Optimizing Anaerobic Digestion of a Local South African Municipal Wastewater Using Box-Behnken Design of Response Surface Methodology

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***Abstract* —** in recent years, water and electricity consumption and demand have increased on both national and international levels. This has resulted in an increase in wastewater generation and greenhouse gas emissions due to inadequate waste management systems. In addressing these challenges, the application of anaerobic digestion (AD) presents a robust solution for the treatment of wastewater with the benefits of biogas production if operated efficiently. In this study, a lab-scale 1 L reactor operated under anaerobic conditions at 37.5 °C was employed for the treatment of local South African municipal wastewater. The influence and synergistic effect of influent pH (5-9), hydraulic retention time (HRT, 15-30 days), and magnetite load (0.125- 0.875 g/L) were operating variables considered as a function of biogas produced and contaminants removal percentage (colour and COD). The experimental matrix was generated and conducted based on the Box-Behnken design (BBD) of the response surface methodology (RSM). The RSM analysis of variance (ANOVA) confirms that the quadratic models obtained are substantial, with a high coefficient of regression (R^2) above 0.98 and p-values below 0.05 for each variable. The optimum conditions of pH of 7.04, magnetite load of 0.42 g/L and HRT of 21 days were obtained. This resulted in desirability of 84% inferring a biogas yield of 93.72 mL/TDS, COD removal of 88.65% and colour removal of 97.55%. The post-analysis of the effluent showed 80 - 98% reduction in the volatile solids, which suggests that the high organic content of wastewater was anaerobically converted into biogas.

***Keywords*—** Anaerobic digestion, wastewater, response surface methodology, Box-Behnken design, magnetite, biogas.

I. INTRODUCTION

Water scarcity and high energy demand have become two of South Africa’s major concerns (Du Plessis and du Plessis 2019). Several communities in South Africa encounter challenges in obtaining consistent and sufficient quantities of potable water and energy to meet their daily needs. This occurs in the context of a declining supply of fresh water and a rising water demand (Adewumi *et al.* 2010). There is a growing interest in the reutilization of wastewater to meet the needs of non-potable water users. Reuse of effluent entails its collection and treatment to qualify it for specific applications. Reusing wastewater can become a significant element in water resource management and wastewater management, providing an environmentally sustainable alternative that mitigates the harmful impacts on surface waters caused by wastewater discharge. As a result of these issues, clean water and clean energy production technologies have attracted both national and global interest. Municipal wastewater treatment facilities have the potential to become net producers of renewable energy by converting the chemically bonded energy content in organic contaminants of municipal wastewater to a usable energy carrier (biogas) while generating clean water for surrounding communities (Sikosana *et al.* 2019). Water is required for energy generation, particularly in coal mining and power generation (cooling coal plants). In turn, water treatment and delivery all necessitate the use of energy. As a result, attempts to create and grow power generation must be balanced with implications of water usage and water footprint associated with power generation.

The application of anaerobic digestion (AD) in the treatment of wastewater faces challenges arising from emerging contaminants. These unidentified contaminants were not incorporated into the design of the existing conventional water treatment facilities including ADs. These turned into a potential danger to our water distribution system. The existing wastewater treatment system is inadequate in eliminating all of these different types of emerging contaminants with unknown characteristics (Rout *et al.* 2021; Sahani *et al.* 2022). In addition to emerging contaminants, energy consumption during the treatment of wastewater has become a major concern. Previously, anaerobic digestion was predominantly used to stabilize sewage sediment; however, in recent times, there has been an increased focus on optimizing energy recovery (Batstone and Virdis 2014; Silvestre *et al.* 2015).

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Therefore, water resources and water consumption should consequently be included in energy generation design. Additionally, predicted growth in population, climate change and urbanization all demand a more comprehensive strategy (Mabhaudhi *et al.* 2016). This study aimed to optimize operating conditions for anaerobic digestion of wastewater to produce clean water for reuse and biogas.

II. MATERIALS AND METHODS

A. Chemicals and wastewater samples

Magnetic nanoparticles (magnetite) used were prepared in-house using the co-precipitation technique and characterized as reported by (Amo-Duodu *et al.* 2021; Tetteh *et al.* 2021; Ngema *et al.* 2023). 3M solutions for hydrochloric acid and sodium hydroxide were prepared to control the pH of mixed liquor in the digester.

Wastewater and activated sludge were sourced from the Umgeni wastewater treatment plant located in Pietermaritzburg, Kwa-Zulu Natal. A Hach spectrophotometer was used to examine the physicochemical properties of wastewater. The presence of organic content (4944 mg COD/L), nutrients and volatile solids warrants anaerobic degradation of this wastewater to produce methane-rich biogas and clean water for re-use.

B. Experimental setup and procedure

The Box-Behnken design (BBD) of the response surface methodology was employed for the design of the experimental matrix. The operating parameters were investigated at three levels defined as low, optimum and high. The BBD defined 15 possible combinations for experimentation which will lead to obtaining optimum conditions for the digestion of this industrial wastewater. The set-up used consisted of a 1L digester heated using a hot plate to keep the temperature inside at 37.5 ˚C. The treated effluent was sampled at the top port for analysis and biogas generated was passed through a scrubber unit to minimize impurities and stored in a small vertical cylinder.

III. RESULTS AND DISCUSSION

In anaerobic digestion (AD), the bacteria remove contaminants via biodegradation of organic contents to produce methane-rich biogas and reduce costs for effluent and sludge discharge. At the end of AD, the effluent was characterized and the removal percentages for the contaminants of interest are summarized in Table 1. The COD removal percentage is a key indicator of the performance of the AD, indicating how much organic content was converted by anaerobic bacteria into biogas. The COD removal ranged from 59% to 92% for all the 15 experimental combinations investigated. This suggests that in experiment 15, where 92% COD removal was achieved, microorganisms were exposed to an environment that promotes growth and stimulates their activity at a faster rate to degrade organic content to produce methane. Volatile solids also indicate the biodegradability of the organics in wastewater as they consist of solids from plants and other organic matter. The reduction of up to 98% in volatile solids indicates the suitability of the digester to digest the solids with other contaminates in wastewater to produce reusable water and biogas. Colour removal is an important factor in measuring the quality of effluents being discharged to the environment or for reuse. It is quite evident that colour removal is affected by both the digestion and settleability of free solids in the effluent. In this study, the application of magnetic nanoparticles permitted microorganisms to degrade contaminants (dissolved and non-dissolved) and longer retention times allowed free solids to settle due to gravity while dissolved are being digested and hence higher removal for colour was achieved across the experiments.

TABLE I

STATISTICAL ANALYSIS POST ANAEROBIC DIGESTION

Run	%VSS reduction	COD removal	Colour removal
1	93,05	89,78	96,79
2	81,39	78,24	97,66
3	78,30	68,62	95,45
4	74,77	74,29	95,65
5	76,98	81,24	99,46
6	78,14	63,36	99,13
7	83,26	87,25	89,29
8	87,58	84,51	95,72
9	77,67	69,23	91,43
10	82,09	70,24	98,93
11	75,81	81,48	93,98
12	75,44	59,31	95,45
13	79,88	59,92	94,11
14	89,53	85,43	94,58
15	98,14	92,11	98,13

After the experimental tests are conducted, the Analysis of Variance (ANOVA) test is employed to analyze the influence of the combination of input variables on the response variables. The fit summary table in ANOVA is a statistical table that may be used for the selection of the type of regression model. This table provides a collection of substantial parameters that are employed as the starting point for the true regression equation. Table 1 and 2 portrays the fit summary for COD removal and colour removal respectively. In Design Expert software, the types of regression equations that are considered are quadratic, cubic, two-factor interaction (2FI), and linear regression equations. The regression equation with the greatest lack-of-fit p-value, predicted R^2, adjusted R^2, F-value, and p-value in statistical analysis is considered the best-fit model (Stat-Ease Inc., 2019). The quadratic regression equation revealed the greatest lack-of-fit p-value, adjusted R^2, and predicted R^2 for both responses, COD removal and colour removal. It was then deduced that the best-fit regression models for all responses were the quadratic equation.

TABLE II
FIT SUMMARY STATISTICS FOR COD REMOVAL

Source	p-value	Lack of fit	Adjusted R^2	Predicted R^2	
Linear	0.9238	0.0005	-0.2209	-0.6678	
2FI	0.9599	0.0004	-0.6202	-2.3052	
Quadratic	< 0.0001	0.439	0.999	0.9958	Suggested
Cubic	0.439		0.9992		Aliased

TABLE III
FIT SUMMARY STATISTICS FOR COLOUR REMOVAL

Source	p-value	Lack of fit	Adjusted R^2	Predicted R^2	
Linear	0.0816	0.0032	0.2917	0.0711	
2FI	0.5805	0.0027	0.2343	-0.2245	
Quadratic	< 0.0001	0.5815	0.9975	0.9910	Suggested
Cubic	0.8656		0.9972		Aliased

After the regression model has been suggested, quadratic equations were developed to predict these responses theoretically for anaerobic digestion. Equations (a) represent COD removal and (b) represents colour removal where factors A, B and C indicate hydraulic retention time, magnetite load and pH respectively. For the quadratic equation (a), it was deduced that magnetite load (B) and pH (C) have a positive influence while the combination of the three input factors have a negative influence in the removal of COD, whereas, for the model (b), all the input factors have positive impact in colour removal and the combination of A and C have the greatest influence with the coefficient of 0.6022.

$$(a)\quad COD\ removal = 89{,}93 - 1{,}69A + 0{,}1498B + 2{,}59C - 0{,}5096AB - 0{,}8698AC - 2{,}71BC - 4{,}49A^2 - 1{,}63B^2 - 19{,}18C^2$$

$$(b)\quad Colour\ removal = 98{,}24 + 0.9702A + 0{,}2761B + 0{,}4360C + 0.5745AB + 0.6022AC - 0.2477BC - 1.02A^2 - 1.11B^2 - 0.4241C^2$$

The results show that pH and magnetite load have a significant effect on COD and colour removal because they affect the activity of bacteria to destroy organic matter into biogas. A low pH in the digester inhibits the activity of microorganisms involved in the digestion process particularly methanogenic bacteria (Guštin and Marinšek-Logar 2011). Various studies confirmed that the optimal pH for anaerobic digestion of wastewater ranges from 6.5 to 8 (Jayaraj *et al.* 2014).

The regression model equation needs to be verified for significance and practical applications. There are numerous ways for model verification by the Design Expert, and this is attained by performing a diagnostic test (Stat-Ease Inc., 2019). The predicted versus actual graph is one of the significant graphs in model validation. Figure 1 and Figure 2 portray the predicted versus actual graphs for COD removal and colour removal. The observational points should be linearly scattered beside the 45-degree line of the predicted versus actual response figure (Stat-Ease Inc., 2019). The data points of all responses (namely, COD removal, and colour removal) were linearly scattered along the 45-degree straight line, suggesting that the regression models were more or less able to predict the true data points.

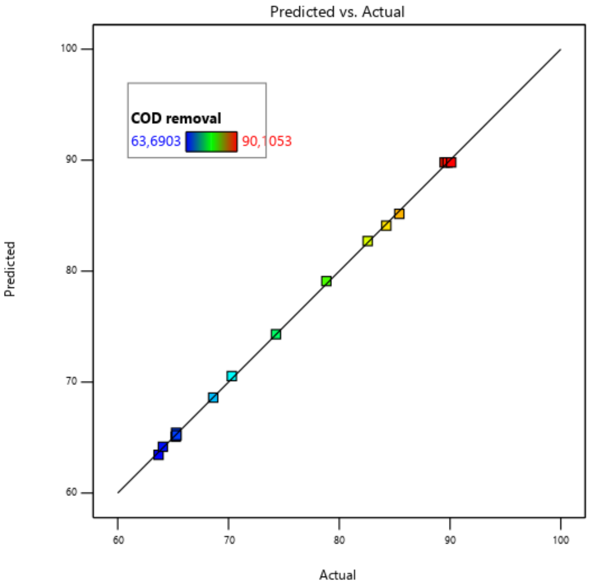

Fig. 1 Predicted versus actual data plot for COD removal.

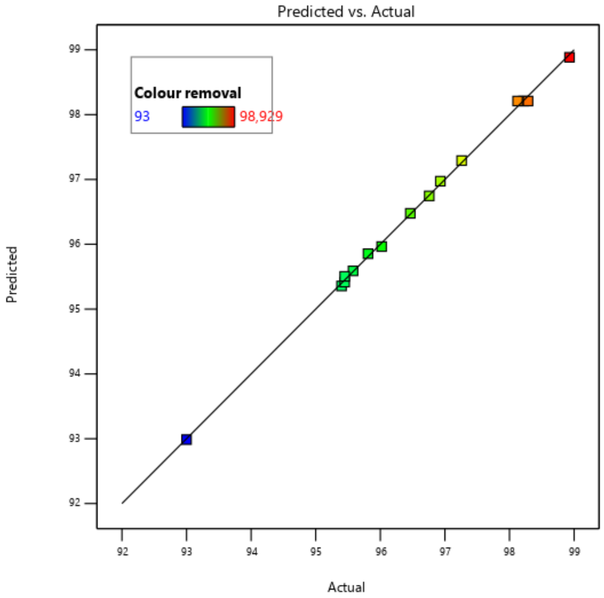

Fig. 2 Predicted versus actual data plot for colour removal.

After the model was analyzed and validated, numerical optimization was conducted to obtain optimum operating conditions for the anaerobic treatment of the local wastewater. The goal was to optimize the removal of colour and COD and also improve the recovery of biogas while operating within the range of the investigated input parameters. The response surface methodology was utilized to optimize the AD and one solution with a desirability of 0.94 was obtained. The predicted optimum conditions for the input variables namely HRT, MNP load and pH were found to be 21 days, 0.42 g/L and 7.04 respectively. Table 4 summarizes and compares the predicted optimum condition (inputs and outputs) and optimum outputs achieved during experimental validation of the predicted optimum conditions. The difference between predicted outputs and validated outputs was found to be below 2% for all responses, which suggests that the predicted optimum conditions can be applicable during AD of municipal wastewater.

TABLE IV
OPTIMIZATION AND VALIDATION

INPUTS		OUTPUTS		
Factors	**Optimum**	**Factors**	**Optimum**	**Validation**
HRT (day)	21	$\%R_{Colour}$	98	96.79
MNP (g/L)	0.42	$\%R_{COD}$	90.1	89.41
pH	7.04	Biogas (mL)	100.04	108.2

The affluent and the digested sludge post-anaerobic digestion at optimum conditions were characterized. The aim was to identify whether the effluent is suitable for discharge based on wastewater discharge regulations. The effluent was found to be clear in colour with normal smell, the analyzed contaminants were pH of 6.98, COD of 68 mg/L, turbidity of 21.74 NTU and colour of 47.9 Pt-Co.

Although optimization of anaerobic digestion allows for the production of clean water and improved biogas, digested sludge is another product that is also produced and discharged to the environment. Figure 3 portrays an SEM image of sludge from conventional digestion, figure 4 represents sludge digested with the addition of magnetite and controlled pH. In Figure 3, sludge particles appear to have formed a solid shape or lumps with the flat surface while in Figure 4 it has a greater surface. The increased surface area as a result of magnetic nanoparticles allows for greater contact between solids and liquid during AD, making it easy for microorganisms to access organic matter for digestion, and thus increases the performance of AD. The small surface area shown in Figure 3 makes it difficult for microorganisms to access organics and hinders contact between solids and liquids. Additionally, the large surface area allows for the use of sludge material into fertilizers and it is easy to handle for further processing or drying and disposal. Landfilling and land application of the sewage sludge are suggested to be the most economical sludge disposal methods. The application of municipal sludge to land is highly advantageous due to its fertilizing and soil-conditioning characteristics, provided that it does not contain any hazardous substances (Singh and Agrawal 2008; Usman *et al.* 2012).

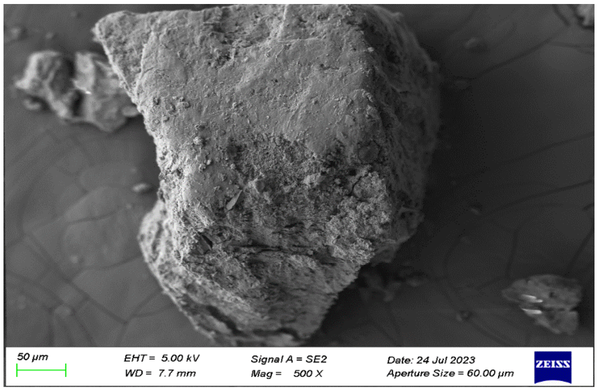

Fig. 3 SEM image for conventional sludge

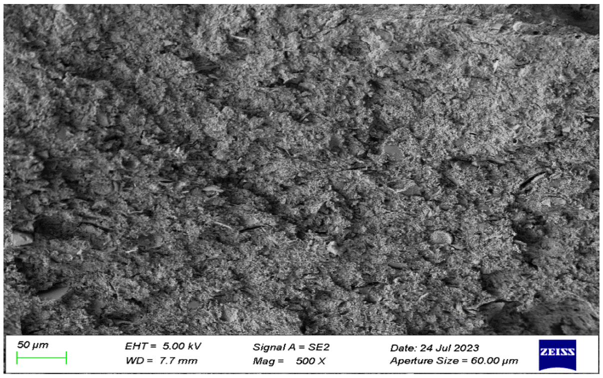

Fig. 4 SEM image for digested sludge at optimum AD conditions

IV. CONCLUSION

The urgency to optimize anaerobic digestion is necessitated by the increased demand for access to clean water to meet daily demands. In this study, it was evident that optimizing the operating conditions of the AD enhances both the removal of contaminants and biogas production compared to conventional AD. The optimum conditions achieved for the digestion of municipal wastewater were HRT of 21 days, pH of 7.04 and magnetite load of 0.42 g/L. The use of magnetic nanoparticles has proved to stimulate microbial activity at a faster rate resulting in improved AD at a faster rate. The impact of controlling both pH and nanoparticle load helps to create an atmosphere that is suitable for microorganisms and enhances methane production. The optimum conditions were found feasible for practical application and are currently investigated/applied at a larger scale for industrial application.

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