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Generalization of artificial neural network for predicting methane production in laboratory-scale anaerobic bioreactor landfills

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ABSTRACT

BACKGROUND AND OBJECTIVES: Leachate recirculation has become a global practice for anaerobic digestion of municipal solid waste. Implementation of artificial neural networks for modeling and prediction of this process still remains challenging. Additionally, there has been a lack of research regarding the generalization capacity of neural networks using the data from other studies. This study aimed to enhance methane production rates and decrease biostabilization time in municipal solid waste treatment. It addressed the research gap in applying and generalizing neural networks to predict biogas production based on laboratory-measured parameters.

METHODS: Two distinct systems were utilized for leachate treatment. System 1 involved collecting the leachate delivered by a new municipal solid waste reactor and transferring it to a recirculation tank. System 2 consisted of passing the fresh municipal solid waste leachate through a degraded municipal solid waste and then returning the obtained liquid back to the waste reactor. The experimental data were employed to develop an artificial neural network to predict methane content and cumulative biogas production. The model was trained and optimized using the experimental data. The effectiveness and generalizability of the optimal neural network were evaluated by using it for the unseen data from other studies, ensuring its ability to make accurate predictions beyond the training dataset.

FINDINGS: The results demonstrated that in System 1, ammonium and chemical oxygen demand concentrations in the leachate progressively increased to high levels. In System 2, the average removal efficiencies for chemical oxygen demand and ammonium were found to be 85 percent and 34 percent respectively. The methane yield in biogas reached 59 liters per kilogram of dry weight, with a corresponding methane fraction of 63 percent. The neural network model showed an excellent performance, with validation performances of 0.716 and 0.634. The overall performance of the dataset resulted in correlation coefficients of 0.9991 and 0.9975. Finally, high correlation coefficients of 0.88 and 0.82 were achieved by incorporating the test data from other studies.

CONCLUSION: Leachate recirculation enhanced the reduction of chemical oxygen demand and the production of methane in bioreactors. Ammonium concentrations initially increased and later decreased due to waste adsorption and bacterial assimilation. The artificial neural network applied for predicting the cumulative methane production from municipal solid waste displayed a robust generalizability when tested on the data from other studies. The neural network was not significantly affected by changes in waste chemical properties, laboratory conditions, and recirculation rate. However, it showed a significant sensitivity to variation of waste mechanical properties.

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NUMBER OF REFERENCES	NUMBER OF FIGURES	NUMBER OF TABLES
63	9	5

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INTRODUCTION

Waste landfills are the ultimate repositories for discarded or unusable materials, particularly municipal solid wastes (MSW). For instance, approximately 81 million tons of waste are landfilled annually in the UK. In 2016, 58 percent (%) of the total solid waste generated in the US was disposed (Alabi *et al.,* 2019). Leachate production and management are acknowledged as significant obstacles associated with the environmentally sound operation of municipal landfills. Recirculating leachates through MSW landfill accelerates solid waste stabilization and, consequently, increases gas production (Liu *et al.,* 2023). Over the past two decades, numerous physical, chemical, and biological treatment processes have been evaluated for their ability to treat landfill leachate (Bah *et al.,* 2023; Guo *et al.,* 2022; Luo *et al.,* 2020). These processes are typically employed for *ex-situ* leachate treatments. Nevertheless, treating leachate *ex-situ* can pose significant challenges and incur substantial costs. Moreover, the characteristics and flow of landfill leachates are influenced by some factors such as composition of solid wastes, precipitation and runoff patterns, landfill age, and permeability and type of cover (Luo *et al.,* 2020). Most leachate components are typically present in elevated concentrations during the first year of landfill operation, and these concentrations tend to decrease as the landfill ages (Kulikowska and Klimiuk, 2008). High levels of ammonia and organic matter in landfill leachate lead to significant treatment challenges (Samimi and Shahriari Moghadam, 2018). There are numerous options for landfill leachate treatment, such as complex and expensive *ex-situ* physical-chemical and biological processes, that address high-strength organics and inorganics including different forms of nitrogen. Numerous studies have documented various leachate treatments, including anaerobic sequencing batch reactors and anaerobic hybrid bed filters (Wei *et al.,* 2021), upflow sludge blanket reactors (Govahi *et al.,* 2012), and electro-Fenton method (Guvenc *et al.,* 2019). These treatment procedures can incur substantial costs. Biological processes have proved to be highly effective when applied to relatively young leachates consisting primary volatile fatty acids, but their effectiveness decreases when applied to older leachates (Bove *et al.,* 2015). Numerous scholars have conducted extensive research documenting the benefits of leachate recirculation in sanitary landfills. According to studies, leachate recirculation generates stabilized leachates with relatively low concentrations of degradable carbon compounds and high concentrations of ammonia (Haydar and Khire, 2005; Hussein and Ibrahim, 2023). In case of biological degradation, the analytical parameters involved exhibit non-linear characteristics (Ćosić *et al.,* 2013). Artificial neural network (ANN) techniques have demonstrated a greater efficiency in accurately modeling these non-linear relationships compared to traditional statistical methods (Desai *et al.,* 2018; Rumaling *et al.,* 2022; Samimi and Mohadesi, 2023). ANNs have become increasingly popular as a useful tool for modeling the environmental systems (Muksin *et al.,* 2023). They have been widely applied in different domains, including air pollution modeling (Cabaneros *et al.,* 2019) and predicting the performance of wastewater treatment plants (El-Rawy *et al.,* 2021). ANNs, however, have not been extensively studied in terms of laboratory settings for anaerobic digestion (Nair *et al.,* 2016; Tufaner and Demirci, 2020). In a study, the utilization of ANN was explored to forecast biogas production and chemical oxygen demand (COD) removal rates in the process of anaerobic digestion (Nair *et al.,* 2016). The results of this experiment demonstrated the effectiveness of the ANN method in accurately predicting biogas production and COD removal rates. Another study indicated a strong correlation between the age of waste and the methane (CH_4) concentration, which was successfully modeled using an ANN (Ozkaya *et al.,* 2007). Additionally, a separate study proposed an ANN approach to simulate the functionality of a biogas wastewater treatment system, accurately predicting the relationship between the system output and its operational parameters (Karamichailidou *et al.,* 2022). An ANN model developed by Behera *et al.* (2015) was utilized to predict the CH_4 concentration in biogas. The input data from this model consisted of the biogas extraction rate and the ratio of landfill leachate to food waste leachate.The results of this study showed that the backpropagation algorithm effectively predicted the percentage of CH_4 in biogas. Despite the utilization of only two input parameters, the ANN model demonstrated a remarkably high prediction accuracy. This could be attributed to the inclusion of the biogas extraction rate as an input, which had a direct relationship with CH_4 production.

The utilization of ANN was justified by its capability to comprehend intricate non-linear relationships, while the association between biogas extraction rate and CH_4 production was characterized by a simple non-linear pattern**.** In a recent study, Bao *et al.* (2023) employed a backpropagation ANN to develop a model for optimizing anaerobic digestion. Their findings indicated that the model successfully achieved a high degree of fitting with the actual data, indicating its accuracy in predicting the biogas production. This finding highlighted the practical application value of the model in anaerobic digestion. However, it is important to note that the study included an excessive number of factors in predicting biogas production, involving ten parameters. One of the primary objectives of ANN design is accurate output prediction with minimal data requirements. The selection of input data plays a critical role in determining the applicability, economy, and accuracy of ANNs. The present study emphasized the importance of selecting the best input data for ANNs based on experimental results and previous studies. The current study explored two distinct anaerobic systems as *in-situ* organic and nitrogen removal methods (System 1 and System 2). In System 1, which consisted of a reactor for fresh waste, the leachate produced by the reactor was recirculated directly into the fresh waste. In contrast, System 2 was established where the fresh waste reactor and a degraded waste reactor were connected, and the process involved recirculating leachate between the two reactors. The primary objective of this study was to investigate the methods for enhancing the rate of CH_4 production and reducing the biostabilization time for MSW treatment. The experiment aimed to examine the impact of operational parameters on the biodegradation of MSW within a simulated anaerobic bioreactor landfill. There was limited exploration of the application and generalization of ANN in predicting cumulative biogas production and CH_4 content, based on the laboratory-measured parameters that influenced the process. This study aimed to address this gap by investigating the potential of ANN in predicting the degradation rate of MSW in the bioreactors. Generalizability of the ANN was assessed by evaluating its performance on completely unseen data from other studies, representing the pioneer application of such test. This study has been conducted in the Environmental Engineering Laboratory, Department of Civil Engineering, University of Birjand, Birjand, Iran in 2023.

MATERIALS AND METHODS

The MSW was sourced from the Saravan Landfill, a municipal landfill located in the northern region of Tehran, Iran. This landfill has been operational since 1984. The waste sorting process involved the removal of plastic bags and inorganic waste. The remaining waste was then pretreated and tattered by blade shredder to ensure optimal flow of leachate in the laboratory-scale landfills**.**

Experimental apparatus

The simulated landfill reactors consisted of square-based columns with internal dimensions of 400 square centimeters (cm^2) and vertical height of 130 centimeters (cm), yielding a volume of 40 liters (L). These columns were made up of steel and Plexiglas. To maintain the internal temperature, 10 cm-thick polystyrene panels were used in order to insulate the columns, which were then placed in a temperature-controlled room at 30±2 °C. To prevent leachate outlet clogging, a 15-cm gravel drainage layer was incorporated into the lower portion of the reactors. Approximately 24 kilograms (kg) of fresh waste was used to load Reactor A and Reactor B, while Reactor C was loaded with approximately 44 kg of degraded waste. Afterward, the 15-cm layer of fine gravel was applied to cover the waste, and a water distributor was installed on top of each reactor. Subsequently, approximately 2 L of deionized water was used to generate the desired amount of leachate.

Sampling and analytical methods

Two distinct experimental methods were employed. In System 1, the leachate from Reactor A was collected and recirculated into a tank every 24 hours (Fig. 1). In System 2, Reactor C was supplied with the leachate from Reactor B. The leachate produced in Reactor C was recirculated back to Reactor B on a 24-hour cycle. The process of leachate recirculation in both systems was facilitated using peristaltic pumps. The schematic diagram of both systems is presented in Fig. 1.

The composition of the MSW was analyzed in terms of its elemental composition including carbon (C), hydrogen (H), nitrogen (N), oxygen (O), and sulfur (S) using the PerkinElmer 2400 Series II CHNS/O Elemental Analyzer. Table 1 presents the physical

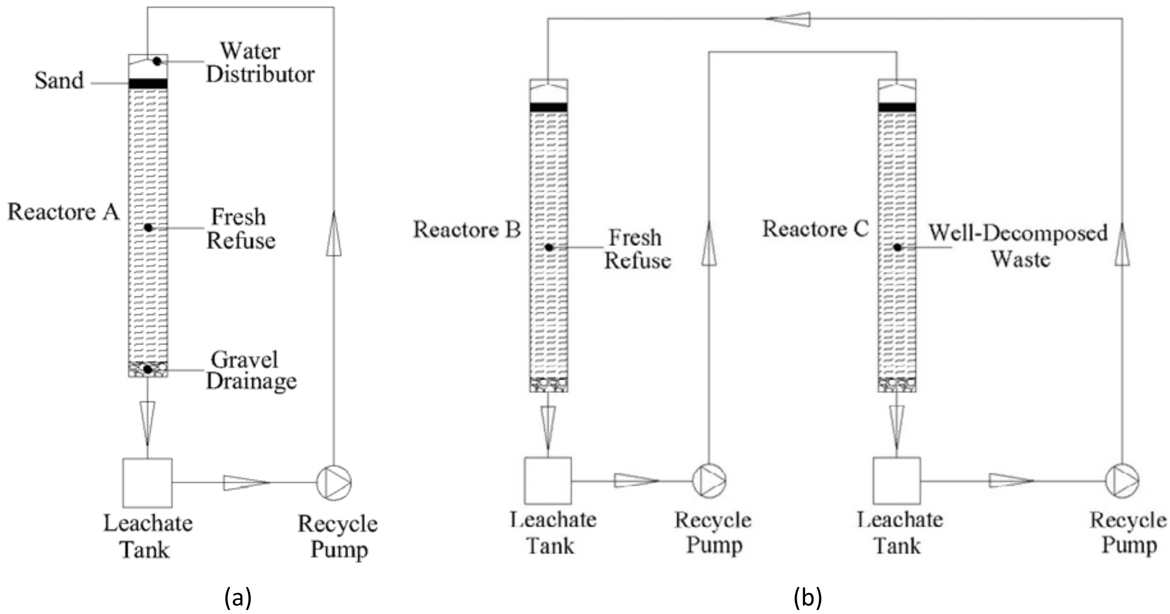


Fig. 1: Schematic diagram of (a) System 1, and (b) System 2

Table 1: Characteristics of fresh and degraded MSW (% of total weight)

Physical composition	Fresh	Degraded	Chemical characteristics	Fresh	Degraded
Food waste	65	0	Moisture content	28.32	46.23
Plastic	12	10.2	C	41.26	24.86
Paper	9	7.5	H	6.28	6.31
Textiles	2	1.2	O	39.72	42.54
Metal	0.5	0.2	N	2.57	2.15
Wood	2	1.5	S	0.51	0.26
Glass and others	9.5	9.7	Volatile solid	39.41	12.46
Soil	0	69.7			

and chemical characteristics of the MSW. Leachate samples were collected from the reactors every 6 days to measure the concentrations of biological oxygen demand (BOD_5), COD, ammonium-nitrogen (NH_4^+-N), and potential of hydrogen (pH) values. The pH value was measured using a HACH pH meter, while BOD_5, COD, and NH_4^+-N concentrations were determined according to the standard methods for the examination of water and wastewater (Rice *et al.,* 2012). A modified water displacement set-up was employed to measure the biogas from various runs. To quantify the total CH_4 produced, biogas was passed through water containing 2% volume per volume (v/v) sulfuric acid (H_2SO_4). The set-up involved connecting the biogas outlet of the reactor to a gas collection vessel filled with a H_2SO_4 solution. As the biogas released from the reactor, it bubbled through the solution. The H_2SO_4 solution was intended to absorb and react with specific components of the biogas, such as CO_2. The volume of CH_4, which did not not react with the solution, was determined by measuring the displacement of water in the gas collection vessel (Sponza and Ağdağ, 2004). To determine the CH_4 content, biogas samples were collected at 6-day intervals during the study. These samples were analyzed using a Young Lin gas chromatograph (model YL6100). The measurement setup was equipped with a PORO PACK Q column and a thermal conductivity detector (TCD).

ANN modelling

The CH_4 content and cumulative biogas production were modeled using ANN methodology. The pH, COD, hydraulic retention time (HRT), and NH_4^+-N were selected as input parameters for the ANN. These parameters were selected based on their profound influence on the microbial processes integral to methane production (Al-Dailami *et al.,* 2022). These parameters served as pivotal indicators, enabling the neural network to unravel both the qualitative and quantitative nuances of biological activity within the reactor. The ANN model was developed using a matrix laboratory (MATLAB) R2018b, a multi-paradigm numerical computing environment, with the support of the Neural Network Toolbox provided by MathWorks, Inc. The ANN architecture utilized included different layers such as input, hidden, and output layers. The neurons in the input layer indicated the independent variables and were connected to the neurons in the hidden layers by weighted connections. These weights determined the importance of the input data for each node, and a bias term was integrated to govern the size of the input data. The obtained values were multiplied by the Tan-Sigmoid activation function. The output layer determined the values of the output variables through the Pure-linear activation function. Tan-Sigmoid activation function is frequently utilized in ANNs to introduce non-linearity, enabling complex mappings within the hidden layer. In contrast, the output layer employs the Linear activation function to create a mapping that is linear in nature without any further non-linear transformation. This pairing of activation functions allowed for efficient modeling and prediction within the ANN architecture (Lee *et al.,* 2020). The values obtained from the ANN model were compared to the experimentally measured values. The error between the predicted and observed values was calculated and used to update the weights and bias of each neuron in the network. This approach enabled precise modeling and prediction of the intended outcomes. The proposed ANN model comprised two distinct ANNs. The first ANN (ANN1) was designed to predict the CH_4 content (%), while the second ANN (ANN2) was developed to estimate the cumulative CH_4 production liter per kilogram (L/kg) dry weight. The inputs to the model included analytical parameters such as pH, COD, NH_4^+-N, and HRT. To ensure objective evaluation, the experimental data were divided into three sets which had been randomly selected from different stages of the experimental study to prevent bias towards any particular stage. The first set comprised 70% of the data and was used for training the model and optimizing its parameters. The second set accounted for 15% of the data and was used for independent testing while serving as a benchmark for evaluating the model performance. The validation set constituted the remaining 15% of the data and was utilized to refine the hyperparameters of the model. In the ANNs training, the Levenberg Marquardt feed-forward back propagation perceptron (LMFFBP) algorithm was utilized, and the performance assessment was done using the mean squared error (MSE) metric. LMFFBP enables faster prediction and correction of limitations by manipulating the flow of input data within the ANN layers. This technique demonstrates excellent capability and robustness in addressing fitting problems (Mougari *et al.,* 2021). Control of the randomly-selected datasets, as well as determining the number of hidden layers, neurons, and activation functions are crucial in identifying the optimal architecture of ANN in terms of accuracy and simplicity. It is also essential to balance model complexity and data learning capacity. An ANN with overabundance of hidden layers and neurons may only store data without effectively learning from it. Hence, the most optimal structure for an ANN is the one that leads to accurate predictions with the least hidden layers and neurons in the hidden layer (Kerdan and Gálvez, 2020). In this study, the adopted ANN architecture included one hidden layer. The optimal number of neurons in the hidden layer was determined through an iterative process of trial and error. The performance of the ANN model was evaluated based on the statistical criteria including MSE and correlation coefficient. Once the optimal ANN was obtained, its effectiveness and generalizability was evaluated by testing it on previously unseen data from other studies. Such an evaluation was done for the first time to ensure the ability of the ANN to generalize beyond the specific dataset used for training, testing, and validation.

RESULTS AND DISCUSSION

Fig. 2 depicts the concentrations of COD in the leachate from Reactors A, B, and C. The experiment showed significant variations in the COD concentrations. At the beginning of the experiment, COD of the leachate from the fresh MSW in System 1 increased rapidly, reaching a maximum of 91,400

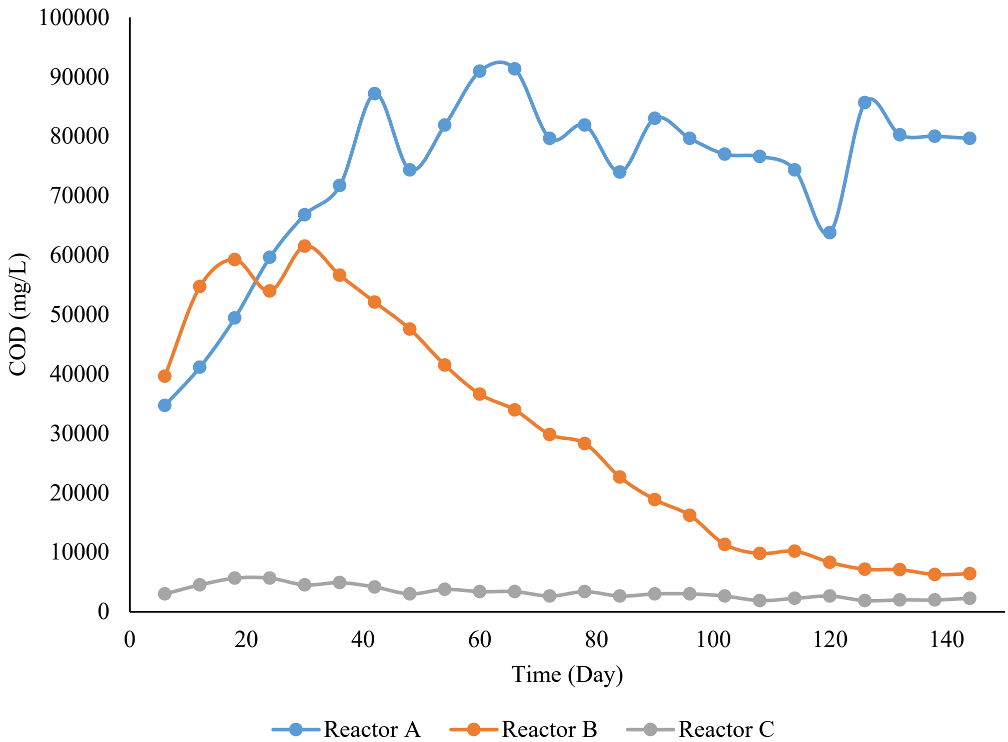


Fig. 2: Changes in the COD concentration of leachate over time in the bioreactors

mg/L on day 66. Notably, no significant decrease in COD concentrations was observed in the leachate produced by System 1. The rapid increase in COD concentrations in Reactor A could be explained by the presence of easily biodegradable organic matter in leachate from young landfills. This result was consistent with the findings of the study conducted by Ahn *et al.* (2002) and Marttinen *et al.* (2013). They observed higher COD concentrations in leachate from young landfill sites due to the higher proportion of biodegradable organic material. In reactor A, the absence of acetogenic bacteria in the fresh waste led to the occurrence of only two stages: hydrolysis and acidogenesis. Acidogenic bacteria (clostridium, bacteroides, and enterococcus), convert complex organic compounds into simpler compounds such as volatile fatty acids (VFAs) (Marttinen *et al.,* 2013). This breakdown of organic matter typically results in the release of COD, leading to an increase in COD concentration in the leachate. In System 2, the leachate COD concentrations in Reactor B increased initially for 30 days after recirculation. This was followed by a gradual decrease from 61,600 mg/L on day 30 to 6,270 mg/L on day 144. (Fig. 2), which could be attributed to the accumulation of carboxylic acid (Saadoun *et al.,* 2021). In contrast, Reactor C consistently maintained low effluent COD, indicating the successful removal of organic contaminants from the leachate by System 2.

Fig. 3 depicts the time-dependent degradation efficiency of COD and NH_4^+-N in System 2. During the entire operation, the degradation efficiency of COD fluctuated between 65% and 90%. In degraded MSW, the presence of acetogenic bacteria (acetobacterium, clostridium, and syntrophomonas) facilitated further metabolism of VFAs generated during acidogenesis. These acetogenic bacteria convert VFAs into acetic acid, hydrogen (H_2), and carbon dioxide (CO_2) through their metabolic activities (Saadoun *et al.,* 2021). Consequently, this conversion process contributed to a notable reduction in COD concentration. COD removal efficiency in System 2 decreased with the decrease of COD concentration in the leachate. The highest COD removal efficiency in Reactor B was

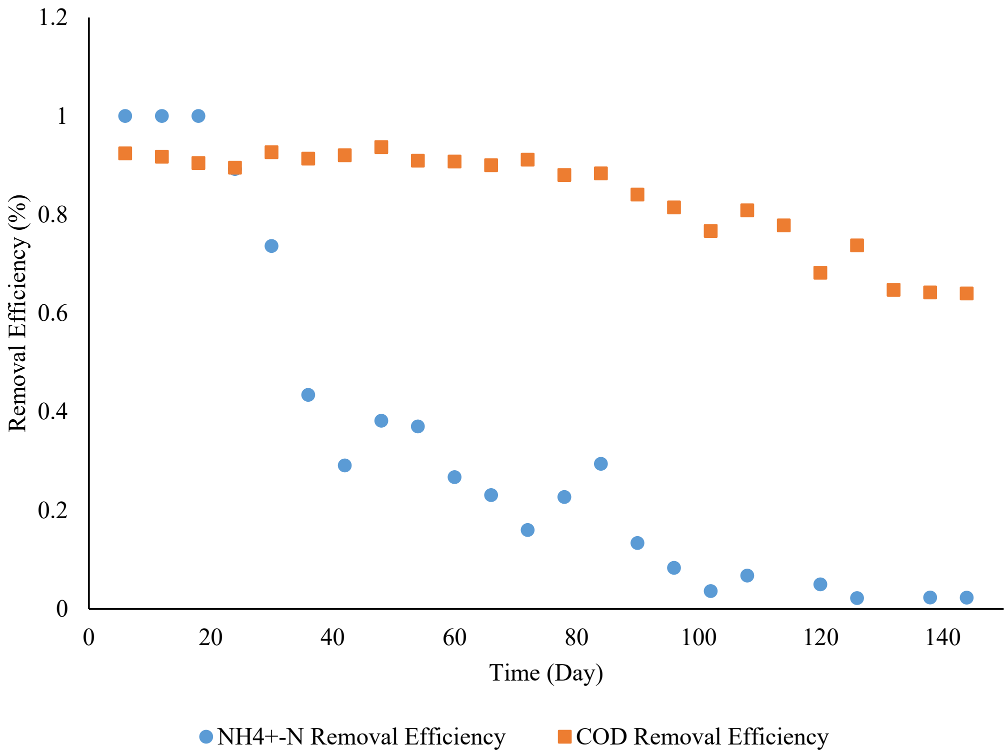


Fig. 3: COD and NH_4^+-N removal efficiencies in System 2

obtained as 90% (Fig. 3). The different biodegradability of organic matter was widely recognized in leachate. The BOD_5/COD ratio is frequently used to evaluate biodegradability; a higher value indicates a greater proportion of biodegradable organic material. On day 144, the BOD_5/COD ratio of the leachate from System 1 was 0.38, while the BOD_5/COD ratio of leachate from System 2 was 0.09. Due to the presence of large molecule compounds, such as humic acids, which are challenging to biodegrade, the residual organic matter in the leachate was nonbiodegradable (Keyikoglu *et al.,* 2021). Consequently, the maximum COD removal efficiency was capped at 90%.

NH_4^+-N

According to Fig. 4, the concentrations of ammonium in leachate in reactor A increased due to the accumulation of ammonium from the recirculated leachate (Feng *et al.,* 2019). Similarly, the concentrations of ammonium in the leachate in Reactors B and C experienced an initial increase within the first 42 days, which could be attributed to the breakdown of nitrogenous compounds specifically in Reactor B. As the two reactors (System 1 and System 2) operated under anaerobic conditions, the absence of nitrification microorganisms, which are effective in aerobic conditions (Peng *et al.,* 2022), caused ammonia-nitrogen to accumulate in them. The concentrations of NH_4^+-N in the leachate in Reactor A showed a significant increase, with the highest amount recorded on day 96 when the NH_4^+-N concentrations reached 3060 mg/L (Fig. 4). During hydrolysis, complex organic compounds are broken down into simpler forms, such as sugars, amino acids, fatty acids, and other organic nitrogen-containing compounds releasing ammonium as a byproduct (Price *et al.,* 2013).

However, under anaerobic conditions in System 2, the NH_4^+-N concentrations in Reactors B and C began to decrease on day 42 due to the degraded MSW adsorption capacity and the assimilation of NH_4^+-N by anaerobic microorganisms to support their growth (Feng *et al.,* 2019). On day 42, the maximum concentrations of ammonium in Reactors B and C were

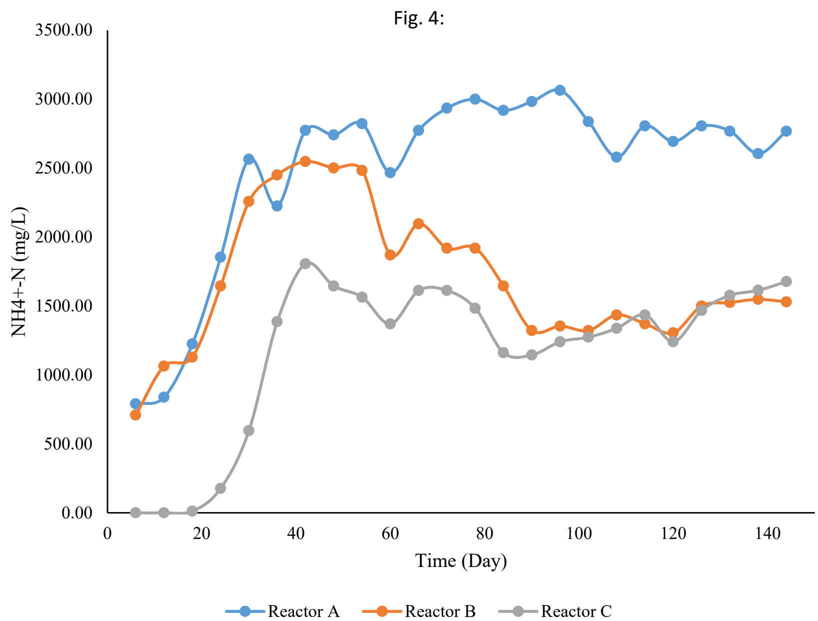


Fig. 4: Changes in the NH_4^+-N concentration of leachate over time in the bioreactors

obtained as 2650 mg/L and 1800 mg/L, respectively. After 42 days, the NH_4^+-N concentrations in leachates in Reactors B and C decreased, with Reactor B exhibiting a greater rate of NH_4^+-N decrease. On day 144, both reactors gave an identical NH_4^+-N concentration of 1600 mg/L. In System 2, NH_4^+-N was removed from the leachate by adsorption in Reactor C. In this system, the initial efficiency of ammonium removal was high and subsequently decreased gradually. Once the absorption capacity of NH_4^+-N in degraded waste reached saturation, the removal efficiency of System 2 decreased to approximately 0.5% on day 96 and remained around 0% in later stages. Obviously, the removal efficiencies of both COD and NH_4^+-N decreased over a certain period, with the NH_4^+-N removal efficiency declining more rapidly compared to the COD removal efficiency (Fig. 3). In biological treatment systems, microorganisms compete for available COD and NH_4^+-N as energy sources. If there is an excess of COD or other easily degradable organic matter, microorganisms may prioritize the utilization of COD over NH_4^+-N (Wang *et al.,* 2020).

pH

Variation in pH levels of anaerobic leachate in all reactors is illustrated in Fig. 5. In Reactor A, the pH of the leachate increased marginally throughout recirculation. Following recirculation, the pH in Reactor A rose from 5.9 on day 6 to 7.2 on day 144. These results indicated a tendency for pH to increase after leachate recirculation, which could be attributed to the stimulation of hydrolytic and fermentative bacteria. Hydrolytic and fermentative bacteria breakdown complex organic compounds into simpler forms through hydrolysis and fermentation as a part of recirculation process. The breakdown of organic compounds results in the release of different byproducts, such as VFAs and organic acids (Ratti *et al.,* 2013). The accumulation of VFAs and organic acids may decrease the pH of the leachate. The accumulation of VFAs and similar compounds in high-temperature reactors has been attributed to their anaerobic degradation in syntrophic reactions. Syntrophic bacteria play a vital role in anaerobic digestion by promoting the conversion of VFAs into CH_4. The bacteria and methanogens form a symbiotic relationship in the final step of CH_4 production (Li et *al.,* 2012). Syntrophic bacteria consume the VFAs produced by fermentative bacteria, thereby producing hydrogen and carbon dioxide as byproducts. Methanogens use these byproducts to produce CH_4. Additionally, sulfate-reducing and homoacetogenic bacteria consume VFAs and organic acids, resulting in the production of hydrogen sulfide

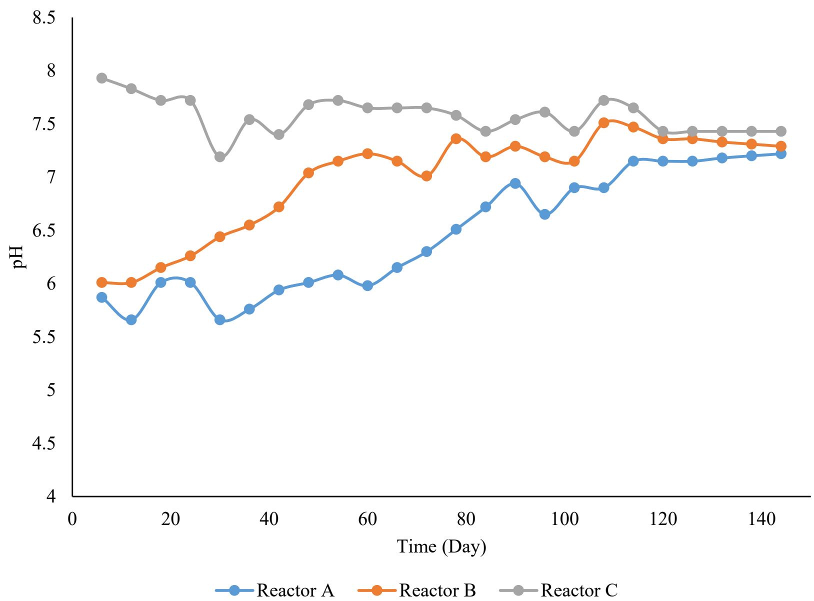


Fig. 5: Changes in the leachate pH over time in the bioreactors

and acetate, respectively (Singh *et al.,* 2021). Acetate, as a compound to other organisms, can be valuable, or it may be converted into CH_4 by methanogens in later steps of anaerobic digestion. This process leads to a decrease in the concentration of VFAs and organic acids and an increase in pH in the leachate. In Reactor B, the pH of the leachate increased from 6 to 7 during the first 48 days of recirculation and then continued to rise to over 7 from day 48 to day 144 (Fig. 5). The highest pH value in this reactor was recorded on day 108 reaching 7.5. Throughout the operation, the pH of leachate in Reactor C remained above 7 as a result of the presence of degraded waste in this reactor. The maximum and minimum pH values for leachate in Reactor C were 8 and 7.2, respectively. In Reactor B, fermentation and bacterial processes acting on biodegradable compounds produced and accumulated acids (Ratti *et al.,* 2013). Methanogenic bacteria in Reactor C converted the acids accumulated in Reactor B into CH_4 and carbon dioxide. As a result, the leachate pH in the Reactor B was found to be lower compared to the pH of the effluent leachate from Reactor C. Therefore, the pH value of the effluent leachate from Reactor C decreased over time as leachate was recirculated from Reactor B to Reactor C. However, the pH variation, occurrin gafter 114 days, was minimal during the stable phase. The stable phase of leachate recirculation is characterized by pH stabilization, which results from the development of a balanced microbial community and stable metabolic processes (Talalaj, 2015). In Reactor B, pH balance resulted from the steady-state consumption of acids by the methanogenic bacteria in Reactor C. The decomposition of solid waste undergoes three distinct phases within the lab-scale landfills (Reactors A and B). Initially, complex organic matter undergoes hydrolysis, resulting in the formation of soluble molecules. During the next stage, these molecules are further transformed into carbon dioxide, hydrogen, simple organic compounds, and VFAs. The third stage involves the production of CH_4 through the decomposition of acids into CH_4 and CO_2, or the reduction of CO_2 with H_2. In this study, Reactor C balanced the growth of the acid-production and CH_4-production phases, accelerating the decomposition of organic matter in System 2. Based on Fig. 5, the recirculation of leachate from degraded MSW to fresh MSW resulted in a notable increase in pH values in Reactor B compared to Reactor A. Consequently, a faster degradation of MSW in System 2 was expected. These findings were consistent with the results of other studies (Luo and Wong, 2019). The characteristics of the leachate effluent from Reactors A and B before and after completion of treatment are

Table 2: Characteristics of the leachate from Reactors A and B

Parameter	Initial leachate	Treated leachate	
	Reactor A and B	Reactor A	Reactor B
COD (mg/L)	31525	79620	6410
NH_4^+-N (mg/L)	772	2768	1530
pH	6	7.22	7.29
BOD (mg/L)	16850	30255	580

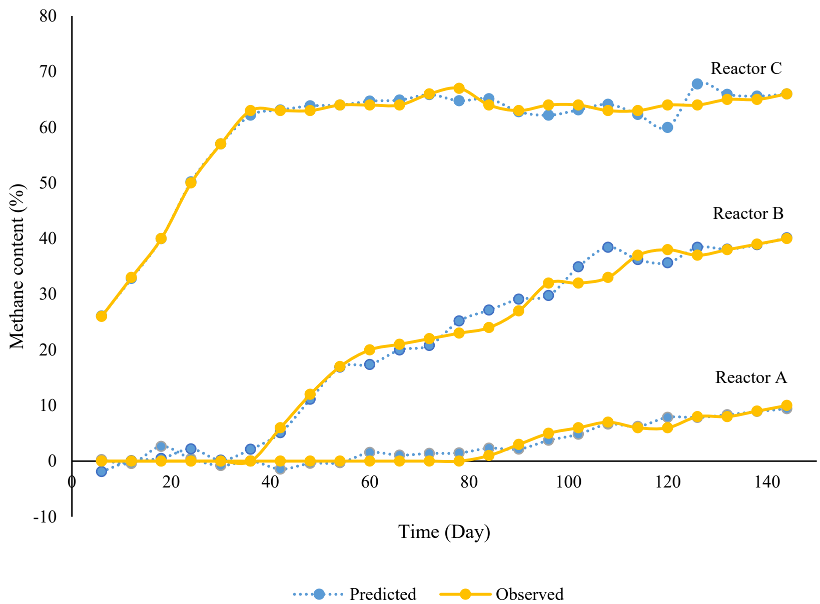


Fig. 6: Methane content observed and predicted over time in the bioreactors

presented in Table 2.

CH_4 content and cumulative CH_4 production

Figs. 6 and 7 illustrate the results for CH_4 content in the biogas and cumulative CH_4 production from Reactors A, B, and C. Fig. 6 shows that the type of recirculation method and the age of waste are involved in the CH_4 gas concentration variations. The CH_4 content in Reactor A was below 10% during the whole experiment. The CH_4 content in biogas from System 2 indicated the stability and performance of anaerobic digestion. The CH_4 content in reactor B showed a rapid increase after 36 days of digestion and reached 48% on day 144. The CH_4 concentration in the biogas from reactor C showed a significant increase from 26% to 63% between day 6 and day 36 and then remained stable (Fig. 6). Cumulative CH_4 production in fresh waste in reactor B reached 26.8 L/kg dry weight, which was 4.31 times higher than those in Reactor A (6.21 L/kg dry weight) (Fig. 7). These findings were consistent with the results reported in previous studies (Behera *et al.,* 2015; Gao *et al.,* 2023). According to Figs. 2, 5, and 6, on day 36, reactor B entered the methanogenesis phase, indicated by the increase of CH_4 content and leachate pH and decrease of leachate COD concentrations. Reactor B reached this stage 48 days earlier as compared to Reactor A. The performances of reactors A and B indicated that the addition of leachate from degraded waste decreased the startup time and enhanced the biogas production from fresh waste. Recirculation of the leachate in fresh waste triggers accelerated hydrolysis and acidification, leading to the accumulation and inhibition of intermediates (Gao *et al.,* 2023). When the leachate is recirculated back into fresh waste, it introduces additional moisture and soluble compounds, providing optimal conditions for microbial activity. It also enhances the

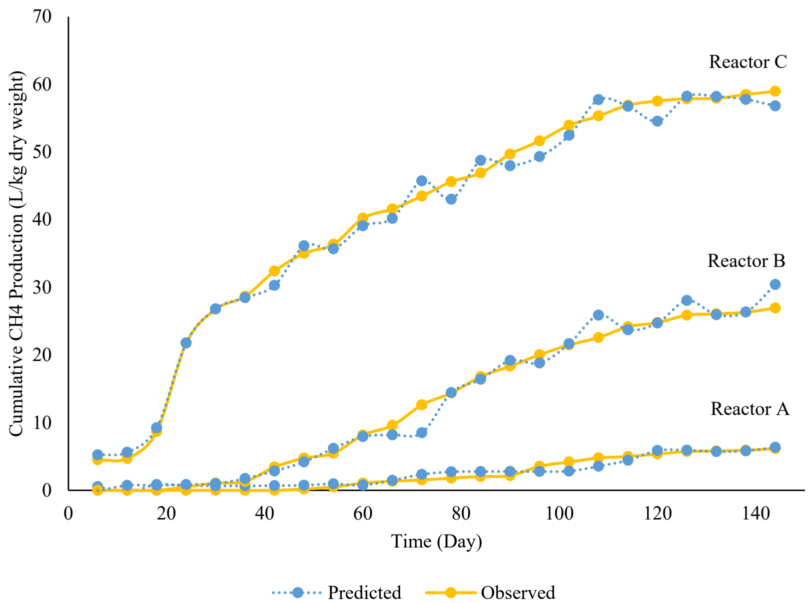


Fig. 7: Cumulative methane production observed and predicted over time in the bioreactors

availability of water and nutrients, promoting the growth and activity of hydrolytic microorganisms.

The stabilization phase in reactor C, as indicated by stable concentration of COD (Fig. 2), was attained prior to the commencement of leachate recirculation. Subsequently, a considerable increase in CH_4 production following the initiation of recirculation indicated the ability of methanogens to effectively utilize organic acids from the ftresh waste leachate, even when the waste decomposition had already reached the stabilization phase. Confirmation of this ability of methanogens has been established in previous studies (Yang *et al.,* 2021). Methanogens utilized these organic acids as a carbon source for their metabolism, converting them into CH_4 gas. Consequently, the consumption of organic acids by methanogens reduced the concentration of organic acids in reactor B and increased the carbon source in reactor C, enhancing CH_4 production in both reactors. As a result of this process, reactor C showed a notable increase in cumulative CH_4 production, with the volume significantly rising from day 24 onwards. By day 144, the cumulative CH_4 production reached 59 L/kg dry weight, indicating a substantial enhancement in CH_4 production within the system. The maximum daily CH_4 production in Reactor C occurred from day 18 to day 24 of the experiment, reaching 2.09 L/day/kg dry weight (Fig. 7). These findings were consistent with the results reported in previous studies. In a study conducted by Sandip *et al.* (2012), the cumulative CH_4 production on day 270 of the experiment was 67 L/kg dry weight, with a maximum daily CH_4 production of 1.68 L/day/kg dry weight. Another study reported a cumulative CH_4 production of 50 L/kg dry weight on day 250 for degraded MSW (Sanphoti *et al.,* 2006). In a study by Ahmadifar *et al.* (2016), the cumulative CH_4 production for degraded MSW was recorded as 54.87 L/kg dry weight on day 180, with a maximum daily CH_4 production of 1.35 L/day/kg dry on day 112. Methanogenic bacteria in a bioreactor containing degraded waste need time to adjust to a new organic load when fresh waste is introduced. Consequently, the initiation of methane production in such reactors can be protracted (Ahmadifar *et al.,* 2016). Starting from day 36 (Fig. 2), COD concentration in reactor B exhibited a simultaneous decrease with the increase of CH_4 production in reactors B and C. This decline could be attributed to the carbon consumption by the methanogenic bacteria existing in reactor C. It has been established that an NH_4^+-N concentration of over 2500 mg/L is toxic to methanogenesis, regardless of temperature and pH levels (Feng *et al.,* 2019; Liu and Sung, 2002). In this study, a comparison was made between NH_4^+-N concentration (Fig. 4) and

CH_4 yields, revealing that NH_4^+-N played a crucial role in determining the initiation time of CH_4 production. Based on the analysis on day 36 (Figs. 4 and 6), a significant increase in the CH_4 content was observed in reactor B. This increase occurred simultaneously with a decrease in NH_4^+-N concentration.

ANN modelling

To perform comprehensive analysis, the modeling samples were randomly selected from different stages of the experiment and included in training, validation, and testing sets. The data used for testing the ANN are presented in Table 3.

The optimal network architecture was determined based on statistical criteria, MSE, and correlation coefficient (R). According to Table 4 in ANN1 and ANN2, as the number of neurons exceeded 4 and 6 respectively, MSE of the training data decreased, but MSE of the test data increased. Increase of the number of neurons in ANN improved the fitting of the training data, potentially decreasing the MSE of the training data. However, it was likely that very high number of neurons could cause the ANN to overfit the training data, leading to an elevated MSE for the test data (McElroy *et al.,* 2021). This increase in MSE for the test data indicated that the ANN was not generalizing effectively and could not preserve the patterns and relationships in the data.

For accurate prediction of CH_4 content, the optimal configuration for ANN1 included one hidden layer with 4 neurons. The best setup for ANN2 to precisely forecast the cumulative CH_4 production entailed one hidden layer containing 6 neurons. Both ANNs utilized the Tan-Sigmoid activation function for the hidden layer and the Pure-linear activation function for the output layer. Utilization of the datasets obtained from MATLAB neural network learning for this analysis, and the results of ANN1 and ANN2 training performances are shown in Fig. 8a and 8b, respectively. By representing the experimental data (target value) and the predicted results (output) in plot form, the regression results demonstrated the relationship between them. Fig. 9a and 9b show the regression results of the significant correlation coefficient for CH_4 content and cumulative CH_4 production in the training, validation, and testing data.

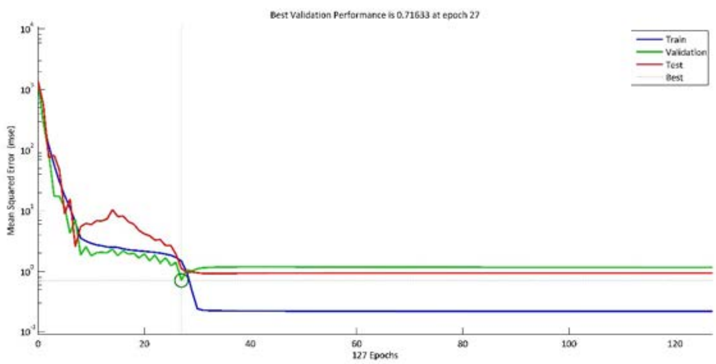
The ANN1 and ANN2 models exhibited exceptional performances during the validation, leading to the best validation performances of 0.716 and 0.634, respectively (Fig. 8). This indicated the models' ability to accurately predict the CH_4 content and cumulative CH_4 production based on the input parameters. The training determination coefficients of 0.9990 for ANN1 and 0.9995 for ANN2 further confirmed the successful learning of the relationship between input and output variables by the ANNs. In addition, the validation R-values of 0.9997 for ANN1 and 0.9981 for ANN2 indicated the strong generalization ability of the models. The overall performance of the dataset showed high R-values of 0.9991 and 0.9975 for ANN1 and ANN2 respectively, suggesting a thorough comprehension of the complex biological process in the system (Fig. 9). The strong correlation in among the test data indicated the model's capacity to understand the complex modeling process. Modeling biological systems is inherently more challenging than physical or chemical processes due to the involvement of living microorganisms and their complex characteristics and responses to changing conditions (Nair *et al.,* 2016). The high R-value obtained for the test data indicated an optimized selection of effective parameters in the input data. ANNs are mathematical models that learn patterns and relationships from data. However, they do not possess an inherent

Table 3: Input data for testing the ANN model

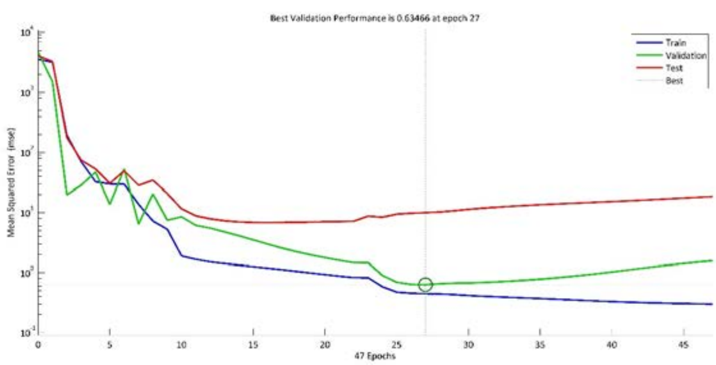
Reactor	HRT (Day)	COD(mg/L)	NH_4^+-N (mg/L)	pH
A	6	34710	790	5.87
	60	91320	2467	5.98
	132	79620	2580	7.22
B	12	54710	1064	6.01
	84	22640	1645	7.19
	114	10180	1371	7.47
	126	7160	1500	7.36
C	6	3010	0	7.93
	72	2640	1612	7.65
	126	1880	1467	7.43

Table 4: MSE values for ANNs with different numbers of neurons in the hidden layer

Number of neurons	CH_4 content		Cumulative CH_4 production	
	Train	Test	Train	Test
3	2.13	9.86	76.24	95.62
4	0.74	0.72	15.75	38.67
5	0.71	3.59	2.61	11.65
6	0.68	9.17	0.52	0.63
7	0.53	23.63	0.48	6.85
8	0.31	22.25	0.41	19.42



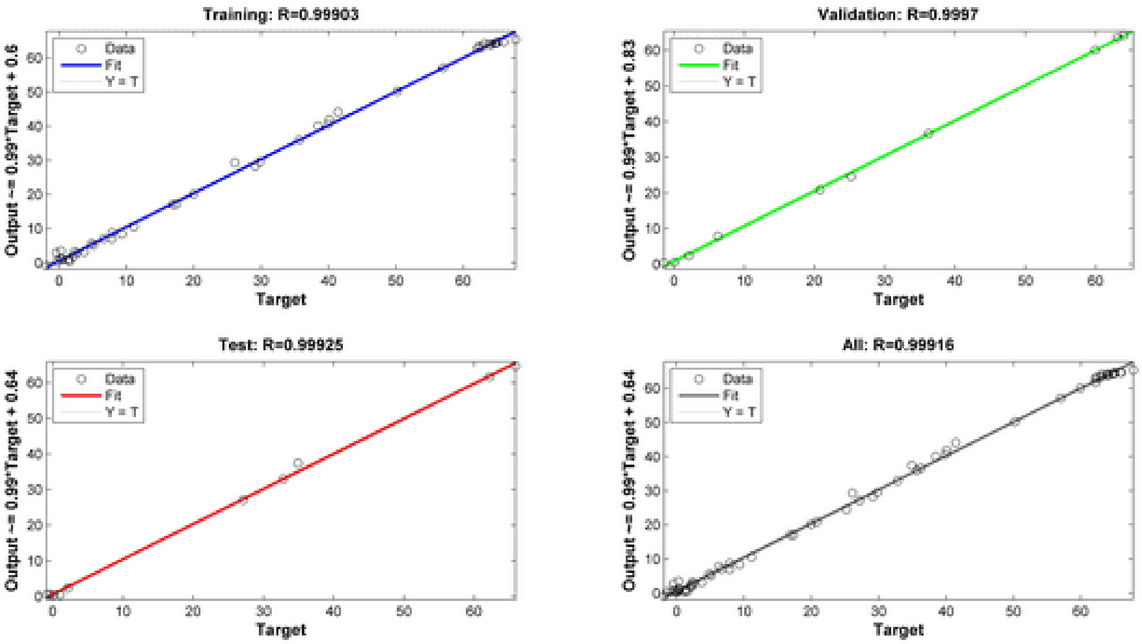
(a)



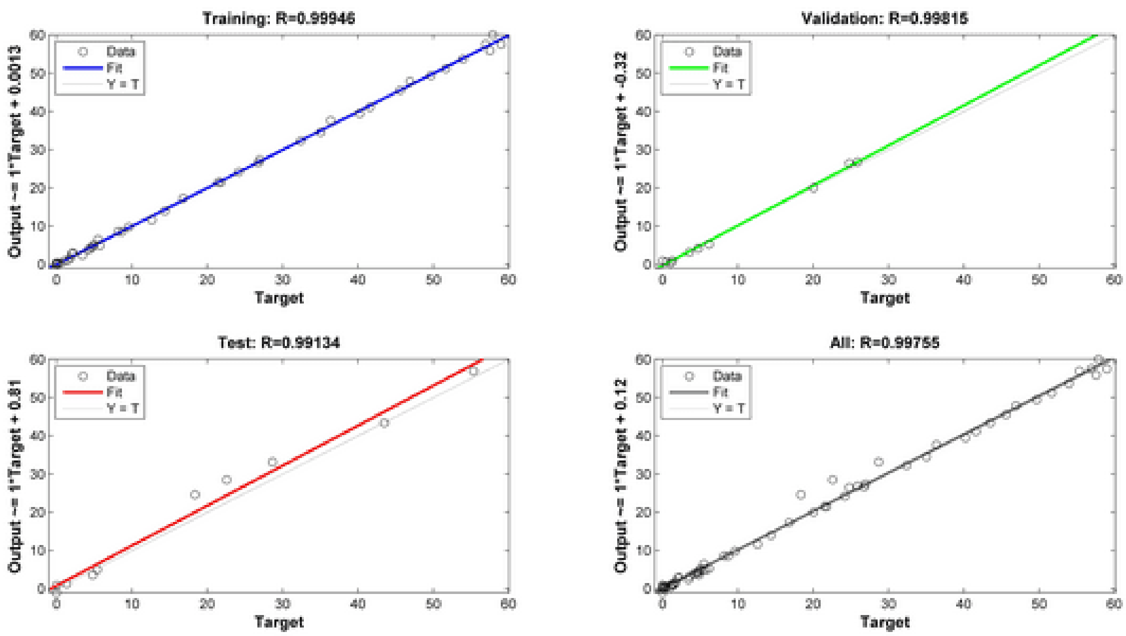
(b)

Fig. 8: MSE variation with Epochs during the training process of (a) ANN1, and (b) ANN2

understanding of the modeled process. For effective learning and predicting, ANNs depend on accurately selected and represented inputs (Hatata *et al.,* 2021). In a study, pH, moisture content, total volatile solids, VFAs, and HRT values were included as the input data for ANN (Nair *et al.,* 2016). In the current study, the analysis of test data revealed a notably higher R-value surpassing the R-value reported by Nair *et al.* (2016). This finding provided a compelling evidence for the advanced proficiency of the developed ANN in understanding the complexities of the biological processes. As discussed earlier, biological factors that affect biogas production could significantly impact the concentration of COD and NH_4^+-N. In the present study, these parameters along with pH and HRT were included in the input data. This approach effectively improved the comprehension of the biological process by the ANN. The performance of ANN in predicting CH_4 production was compared with other methods (Table 5). The ANN demonstrated a better performance compared to other methods for predicting CH_4 production (Table 5). However, it is



(a)



(b)

Fig. 9: Comparison of the observed and predicted (a) CH_4 content, and (b) cumulative CH_4 production for training, validation, and testing sets

Table 5: Comparison of the CH_4 production prediction models

Feedstock	Reactor type	Input parameters	Method	R-value	MSE	References
MSW	Landfill	Total waste landfilled, organic content, temperature, precipitation, landfill age, depth, and landfill cover	Fuzzy	0.951	71.31	Mohsen and Abbassi, 2020
			LandGEM	0.804	96.75	
MSW	Anaerobic bioreactor landfill	Amount of leachate, temperature, methane content, pH, COD	Neuro-fuzzy	0.71	3.62	Mehrdad *et al.*, 2021
			Support vector machine	0.90	3.37	
			ANN	0.98	3.21	
Sewage sludge	Anaerobic digester reactor	Sludge inflow, temperature, pH, total solid, volatile solid, organic acid, alkalinity, HRT, and organic loading rate (OLR)	Multiple linear regression	0.722	11.03	Bao *et al.*, 2023
			ANN	0.794	10.23	
Molasses wastewater	Pilot-scale upflow anaerobic sludge blanket	OLR, COD removal rate, influent and effluent alkalinity, and pH	Fuzzy-logic	0.8721	8.15	Turkdogan-Aydınol and Yetilmezsoy, 2010
			ANN	0.9847	4.86	
Palm oil mill effluent	Anaerobic digester reactor	Recirculation ratio, pH and temperature	Response surface methodology	0.9512	0.51	Chong *et al.*, 2023
			ANN	0.977	0.23	
			ANFIS	0.976	0.23	
High polluted wastewater	Anaerobic reactor	Reactor fill ratio, OLR, influent and effluent pH, alkalinity, COD, suspended solids	Nonlinear regression	0.9852	18.22	Tufaner and Demirci, 2020
			ANN	0.9878	14.74	
MSW	Anaerobic bioreactor landfill	COD, HRT, NH_4^+-N, pH	ANN	0.998	0.634	The current study

worth noting that there was no significant difference between the ANN and the adaptive neuro-fuzzy inference system (ANFIS) in terms of performance. The results presented in Table 5 also demonstrated that the achieved prediction quality was superior to the prediction quality reported in recent studies focusing on biogas production modeling.

The impact of removing specific input variables on the performance of ANN was investigated for their combined effect on cumulative CH_4 production. Eliminating the pH parameter resulted in a lower R-value (0.8801) and a higher MSE (21.79) compared to ANN2 in the test and validation data, respectively. Exclusion of the COD concentration led to a lower R-value (0.751) and higher MSE (44.52). Similarly, removing the HRT parameter resulted in a R-value of 0.7731 and a MSE of 29.73. Moreover, exclusion of the NH_4^+-N concentration yielded a R-value of 0.835 and a MSE of 23.39. The results suggested that the combined influence of the selected parameters could affect cumulative CH_4 production. According to the results obtained from MSE and R-values in the test and validation data, it could be inferred that the COD concentration had the highest influence on cumulative CH_4 production. Conversely, the pH level was found to have the lowest impact on cumulative CH_4 production among the studied parameters. The weights of the ANN were employed to estimate the relative importance of the operational parameters on cumulative CH_4 production. The results showed that COD and HRT had a stronger influence, with relative importance values of 39.03 and 28.34, respectively. NH_4^+-N and pH had relative importance values of 18.54 and 14.09, respectively. After successful development of a reliable and high-performing ANN model, the trained ANN was employed to optimize the process conditions with the aim of maximizing cumulative CH_4 production. Utilizing the trained ANN as a fitness function, a genetic algorithm (GA) was implemented for this purpose. For each GA iteration, the ANN model assessed methane production, yielding a fitness value. The GA was executed for 200 iterations with a population size of 25 individuals. During each generation, the individuals producing the highest methane estimates, as determined by the ANN, underwent reproduction, mutation, and crossover to form the subsequent generation. The optimized values for HRT, COD, NH_4^+-N, and pH were determined as 81 days, 12680 mg/L, 1712 mg/L, and 7.28, respectively. The maximum cumulative CH_4 production under these optimum parameters was found to be 61.94 L/kg dry weight.

Assessing the generalizability of the ANN

If all the influential parameters were selected and the ANN was properly trained, the ANN would have a potential to accurately predict outputs under different conditions. In this study, the input data for the ANN consisted of COD and NH_4^+-N concentration, pH value, and HRT. These parameters were selected based on their known influence on the biogas production process. By incorporating these input data, the ANN was able to effectively capture and accommodate the influence of variations in biological processes on the output. The hypothesis was that any variation in the biological characteristics of the waste, the waste quality parameters (such as carbon, nitrogen, moisture, sulfur, etc.), laboratory conditions (temperature, volume, number of reactors, etc.), and recirculation rate might not have a substantial impact on accuracy of the ANN in forecasting the output. Since these variations affected the input data, the ANN accounted for their effects on the output. In this section, the data from four additional studies were utilized as inputs to the ANN. These studies were selected to provide a diverse range of conditions and inputs in order to ensure the robustness of the ANN model. In the first study, three types of bioreactors containing MSW had been employed. One of these bioreactors operated without leachate recirculation, while the others (remaining two bioreactors) had leachate recirculation systems. The leachate recirculation rate in one of the reactors was 9 liters per day (L/day), while the other reactor had a recirculation rate of 21 L/day (Sponza and Ağdağ, 2004). The simulation results obtained using ANN1 demonstrated significant effectiveness in predicting the performance of the bioreactors. By incorporating the data from the study conducted by Sponza and Ağdağ (2004) into the input data of ANN1, the R-value and MSE for the experimental data and the predicted data were determined to be 0.88 and 5.35, respectively. In the second study, the biological degradation of MSW had been investigated in both anaerobic and hybrid bioreactors (Xu *et al.,* 2015). To assess the generalization ability of ANN1, only the anaerobic bioreactor data were employed. By incorporating the data from the study conducted by Xu *et al.* (2015) into the ANN1, the values of MSE and R-value for the observed and predicted data were determined as 9.06 and 0.83, respectively. Despite the reduction of the R-value in these two studies, it should be considered that any R-value exceeding 0.8 was widely accepted as suitable for modeling the biological processes (Bao *et al.,* 2023; Turkdogan-Aydınol and Yetilmezsoy, 2010). By comparing these results with those presented in Table 5 and taking into account that the data utilized at this stage were completely unseen by the ANN, the performance of the ANN could be deemed appropriate. Additionally, it was important to consider other statistical indices, such as MSE, in conjunction with the R-value (Rahman *et al.,* 2022). Considering the predicted data range, with the highest value of 70 and the lowest value of 0, a MSE of 9.06 represented an acceptable level of accuracy. In the third and fourth studies, the MSW had undergone mechanical treatment followed by biological treatment (Di Addario and Ruggeri, 2018; Sormunen *et al.,* 2008). The primary objective of the mechanical treatment of MSW was to separate different components such as organic materials for recycling and metals for reuse, and prepare the remaining waste for subsequent treatment. This process involved sieving and sorting methods, resulting in smaller particle sizes in the residual waste. It should be noted that when the smaller particles are landfilled, the initial degradation phase may be impacted and the leaching of organic materials and nitrogen from waste may be increased (Di Addario and Ruggeri, 2018). The mechanical properties of waste, including porosity, hydraulic conductivity, particle sizes, and surface areas, play a crucial role in anaerobic decomposition. The hydraulic conductivity of waste can directly affect the availability of water for microbial activity, which is essential for the growth and metabolism of methanogenic bacteria. Additionally, smaller particle sizes and larger surface areas provide more contact sites for microbial colonization and enhance the breakdown of organic compounds, leading to an increased methane production (Johnravindar *et al.,* 2021). Reducing the porosity of MSW can improve mass transfer and increase the rate of CH_4 production (Ko *et al.,* 2015). It is important to note that the ANN trained in this study did not consider the input parameters related to the mechanical characteristics of the waste, such as porosity, particle size, and hydraulic conductivity. Consequently, ANN1 did not consider the potential impact of these variables on the prediction of cumulative CH_4 production from the waste. Therefore, the performance of ANN1 in accurately predicting the CH_4 content was significantly reduced for the data from the latter two studies. In the study conducted by Di Addario and Ruggeri (2018), the R-value and MSE of ANN1 were determined as 0.28 and 186, respectively. Similarly, in the study done by Sormunen *et al.* (2008) the R-value and MSE of ANN1 were found to be 0.39 and 137, respectively.

CONCLUSION

This study demonstrated that the recirculation of leachate between fresh and degraded waste led to a substantial improvement in COD reduction in System 2. Acetogenic bacteria in Reactor C contributed to a notable reduction in COD concentration. The COD removal efficiency in System 2 fluctuated between 65% and 90%. NH_4^+-N concentrations in Reactors B and C initially increased to maximum levels of 2650 mg/L and 1800 mg/L, respectively. However, after day 42, the concentrations started to decrease. By day 144, both reactors reached the same NH_4^+-N concentration of 1600 mg/L. The decrease in NH_4^+-N was mainly due to the adsorption of degraded waste and assimilation by anaerobic bacteria. Throughout the experiment, reactor A had a CH_4 content of below 10%, while reactors B and C showed significant increases. Reactor B reached 48% CH_4 content on day 144, with its cumulative CH_4 production being 4.31 times higher compared to reactor A. Leachate recirculation in Reactor B accelerated the startup time, enhancing biogas production. Reactor C exhibited a notable CH_4 production reaching 59 L/kg dry weight by day 144. The performance of the ANN models during the validation was exceptional, with validation performances of 0.716 and 0.634 achieved for ANN1 and ANN2, respectively. The high R value obtained for the test data demonstrated the model's capacity to understand the complex modeling process, despite the inherent challenges in biological systems modeling. The accuracy of ANN1 prediction was evaluated under different experimental conditions. The data from two previous studies supported its effectiveness and significant accuracy in predicting the bioreactor performance. However, ANN1 did not account for variations in mechanical characteristics of waste, impacting its ability to accurately predict cumulative CH_4 production. Specifically, the ANN performance in predicting the CH_4 content for the two studies, which involved mechanical treatment and smaller particle sizes in the waste, showed a significant decline.

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CONFLICT OF INTEREST

The author declares that there is no conflict of interests regarding the publication of this manuscript. In addition, the ethical issues, including plagiarism, informed consent, misconduct, data fabrication and/or falsification, double publication and/or submission, and redundancy have been completely observed by the authors.

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ABBREVIATIONS

%	Percent
ANFIS	Adaptive Neuro-Fuzzy Inference System
ANN	Artificial Neural Network
ANN1	First type of artificial neural network
ANN2	Second type of artificial neural network
BOD_5	Biochemical Oxygen Demand
C	Carbon
CH_4	Methane
cm	Centimeter
cm^2	Square centimeter
CO_2	Carbon Dioxide
COD	Chemical Oxygen Demand
GA	Genetic Algorithm
H	Hydrogen
H_2SO_4	Sulfuric Acid
HRT	Hydraulic Retention Time
kg	Kilogram
L	Liter
L/day	Liter per Day
L/kg	Liter per Kilogram
LMFFBP	Levenberg Marquardt Feed-Forward Back Propagation Perceptron
m	Meter
m^2	Meter square
MATLAB	Matrix Laboratory
mg/L	Milligram per Liter
MSE	Mean Squared Error
MSW	Municipal Solid Waste
N	Nitrogen
NH_4^+*-N*	Ammonium-Nitrogen
O	Oxygen
OLR	Organic Loading Rate
S	Sulfur
TCD	Thermal Conductivity Detector
v/v	Volume per Volume
VFA	Volatile Fatty Acid

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