

Optimizing SO₂ Adsorption from Flue Gas Using Microporous Polypropylene Hollow Fiber Membrane Contactor

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ARTICLE INFO	ABSTRACT
<p>Article History: Received: 31 July 2021 Revised: 01 September 2021 Accepted: 03 September 2021</p> <p>Article type: Research</p> <p>Keywords: Flue Gas, Membrane Contactor, Optimization Operational Parameters, RSM, SO₂ Removal</p>	<p>This study optimized the operational parameters of removing SO₂ from flue gas via a polymeric hollow fiber membrane contactor (HFMC) using the response surface methodology (RSM). The distilled water and polypropylene hollow fibers were applied as the adsorbent and membrane material, respectively. Three independent variables were selected as experimental parameters: liquid flow rate, gas flow rate, and initial SO₂ concentration. The initial SO₂ concentration had a remarkable effect on SO₂ removal efficiency. The optimal ratio of liquid-to-gas flow rate was found to be 0.25 to reach maximum separation efficiency (98.81%). The optimal value of the liquid flow rate was 33 l/h, and the optimal gas flow rate was 131 l/h. The effect of CO₂ presence, module length, fibers number, temperature, and the adsorbent nature were also investigated under optimal values obtained for the ratio of liquid-to-gas flow rate. The findings indicated that CO₂ presence in the flue gas slightly affects SO₂ removal using water as an absorbent in HFMC. Furthermore, it was indicated that the SO₂ removal efficiency was a function of the flue gas temperature and number of fibers: it decreased as the temperature rose from 20 to 50°C and the fiber numbers increased from 300 to 1000. This study offers a model to predict the efficiency of SO₂ removal using HFMC under different conditions and provides the ground to further explore the industrial applications of this technology.</p>

Introduction

SO₂ emissions are a major environmental concern due to detrimental impacts on human health and ecosystems. Respiratory problems, the increased risk of asthma attacks, and cardiovascular issues have been associated with high SO₂ emissions [1, 2]. Moreover, gaseous SO₂ is the main element behind the acidic precipitation phenomenon with harmful impacts on ecosystems. As one of the most pervasive air pollutants, SO₂ is primarily generated from the combustion of fossil fuels in power plants [3, 4]. It is necessary to develop appropriate SO₂ elimination technologies from flue gas to overcome the challenges associated with such emissions. The wet scrubbing technology, known as the flue gas desulfurization (FGD) process, is the most common technique for SO₂ adsorption from flue gas stream [3]. However, this process faces several limitations: it needs large space and has high operational costs. Besides,

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foaming, flooding, and entrainment have also been considered operational problems related to conventional wet scrubbing techniques [5, 6].

A hollow fiber membrane contactor (HFMC), known as an alternative technology for acid gas removal, has attracted great attention in recent years [5, 7-12]. In these devices, mass transfer of two phases are achieved without dispersing one phase into the other. The independent liquid and gas flow, operational flexibility and compact size, and high surface contact per unit volume have been reported as the main advantages of this process over the conventional method [13].

A few experimental works have studied SO₂ removal from flue gas using an HF contactor membrane. Park et al. [5] carried out some experiments to evaluate the effect of operational parameters including flow rate of gas/liquid, initial SO₂ concentration, length of the module, concentration, and absorbent nature on the SO₂ adsorption via polyvinylidene fluoride (PVDF) membranes. They also studied the impact of liquid flow rate on the SO₂ removal using PEG-modified PVDF elsewhere [14]. Kim et al. [7] used the SO₂/CO₂/N₂ mixed gas in the hollow fiber composite membrane to evaluate the effect of CO₂ presence on SO₂ removal. In a relevant study, the effect of seawater as the adsorbent for SO₂ removal through polypropylene (PP) hollow fiber membrane contactor was investigated by Sun et al. [15]. Other studies have addressed the effect of membrane properties such as pore size of fiber [16], pore size distribution [17, 18], wall thickness [16], and porosity of membrane [19] on SO₂ separation performance using hollow fiber contactor.

However, all previous researches have used the traditional one-factor-at-a-time (OFAT) approach to study the effect of parameters on the SO₂ separation performance using the HFMC process. As know, the operational parameters may change simultaneously in the large usage, while univariate analysis of OFAT approach remains silent about the simultaneous impacts of operational factors of the process [20]. Based on our current knowledge, there is no investigation reported in the literature to study the interaction between influencing factors on SO₂ efficiency separation using hollow fiber membrane contactor. This study offers a model to predict the efficiency of SO₂ removal using HFMC under different conditions providing further development for the industrial application of this technology. Therefore, for the first time, in this work, response surface methodology (RSM) was used to study the interactions among the investigated parameters on the efficiency of SO₂ removal using the HFMC process.

Most previous studies have reported that the initial SO₂ concentration, gas flow rate, and liquid flow rate significantly impact the removal of SO₂ using the HFMC process [5, 7, 14, 15]. Consequently, these factors were selected as independent variables and RSM was applied to examine their effects and optimize the SO₂ removal efficiency using the contactor membrane. In the second stage, according to the optimal results obtained from the experimental design, the effect of important operating parameters including module length, temperature, fibers number, the nature of adsorbent, and the presence of carbon dioxide was also investigated SO₂ separation efficiency.

Experimental

Materials

Various liquids have been considered as the absorption medium for SO₂ removal in membrane contactors. The absorbents include pure water, seawater [15], aqueous solutions of NaOH, Na₂CO₃, Na₂SO₃, NaHCO₃ [5, 21, 22], amines (DMA) [23], and ionic liquids [24]. The good chemical compatibility with the membrane materials, feasibility of regeneration, environmental compatibility, low cost, and ease of access have been reported as important factors for the absorbent selection. Based on these criteria, in this work, distilled water was used

as the adsorption solvent. To make a comparison, a series of experiments was also conducted using tap water as the solvent to study the effect of solvent nature.

Among various polymeric membranes, PP and PVDF have been reported as the most common membrane materials for SO₂ removal using HFMC [5, 14, 15]. PVDF membranes are excellently thermal and chemical stable against alkaline adsorbent. However, they have a higher cost relative to PP membranes. Thus, according to the utilization of water as an adsorbent in this work, a polymeric hollow fiber membrane composed of PP was used as the membrane material. Some features of the membrane have been shown in Table 1.

Table 1. Physical properties of the membrane

Fiber material	PP
Fiber o.d. (d _o), m	4×10 ⁻⁴
Fiber i.d. (d _i), m	3.5×10 ⁻⁴
Fiber length (L), m	0.3
Number of fibers (n)	310
Membrane porosity (ε)	0.17
Average Pore Radius (r _p)	4.17×10 ⁻⁶

Microporous polypropylene hollow fibers were prepared from Parsian Advanced Industry Co, Iran. SO₂ (1.00 cmol/mol, N₂ balance) and CO₂ gas (99.99%) were provided by Tehran Farafan Gas Co, Iran. Air was provided by an air compressor (EURO 1500-1 model, Einhell).

Experimental Design and Optimization by RSM

The initial SO₂ concentration, gas flow rate, and liquid flow rate have been reported as the most significant parameters affecting the SO₂ removal efficiency using HFMC. In this work, these parameters were selected as independent variables to evaluate their simultaneous effect on SO₂ adsorption. For this purpose, a central composite design (CCD) at five levels was employed to study the effect of liquid flow rate (x₁), gas flow rate (x₂), and initial SO₂ concentration (x₃) in 20 runs. The SO₂ removal efficiency was chosen as the dependent variable (response). Table 2 shows the ranges and levels defined for parameters selected in this work. To evaluate the response variable, all runs were conducted according to Table A.1 of the appendices. The experimental results of CCD were fitted by a second-order polynomial model as follows:

$$Y(\%) = b_0 + b_1x_1 + b_2x_2 + b_3x_3 + b_{12}x_1x_2 + b_{13}x_1x_3 + b_{23}x_2x_3 + b_{11}x_1^2 + b_{22}x_2^2 + b_{33}x_3^2 \quad (1)$$

where Y stands for the response variable, x_i 's represents the independent variables and b_i and b_{ii} indicate the linear and quadratic terms, respectively, and b_{ij} is the interaction coefficients of parameters. The RSM concepts with more details have been basically described elsewhere [25-26].

Table 2. Experimental range and levels of independent variables

Variables	Ranges and levels				
	-2	-1	0	+1	+2
Liquid flow rate (l/h) (x ₁)	10	18.1	30	41.8	50
Gas flow rate (l/h) (x ₂)	76.8	94.3	120	145.6	163.2
Initial SO ₂ concentration (ppm) (x ₃)	200	402.7	700	997.3	1200

Experimental Setup

The SO₂ separation was experimentally investigated in a lab-scale setup. The membrane module consisted of a 5 cm diameter and, 30 cm length was designed for the experimental studies. The number of hollow fiber membranes was 310. The gaseous mixture of SO₂ and air with a certain concentration of SO₂, according to Table 2, flowed through the tube side, and the liquid absorbent stream was passed co-currently inside the shell side of the polypropylene hollow fiber.

To avoid the air bubble formation on the surface of the liquid, the fluid pressure was considered more than 0.2×10^5 Pa. The operating pressure and temperature were constant at atmospheric pressure (10^5 Pa) and room temperature (19 °C). The experimental setup has been schematically illustrated in Fig.1.

The pure gases stream flow rates of SO₂ and air was controlled by two flow meters (Fischer and Porter Co, Germany). The mixing of SO₂ and air streams was done by a gas mixing device. A portable gas analyzer (Model Testo 350, Germany) was used to measure SO₂ concentration in the inlet and outlet gas stream. All results were recorded at a steady state. After 20 minutes of run time, the steady-state was obtained when the concentration of SO₂ in the outlet stream remained constant. The inlet and outlet pressures of the gaseous mixture and liquid adsorbent were controlled by four pressure regulators (VSR-2 series, Vigour Co, Germany). The flow rates of liquid solution and gas mixture were adjusted according to Table 2 using two flow meters (Platon Co, France). A bubble flow meter was also used to calibrate flow meters. Two solution containers were applied to storage the feed/ rich absorbent liquid solution. The absorbent solution was delivered into the HFMC system through a pump. During the tests, the membrane module was kept in a thermostat water bath, allowing the exact control of the temperature. Before all experiments, the fiber membrane modules were pressurized with air for at least one h without liquid absorbent to ensure dryness of membrane pores. The probable leakage of SO₂ from all setup connections was continually controlled using a portable multi-gas monitor (VRAE, PGM-7840 model, USA) during experiments.

The SO₂ removal efficiency (response) can be calculated using Eq. 2 [27]:

$$\begin{aligned} \text{SO}_2 \text{ removal efficiency (\%)} \\ = 100(1 - C_{\text{outlet}}/C_{\text{inlet}}) \end{aligned} \quad (2)$$

where C_{inlet} and C_{outlet} are the concentration of SO₂ at the inlet and outlet of module, respectively.

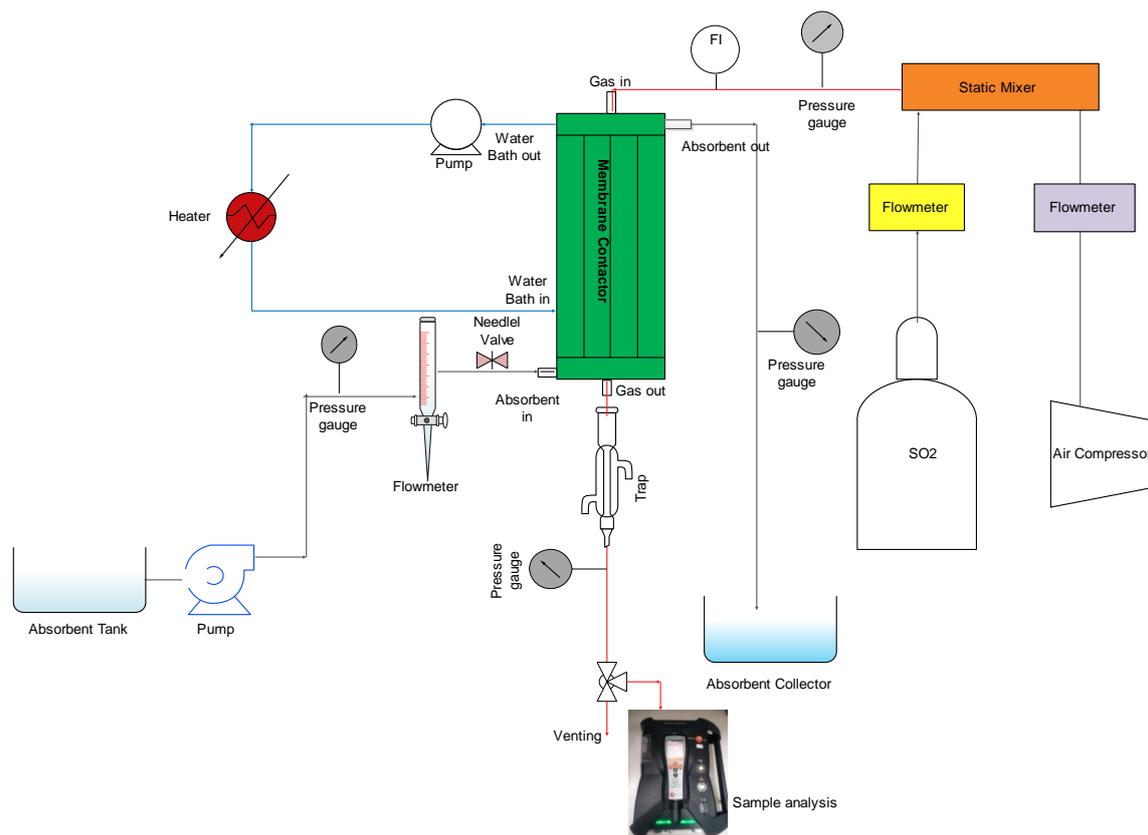


Fig. 1. Schematic diagram of the experimental setup

Results and discussion

Model Fitting and Analysis of Variance (ANOVA)

Based on the experimental runs conducted according to Table A.1 of the appendices and the value of predicted data, a second degree polynomial equation was obtained expressing the correlation between the response and independent variables as follows:

$$\begin{aligned} \text{Efficiency (\%)} = & 0.6334 + 2.11 \times 10^{-3} \times [\text{Liquid flow rate}] + 4.37 \times 10^{-3} \times [\text{Gas flow rate}] + 2.05 \times 10^{-4} \times [\text{SO}_2 \text{ Concentration}] + 2.05 \times 10^{-5} \times \\ & [\text{Liquid flow rate}] \times [\text{Gas flow rate}] + 2.44 \times 10^{-6} \times [\text{Liquid flow rate}] \times [\text{SO}_2 \text{ Concentration}] - 1.98 \times 10^{-6} \times [\text{Gas flow rate}] [\text{SO}_2 \text{ Concentration}] - \\ & 8.56 \times 10^{-5} \times [\text{Liquid flow rate}]^2 - 1.66 \times 10^{-5} \times [\text{Gas flow rate}]^2 - 8 \times 10^{-8} \times [\text{SO}_2 \text{ Concentration}]^2 \end{aligned} \quad (3)$$

The negative coefficients of quadratic terms, x_{ii}^2 , in the polynomial equation indicate that an extra value of independent variables in the system induces a negative impact on SO_2 adsorption [28].

The analysis of variance (ANOVA) was used to assay the model adequacy.

Table 3. ANOVA results for the response surface quadratic model

Source	Sum of squares	Degree of freedom	Mean squares	F value	p value
Model	0.021	9	2.35×10^{-3}	11.55	0.0003
Residual	2.036×10^{-3}	10	2.036×10^{-4}		

Lack of fit	2.35×10^{-3}	5	4.07×10^{-4}
Pure error	0	5	0
Total	0.023	19	

According to ANOVA data (Table 3), the model F-value of 11.55 indicates that the model is significant. There is a 0.03% probability that the model F value could occur due to noise. In addition, the p-value for the model implies the significance level of the model. The insignificant lack-of-fit term indicates that the model can accurately predict the experimental data. The value of “Pred R-Squared” of 0.8333 is in reasonable agreement with the Adj R-Squared of 0.9123, approving the model's ability for accurate prediction of the SO₂ removal efficiency. Fig. 2 presents the predicted values of responses in terms of the actual ones implying the good predictability of the model.

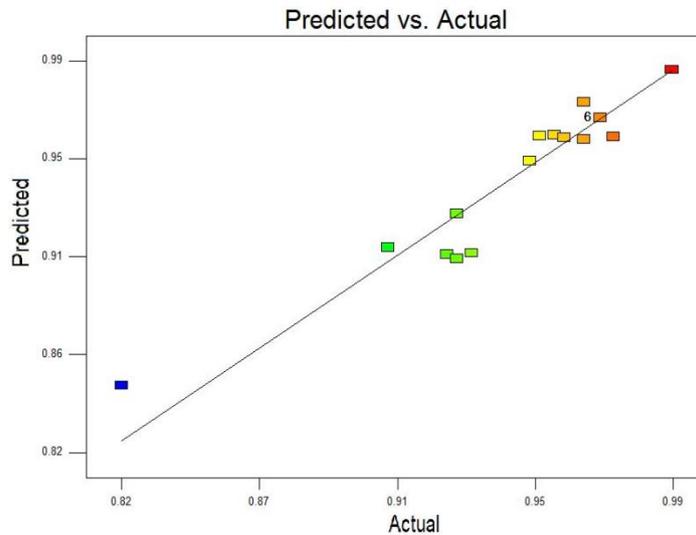


Fig. 2. Experimental data plotted versus the predicted values obtained from the model

The data at a 95% confidence level have been presented in Table A. 2 of the appendices. The p-values were applied as a tool to estimate the significance level of each parameter. Among the variables studied in this work, the initial SO₂ concentration (x_3) is a highly significant factor with $p < 0.0001$. Furthermore, the liquid flow rate (x_1), the gas flow rate (x_2), the interaction effect between the initial concentration of SO₂ and gas flow rate (x_2x_3), the second-order of liquid flow rate (x_1^2), and the second-order of gas flow rate (x_2^2) are significant at $p < 0.05$. Furthermore, the p value > 0.05 means that the model term is insignificant. Table A. 2 shows that the interactions between liquid flow rate and gas flow rate and liquid flow rate and initial concentration of SO₂ are insignificant.

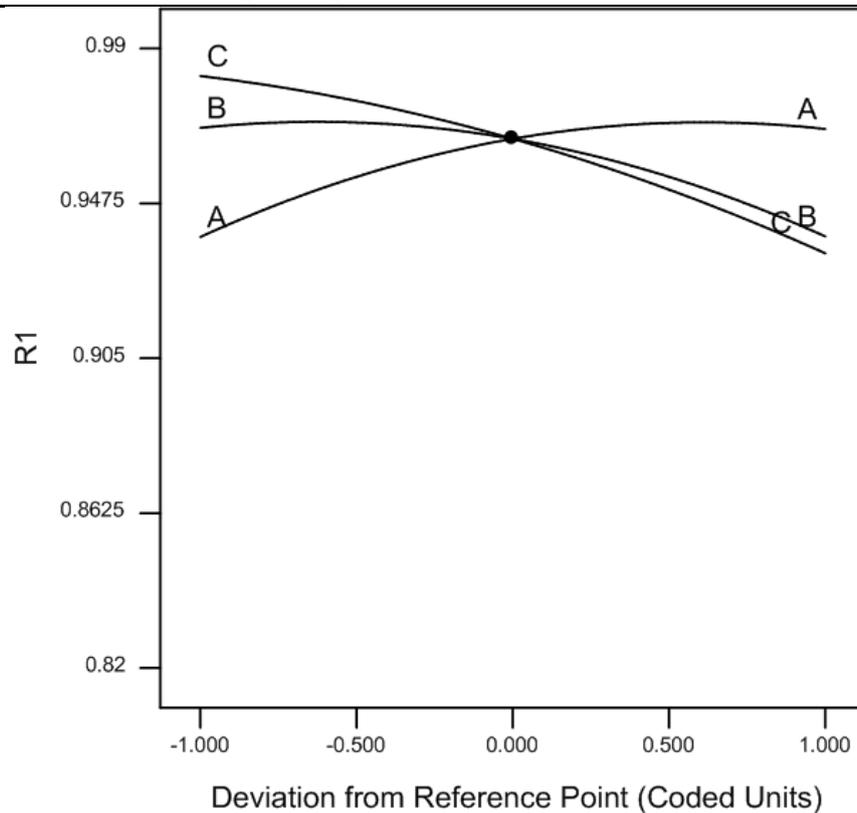


Fig.3. Perturbation plot of SO₂ removal

A comparison of the effects of all factors on the SO₂ removal percentage was performed using a perturbation plot (Fig. 3). The steep curvature of the initial concentration of SO₂ (C) indicates the SO₂ adsorption is highly affected by this parameter.

Based on the monomial coefficient values for each parameter, $p(x_3) < 0.0001$ (initial SO₂ concentration), $p(x_2) = 0.0033$ (gas flow rate) and $p(x_1) = 0.0032$ (liquid flow rate), the significance ranking among the factors is initial SO₂ concentration (x_3) > gas flow rate (x_2) > liquid flow rate (x_1). It is noted that the resultant order can also be ascribed to the experimental range determined for independent variables in the present work.

Response Surface Analysis

To find a better understanding of the interaction between variables, three-dimensional surface plots were applied. The response changes (SO₂ removal efficiency) as the function of the interactions between independent parameters have been presented in Fig. 4.

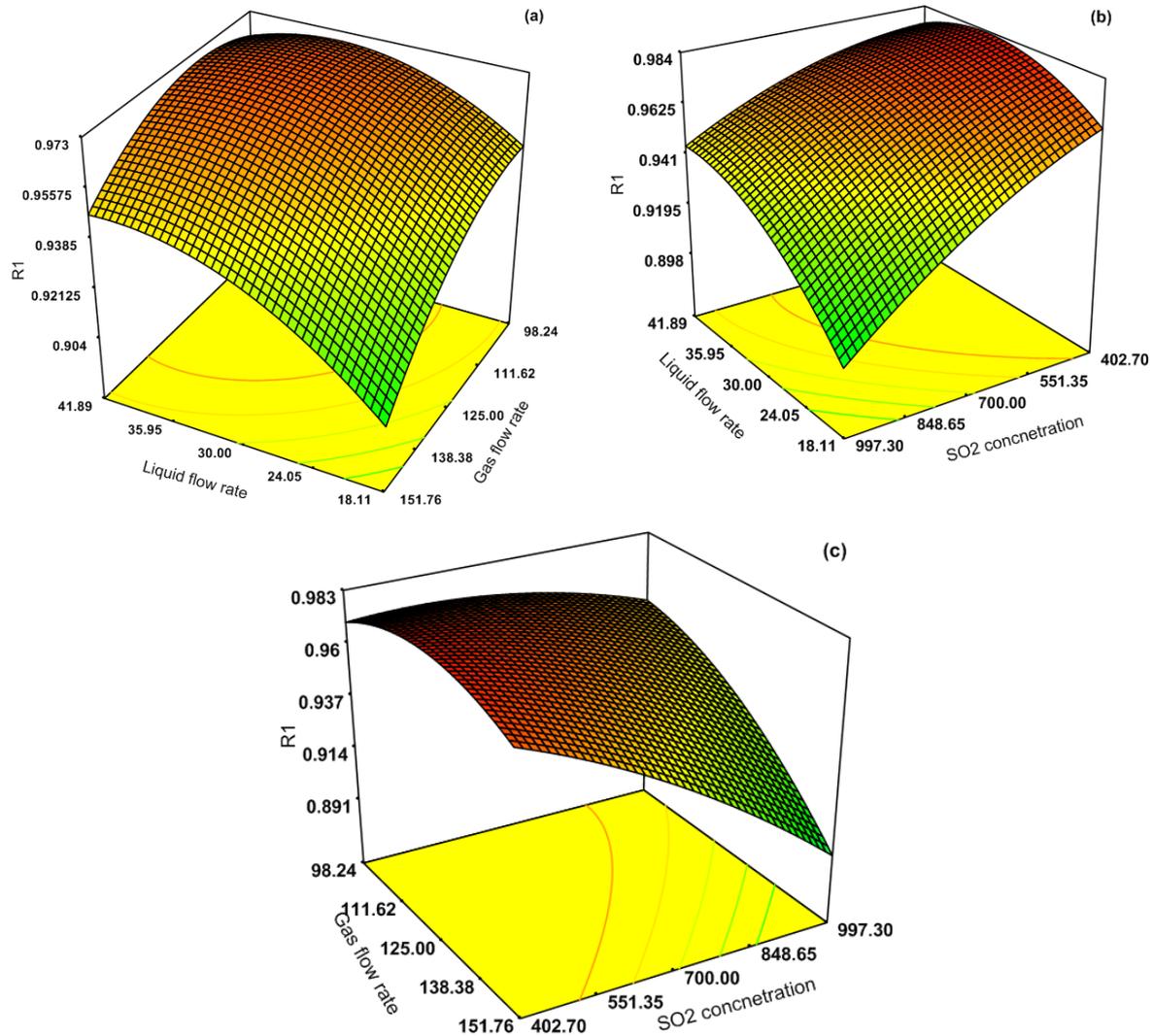


Fig. 4. Simultaneous effect of (a) liquid flow rate and gas flow rate (initial SO₂ concentration: 700 ppm) (b) liquid flow rate and initial SO₂ concentration (gas flow rate: 120 l/h) (c) gas flow rate and initial SO₂ concentration (liquid flow rate: 30 l/h) on SO₂ removal efficiency

Fig. 4a shows the simultaneous effect of liquid flow rate and gas flow rate on SO₂ removal using polymeric hollow fiber. By increasing the liquid flow rate from 10 to 30 l/h, the SO₂ removal percentage remains constant after passing a maximum value. The positive effect of liquid flow rate can be attributed to the decreased resistance of liquid flow rate at a higher flow rate [5]. As a result, the gas diffusion and the mass transfer performance are enhanced. In addition, the increased surface area of gas-liquid contact has been reported as another reason for the enhanced SO₂ removal efficiency at a higher flow rate of the liquid phase [29]. However, results show that the SO₂ removal efficiency achieves a relatively steady state when the liquid flow rate increases over 30 l/h. This is because the transfer of SO₂ might not be controlled by the liquid film resistance at a flow rate over 30 l/h. Results in Fig. 4a show that the simultaneous increase in liquid and gas flow rate leads to a decrease in the SO₂ adsorption using HFMC. It can be attributed to the decreased residence time of gas associated with the increased gas flow rates, which leads to the lower diffusion rate of SO₂ gas inside the membrane [30].

The effects of liquid flow rate and initial SO₂ concentration on SO₂ removal efficiency have been shown in Fig. 4(b). The SO₂ removal decreases significantly with increasing the

concentration of SO₂ in the inlet gas stream. As known, a larger amount of adsorbent is required when the feed SO₂ concentration increases. Therefore, it is expected that the SO₂ removal percentage reduces when the flow rate of liquid is constant. By increasing the liquid flow rate while the feed SO₂ concentration increases, the active sites for adsorption of SO₂ increase suppressing of the negative impact of SO₂ concentration on the removal efficiency, as shown in Fig. 3 (b).

Fig. 4(c) shows the effects of gas flow rate and initial SO₂ concentration on the removal efficiency of SO₂ using polymeric HF. As can be seen, the SO₂ removal efficiency increases with an increase in the gas flow rate from 76 l/h to 120 l/h and decreases gradually when the flow rate of gas increases beyond 120 l/h. The positive effect of gas flow rate can be attributed to the reduced gas boundary layer thickness due to the increased gas flow rate. A similar result has been reported in the literature [30]. By increasing the gas flow rate over an optimum value, the removal efficiency decreases due to the reduced residence time of gas at the higher gas flow rates, as implied earlier. However, the increase in initial SO₂ concentration accelerates the negative impact of gas flow rate on the system performance. This behavior can be related to the insufficient active site for SO₂ adsorption when the ratio of gas-to-liquid flow rate increases.

Optimization of the operational conditions

In order to determine the optimal operational conditions for the SO₂ removal using HFMC, a desirability function was applied for optimizing. The criteria for all parameters according to the removal percentage have been presented in Table A. 3 of the appendices. The weight, or importance, implies added emphasis to the upper/lower bounds or target values. According to the settings given in Table A. 3, the optimal operational conditions for maximum SO₂ removal efficiency (98.81 %) using HFMC were found to be a liquid flow rate of 33 l/h and a gas flow rate of 131 l/h.

Model Validation and Confirmation

To confirm the adequacy of the model for the prediction of the maximum percentage efficiency of SO₂, validation of the model was performed under the optimal operating conditions derived from the software. Average maximum efficiency of 97.74 % was obtained through three repeated experiments carried out in optimal operational conditions. The reasonable agreement between the predicted (98.81 %) and the actual results (97.74 %) demonstrates the model's ability for valid simulation of the SO₂ removal using polymeric HFMC.

Under optimal operational conditions for liquid and gas flow rate and initial SO₂ concentration, the effect of some parameters was investigated. The obtained results have been presented in Figs. 4 to 6.

Effect of operational parameters

Effect of the Presence of CO₂

To evaluate the effect of CO₂ presence on the SO₂ removal percentage, the gas mixture of SO₂ and air with 10% CO₂ concentration was introduced into the system at room temperature. The CO₂ concentration in the inlet gas stream was adjusted by a programmable gas divider (SONIMIX 2106-X-Corrosive model, Switzerland). A gas analyzer (Model Delta 1600-S, MRU Instruments Inc., USA) was applied to measure the CO₂ concentration in the outlet gas stream.

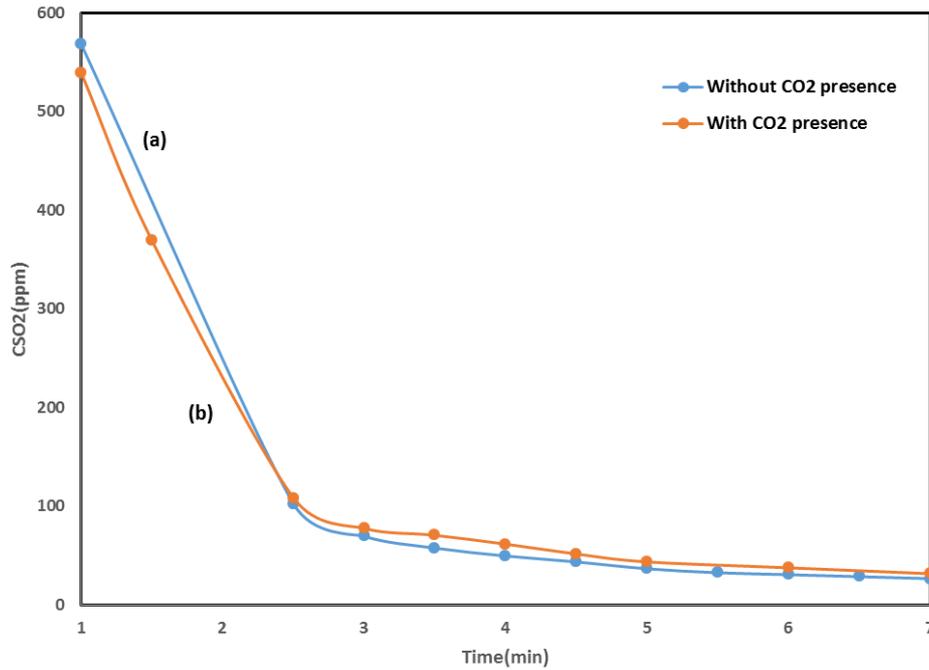


Fig. 5. Effect of CO₂ presence on SO₂ adsorption (L: 33 l/h, G: 131 l/h, C_{SO₂}: 700ppm)

The results demonstrated that the presence of CO₂ gas along with SO₂ in the inlet gas has no significant effect on the separation efficiency of the sulfur dioxide (Fig. 5). In fact, the CO₂ does not compete with the SO₂ gas when being absorbed by the distilled water, which is due to the low reactivity of CO₂ with the distilled water absorbent.

It is worth mentioning that during this experiment, the CO₂ concentration insignificantly declines from 10 to 7% by volume, confirming the lower reactivity of CO₂ with water.

Effect of Temperature

It is noted that some experiments were performed at 30 °C and 50 °C to study the effect of temperature on SO₂ adsorption using HFMC. For this purpose, the water bath was heated up to the desired temperature using a heater connected to the water bath. Before the tests, the gas mixture was flowed in the membrane module for two h to achieve thermal equilibrium.

The effect of temperature on the SO₂ separation efficiency by distilled water in the Membrane contactor is indicated in Fig. 6. According to the results, as the temperature increases, the separation efficiency of the SO₂ declines. Concerning the sulfur dioxide's exothermic dissolution in water, such an observation is expected. Similar results are reported by other researchers [7].

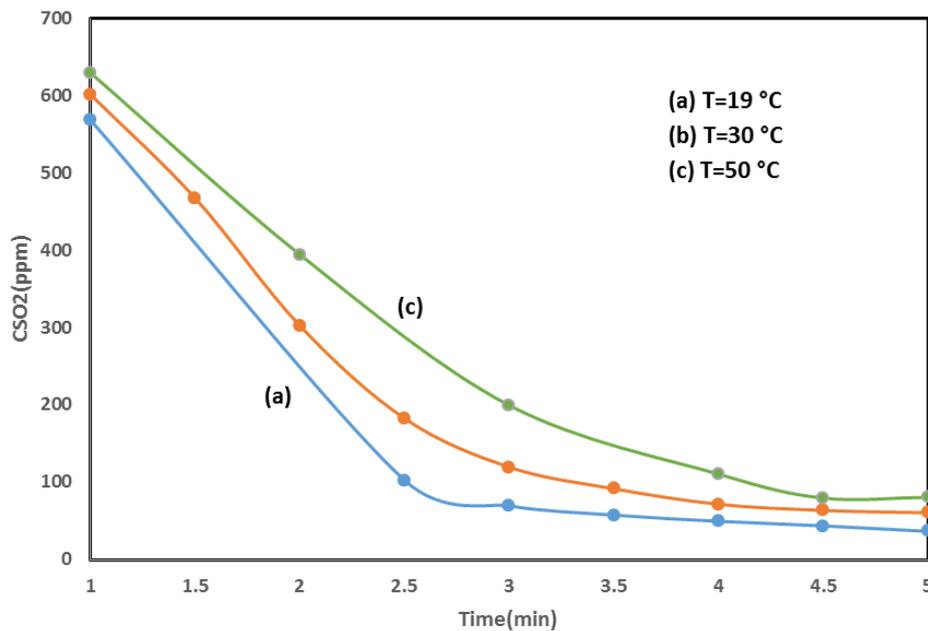


Fig. 6. Effect of temperature on SO₂ adsorption (L: 33 l/h, G: 131 l/h, C_{SO₂}: 700ppm)

Effect of Absorbent Nature

To investigate the effect of the absorbent nature on the separation efficiency of the SO₂ by the membrane contactor, its performance using the tap water as adsorbent was examined. The results indicated that in the case of using tap water and after 5 min of reaction initiation and reaching the equilibrium, the separation percentage of the SO₂ is 92% which is lower than that of the distilled water (96%). However, based on these findings, when the efficiency less than 95% is desirable, tap water instead of distilled water can be used, and the costs related to the solvent could be saved.

Effect of Number of Fiber

Fig. 7 shows the impact of fiber number on the separation efficiency of SO₂ by the distilled water in the membrane contactor.

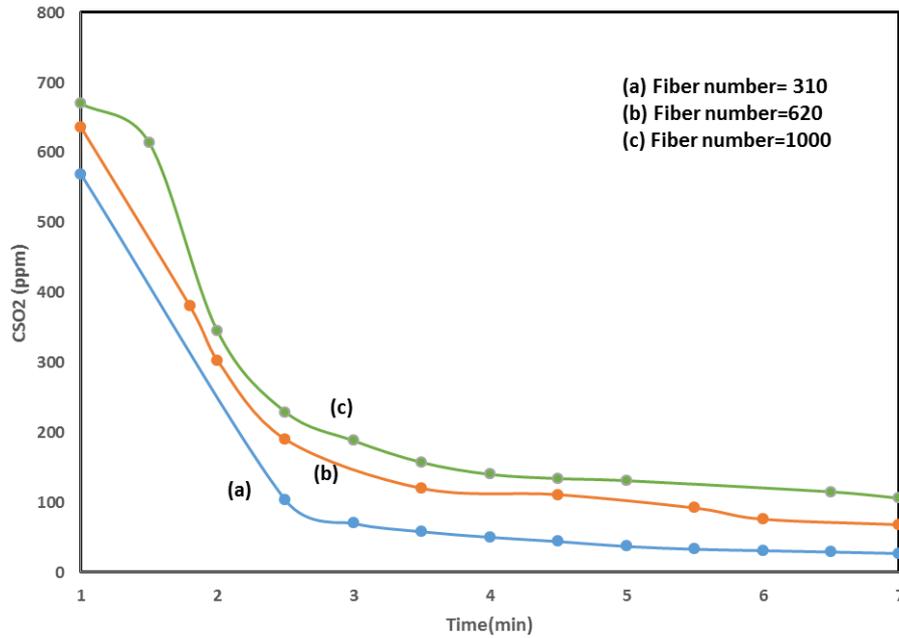


Fig. 7. Effect of temperature on SO₂ adsorption (L: 33 l/h, G: 131 l/h, C_{SO₂}: 700ppm)

According to the results, as the number of fibers in the membrane modulus increases, the separation efficiency of sulfur dioxide declines. This result can be because of the decrease in the space between fibers by virtue of their high density in the fibers with more than 300 numbers. Such being a case, the mass transfer between the gas and liquid phases diminishes [16]. Besides, some researchers believe that as the number of fibers increases, the dead regions are created inside the shell, reducing the absorption of SO₂ [31].

Effect of Module Length

The effect of the module length on the separation percentage of the SO₂ by the distilled water in the membrane contactor was investigated by increasing the module length from 30 to 100 cm. The results indicate that the separation efficiency changes from 96 to 98% when the module length increases by three times. These results may be due to the independence of the mass transfer coefficient and the SO₂ diffusion in the module with a length greater than 30 cm. Such results have also been reported by Park et al. [5].

Conclusion

The experimental design methodology was used to optimize parameters in the SO₂ separation from flue gas using HFMC. A quadratic model expressed the correlation between the removal efficiency of SO₂ and independent parameters. Under the optimal ratio of liquid-to-gas flow rate (0.25), the separation efficiency of SO₂ approached 98.81%. The CO₂ presence exhibits an insignificant effect on SO₂ removal using water as an adsorbent under optimal conditions. By increasing the number of fibers and temperature, the removal efficiency decreases while the SO₂ adsorption changes independently as the module length increase. This study can be considered as beneficial research for the further development of SO₂ removal from flue gas using HFMC.

Nomenclature

ANOVA	Analysis of variance
r_p	Average pore radius (m)
d_i	Fiber inner diameter (m)
L	Fiber length (m)
d_o	Fiber outer diameter (m)
ε	Membrane porosity
n	Number of fibers

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Appendices

Table A. 1. Experimental designs and experimental results with predicted values

Run	Expeiremental conditions			SO ₂ removal efficiency (%)	
	x ₁	x ₂	x ₃	Experimental	Predicative
1	30	120	700	97	96
2	30	120	700	97	96
3	41.89	94.31	997.30	91	92
4	18.11	94.31	997.30	83	86
5	30	120	700	96	96
6	41.89	145.68	997.30	95	95
7	30	76.8	700	95	92
8	30	163.2	700	93	94
9	50	120	700	96	96
10	18.11	145.68	997.30	92	92
11	41.89	94.31	402.70	96	98
12	30	120	700	96	96
13	41.89	145.68	402.70	97	95
14	18.11	145.68	402.70	94	93
15	30	120	200	99	99
16	30	120	1200	92	91
17	18.11	94.31	402.70	95	96
18	30	120	700	95	96
19	10	120	700	92	91
20	30	120	700	96	96

Table A. 2. Coefficients of regression and their significances

Factor	Coefficient Estimate	Degree of freedom	Standard Error	F-value	95% confidence interval Low	95% confidence interval High	p-value
Intercept	0.97	1	5.82E-3	-	0.95	0.98	-
x ₁	0.015	1	3.86E-3	14.71	6.20E-3	0.023	0.0033
x ₂	-0.015	1	3.86E-3	14.87	-0.023	6.28E-3	0.0032
x ₃	-0.025	1	3.86E-3	39.68	-0.033	-0.016	< 0.0001
x ₁ x ₂	6.52E-3	1	5.04E-3	1.67	-4.71E-3	0.018	0.2250
x ₁ x ₃	8.65E-3	1	5.04E-3	2.94	-2.59E-3	0.02	0.1172
x ₂ x ₃	-0.016	1	5.04E-3	9.81	-0.027	-4.55E-3	0.0107
x ₁ ²	-0.012	1	3.75E-3	10.39	-0.02	-3.54E-3	0.0091
x ₂ ²	-0.012	1	3.75E-3	10	-0.02	-3.74E-3	0.0101
x ₂ ³	-7.08E-3	1	3.75E-3	3.55	-1.29E-3	-0.015	0.0890

Table A. 3. Optimization of the individual responses (di)

Name	Goal	Lower Limit	Upper Limit	Lower weight	Upper weight	Importance
Liquid flow rate	In the range	10	50	1	1	3
Gas flow rate	In the range	76.8	163.2	1	1	3
Initial SO ₂ concentration	In the range	200	1200	1	1	3
Response (degradation %)	Maximize	91	99	1	1	5