed height and superficial gas velocity on percentage removal of SO_2 of the three stage fluidized bed have been studied using RSM based CCD. Fig.5 shows the combined effect of each of two variables on percentage removal of SO_2 at a constant value of the third variables for the lower bed. The combined effect of particle size and static bed height on percentage removal of SO_2 at superficial gas velocity 1.13m/s is shown in

Table 4
Analysis of Variance(ANOVA) for lower bed % removal efficiency for eq.(5)

Source	Sum of squares	degrees of freedom	Mean square	F value	p-val Prob > F	Remark
Model	82.150	9.000	9.128	72.895	0.000	significant
A	6.896	1.000	6.896	55.071	0.000	significant
B	67.104	1.000	67.104	535.893	0.000	significant
C	0.015	1.000	0.015	0.118	0.739	
AB	0.281	1.000	0.281	2.246	0.165	
AC	0.031	1.000	0.031	0.250	0.628	
BC	7.031	1.000	7.031	56.152	0.000	significant
A^2	0.027	1.000	0.027	0.216	0.652	
B^2	0.709	1.000	0.709	5.664	0.021	significant
C^2	0.006	1.000	0.006	0.052	0.824	
Residual	1.252	10.000	0.125			
Lack of Fit	1.252	5.000	0.250			
Pure Error	0.000	5.000	0.000			
Cor Total	83.402	19.000				
R^2						98
R_{adj}^2						97
R_{pred}^2						88

Table 5
Analysis of Variance(ANOVA) for middle bed % removal efficiency for Eq.(6)

Source	Sum of squares	Degrees of freedom	Mean squar	F value	p-value Prob > F	Remark
Model	152.207	9.000	16.912	23.688	0.000	significant
A	13.798	1.000	13.798	19.326	0.001	significant
B	129.388	1.000	129.388	181.229	0.000	significant
C	0.159	1.000	0.159	0.222	0.648	
AB	0.180	1.000	0.180	0.252	0.626	
AC	0.000	1.000	0.000	0.000	1.000	
BC	7.605	1.000	7.605	10.652	0.009	significant
A^2	0.232	1.000	0.232	0.325	0.581	
B^2	0.527	1.000	0.527	0.739	0.031	significant
C^2	0.258	1.000	0.258	0.362	0.561	
Residual	7.139	10.000	0.714			
Lack of Fit	7.139	5.000	1.428			
Pure Error	0.000	5.000	0.000			
Cor Total	159.347	19.000				
R^2						95
R_{adj}^2						91
R_{pred}^2						84

Table 6
Analysis of Variance(ANOVA) for upper bed % removal efficiency for Eq.(7)

Source	Sum of squares	degrees of freedom	Mean squares	F value	p-value Prob > F	Remark
Model	182.397	9.000	20.266	30.162	0.000	significant
A	16.801	1.000	16.801	25.004	0.001	significant
B	156.442	1.000	156.442	232.827	0.000	significant
C	0.086	1.000	0.086	0.128	0.728	
AB	0.405	1.000	0.405	0.603	0.456	
AC	0.000	1.000	0.000	0.000	1.000	
BC	7.220	1.000	7.220	10.745	0.008	significant
A^2	0.142	1.000	0.142	0.212	0.655	
B^2	1.136	1.000	1.136	1.690	0.023	significant
C^2	0.107	1.000	0.107	0.160	0.698	
Residual	6.719	10.000	0.672			
Lack of Fit	6.719	5.000	1.344			
Pure Error	0.000	5.000	0.000			
Cor Total	189.116	19.000				
R^2						96
R_{adj}^2						93
R_{pred}^2						86

Fig.5(a).It is observed that with increase in particle size from 0.82mm to 1.63mm the percentage removal of SO_2 decreases from 13% to 10%.This is due to the fact that smaller particle has more surface area than that of larger particle and hence the adsorption on smaller particle is more. It is also observed from the same figure that the percentage removal of SO_2 increases from 7% to 13% with an increases of static bed height from 4.42cm to 8.58cm.This is due to increased surface area of adsorbent which in turn is due to more adsorbent in the bed leading to more adsorption and hence more percentage removal of SO_2. Additionally increased static bed height in the bed

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increases the force of gravity [40] which increases the mean residence time of the gas in the bed for which the removal

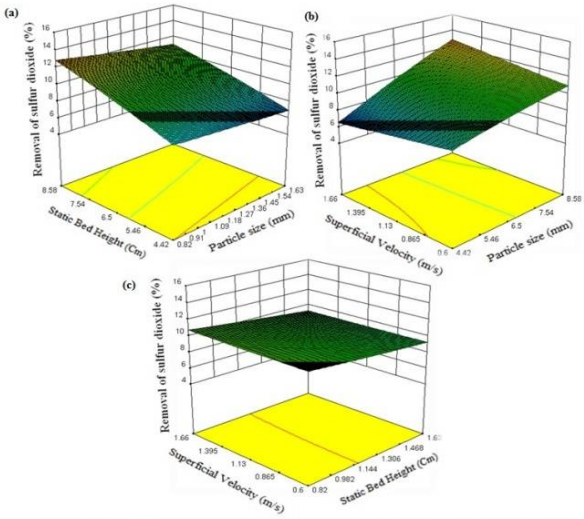

Fig. 5. Combined effects of (a) static bed height and particle size on lower bed % removal of $SO_2$for for superficial gas velocity of 1.13m/s (b) superficial gas velocity and particle size on lower bed % removal of $SO_2$for static bed height of 6.5 cm, (c) static bed height superficial gas velocity on lower bed % removal of $SO_2$for particle size of 1.25 mm.

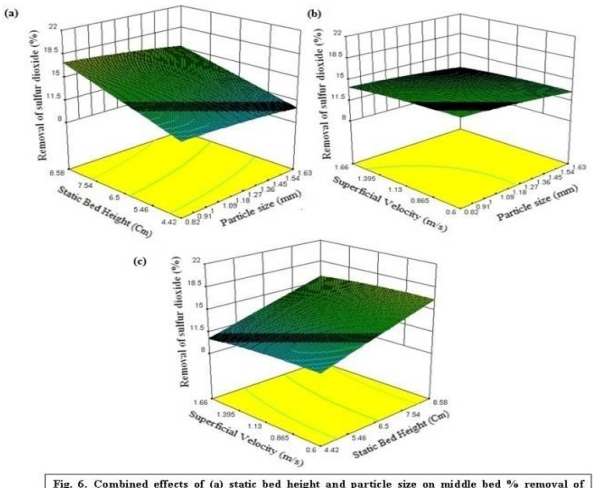

Fig. 6. Combined effects of (a) static bed height and particle size on middle bed % removal of $SO_2$for for superficial gas velocity of 1.13m/s (b) superficial gas velocity and particle size on middle bed % removal of $SO_2$for static bed height of 6.5 cm, (c) static bed height superficial gas velocity on middle bed % removal of $SO_2$for particle size of 1.25 mm

efficiency increases with an increase of static bed height. Similarly Fig. 5(b) shows the combined effect of superficial gas velocity and particle size at a constant value of static bed height of 6.5 cm.

It is seen from the figure that percentage removal of SO_2 goes on decreasing from 10% to 7% while the superficial velocity increases from 0.6 m/s to1.66m/s for the same lower bed. Increased superficial gas velocity reduces the mean residence time of the gas inside the bed for which the gas leaves the bed promptly without having much contact with the adsorbent, thus leading to decreased removal efficiency. The combined effect of superficial gas velocity and static bed height at particle size of 1.225mm is shown in the Fig.5(c).

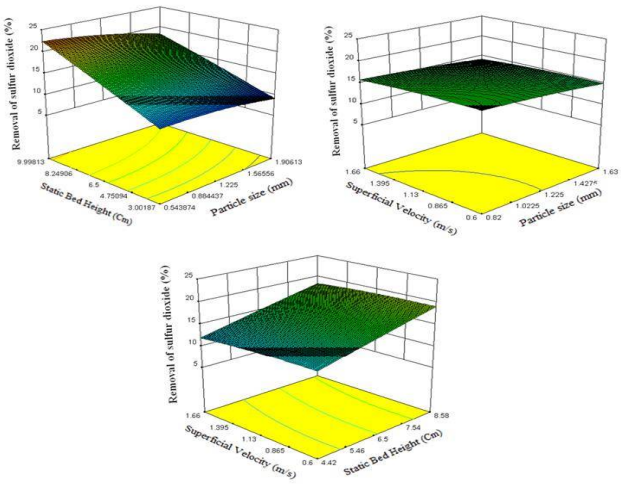

Fig. 7. Combined effects of (a) static bed height and particle size on upper bed % removal of $SO_2$for for superficial gas velocity of 1.13m/s (b) superficial gas velocity and particle size on upper bed % removal of $SO_2$for static bed height of 6.5 cm, (c) static bed height superficial gas velocity on upper bed % removal of $SO_2$for particle size of 1.25 mm

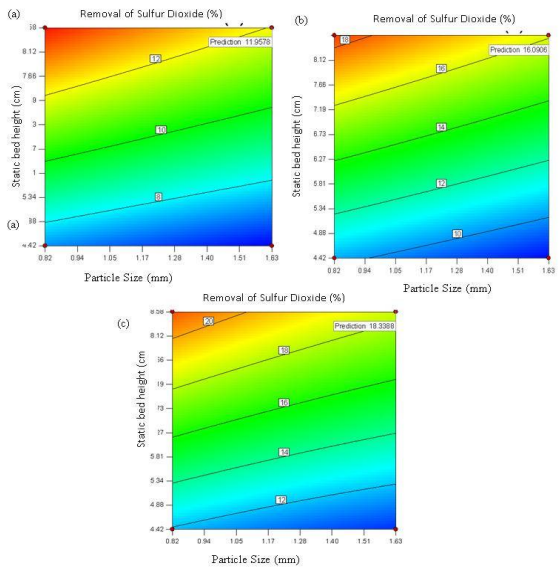

Fig 8. Optimum region for SO2 removal on the particle size–static bed height–removal (%) response surface plot for(a)lower bed(b)middle bed(c)Upper bed

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Similarly for middle bed and upper bed, the combined effect of operating variables on percentage removal is shown in Fig.6 and Fig.7 respectively. A similar trend as discussed above have been observed between the percentage removal and the operating variables. A careful observation of Figs. 5-7 reveals that the removal efficiency is highest in the upper bed with lowest and intermediate in lower and middle bed respectively. Under identical condition the percentage removal of SO_2 is 13%,17% and 23% in lower, middle and upper bed respectively(Fig. 5(a), Fig. 6(a) and Fig.7(a)).This increase in removal efficiency from lower to upper bed is due to the fact that the mean residence time of the gas in the bed increases as the gas moves from lower to upper bed through the middle one. In addition to this the flow pattern of adsorbents changes from well mixed in the lower bed to plug flow pattern in upper bed with an intermediate mixing in the middle bed [41]. As the perfect mixing of solids reduces the removal efficiency, it is least in the lower bed and highest in the upper bed.

Optimization of removal efficiency of SO_2 : The determination of the optimum operating variables for maximizing the percentage of removal of SO_2 was one of the vital parts of this experimental study. The developed quadratic model equation was optimized using a quadratic programming to maximize the removal efficiency. The optimum region for the removal of SO_2 for the three beds are shown in Fig.8. Table 7 shows the optimum values of percentage removal of

Table 7

Optimum condition and model validation

Particle size, A(mm)	Static bed height, B (cm)	Superficial gas velocity, C (m/s)	Removal efficiency, Y (%)		
			Lower bed	Middle bed	Upper bed
1.51	8.12	1.6	11.95	16.09	18.388

SO_2 corresponding to the optimum operating variables. It is found that total percentage removal of SO_2 in three stages at normal temperature is 56% corresponding to the particle size of 1.51mm, static bed height of 8.12cm and superficial gas velocity of 1.6m/s.

V.CONCLUSIONS

In this paper, we examined the adsorption of SO_2 on limestone particles in a multistage fluidized bed column without downcomer facility. The parameters affecting the percentage removal efficiency are found to be the particle size, superficial gas velocity, and static bed height. It has been observed that both particle size of the adsorbent and superficial gas velocity has an inverse effect on percentage removal of SO_2, where as the static bed height has a direct effect. Further the removal efficiency of different stages of multistage fluidized bed column was compared and was concluded that the removal efficiency of SO_2 is highest in upper bed and least in lower bed with intermediate one in the middle bed. An increase in SO_2 removal efficiency is found to result from the increase of mean residence time of gas in the reactor It has been found that the maximum removal efficiency was 56% at normal temperature. The models developed in this study for the prediction of percentage removal of SO_2 can be effectively used within the design space described in this study The models exhibits a very good correlation coefficient, and its output is within the 95% confidence level. The result of this study can be effectively used for the removal of harmful gaseous pollutants such as SO_x and NO_x from flue gas.

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