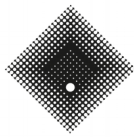# VICTORIA UNIVERSITY

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ABSTRACT

Recent research studies on the innovative concept of submerged anaerobic membrane bioreactor (SAnMBR) technology have demonstrated superior treatment and operational performance for treating a broad range of waste streams discharged from various industries. This study aimed to investigate the treatment and recovery of biomethane (bio-CH_4) performance of ceramic ultrafiltration (UF) coupled with "*co-digestion based SAnMBR*", which was not previously studied by others, for treating an organic fraction of food waste (OFFW) blended with domestic wastewater (DWW) at surge organic loading rates (OLRs) disposed at modern high-rise establishments and similar residential clusters. The SAnMBR was operated in five phases (Phase 1–5), with different organic loading rates (OLRs) varying from 0.49 to 22.57 kg-COD/m^3/d. All bio-CH_4, mixed liquor sludge, and treated permeate samples were analyzed using standard methods. The key parameters representing the cumulative bio-CH_4 yield during each phase were estimated using sigmoidal models, and the simulated results were validated using ANOVA. It was found that the SAnMBR produced high-quality, low-turbid reclaimed water showing an increasing trend in yield of bio-CH_4 with an increase of OLR. It was also observed that the SAnMBR demonstrated stable and superior treatment performance at shock-loads of organics. The maximum bio-CH_4 yield recorded during the study was $73.06 \pm 6.48\%$. The findings of this study confirmed the suitability of applying this novel concept of "*co-digestion-based SAnMBR*" towards sustainable and efficient waste management in modern-high rise establishments.

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List of abbreviations

Abbreviation	Description
SAnMBR	Submerged Anaerobic Membrane Bioreactor
bio-CH$_4$	Biomethane
UF	Ultrafiltration
OFFW	Organic Fraction Of Food Waste
DWW	Domestic Wastewater
OLR	Organic Loading Rate
ANOVA	Analysis Of Variance
COD	Chemical Oxygen Demand
FW	Food Waste
AD	Anaerobic Digestion
AcoD	Anaerobic Co-Digestion
SRT	Solid Retention Time
HRT	Hydraulic Retention Time
TOC	Total Organic Carbon
MLSS	Mixed Liquor Suspended Solids
MLVSS	Mixed Liquor Volatile Suspended Solids
VFA	Volatile Fatty Acids
PVC	Polyvinyl Chloride
TMP	Transmembrane Pressure
SSW	Semi-solid Waste
WW	Wastewater
CI	Confidence Interval
ODE	Ordinary Differential Equation
RPM	Revolution Per Minute
SBR	Sequential Batch Reactor

Nomenclature

Symbol	Description (**Unit/value**)
V	Hydraulic volume of the bioreactor (L)
t	Time (D)
Q_i	Influent flow rate (L/d)
S_i	Influent COD (g/L)
S_e	Effluent COD (g/L)
L	Organic loading rate (g/m^3/d)
x	MLSS concentration in the bioreactor (g/L)
Y	MLSS yield (g-MLSS/g-COD)
k_{dx}	Death (endogenous decay) rate of MLSS (/d)
μ_s	Specific growth rate of MLSS (/d)
ΔP	Transmembrane pressure (Pa)
P	Bio-CH$_4$ production potential (L-CH$_4$/g-COD)
R	Maximum bio-CH$_4$ production rate (/d)
L	Lag time (D)

1. Introduction

The quality of life assured by metropolitanized modern high-rise establishments has attracted people's attention worldwide. Nonetheless, these residential establishments lack strategies to mitigate the cost of high-energy consumption, trade charges related to wastewater discharge and waste management challenges. Food waste (FW) has been identified as the primary constituent of domestic solid waste in many urban areas (Pandey and Mukherjee, 2022), including modern high-rise establishments. FW generated at these modern high-rise clusters constitutes the bulk share of total FW in developed and developing countries (Reutter et al., 2017). Due to the presence of complex organic and particulate matter in the organic fraction of food waste (OFFW), improper management practices can cause severe environmental constraints

(Satayavibul and Ratanatamskul, 2021; Shooshtarian et al., 2020), such as contamination of surface and groundwater, production of leachate, and emission of greenhouse gases. Also, inefficient FW management can cause significant economic expense worldwide, affecting national FW management plans (Ananda et al., 2021).

Further, the treatment and discharge of domestic wastewater (DWW) generated at these residential establishments are becoming more expensive and challenging due to stringent laws and strict environmental regulations (Anjum et al., 2021). Due to the unavailability of efficient pretreatment systems, these residential establishments have to incur high costs and penalties to achieve the required treated effluent quality standards specified by the water authorities before discharge. Conventional wastewater treatment methods applied at municipal and highly polluted industrial point sources containing high-organics, toxic and complex particulate matter, such as DWW (Abeysiriwardana-Arachchige et al., 2020; Gautam et al., 2017), food processing industries (Alexander et al., 2020), and abattoirs (Alfonso-Muniozguren et al., 2018), are found inefficient. Additionally, conventional wastewater treatment facilities incur considerable energy and space footprints. Therefore, there is a sense of urgency in seeking an alternative, efficient, and sustainable wastewater treatment system that demonstrates superior performance.

Past studies show that blended DWW and OFFW have a high energy recovery potential if treated using anaerobic digestion (AD) (Cheng et al., 2021; Pramanik et al., 2019; Vinardell et al., 2021; Zamorano-López et al., 2018). Anaerobic digestion (AD) is a conventional biological process that occurs in strict anoxic conditions and converts organic matter into biogas containing biomethane (bio-CH_4). Biogas is primarily a mixture of bio-CH_4, carbon dioxide (CO_2) and some trace gases (Nwokolo et al., 2020). Bio-CH_4 is a clean-fuel component of biogas that can produce electricity, co-generation assisted space heating and cooling (Ciampi et al., 2018; Ghersi et al., 2021) and could help in reducing reliance on conventional energy resources.

Water industries have significantly practiced the AD process to treat sewage sludge. However, due to slow hydrolysis, digestion takes a longer time (Shi et al., 2017), reducing the overall efficiency of AD (Hansen et al., 2021; O'Shea et al., 2021). Therefore, anaerobic co-digestion (AcoD) has been adopted in recent years to mitigate this issue and recover high-quality bio-CH_4 (Cheng et al., 2021; Mancini et al., 2021; Nghiem et al., 2017; Tabatabaei et al., 2020a,b; Vinardell et al., 2021), and some of these studies are summarized in Table 1. Recently, submerged anaerobic membrane bioreactors (SAnMBRs) have attracted the scientific community through growing concerns about treating AcoD effluent due to high chemical oxygen demand (COD) and a deficit of microbes due to washout leading to poor methanogenesis (Rabii et al., 2019). SAnMBRs have been widely accepted as a robust technology due to numerous potential benefits; including the ability to operate at higher organic loading rate (OLR) (Gautam et al., 2022; Vu et al., 2020), i.e., process more significant quantities of biodegradable waste (Du et al., 2020), complete retention of microbial biomass, ability to disengage solid retention time (SRT) and HRT at higher OLRs (Berkessa et al., 2018), lower sludge production (Abuabdou et al., 2020) and tri-resource recovery (energy, nutrient and reclaimed water) at a low footprint (Gautam et al., 2022).

As shown in Table 1, previous studies conducted by Cheng et al. (2020), Lei et al. (2019), Moñino et al. (2017) and Xiao et al. (2017) using SAnMBR coupled with polymeric membranes treating wastewater containing high organic and particulate matter has shown superior treatment performance. However, most of these studies were conducted using polymeric membranes at OLR ranging from 0 – 9.5 kg-COD/m^3/d. There is no evidence that SAnMBR coupled with ceramic ultrafiltration (UF) flat-sheet membrane treating OFFW blended DWW has been significantly researched at shock-loads of OLR to anticipate their treatment performance and bio-CH_4 yield. Therefore, the treatment performance and bio-CH_4 recovery using SAnMBR at surge OLRs are unknown. Hence, this research investigates the treatment performance and bio-CH_4 yield of a SAnMBR coupled with ceramic flat-sheet UF membrane treating domestic wastewater and a high-solid concentration of OFFW under surge OLR variations.

In summary, the investigation of SAnMBR performance using DWW and OFFW in terms of COD and total organic carbon (TOC) removal (%), mixed liquor suspended solids (MLSS) and mixed liquor volatile suspended solids (MLVSS) concentrations, VFA (acetic acid) formation (%), and bio-CH_4 production (%) were conducted in 5 phases at OLR 5.16, 0.49, 3.18, 6.14 and 22.57 kg-COD/m^3/d. The MLSS and MLVSS behavior was studied using Nagaoka's model (Nagaoka et al., 1998). Sigmoidal models were used to simulate the cumulative daily bio-CH_4 yield during each phase to estimate critical model parameters representing bio-CH_4 yield, and the obtained results were statistically validated using ANOVA. The results of this study are anticipated to facilitate further research and development of SAnMBRs and promote their application in treating blended DWW and OFFW generated at modern-high rise and similar residential establishments.

2. Material and method

2.1. Submerged anaerobic membrane bioreactor (SAnMBR) experimental setup

The lab-scale SAnMBR experiment setup consists of a water-jacketed, continuously stirred glass fermenter (bioreactor) vessel with a 5 L hydraulic capacity, as shown in Fig. S1. The SAnMBR was operated using a BIOSTAT® automated controller (Applikon Bio Console ADI 1035). Polyvinyl chloride (PVC) feed and permeate containers of 20 L capacity were used to feed and collect raw and treated wastewater through a submersible pump connected to the BIOSTAT®. Level sensors attached with SAnMBR were connected to the BIOSTAT® to draw the wastewater from the feed tank using the submersible pump. The inoculum was kept in suspension by maintaining consistent and thorough stirring using a mechanical stirrer attached with the SAnMBR and controlled using the BIOSTAT®. Strict anoxic conditions were held in

Table 1

Comparison of performance of recent AnMBR studies.

Substrate	Configuration	Operating parameters and influent characteristics	Organic removal efficiency and methane yield	Reference
Organic fraction of food waste and domestic wastewater	SAnMBR, FS, UF, Ceramic	COD: 1.10–13.15 g/L HRT: 0.58–2.24 d Temperature: 37 $\pm$ 2 °C OLR$_{max}$: 22.57 kg-COD/m^3/d MLSS: 5.82–16.36 g/L MLVSS: 5.52–15.46 g/L	COD removal : >97.4%; 0.34–0.70 L-CH4/ g-COD$_{removed}$	This study
Urban wastewater and kitchen food waste	External-SAnMBR, HF, PVDF	HRT: 0.74–1.25 d Temperature: 25 $\pm$ 5 °C OLR$_{max}$: 1.04 kg-COD/m^3/d TS: 12.8–16.5 g/L VS: 8.9–11.4 g/L	COD removal : >97%; 0.07–0.34 L-CH4/ g-COD$_{removed}$	Moñino et al. (2017)
Food waste and sewage sludge	High-solid external SAnMBR, PTFE	HRT: 15–30 d Temperature: 37 $\pm$ 2 °C MLTS: 25–30 g/L	COD removal : >99.4%; 0.30–0.54 L-CH4/g-VS$_{fed}$	Cheng et al. (2021)
Canned coffee processing wastewater and waste activated sludge	SAnMBR, FS, Polyethylene	TCOD:42.4 $\pm$ 9.9 g/L HRT: 36–3 d Temperature: 55 $\pm$ 1 °C OLR$_{max}$: 9.18 kg-COD/m^3/d MLVSS: 21.4–47.3 g/L	COD removal: >90%; 0.251–0.307 NL/g-COD$_{removed}$	Lei et al. (2019)
High-strength kitchen waste slurry	External-AnMBR, HF, UF, PVDF	Total COD: 78–100 g/L HRT: 20.8–10.2 d Temperature: 39 $\pm$ 1 °C OLR$_{max}$: 9.3 kg-COD/m^3/d MLVSS: 15.2–28.8 g/L	COD removal:68.8–84.4%; 0.192–0.274 NL/g-COD$_{removed}$	Xiao et al. (2017)

SAnMBR–Submerged anaerobic membrane bioreactor, FS—Flat sheet, UF—Ultrafiltration, HF—Hollow fiber, PVDF—Polyvinylidene fluoride, PTFE—Polytetrafluoroethylene.

Table 2a

SAnMBR operating condition and flow rates.

Phase*	OLR (kg-COD/m^3/d)	HRT/(d)	Feed flow rate (L/d)
1	5.16	2.24	2.23
2	0.49	2.24	2.23
3	3.18	2.24	2.23
4	6.14	0.58	8.58
5	22.57	0.58	8.58

*pH - 7, *Temp - 35 $\pm$ 2 °C.

the SAnMBR and the pH and temperature maintained at 7 $\pm$ 0.5 and 35 $\pm$2.5°C, respectively, using a pH regulator system connected to the BIOSTAT$^{\circledR}$ and hot bath attached to the bioreactor. To maintain the desired pH level in the bioreactor, two electronically driven acid (0.1M HCl) and base (0.1M NaOH) dosing pumps were used to inject the acid/alkali solutions into the system in a controlled manner. Two ceramic flat-sheet UF (ITN Germany) with a mean pore size of 0.1 μm and an effective filter area of 0.01 m^2 were immersed in the bioreactor to separate the biomass and produce high-quality treated effluent using a peristaltic precision pump (Masterflex L/s 07551-20) (Gautam et al., 2022).

2.2. SAnMBR operation

Excluding the stabilization and acclimatization phase, the SAnMBR was continuously operated in 5 phases with different operating conditions for 91 days, as shown in Table 2a. OLRs of 5.16, 0.49, 3.18, 6.14 and 22.57 kg-COD/m^3/d were supplied to the SAnMBR system to observe its performance with respect to sudden down-surge and shock-load of OLR. During Phase 1–5, feed flowrate and backwash rate were regulated to obtain the designed OLR values, as shown in Table 2a. The SAnMBR system was operated in 30-minute cycles (2 cycles per hour), consisting of 27 min filtration and 3 min of high-intensity backwash. During Phase 1–3 and Phase 4–5, the average operational flux was maintained at 0.116 m/d and 0.429 m/d, respectively, resulting in a decrease in HRT from 2.24/d to 0.58/d. A suction/vacuum pressure transducer was also installed to observe and record TMP during the study.

During the study, ex-situ cleaning of the membrane was conducted when the transmembrane pressure (TMP) reached 40 kPa. As the TMP approached 40 kPa, the membrane module was taken out, cleaned with Milli-Q, and later soaked in 0.8 g/L sodium hypochlorite for 1 h to remove irreversible and persistent foulants. The backwashing flux was 0.504 m/d. The reactor was sealed using silicon sealant (RS-PRO), thoroughly checked for leaks using leak detectors, and then purged with nitrogen to maintain a strict anoxic environment inside the bioreactor. The produced biogas composition was manually measured with the help of a biogas analyzer (Geotech Biogas 5000) daily.

Table 2b
Domestic wastewater recipe for quantity equivalent to 20 L.

Chemical component	Composition	Conc. (mg/L) $\pm 5\%$	Qty per 20 L (g)
Glucose	$C_6H_{12}O_6$	710	14.2
Ammonium Acetate	CH_3COONH_4	200	4
Sodium Bicarbonate	$NaHCO_3$	750	15
Ammonium Chloride	NH_4Cl	30	0.6
Mono-potassium Phosphate	KH_2PO_4	30	0.6
Di-potassium Phosphate	K_2HPO_4	60	1.2
Magnesium Sulphate	$MgSO_4.7H_2O$	50	1
Calcium Chloride Di-hydrate	$CaCl_2.H_2O$	30	0.6
Sodium Chloride	$NaCl$	30	0.6

Table 3
Wastewater characteristics and composition.

Phase	Days of operation	WW composition	SSW (g/L)	COD_{in} (g/L)	OLR (kg-COD/m^3/d)	MLSS (g/L)[a]	MLVSS (g/L)[a]
Phase 1	0–13	DW + SSW	10	11.55	5.16	16.36	15.46
Phase 2	14–25	DW only	0	1.10	0.49	12.35	11.67
Phase 3	29–62	DW + SSW	6	7.13	3.18	13.01	11.23
Phase 4	63–75	DW + SSW	2.5	3.58	6.14	6.48	5.89
Phase 5	76–91	DW + SSW	12.5	13.15	22.57	5.82	5.52

[a]Values at the beginning of Phase 1–5, WW—Wastewater, DWW—Domestic wastewater, SSW—Semi-solid waste.

2.3. Experimental conditions

(a) Domestic wastewater, semi-solid slurry and inoculum

20 L of domestic wastewater (DWW) was prepared as per the recipe shown in Table 2b to feed the SAnMBR. The wastewater was then transferred into 5 L containers and stored at -18 °C to reduce the rate of chemical reactions. For making batches of 5 L semi-solid waste (SSW) feed solution, collection of 10 g of each organic ingredient, including wheat flour, tomatoes, onion, green leaves, herbs (garlic), minced meat (lamb or chicken), were used. When required, these ingredients were blended into a smooth paste and stored in batches of 250 g for mixing with DWW. The SSW prepared paste was then stored in laboratory zipper bags at -18 °C to reduce decay. The SSW prepared was fed to the SAnMBR in the desired quantity during Phase 1–5, as shown in Table 3.

The inoculum was collected from a mesophilic anoxic bio-digester of a conventional wastewater treatment plant located in West Melbourne and later acclimatized in the SAnMBR with domestic wastewater. To maintain a specific solid retention time (SRT), the biomass (SAnMBR sludge) was not intentionally wasted. However, 100 mL of sludge was collected from the bioreactor for analytical purposes. The bioreactor was occasionally purged using nitrogen gas (1.5 L/min) for 20 min to eliminate oxygen to ensure strict anoxic conditions in the SAnMBR. Table 3 summarizes the experimental study phases, days of SAnMBR operation during these phases, influent chemical oxygen demand COD (COD_{in}) characteristics, OLR (kg-COD/m^3/d) and concentration of mixed liquor suspended solids (MLSS) and mixed liquor volatile suspended solids (MLVSS) in the SAnMBR.

2.4. Analytical methods

COD was measured using the HACH colorimetric method (DR 5000™ UV–Vis Spectrophotometer) to determine the difference in the COD of the influent and the COD of permeate. TOC was measured using a Shimadzu TOC-V analyzer. The MLSS and MLVSS concentration of bioreactor sludge samples was measured using the standard methods (Federation and Association, 2005). The detailed analytical procedure for measuring COD, MLSS and MLVSS is described in the supplementary section (S.1). The system's bio-CH$_4$ composition and flowrate as measured using a high-end gas analyzer (Geotech-Biogas Sampler 5000). The gas samples were taken from the SAnMBR on alternate days through the gas analyzer, and the data obtained were logged and processed through a Microsoft Excel spreadsheet.

2.5. Evaluation of cumulative biomethane production curves using kinetic modeling

The growth curves describe how a variable increases over a particular time interval until it reaches its saturation value. The Bacterial growth curves generally indicate a phase in which the specific growth rate starts at zero (minimum asymptote) and then reaches a maximum growth rate in a certain period, denoted as the lag phase (Ma et al., 2013), see Fig S.2. The growth rate gradually decreases and finally reaches zero, a point of saturation or the maximum asymptote (Ware and Power, 2017). This can be related to the typical growth curves of cumulative bio-CH$_4$ production during Phase 1–5 under various operating conditions.

Bio-CH$_4$ production rates of blended DWW–SSW were simulated and analyzed during Phase 1–5 using sigmoidal models found in literature: Modified Gompertz model (Eq. (1)), modified logistic model (Eq. (2)), and the modified

Richard's model (Eq. (3)) (Zwietering et al., 1990). These sigmoidal models were used to estimate multiple parameters, such as (1) bio-CH_4 production potential (P) (L-CH_4/g-COD), (2) Maximum bio-CH_4 production rate (R) (/d), (3) Lag time (L) (d), (4) Shape coefficient (v), and (5) Correlation coefficient (R^2) for Phase 1–5 under different operating conditions. The modified Gompertz model equation (Dhamodharan et al., 2015) is given by

$$y = P * \exp\left\{-\exp\left[R * \frac{e}{P} * (L - t) + 1\right]\right\} \tag{1}$$

The modified logistic model equation (Ware and Power, 2017) is given by

$$y = P/\{1 + \exp[4 * R/L * (L - t) + 2]\} \tag{2}$$

The modified Richard's model equation (Teleken et al., 2018) is given by

$$y = P * \left\{1 + v * \exp(1 + v) * \exp\left[R * (1 + v) * \left(1 + \frac{1}{v}\right) * \frac{L - y}{P}\right]\right\}^{-\frac{1}{v}} \tag{3}$$

where y is the cumulative bio-CH_4 production (L-CH_4/g-COD) at a given time t (d), P is the maximum bio-CH_4 production potential (L-CH_4/g-COD), R is the maximum bio-CH_4 production rate (L-CH_4/g-COD/d), L is the lag phase time (d), v is shape coefficient, and e is a constant equal to 2.718282.

2.6. Modeling mixed-liquor suspended solid concentration in the SAnMBR

The decay and growth rate of biomass in the SAnMBR system can be expressed using Eq. (4) (Gautam et al., 2022; Nagaoka et al., 1998; Navaratna et al., 2012) below:

$$\frac{dx}{dt} = \mu_s x - k_{dx} x \tag{4}$$

where μ_s is specific MLSS growth rate (/d), x is the concentration of MLSS (g/L) in the SAnMBR and k_{dx} is the endogenous decay rate of MLSS (/d). Eq. (4) can be further simplified as follows:

$$\mu_s x = \frac{Q_i}{V} (S_i - S_e) Y \tag{5}$$

Q_i is the influent (organic feed in L/d), V is the hydraulic volume of the bioreactor (L), S_i, and S_e are the concentration of COD (g/L) in the influent and effluent, Y represents the MLSS yield due to S_i.

Since the SAnMBR operates at a very high biomass concentration, i.e., $S_i \gg S_e$ (Berkessa et al., 2018; Gimenez et al., 2020; Inaba et al., 2020), $S_i = YL$. Where L represents OLR (kg-COD/m^3/d). Eq. (5) (Radjenović et al., 2008), therefore, can be further simplified as,

$$\frac{dx}{dt} = YL - k_{dx} x \tag{6}$$

2.7. Data analysis using statistical tools

SPSS V.21 was used to conduct the non-linear regression analysis of the modified Gompertz, logistic and Richards model (Eqs. (1), (2) and (3)). The value of P, R, L and v were obtained using a minimum residual sum of squares at 95% confidence interval (CI). The best curve fitting results represent a higher correlation coefficient value (R^2). T-test was performed on the fitting results obtained from the modified Gompertz, logistic and Richards model. Further, an analysis of variance (ANOVA) was also performed to validate the fitting results.

Further, the model equation (Eq. (6)) was solved and simulated using Runge–Kutta 4th order differential equation (ODE) solver in Microsoft solver in Excel with a step size of 0.05. The experimental and model-simulated results were fitted to estimate the values of Y and k_{dx}.

3. Results and discussion

The co-digestion based SAnMBR system was fed with SSW-DWW and various ranges of OLR, complex organic fatty lipid content, and high solids concentrations. The primary aim was to observe the SAnMBR's treatment performance under realistic conditions if applied at a pilot scale in modern high-rise or similar residential establishments. Co-digestion aimed to improve the system's efficiency for bio-CH_4 production and improve reclaimed water quality. The objective of changing the operating conditions of the SAnMBR system was to determine the optimum condition that would yield the highest bio-CH_4 and generate high-quality reclaimed water.

Table 4
Performance of SAnMBR under different conditions.

Phase	Days of operation	OLR$_{kg\text{-}COD/m^3/d}$	COD$_{in\ (g/L)}$	Avg. COD removal $_{(\%)}$	TOC$_{in\ (g/L)}$	Avg. TOC removal $_{(\%)}$	MLSS $_{(g/L)}$[a]	MLVSS $_{(g/L)}$[a]
Phase 1	0–13	5.16	11.55	94.38 $\pm$ 1.12	8.72	91.88 $\pm$ 2.79	16.36	15.46
Phase 2	14–25	0.49	1.10	84.88 $\pm$ 2.99	0.73	80.88 $\pm$ 6.94	12.35	11.67
Phase 3	29–62	3.18	7.13	93.33 $\pm$ 1.30	4.70	88.67 $\pm$ 3.37	13.01	11.23
Phase 4	63–75	6.14	3.58	96.67 $\pm$ 1.02	1.15	91.89 $\pm$ 0.67	6.48	5.89
Phase 5	76–91	22.57	13.15	98.05 $\pm$ 0.72	10.72	94.88 $\pm$ 0.78	5.82	5.52

[a]MLSS and MLVSS values at the beginning of each phase.
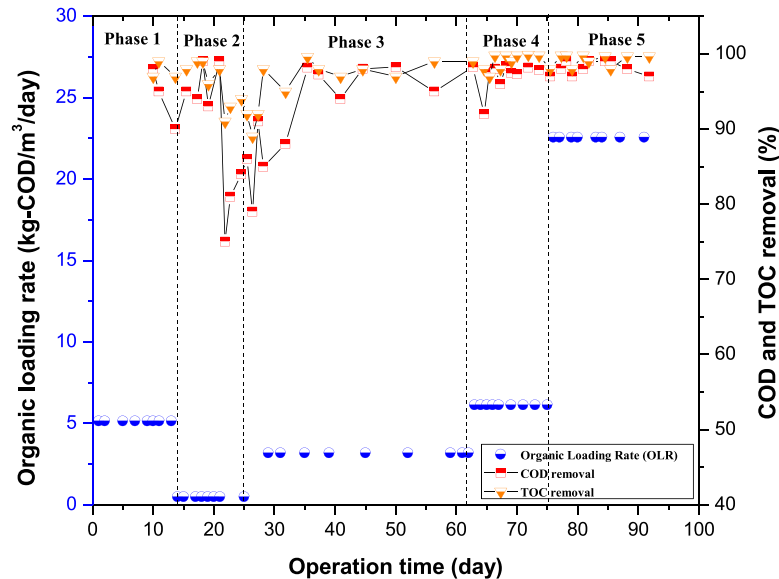
Fig. 1(a). Variation of COD and TOC removal (%) during different Phase 1–5 at OLR 5.16, 0.49, 3.18, 6.14 and 22.57 kg-COD/m³/d.

3.1. Treatment performance of submerged anaerobic membrane bioreactor (SAnMBR)

The removal of organics through SAnMBR was thoroughly investigated to measure COD and TOC at various concentrations of SSW blended with DWW. As shown in Table 4, the long-term continuous SAnMBR experiment was divided into five phases (Phase 1–5) and performed with a periodically shortening HRT from 2.24/d during Phase 1–3 to 0.58/d during Phase 4–5, at OLR 5.16, 0.49, 3.18, 6.14 and 22.57 kg-COD/m³/d respectively.

Ideally, Phase 1 would yield the highest treatment performance in terms of COD removal at a high organic loading rate (Boonyungyuen et al., 2014; Burman and Sinha, 2020; Gautam et al., 2022; Vinardell et al., 2021; Yu et al., 2021) of 5.16 kg-COD/m³/d. This phenomenon was in agreement with the SAnMBR performance during Phase 1 in the presence of SSW-DWW, and the SAnMBR demonstrated superior treatment performance in COD and TOC removal averaging 94.38 $\pm$ 1.12% and 91.88 $\pm$ 2.79% removal efficiencies, as shown in Fig. 1(a) and Table 4. The SSW-DWW composition in the bioreactor during Phase 1, Phase 3, Phase 4 and Phase 5 caused the reactor to produce good effluent quality with good average COD and TOC removal (%). However, visible foaming was seen inside the bioreactor during Phase 1. Initially, the foaming was eliminated by constant nitrogen purge (2 L/min) at regular intervals and by increasing the magnetic stirrer speed to 120 RPM. However, Zhang et al. (2019) reported that increasing the stirring would agitate microbial activity, and nitrogen purging reduces the quality of methane gas (Yu et al., 2021). Therefore, once the foaming was controlled, the nitrogen supply was intermittently supplied at weekly intervals, and the magnetic stirrer speed was reduced to 90 RPM.

It was observed that during Phase 2, the average COD and TOC removal were subpar compared to Phase 1 and Phases 3–5. This was due to the absence of SSW in Phase 2, which led to a 10% drop in COD and TOC removal efficiency, averaging 84.88 $\pm$ 2.99 % and 80.88 $\pm$ 6.94%, respectively. The primary purpose of the co-digestion of SSW with DWW was to improve the system's efficiency in both methane production and effluent quality. Nghiem et al. (2017) reported that food waste is the most common co-substrate in anaerobic co-digestion experiments, while Vinardell et al. (2021) reported that co-digestion of sewage sludge and food waste in an AnMBR system has an affinity for higher methane production and producing superior effluent quality.

The SSW was re-introduced in Phase 3 with an OLR of 3.18 kg-COD/m³/d, and expectedly, the COD and TOC removal efficiency increased by 10%, averaging 93.33 $\pm$ 1.30% and 88.67 $\pm$ 3.37% respectively. Furthermore, the OLR in Phase
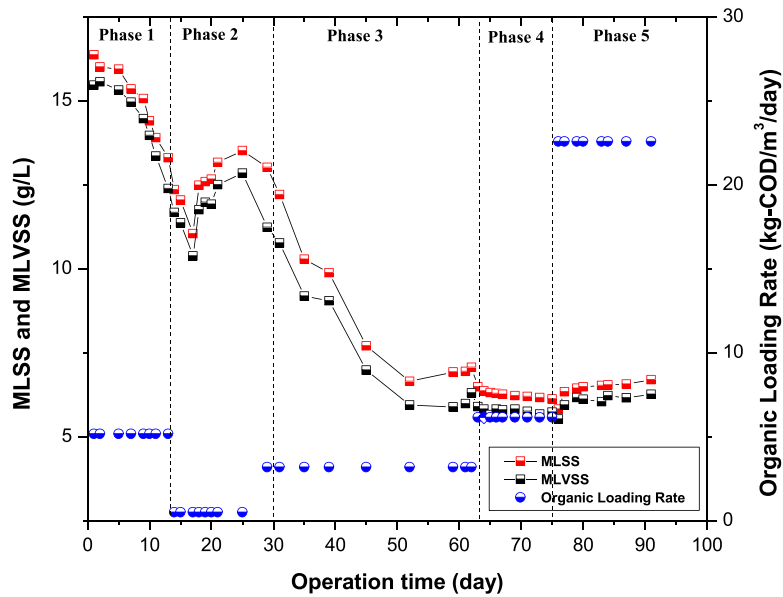
Fig. 1(b). Variation of MLSS and MLVSS during different phases at OLR 5.16, 0.49, 3.18, 6.14 and 22.57 kg-COD/m³/d.

Table 5
Model estimated parameters for different phases.

| Symbol	Description	Unit	Phase 1	Phase 2	Phase 3	Phase 4	Phase 5
L	OLR	kg-COD/m³/d	5.16	0.49	3.18	6.14	22.57
x	MLSS	g/L	16.36	12.35	13.01	6.48	5.82
Parameter			Estimated parameter values				
Y	Yield coefficient	g-MLSS/g-COD	0.15	0.20	0.10	0.12	0.12
K_{dx}	Decay rate of MLSS	(/d)	0.08	0.02	0.18	0.20	0.21

4 was increased to 6.14 kg-COD/m³/d, and SAnMBR showed a superior treatment performance by demonstrating an average COD and TOC removal of 96.67 ± 1.02% and 91.89 ± 0.67%, respectively. In phase 5, a sudden shock-load of OLR 22.57 kg-COD/m³/d was applied to the SAnMBR system. The system showed unexpectedly superior performance, showing an average COD and TOC removal efficiency of 98.05 ± 0.72% and 94.88 ± 0.78%, respectively. It should be noted that no visible fouling was seen during Phase 2–5; hence, the fouling data is not included in this study.

It should be noted that the highest removal of COD and TOC was observed during Phases 1, 4 and 5, as shown in Fig. 1(a), at a high OLR concentration in the presence of SSW-DWW, while the SAnMBR demonstrated a low COD and TOC removal during Phase 2, at low OLR in the absence of SSW. These indicate that SAnMBRs can show superior treatment performance at high OLRs if fed with DWW containing high organic content, such as OFFW.

3.2. Long-term variation of mixed liquor suspended solid (MLSS) and mixed liquor volatile suspended solids (MLVSS)

The biomass yield (*Y*) and decay rate (*k_dx*) also affect the amount of waste anaerobic sludge production and affects the overall stability and treatment performance of SAnMBR. At the same time, the OLR is known to have a positive correlation with *Y* (Gautam et al., 2022). Similarly, *k_dx* is known to positively correlate with the microbial population in the bioreactor (Rittmann and McCarty, 2001). As shown in Fig. 1(b) and Table 4, the MLSS and MLVSS at the beginning of Phase 1 were 16.36 g/L and 15.46 g/L, which gradually decreased and reached 13.3 g/L and 12.38 g/L by the end of Phase 1 at OLR 5.16 kg-COD/m³/d. This gradual decrease in MLSS concentrations during Phase 1 (refer to Fig. 2) was studied using Eq. (6) (Nagaoka et al., 1998), and parameters were estimated using model-simulated and experimental results, as shown in Table 5. The values of *Y and k_dx* during Phase 1 were estimated as 0.15 g-MLSS/g-COD and 0.08/ d respectively as shown in Table 5.

The estimated parameter shows that the decay of MLSS (*k_dx*) was significantly less, while the yield coefficient (*Y*) was high during Phase 1, leading to a gradual decline in MLSS and MLVSS concentrations until the beginning of Phase 2. However, on removing the SSW composition from DWW, MLSS and MLVSS gradually increased from 12.35 to 13.51 g/L and 13.51 to 12.84 g/L during Phase 2, but showed a slight decline by the end of Phase 2 at OLR 0.49 kg-COD/m³/d. The estimated values of *Y and k_dx* for Phase 2 were 0.20 g-MLSS/g-COD and 0.02/d, as shown in Table 5. The model-estimated
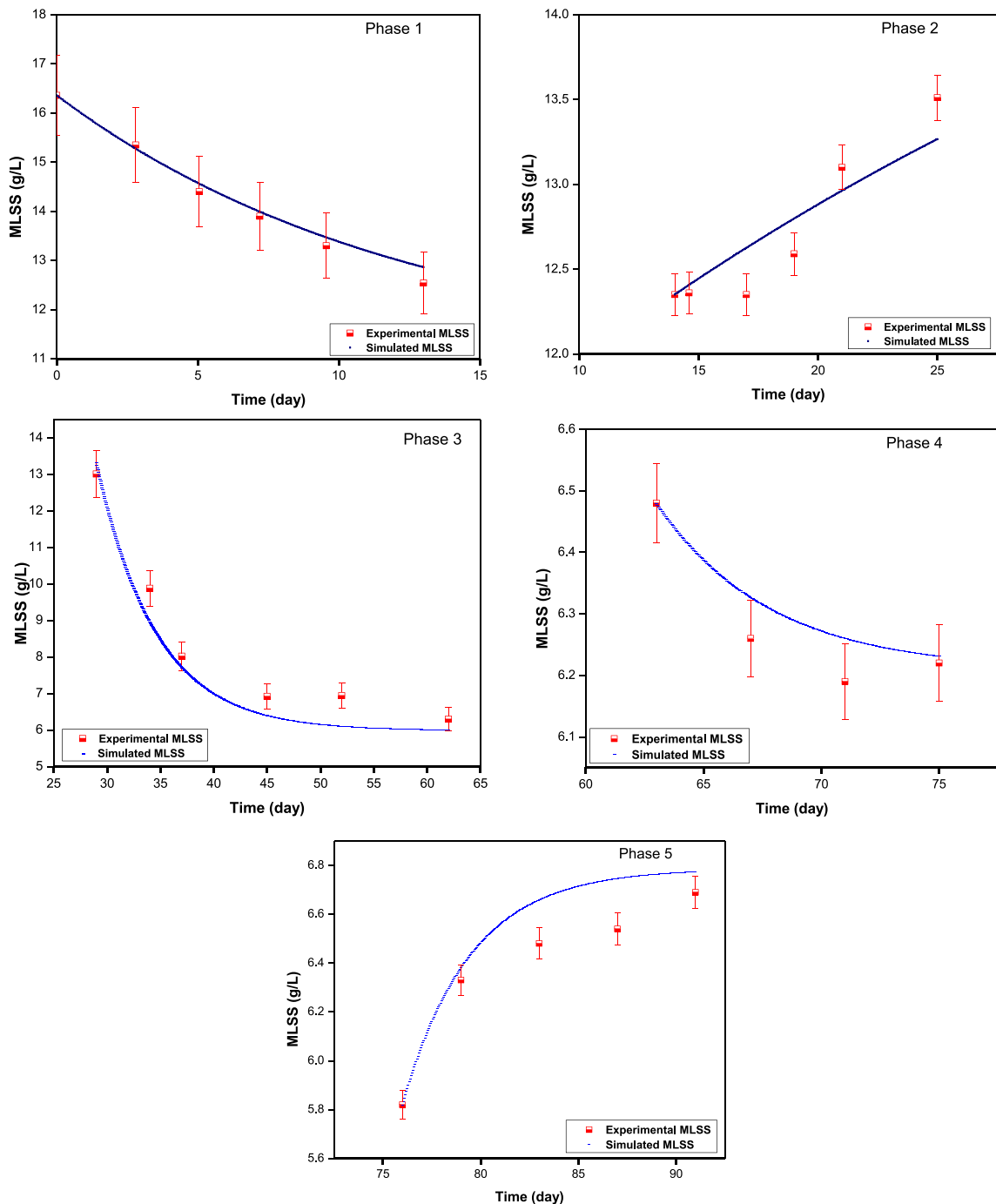
Fig. 2. Model fitting results with experimental data for Phase 1–5 at OLR 5.16, 0.49, 3.18, 6.14 and 22.57 kg-COD/m^3/d.

parameter shows that Y increased during Phase 2, while the value of k_{dx} decreased, allowing the MLSS and MLVSS to increase and stabilize.

On further increasing the OLR by threefold in Phase 3, i.e. 3.18 kg-COD/m^3/d, Y decreased to 0.10 g-MLSS/g-COD while k_{dx} increased nine-fold and reached 0.18/d. It should be noted that Phase 3 was conducted for the most prolonged duration, i.e. for 33 days. As depicted in Fig. 1(b), MLSS and MLVSS showed a decreasing trend from 13.01 g/L to 7.06 g/L (45%) and 11.23 g/L to 6.3 g/L (43.9%) in 33 days during Phase 3. Previous studies (Navaratna et al., 2012; Wei et al., 2014; Yu et al., 2021; Zhou et al., 2012) reported that OLR has a proportional relation with MLSS concentration; however, the behavior of MLSS may change under a different set of operating conditions (Dvořák et al., 2011). Previous studies conducted by Cheng

et al. (2020) and Gautam et al. (2022) has shown a positive correlation between MLSS and OLR in a SAnMBR treating high strength wastewater.

The applied OLR, MLSS and MLVSS are critical parameters in determining the SAnMBR filterability and operational stability. Lousada-Ferreira et al. (2015) and Mohan and Nagalakshmi (2020) found that a concentration of MLSS greater than 10 g/L caused entrapment of all particles greater than 20 μm. The entrapped particles caused the solids to bulk, increasing the membrane resistance and resulting in system overloading. The overloading of the system reduces the sludge settling ability and reduces the overall treatment efficiency of SAnMBR. Similarly, suppose the MLSS/MLVSS concentration is too low; in that case, the anaerobic digestion process may not remove sufficient amounts of organic matter from the wastewater (Tran et al., 2022) due to lesser organics that may inhibit the biological filtration process. However, the influence of MLSS and MLVSS on fouling and treatment efficiency is not very consistent and sometimes contradictory (Deowan et al., 2015).

On further increasing the OLR to 6.14 kg-COD/m^3/d during Phase 4, the decrease in MLSS and MLVSS concentration was significantly lower, i.e. 5.55% and 2.71%. Y and k_{dx} slightly increase during Phase 4 and reached 0.12 g-MLSS/g-COD and 0.20/d, respectively. During Phase 5, the OLR was increased by four-folds, i.e., 22.57 kg-COD/m^3/d and the MLSS and MLVSS concentrations gradually increased by 14.94% and 13.4% and stabilized at 6.69 g/L and 6.26 g/L by the end of Phase 5. It was also found that Y and k_{dx} almost got stable and reached 0.12 g-MLSS/g-COD and 0.21/d, respectively, as shown in Table 5. This phenomenon was in agreement with past AnMBR studies conducted by Aramrueang et al. (2016), Burman and Sinha (2020) and Wei et al. (2014). However, at high OLR (Phase 1, Phase 3, Phase 4 and Phase 5), the value of k_{dx} was found higher than the range (0.04–0.1 g-COD/g-COD at 35 °C) reported by Batstone et al. (2002), Henze et al. (2000) and Metcalf et al. (1991) for a conventional anaerobic reactor. This might be due to the substantial decay of cells due to endogenous respiration (Gautam et al., 2022). The HRT during Phase 1–3 was 2.24 and 0.58 (/d) during Phase 3–5, and it was observed that Y correlated negatively with HRT and OLR while k_{dx} correlated positively with OLR and HRT. This observation was in agreement with the study conducted by Muda et al. (2011) to understand the influence of loading rate (OLR) on Y and k_{dx} in an sequential batch reactor (SBR) treating textile wastewater. Additionally, the MLSS/MLVSS ratio averaged 0.93 ± 0.02 throughout the study, which shows the positive stability of SAnMBR. This shows that SAnMBR system coupled with ceramic UF membrane could be applied at pilot scale to treat OFFW mixed with municipal sewage at surge OLRs, low HRT and high MLSS and MLVSS concentrations.

3.3. Biomethane (bio-CH$_4$) production

The gas counter used in this study recorded an average biogas yield of 21 L/d. The produced gas composition includes H$_2$S, CO$_2$, and CH$_4$. Considering the ratios of the gas components, the average bio-CH$_4$ production during the stable SAnMBR operation in Phases 4 and 5 was estimated as 70.09 ± 7.44%, showing a very promising outcome from this study to apply this technology commercially. Previous studies on the AD for bio-CH$_4$ production, sludge accumulation, and mass balancing of carbon have concluded that 74% of removed organic carbon is converted into bio-CH$_4$, and 15% is stored in sludge (Ali et al., 2020; Bekiaris et al., 2015; Cheng et al., 2018; Ware and Power, 2017).

At the beginning of Phase 1, the SAnMBR demonstrated a decline in bio-CH$_4$ production, but it slightly increased by the end of Phase 1. It was noted that the average bio-CH$_4$ and acetic acid concentration (%) during Phase 1 were 44.5 ± 6.81% and 44.03 ± 1.56%, respectively, at an OLR of 5.16 kg-COD/m^3/d. During this period, the SAnMBR demonstrated a high TOC removal (91.88 ± 2.79 %).

As illustrated in Table 3, during Phase 2, only DWW (without SSW) was fed into the SAnMBR, and an OLR of 0.49 kg-COD/m^3/d was maintained. This led to a decline in the average daily bio-CH$_4$ concentration by 9% compared to Phase 1. The average daily bio-CH$_4$ and acetic acid concentration (%) during Phase 2 was 39.72 ± 5.65 % and 37.23 ± 3.29%, respectively. It should be noted that the TOC removal during this period declined by approximately 10% and averaged 80.88 ± 6.94%. In Phase 3, the feeding OLR was increased up to 3.18 kg-COD/m^3/d by adding 6 g/L of SSW to DWW, and this resulted in improving methane yield (%) and the SAnMBR effluent quality. The SAnMBR showed a rapid increase in bio-CH$_4$ production and TOC removal, averaging 64.58 ± 11.70 % and 86.7 ± 3.37 %, respectively, with an average acetic acid concentration of 40.74 ± 3.22%.

Acetic acid is produced due to acetogenesis and is a crucial substrate for methanogenesis (Ali et al., 2020; Mancini et al., 2021). The availability of acetic acid indicates that digestion is in constant process with the simultaneous production of bio-CH$_4$ (Liu et al., 2018), assisting the micro-organisms to efficiently combine and convert acetic acid, hydrogen and carbon dioxide into bio-CH$_4$. This phenomenon was in agreement with acetic acid production during Phase 1–3. The decrease in the concentration of acetic acid during Phase 2 correlates positively with less production of bio-CH$_4$ and vice-versa for Phase 1 and Phase 3.

The bio-CH$_4$ concentration continued stable and peaked at 67.12 ± 7.15 % and 73.06 ± 6.48 % during Phase 4 and Phase 5 at a high OLR of 6.14 and 22.57 kg-COD/m^3/d, respectively. During this period, the acetic acid concentration averaged 41.04 ± 2.52 % and 43.05 ± 2.65 %, respectively. Additionally, the SAnMBR system demonstrated a stable and high TOC removal, averaging 91.89 ± 0.67% and 94.88 ± 0.78, respectively, during Phase 4 and Phase 5, as shown in Fig. 3. This clearly indicates that the bio-CH$_4$ gas production in a SAnMBR positively correlates with applied OLR and acetic acid concentration in the reactor.

Based on the above results, it can be articulated that the amount of carbon in the sludge varies linearly with the type of semi-solid feed applied to the SAnMBR system. The importance of calculating the removal efficiency of TOC was to evaluate
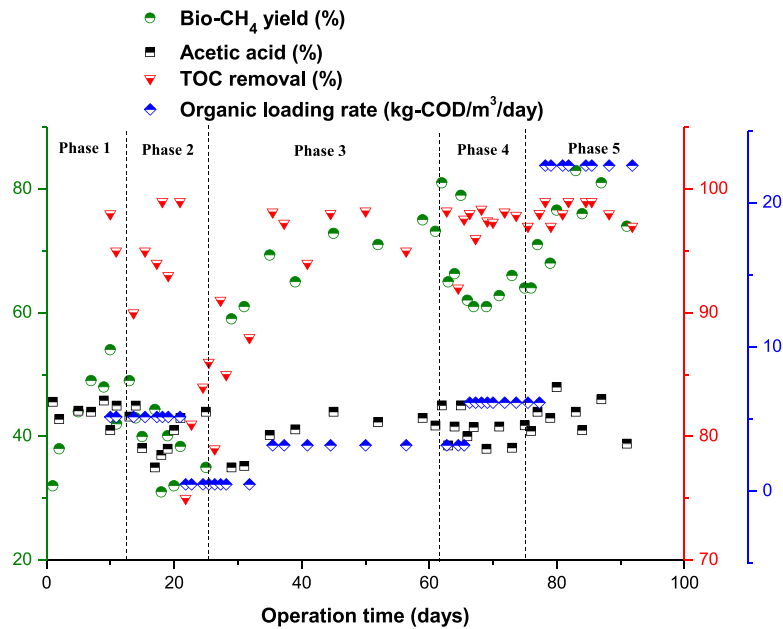
Fig. 3. Variation of bio-CH$_4$ concentration (%), acetic acid production (%) and TOC removal (%) at OLR 5.16, 0.49, 3.18, 6.14 and 22.57 kg-COD/m^3/d.

the efficiency of SAnMBR in producing bio-CH$_4$. The purpose of running the AnMBR system in various conditions was to determine the system efficiency at different organic loading when producing bio-CH$_4$. The expectations of increasing OLR past Phase 5 are vague but not covered in this study. Since fouling did not happen during Phase 1–5, the relationships covering trans-membrane pressure (TMP) are not presented in this paper.

3.3.1. Modeling biomethane (bio-CH$_4$) production using sigmoidal models and validating results using analysis of variance (ANOVA)

The cumulative daily bio-CH$_4$ production was calculated for all five phases (Phase 1–5) during the study, and the obtained results were fitted with the model-simulated results. From the visual representation, modified Gompertz, modified logistic, and modified Richards demonstrated a good fit, as shown in Fig. 4 and Table S.1. The bio-CH$_4$ production parameters (P, R, L, and (v)) were well represented by these sigmoidal models with minimum variance, as summarized in Table S.1. The non-linear regression analysis was conducted using SPSS v.21 for experimental and model-simulated results, showing a correlation coefficient (R^2) of 0.99, 0.96 and 0.96 for modified Gompertz, modified logistic and modified Richards models, respectively, as shown in Table S.1.

Further, an analysis of variance (ANOVA) was also performed and found, $p < 0.05$, which indicated a significant fit. While conducting the statistical analysis of model-simulated results in ANOVA and estimating the model parameters (P, R, L and v), it was found that the parameters estimated using the modified Gompertz model were in the range previously reported in literature (Zhang et al., 2021; Zwietering et al., 1990).

The estimated value of maximum bio-CH$_4$ production rate (R) for Phase 1–5 using the modified Gompertz model was 0.664 L-CH$_4$/g-COD/d. On the contrary, modified logistic and modified Richards models demonstrated unacceptably low values of R, i.e., 0.074 and 0.013 L-CH$_4$/g-COD/d, respectively. The modified Gompertz model represented R well and gave acceptable values similar to results reported by Nguyen et al. (2016), which match our experimental results. On the other hand, the modified logistic and Richards model showed unacceptable R values (Matheri et al., 2016; Ware and Power, 2017) for Phase 1–5.

Furthermore, when bio-CH$_4$ production potential (P) was calculated using the modified logistic model, it was too low for all phases combined, i.e., 50.91 L-CH$_4$/g-COD. At the same time, modified Gompertz and modified Richards gave similar results values, as shown in Table S.1. On comparing each parameter critically, it was also found that it is hard to predict the efficiency and reliability of a sigmoidal mode by its visual fit and R^2 results only. Therefore the residual sum of squares (RSS) was calculated (Table S.1) to validate the reliability of the results obtained. The obtained results concluded that the Gompertz model overall demonstrated more accurate parameter estimation, graphical representation (Fig. 4), and correlation coefficient (R^2) compared to the modified logistic and Richards model and was found suitable for this study.

4. Conclusions and future perspectives

A Submerged AnMBR (SAnMBR) with 5 L hydraulic capacity was researched for highly fluctuating OLRs for over 90 days, feeding a highly organic semi-solid wastewater consisting of a mixture of blended organic matter and synthetic
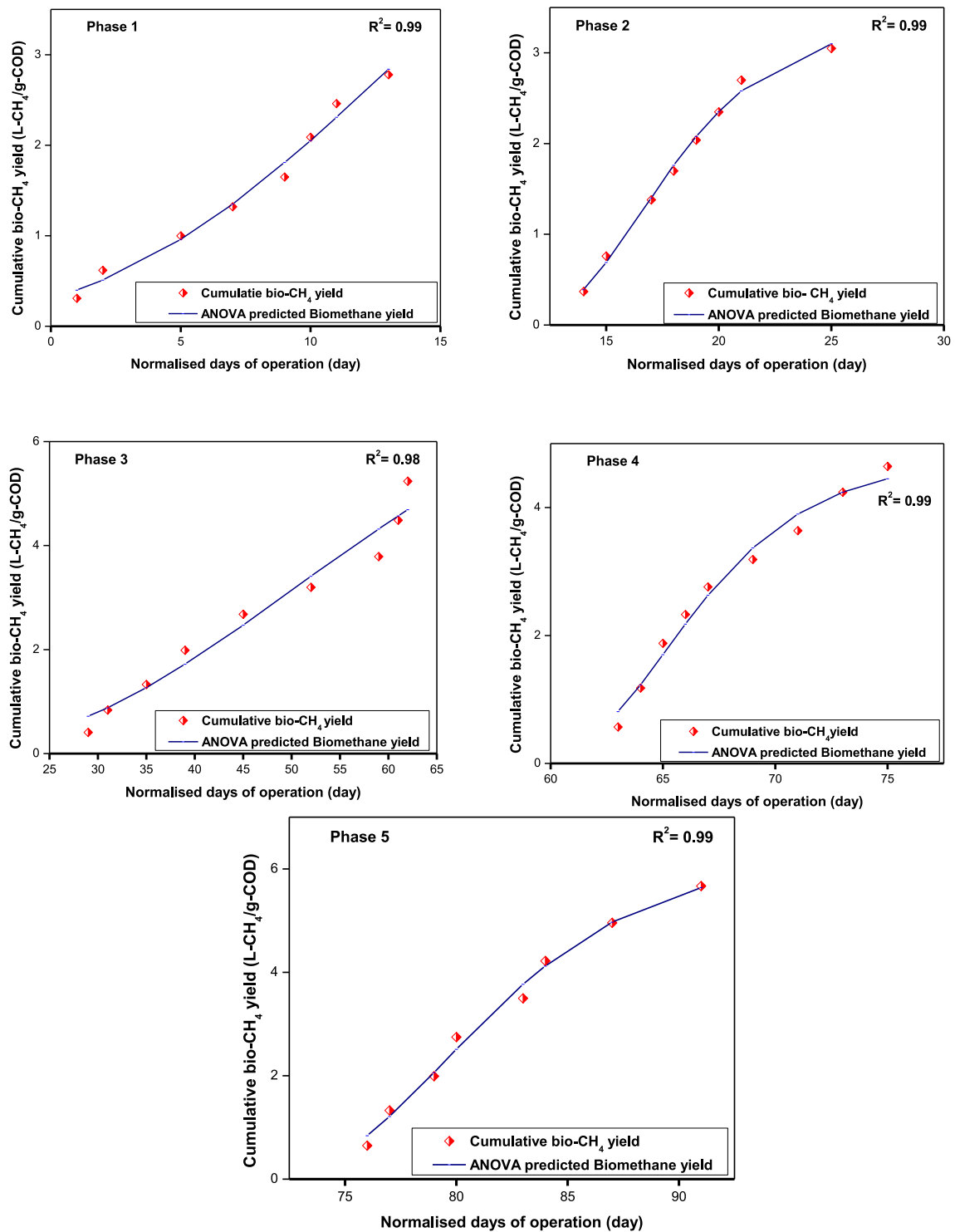
Fig. 4. Modified Gompertz model-simulated fitting results with experimental data for phases 1–5 at OLR 5.16, 0.49, 3.18, 6.14 and 22.57 kg-COD/m^3/d.

DWW. The feeding OLR varied from 0.49 kg-COD/m^3/d to 22.57 kg-COD/m^3/d and maintained an HRT of 0.58–2.24 days. During the stable phases (Phase 4 and 5), operated at OLR of 6.14 and 22.57 kg-COD/m^3/d, the SAnMBR system recorded an average COD and TOC removal of 97.43 ± 4.7 %5% and 94.20 ± 4.7 %, respectively. The SAnMBR also showed its resilience for operating at highly fluctuating shock-loads of semi-solid organic waste streams and produced a significantly high yield

(70.09 $\pm$ 7.44%) of bio-CH$_4$, evidencing the suitability of applying this co-digestion based SAnMBR technology for high-rise establishments. The behavior of MLSS and MLVSS was thoroughly investigated using a numerical model and validated using statistical tools. It was observed that *Y and k$_{dx}$* during each phase demonstrated a good correlation between applied OLR and the subsequent MLSS variation. The outcomes of this study also confirmed the application of SAnMBR for an efficient and sustainable pretreatment system for treating wastewater consisting of high-organic content and a complex array of particulate matter discharging from various food industry processes such as abattoirs. Future efforts should look into the sustainability features of the results obtained using advanced sustainability assessment tools, including life cycle assessment, exergy and its combinations with environmental and economic analysis, respectively.

CRediT authorship contribution statement

Rajneesh Kumar Gautam: Mathematical modelling, Investigation, Data curation, Writing – original draft, Visualization, Statistical analysis, Writing – review & editing. **Robert Valente:** Investigation, Writing – review & editing. **Haitham Abbas:** Investigation, Writing – review & editing. **Anh Bui:** Investigation, Writing – review & editing. **Nandkishor More:** Writing – review & editing, Supervision. **Stephen Gray:** Writing – review & editing, Supervision. **Shobha Muthukumaran:** Writing – review & editing, Supervision. **Dimuth Navaratna:** Conceptualization, Investigation, Mathematical modelling, Data curation, Writing – review & editing, Supervision.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Appendix A. Supplementary data

Supplementary material related to this article can be found online at https://doi.org/10.1016/j.eti.2022.102763.

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