



Modelling and Optimisation of High Pressure Water Scrubbing of Biogas for CO₂ removal using Response Surface Methodology and Artificial Neural Networks

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ABSTRACT

Biogas produced from anaerobic digestion of organic wastes consists of mainly methane (CH₄) and undesirable gases like carbon dioxide (CO₂), hydrogen sulphide (H₂S) and other trace gases. This study focused on the modelling and optimisation of biogas upgrading using high pressure water scrubbing (HPWS). The biogas upgrading process was simulated using Aspen HYSYS while the experiments were designed using Design Expert software. Response surface methodology (RSM) and artificial neural network (ANN) was adopted to model and optimise the process. The four factors considered were absorber pressure, biogas flowrate, and water flow rate and absorber temperature. RSM analysis yielded a quadratic model for predicting the chosen responses (CH₄ recovered and CO₂ captured) as a function of the four independent variables. For ANN, a multilayer full feed forward neural network trained with the Levenberg-Marquardt algorithm was found suitable for modelling the upgrading process. Both the RSM and ANN models described the upgrading process (methane recovery) with high accuracy as indicated by their high R² (0.9826 and 0.9999), low RMSE (1.3937 and 0.1200) and low AAD (0.0172 and 0.0006) values respectively. Optimisation results showed that up to 93% CH₄ recovery and 87% CO₂ capture was achieved using HPWS and this was obtained at a condition of absorber pressure (18 bar), biogas flowrate (7 m³/h), water flow rate (148 m³/h) and absorber temperature (11 °C). When compared, the ANN model performed better in predicting the responses than RSM.

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INTRODUCTION

Biogas is produced by anaerobic decomposition of organic substances, a process that is catalysed by a consortium of microorganisms (Zhang et al., 2017). Biogas consists mainly of methane (CH₄), carbon dioxide (CO₂), hydrogen sulphide (H₂S) and other gases such as water

vapour, ammonia and nitrogen (Ajeh et al., 2016; Malinauskaite et al., 2017).

In its purified form, biogas can be used as a substitute for natural gas and be compressed and injected into the national gas pipeline network, thus becoming a valuable renewable energy source (Maria, 2008). However, the presence of undesirable components like



carbon dioxide, water vapour, ammonia, hydrogen sulphide etc in biogas reduces its heating value and could potentially cause corrosion problems in the gas pipelines. The presence of these impurities in biogas can also potentially increase compression and transportation costs (Abdeen et al., 2016). This thus limits the application of biogas for power generation (Awe et al., 2017).

Thus, it is necessary to remove the impurities in biogas to improve its calorific value and enhance its fuel properties through an upgrading process (Sun et al., 2015). Currently, biogas upgrading strategies developed for industrial-scale operation include adsorption, chemical absorption, membrane separation and cryogenic processes (Khan et al., 2017). These methods are characterised by certain significant demerits such as high cost of deployment and environmental unfriendliness (IEA 2000).

However, high pressure water scrubbing has emerged as the most common strategy for biogas upgrading. This process is built on the basis of the fact that the impurity gases are more soluble in water than methane. For this process, water is used as a scrubbing solvent and the process takes place in a packed column in which water and biogas are contacted counter-currently (Pettersson and Wellinger, 2009). Methane purity of as much as 80-99% have been recorded for biogas upgraded with high pressure water scrubbing pointing to the efficiency of the process (Cozma et al., 2015). The process is also accompanied by very little methane loss (3-5%) originating from the dissolution of methane in water.

The performance of the HPWS upgrading process has been reported to be dependent on a number of process variables such as pressure, temperature, composition, gas and liquid flowrate (Cozma et al., 2015; Kapoor et al., 2017). The efficiency of the process can be

improved by optimizing these variables to give maximum methane recovery and carbon dioxide removal. This can be achieved by deploying techniques like design of experiment for response surface methodology (RSM) which is an empirical statistical tool for modelling and optimising multivariable processes (Amenaghawon et al., 2015). Recently, machine learning tools like artificial neural networks (ANN) have been deployed for modelling chemical engineering processes as a result of some of the limitations of RSM (Bas, and Boyaci, 2007). ANN is particularly useful when it is not possible to deploy conventional empirical models or when such models are inadequate in describing real life systems (Velu et al., 2016).

Thus, the aim of this study was to model and optimise high pressure water scrubbing of biogas for improved methane composition using RSM and ANN.

METHODOLOGY

Description of Biogas Upgrading Process

High pressure water scrubbing was adopted to upgrade the biogas produced in order to improve its fuel properties. The primary basis for achieving this was to remove the major impurities such as carbon dioxide and hydrogen sulphide. The description of the procedures for the production and characterisation of the biogas has been previously reported (Ajieh et al., 2018).

A simple process flow diagram of the water scrubbing process is shown in Figure 1. The basis of separation of the impurities is the difference in their solubility in water. When compared to CH₄, CO₂ and H₂S are relatively more soluble in water, hence they are preferentially separated from raw biogas. A high column pressure is usually used for this process to facilitate the separation process by increasing the driving force of solubility

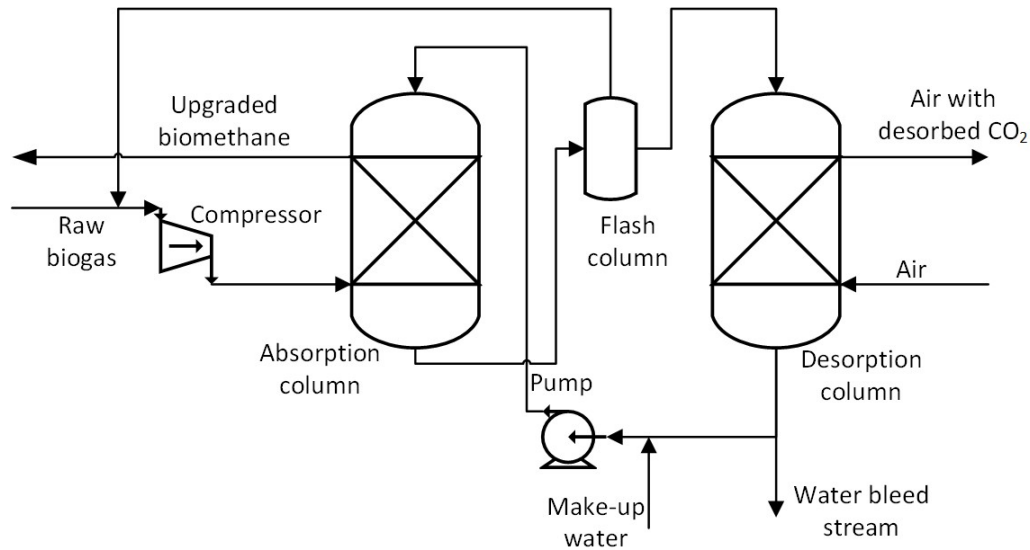


Figure 1: Process flow for water scrubbing process

Design of Experiment

In this study, a four variable Box-Behnken design (BBD) was used to plan the biogas upgrading experiments. The BBD was chosen for this purpose because it is a very useful experimental design method and it is very suitable for fitting quadratic response surfaces, which are mostly encountered in many nonlinear processes in chemical engineering (Foroughi et al., 2018). For this work, the Design Expert® software version 7.0.0, (Stat-ease, Inc. Minneapolis, USA) was used to design the experiments and analyse the data obtained. The factors considered for optimisation are shown in Table 1. The range of these factors was selected based on previous studies and the operating conditions of real life biogas upgrading plants. The values of the factors were calculated using Equation 1.

$$Y = b_0 + \sum_{i=1}^N b_i X_i + \sum_{i,j=1}^N b_{ij} X_i X_j + \sum_{i=1}^N b_{ii} X_i^2 + \sum_{i=1}^N e_i \quad (2)$$

The optimisation of process variables and experimental conditions using RSM was

$$x_i = \frac{X_i - X_0}{\Delta X_i} \quad (1)$$

X_i and x_i are the actual and coded values of the factors respectively while X_0 is the actual value of the factors at the centre point, and ΔX_i is the step change in the value of the actual values of the factors.

Equation 2 is a quadratic response model which was used to fit the experimental data and this was achieved by using multiple regression analysis to estimate the values of the coefficients of the model. The second order model was chosen because it is the most widely used model for response surface methodology is the most commonly used model for modelling real life processes (Asfaram et al., 2016). Analysis of variance (ANOVA) was then used to assess the quality and significance of the model.

through numerical optimisation methods (Myers et al., 2009).

Table 1: Coded and actual level of factors for HPWS

Factors	Unit	Symbols	Actual and Coded Levels		
			-1	0	1
Absorber pressure	Bar	X ₁	5	12.5	20
Biogas flowrate	m ³ /h	X ₂	7	9.5	12
Water flow rate	m ³ /h	X ₃	50	125	200
Absorber temperature	°C	X ₄	5	22.5	40

Artificial Neural Network Modeling

For this study, artificial neural networks used to model the responses representing the biogas upgrading process (CH₄ recovered and CO₂ captured). To achieve this, a commercially available ANN Software, NeuralPower, version 2.5 (C.P.C-X Software USA) was used. In order to choose the most appropriate network architecture, two options were evaluated and these are multilayer full feed forward (MFFF) and a multilayer normal feed forward (MNFF) neural networks. A number of training algorithms were also evaluated to train the networks. The two networks evaluated were trained using algorithms such as incremental back propagation (IBP), quick propagation (QP), generic algorithm (GA), batch back propagation (BBP), and Levenberg- Marquardt (LM) algorithm. The most suitable transfer function for the hidden and output layer was chosen amongst the sigmoid, hyperbolic tangent, Gaussian, linear and threshold linear. The optimum number of neurons needed for the hidden layer of the network was determined following an iterative process. For this process, 70% of the experimental data was used to train

the networks, 15% was used for validation while the remaining 15% was used for testing the network. The selection of the best network architecture, training algorithm, best transfer function and optimum number of neurons was done on the basis of the root mean square error (RMSE) and coefficient of determination (R²). The best alternative was chosen to be the one with the highest R² value and smallest RMSE value (Betiku et al., 2014).

RSM and ANN Data Verification

The capacity of RSM and ANN to predict the responses was evaluated by comparing the results predicted by the RSM and ANN models with those of the actual experiments. The level of fit between the RSM and the ANN was assessed by using the coefficient of determination (R² value), absolute average deviation (AAD) and root mean square error (RMSE), as defined in Equations 3 to 5 (Amenaghawon and Amagbewan, 2017). It is desirable that the R² value be as close to unity as possible while the RMSE and AAD should be as small as possible (Yi et al., 2009).

$$R^2 = 1 - \frac{\sum_{i=1}^n (x_{a,i} - x_{p,i})^2}{\sum_{i=1}^n (x_{p,i} - x_{a,ave})^2} \quad (3)$$

$$RMSE = \sqrt{\frac{1}{n} \sum_{i=1}^n (x_{p,i} - x_{a,i})^2} \quad (4)$$

$$AAD = \frac{1}{n} \left(\sum_{i=1}^n \left(\frac{|(x_{a,i} - x_{p,i})|}{x_{a,i}} \right) \right) \times 100 \quad (5)$$

Where n is the number of experimental data, x_{p,i} is the predicted

value, $x_{a,i}$ is the experimental values, $x_{a,ave}$ is the average experimental values, and $x_{p,ave}$ is the average predicted values.

HYSYS Simulation of the HPWS Process

The Aspen HYSYS software version 2006 was used to carry out all the simulation reported in this work. For this study, the components added for the water scrubbing process were methane, (main component in raw biogas), carbon dioxide, hydrogen sulphide and water. The selection of the components was on the basis of the composition of the actual biogas produced as reported in Ajieh et al. (2018). After creating the Component List, it was then associated with a Fluid Package. It is necessary to select the appropriate Fluid Package because the Fluid Package contains the thermodynamic models that are used by the Aspen HYSYS software to carry out vapour liquid equilibrium calculations (AspenTech 2017).

For the base case simulation, the raw biogas was compressed (in Comp. 1) to increase its pressure from 1 bar to about 2 bar. The composition of the raw biogas feed was determined through chromatographic analysis of the biogas produced from the anaerobic digestion of the organic waste feedstock as reported in Ajieh et al. (2018). After compressing

the raw biogas to 2 bar, it was then cooled and further compressed and cooled in order to take it to the operating pressure of the absorber. The compressed raw biogas was fed to the absorption column (Absorber) from the bottom and it was made to contact the scrubbing water in a counter current manner (Figures 2 and 4). During this process, the water absorbs the CO_2 in the raw biogas and the upgraded biogas that is essentially methane (biomethane) is collected from the top of the absorption column. The water containing the absorbed impurities was sent to a flash drum (Flash separator) where the pressure was suddenly reduced to effect flash vapourisation. The gas stream from the flash separator was combined with the raw biogas at the start of the process using the mixer. The liquid stream from the flash separator which is mainly rich in CO_2 was then fed to the desorption column (stripper) and the absorbed CO_2 was removed using counter current flow of air at atmospheric pressure (Figures 3 and 4). The regenerated water was then recirculated back to the absorber. For the simulation process, some operating variables of the absorber were varied to determine the effect on methane recovery and CO_2 capture and this was done using the experimental design.

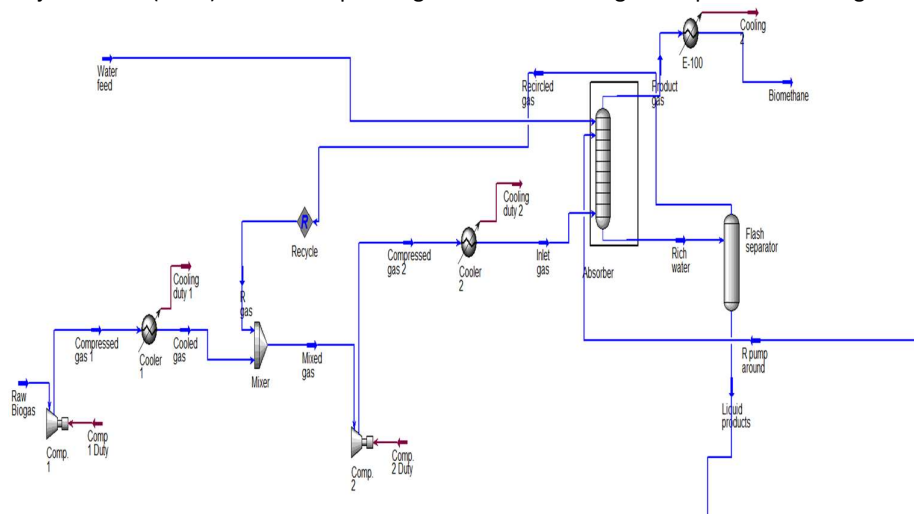


Figure 2: HYSYS process flow diagram for HPWS (absorber portion)

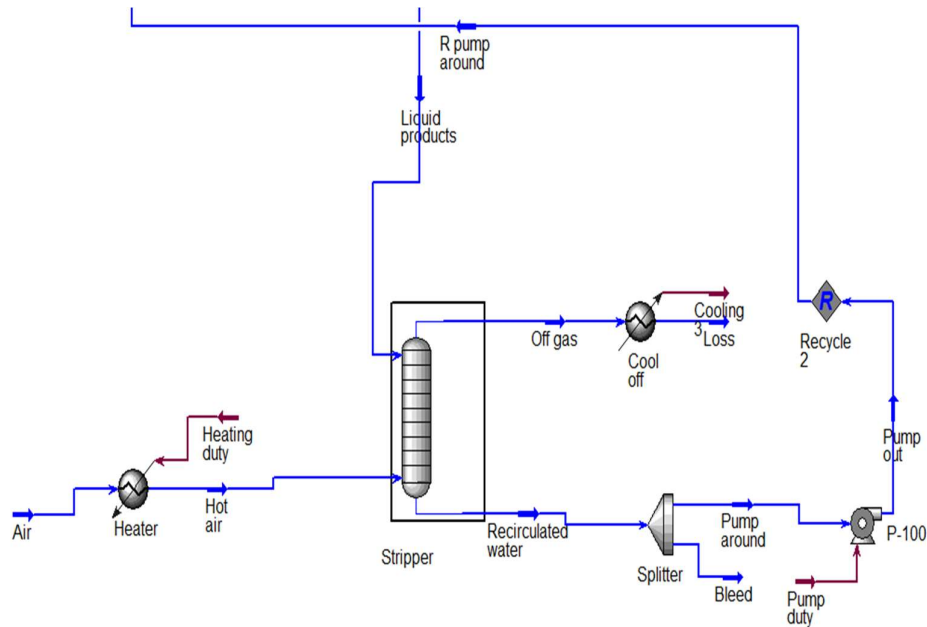


Figure 3: HYSYS process flow diagram for HPWS (stripper portion)

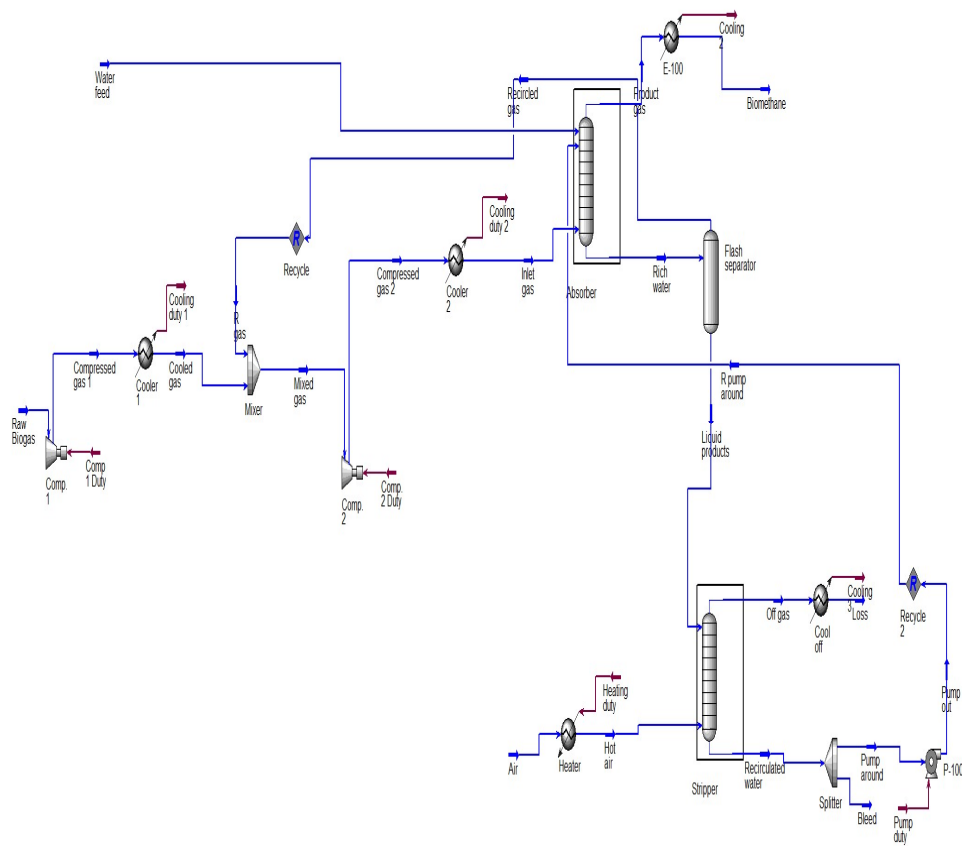


Figure 4: HYSYS process flow diagram for HPWS (full process flow diagram)

Validation of HYSYS Thermodynamic Model

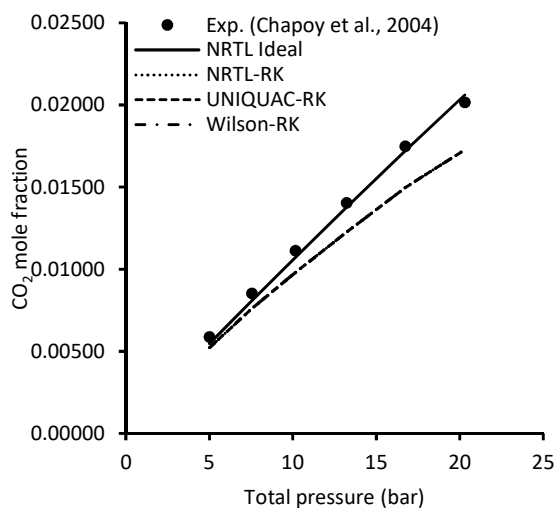
For this work, different thermodynamic models were evaluated to determine their suitability for the intended purpose. This was done because if the wrong thermodynamic model is chosen, it could lead to incorrect simulation results. The accuracy of the simulation results is tied to the selection of the appropriate thermodynamic model (Cozma et al., 2013). The models investigated were NRTL-Ideal (Non-Random-Two-Liquid with ideal gas and Henry's law), NRTL-RK (Non-Random-Two-Liquid/Redlich-Kwong equation of state with Henry's law), UNIQUAC-RK (Universal Quasi Chemical/Redlich-Kwong equation of state with Henry's law) and Wilson. These different models were validated using experimental data reported in the literature (Chapoy et al., 2004; Valtz et al., 2004). The thermodynamic model deemed to be most appropriate for the intended purpose was the one that showed the

best fit with the experimental data. The most appropriate thermodynamic model was thus chosen to run all the simulations reported in this work.

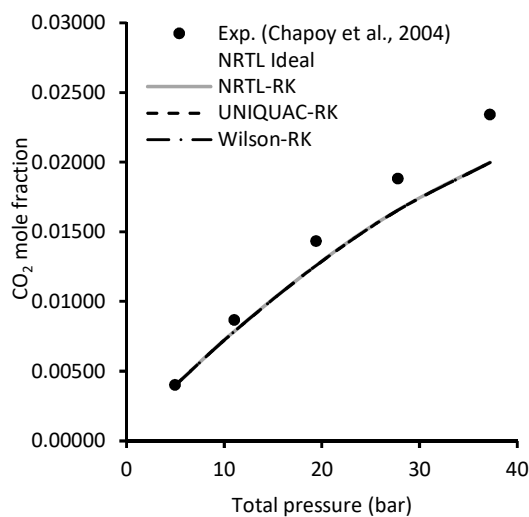
RESULTS AND DISCUSSION

Validation of HYSYS Model

The applicability of the thermodynamic models in the HYSYS software was evaluated by comparing the model results with the available experimental literature data for CO₂ and CH₄ solubility in solution (Chapoy et al., 2004; Valtz et al., 2004). The results are shown in Figures 5 and 6. Figures 5 and 6 for CO₂ and CH₄ respectively show the comparison of the experimental data with the HYSYS models (NRTL-Ideal, NRTL-RK, UNIQUAC-RK, and Wilson). After comparing the results obtained for all four thermodynamic models, it was found that the NRTL-Ideal (Non-Random-Two-Liquid with ideal gas and Henry's law) model best fitted the experimental data and was thus used for the simulations done in the remainder of this work.



(a)



(b)

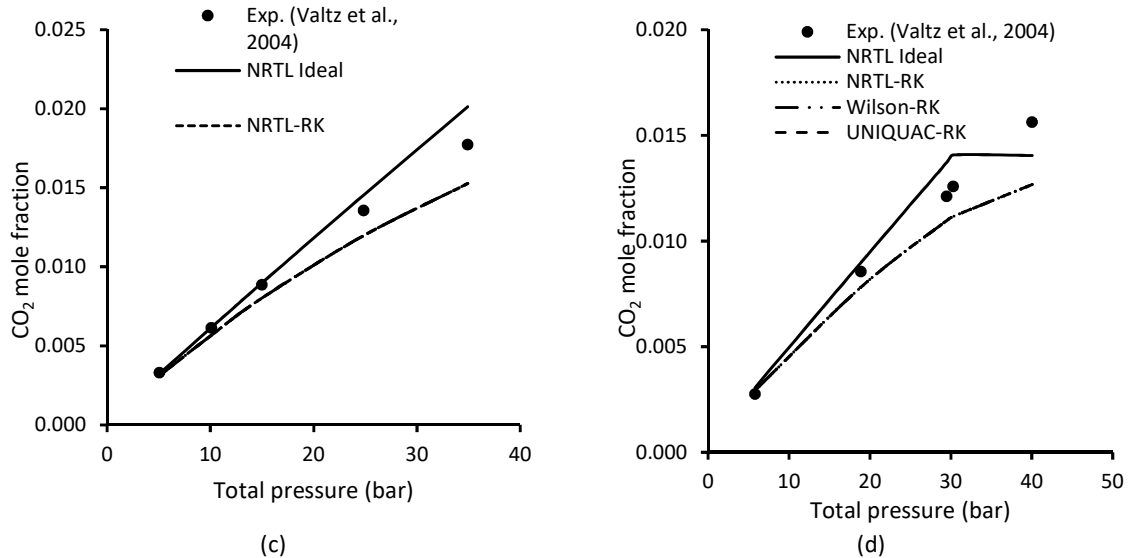
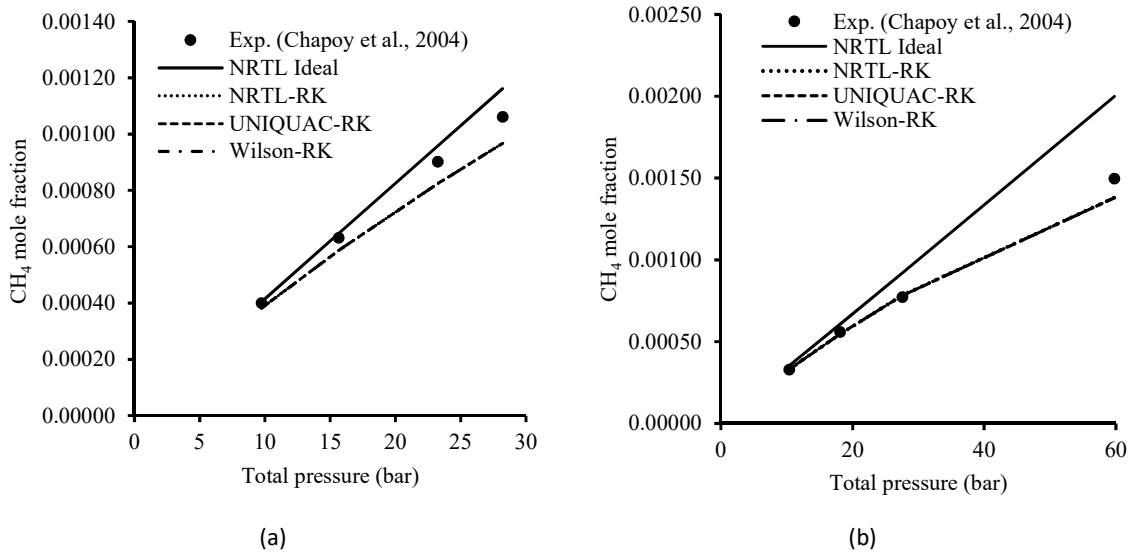


Figure 5: Comparison of model and experimental results of CO₂ solubility at (a) 5 °C (b) 15 °C (c) 25 °C (d) 35 °C



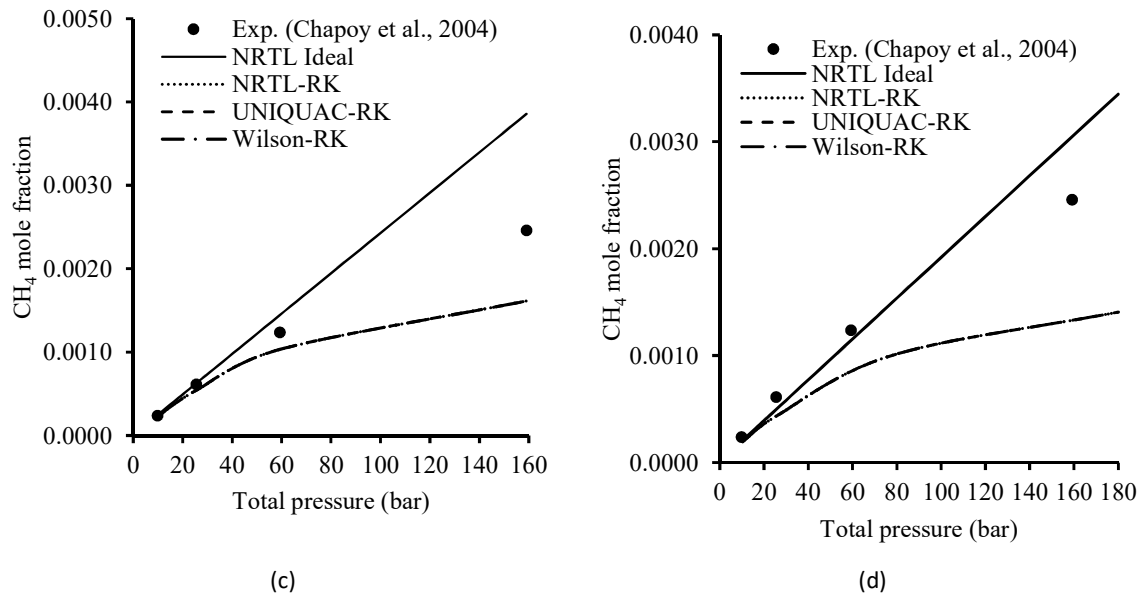


Figure 6: Comparison of model and experiment results for CH₄ solubility at (a) 2 °C (b) 10 °C (c) 25 °C (d) 40 °C

Statistical Analysis of Models

The quadratic model was fitted to the data generated from the HYSYS simulations in order to estimate the model parameters. A total of 29 experimental runs were obtained by the Box-Behnken design for this purpose. Multiple regression analysis was used for the fitting process and the model parameters were estimated to obtain the final model equations in terms of actual experimental factors. The model equations for the two responses are shown in Equations 6 and 7. The equations represent CH₄ recovered and

CO₂ captured as a function of absorber pressure (X_1), gas flow rate (X_2), liquid flowrate (X_3) and absorber temperature (X_4). These equations were used to predict their corresponding responses and the results are shown in Table 2. Comparison of the experimental values (HYSYS simulation) of the response and those predicted by the statistical models showed that there was an acceptable level of fit between the two as seen in the fact that both set of values were very close.

$$CH_4 \text{ recovered} = 63.45 + 3.56X_1 - 5.30X_2 + 0.34X_3 - 0.16X_4 + 0.034X_1X_2 - 0.016X_1X_3 - 0.011X_2X_3 + 0.0012X_2X_4 + 0.0018X_3X_4 - 0.057X_1^2 + 0.16X_2^2 - 0.00080X_3^2 + 0.0029X_4^2 \quad (6)$$

$$CO_2 \text{ captured} = 63.45 + 3.56X_1 - 5.30X_2 + 0.35X_3 - 0.17X_4 + 0.034X_1X_2 - 0.016X_1X_3 - 0.0000092X_1X_4 + 0.011X_2X_3 + 0.0012X_2X_4 + 0.00018X_3X_4 - 0.057X_1^2 + 0.16X_2^2 - 0.00080X_3^2 + 0.0029X_4^2 \quad (7)$$



Table 2: Experimental and RSM predicted results

Run	Factors								Responses			
	Coded values				Actual values				CH ₄ recovered (%)		CO ₂ captured (%)	
	X ₁	X ₂	X ₃	X ₄	X ₁	X ₂	X ₃	X ₄	Exp.	RSM	Exp.	RSM
1	0	0	-1	-1	12.5	9.5	50	5.0	73.7	73.6	81.7	77.6
2	0	1	0	-1	12.5	12.0	125	5.0	77.5	78.8	85.4	86.7
3	1	-1	0	0	20.0	7.0	125	22.5	88.9	91.0	85.7	87.7
4	-1	0	1	0	5.0	9.5	200	22.5	64.7	64.6	84.9	87.7
5	-1	-1	0	0	5.0	7.0	125	22.5	65.2	67.5	85.4	81.2
6	0	-1	1	0	12.5	7.0	200	22.5	89.4	88.7	85.7	85.9
7	0	0	0	0	12.5	9.5	125	22.5	81.7	81.2	85.9	85.7
8	0	0	-1	1	12.5	9.5	50	40.0	73.4	74.4	80.8	77.0
9	1	0	0	-1	20.0	9.5	125	5.0	85.1	85.7	85.7	87.3
10	0	1	1	0	12.5	12.0	200	22.5	80.7	80.0	85.7	88.2
11	0	0	1	1	12.5	9.5	200	40.0	82.9	84.0	85.7	87.1
12	1	1	0	0	20.0	12.0	125	22.5	85.1	83.7	85.7	87.1
13	-1	1	0	0	5.0	12.0	125	22.5	64.0	62.8	82.8	78.0
14	0	0	0	0	12.5	9.5	125	22.5	81.1	81.2	85.7	85.7
15	0	0	0	0	12.5	9.5	125	22.5	81.1	81.2	85.7	85.7
16	1	0	-1	0	20.0	9.5	50	22.5	78.4	77.2	84.5	85.8
17	1	0	1	0	20.0	9.5	200	22.5	89.8	90.3	85.7	77.9
18	0	1	0	1	12.5	12.0	125	40.0	79.3	80.3	85.6	86.6
19	1	0	0	1	20.0	9.5	125	40.0	88.3	87.8	85.7	87.2
20	0	1	-1	0	12.5	12.0	50	22.5	71.9	73.0	75.8	74.3
21	-1	0	0	1	5.0	9.5	125	40.0	64.2	64.0	82.3	79.5
22	-1	0	0	-1	5.0	9.5	125	5.0	64.2	65.0	82.4	79.6
23	0	0	0	0	12.5	9.5	125	22.5	81.1	81.2	85.7	85.7
24	0	-1	-1	0	12.5	7.0	50	22.5	75.3	76.3	84.3	80.5
25	0	0	1	-1	12.5	9.5	200	5.0	83.7	83.5	85.7	86.7
26	0	0	0	0	12.5	9.5	125	22.5	81.1	81.2	85.7	85.7
27	0	-1	0	1	12.5	7.0	125	40.0	87.9	85.4	85.7	88.5
28	-1	0	-1	0	5.0	9.5	50	22.5	60.2	58.4	48.8	60.6
29	0	-1	0	-1	12.5	7.0	125	5.0	87.9	85.7	85.7	88.7

Analysis of Variance of Models

Analysis of variance (ANOVA) was carried out to assess the statistical significance and fit of the models representing the responses and the

results are shown in Tables 3 and 4. Tables 3 and 4 show the ANOVA results for the models representing CH₄ recovered and CO₂ captured for the high-pressure water scrubbing process. For



these results, model terms having p values less than 0.05 are significant. The implication of the statistical significance of a model is that changes in the values of the actual physical factor represented by that model term could potentially significantly affect the response under consideration. Conversely, model terms with p value greater than 0.05 are considered not to be significant and this also means that changes in the values of the actual physical factor represented by that model term does not significantly affect the response under consideration (Asfaram et al., 2016). The model terms representing absorber pressure (X_1), biogas flowrate (X_2), water flowrate (X_3)

were significant in predicting both CH₄ recovered and CO₂ captured as shown in Tables 3 and 4. Considering the results presented in Tables 3 and 4, it was observed that the "model p value" was in most cases < 0.0001. This indicates that the statistical models chosen to represent the responses for were very significant, meaning that the models are very useful for predicting the responses (Yi et al., 2009). Furthermore, the "lack of fit" p value for all the response models were greater than 0.05 indicating that the lack of fit was not significant. Significant lack of fit is not desirable as it implies that the model does not fit the experimental data (Myers et al., 2009).

Table 3: ANOVA results for model representing CH₄ recovered

Source	Sum of squares	Degree of freedom	Mean square	F value	p value
Model	2194.48	14	156.75	56.33	< 0.0001
X_1	1483.13	1	1483.13	532.99	< 0.0001
X_2	108.67	1	108.67	39.05	< 0.0001
X_3	283.92	1	283.92	102.03	< 0.0001
X_4	1.18	1	1.18	0.43	0.5248
X_1X_2	1.73	1	1.73	0.62	0.4432
X_1X_3	11.94	1	11.94	4.29	0.0573
X_1X_4	2.45	1	2.45	0.88	0.3636
X_2X_3	6.96	1	6.96	2.50	0.1361
X_2X_4	0.78	1	0.78	0.28	0.6050
X_3X_4	0.044	1	0.044	0.016	0.9014
X_1^2	226.22	1	226.22	81.29	< 0.0001
X_2^2	6.24	1	6.24	2.24	0.1564
X_3^2	45.69	1	45.69	16.42	0.0012
X_4^2	0.82	1	0.82	0.29	0.5965
Residual	38.96	14	2.78		
Lack of fit	38.67	10	3.87	54.64	0.0801
Pure error	0.28	4	0.071		
Cor total	2233.44	28			

Table 4: ANOVA results for model representing CO₂ captured

Source	Sum of squares	Degree of freedom	Mean square	F value	p value
Model	1021.38	14	72.96	2.91	0.0275
X_1	180.22	1	180.22	7.18	0.0179



Source	Sum of squares	Degree of freedom	Mean square	F value	p value
X ₂	11.13	1	11.13	0.44	0.5162
X ₃	277.33	1	277.33	11.06	0.0050
X ₄	0.048	1	0.048	1.91E-03	0.9658
X ₁ X ₂	1.66	1	1.66	0.066	0.8007
X ₁ X ₃	305.00	1	305.00	12.16	0.0036
X ₁ X ₄	5.87E-06	1	5.87E-06	2.34E-07	0.9996
X ₂ X ₃	18.04	1	18.04	0.72	0.4107
X ₂ X ₄	0.012	1	0.012	4.59E-04	0.9832
X ₃ X ₄	0.23	1	0.23	9.36E-03	0.9243
X ₁ ²	67.54	1	67.54	2.69	0.1231
X ₂ ²	6.49	1	6.49	0.26	0.6189
X ₃ ²	132.49	1	132.49	5.28	0.0375
X ₄ ²	5.04	1	5.04	0.20	0.6608
Residual	351.16	14	25.08		
Lack of fit	351.12	10	35.11	3149.34	0.1011
Pure error	0.045	4	0.011		
Cor total	1372.54	28			

The level of fit between the experimental data and the models were assessed using other statistical parameters such as coefficient of determination, coefficient of variation, standard deviation, adequate precision etc as shown in Table 5. As seen from the results presented in Table 5, all the models were characterised by high R² values indicating very good fit between the experimental observations and model predictions. Furthermore, the adjusted R² values obtained were within reasonable agreement with the corresponding R² values further confirming the fit of the models (Yi et al., 2009). The values of standard deviation were small compared to the mean of the observations showing that there was minimal dispersion of each

individual observation about the mean (Myers et al., 2009). This further confirmed the significant fit of the models. The coefficient of variation (CV) is the standard deviation expressed as a percentage of the mean. It is a statistical parameter used to assess the reliability and repeatability of the experiments. Low CV values like those presented in Table 5 usually indicate that the experimental runs are reliable and repeatable. The adequate precision is a measure of the signal to noise ratio. Values greater than 4 are usually recommended. The values obtained as shown in Table 5 were all greater than 4 and this is an indication of adequate signal and the models can thus be used to navigate the design space (Myers et al., 2009).

Table 6: Goodness of fit statistics for RSM models

Response	R ²	Adjusted R ²	Mean	Standard deviation	CV %	Adeq. precision
CH ₄ recovered	0.9826	0.9651	78.20	1.67	2.13	27.23
CO ₂ captured	0.7442	0.6552	83.31	5.01	6.01	7.81



Artificial Neural Network Modelling

The training algorithm for a neural network is unique to every process (Dorofki et al., 2012). Similarly, it is also not possible to determine a priori, the algorithm that will be most suitable for training a network. Thus, there was the need to iteratively investigate several training algorithms with the aim of determining the one that is most suitable for the process. The results presented in

Table 7 shows that a multilayer full feed forward neural network trained with Levenberg-Marquardt learning algorithm was the best network architecture for predicting for the upgrading process. This was chosen because it had the highest R^2 value (0.9999 and 1.0000) and the lowest RMSE value (0.1014 and 0.0338) respectively for CH_4 recovery and CO_2 capture.

Table 7: ANN training algorithm for predicting CH_4 recovered and CO_2 captured

Network architecture	Training algorithm	CH_4 recovered		CO_2 captured	
		R^2	RMSE	R^2	RMSE
MNFF	IBP	0.9996	0.1866	0.9996	0.1437
	BBP	0.9981	0.3897	0.9998	0.0883
	QP	0.9963	0.5421	0.9991	0.2150
	GA	0.9947	0.6487	0.9965	0.4114
	LM	0.9999	0.1061	1.0000	0.0338
MFFF	IBP	0.9988	0.3152	0.9996	0.1478
	BBP	0.9995	0.1982	0.9999	0.0541
	QP	0.9801	1.251	0.9995	0.1557
	GA	0.9971	0.4776	0.9557	1.4729
	LM*	0.9999	0.1014	1.0000	0.0338

*Best learning algorithm and network

An iterative process was also used to determine the optimum number of neurons in the network. Different neural networks with different number of neurons were investigated and the results are shown in Table 8. As can be seen from the results obtained, the R^2 value increased as the number of neurons was increased from 1 to 4. The RMSE of both responses was observed to decrease as the number of neurons was increased from 1 to 4. This is an indication that the predictive capability of the network was increasing. However, increasing the number of neurons beyond 4 did not result in any change in the R^2 and RMSE values showing that the predictive capability of the network was not enhanced beyond 4 neurons. Thus, the optimum number of neurons for the

network was chosen as 4. Hence, for a neural network with four input factors, 4 neurons in the hidden layer and 1 factor in the output layer, the corresponding optimum neural networks will be 4-4-1. The network architectures for both responses are represented schematically as shown in Figures 7 and 8 for both responses.

The predicted response levels of CH_4 recovery and CO_2 capture using the optimum neural network architecture and the actual experimental values are presented in Table 9 for comparison. It can be seen from the results presented that the values predicted by the ANN model were very close to the experimental values indicating validity and reliability of the ANN model.

Table 8: Optimum number of neurons

No of neurons	CH ₄ recovered		CO ₂ captured	
	R ²	RMSE	R ²	RMSE
1	0.8973	2.8577	0.9293	1.8600
2	0.9909	0.8498	0.9983	0.2851
3	0.9978	0.4178	0.9988	0.2394
4*	0.9999	0.1014	1.0000	0.0338
5	0.9999	0.1014	1.0000	0.0338

* Optimum number of neurons

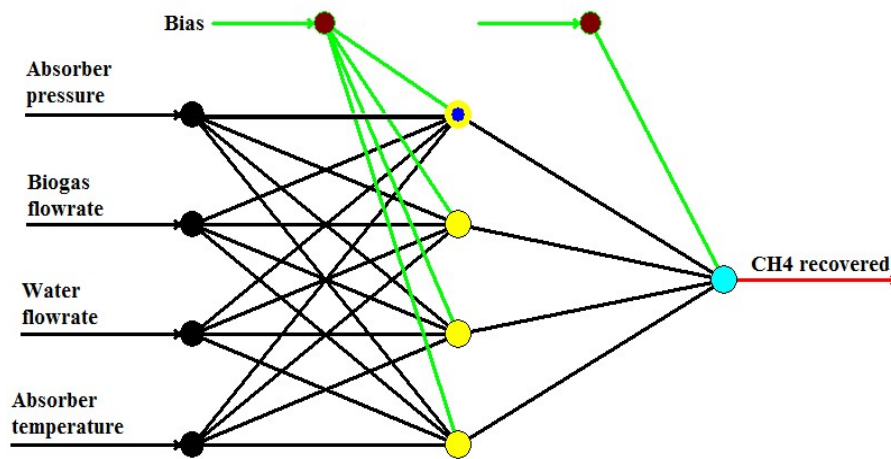


Figure 7: Optimal ANN architecture for predicting CH₄ recovered

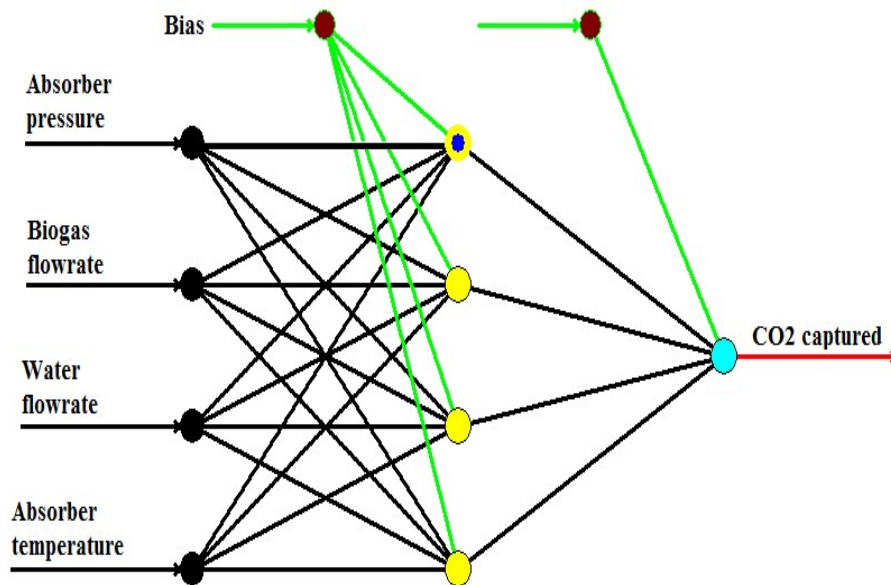


Figure 8: Optimal ANN architecture for predicting CO₂ captured



Table 9: Comparison of experimental results with ANN predicted results

Run	Factors								Responses			
	Coded values				Actual values				CH ₄ recovered (%)		CO ₂ captured (%)	
	X ₁	X ₂	X ₃	X ₄	X ₁	X ₂	X ₃	X ₄	Exp.	RSM	Exp.	RSM
1	0	0	-1	-1	12.5	9.5	50	5.0	73.7	73.7	81.7	81.7
2	0	1	0	-1	12.5	12.0	125	5.0	77.5	77.5	85.4	85.4
3	1	-1	0	0	20.0	7.0	125	22.5	88.9	88.9	85.7	85.7
4	-1	0	1	0	5.0	9.5	200	22.5	64.7	64.7	84.9	84.9
5	-1	-1	0	0	5.0	7.0	125	22.5	65.2	65.2	85.4	85.4
6	0	-1	1	0	12.5	7.0	200	22.5	89.4	89.4	85.7	85.7
7	0	0	0	0	12.5	9.5	125	22.5	81.7	81.2	85.9	85.7
8	0	0	-1	1	12.5	9.5	50	40.0	73.4	73.4	80.8	80.8
9	1	0	0	-1	20.0	9.5	125	5.0	85.1	85.1	85.7	85.7
10	0	1	1	0	12.5	12.0	200	22.5	80.7	80.7	85.7	85.7
11	0	0	1	1	12.5	9.5	200	40.0	82.9	82.9	85.7	85.7
12	1	1	0	0	20.0	12.0	125	22.5	85.1	85.1	85.7	85.7
13	-1	1	0	0	5.0	12.0	125	22.5	64.0	64.0	82.8	82.8
14	0	0	0	0	12.5	9.5	125	22.5	81.1	81.2	85.7	85.7
15	0	0	0	0	12.5	9.5	125	22.5	81.1	81.2	85.7	85.7
16	1	0	-1	0	20.0	9.5	50	22.5	78.4	78.4	84.5	84.5
17	1	0	1	0	20.0	9.5	200	22.5	89.8	89.8	85.7	85.7
18	0	1	0	1	12.5	12.0	125	40.0	79.3	79.3	85.6	85.6
19	1	0	0	1	20.0	9.5	125	40.0	88.3	88.3	85.7	85.7
20	0	1	-1	0	12.5	12.0	50	22.5	71.9	71.9	75.8	75.8
21	-1	0	0	1	5.0	9.5	125	40.0	64.2	64.2	82.3	82.3
22	-1	0	0	-1	5.0	9.5	125	5.0	64.2	64.2	82.4	82.4
23	0	0	0	0	12.5	9.5	125	22.5	81.1	81.2	85.7	85.7
24	0	-1	-1	0	12.5	7.0	50	22.5	75.3	75.3	84.3	84.3
25	0	0	1	-1	12.5	9.5	200	5.0	83.7	83.7	85.7	85.7
26	0	0	0	0	12.5	9.5	125	22.5	81.1	81.2	85.7	85.7
27	0	-1	0	1	12.5	7.0	125	40.0	87.9	87.9	85.7	85.7
28	-1	0	-1	0	5.0	9.5	50	22.5	60.2	60.2	48.8	48.8
29	0	-1	0	-1	12.5	7.0	125	5.0	87.9	87.9	85.7	85.7

The goodness of fit statistics for the models representing CH₄ recovered and CO₂ captured for the three upgrading strategies are presented in Table 4.43. The results show that the ANN prediction was characterised by very high R² and

adjusted R² (R² and adjusted R²~1.0000). They were also characterised by very low RMSE and AAD values. This shows that there was almost a perfect fit between the ANN model predictions and the experimental values.

Table 10: Goodness of fit statistics for ANN

Parameter	Response	
	CH ₄ recovered	CO ₂ captured
R ²	0.9999	1.0000
Adj. R ²	0.9998	1.0000
RMSE	0.1200	0.0400
AAD	0.0003	0.0009

Comparison of RSM and ANN Performance

A performance evaluation was carried out to determine which of RSM and ANN was better in terms of their accuracy in predicting the responses (CH₄ recovered and CO₂ captured). Models with more predictive capability are more accurate when used in predicting responses (Betiku et al., 2014). The predictive capability of RSM and ANN was assessed using R² value, adjusted R² value, root mean square error (RMSE) and absolute average deviation (AAD) as shown in Table 11. Very accurate models

are characterised by high R² and adjusted R² values as well as low RMSE and AAD values (Velu et al., 2016). The results presented in Table 11 show that the ANN prediction was characterised by very high R² and adjusted R² (R² and adjusted R² ~ 1.0000). They were also characterised by very low RMSE and AAD values. This shows that there was almost a perfect fit between the ANN model predictions and the experimental values. Based on these observations, it is clearly evident that the ANN model performed better in predicting the responses compared to the RSM model.

Table 11: Comparison of RSM and ANN predictive performance

Parameters	RSM		ANN	
	CH ₄ recovered	CO ₂ captured	CH ₄ recovered	CO ₂ captured
R ²	0.9826	0.7442	0.9999	1.0000
Adj. R ²	0.9651	0.9521	0.9998	1.0000
RMSE	1.3937	4.2042	0.1200	0.0400
AAD	0.0172	0.0475	0.0003	0.0009

EFFECT OF INPUT FACTORS ON RESPONSES

The effect of the input variables (absorber pressure, biogas flowrate, liquid flowrate and absorber temperature) on the responses (CH₄ recovered and CO₂ captured) was evaluated by using response surface plots. These are three-dimensional (3D) plots, and they were produced by varying two variables within the experimental range, and keeping the remaining two variables constant at their centre point values. Figure 9 shows the effect of the interaction between absorber pressure and biogas flowrate on the amount of

methane recovered for the high-pressure water scrubbing process. The trend observed show that high values of pressure favoured the water scrubbing process. This is seen in the fact that the amount of methane in the upgraded biogas increased as the pressure in the absorber was increased. This could be attributed to the removal of the impurities in the biogas, which is facilitated by the increase in their solubility at high pressures (Cozma et al., 2015). The amount of methane in the upgraded biogas was not significantly influenced by the biogas flowrate compared to absorber pressure.

Nevertheless, there was an inverse relationship between methane recovery and the biogas flowrate. A combination of high absorber pressure and low biogas flowrate resulted in optimal methane

recovery. Similar observations have been reported by previous researchers (Cozma et al., 2015; Sugiharto and Hidayat, 2015; Morero et al., 2017).

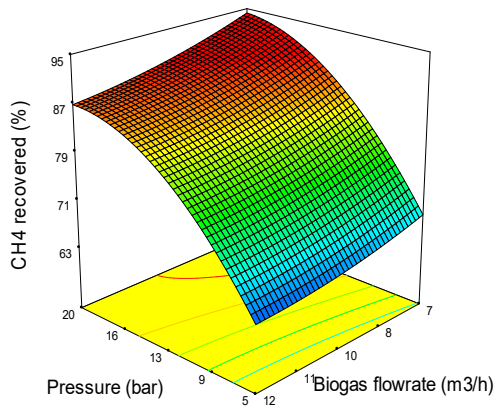


Figure 9: Response surface plot showing effect of absorber pressure and biogas flowrate on CH₄ recovered

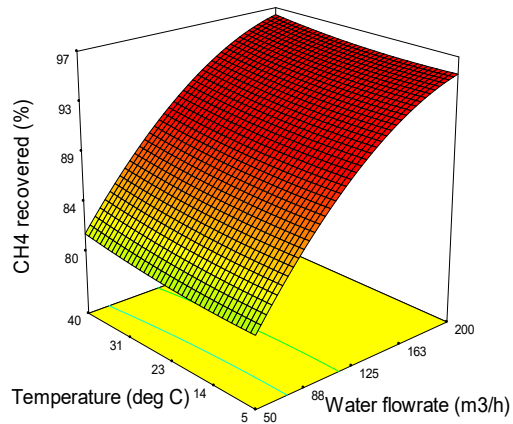


Figure 10: Response surface plot showing effect of temperature and water flow rate on CH₄ recovered

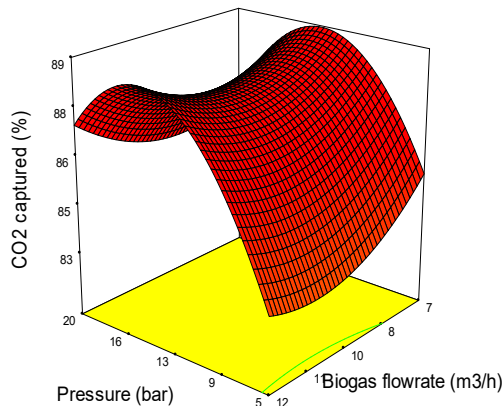


Figure 11: Response surface and contour plot showing effect of absorber pressure and biogas flowrate on CO₂ captured

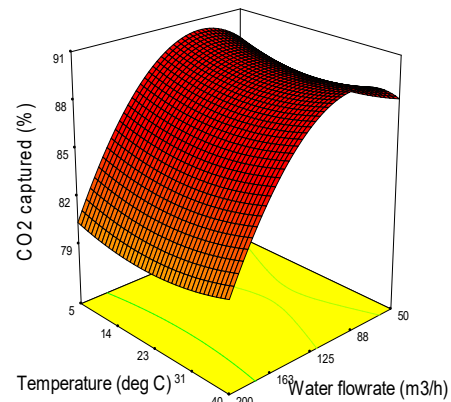


Figure 12: Response surface and contour plot showing effect of temperature and water flowrate on CO₂ captured

The effect of water flowrate and absorber temperature on methane recovery is shown in Figure 10. Increasing the water flowrate resulted in a significant increase in the amount of methane recovered in the upgrade biogas. This could be attributed to an

increase in the mass transfer driving force, which facilitates the enhanced removal of the impurities from the raw biogas (Ndiritu et al., 2013). This observation is in line with the results of Aroonwilas et al. (2001) who attributed the increase in absorption efficiency to



enhanced bulk absorption capacity of the liquid due to the increase in liquid molecules. Similar observations were also reported by previous researchers (Oi, 2007; Cozma et al., 2015; Sugiharto and Hidayat, 2015; Morero et al., 2017). Temperature did not have any significant effect on the absorption process as seen from Figure 10. This observation is corroborated by the fact that the p value for the model term representing temperature in the statistical model was greater than 0.05 indicating that it was not significant. Nevertheless, a combination of low temperature and high water flowrate resulted in optimum methane recovery.

The removal of CO₂ from the raw biogas as a function of absorber pressure and biogas flowrate is presented in Figure 11. The absorber pressure significantly enhanced the removal of CO₂ from the biogas stream as seen in the steep increase in CO₂ captured when the absorber pressure was increased. This observation can be directly linked to the fact that absorption of CO₂ by water is governed by Henry's law. Thus, the absorption process will be facilitated at high pressures (Cozma et al., 2013). The absorption of CO₂ was better at lower biogas flowrates. This could be as a result of the increase in the CO₂ loading as the biogas flowrate was increased. The

increased CO₂ loading does not favour the process in terms of mass transfer characteristics because the mass transfer driving force would have been increased (Abdul Halim et al., 2015). The increase in the removal of CO₂ from the raw biogas as a function of absorber pressure and water flowrate is directly related to the increase in the recovery of methane in the upgraded biogas. Higher water flowrates was found to be favourable for the removal of CO₂ while temperature did not have any significant influence on the process as shown in Figure 12.

Optimisation of Input Factors and Responses

Numerical optimisation of the responses was carried out using the Design Expert software. The values of the independent variables were fixed within the experimental range while the responses were maximized. The results of the analysis are shown in Table 12. The high-pressure water scrubbing process recorded 93% methane in the upgraded biogas stream with a corresponding CO₂ removal level of 87%. These results were validated by carrying out triplicate experiments at the recorded optimum conditions. The mean values of the triplicate experiments were comparable to the prediction of the statistical models.

Table 12: Optimization results

Variable	Value
Absorber pressure	18 bar
Biogas flow rate	7 m ³ /h
Water flow rate	148 m ³ /h
Absorber temperature	11 °C
Maximum CH ₄ recovered	93%
Maximum CO ₂ captured	87%

CONCLUSION

The following conclusions can be drawn from the results of this work.

1. ANN is a better predictive tool compared to RSM

2. CH₄ recovery and CO₂ captured during biogas upgrading via HPWS can be represented by validated quadratic models



3. CH₄ recovery is significantly influenced by absorber pressure, biogas flow rate, water flow rate but not temperature
4. CO₂ captured is significantly influenced by absorber pressure and water flow rate but not biogas flow rate and temperature

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