

Research Article

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Simultaneous removal efficiency of H₂S and CO₂ by high-gravity rotating packed bed: Experiments and simulation

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Abstract: This study explores the possibility of applying high-gravity rotating packed bed (HGRPB) in removing H₂S and CO₂ from biogas. Ca(OH)₂ aqueous solution was used as the absorbent in this study. Different experimental conditions including solution pH, rotating speed (R_S) of HGRPB, gas flow rate (Q_G), and liquid flow rate (Q_L) were investigated with respect to the removal efficiency (E) of H₂S and CO₂. The experimental and simulated results show that the optimal removal efficiency of H₂S and CO₂ using HGRPB achieved nearly the same as 99.38 and 99.56% for removal efficiency of H₂S and 77.28

and 77.86% for removal efficiency of CO₂, respectively. Such efficiencies corresponded with the following optimal conditions: a solution pH of 12.26, HGRPB reactor with the rotating speed of 1,200 rpm, the gas flow rate of 2.46 (L/min), and the liquid flow rate of 0.134 (L/min).

Keywords: biogas upgrading, high-gravity rotating packed bed, removal efficiency, carbon dioxide, hydrogen sulfide

1 Introduction

The current trend of energy production is shifting toward environmentally friendly forms of green energy due to the scarcity and depleting status of fossil fuel sources. Of which, biogas fuels have been attracting research attention from scientists in recent years. Biogas is a mixture of many different gases generated from the decomposition of organic compounds in an anaerobic process with the main participation of methane-producing bacteria (methanogen) or anaerobic digestion organisms [1]. The usable materials in biogas production are diverse, ranging from agricultural waste, manure, municipal waste, wastewater, food waste, to sludge from the wastewater treatment plant. Biogas is primarily composed of methane (CH₄), carbon dioxide (CO₂), hydrogen sulfide (H₂S), moisture, and siloxane [2]. A large proportion of biogas components belong to hydrogen sulfide (H₂S) and CO₂; however, they are responsible for a number of drawbacks. At high concentrations, H₂S is a toxic gas and could cause erosion at metal contact surfaces [3–5]. Although CO₂ is not as corrosive as H₂S, it could reduce the calorific value of biogas fuel when existing in large amounts. To improve the applicability of biogas, especially for electricity generation, the purification of biogas to biomethane is considered as a crucial process. Two main steps to convert biogas to biomethane are as follows: (1) cleaning process to remove polluting components such as H₂S, CO₂, H₂O, and NH₃ and (2) upgrading

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process to improve the thermoelectric response of the gas for power generation [2].

Recent studies involving the removal of contaminants such as H₂S, CO₂, H₂O, NH₃, or siloxanes in biogas often adopt diverse methods. The biological process is commonly used to utilize living organisms in the conversion of contaminants into less harmful forms. Another popular pathway is to use physicochemical techniques that include reactive and nonreactive absorption [6–9]. The latter has been attracting more attention due to its advantage in terms of removal efficiency and applicability in larger-scale implementations. Water and organic solvent are frequently used in physicochemical processes to eliminate CO₂ and H₂S. Water has been referred as a popular absorbent [10]; other publications presented that CO₂ and H₂S can be removed from using absorption processes that rely on binding force difference between more polar CO₂ or H₂S and nonpolar methane [11–13]. Previous results presented another technique that involves high-pressure water scrubbing technology to remove CO₂ and H₂S [14,15]. The aqueous absorbents that were frequently used to reduce the contaminants include mono ethanolamine (MEA), di-methyl ethanolamine (DMEA), KOH, K₂CO₃, NaOH, Fe(OH)₃, and FeCl₂ [16,17]. On another aspect, specialized microorganisms might also be considered to use biological methods to reduce H₂S in biogas via converting it to elemental sulfur and some sulfates [18,19].

Among three operating modes of gas–liquid flow: forward, backflow, and cross-flow, backflow mode has the largest mass transfer force but requires a higher gas pressure to push gas from the bottom to the top of the device. Crossflow mode is rarely used in practical applications due to the complex device equipment. High-gravity rotating packed bed (HGRPB) has been the typical process that enhances contact and mass transfer between phases [20]. In this technique, the flow of gas–liquid into the device forms a centrifugal eddy current between the gas and liquid phases, which greatly aids the process of transferring the mass between the gas and liquid phases. The gas–liquid contacting inside HGRPB creates thinner film membranes (1–10 μm) or smaller droplets (10–100 μm) that increase the contact surface between the liquid and gas phases [20,21]. This process has been widely applied in several fields such as nanoparticle fabrication [22,23], extract separation process [24,25], and reaction acceleration [26,27]. In the field of environmental engineering, HGRPB is used in the wastewater treatment [28,29], CO₂ capturing [30–32], and NO₂ and H₂S removal [33–37]. Therefore, the HGRPB reactor is applied in this study to enhance removal efficiency of CO₂ and H₂S in biogas production. The results are expected

to contribute to the process intensification of biogas-based power generation and demonstrate the feasibility of HGRPB in real-scale applications.

In this study, the main objective was to investigate the performance of the HGRPB used for upgrading the simultaneous removal of CO₂ and H₂S from biogas using Ca(OH)₂ solution as an absorbent. Effects of pH correlated to Ca(OH)₂ concentration added, rotating speed (R_S) of HGRPB reactor, gas flow rate (Q_G), and liquid flow rate (Q_L) on removal efficiency (E) of H₂S and CO₂ in raw biogas production were evaluated and simulated through absorption experiments in a pilot-scale HGRPB system. The optimal obtained results were simulated and calculated by modeling. The experimental and simulated results are expected to contribute to the process intensification of biogas-based power generation and demonstrate the feasibility of HGRPB in real-scale applications.

2 Materials and methods

2.1 Materials and experimental apparatus

The raw biogas used for this study was obtained from an anaerobic sludge system of a brewery wastewater treatment facility, and the composition of the gas was illustrated in Table 1. The composition of the input biogas was setup for all experiments with a CO₂ concentration of 19.44–28.01%, H₂S concentration of 800–900 ppm, and CH₄ concentration of 40.3–55.5%. The organic sludge, after being collected from the thickening tank, was pumped into the equalization tank for pH, organic, and nutrient control. Afterward, the sludge was transferred into the anaerobic system with the real pilot operation parameters as follows: digestion tank volume of 80 m³, sludge rotating speed of 20 rpm, sludge retention time of 25 days, and digested temperature of around 32–35°C (Figure 1). The digested sludge has been composted to produce organic composting, and the electricity is used to supply for the wastewater system. In each experiment setup for the removal of hydrogen sulfide (H₂S) and carbon dioxide (CO₂), raw biogas is introduced from the

Table 1: Composition of input biogas

Parameter	Unit	Concentration
CO ₂	(%)	19.44–28.01
H ₂ S	(ppm)	800–900
CH ₄	(%)	40.3–55.5

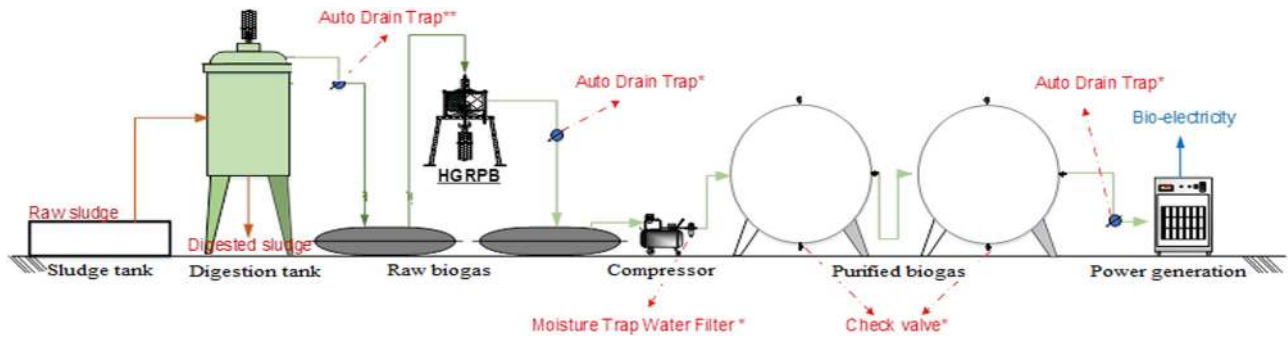


Figure 1: Scheme of a pilot-scale process to generate raw biogas from sludge anaerobic digestion system and purification by HGRPB for generation.

bottom of the HGRPB device at the confirmed initial concentration of CO_2 and H_2S through the HGRPB instrument containing absorbents.

In this study, the high-gravity rotating packed bed instrument is made from stainless steel with a body height of 6 cm, an internal diameter of 21 cm, an outer diameter of 38 cm, a porosity of 97, and a total specific surface area of $50 \text{ m}^2/\text{m}^3$ (Figure 2). The HGRPB instrument is motorized by a centrifugal rotary motor with adjustable rotating speed ranging from 50 to 1,700 rpm.

2.2 Experimental procedure

The raw biogas mixture is introduced from the bottom of the HGRPB device. The $\text{Ca}(\text{OH})_2$ absorbent ($M = 315.47$, 98% purity; Merck, Darmstadt, Germany) solution with different initial concentrations (0.01 and 0.1M) enters from the top of the device and is evenly sprayed inside by centrifugal force and a mesh distribution system. It is ensured that the process of mass transfer from the gas phase to the liquid phase takes place steadily. After the

process, the purified biogas flow is released from the top of the HGRPB device and the adsorbed solution is removed below the HGRPB device (Figure 2). All the experiments were carried out in triplicate for each condition setup. The feed gas and the treated gas of H_2S and CO_2 were sampled for the analysis of H_2S and CO_2 concentration for removal efficiencies.

2.3 Methods of analysis

The concentrations of H_2S and CO_2 in the input and treated gas stream are determined according to the method of ASTM D 5504-12 by gas chromatography. A chromatography (GC 2010 Shimadzu, Japan) with a thermal conductivity detector (TCD) was introduced. Samples were injected using a gas-tight syringe (Hamilton, USA). The pH value of the solution is measured according to the method of ISO 10523:2008 by Horiba U-52G multi-parameter water quality meter.

The removal efficiency (E) of H_2S and CO_2 is calculated by the following equation:

$$E(\%) = \frac{C_0 - C_1}{C_0} \times 100,$$

where C_0 is the initial concentration of H_2S (ppm) or CO_2 (%) and C_1 is the concentration of H_2S (ppm) or CO_2 (%) after purification.

All the experiments were carried out in triplicate to produce mean values and standard deviations. Microsoft Excel was used to analyze the data.

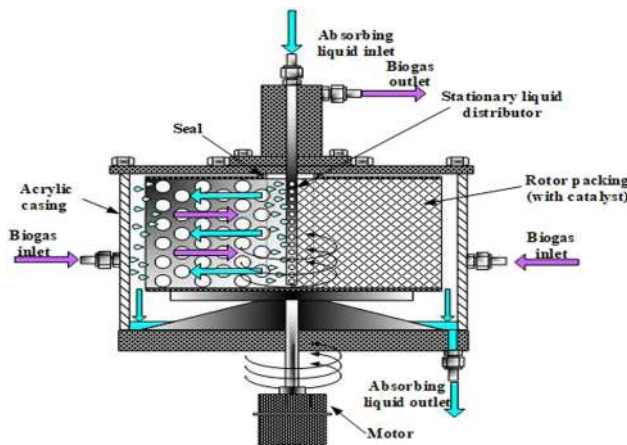


Figure 2: Principle of operation of the HGRPB for biogas purification.

2.4 Response surface methodology (RSM) optimization and its experimental design

The response surface methodology, in conjunction with the Box–Behnken design, was used to estimate the effect

of four parameters (rotating speed, liquid flow rate, gas flow rate, and effect of pH correlated to Ca(OH)₂ concentration added) on removal efficiency (*E*) of H₂S and CO₂ in raw biogas production. The procedure for the construction of 29 experiments – designed matrix with the mathematical-statistical treatments and the determination of optimal conditions were executed using Design-Expert 7.0 software.

To optimize four experimental parameters (rotating speed, liquid flow rate, gas flow rate, and pH correlated to Ca(OH)₂ concentration added) with respect to removal efficiency (*E*) of H₂S and CO₂ in raw biogas production, a response surface methodology routine was used in combination with Box–Behnken experimental design. Based on single-factor investigation results, 29 combinations of parameters were produced using the experimental design, and these parameters were attempted in actual experimental runs to generate the data for model estimation. Then, ANOVA analysis was employed to test model validity. From the estimated model, optimal conditions were calculated. Final optimized parameters were then verified by another actual experiment. The generation of experiment design and estimation of model parameters was performed in Design-Expert software. The second-order equation representing the influence of variables and their interaction on the response is shown as follows:

$$Y = b_0 + \sum_{j=1}^k b_j X_j + \sum_{u,j=1}^k b_{uj} X_u X_j + \sum_{j=1}^k b_{jj} X_j^2,$$

where *Y* is the predicted response, *b*₀ is the intercept coefficient, *b*_{*j*} is the linear coefficient, *b*_{*jj*} is the square coefficient, *b*_{*uj*} is the interaction coefficient, *X*_{*u*} and *X*_{*j*} are the coded independent variables, and *X*_{*u*}*X*_{*j*} and *X*_{*j*}² are the interaction and quadratic terms, respectively.

Ethical approval: The conducted research is not related to either human or animal use.

3 Results and discussion

3.1 Effect of pH on H₂S and CO₂ removal

A solution of Ca(OH)₂ was used as an absorbent and controlled the desired pH value for evaluation of the removal efficiency of H₂S and CO₂. Initial aqueous solutions of Ca(OH)₂ 0.01 and 0.1M are used to control pH for experiments in the investigation of CO₂ and H₂S removal.

However, experiments of pH 13 are difficult to obtain as desired; thus, NaOH solution 1 M is added into the reactor to evaluate compared experiments. The input gases including CO₂ concentration of 19.44–28.01%, H₂S concentration of 800–900 ppm, and CH₄ concentration of 40.3–55.5% were setup in the device of the HGRPB with a rotating speed of 900 rpm, gas flow rate of 2.5 (L/min), Ca(OH)₂ solution – liquid flow rate of 0.125 (L/min). Experiments with pH 10, 11, 12, and 13 in the HGRPB reactor were investigated. The obtained results of H₂S and CO₂ removal by the HGRPB are shown in Figure 3.

Generally, increasing the pH of the solution led to improved removal efficiencies of both CO₂ and H₂S in biogas. This is explained by the greater number of OH[−] ions per unit volume caused by higher Ca(OH)₂ concentrations, which promote the absorption of H₂S and CO₂. Moreover, the movement of H₂S and CO₂ from gas to gas–liquid interface and eventually to the liquid phase in the Ca(OH)₂ solution in HGRPB could accelerate the reactions. It was also indicated that the removal efficiency increases rapidly when increasing the pH from 10 to 12, corresponding to the fast increase efficiency from 59.67 to 98.62% for H₂S, and from 40.45 to 76.23% for CO₂. However, increasing the pH from 12 to 13 did not cause the removal performances to change significantly. Referring to several previous works, it was shown that one study reported CO₂ removal of only 47% [4] while another also used HGRPB to eliminate H₂S content and H₂S was reduced less than 100 ppm after purification [35]. Almost all of the above previous works showed positive results; however, complicated techniques were applied to purify contaminant compounds. The significant results in this step indicated that HGRPB with Ca(OH)₂ solution of pH = 12 is highly effective and beneficial, which serves biogas purification for generation. Therefore, Ca(OH)₂

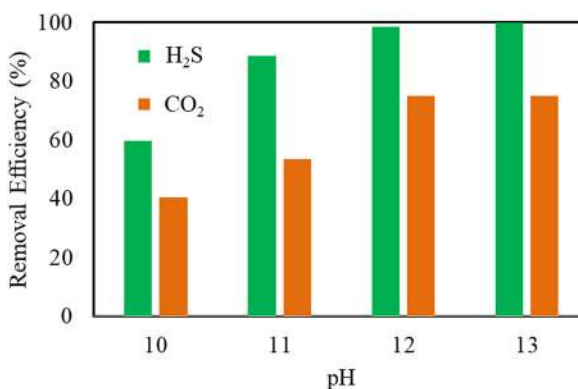


Figure 3: Effect of pH on the removal efficiency of H₂S and CO₂.

solution of 0.01 M added at pH = 12 was selected for subsequent investigations.

3.2 Effect of the rotating speed (R_S) of HGRPB on the removal efficiency of H_2S and CO_2

Figure 4 shows the variation of removal efficiency of H_2S and CO_2 , corresponding to test conditions of rotating speed of HGRPB at 600, 900, 1,200, and 1,500 rpm. In this experiment, a concentration of 0.01 M $Ca(OH)_2$ solution was used to maintain pH 12 of the solution, Q_G of 2.5 (L/min), and Q_L of 0.125 (L/min).

Data in Figure 4 indicated that the removal efficiency of H_2S and CO_2 increases with increasing R_S of HGRPB from 600 to 1,200 rpm. This result is due to the fact that when increasing the rotation speed of the device, the liquid solution produces a thinner film or smaller droplets. This causes the mass transfer process between the gas and the liquid phases, and in turn, the absorbing process to occur thoroughly. Particularly, when R_S accelerates from 600 to 1,200 rpm, the removal efficiency of H_2S and CO_2 increases rapidly from 86.63 to 99.81% and from 50.56 to 76.23%, respectively. As the R_S exceeds 1,500 rpm, improvement in efficiencies was not significant, achieving the maximum efficiency of 77.74% for CO_2 and 99.68% for H_2S . The obtained efficiencies in this work are similar to that where optimal removal efficiency of H_2S achieved 99.8% at R_S of 1,100 rpm and higher than that in literature where the R_S of 1,200 rpm gave optimal SO_2 removal [35,36]. Thus, the rotating speed at 1,200 rpm was selected for all next experiments.

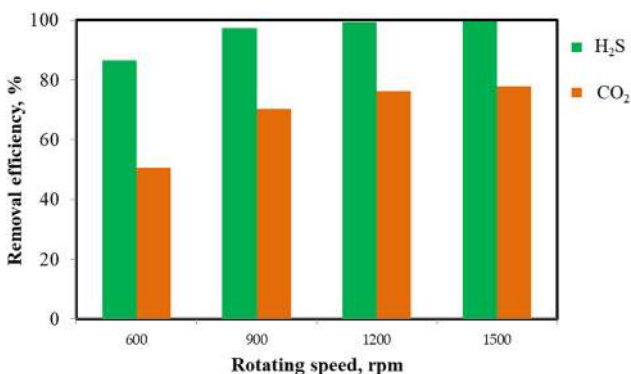


Figure 4: Effect of rotating speed (R_S) on the removal efficiency of H_2S and CO_2 .

3.3 Effect of Q_G on H_2S and CO_2 removal

Effects of Q_G on the removal efficiency of H_2S and CO_2 were tested under varying Q_G of 1.25, 2.5, 3.75, 5, and 7.5 L/min. The pH of solutions, R_S , and Q_L were kept constant at 12, 1,200 (rpm), and 0.125 (L/min), respectively. The obtained values are shown in Figure 5.

It was indicated that the removal efficiency of H_2S and CO_2 decreases when increasing Q_G speed while keeping the velocity of fluid flow and R_S constant. Accelerating the gas flow from 1.25 to 7.5 L/min seemed to reduce the removal efficiency of H_2S and CO_2 from 99.86 to 73.09% and from 77.85 to 51.33%, respectively. The trend could be explained by the greater amount of H_2S and CO_2 and reduced contact time between gas and liquid, thus leading to the lowered removal of H_2S and CO_2 . This trend is similar to that of literature [35,36]. From the experimental results shown in Figure 5, Q_G of 2.5 L/min was selected as the basic condition for subsequent experiments.

3.4 Effect of the Q_L on the removal efficiency of H_2S and CO_2

The dependence of removal efficiency of H_2S and CO_2 on the liquid flow rate is shown in Figure 6. Parameters in this experiment included concentration of $Ca(OH)_2$ of 0.01 M, pH of the solution of 12, R_S of the HGRPB of 1,200 rpm, and Q_G of 2.5 L/min. Q_L was allowed to vary at 0.05, 0.083, 0.125, 0.167, and 0.25 L/min.

Rising the $Ca(OH)_2$ flow rate improved the removal efficiency of CO_2 . The maximum removal efficiency of CO_2 was 77.15%, reached at the flow rate of 0.125 (L/min). Regarding the H_2S elimination, the gas was almost eliminated (remaining amount of lower than 0.1 ppm) even at the liquid flow rate of 0.125 (L/min). This is explained by

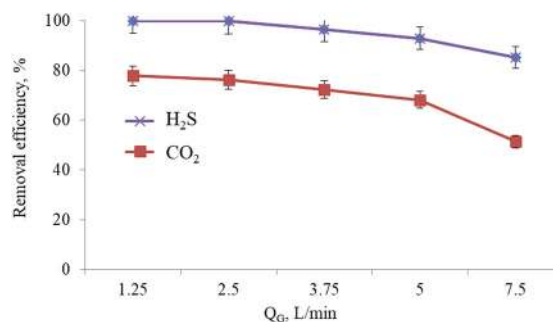


Figure 5: Effect of Q_G on the removal efficiency of H_2S and CO_2 .

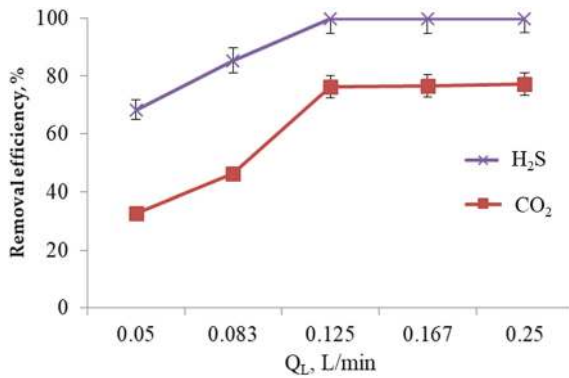
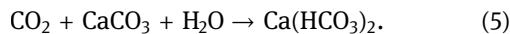
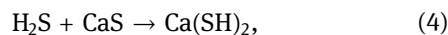
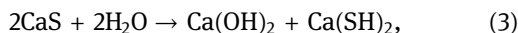
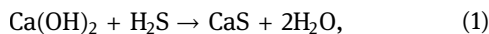


Figure 6: The effect of Q_L on the removal efficiency of H₂S and CO₂.

the increase in the flow of the absorbent solution in a given interaction period and volume of gas, which in turn improves the volume of passing solution and the purification process. However, rising the flow rate to very high levels (Q_L of 0.25 L/min) did not improve the removal efficiency of H₂S and CO₂ significantly. Comparing with the results in the literature to explore the effects of Q_G and Q_L on the removal efficiency of H₂S, current results obtained the same trend [35]. Therefore, Q_L of 0.125 (L/min) was selected as the appropriate Ca(OH)₂ flow rate value for obtained efficiency of H₂S and CO₂ removal.

Hence, removal of H₂S and CO₂ in biogas by absorption mechanism of Ca(OH)₂ solution into the reactor gives high efficiencies with optimal conditions: pH of 12, rotating speed of HGRPB of 1,200 (rpm), gas flow rate of 2.5 (L/min), and liquid flow rate of 0.125 (L/min), and it is explained by reactions as follows [38,39]:

In the high alkaline at pH 12:



With a higher concentration of H₂S in the gas, reactions then occur as



Reactions (5) and (6) simultaneously happened; thus, it is difficult to form precipitation of CaCO₃ in the reactor due to HGRPB apparatus with its performance and all reactions happened continually in the solution. Therefore, H₂S and CO₂ concentrations were removed simultaneously.

3.5 Predicted model and statistical analysis

From the experimental data of the effect of univariate technological parameters to the objective functions Y1 and Y2, experiments were planned according to the Box–Behnken's description. The original (0), low (−1), and high (+1) levels of the factors (with $k = 4$) and the variable interval are listed in Table 2.

The Design-Expert 7.0.0 software was used to build an experimental matrix with 29 experiments. The objective functions are Y1 (%) and Y2 (%) of the removal efficiency of H₂S and CO₂, respectively. Experimental results are listed in Table 3.

The statistical model and the factors conducted by an analysis of variance (ANOVA) are listed in Table 4.

The analysis results listed in Table 4 show that this model is completely compatible with practical experiments; thus, the model is convergent. This is proved with the standard F (Fisher) of a valuable model with Y1 (32.74) and Y2 (14.06). This model is statistically significant with high reliability with all P values (<0.0001).

The fitting of the experimental modeling is also verified by the multiple correlation coefficient R^2 . The closer R^2 value is 1, the nearer the experimental value is the predicted value of the model. According to the analysis in shown in Table 4, the multiple correlation coefficients of the two models Y1 and Y2 are 0.9704 (97.04%) and 0.333 (93.36%), respectively. Besides, adj- R^2 values of Y1 and Y2 are 0.9407

Table 2: The experimental levels of technological variables

Real variable	Varied code	Variable range (Δ)	Research level		
			−1	0	1
Rotating speed (rpm)	A	300	900	1,200	1,500
Liquid flow rate Q_L (L/min)	B	0.042	0.083	0.125	0.167
Gas flow rate Q_G (L/min)	C	1.25	1.25	2.5	3.75
pH	D	1	11	12	13

Table 3: Experimental planning matrix of the removal efficiency of H₂S and CO₂

No.	Varied code				Objective functions	
	A	B	C	D	Y1 (%)	Y2 (%)
1	-1	-1	0	0	72.70 ± 0.17	54.31 ± 0.14
2	+1	-1	0	0	74.71 ± 0.15	60.51 ± 0.15
3	-1	+1	0	0	83.53 ± 0.18	66.69 ± 0.14
4	+1	+1	0	0	82.64 ± 0.21	62.74 ± 0.18
5	0	0	-1	-1	81.04 ± 0.11	63.43 ± 0.19
6	0	0	+1	-1	63.52 ± 0.16	49.01 ± 0.15
7	0	0	-1	+1	79.85 ± 0.15	62.77 ± 0.17
8	0	0	+1	+1	81.69 ± 0.22	64.46 ± 0.11
9	-1	0	0	-1	79.11 ± 0.19	60.80 ± 0.17
10	+1	0	0	-1	78.78 ± 0.13	60.91 ± 0.21
11	-1	0	0	+1	86.68 ± 0.14	67.40 ± 0.12
12	+1	0	0	+1	86.97 ± 0.25	71.44 ± 0.14
13	0	-1	-1	0	76.88 ± 0.14	62.02 ± 0.21
14	0	+1	-1	0	75.99 ± 0.22	64.60 ± 0.18
15	0	-1	+1	0	58.18 ± 0.21	40.84 ± 0.21
16	0	+1	+1	0	77.48 ± 0.11	59.16 ± 0.21
17	-1	0	-1	0	73.62 ± 0.12	57.33 ± 0.18
18	+1	0	-1	0	83.56 ± 0.15	65.07 ± 0.11
19	-1	0	+1	0	82.31 ± 0.17	64.10 ± 0.15
20	+1	0	+1	0	74.21 ± 0.17	57.79 ± 0.19
21	0	-1	0	-1	78.28 ± 0.14	60.97 ± 0.13
22	0	+1	0	-1	68.96 ± 0.18	53.70 ± 0.12
23	0	-1	0	+1	66.73 ± 0.22	51.98 ± 0.14
24	0	+1	0	+1	87.00 ± 0.13	67.76 ± 0.16
25	0	0	0	0	94.40 ± 0.23	73.53 ± 0.19
26	0	0	0	0	99.80 ± 0.19	77.72 ± 0.13
27	0	0	0	0	99.74 ± 0.25	77.68 ± 0.25
28	0	0	0	0	99.68 ± 0.23	77.62 ± 0.17
29	0	0	0	0	99.86 ± 0.22	77.86 ± 0.18

(94.07%) and 0.8972 (89.72%), respectively. Adeq. precision values are 20.761 and 13,688, respectively, and both of all are higher than 4. This shows that the value of the objective functions depends largely on the influential variables, and this model is compatible with the real experiments.

After eliminating the nonsignificant variables ($P > 0.05$), the objective functions Y1, Y2 and model are also identified and presented by the second-order regression equation as follows:

$$\begin{aligned}
 Y1 = & 98.69 + 4.01B - 2.80C + 3.27D - 4.51AC \\
 & + 5.05BC + 7.40BD + 4.84CD - 6.76A^2 \\
 & - 13.73B^2 - 13.07C^2 - 9.29D^2,
 \end{aligned} \tag{7}$$

$$\begin{aligned}
 Y2 = & 76.88 + 3.67B - 3.32C + 3.08D - 3.51AC \\
 & + 3.94BC + 5.76BD + 4.03CD - 5.21A^2 \\
 & - 10.69B^2 - 10.03C^2 - 7.02D^2.
 \end{aligned} \tag{8}$$

The influence of linear factors (A, B, C, D) on the value of the objective function is greatest, followed by

Table 4: Regression analysis of the objective functions Y1 and Y2

Source	Y1 (%)		Y2 (%)	
	F-value	P-value	F-value	P-value
Model	32.74	<0.0001*	14.06	<0.0001*
A	0.11	0.7480 ^{NS}	0.5	0.4932 ^{NS}
B	28.79	<0.0001*	15.62	0.0014*
C	13.99	0.0022*	12.82	0.003*
D	19.15	0.0006*	11.04	0.005*
AB	0.32	0.5831 ^{NS}	2.49	0.1366 ^{NS}
AC	12.15	0.0036*	4.78	0.0463*
AD	0.014	0.9059 ^{NS}	0.37	0.5505 ^{NS}
BC	15.20	0.0016*	6.0	0.0281*
BD	32.67	<0.0001*	12.85	0.003*
CD	13.97	0.0022*	6.28	0.0252*
A ²	44.25	<0.0001*	17.05	0.0010*
B ²	182.44	<0.0001*	71.73	<0.0001*
C ²	165.41	<0.0001*	63.13	<0.0001*
D ²	83.50	<0.0001*	30.93	<0.0001*
R ²	0.9704		0.9336	
Adj-R ²	0.9407		0.8972	
Adeq. precision	20.761		13.688	

* $P < 0.05$: meaningful values; ^{NS} $P > 0.05$: non-meaningful values.

the influence of convolution factors (AB, AC, AD, BC, BD, CD), and the least effect on the objective function value is the square factor (A², B², C², D²).

From the regression equation (7), the influence of the factors on the objective function Y1 (removal efficiency of H₂S) is seen. The three factors including B, C, and D affect the Y1 function. Factor A shows a weaker level of influence through convolutional and squared interactions (AC, A²). In which, the level of influence of three technological factors in descending order is B > D > C. Two factors B and D positively affect (positive interaction) Y1 and factor C inversely affects (negative interactions) Y1 corresponding to their coefficients in the regression equation (7).

The regression equation (8) also shows impacts of experimental parameters on the objective function Y2 (removal efficiency of CO₂). Specially, parameters that influence the removal efficiency of CO₂ are AC and A². The indirect influence of factor A into the response is also weak, demonstrated by the low magnitude of AC and A². Based on the coefficients of the linear terms, the order of magnitude of experimental factors is as follows B > C > D. Similar to the previous function, the two factors B and D showed positive interactions with Y2 while factor C has a negative impact on the response.

The effect of the double-factor technology interaction on the objective functions is expressed through the response surfaces in Figure 7.

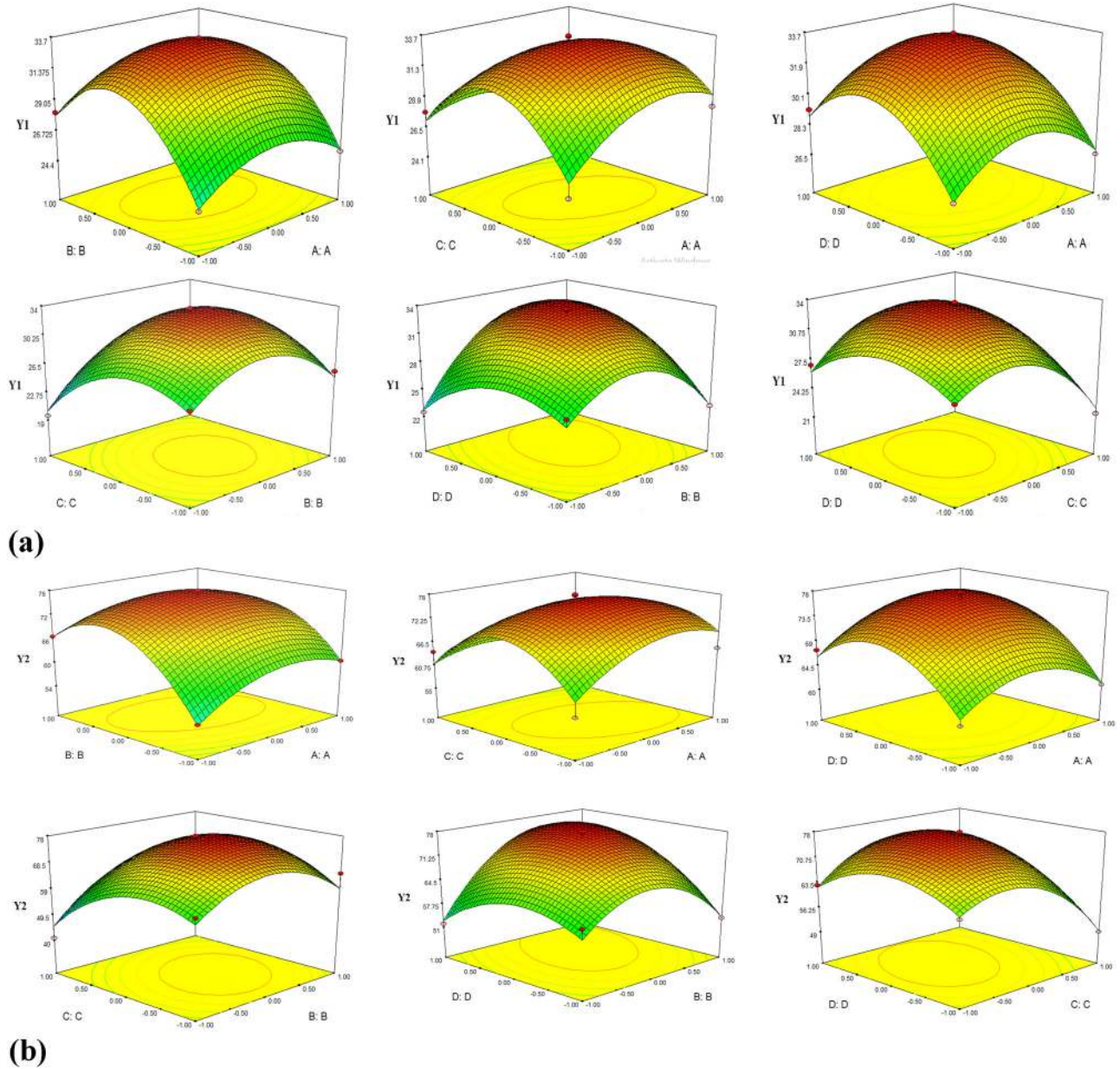


Figure 7: The response surfaces of the objective functions: (a) Y1 corresponding surface and (b) Y2 corresponding surface.

Table 5: Optimization of real experimental variables

Independent variables				Real variables			
A	B	C	D	Rotating speed (rpm)	Q _L (L/min)	Q _G (L/min)	pH
0.02	0.21	-0.03	0.26	1,200	0.134	2.46	12.26

On response surfaces, the dark red zone is the optimal area. Therefore, the objective function values Y1 and Y2 are in the largest value region. From the response surfaces in Figure 7, there are some observations as follows:

With the expressed response surfaces for the function Y1: three pairs of interactive factors (BC, BD, CD) have a greater influence on the objective function than the other three pairs of factors (AB, AC, AD). In three pairs of factors (BC, BD, CD), the order of strong effect on the

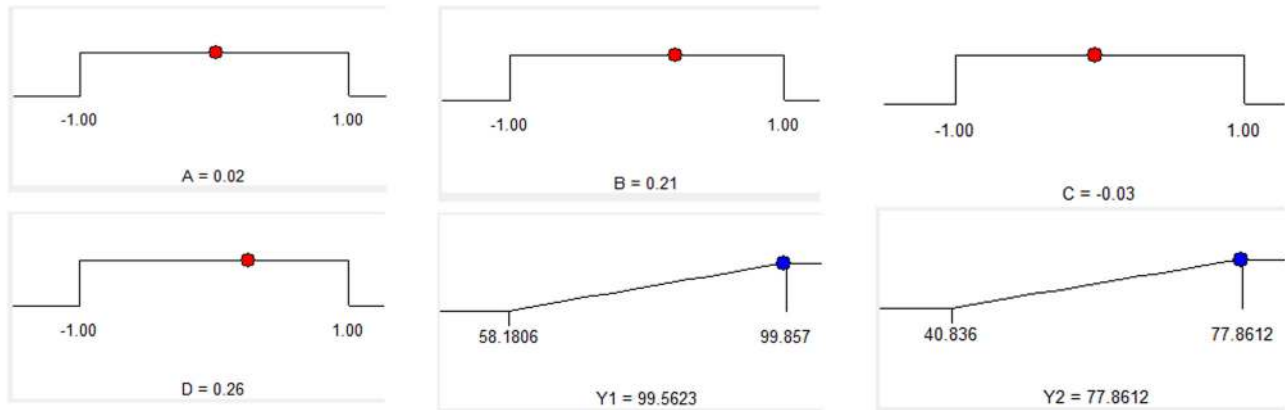


Figure 8: Optimum conditions by the solution of ramps.

Table 6: Comparison in experimental and modeling data at the optimal conditions

Experimental factors				The objective function	Result	
Rotating speed (rpm)	Q_L (L/min)	Q_G (L/min)	pH		Experiment	Predicted
1,200	0.134	2.46	12.26	Y1 (%)	99.38 ± 0.56	99.56
				Y2 (%)	77.28 ± 0.41	77.86

objective function Y1 is $BD > CD > BC$. This is consistent with the results shown in the regression equation (1).

Similar to the Y1 function, the response surfaces for the function Y2 show that three pairs of interaction factors (BC, BD, CD) have a greater influence on the objective function than the three remaining pairs of factors (AB, AC, AD). In three pairs of elements (BC, BD, CD), the order of strong impact on the objective function Y2 is $BD > CD > BC$. This is fitted with the results shown in the regression equation (2).

3.6 Optimization and model verification

The highest removal efficiency of H_2S and CO_2 is expected to be optimized, so both the objective functions Y1 and Y2 need to reach the maximum values. This is solved by using Design-Expert 7.0.0 software for resolution according to the method of aspiration with priority levels (from 1 to 5). In this way, with the goals, the priority levels for the objective functions are selected as follows:

Objective function Y1 (level 5)

Objective function Y2 (level 5)

The optimized results by Design-Expert 7.0.0 software gives out a resolution corresponding to a set of technological data. The optimal data of real experimental variables are shown in Table 5 and Figure 8. In terms of

real variables, the predicted values of the objective functions are $Y1 = 99.56$ (%) and $Y2 = 77.86$ (%) by modeling (Table 6).

Experiments are repeated thrice with the set of the technological factors at the optimal conditions including rotating speed 1,200 (rpm), Q_L 0.134 (L/min), Q_G 2.46 (L/min), and solution pH of 12.26. The test results are given in Table 6.

The results in Table 6 show that the experimental results at optimal conditions are nearly equal to the predicted values of the objective function in the predicted result. Therefore, the model of optimal calculation is consistent with the experimental factors.

4 Conclusions

This work applied high-gravity rotating packed bed, a novel apparatus, to eliminate H_2S and CO_2 in biogas and investigated the effects of some parameters including absorbent solution pH, rotating speed, and gas-liquid flow rate on the removal efficiencies. The obtained experimental results indicated that the optimal removal efficiencies of H_2S and CO_2 could be achieved at a pH of 12.26 for $Ca(OH)_2$ absorbent solution, rotating speed of HGRPB of 1,200 (rpm), gas flow rate of 2.46 (L/min),

and liquid flow rate of 0.134 (L/min). Referring to some previous works, current removal efficiencies are relatively higher, which suggest the potential of the high-gravity rotating packed bed reactor for the removal of H₂S and CO₂ in biogas. Results are approved by the optimal conditions in experiments and simulation with calculated modeling.

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Data availability statement: The datasets generated during and/or analysed during the current study are available from the corresponding author on reasonable request.

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