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# Food waste-derived volatile fatty acids platform using an immersed membrane bioreactor

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## Abstract

Volatile fatty acids (VFAs) are the key intermediates from anaerobic digestion (AD) process that can be a platform to synthesize products of higher value than biogas. However, some obstacles still exist that prevent large-scale production and application of VFAs, key among them being the difficulty in recovering the acids from the fermentation medium and low product yields. In this study, a novel anaerobic immersed membrane bioreactor (iMBR) with robust cleaning capabilities, which incorporated frequent backwashing to withstand the complex AD medium, was designed and applied for production and *in situ* recovery of VFAs. The iMBR was fed with food waste and operated without pH control, achieving a high yield of 0.54 g VFA/g $VS_{added}$. The continuous VFA recovery process in the iMBR was investigated for 40 days at OLR 2 gVS/L/d and 4 gVS/L/d without significant change in the permeate flux at a maximum suspended solids concentration of 31 g/L.

Keywords: Volatile fatty acids; In situ recovery; Immersed membrane bioreactor; Fouling control; Food waste

## 1. Introduction

About 1.3 billion tons of food waste is generated globally along the food supply chain every year (FAO, 2011). The available treatment methods of this waste, that also makes up a significant portion of the municipal solid waste, include composting, incineration, landfilling and anaerobic digestion (AD). Among these methods, AD is an established and environment-friendly technology (Xu et al., 2018; Zhang et al., 2014). The main application of this technology is normally production of biogas by completing all the four steps of the biodegradation process; i.e. hydrolysis, acidogenesis, acetogenesis and methanogenesis (Castellano-Hinojosa et al., 2018). Biogas is primarily used for the production of electricity, heat and transport fuel (Kleerebezem et al., 2015). However, due to the increasing demand of essential bio-based materials of higher value than biogas, alternative applications of the AD process are needed (Kleerebezem et al., 2015). Volatile fatty acids (VFAs), which are the key intermediate metabolites during AD and consisting of up to six carbon atoms, have therefore been considered as a platform for developing a wide spectrum of essential products that are currently derived from fossil-based feedstocks such as polymers, alcohols, olefins and ketones (Aydin et al., 2018; Chen et al., 2013).

In order to hinder the bioconversion of the VFAs into biogas by methanogens in dedicated VFAs-producing AD procedures, external pH adjustment using chemicals, such as NaOH and HCl, is a common practice. The use of these chemicals, however, could reduce the activity of the other microorganisms enriched in the microbial consortium, thereby decreasing the VFA yields, in addition to increasing the process costs. Alternatively, the pH drop as a result of the accumulating VFAs in the bioreactor can be utilized for preventing the bioconversion of VFAs into methane. Nevertheless, appropriate measures are required to compensate for the product

inhibition brought along mainly by the undissociated acids that dominate at low pH levels (López-Garzón & Straathof, 2014).

Although the VFA platform is a promising approach for the application of the AD technology, there are only a few reports on its large-scale implementation (Holtzapple et al., 1999; Kim et al., 2018). This shortage of industrial VFA-based processes originating from organic waste is attributed to difficulty in recovery of the VFAs from the culture medium and the low product yields (Kim et al., 2018; Rebecchi et al., 2016). Indeed, several reports in literature have focused on VFA production from food waste using batch cultivations in which key operating conditions were studied (Chen et al., 2013; He et al., 2019; Hussain et al., 2017; Sheng et al., 2019; Xu et al., 2012). However, these processes were not scaled up to continuous operations that are suitable for modeling large-scale procedures. Furthermore, for the VFAs produced in such batch processes to be applied in the subsequent downstream stages, a shutdown of the operation is required followed by a centrifugation-based VFAs recovery (Chen et al., 2013). In addition to the operation downtime caused by the centrifugation procedure, it is an energy intensive and laborious separation technique when high volumes of VFAs are to be extracted from the AD fermentation broths that are usually highly concentrated with solids.

To fulfil the requirements of optimized large-scale continuous or semi-continuous VFA production processes, a recovery method that is easy to retrofit in existing AD installations and minimal utilization of chemicals is preferable. The use of membrane separation technique has been proposed as a favorable means to continuously recover the VFAs while they are being formed in the bioreactor (Trad et al., 2015). While a number of studies using membrane separation techniques for the recovery of VFAs have been carried out, they were either performed in short filtration periods (Grzenia et al., 2008; Zacharof & Lovitt, 2014), applied synthetic

medium (Aydin et al., 2018; Béligon et al., 2016; Tugtas, 2014; Zhou et al., 2013) or their focus was on hydrogen production (Lee et al., 2014; Trad et al., 2015). A promising VFAs production and recovery strategy that potentially fulfils the criteria for an AD process treating solid organic waste involves applying an immersed membrane bioreactor (iMBR) constructed using a 2$^{nd}$ generation integrated permeate channel (IPC) flat-sheet membrane panel that is robust, compact and backwashable (Doyen et al., 2010). In addition to providing utilizable organic acids from the AD system, integrating immersed membrane separation in VFA production processes would minimize cell washout from the bioreactor as well as reduce the process-inhibiting influence of the acids, consequently enhancing the biodegradation process (López-Garzón & Straathof, 2014).

The aim of the present study was therefore to develop a novel concept to continuously produce and recover VFAs at high yields. To achieve this, food waste was biodegraded in an iMBR in acidic conditions without external chemical addition. Using this strategy, the cells were maintained in the bioreactor at low hydraulic retention time during the AD process without the production of biogas. The selection of the membrane panel used for the VFA extraction was based on its capability to allow for continuous and vigorous cleaning. The flow characteristics through the membrane panel at varying solids concentrations and the performance of the AD process were assessed.

## 2. Materials and methods

### 2.1 Food waste and seeding inoculum

Synthetic food waste mimicking a typical mixture from the European Union (Ariunbaatar et al., 2016) was used as substrate and consisted of (on wet basis): fruits and vegetables 79%, pasta and rice 5%, bread and bakery 6%, meat and fish 8% and dairy 2%. To ensure consistency,

a bulk of the substrate was prepared by milling the food mixture using an electric blender (Waring® CB15, CT, USA). It was then divided in appropriate portions, stored in a freezer and thawed in a cold room (4 – 5 °C) prior to use as feed in the iMBR. Some characterization data for this substrate were (average ± 2 standard deviations) 16.11 ± 0.98% total solids (TS), 15.41 ± 0.94% volatile solids (VS), 160 ± 9.90 g/L total chemical oxygen demand (tCOD), 60.00 ± 5.66 g/L soluble chemical oxygen demand (sCOD), 27.02 ± 0.67% TS crude protein, 19.67 ± 2.68% TS total lipids and pH 4.5 ± 0.1**.** The seeding inoculum used in the AD process was collected from an upflow anaerobic sludge blanket reactor treating wastewater (Hammarby Sjöstad, Stockholm, Sweden), and contained 9.55 ± 0.35% TS and 6.48 ± 0.25% VS. The inoculum was acclimatized to the new cultivation conditions by feeding 0.2 gVS/L/d of food waste for 18 days prior to the actual fermentation.

### 2.2 Immersed membrane bioreactor (iMBR) set up and operation

A continuous stirred tank reactor (CSTR) equipped with a radial impeller (BBI-biotech GmbH, Berlin, Germany) was used. The 2nd generation IPC membrane panel used was obtained from Flemish Institute of Technological Research (VITO NV, Mol, Belgium). The flat-sheet membrane had an effective area of 68.6 $cm^2$ and the membrane coating was prepared from hydrophilic polyethersulfone (PES) with an average pore size of 0.3 μm and a clean water permeability of 3000–4000 $L/h/m^2/bar$. The panel was inserted into the CSTR below the medium surface as shown in Fig 1. The gas sparging on the membrane surface, during the filtration cycles, was enabled by 12 inbuilt gas diffusers at the bottom of the panel (6 on each side with a diameter of 0.5 mm each) using $N_2$. The permeate flow was measured using Atrato 710-V11-D ultrasonic flowmeter (Titan Enterprises Ltd, Dorset, England). The bioreactor was inoculated with 90 g granules and then food waste was added. Afterwards, the reactor was filled with tap water to

achieve a working volume of 2 L. The initial pH was set at 6.0 using 2M HCl and 2M NaOH after which the AD process was allowed to proceed without further external pH adjustment. The reactor was then sparged with $N_2$ for 5 min to create anaerobic conditions. The impeller speed was set at 75 rpm and the temperature was maintained at 37 °C. The gas produced in the headspace was measured by a volumetric gas flow meter (μFlow, Bioprocess Control, Lund, Sweden). The permeate pump was set at 10 rpm. The forward filtration and backwash cycles were automatically operated for 210 sec and 30 sec respectively using an electric relay (Zelio Logic SR2A101BD, Schneider Electric, USA).

During the actual anaerobic fermentation process, a hydraulic retention time (HRT) of 6.67 days and an organic loading rate (OLR) was 2 gVS /L/d were applied and the OLR was increased to 4 gVS /L/d from day 31 (i.e. after approximately 3 times the HRT after achieving stability in the initial OLR) until day 40 to assess the membrane performance at high solids concentration. The iMBR was operated in semi-continuous mode by carrying out the filtration cycle before feeding the reactor once every day.

## 2.3 Analytical methods

Gas samples were collected using a 0.25 mL gas-tight syringe (VICI, Precision Sampling Inc., Baton Rouge, LA, USA) and were analyzed by using a Perkin-Elmer gas chromatograph (Clarus 550; Perkin Elmer, Norwalk, CT, USA) equipped with a packed column (CarboxenTM 1000, 6’ × 1.8” OD, 60/80 Mesh, Supelco, Shelton, CT, USA) using a thermal conductivity detector (Perkin-Elmer, Norwalk, CT, USA) with an injection temperature of 200 °C. The carrier gas was $N_2$ with a flow rate of 30 mL/min at 75 °C. The VFAs were analyzed using a high-performance liquid chromatograph (HPLC) (Waters Corporation, Milford, CT, USA) with a hydrogen-based column (Aminex HPX87-H; BioRAD Laboratories, München, Germany) at 60

°C. The mobile phase was 5 mM $H_2SO_4$ at 0.6 mL/min and the peak detection was achieved using ultraviolet (UV) absorption detector at 210 nm (Waters 2487, Waters Corporation, Milford, CT, USA).

The total nitrogen, suspended solids (SS), TS, VS, tCOD, sCOD and $NH_4^+$-N concentrations were determined according to standard methods (APHA-AWWA-WEF, 2005). Crude protein was calculated using a factor of 6.25 (Sosulski & Imafidon, 1990). The total lipid content were determined according to Majdejabbari et al. (2011). For determination of the COD equivalents of the VFAs, the conversion factors used were 1.07 g COD/g acetic acid, 1.51 g COD/g propionic acid, 1.82 g COD/g butyric and iso-butyric acids, 2.04 g COD/g valeric and iso-valeric acids, and 2.20 g COD/g caproic acid.

The undissociated acid concentrations in the permeate were determined using Henderson-Hasselbalch equation. Equation 1 was applied to include the temperature correction factor for the calculation of the permeate flux (Fan et al., 2006).

$$J_{20} = J_T * 1.025^{(20 - T)} \quad (1)$$

where $J_{20}$ is the permeate flux at 20 °C and $J_T$ is the permeate flux at the operating temperature.

## 3. Results and discussion

In this study, a semi-continuous VFAs production process was established by circumventing the bioconversion of the VFAs into methane by operating in low pH conditions and the membrane-assisted *in situ* extraction of the VFAs using an iMBR that ensured a continuous supply of a VFA solution for further bioprocessing or purification. Unlike other membrane-assisted VFA separation processes, the current study was performed using food waste as substrate

which forms a complex matrix in the fermentation medium. The complexity of the medium is due to the presence of high solid concentrations and macromolecules such as proteins that propagate membrane fouling (Ho & Zydney, 1999). Therefore, a flat-sheet membrane panel that allows for constant backwashing was selected to deal with the aforementioned inherent properties that would drastically decrease the membrane performance. Although gas sparging has been applied in previous iMBRs for $H_2$ production (Lee et al., 2008; Lee et al., 2014), this is the first report on the use of backwashing for sustaining a stable filtration during VFAs recovery. The iMBR was therefore constructed using a CSTR and an immersed 2$^{nd}$ generation IPC membrane panel which, in addition to its robustness, is easy to operate, has a small footprint and its energy demand is low. Exceptional membrane performance was observed as the permeate flux experienced minimal changes over a period of 40 days for the fermentation medium at high suspended solids concentration as discussed in the following sub-sections. Moreover, using the iMBR set-up resulted in a high yield of VFAs even with high presence of undissociated acids.

### 3.1 Continuous membrane-assisted recovery of VFAs

The anaerobic iMBR set-up applied here was developed for high production and *in situ* recovery of VFAs during biotreatment of food waste is the first of its kind. It was operated by conducting a filtration cycle in which a VFA-concentrated solution was extracted daily and replaced by a dilute food waste solution at a HRT of 6.67 days. The short retention time was designed to assess the performance of the system at high volumetric productivity. The robust membrane cleaning system in this iMBR set-up benefitted from both the mechanical agitation (using the radial impeller) and the membrane module- related gas sparging. The membrane fouling was further controlled by backwashing as well as the membrane relaxation, which was the idle time (approximately 20 h) between the filtration cycles. The inbuilt panel diffusers sparge

gas adjacent to the membrane surface and double-layer membrane panel allowed for backwashing pressures of up to 2 bar. Unlike previous practices with AD media filtration, these qualities were synchronized in this work so that the high shear stress applied on membrane surface could remediate fouling-related issues.

The effect of the AD medium on membrane fouling is exacerbated in case of immersed membranes as the membrane is in direct contact with the medium. Therefore, any change in the medium quality and viscosity influences shear stress over the membrane and deteriorates membrane-cleaning efficiency. Due to excessive membrane fouling and solid accumulation, most membrane-separation systems tested previously cannot guarantee a continuous or semi-continuous VFAs separation that would lead to high process productivity using solid organic waste. However, since the continuous reactor operation is a key element for sustainable industrial processes, the current iMBR set-up was designed to overcome the mentioned drawbacks and was applied for continuous extraction of VFAs in a harsh AD process.

### 3.2 Permeate flux though the membrane panel

During the filtration cycles, the permeate pump provided the suction and the soluble compounds passed through the semi-permeable membrane barrier. After a forward cycle (run for 210 sec), a backwash cycle using the permeate was run for 30 sec to constantly unblock the membrane pores as observed in a typical profile in Fig 2. The average forward flow was 0.65 L/h on day 1 at a maximum of 0.77 L/h (average permeate flux of 62.5 $L/m^2/h$). From day 14 some stability in the flow was established for the remaining of the filtration periods at an average value of 0.56 L/h with the average permeate flux ranging between 52 – 53 $L/m^2/h$. Due to the backwashing strategy, membrane fouling was well controlled and the permeate flux reduced only by an average 0.22 $L/m^2/h$ per day and the overall reduction was 16.4 % by day 40.

The performance of a similar iMBR set-up was first evaluated in our research group in a less complex lignocellulosic fermentation medium with high-suspended solid content (Mahboubi et al., 2017). It was observed that in a wheat straw hydrolysate containing up to 85 g/l of polysaccharides and 20% w/v SS (undigested residuals mainly consisting of lignin) the iMBR could provide a stable filtration performance during bioethanol fermentation. In the abovementioned experiment, excessive fouling was prevented using frequent backwashing and gas sparging. However, the complexity in the composition and content of an AD system using food waste as the substrate is incomparable to that of controlled lignocellulosic fermentation. In case an effective immersed membrane filtration system is to be applied for the AD system, different parameters such as SS concentration, lipid and protein content, medium pH, bacterial colony morphology (suspended or flocculated), extracellular polymeric substance (EPS) and soluble microbial product (SMP) content of the media should be taken into account.

### 3.3 Performance of the iMBR at high suspended solids

The retained bacterial cells and food waste unhydrolyzed solids accumulated with time in the iMBR at a rate of 0.62 g/L/d (Fig 3). This resulted in a progressively SS-rich fermentation medium in the iMBR. Nevertheless, from the permeate flux profiles (Fig 3), it was observed that the iMBR performed exceptionally well in sustaining the filtration and VFAs recovery rate with a maximum SS of 31 g/l (Fig 3). In pressure-driven membrane processes used for product recovery in bioprocesses, maintaining a stable permeate flux in order to guarantee high productivity rates is of great importance (Carstensen et al., 2012; Singhania et al., 2012). However, the filtration efficiency is constantly jeopardised by different fouling mechanisms such as concentration polarization and cake layer deposition (Park et al., 1997). One of the main contributors to filtrate flux deterioration is increase in medium viscosity (Yoon, 2015). It is noteworthy that the

viscosity of the fermentation medium is in direct relation with cell (biomass) concentration in the iMBR and the substrate unhydrolyzed SS. A high medium viscosity leads to high propensity to membrane fouling (Lee & Yeom, 2007; Wicaksana et al., 2006). Thus, there was need for complimenting the backwash with consistent gas sparging and vigorous medium agitation in the iMBR.

It has been observed that filtration performance in iMBRs is susceptible to deterioration by the increase in suspended solid concentration (Jeison & van Lier, 2006). The other problem confronted in recovery of VFAs using membrane filtration is the protein and lipid content of the as-received food waste and hydrolysis products. As highlighted earlier, the food waste used in this research contained up to (on w/w dry basis) 27.02% proteins and 19.67% lipids. The hydrophobic nature of these compounds and their inclination towards adhesion to abiotic surfaces, such as the polymeric membrane applied here, aggravates the fouling tendency. The adhesion of lipid and protein macromolecules originating from the food waste, the hydrolysate (fatty acids and amino acids), the EPS and semi-soluble microbial products to the membrane surface during fermentation prepares a perfect adhesive and nutritional base for bacteria attachment (Rosenberger et al., 2005; Vanysacker et al., 2013).

### 3.4 High VFA yield at low pH

In the iMBR set-up used in this study, the pH is intentionally kept low in order to inhibit methanogens and to circumvent the use of chemicals. The initial pH was set at 6.0 after which no further external pH control was made. The self-sustaining fermentation process proceeded at pH values ranging between 3.6 – 4.1. The by-product collected from the iMBR headspace contained $H_2$ produced at a yield of 2.36 NmL/g $VS_{added}$ during the initial 30 days. The $NH_4^+$-N released during the biodegradation of the proteins in the substrate was 70.48 ± 25.98 mg/L during the

stable phase. Since the pH levels remained low throughout the fermentation process, it was concluded that the ammonia released was insufficient to provide a buffering capacity in the bioreactor that would maintain the pH close to the initially set value.

By preventing the utilization of the VFAs by methanogens, the acidic environment further stimulated the accumulation of the acids in the iMBR, in agreement with Dechrugsa et al. (2013). Consequently, the hydrolyzed organic material in the iMBR was highly acidified as the VFA fraction in the solubilized organic content made up 83% of the sCOD. The low pH condition has also been associated with overall microbial toxicity due to the presence of undissociated acids. These undissociated acids can easily pass across the cell membrane and dissociate inside the cell causing the energy generated in the cell to be directed towards cell maintenance which eventually hinders the microbial productivity (van der Wielen et al., 2000). The low pH condition sustained in the iMBR was therefore a possible cause for reduction in cell growth and this made cell retention of importance. Furthermore, the current process seemed to have been tolerant to the potentially toxic environment since the fermentation proceeded successfully for 30 days in a low pH environment, where the undissociated acids comprised more than 80% of the VFAs produced. Moreover, despite the low pH levels, a high yield of 0.54g VFA/g $VS_{added}$ was realized, which was a better performance than what was previously achieved during biodegradation of food waste with pH control. For instance, Zhang et al. (2005), Jiang et al. (2013) and Lim et al. (2000) used batch reactors and the yields obtained from their processes were 0.31g VFA/g $VS_{added}$ (at pH 7.0), 0.50g VFA/g $VS_{added}$ (at pH 6.0) and 0.40g VFA/g $VS_{added}$ (at pH 5.5) respectively. Lim et al. (2008) conducted a semi-continuous acidogenic fermentation process using continuously stirred reactors at various pH levels. They also reported lower yields of up to 0.26, 0.35, 0.37 VFA/g $VS_{added}$ at pH values of 5.0, 5.5 and 6.0 respectively. The *in situ* removal of the VFAs in the

current iMBR process might have relieved the acid stress on the microbes, leading to the high product yield and could therefore be a promising approach to ensure long-term fermentation processes.

The VFA production and distribution of the individual acids in the filtrate are shown in Fig 4. It can be observed that caproic acid was the predominant metabolite (31 – 47%) at a near constant concentration of 3.0 g/L while acetic acid was the second most predominant product (20 – 30%) at concentrations ranging between 1.3 – 1.9 g/L. This was followed by butyric acid (14 – 23%) at 1.0 – 1.3 g/L and valeric acid (4 – 18%) although the latter reduced steadily from day 16 to day 27. The concentration of propionic acid (3 – 10%) stayed at a level of 0.25 g/L until day 26 and then increased to reach the highest concentration of 0.35 g/L on day 28. The production of branched-chain acids (iso-butyric and iso-valeric) was less than 0.1 g/L throughout the process. The maximum VFA concentration was on day 13 at 7.5 g/L (Fig 4). The aforementioned product distribution can be attributed to a combination of operating conditions in the iMBR. The level of pH is one of the key factors that influence metabolic pathways and thereby controls the VFA composition (Zhou et al., 2018). According to previous studies, the production of acetic acid dominates when the pH is kept in the acidic region while operating around the neutral region has been reported to favor the production of longer-chain carboxylic acids (Kim et al., 2016). A study by Lim et al. (2008) for instance revealed that at pH 5.0, the acetic, butyric and caproic acids comprised of up to 18.2%, 18.4% and 13.0% of the total VFA respectively. The differences in the metabolic activities towards the production of different VFAs in undefined mixed culture systems, such as the one applied in our study, can also be ascribed to the microbial community structure enriched in the seeding inoculum due to the diversity of the bacteria and the available nutrients (Zhou et al., 2018). Therefore, the high production of butyric and caproic acids could be

attributed to possible high activity of the specific acid-producing bacteria (Ai et al., 2016; Jeon et al., 2013). Moreover, the low OLR applied here probably impacted on the metabolic pathways leading to the present VFA composition similar to observations made by Jiang et al. (2013). Their study suggested that the share of acetic acid concentration could decrease at low OLR compared to the share of the longer-chain acids, such as butyric and valeric acids.

## 4. Conclusions

A novel anaerobic iMBR with a robust membrane cleaning system was successfully applied for production and *in situ* recovery of VFAs during biodegradation of food waste without external pH regulation. A high yield of 0.54 g VFA/g $VS_{added}$ at an OLR of 2 gVS/L/d was achieved due to the continuous extraction of the highly undissociated acids through the membrane panel. Due to the frequent backwashing, the membrane filtration performed remarkably well with only a 16.4% reduction in the permeate flux after 40 days of operation at maximum suspended solids concentration of 31 g/L.

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## Figures captions

**Figure 1.** Schematic diagram of the immersed membrane bioreactor (iMBR).

**Figure 2.** The typical volumetric flow profiles during the forward and backwash cycles of the VFA filtration through the immersed IPC membrane panel on day 2.

**Figure 3.** Profiles of the average temperature-corrected flux (triangles) and the suspended solids (circles) in the iMBR during the filtration cycles of the VFA production process at OLR 2 gVS/L/d (I) and 4 gVS/L/d (II).

**Figure 4.** Concentration and percentage distribution of VFAs during AD of food waste in the immersed membrane bioreactor at OLR 2 gVS/L/d.

**Figures**

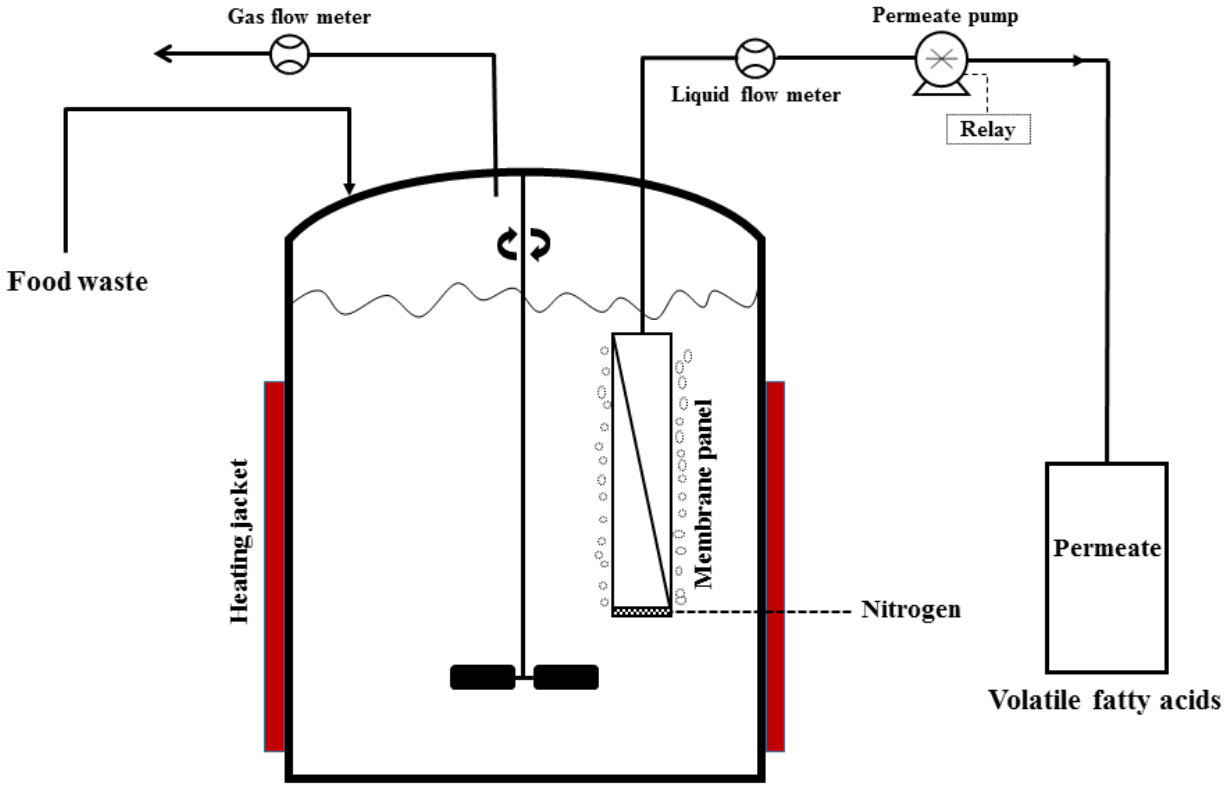



**Fig. 1.**

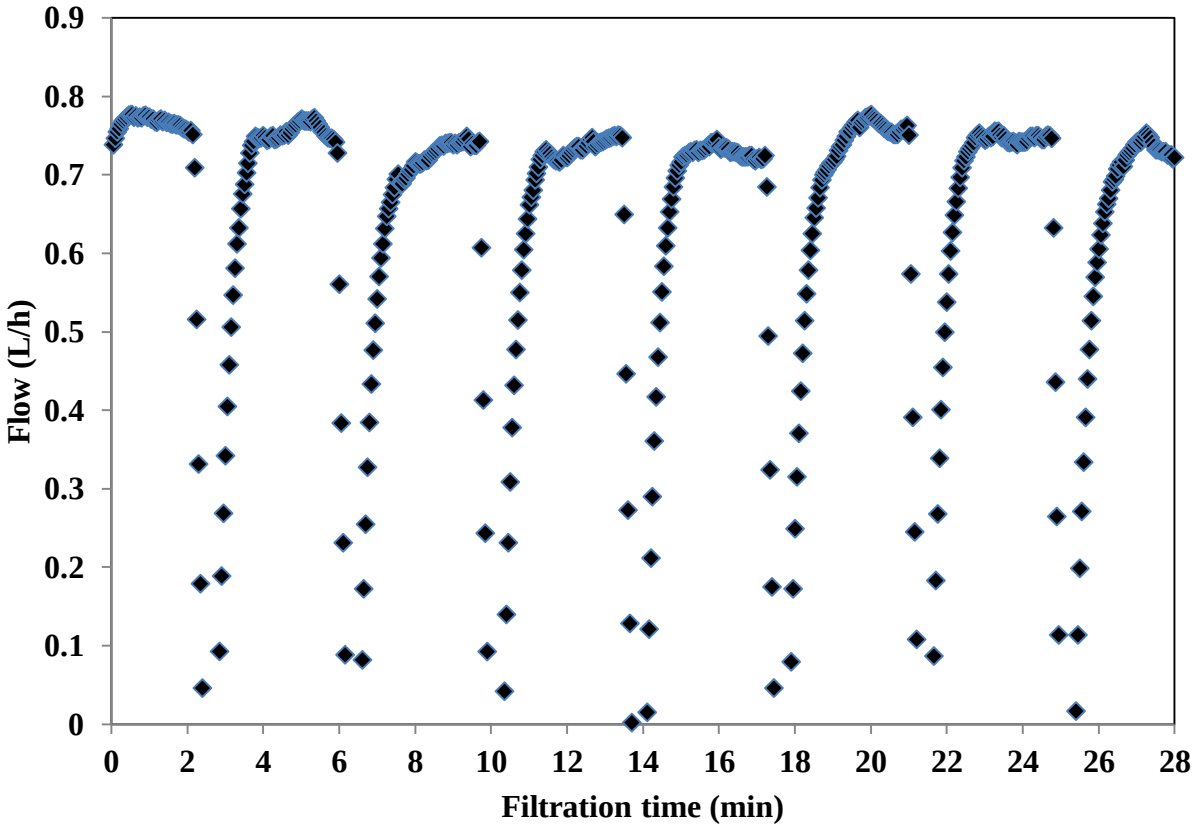



**Fig. 2.**

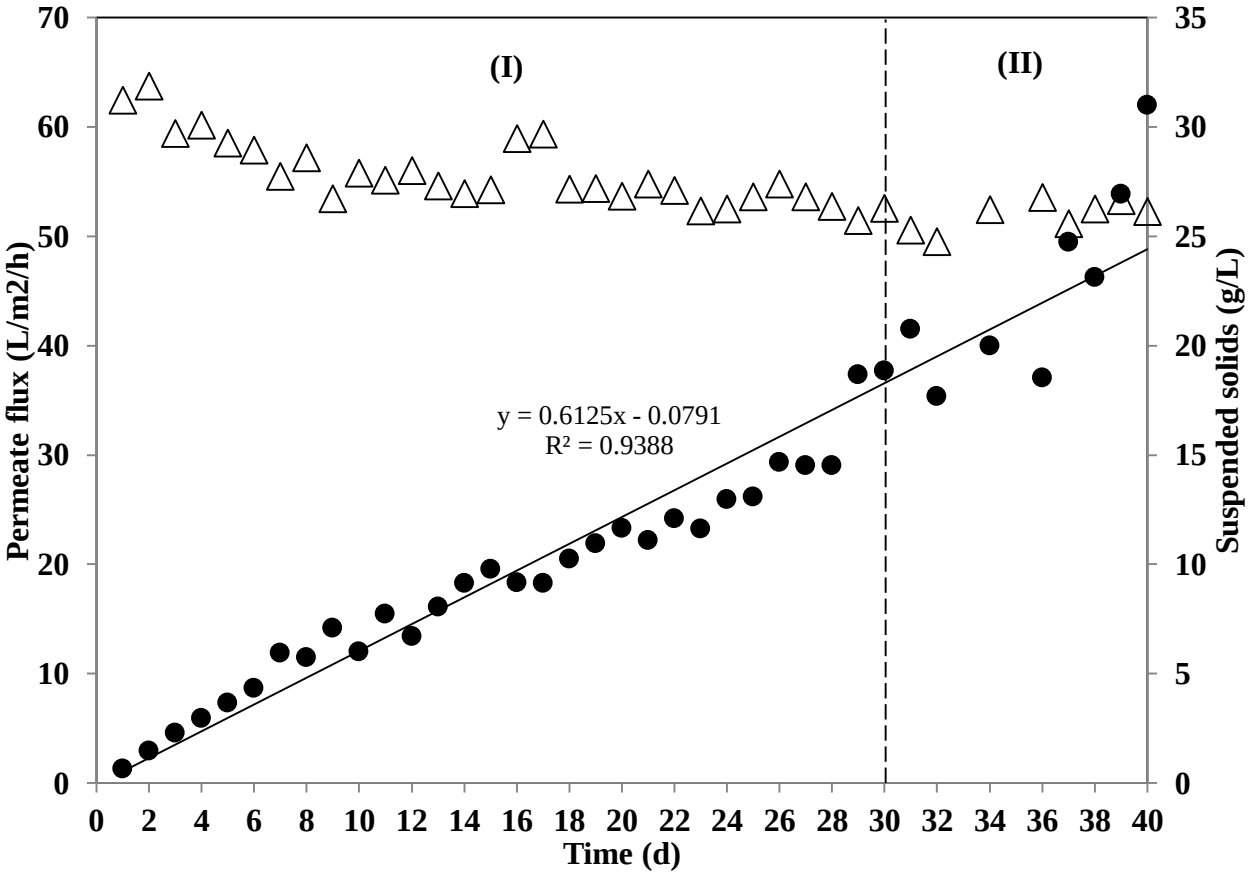



**Fig. 3.**

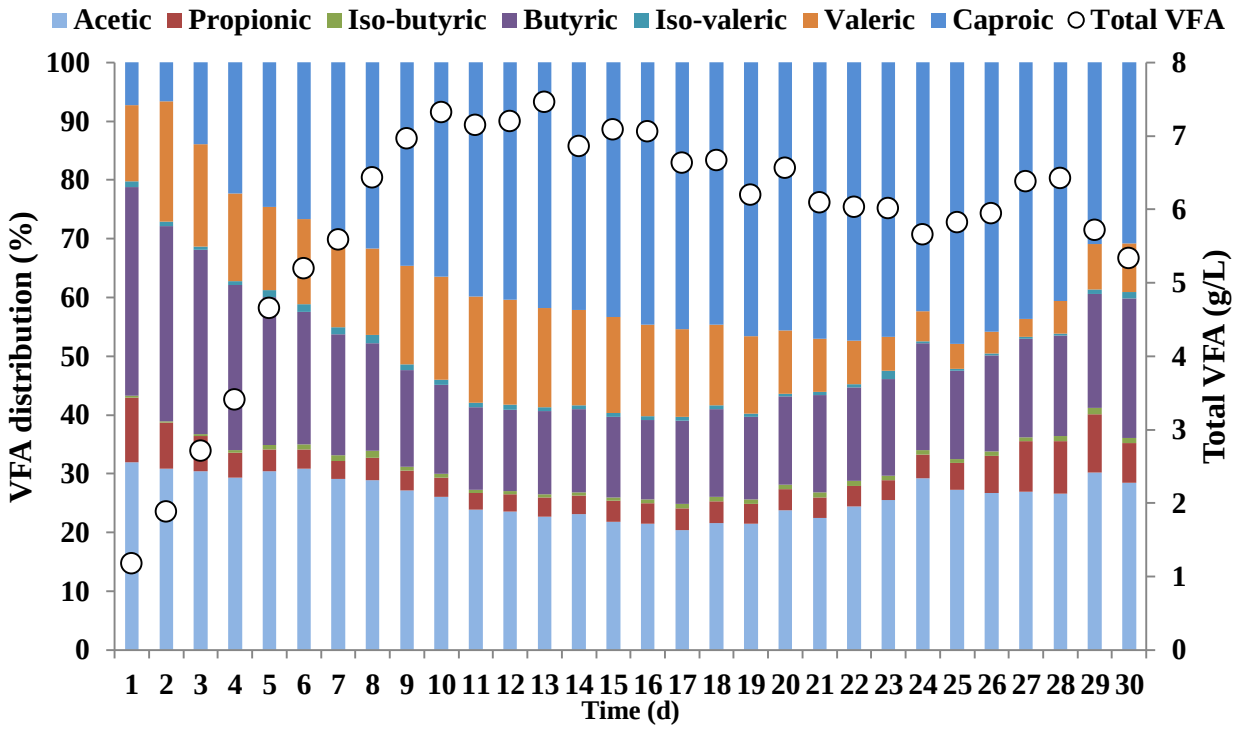
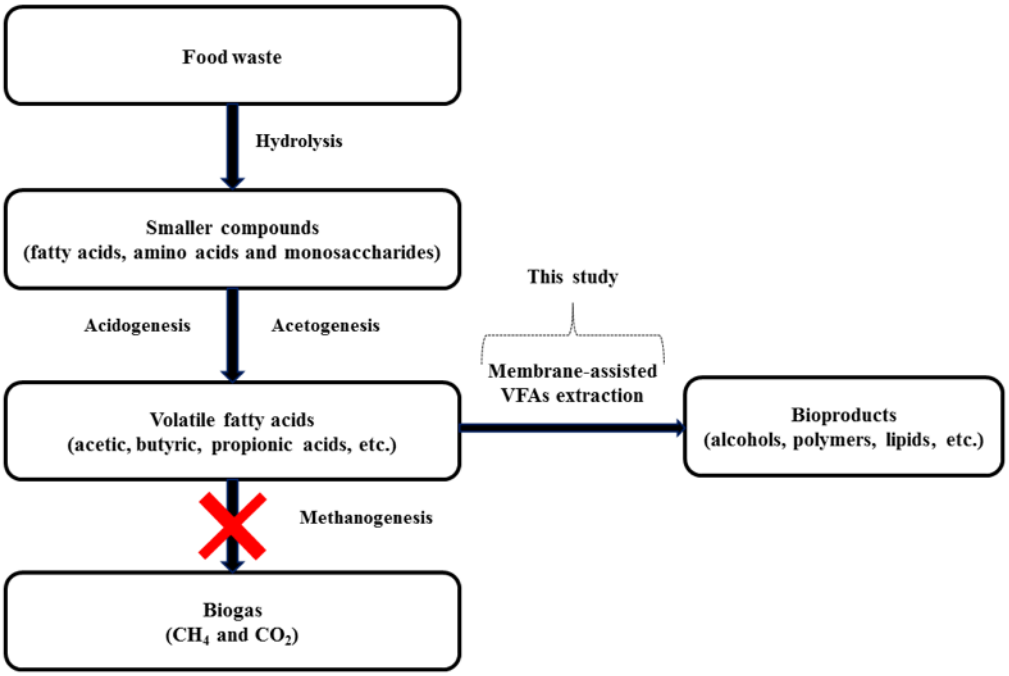



**Fig. 4.**


Food waste
Hydrolysis
Smaller compounds
(fatty acids, amino acids and monosaccharides)
Acidogenesis
Acetogenesis
This study
Membrane-assisted
VFAs extraction
Volatile fatty acids
(acetic, butyric, propionic acids, etc.)
Bioproducts
(alcohols, polymers, lipids, etc.)
Methanogenesis
Biogas
($CH_4$ and $CO_2$)

**Highlights**

- Membrane bioreactor was developed for continuous production of Volatile fatty acids
- Food waste was continuously biodegraded in a novel iMBR without pH control.
- Methanogens were suppressed and *in situ* VFAs recovery performed daily.
- Reduction in permeate flux was 16.4% at maximum solids concentration of 31 g/L.
- A high product yield of 0.54 g VFA/ g $VS_{added}$ was attained.