Int. J. Renew. Energy Dev. 2023, 12(1), 99-110

Contents list available at IJRED website

International Journal of Renewable Energy Development

Journal homepage: https://ijred.undip.ac.id

Research Article

Optimization and Analysis of a Low-Pressure Water Scrubbing Biogas Upgrading System via the Taguchi and Response Surface Methodology Approaches

Gad Reuben Mugagga[a*], Isaiah Bosire Omosa[b], Thomas Thoruwa[c]

[a]*Department of Energy Technology, School of Engineering and Technology, Kenyatta University, Kenya*

[b]*Department of Civil Engineering, School of Engineering and Technology, Kenyatta University, Kenya*

[c]*School of Pure and Applied Sciences, Pwani University, Kenya*

Abstract. Biogas upgrading is essential in order to increase the calorific value and improve the quality of raw biogas. This present study aims at investigating the optimum performance of a near atmospheric pressure water scrubbing (NAPWS) system for biogas upgrading while using both the adsorption and absorption techniques. This was achieved through a two-stage process: namely, the Taguchi approach followed by the response surface methodology (RSM). The Taguchi orthogonal array design consisted of 27 runs where the raw biogas pressure (10 - 30 kPa), liquid flow rates (2.6 - 4.2 l/ min.) and variations of the steel wool height (0 - 45.72 cm) in the adsorption column were experimentally studied with respect to the methane (CH_4) yield and removal efficiency of hydrogen sulfide (H_2S) and carbon dioxide (CO_2). From the experiments, the removal efficiency of hydrogen sulfide was greater than 87% with the average bio-methane content of 77.67%. During the second-stage, the analysis of variance (ANOVA) and the RSM were undertaken for optimization of the process parameters. The optimum bio-methane concentration of 84.71 (%v/v) CH_4 and 13.31 (%v/v) CO_2 was attained at gas pressure of 14kPa, liquid flow rate of 4.2 l/min., and steel wool height at 22.86cm obtained through numerical optimization. These results revealed that the utilization of the Taguchi and the RSM yielded to the best optimal system performance with the liquid flow rate as the most significant factor.

Keywords: Biogas upgrading, Biogas yield, Packings, Taguchi method, Response surface methodology

Received: 11th August 2022; Revised: 27th Sept 2022; Accepted: 12th Oct 2022; Available online: 19th Oct 2022

1. Introduction

Biogas usage in cooking applications is a sustainable option in several developing countries as the need for cooking fuel increases (IRENA, 2016). It is a product of Anaerobic Digestion (AD) of biodegradable waste and bio-slurry, a potential fertilizer. Biogas is comprised of constituent gases namely methane (CH_4) (55-70%), carbon dioxide (CO_2) (30-40%), hydrogen sulfide (H_2S) (0-20,000ppm), nitrogen (N_2) (0-2%), ammonia (NH_4) (0.05%), water vapor (H_2O) (0-7%) and other trace elements (Awe *et al.*, 2017; Singhal *et al.*, 2017). The raw biogas composition largely depends on the pH, temperature, retention time, pretreatment of the substrate and the loading rate (Djimtoingar *et al.,* 2022). The methane component in biogas is the only combustible gas, while the presence of carbon dioxide reduces the calorific value of the gas. The presence of hydrogen sulfide causes corrosion because of a chemical reaction with water vapor forming sulfur dioxide which is both corrosive and toxic (Abdeen *et al.*, 2018). In contrast, water vapor leads to fouling of biogas equipment. It is noteworthy that using raw biogas as a fuel has limitations for various applications, which necessitates its upgrading to improve its quality. The upgrading consequently improves the calorific value of biogas from 22MJ/m^3 to 34MJ/m^3 of the now bio-methane (IRENA, 2016). The bio-methane could be compressed into cylinders or used as a fuel for transportation, electricity generation, and heating.

Biogas upgrading has widely been carried out using various techniques, including absorption, adsorption, membrane, biological and cryogenic separation (Awe *et al.*, 2017; Kapoor *et al.*, 2019). Previous research on this subject indicates that the absorption and adsorption techniques have been widely employed due to their simplicity, cost, and environmental friendliness (Bauer *et al.*, 2013; Gantina *et al.*, 2020). The water scrubbing technology employs the difference in solubility of H_2S and CO_2, which are much lower than CH_4 i.e., the solubility of CO_2 in water is 25 times higher than that of methane at 25^{0}C (Bauer *et al.*, 2013). Since the process can also be carried out with the near atmospheric pressure water scrubbing (NAPWS) and at ordinary temperatures, it renders it more versatile as it can quickly be adopted at various locations. Water scrubbing has been reported to be the most extensively used technology around the world (Kapoor *et al.*, 2017). Though numerous studies (Gantina *et al.*, 2020; Ilyas, 2006; Rajivgandhi & Singaravelu, 2014; Vijay *et al.*, 2006) have focused on the use of High-Pressure Water Scrubbing (HPWS), it is characterized by

* Corresponding author
Email: gadrobben@gmail.com (G.R. Mugagga)

https://doi.org/10.14710/ijred.2023.48269
ISSN: 2252-4940/© 2023.The Author(s). Published by CBIORE

high power requirement for gas compression and are mostly used at a commercial scale which facilitates bio-methane injection onto natural gas pipelines (Yousef *et al.*, 2016). The necessity to increase raw biogas and water to high pressures while also in-putting energy for cooling the scrubbing water renders the HPWS process expensive due to the energy intensity required (Walozi *et al.*, 2016). However, this high energy requirement could be offset by utilizing the NAPWS systems, which are reported to have lower specific power requirement, longer equipment lifespan, and requires no flashing process yet appropriate for smaller installations (Budzianowski *et al.*, 2017). Few studies have experimented on the application of the NAPWS for biogas purification. Noorain *et al.*, (2019) created a mathematical simulation model whilst using sponge carriers as packing materials and obtained a methane percentage of greater than 90%. Walozi *et al.*, (2016) investigated the use of granite stones, candlenut seed, plastic balls, calcite marble and steel wire mesh in a low-pressure pilot scale scrubbing system and achieved a maximum purification methane concentration of 80%. These studies show that literature on the NAPWS utilizing conventional packing materials is limited.

For smooth operation, proper packing selection is also considered a vital parameter in absorption column design as the packings influence the column's efficacy and investment costs (Tayar *et al.*, 2019). This is because they determine the mass transfer efficiency, void fraction, and packing factor between the liquid and the gas (Benizri *et al.*, 2019). Using random packings in gas absorption columns is advantageous because they are low cost, corrosion resistant, have a low-pressure drop, are suitable for high liquid loads, and have easy adaptability for smaller diameter columns (Kister *et al.*, 2008). Previous studies undertaken with the utilization of the most common random packings like the ceramic raschig (89.3-99%CH_4) (Gantina *et al.*, 2020; Ilyas, 2006; Vijay *et al.*, 2006), plastic bioball (>90% CO_2 absorption) (Abdeen *et al.*, 2018; Mel *et al.*, 2016; Tippayawong & Thanompongchart, 2010), intalox saddle (96-97%CH_4) (Rajivgandhi & Singaravelu, 2014), Pall ring (96-97% CH_4) (Nock *et al.*, 2014; Rajivgandhi & Singaravelu, 2014) demonstrate that there has been numerous research works undertaken with the first and second generation packings for biogas upgrading. However, only a few studies have been undertaken using the third and fourth-generation packings in the same regard. For instance, Kapoor *et al.*, (2017) utilized the IMTP - a third-generation metal packing, to study how methane concentration, column pressure, water flow rate, and the pressure difference between the vessel and scrubbing column affect the methane loss. The highest methane loss recorded was 9.9 percent. However, their study highlighted the need to study more about how the interaction of multiple factors leads to methane losses. Benizri *et al.*, (2019) employed the plastic Raschig Super Ring®, a fourth-generation packing in an improved absorption column. These authors studied the effect of pressure, temperature, gas, and liquid flow on biogas upgrading. The biogas was enriched to 77% methane with a high methane recovery ratio of 94%. Correspondingly, these recent packings are known for having superior performance as they offer lower pressure drop, higher capacity and higher interfacial area (Hegely *et al.*, 2017). With regards to the packing material, the comparative study (Kolev *et al.*, 2006) conducted on both plastic and metallic third and fourth generation packings (Raschig Super Ring®, metal Intalox random packing, and Ralu flow) revealed that at the same liquid superficial velocity and specific surface, the metallic packings had superior effective surface area and lower pressure drop. However, literature on the utilization of the third and fourth-generation metallic packings for biogas purification is rare mainly because their significant areas of application are on an industrial scale.

Response Surface Methodology (RSM) is a set of both mathematical and statistical techniques useful for the modeling and analysis to fully understand the correlation of several control variables on the process responses (Madondo *et al.*, 2022). The performance of water scrubbing systems is dependent on the gas pressure, water flow rate, raw biogas composition, the purity of the water used, the hydraulic retention time, and the dimensions of the scrubbing tower (Noorain *et al.*, 2019). The system efficiency can be improved by optimizing the above-named process parameters whilst utilizing the RSM thereby reducing experimental replications, cost, as well as the time for conducting the experiments. Manmai and his counterparts studied the application of RSM with particular emphasis on bioethanol production from both sunflower stalk (Manmai *et. al.*, 2020) and the comparison between the sorghum stalk and sugarcane leaf (Manmai *et al.*, 2020). However, these studies were mainly focused on biogas generation. Some previous studies applied the same technique but in carbon dioxide capture and omitted hydrogen sulfide removal. Carbon dioxide capture using the vacuum pressure swing adsorption with silica gel was conducted through a pilot and simulation study recently (Shen *et al.*, 2018). Besides, a previous study (Song *et al.*, 2014) used a novel cryogenic CO_2 capture process based on three levels of the Central Composite Design (CCD) to obtain the desired optimum conditions. The CCD happens to be the most popular design method in RSM (Djimtoingar *et al.*, 2022). Previous efforts to capture the application of the CCD for biogas upgrading were undertaken by (Morero *et al.*, 2017) through a simulation study with three varied absorber–stripper configurations consisting of solvents like water, dimethyl ethers of polyethylene glycol and diglycolamine. It is just one recent study (Tran *et al.*, 2021) that investigated biogas purification by removal of both hydrogen sulfide and carbon dioxide by employing a high gravity rotating packed bed (HGRPB). In their study, they investigated various factors like the pH of the solution used, the speed of the HGRPB, biogas pressure and the liquid flow rate with the Box-Behnken Design (BBD) in conjunction with RSM. Up to this point, to the best of the authors' knowledge, there is no published research where both Taguchi and RSM are used for process optimization of a NAPWS packed bed with third-generation packings for biogas upgrading.

Therefore, the main objective of this paper is aimed at upgrading biogas with the NAPWS using third-generation packing material while applying both the Taguchi and RSM for the process optimization. For this reason, the performance evaluation was undertaken with varying biogas pressures, liquid flow rates, and varying heights of the adsorption material (steel wool) in order to obtain the optimal level parameters for bio-methane production.

2. Materials and Methods.

2.1 Materials.

Raw biogas was generated from a 2m^3 balloon type digester located in Mwiki, Nairobi County (1°13'30.5"S, 36°55'30.4"E). The organic substrate was composed of water and cow manure in a ratio of 1:1 totaling to 80 liters per day. Tap water was used as the scrubbing liquid inflowing to the absorption column without any purification. Biogas from the digester was obtained under mesophilic conditions. The experiment was conducted during both the hot and cold days of the months of November and December, 2021 so as to account for the variability in gas production due to seasonal changes.

2.2 Methods

2.2.1 Experimental setup and procedure.

The biogas digester was hooked to the upgrading system with the aid of a plastic hose which was attached to a PVC inlet pipe (diameter 2.54 cm) at the bottom of the adsorption column. Prior to experimentation, a leak test was carried out to ensure that the upgrading system (Fig. 1) had no leakage. The biogas upgrading system was comprised of two columns (the adsorption and the absorption column) and the dehydrating unit which were all fixed onto a vertical chaff board with the aid of metallic clips.

The two columns were constructed from PVC pipes of diameters 10.16 and 15.42 cm respectively. PVC pipes were selected as they have been widely used (Abdeen *et al*., 2018; Riyadi *et al*., 2018) and are economical for column construction. Subsequent to the sampling of the inlet gas, the raw biogas was then introduced to the adsorption chamber which either was empty or contained steel wool depending on the particular experimental variation being conducted. This column was intended for hydrogen sulfide adsorption. In addition to the atmospheric pressure acting on the digester, the biogas was further propelled to the purification system by the aid of a variable biogas pump procured from Nairobi. The pump enabled the regulation of gas pressures used and also ensured constant inflow of the gas to the absorption column through the adsorption column. The gas was passed through a series of flexible and PPR pipes to ensure no leakage in the system. The gas flow rate was determined along this path with the aid of an air flow meter (Model 8357, USA). The gas was introduced into the absorption chamber at close to the bottom of the absorption chamber which had a concurrent downward water flow that was regulated with the aid of a liquid flow meter. More generally, the absorption column had an inlet for the water (from a raised tank) at the top and a water outlet at the bottom whereas the reverse was true for the gas in- and out-lets. The effluent water from this column was discharged through its bottom section and its pH and temperature measured. The absorption column had a height of 45.47 cm with an internal volume corresponding to 7.3 litres and was filled with packings (Fig. 2) to a height of 30.48cm. The packing material characteristics are further detailed in table 1. The packings were employed to increase the mass transfer efficiency between the liquid and gas interface. Additionally, two dry type pressure gauges (LANNENG, China) were used to monitor the absorption column pressures. This column was intended for the removal of both carbon dioxide and the remaining hydrogen sulfide. During the system construction, a ½'' non-return valve was used in order to avoid the liquid back flow towards the gas line from the biogas pump.

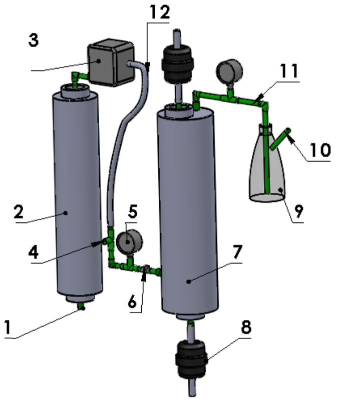

Fig.1: 3D schematic view of the setup

Fig. 2: The Cascade Mini Ring®

Table 1
Packing material characteristics

S/No.	Parameter	Detail
1.	Packing type	Cascade Mini Ring®
2.	Manufacturer	Pingxiang
3.	Material	Metallic (SS304)
4.	size (mm)	17*6*0.3
5.	Void fraction, ε (m^3/m^3)	0.955
6.	Specific surface area, a (m^2/m^3)	420

In order to prevent gas short circuiting through the bottom of the absorbing column, a water seal was designed with the packings held raised to 10cm from the bottom of the column. The cleaned gas escaped through a PPR pipe at the top of the absorption column to the dehydrating unit made of a plastic containment of 500 mL volume filled with blue silica gel (5-20 mesh size). The gas was collected and analyzed with both the OT-600 and GC-SRI at the exit of the dehydrating unit.

2.2.2 Gas sampling.

It was paramount to pay keen attention to the gas sampling techniques. In the present study, two test points were considered i.e. at the inlet and the outlet of the purification system. For the incoming gas composition, triplicate samples for each test were done in intervals of 30 seconds for the on- and off-system gas tests to factor in the variability. The on-system tests were carried out to measure the composition of the hydrogen sulfide with the aid of the OT-600 biogas analyzer while the off-system analysis was undertaken with the GC-SRI gas chromatography for further methane and carbon dioxide analysis. In this analysis however, all other poorly soluble gases like oxygen and nitrogen were neglected. The first gas samples were collected in the intervals as mentioned. The samples from the purification system were obtained directly from the gas exit pipe. All on-system tests for the H_2S where measured and recorded directly while samples for the off-system gas tests were collected in propylene syringes. The collected samples were then immediately transferred to 10 mL transparent gas vials that were fitted with both plastic and crimp seals. The gas vials were differentiated by marking them with individual experimental numbers with either the -1 or -2 suffix to denote whether the test was either for the incoming or outgoing gas.

2.2.3 Analytical methods.

During the experiments, the on-system measurements performed using the OT-600 portable multi-gas analyzer (Henan Zhongan, China) were recorded directly. According to the manufacturer's specification, the actual response times were less than 20 seconds. For this study however, the analyzer readings were undertaken after 30 seconds to ensure that enough gas had been suction for more accurate readings to be displayed. As for the off-system testing, this was carried with the gas chromatography machine (model 8610C, GC-SRI, USA) at the International Livestock Research Institute (ILRI) facility in Nairobi - Kenya to obtain the CH_4 and CO_2 variations for each test undertaken. The analysis of the gas samples was done within 2-3 weeks from the time the gas samples were taken. The GC-SRI was comprised of a methanizer and coupled with a Flame Ionization Detector (FID) and a 63Ni Electron Capture Device (ECD). Additionally, the GC-SRI was operated using Haysep D packed columns (3m, 1/8'') with oven temperatures ranging from 65-70^{0}C. However, both the ECD detector and the methanizer temperatures were fixed to 350^{0}C. Nitrogen having flow rates of 25 ml/min through both the ECD and FID lines was used as the carrier gas. In order to obtain the concentration of the gas samples, the differences in the peak areas were measured for each sample vis-à-vis the peak samples of the calibration gases. The attained concentrations were later converted to mass per volume. An AC 20W biogas booster pump (LANNENG, China) was used for the variation of raw biogas pressures whereas the liquid flow rates where controlled by the use of water flow meters (LANNENG, China). The gas flow rate was measured with the TSI VELOCICALC® airflow meter (Model 8357, USA). The pH and water temperature were measured with a SensoDirect 150 meter (Lovibond 724200, Germany). This pH meter was recalibrated after every test with the use of distilled water to ensure its accuracy. Additionally, the pH results were recorded only after the pH meter reading had stabilized while those for the water temperature were taken immediately after the effluent water left the absorption column.

2.2.4 Optimization and process parameters.

The Taguchi method was utilized so as to analytically study the effect of the variable control factors for the first stage analysis. This was essential in order to determine the ranking sequence of the individual factors from the one having the greatest influence on the methane yield to the one with the least. This method aids in studying the outstanding factors for optimal level performance by measuring the variance of the factor levels and also in attaining the highest signal to noise (S/N) ratio pertaining to the desired response. For this study, three control variables namely; the liquid flow rate, biogas pressure and the height of the steel wool in the adsorption column were studied. The Taguchi experimental design consisted of 27 experiments performed through the L_{27} Orthogonal Array (OA) with each experiment lasting one minute. All the control factors were varied between three levels which are represented as 1, 2 and 3 in the subsequent sections. The liquid flow rate was varied between 2.6 – 4.2 liters/minute, with an increment of 0.8 l/min. The biogas flow rate was varied between 10 – 30 k/Pa with an increment or 10 k/Pa and finally the height of the steel wool was varied between 0 – 45.72 cm with an increment of 22.86 cm. In order to determine the factor dispersion about the desired optimum value, the Signal to Noise (S/N) ratio was determined. The design of experiments and analysis was performed using Minitab version 18 (Minitab Inc., USA).

The results of the Taguchi design were subjected to a second optimization stage using the RSM to further analyze the combined effect of the key operating factors on the response variable(s). Experimental data was used to generate predicted yield studentized residual plots, predicted versus actual value plots and pareto charts. ANOVA analysis was also carried out to test the developed quadratic model validity. From the estimated models, response surfaces were developed using Design-Expert 13 (Stat-Ease Inc., Minneapolis, USA) which is the most widely used softwares (Djimtoingar *et al.,* 2022).

3. Results and Discussions.

3.1 Biogas generation.

From the chemical compositional analysis of raw biogas, the average biogas composition is represented in Table 2. This methane composition had a fairly good quality preferable for domestic cooking. A similar behavior in the variation of biogas was reported by Singhal *et al.*, (2017).

Table 2
Chemical composition of biogas

Constituent	This study	Singhal *et al.*, (2017)
CH_4 (%)	56.63±3.69	50-70
CO_2 (%)	47.32±3.691	30-50
H_2S (ppm)	>100	0-4000

3.2 Effect of the adsorption column to the purification performance.

The inlet gas concentration was observed as 100ppm for most tests. This was mainly attributed to the technical limitation of the OT600 portable multi-gas detector whose design limit was up to that level. However, upon analysis of the purified gas, results in Fig. 3 showed a marked reduction of the levels of the hydrogen sulfide concentration to levels less than 13ppm, corresponding to a removal efficiency of greater than 87%. This hydrogen sulfide level renders the gas suitable for use in boilers, internal combustion engines, in selected fuel cells and also for kitchen stoves (Awe *et al.*, 2017). Remarkably, most of the results were below undetectable levels i.e. 0ppm. This subsequently resulted into a mild change of color of the steel wool to a rusty brown precipitate due to its reaction with the hydrogen sulfide in the adsorption column to form Iron (II) sulfide. As for the experimental runs 22-27, it was observed that for the same pressure i.e. 30kPa, increasing the liquid flow rates i.e. from 3.4 to 4.2 l/min resulted into the reduction of the average hydrogen sulfide concentrations to 8 and 0 respectively.

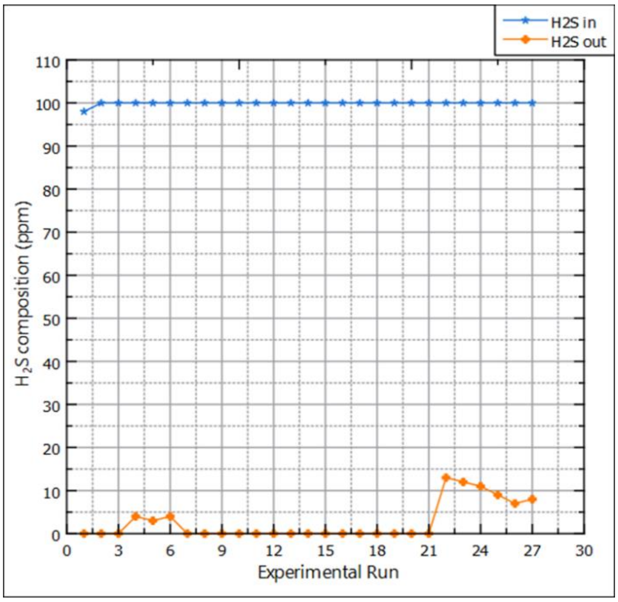

Fig. 3. Effect of the purification system on H_2S removal between the input and output gas compositions.

However, this increase in pressure resulted into faster gas escape through the adsorption column which consequently led to the reduction of the gas residence time in the adsorption chamber. The remainder of the hydrogen sulfide gas was otherwise then reduced in the absorption chamber. Indeed, the height of the steel wool in the absorption column influences the removal efficiency of the hydrogen sulfide. A similar study investigating this parameter with heights of 10, 30 and 50 cm attained 55.6%, 63.2% and 70.4% hydrogen sulfide reduction to levels less than 8 ppm (Mohanakrishnan & Kurian, 2016). This was also in agreement with the study conducted by the authors (Riyadi *et al.*, 2018) where steel wool packed to heights of 100, 75 and 50 cm attained the best hydrogen sulfide removal efficiency of 97% at 100cm.

For this reason, though hydrogen sulfide has a small effect to the overall biogas percentage composition, its removal efficiency is slightly dependent on the rate of gas flow. Comparing of the present study with the above reported studies, it is then clearly indicated that the coupling of both the absorption and the adsorption column effectively removes the hydrogen sulfide from raw biogas.

3.3 Effect of scrubbing water pH on purification performance.

The purification of the biogas proceeds with the washing out of both the hydrogen sulfide and carbon dioxide which react with the influent water to form an acidic solution. Fig. 4 demonstrated that there was a reduction in the pH of the scrubbing water owing to the dissolution of these gases into the scrubbing water. The overall incoming water pH to the absorption column was neutral and tending to alkalinity in a few instances (for runs 19-21) characteristic of the used tap water. As for the effluent water, a gradually reducing trend of the pH was noted as the working pressures increased corresponding to experimental runs 16-27. In this respect, the water pH was studied to assess the level of solubility of the acid gases. It was observed that as the presence of hydrogen sulfide in the final bio-methane increased (Fig. 3), the more acidic the effluent water was. This was observed in experimental runs 4-6 and more specifically in runs 22-27. Consequently, this lowered the possibility of increased carbon dioxide solubility in the scrubbing water. This implied that the methane percentage should have been lowered which actually was not the case but was rather offset by the increased pressures for the later mentioned runs i.e. 22-27. In these runs, a similar trend was observed where the solubility of hydrogen sulfide decreased with decreasing pH. Remarkably, when incoming water of increased average pH i.e. 7.42 was used for runs 19-21, there was no sudden change in the output water pH trend it had previously followed when the influent water pH was in the 6-7 pH range. This implied that even when the influent water has varying pH ranges, biogas purification is still possible. Given that our anticipation prior to experimentation was that the scrubbing water pH would have a considerable effect on the level of biogas purification, the results further showed only a slight effect indicating that the pH has a minimal impact on the purification performance which concurs with the study by the authors (Noorain *et al.*, 2019).

During the experiments, the temperature of the incoming water into the absorption column kept fluctuating as it was not controlled and was thus considered a noise factor. The average influent scrubbing water temperature used was 26.11^{0}C while the average exit water temperature was 29.35^{0}C (Fig. 5). As the temperature of the influent water increased, the carbon dioxide absorption efficiency was decreased.

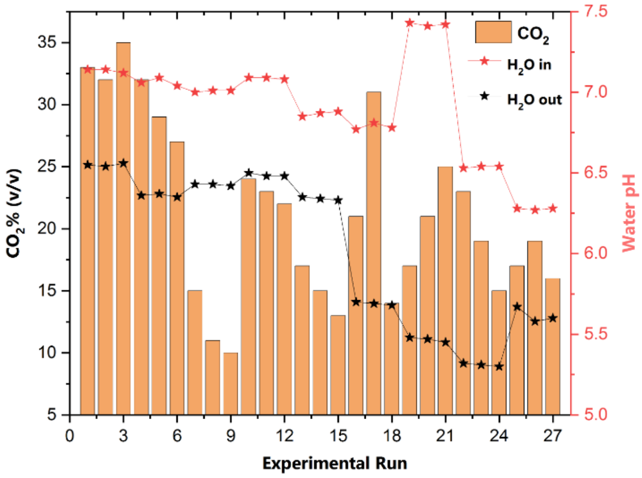

Fig. 4. Effect of scrubbing water pH on CO_2 (%v/v) removal.

3.4 Effect of the scrubbing water temperature on purification performance.

This was mainly because, at higher water temperatures as is explained by Henry's law, the liquid molar fraction of carbon dioxide is lowered due to reduced carbon dioxide solubility. Specifically, this was experienced in the experimental runs 4-6, 19-21 which were characterized with lower methane and higher carbon dioxide concentrations. This implied that the solubility of carbon dioxide in water increased with a reduction in temperature. During the experiment, the temperature variation range of the influent water was 3.9 Kelvin which was rather low and could not yield a substantial effect to the performance index as compared to the 10 Kelvin range reported in the model developed by authors (Nock *et al.*, 2014).

There was a slight relationship between the methane content in the effluent gas and the scrubbing water output temperatures. This was depicted in runs 1-6 and 16-18 where the effluent water temperature was relatively higher compared to the rest of the experimental runs. Incidentally, the aforementioned runs corresponded to the runs with the lowest methane content i.e. mostly less than 70% v/v. Fig. 5 below showed a gradually reducing trend for the output water temperature in subsequent runs as the experiments were carried on from a maximum of 32.2 to a minimum of 28.8^{0}C. This temperature reduction was associated to the increasing water flow rates leading up to reduced hydraulic residence time in the absorbing column.

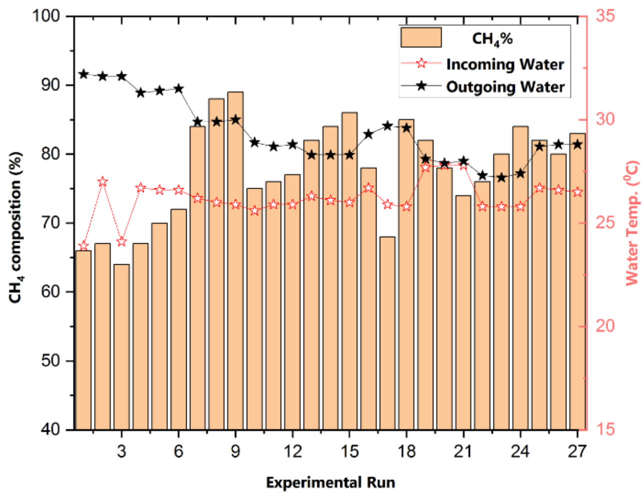

Fig. 5. Effect of scrubbing water temperature on purification performance.

3.5 Performance optimization with the Taguchi orthogonal array.

In order to determine the influence of the control factors on the methane yield, the Taguchi method of experimental design was applied. This was intended to limit the time of experimentation, number of experiments, and the cost while keeping the experiment robust (Nguyen *et al.*, 2017). This was done by varying different sets of combinations of the said control factors (Table 3) and recording the corresponding response data (Table 4). From this data, it was observed that the average methane concentration was 77.67%. Correspondingly, it is evident from Fig. 6 that the liquid flow rate has the highest influence on the carbon dioxide absorption rate. This observation correlates with why the liquid/gas (L/G) ratio is an important factor in determining the rate of absorption (Pirola *et al.*, 2015; Rajivgandhi & Singaravelu, 2014). Using the Taguchi methodology, the response means were arrived at by variation of the control factors as shown in Table 4.

Table 3
Control factors and their corresponding levels and notations in Taguchi design.

Notation	Control Factor	Units	Actual levels (coded levels)		
			1 (-1)	2 (0)	3(+1)
A	Pressure	kilopascals	10	20	30
B	Height*	centimetres	0	22.86	45.72
C	Liquid flow rate	Litres/minute	2.6	3.4	4.2

* height of steel wool in Column 1

Table 4
The L_{27} orthogonal array in coded units and the response data of the control factors.

L_{27}	Control factors and levels			Response 1: CH_4 (%vol.)		Response 2: CO_2 (%vol.)	
	A	B	C	Actual	Predicted	Actual	Predicted
1	1	1	1	66.0	65.6	33.0	33.4
2	1	1	1	67.0	65.6	32.0	33.4
3	1	1	1	64.0	65.6	35.0	33.4
4	1	2	2	67.0	69.6	32.0	29.4
5	1	2	2	70.0	69.6	29.0	29.4
6	1	2	2	72.0	69.6	27.0	29.4
7	1	3	3	84.0	86.9	15.0	12.0
8	1	3	3	88.0	86.9	11.0	12.0
9	1	3	3	89.0	86.9	10.0	12.0
10	2	1	2	75.0	75.9	24.0	23.0
11	2	1	2	76.0	75.9	23.0	23.0
12	2	1	2	77.0	75.9	22.0	23.0
13	2	2	3	82.0	83.9	17.0	15.0
14	2	2	3	84.0	83.9	15.0	15.0
15	2	2	3	86.0	83.9	13.0	15.0
16	2	3	1	78.0	76.9	21.0	22.0
17	2	3	1	68.0	76.9	31.0	22.0
18	2	3	1	85.0	76.9	14.0	22.0
19	3	1	3	82.0	77.9	17.0	21.0
20	3	1	3	78.0	77.9	21.0	21.0
21	3	1	3	74.0	77.9	25.0	21.0
22	3	2	1	76.0	79.9	23.0	15.7
23	3	2	1	80.0	79.9	19.0	15.7
24	3	2	1	84.0	79.9	15.0	15.7
25	3	3	2	82.0	81.6	17.0	17.3
26	3	3	2	80.0	81.6	19.0	17.3
27	3	3	2	83.0	81.6	16.0	17.3

3.6. Parametric analysis.

The increase of the L/G ratio increases the CO_2 solubility and thereby reduces the CO_2 composition in the product gas. Increasing liquid flow rates increases the efficiency of the absorption column and therefore leads to more CO_2 absorption. It was also observed that progressive increments in methane enrichment were attained due to increasing liquid flow rates and reducing gas flow rates. This suggests that a higher absorbent flow rate compensates for the reduced pressures provided the liquid gas interface is high (Rasi *et al.*, 2008). Relatively increasing trends were observed between runs 7-9 and 13-15. The highest removal efficiency of carbon dioxide of 73.7 percent was obtained at an L/G ratio corresponding to 0.371. However, this ratio seemed abnormally low compared to similar studies corresponding to 2 (Walozi *et al.*, 2016), 1.5 – 2.0 (Noorain *et al.*, 2019). The reason for this deviation is likely attributed to the packing material used in the reported studies i.e. steel wire mesh and sponge carriers respectively as opposed to the re-known packing materials. The L/G ratio of the present study was found to be higher than that of (Rasi *et al.*, 2008) studied at higher pressures ranging from 10 - 30 bars. In their study, the removal efficiency of carbon dioxide ranged from 88.2 to 91.1 percent attained at L/G ratios in the range of 0.05 to 0.2. However, their removal efficiencies were considerably higher than those of the present study due to the experiment being conducted at 30bars. Accordingly, the L/G ratio was the most outstanding factor which confirms that it is a vital parameter in biogas purification.

Surprisingly, the height of the steel wool came as the second influential factor. Experiments with the fully packed adsorption column i.e. B equal to 3, had some of the best methane percentages i.e. averagely greater than 80%. This however is explained by the fact that the more the hydrogen sulfide gas in absorbed in the adsorption column, the less acidic the scrubbing water becomes which eventually facilitates the increased rate of carbon dioxide absorption by the influent water. The existence of hydrogen sulfide past the adsorption column either due to high gas pressures leading to its escape or due to low steel wool heights in the adsorption column ultimately lowers the purification performance of the system.

With regards to the control factors studied in this experiment, pressure was obtained as the third influential factor. From the response data obtained, this deviates from the critical importance of pressure in the conventional HPWS systems since it contributes to the rate of mass transfer and the liquid hold up. However, pressure is known for its alternating behavior since it could have a positive, negative or a minor effect to gas dissolution rate depending on the design of the apparatus (Nguyen *et al.*, 2017). The reason for being in the third place is likely due to its variable effect on the response. That notwithstanding, its prominence was witnessed by the increased rates of carbon dioxide dissolution in the interactions of other factors with pressure level 3.

Fig. 6 represents the influence of the control factors graphically at different levels for the multiple quality characteristics like the pressure, height and the liquid flow rate. The highest S/N ratio was indicative of the best quality characteristic using the “larger is better” concept as this research was aimed at maximizing the methane yield. The slope of the graphs were used to compare and determine the magnitude of the individual main effects. There was a positive correlation for the pressure by increasing sharply from levels 1 to 2, but the increment reduces gradually thereafter as the pressure approaches to level 3. In regards to the steel wool height in the adsorption column, a trend similar to that of the pressure was observed only that a sharp increase was maintained after level 2.

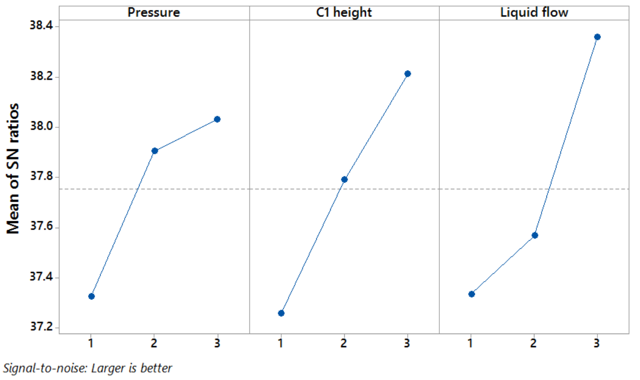

Fig. 6. Influence of the control factors on methane enrichment based on the S/N ratios.

Table 5
Response Table for the signal-to-noise ratios

Level	A (kPa)	B (cm)	C (l/min.)
1	37.32	37.26	37.33
2	37.90	37.79	37.57
3	38.03	38.21	38.36
Delta	0.71	0.95	1.03
Rank	3	2	1

Table 6
Response table for the average means.

Level	A (kPa)	B (cm)	C (l/min.)
1	74.11	73.22	74.22
2	79.00	77.89	75.78
3	79.89	81.89	83.00
Delta	5.78	8.67	8.78
Rank	3	2	1

For the liquid flow rate, a slow increment was observed from levels 1 to 2. However, a sharp rise was witnessed from level 2 to 3. This implies that the liquid flow had a more significant effect to the response variable – methane enrichment, as it exhibited the largest main effect.

In the determination of the deviation of the quality characteristics of the chosen levels, the Signal-to-Noise (S/N) ratios (Table. 5) and the average means (Table. 6) were also determined. From these tables, the S/N ratio was 37.75 with the overall mean given as 77.67. Correspondingly, the parameter with the largest S/N ratio and the highest average mean corresponded to the better quality characteristic. From the optimum parameter analysis, the order of influence was C > B > A with the corresponding delta figures of 1.03, 0.95 and 0.71 respectively.

3.6 Performance optimization with the Response Surface Methodology.

For the second stage analysis, RSM-based quadratic models were developed to predict the high gas quality characteristics (i.e. increased CH_4 yield and reduced CO_2) under the influence of the aforementioned experimental factors. With the aid of graphs, experimental values obtained were plotted against the predicted values so as to assess the prediction capabilities of the quadratic regression equations (1 and 2) below.

The respective models accounted for 80.8% and 78.18% of the variability in the experimental data. The reason behind this was in fitting the expected normal values that had outliers i.e. experimental runs 17 and 18 that caused low statistical fitting (Wang *et al.*, 2013). These outliers were however not discarded as they were considered important (Montgomery & Runger, 2003). The results of the model predictions and their variations with the externally studentized residuals and the actual methane yield and carbon dioxide removal are presented in Figs. 7 and 8 respectively. Fig. 7(a) and 8(a) show the externally studentized student residual versus the methane yield and the carbon dioxide removal. From the above figures, it was evident that the scatter plot and the variance of the experimental percentages was constant hence not requiring any need for any data alteration to the response variables. The predicted versus actual graphs are shown in Figs. 7(b) and 8(b).

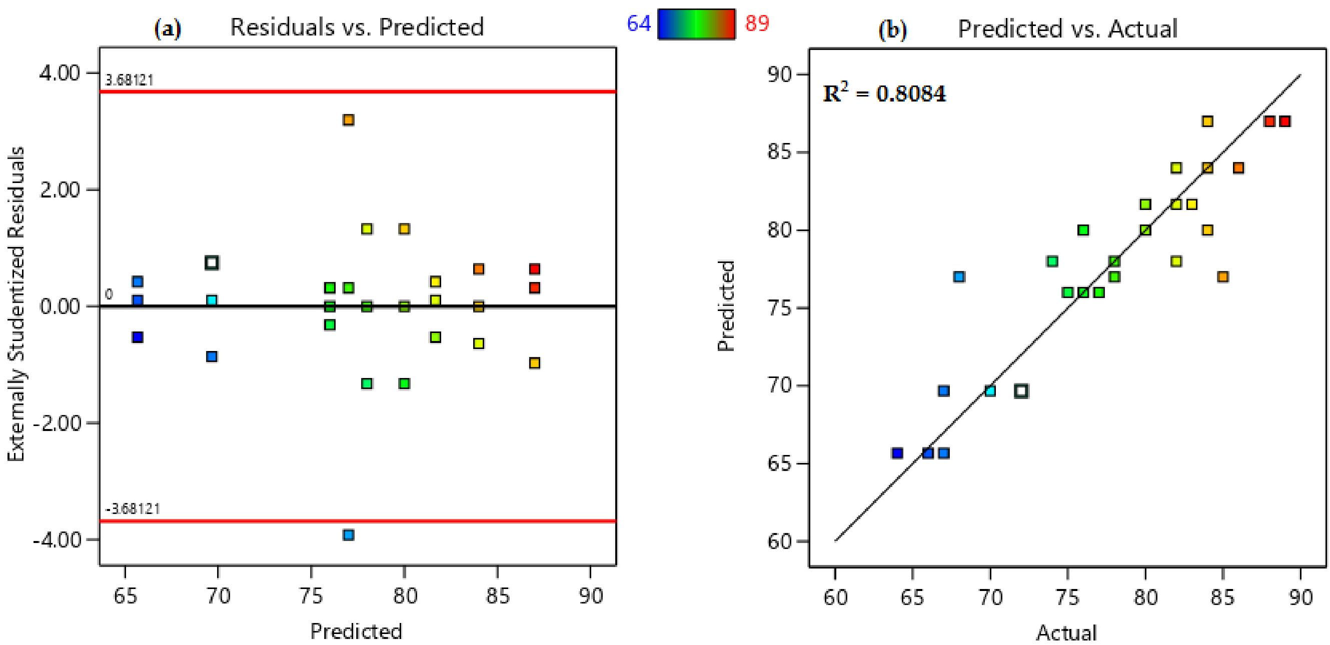

Fig. 7. CH_4 yield analysis. (a) Predicted yield studentized residual plots, (b) Predicted and actual plots.

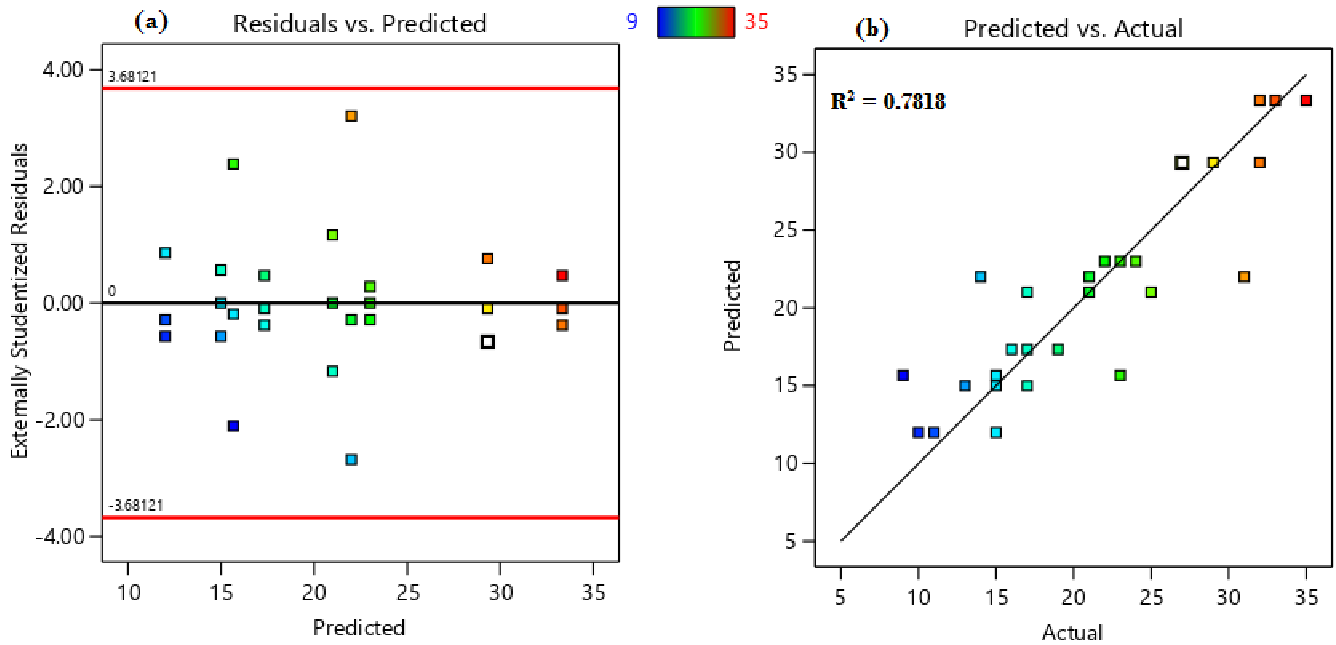

Fig. 8. Carbon dioxide removal analysis. (a) Predicted removal studentized residual plots, (b) Predicted and actual plots.

The quadratic response surface regression models generated by the ANOVA can be used to predict the methane yield and carbon dioxide removal efficiency in terms of the previously defined adopted factors. However, it is noteworthy that the BC interaction term was eliminated from the model as it could not be estimated.

$$CH_{4\,yield} = 58.3 + 18.67A - 14.06B + 1.94C + 2.22AB - 6.11AC - 2.00A^2 + 2.72B^2 + 3.94C^2 \quad (1)$$

$$CO_{2\,removal} = 41.8 - 19.22A + 12.4B - 1.39C - 2.22AB + 7.22AC + 1.44A^2 - 2.17B^2 - 4.50C^2 \quad (2)$$

Table 7
ANOVA for the quadratic model of CH_4 yield

Source	DF	SS	Mean	F-Value	P-Value
Model	8	1080.00	135.00	9.49	0.000*
A: Pressure	1	150.22	150.22	10.56	0.004*
B: C1 Height	1	14.69	14.69	1.03	0.323
C: Liquid flow	1	272.25	272.25	19.14	0.000*
AB	1	22.22	22.22	1.56	0.227
AC	1	168.06	168.06	11.82	0.003*
A^2	1	24.00	24.00	1.69	0.210
B^2	1	33.35	33.35	2.34	0.143
C^2	1	70.01	70.01	4.92	0.040*
Error	18	256.00	14.22		
Total	26	1336.00			
Std. Dev.	3.77		R^2		0.8084
Mean	77.67		Adjusted R^2		0.7232
C.V %	4.86		Predicted R^2		0.5689
			Adeq. Precision		9.7980

DF – Degrees of freedom, SS - Sum of Squares, Std. Dev. – Standard Deviation, C.V% - coefficient of variations, Adeq. – Adequate, * - significant

Table 8
ANOVA for the quadratic model of CO_2 yield

Source	DF	SS	Mean	F-Value	P-Value
Model	8	1156.30	144.537	8.06	0.000*
A: Pressure	1	213.56	213.556	11.91	0.003*
B: C1 Height	1	4.69	4.694	0.26	0.615
C: Liquid flow	1	220.03	220.028	12.27	0.003*
AB	1	22.22	22.222	1.24	0.280
AC	1	234.72	234.722	13.09	0.002*
A^2	1	12.52	12.519	0.70	0.414
B^2	1	21.12	21.125	1.18	0.292
C^2	1	91.12	91.125	5.08	0.037*
Error	18	322.67	17.926		
Total	26	1478.96			
Std. Dev.	4.23			R^2	0.7818
Mean	20.96			Adjusted R^2	0.6849
C.V %	20.2			Predicted R^2	0.5091
				Adeq. Precision	8.7273

DF – Degrees of freedom, SS - Sum of Squares, Std. Dev. – Standard Deviation, C.V% - coefficient of variations, Adeq. – Adequate, * - significant

3.7. The Analysis of Variance (ANOVA) responses.

From the ANOVA Tables 7 and 8 above, the models have their F-values as 9.49 and 8.06 respectively which implies that both models are significant. Both models have only a 0.01% chance that an F-value this large could actually occur which might only be due to noise. Similarly, from both models, the significant terms are A, C, AC and C^2 as they have p values less than 0.05 (Manmai *et al.*, 2020). All model terms greater than 0.10 were considered insignificant. Statistical significance implied that changing the actual factor would lead to a significant change in the response. Considering that the predicted and adjusted R^2 values have a difference of less than 0.2, then there is a reasonable agreement between the two values for both models. The adequate precision is measure of the signal to noise ratio and usually, a ratio of greater than 4 is required for more accurate predictions. Given that the models have adequate precisions of 9.7980 and 8.7273 respectively, then they indicate an adequate signal and that both models can be used to navigate the design space effectively. The C.V value of the CH_4 yield i.e. 4.86 was lower than that of CO_2 i.e. 20.2 implying that from the deduced models, the one of CH_4 yield is more significant.

As for the H_2S yield ANOVA analysis presented in S1, the model developed was significant with F-value as 6.86 with a 0.04% chance of occurrence due to noise. The only significant model factors are A and A^2 with p values less than 0.05. The predicted R^2 of 0.4441 is in reasonable agreement with the adjusted R^2 of 0.6431 with a difference of less than 0.2. Though the adequate precision is greater than 4 implying that the obtained ratio indicates an adequate signal and that the model can be used to navigate the design space, the C.V value corresponding to 89.7 is very high. The C.V is the dispersion around the mean and the lower the C.V value, the more precise the model becomes (Armah *et al.*, 2020). The F-value is a representation of the mean square of the divided term by the mean square of the residual term. Consequently, the larger the F-values, the more significant the terms is. From Table 7, the rate of the liquid flow (i.e. 19.14) followed by its interaction with gas pressure (i.e. 11.82) were the more significant factors that affected the methane yield as can also be seen in the pareto chart shown in Fig. 9(a). As for the rate of carbon dioxide removal, a slight inverse of the above-mentioned factors was witnessed followed by the gas pressure and the liquid-liquid flow rate interaction as shown in Fig. 9(b) Both factors that is; pressure and the liquid flow play a key role in the L/G ratio as earlier discussed in section 3.5 above.

With regards to the H_2S removal efficiency, this was greatly affected by the rate of the biogas pressure as the biogas passed through the adsorption column as shown by the pareto chart in Fig. 9(c). Higher pressures tend to let the biogas to escape through the adsorption chamber quicker which renders it less effective in hydrogen sulphide adsorption. A similar behaviour was also reported in a related study (Vakili *et al.*, 2012).

3.8 Surface graphs and analysis.

The interaction effects of three independent factors were studied using 3D plots. The independent factors were considered as study variables to realize their interaction effects on the response value i.e. the CH_4 and CO_2 percentage compositions. The optimization of the response value(s) was carried out using the numerical optimization method following the most significant interaction effect of AC from the attained ANOVA Tables (7 and 8 above). The two-dimensional contours and three-dimensional plots for the optimum CH_4 and CO_2 percentage compositions are shown in Figs. 10 (a) and (b) and 10 (c) and (d) respectively. The optimum methane yield of 84.71% was obtained at gas pressure of 14kPa, steel wool height of 22.86cm and liquid flow rate of 4.2 litres per minute. The corresponding CO_2 percentage under the above-mentioned conditions was 13.31%. The optimum methane yield obtained in this study is lower than the reported study (Noorain *et al.*, 2019) but was higher than other studies (Benizri *et al.*, 2019; Walozi *et al.*, 2016). The profiles from Figs. 10(a) and (b) depict that the CH_4 yield is sensitive to increasing liquid flow rates but only slightly sensitive to pressure.

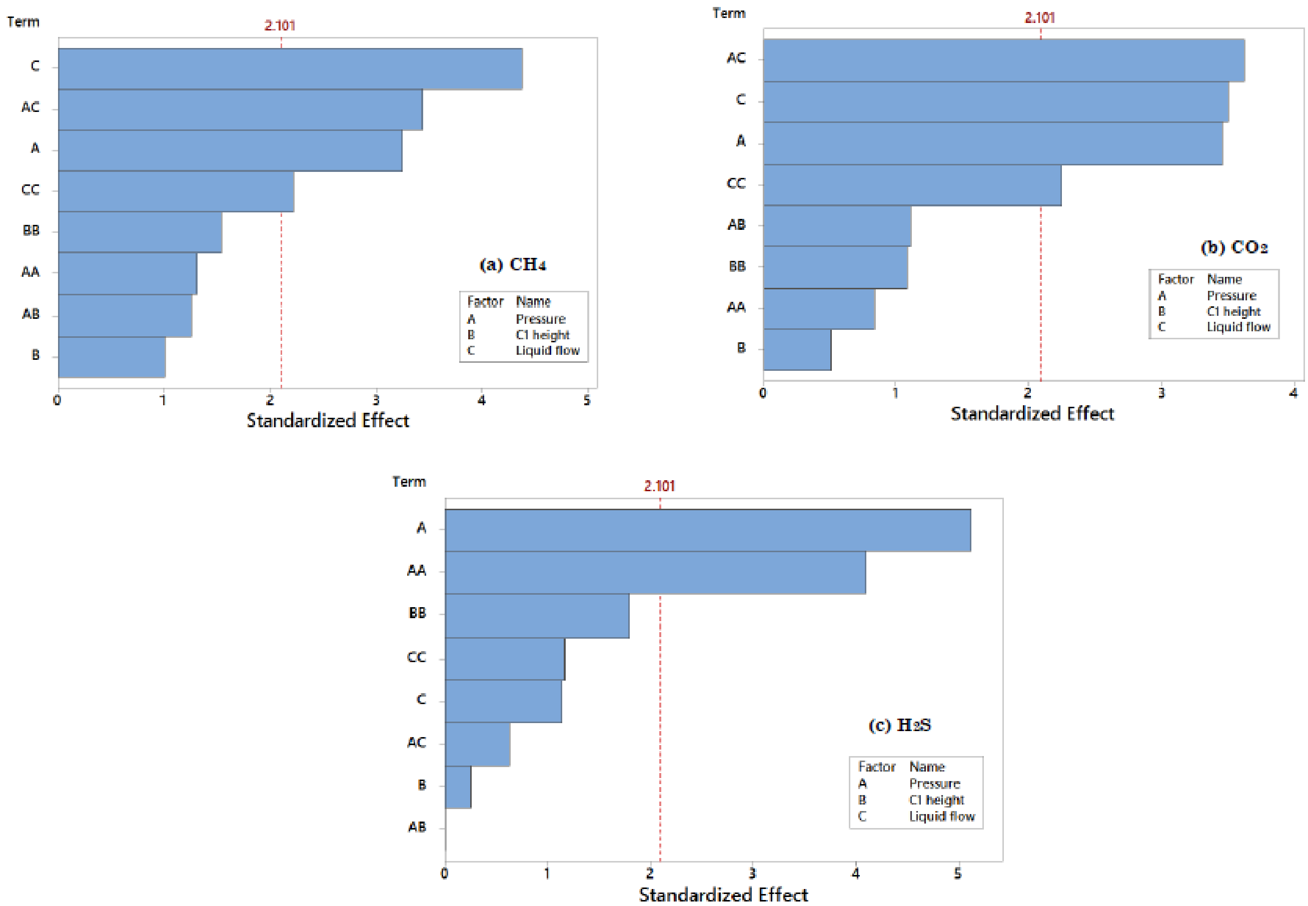

Fig. 9. Pareto charts of the standardized effects from the investigated factors; (a) on methane yield, (b) carbon dioxide removal, (c) hydrogen sulfide removal

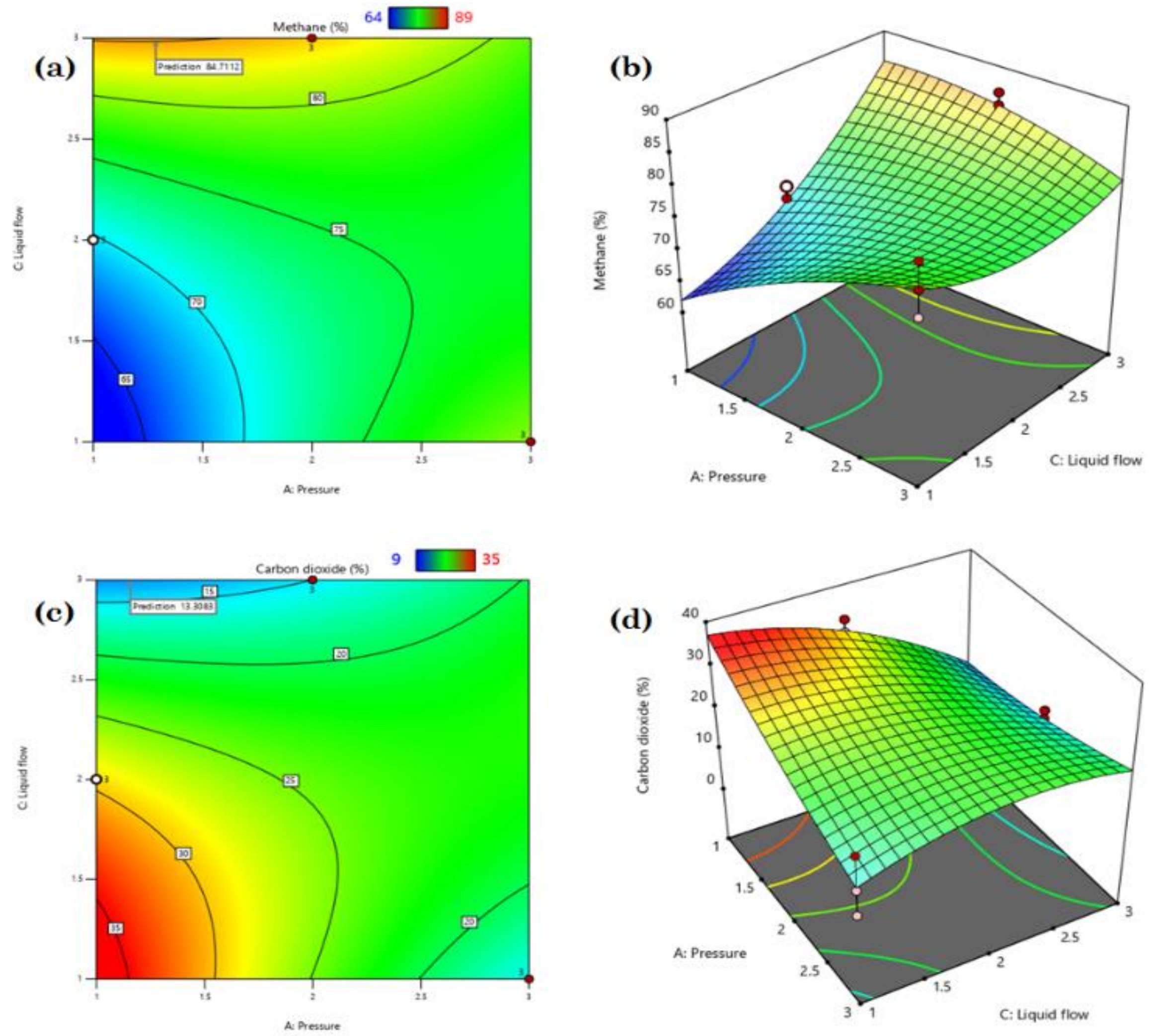

Fig. 10: Pressure – liquid flow interaction on CH_4 yield; (a) 2D Contour plot, (b) 3D surface plot, and on CO_2 removal; (c) 2D Contour plot, (d) 3D surface plot.

From the first-stage optimization using the Taguchi method, the order of factor influence was C > B >A. However, after the second-stage optimization, it changed to C > A > B. This therefore renders the height of the steel wool in the adsorption column relatively less important in the optimization of the CH_4 yield

4. Conclusions.

In this paper, a pilot biogas to bio-methane upgrading system utilizing the NAPWS system (comprising of two columns - the adsorption column packed with steel wool and the absorption column packed with the cascade mini rings, and the dehydrating unit packed with silica gel) was developed and its performance analysed using both the Taguchi and RSM techniques. The optimal performance of the system was investigated with an experimental matrix consisting of 27 experimental runs. Three control factors were varied to attain their rank of influence to the performance characteristic. From the experiments conducted, the purification performance of the system revealed the possibility of up-grading raw biogas from 53-63 (%v/v) to an optimum bio-methane concentration of 84.71 (%v/v) while reducing hydrogen sulfide by 87% to less than 13 ppm. This experimental study differs from existing experiments as it; investigated the upgrading performance using the CMR® with the NAPWS system which revealed superior specific flow parameters and mass transfer characteristics, revealed the possibility of good purification characteristics even for a short column other than the conventional long columns. The ranking of the studied factors was found to be in the order of; the water flow rate, the system pressure and the height of the steel wool respectively. The optimal performance of the system was attained at gas pressure of 14kPa, steel wool height at 22.86cm and liquid flow rate of 4.2 litres per minute. This study further demonstrates that the use of lower gas pressure has benefits of lowering the power requirements and the overall capital expenditure as opposed to the conventional high pressure water scrubbing systems. It can be concluded that the developed low cost biogas upgrading system has great potential for bio-methane production for domestic cooking and heating applications in developing countries with further research in using scrubbing water at temperatures less than 10^0C.

Acknowledgments

The first author acknowledges the support from Kenyatta University Energy Department and the German Academic Exchange Service (DAAD) for the scholarship.

Author Contributions: G.R.M.: Conceptualization, methodology, formal analysis, writing—original draft, I.B.O.; supervision, editing, validation, resources, T.T; writing—review, methodology, supervision and editing. All authors have read and agreed to the published version of the manuscript.

Conflicts of Interest: The authors declare no conflict of interest.

References

Abdeen, F. R. H., Mel, M., Jami, M. S., Ihsan, S. I., & Ismail, A. F. (2018). Improvement of Biogas Upgrading Process using Chemical Absorption at Ambient Conditions. *Jurnal Teknologi*, *80*(1), 107–113. https://doi.org/ 10.11113/jt.v80.10382

Armah, E., Chetty, M., & Deenadayalu, N. (2020). Biogas production from sugarcane bagasse with South African industrial wastewater and novel kinetic study using response surface methodology. *Scientific African*, *10*. https://doi.org/10.1016/j.sciaf.2020.e00556

Awe, O. W., Zhao, Y., Nzihou, A., Minh, D. P., & Lyczko, N. (2017). A Review of Biogas Utilisation, Purification and Upgrading Technologies. *Waste and Biomass Valorization* *8*(2), 267–283. https://doi.org/10.1007/s12649-016-9826-4

Bauer, F., Hulteberg, C., Persson, T., & Tamm, D. (2013). *Biogas upgrading – Review of commercial technologies. Swedish Gas Center Report SGC 2013:270 Available at* http://vav.griffel.net/filer/C_SGC2013-270.pdf (Last access August 2017).

Benizri, D., Dietrich, N., Labeyrie, P., & Hébrard, G. (2019). A compact, economic scrubber to improve farm biogas upgrading systems. *Separation and Purification Technology*, *219*, 169–179. https://doi.org/10.1016/j.seppur.2019.02.054

Budzianowski, W. M., Wylock, C. E., & Marciniak, P. A. (2017). Power requirements of biogas upgrading by water scrubbing and biomethane compression: comparative analysis of various plant configurations. *Energy Conversion and Management*, *141*, 2–9. doi: https://doi.org/10.1016/ j.enconman.2016.03.018

Djimtoingar, S. S., Derkyi, A., Kuranchie, F., & Yankyera, J. K. (2022). A review of response surface methodology for biogas process optimization. *Cogent Engineering*, *9*(2115283). https://doi.org/10.1080/23311916.2022.2115283

Gantina, T. M., Iriani, P., Maridjo, & Wachjoe, C. K. (2020). Biogas purification using water scrubber with variations of water flow rate and biogas pressure. *Journal of Physics: Conference Series*, *1450*(012011). https://doi.org/10.1088/1742-6596/1450/1/012011

Hegely, L., Roesler, J., Alix, P., Rouzineau, D., & Meyer, M. (2017). Absorption methods for the determination of mass transfer parameters of packing internals: A literature review. *AIChE Journal,* 63(8), 3246–3275; https://doi.org/10.1002/aic.15737.

Ilyas, S. Z. (2006). A Case Study to Bottle the Biogas in Cylinders As Source of Power for Rural Industries Development in Pakistan. *World Applied Sciences Journal*, *1*(2), 127–130. https://doi.org/10.1016/0960-1481(96)88447-3

IRENA. (2016). *Measuring small-scale biogas capacity and production*. International Renewable Energy Agency, Abu Dhabi.

Kapoor, R., Ghosh, P., Kumar, M., & Vijay, V. K. (2019). Evaluation of biogas upgrading technologies and future perspectives: a review. *Environmental Science and Pollution Research*. https://doi.org/10.1007/s11356-019-04767-1

Kapoor, R., Subbarao, P. M. V, Kumar, V., Shah, G., Sahota, S., Singh, D., & Verma, M. (2017). Factors affecting methane loss from a water scrubbing based biogas upgrading system. *Applied Energy*, *208*, 1379–1388. https://doi.org/10.1016/j.apenergy.2017.09.017

Kister, H. Z., Mathias, P. M., Steinmeyer, E. D., Penney, W. R., Crocker, B. B., & Fair, J. R. (2008). Equipment for Distillation, Gas Absorption, Phase Dispersion, and Phase Separation. In D. W. Green & R. H. Perry (Eds.), *Perry's Chemical Engineers' Handbook* (8th ed., pp. 14-1-14–57). McGraw-Hill.

Kolev, N., Nakov, S., Ljutzkanov, L., & Kolev, D. (2006). Comparison of the Effective Surface Area of Some Highly Effective Random Packings Third and Forth Generation. *IChemE Symposium series*, *152*, 754–763; https://folk.ntnu.no/skoge/prost/proceedings/distillation06/CD-proceedings/paper073.pdf

Madondo, N. I., Rathilal, S., & Bakare, B. F. (2022). Utilization of Response Surface Methodology in Optimization and Modelling of a Microbial Electrolysis Cell for Wastewater Treatment Using Box – Behnken Design Method. *Catalysts*, *12*(1052). https://doi.org/https:// doi.org/10.3390/catal12091052.

Manmai, N., Unpaprom, Y., & Ramaraj, R. (2020). Bioethanol production from sunflower stalk: application of chemical and biological pretreatments by response surface methodology (RSM). *Biomass Conversion and Biorefinery*. https://doi.org/10.1007/s13399-020-00602-7

Manmai, N., Unpaprom, Y., Ponnusamy, V., & Ramaraj, R. (2020). Bioethanol production from the comparison between optimization of sorghum stalk and sugarcane leaf for sugar production by chemical pretreatment and enzymatic degradation. *Fuel*, *278*, 118262. https://doi.org/10.1016/j.fuel.2020.118262

Mel, M., Ibrahim, M. M. A., & Setyobudi, R. H. (2016). Preliminary study of biogas upgrading and purification by pressure swing adsorption. *AIP Conference Proceedings*, *1755*, 130010–130011–130010–130015. https://doi.org/10.1063/1.4958554

Mohanakrishnan, L., & Kurian, J. (2016). Chemical Scrubbing for removal of CO_2 from Biogas using Algae and H_2S using Sponge Iron. *International Journal of Renewable Energy and Environmental Engineering*, *4*(2), 35–41.

Montgomery, D. C., & Runger, G. C. (2003). *Applied Statistics and Probability for Engineers* (W. Anderson, J. Welter, & N. M. Pigliucci (eds.); 3rd ed.). John Wiley & Sons, Inc. https://doi.org/10.1080/03043799408928333.

Morero, B., Groppelli, E., & Campanella, E. (2017). Evaluation of biogas upgrading technologies using a response surface methodology for process simulation Related papers. *Journal of Cleaner Production*, *141*, 978–988. https://doi.org/10.1016/j.jclepro.2016.09.167.

Nguyen, K. D. M., Imai, T., Yoshida, W., Dang, T. L., Higuchi, T., Kanno, A., Yamamoto, K., & Sekine, M. (2017). Performance of a Carbon Dioxide Removal Process Using a Water Scrubber with the Aid of a Water-Film-Forming Apparatus. *Waste and Biomass Valorization*. https://doi.org/10.1007/s12649-017-9951-8

Nock, W. J., Walker, M., Kapoor, R., & Heaven, S. (2014). Modeling the water scrubbing process and energy requirements for CO_2 capture to upgrade biogas to biomethane. *Industrial and Engineering Chemistry Research*, *53*, 12783–12792. https://doi.org/10.1021/ie501280p

Noorain, R., Kindaichi, T., Ozaki, N., Aoi, Y., & Ohashi, A. (2019). Biogas purification performance of new water scrubber packed with sponge carriers. *Journal of Cleaner Production*, *214*(20), 103–111. https://doi.org/10.1016/j.jclepro.2018.12.209

Pirola, C., Galli, F., Manenti, F., & Bianchi, C. L. (2015). Biogas Upgrading by Physical Water Washing in a Micro-Pilot Absorption Column Conducted at Low Temperature and Pressure. *Chemical Engineering Transactions.*, *43*, 1207–1212. doi: https://doi.org/10.3303/CET1543202

Rajivgandhi, M. M. C., & Singaravelu, M. (2014). Upgrading Biogas to Biomethane by Physical Absorption Process. *International Journal of Agriculture, Environment and Biotechnology*, *7*(3), 639. https://doi.org/10.5958/2230-732x.2014.01370.9

Rasi, S., Lantela, J., Veijanen, A., & Rintala, J. (2008). Landfill gas upgrading with countercurrent water wash. *Waste Management*, *28*(9), 1528–1534. https://doi.org/10.1016/j.wasman.2007.03.032

Riyadi, U., Kristanto, G. A., & Priadi, C. R. (2018). Utilization of steel wool as removal media of hydrogen sulfide in biogas. *IOP Conference Series: Earth and Environmental Science*, *105*(1). https://doi.org/10.1088/1755-1315/105/1/012026

Shen, Y., Shi, W., Zhang, D., Na, P., & Fu, B. (2018). The removal and capture of CO_2 from biogas by vacuum pressure swing process using silica gel. *Journal of CO_2 Utilization*, *27*, 259–271. https://doi.org/10.1016/j.jcou.2018.08.001

Singhal, S., Agarwal, S., Arora, S., Sharma, P., & Singhal, N. (2017). Upgrading techniques for transformation of biogas to bio-CNG: a review. *International Journal of Energy Research*. https://doi.org/10.1002/er.3719

Song, C., Kitamura, Y., & Li, S. (2014). Optimization of a novel cryogenic CO_2 capture process by response surface methodology (RSM). *Journal of the Taiwan Institute of Chemical Engineers*, *45*(4), 1666–1676. https://doi.org/10.1016/j.jtice.2013.12.009

Tayar, S. P., Guerrero, S., Hidalgo, L. F., & Denise Bevilaqua. (2019). Evaluation of Biogas Biodesulfurization Using Different Packing Materials. *ChemEngineering*, *3*(27), 1–12. https://doi.org/10.3390/chemengineering3010027

Tippayawong, N., & Thanompongchart, P. (2010). Biogas quality upgrade by simultaneous removal of CO_2 and H_2S in a packed column reactor. *Energy*, *35*(12), 4531–4535. https://doi.org/10.1016/j.energy.2010.04.014

Tran, L. T., Le, T. M., Nguyen, T. M., Tran, Q. T., Le, X. D., Pham, M. Q., Lam, V. T., & Do, M. Van. (2021). Simultaneous removal efficiency of H_2S and CO_2 by high - gravity rotating packed bed: Experiments and simulation. *Open Chemistry*, *19*, 288–298. https://doi.org/10.1515/chem-2020-0187

Vakili, M., Gholami, Z., & Gholami, F. (2012). Removal of Hydrogen sulfide from gaseous streams by a chemical method using ferric sulfate solution. *World Applied Sciences Journal*, *19*(2),241–245. https://doi.org/10.5829/idosi.wasj.2012.19.02.1541

Vijay, V. K., Chandra, R., Subbarao, P. M. V, & Kapdi, S. S. (2006). Biogas Purification and Bottling into CNG Cylinders: Producing Bio-CNG from Biomass for Rural Automotive Applications. *The 2nd Joint International Conference on "Sustainable Energy and Environment (SEE 2006)" 21-23 November, Bangkok, Thailand.*

Walozi, R., Nabuuma, B., & Sebiti, A. (2016). Application of Low Pressure Water Scrubbing Technique for Increasing Methane Content in Biogas. *Universal Journal of Agricultural Research*, *4*(2), 60–65. https://doi.org/10.13189/ujar.2016.040206

Wang, K., Chen, J., Huang, Y., & Huang, S. (2013). Integrated Taguchi method and response surface methodology to confirm hydrogen production by anaerobic fermentation of cow manure. *International Journal of Hydrogen Energy*, *38*, 45–53. https://doi.org/10.1016/j.ijhydene.2012.03.155

Yousef, A. M. I., Eldrainy, Y. A., El-Maghlany, W. M., & Attia, A. (2016). Upgrading biogas by a low-temperature CO_2 removal technique. *Alexandria Engineering Journal*, *55*, 1143–1150. https://doi.org/10.1016/j.aej.2016.03.026