ATIPC – 2020 Special Issue

J. Indian Chem. Soc.,
Vol. 97, No. 12b, December 2020, pp. 2783-2790

Optimization of process variables for particulate laden H_2S removal from gasifier syngas in a multistage dual-flow sieve plate scrubber

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Manuscript received online 08 December 2020, accepted 28 December 2020

This study describes the optimization of process parameters – flow rates of gas and liquid, inlet particulate loading and inlet H_2S loading on particulate laden H_2S removal efficiency in a dual-flow sieve plate scrubber by using response surface methodology. The process parameters for particulate laden H_2S removal in the scrubber were optimized to maximize the H_2S removal efficiency using response surface methodology and in the operations range, the optimum conditions to get 93.1% H_2S removal are 25×10^{-3} kg/Nm^3, 47.9×10^{-6} m^3/s liquid flow rate, 27.3×10^{-4} Nm^3/s gas flow rate and 383 ppm inlet H_2S loading and the experiments were conducted at same conditions and achieved 92.8% H_2S removal.

Keywords: Wet scrubber, pollution, fly ash, RSM, flow rate.

Introduction

Gasification process produces gaseous pollutants hydrogen sulphide and hydrogen chloride along with particulate matter which affect the energy production equipment and environment synergistically. The studies on the simultaneous removal of particulate matter and gaseous pollutants by various wet scrubbers revealed that the removal efficiency range of gaseous pollutants increases in presence of particulate matter. Dual-flow sieve plate columns are effective in removing the acid gases and particulate matter. Therefore, it is very much required to optimize the process variables for particulate laden H_2S removal efficiency of the multi-stage dual-flow sieve plate column. The optimization can be achieved by single parameter change and maintaining the other variables at unchanged conditions. It is laborious and cannot address the process variables effect completely. It also neglects the variable-variable combination effects. Whereas the response surface methodology (RSM) establishes the experiment parameters relationship with the results of experiments[1]. The RSM analyses the independent variables combined and individual effects[2].

The major objective of RSM is the determination of optimum operating conditions or the zone which gives the satisfied operating specifications[3]. In general, RSM is the union of techniques (mathematical and statistical) to optimize the processes along with the development and improvement. RSM can estimate the importance of many factors that affect the response individually and in combination. The RSM design will have generally four major steps[4]. They are experiments series design to response of interest measurement, development of mathematical model, estimating the experimental parameters optimal set which gives a response value of minimum/maximum and represents synergetic or individual effects of process parameters through plots of two dimensional (2D) and 3D. The RSM is being used extensively for optimization of processes in many areas of scientific research[5–8]. The quantitative data from the experiments will be used in RSM statistically for determination of model and conditions for optimum operation[9]. The central composite design (CCD) of RSM design can be used to observe the variables effect on response[10–12]. Thus, an approach towards vital parameters of process optimization for particulate laden

H_2S removal efficiency in the domain of design of experiment (DOE) has been carried out.

The objective is to model and get the optimum conditions to achieve maximum particulate laden H_2S removal efficiency by simultaneously considering the process parameters. To observe the effect of different variables on particulate laden H_2S removal efficiency, the Central composite design (CCD) was employed. The particulate matter inlet concentration, H_2S inlet concentration, liquid flow rate and gas flow rate were taken as parameters for particulate laden H_2S removal efficiency. Development of a quadratic model for particulate laden H_2S removal efficiency was done by using the Design-Expert software and the optimum conditions of operation to get maximum particulate laden H_2S removal efficiency have been calculated by the model.

Materials and methods

Multivariate experimental design:

The CCD is largely applied method for fitting a second order model and the model development can be achieved with the experiments in lesser number. The knowledge of detailed mechanism is not required for the development of model in this method since the model is an empirical one. It consists of n_c center, 2n axial and 2^n factorial runs. These designs will have n_c center points (0, 0, 0, . . .,0), 2^y factorial/ fractional (usually coded to notation of ±1) and 2y axial points augmented $(\pm\alpha, 0, 0 \ldots, 0), (0, \pm\alpha, 0, \ldots, 0), \ldots, (0, 0, \ldots, \pm\alpha)$[13]. The increase in factors (n) enhances the number of runs for a total design copy and the estimation of important interactions can be understood by running minimum experiments. The 2^y factorial designs cannot estimate individual second-order effects separately. The estimation of statistical parameters with respect to methods of response surface was done by analysis of variance (ANOVA) along with modeling and optimization of parameters and the responses. The process of optimizing the parameters follows three important steps in which the first step is carrying out the experiments that are designed statistically and the second one is coefficients estimation in a mathematical model. The prediction of the response and the model adequacy check is the third step. The response is the function of independent variables as given in eq. (1).

$$Y = f(X_1, X_2, X_3, X_4 \ldots \ldots X_n) \tag{1}$$

where, Y is the system response and X_i is the action variables and called as factors. The objective is the optimization of response (Y). The assumption of the method is that the action variables are continuous and they are experimentally controlled with negligible errors. The major requirement is that the finding of appropriate functional relation between the response surface and independent variables[4]. The effects of factors that are not controlled can be diminished by using random experimental order. An empirical model can be developed by using response by correlating the responses to maximize the particulate laden H_2S removal process parameters using a second-degree polynomial equation as given by eq. (2):

$$Y = b'_0 \sum_{i=1}^{n} b_i X_i + \sum_{i=1}^{n} b_{ii} X_i^2 + \sum_{i=1}^{n} \sum_{j>1}^{n} b_{ij} X_i X_j \tag{2}$$

where b'_0, b_i, b_{ij} and b_{ii} are constant, linear, interaction and quadratic coefficients respectively and X_i and X_j are independent variables' coded values. The required tests number for CCD to generate the quadratic terms comprise of 2^y factorial (origin at the center), points fixed axially at a distance a from the center are 2y. The rotatability can be allowed by choosing the axial points and model prediction variance is constant at the points that are equally distant from the center of the design. The experimental error can be estimated by the test replicates at the center and six tests are at the center for four parameters. Total number of tests required for this method can be estimated by eq. (3) and for four parameters it can be calculated as thirty[14].

$$N = 2^y + 2y + y_c \tag{3}$$

After defining the desired variables ranges, then they are coded to lie at ±1 for the factorial points, 0 for the center points and $\pm\alpha$ for the axial points. The calculation of codes as the interest range function is shown in Table 1[14].

Table 1. Relationship between coded and actual values of the variables

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

where X_{max} and X_{min} are maximum and minimum values of X respectively and $\beta = 2^{n/4}$.

Experimental

The schematic diagram of the multistage dual-flow sieve plate column wet scrubber for simultaneous removal of particulate laden H_2S is shown in Fig. 1. The experimental air was supplied by an air compressor. The air flows through an air rotameter which is connected to the inlet of the column

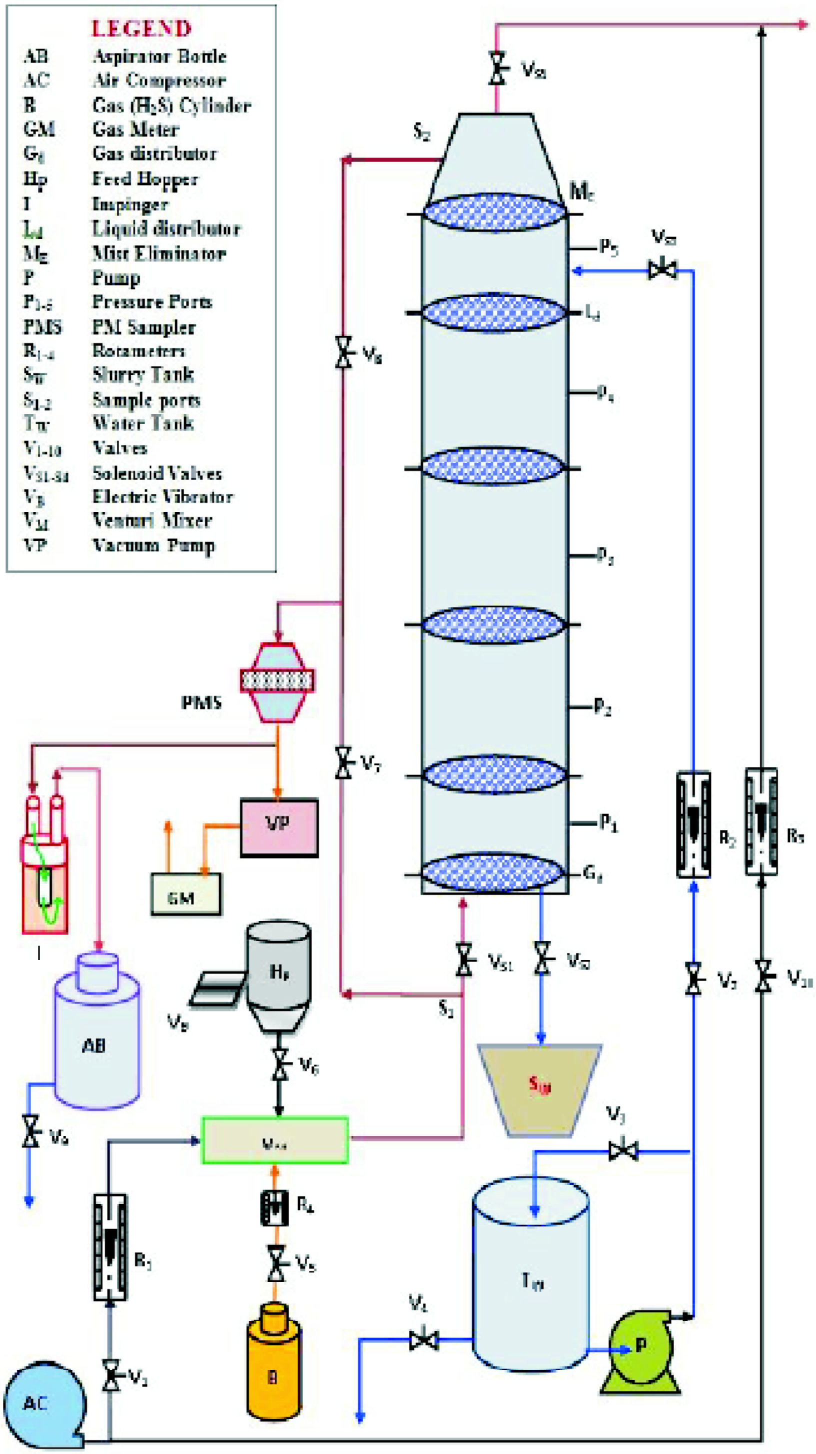

Fig. 1. Schematic diagram of multistage dual-flow sieve plate column scrubber.

through a venturi mixer to which the particulate matter and H_2S gas were added from a steel hopper and a H_2S cylinder. The fly ash was used as particulate matter. The particulate matter was fed to the venturi mixer from hopper at controlled rates and introduced to the experimental air by regulating the intensity of vibration by the variable rheostat connected to the power line of the vibrator. The H_2S concentrations were controlled by a rotameter (R_4) and the H_2S inlet concentrations of were estimated with the flow rates of experimental air and H_2S. The air mixes thoroughly in the venturi mixer with particulates and H_2S. The thoroughly mixed particulates-H_2S-air then leaves the mixer to enter into the column from the inlet provided at the scrubber bottom. The liquid for the scrubbing enters the system from the top of the column at controlled flow rates using rotameter (R_2) and the uniform liquid flow is facilitated by a liquid distributor.

The sampling was done simultaneously using provisions (S_1 and S_2) by passing sampling air first through the particulate sampling assembly (PMS) and then through the impinger (I). Indian Standard methods IS 5182: Part-IV (1973) and IS 5182: Part-VII (1973) were used for sampling of PM laden H_2S[15]. The percentage removal of H_2S in presence of particulate matter have been calculated for each run.

Results and discussion

The regression analysis of data from experiments was done by statistical software package Design-Expert 7® (trial version). The particulate concentrations (15×10^{-3} to 25×10^{-3} kg/Nm^3), H_2S concentrations (200–400 ppm), gas flow rate (16.59×10^{-4} to 27.65×10^{-4} Nm^3/s) and liquid flow rate (20.649×10^{-6} to 48.183×10^{-6} m^3/s) are used as variables for to model and optimize for particulate laden H_2S removal efficiency by RSM.

Regression model development:

The correlation between the particulate laden H_2S removal efficiency and the operating variables is developed by using CCD. The experiments design and the results from the experiments for particulate laden H_2S removal is given in Table 2. The error of the experiments was found by the center point runs (15–20). The selection of models was based on the polynomials of highest order with the significant additional terms and the not aliased models as per the sequential model sum of squares. To get a quadratic model with 24 trials and a star configuration ($\alpha = \pm2$), the experiments were planned. Regression analysis was done to fit the response function of particulate laden H_2S removal. The expression of model given in eq. (4) (with the coded values of variables) represents particulate laden H_2S removal efficiency (Y) as a

Table 2. Design matrix of experiments with results for $\eta_{H_2S\text{-}PM}$

Run	Variables coded level				Variables actual level				PM laden
	X_1	X_2	X_3	X_4	$X_1\times10^4$ (Nm^3/s)	$X_2\times10^6$ (m^3/s)	X_3 (ppm)	$X_4\times10^3$ (kg/Nm^3)	H_2S removal efficiency
1	–1	–1	–1	–1	16.59	20.64	200	15	0.2085
2	+1	–1	–1	–1	27.65	20.64	200	15	0.4677
3	–1	+1	–1	–1	16.59	48.18	200	15	0.5084
4	+1	+1	–1	–1	27.65	48.18	200	15	0.7787
5	–1	–1	+1	–1	16.59	20.64	400	15	0.2901
6	+1	–1	+1	–1	27.65	20.64	400	15	0.5984
7	–1	+1	+1	–1	16.59	48.18	400	15	0.6208
8	+1	+1	+1	–1	27.65	48.18	400	15	0.8449
9	–1	-1	–1	+1	16.59	20.64	200	25	0.2718
10	+1	-1	–1	+1	27.65	20.64	200	25	0.501
11	+1	+1	–1	+1	16.59	48.18	200	25	0.5821
12	+1	+1	–1	+1	27.65	48.18	200	25	0.8342
13	–1	–1	+1	+1	16.59	20.64	400	25	0.4244
14	+1	–1	+1	+1	27.65	20.64	400	25	0.6973
15	–1	+1	+1	+1	16.59	48.18	400	25	0.7108
16	+1	+1	+1	+1	27.65	48.18	400	25	0.9051

Table-2 (contd.)

17	$-\alpha$	0	0	0	11.06	34.41	300	20	0.3598
18	$+\alpha$	0	0	0	33.18	34.41	300	20	0.7858
19	0	$-\alpha$	0	0	22.12	6.87	300	20	0.1135
20	0	$+\alpha$	0	0	22.12	61.95	300	20	0.8832
21	0	0	$-\alpha$	0	22.12	34.41	100	20	0.4271
22	0	0	$+\alpha$	0	22.12	34.41	500	20	0.7252
23	0	0	0	$-\alpha$	22.12	34.41	300	10	0.5332
24	0	0	0	$+\alpha$	22.12	34.41	300	30	0.8035
25	0	0	0	0	22.12	34.41	300	20	0.5732
26	0	0	0	0	22.12	34.41	300	20	0.5772
27	0	0	0	0	22.12	34.41	300	20	0.5795
28	0	0	0	0	22.12	34.41	300	20	0.5782
29	0	0	0	0	22.12	34.41	300	20	0.5752
30	0	0	0	0	22.12	34.41	300	20	0.5762

function of gas flow rate (X_1), liquid flow rate (X_2), H_2S concentration (X_3) and particulate loading (X_4).

$$Y = 0.58 + 0.12X_1 + 0.16X_2 + 0.064X_3 + 0.048X_4 - 0.0085X_1X_2 - 0.00070X_1X_3 - 0.0071X_1X_4 - 0.0114X_2X_3 - 0.0031X_2X_4 + 0.0099X_3X_4 - 0.0011X_1X_1 - 0.02X_2X_2 - 0.00030X_3X_3 + 0.023X_4X_4 \quad (4)$$

Particulate laden H_2S removal efficiency:

From the ANOVA results, flow rates of gas and liquid, PM concentration and concentration of H_2S were found to have significant effects on the particulate laden H_2S removal efficiency. The gas flow rate imposed the greatest effect on the response whereas the least effect was imposed by H_2S concentration. The 3D response surfaces constructed to show the effect of gas flow rate and particulate concentration on

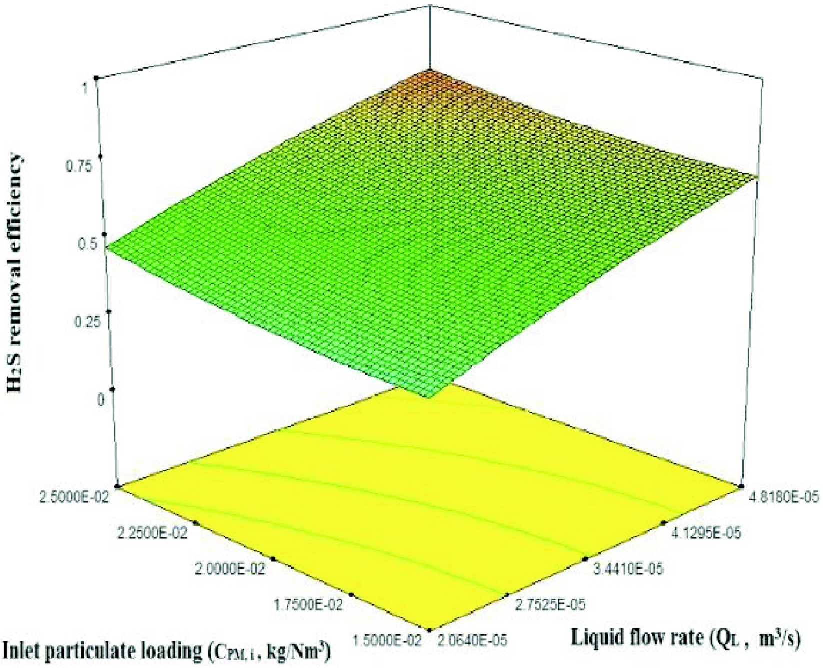

Fig. 2. Effect of $C_{PM,i}$ and Q_G on $\eta_{H_2S\text{-}PM}$ at $C_{H_2S,i}$ = 300 ppm and $Q_L = 48.183\times10^{-6}$ m^3/s.

particulate laden H_2S removal efficiency at liquid flow rate 48.183×10^{-6} m^3/s and 300 ppm H_2S concentration can be seen in Fig. 2. It can be seen from the Fig. 3 that 3D response surfaces which are developed to observe the effect of particulate concentration and inlet H_2S concentration on particulate laden H_2S removal efficiency at gas flow rate 27.653×10^{-4} Nm^3/s and 48.183×10^{-6} m^3/s liquid flow rate.

The objective of the work is to get the optimum values of the process parameters to maximize the particulate laden H_2S removal efficiency by using RSM.

The gas flow rate of 27.3×10^{-4} Nm^3/s, liquid flow rate of 47.9×10^{-6} m^3/s, inlet H_2S concentration of 383 ppm and 25×10^{-3} kg/Nm^3 particulate concentration are observed as optimum values of parameters to get maximum particulate laden H_2S removal efficiency. The maximum particulate laden H_2S removal efficiency of 93.1% was predicted from the model at optimum conditions of the parameters whereas 92.8% maximum particulate laden H_2S removal efficiency was obtained from the experiments. The contour plot for optimization is given in Fig. 4.

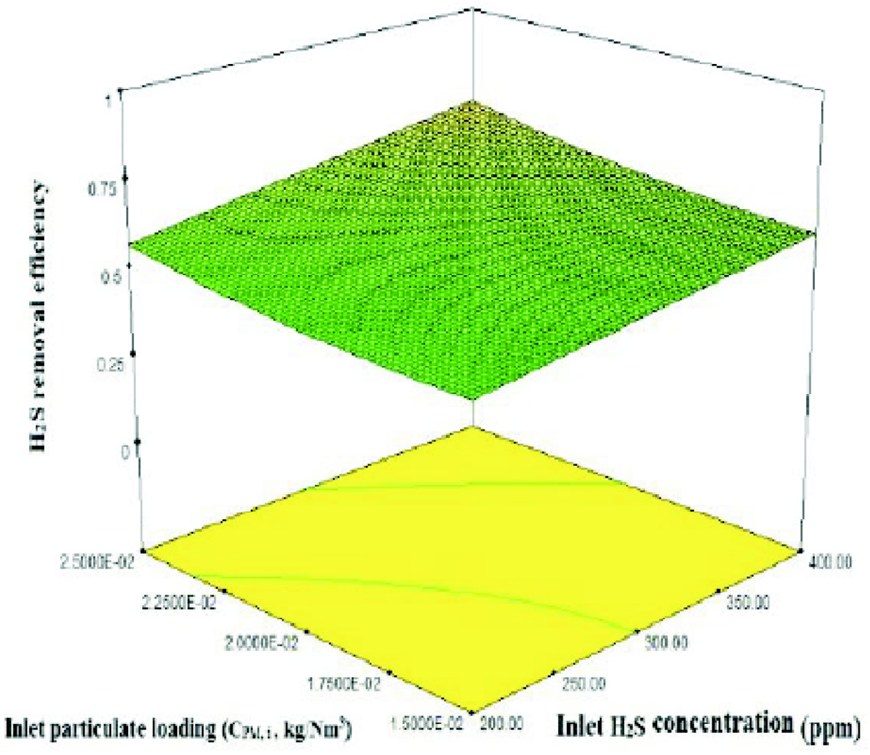

Fig. 3. Effect of $C_{PM,i}$ and $C_{H_2S,i}$ on $\eta_{H_2S\text{-}PM}$ at $Q_G = 27.65\times10^{-4}$ Nm^3/s and $Q_L = 48.183\times10^{-6}$ m^3/s.

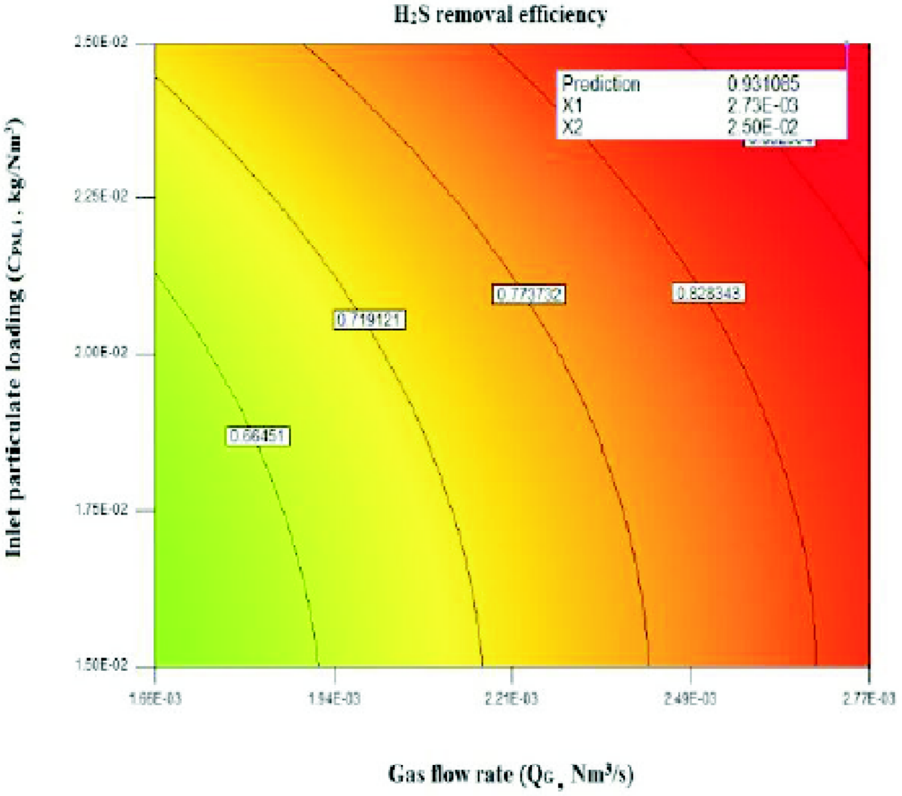

Fig. 4. Contour plot for optimum level of parameters.

Conclusions

The optimization of process parameters - flow rates of gas and liquid, liquid flow rate, inlet particulate loading and inlet H_2S loading on particulate laden H_2S removal efficiency was done by using response surface methodology. The effects of the process variables on particulate laden H_2S removal were described using three dimensional plots of response surface developed from simulations. These parameters were determined to be the most critical operating parameters as they enhance the removal efficiency. The process parameters for particulate laden H_2S removal were optimized to maximize H_2S removal efficiency using response surface methodology and the optimum conditions to get 93.1% H_2S removal are 25×10^{-3} kg/Nm^3, 47.9×10^{-6} m^3/s liquid rate, 27.3×10^{-4} Nm^3/s gas rate and 383 ppm inlet H_2S loading and the experiments were conducted at same conditions and achieved 92.8% H_2S removal.

Acknowledgements

The authors thankfully acknowledge the Department of Science and Technology, Government of India and Indian Institute of Technology Kharagpur, India for their financial and support (Sanction No: DST/INT/SA/P-14/2011).

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