

# Strategies for the improvement of VFAs recovery in the nanofiltration process within the circular economy framework

PERVEZ MN.<sup>1</sup>, UWINEZA C.<sup>2</sup>, SAPMAZ T.<sup>2,3</sup>, MAHBOUBI A.<sup>2</sup>, HASAN SW.<sup>4</sup>, CAI Y.<sup>5</sup>, ZARRA T.<sup>1</sup>, BELGIORNO V.<sup>1</sup>, TAHERZADEH MJ.<sup>2</sup>, and NADDEO V.<sup>1\*</sup>

<sup>1</sup>Sanitary Environmental Engineering Division (SEED), Department of Civil Engineering, University of Salerno, via Giovanni Paolo II 132, 84084 Fisciano (SA), Italy

<sup>2</sup>Swedish Centre for Resource Recovery, University of Borås, 501 90, Borås, Sweden

<sup>3</sup>Department of Environmental Engineering, Istanbul Technical University, Maslak, 34469 Istanbul, Turkey

<sup>4</sup>Center for Membranes and Advanced Water Technology (CMAT), Department of Chemical Engineering, Khalifa University of Science and Technology, P.O. Box 127788 Abu Dhabi, UAE

<sup>5</sup>Engineering Research Centre for Clean Production of Textile Dyeing and Printing, Ministry of Education, Wuhan Textile University, Wuhan, 430200, China

\*corresponding author:

e-mail: vnaddeo@unisa.it

**Abstract.** Over the past decades, the anaerobic digestion (AD) process has been employed as a potential medium to produce valuable resources from the waste-based feedstock. Moreover, the use of AD technology can generate not only resources, but it also creates an avenue for useful chemicals recovery to further use in commercial purpose, which is a part of the circular economy consortium. Volatile fatty acids (VFAs) are one of the most precious chemical feedstock that can be recovered from anaerobically digested effluent through the use of a pressure-driven membrane filtration process. In an effort to improve the recovery percentages of VFAs from anaerobically digested effluent, this study proposed a sustainable pathway by reusing the permeate effluent in the nanofiltration process. For this purpose, particles free feed solution was produced using the ultrafiltration process and sequentially subjected to the nanofiltration process in the presence of a 200-300 Da nanofiltration membrane. Each permeate was mixed with a known amount of fresh feed and continues up to 3 cycles to be improved the recovery percentages of total VFAs concentrations. Results indicated that recycling strategies could be a potential way to modulate the concentration of VFAs compounds.

**Keywords:** Food waste, volatile fatty acids, nanofiltration, resource recovery, recyclability

## 1. Introduction

The growing number of global populations are generated a vast amount of food waste that requires suitable treatment methods (Caldeira et al. 2021; Papargyropoulou et al. 2014; Schanes et al. 2018). Consequently, tremendous research efforts have been made on food waste valorization into high-value-added products towards the development of circular bioeconomy (Bigdeloo et al. 2021; Ingrao et al. 2018; Naddeo and Taherzadeh 2021; Ong et al. 2018; Pervez et al. 2021;

Slorach et al. 2019). Compared to other techniques, including composting, landfilling and incineration, the anaerobic digestion (AD) process has particularly attracted widely for sustainably managing food waste (Lytras et al. 2021; Wainaina et al. 2019a).

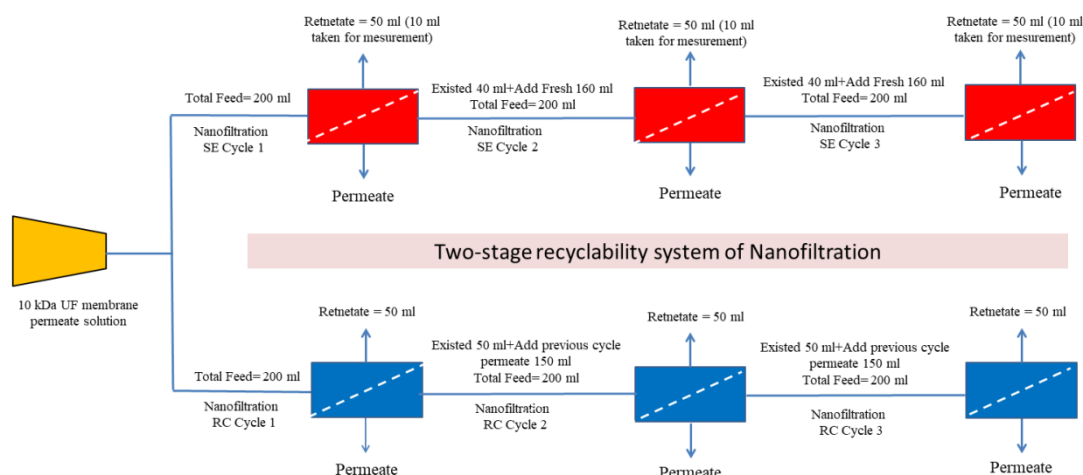
Typically, AD process has been carried to produce methane containing biogas, which is of interest in the renewable bioenergy field (Pramanik et al. 2019; Zhang et al. 2014). However, bio-based volatile fatty acids (VFAs) production in the AD process to be selected as an emerging research field due to their higher market value and large industrial applications compared to biogas (Kleerebezem et al. 2015; Lukitawesa et al. 2020; Strazera et al. 2018; Wainaina et al. 2019b). VFAs are primarily collected in a mixed liquid digested form that needs to be purified (higher concentrations of VFAs) for their promising economic value (Atasoy et al. 2018; Ramos-Suarez et al. 2021).

The utilization of pressure-driven membrane filtration has become common technology for VFAs recovery due to their energy positivity, sustainability and high recovery percentage (Aghapour Aktij et al. 2020; Rongwong et al. 2018; Zhu et al. 2021). Among them, nanofiltration (NF) membranes with 0.2–2 nm and molecular weight cut-off (MWCO) from 200 to 1000 Da appears to have great potential for small organic molecules (<1 kDa) recovery. Chemically, the surface of NF membranes is negatively charged that enhanced the retention efficiency of charged compounds and dissolved ions (Wu et al. 2021; Zacharof et al. 2016; Zhu et al. 2020). However, the recovery percentage of VFAs in the nanofiltration process could be upgraded using the introduction of the recycling system. Although previous studies showed that recycling is an effective way to enhance process efficiency for anaerobic digestion (Romli et al. 1994; Wu et al. 2015; Yin et al.

2021), a recycling system of nanofiltration effluent for VFAs recovery is still relatively limited in the literature.

Therefore, the current study has undertaken an innovative protocol for VFAs recovery in the nanofiltration system. Two-stage batch nanofiltration including sequencing and recirculation treatments, were applied to improve the recovery percentage of VFAs.

## 2. Experimental



**Figure 1. Batch nanofiltration of Sequencing (SE) and Recirculation (RC)**

## 3. Results and discussion

Downstream technologies for the recovery of valuable resources in a waste biorefinery process are expensive in terms of market overview; for this, the implications of recycling strategies for VFAs recovery in the nanofiltration demonstrates an economical approach. The key point of using nanofiltration lies in the interest of achieving a higher concentration of VFAs due to their membrane pore size properties. In this study, TS 40 nanofiltration membrane was chosen because of their tailored molecular cutoff weight (pore size: 200-300 Da) and having affinity to retentate VFAs compounds (Lyu et al. 2016; Zaman et al. 2017).

While the sequencing nanofiltration process was carried out up to 3 cycles, the obtained results are shown in Figure 2 a and c. It was found that the introduction of the sequencing cycle increased the concentration and recovery percentages of VFAs with individual acids, with the addition of fresh effluent feed into existed retentate solution generally increased VFAs amount with the increased number of cycles. It can be noticed that acetic acid was the dominant recovered product up to 2 cycles at a concentration from 1.99 to 9.36 g L<sup>-1</sup> and goes to 11.93 g L<sup>-1</sup> at the 3<sup>rd</sup> cycle. On the other hand, the recovered concentration of butyric acid was low up to 2 cycles (from 1.78 to 9.15 g L<sup>-1</sup>), but at the 3<sup>rd</sup> cycle, the concentration increased to 13.26 g L<sup>-1</sup>. Also, enhanced concentration trends with respect to cycle have been found for caproic acid up to 3 cycles

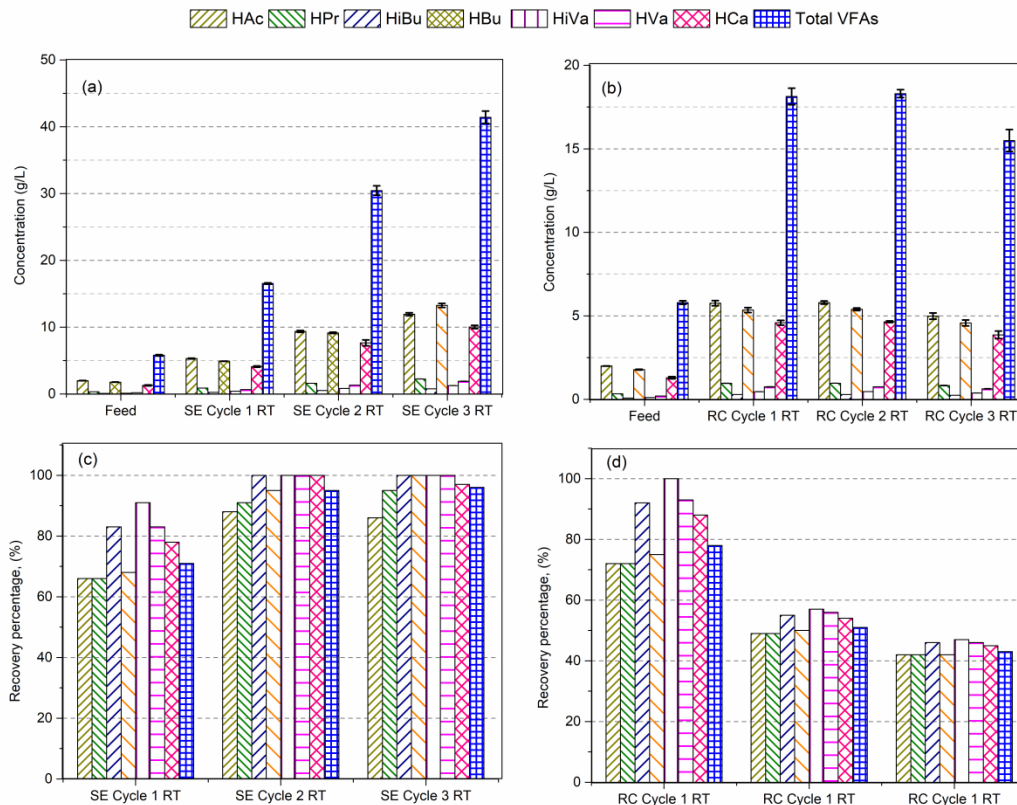
A certain amount of anaerobically digested decanted ultrafiltered effluent (UP010, 10 kDa, PES UF membrane, Microdyn Nadir GmbH, Germany) was used as a feed solution for the nanofiltration process. After that, two-stage batch nanofiltration was conducted using a commercial nanofiltration membrane (TS 40, 200-300 Da, PES, Microdyn Nadir GmbH, Germany) and operated 20-21°C, pH 9 and 15 bar pressure, as shown in Figure 1. The concentration of VFAs was measured using gas chromatography (Clarus 550; Perkin-Elmer, Norwalk, CT, USA).

(from 1.29 to 10.02 g L<sup>-1</sup>). These results could be corroborated with our previous reports, where the production rate of acetic, butyric and caproic was higher in the anaerobic MBR digested microfiltered effluent (Parchami et al. 2020; Wainaina et al. 2020; Wainaina et al. 2019b). Moreover, total VFAs concentration was significantly increased from 5.79 to 41.42 g L<sup>-1</sup> and contributes to more than 96% recovery, which is very promising in terms of recyclable VFAs strategy (Ramos-Suarez et al. 2021; Young et al. 2013).

Building up to the recycling strategy, second stage experiments were followed up by the use of the recirculation method up to 3 cycles, and the results are shown in Figure 2 b and d. Using existing retentate solution with added previous cycle permeate solution, concentrated VFAs with individual acids were calculated and found a decreasing trend for overall efficiency. For the 1<sup>st</sup> and 2<sup>nd</sup> cycle, the concentration of acetic acid increased from 5.75 to 5.79 g L<sup>-1</sup>, but in the 3<sup>rd</sup> cycle, it reduced to 4.98 g L<sup>-1</sup>. And the same results were also obtained for butyric and caproic acid. The recovery percentage indicated that recovery percentages for each dropped almost 40-50 % of the previous cycle. This result may occur due to insufficient adsorption sites on the membrane surface in order for capturing VFAs compounds in the recirculation process (Ahsan et al. 2014; Jiang et al. 2018; Yang et al. 2013), which strongly influences the recovery percentage attributes. In addition, these lower recovery percentages of VFAs are also caused due to membrane structure,

such as our selected membrane TS 40 was polypiperazine amide, which is less stable and more prone to membrane fouling compared to fully aromatic

polyamide-based membranes (Tang et al. 2009; Wadekar and Vidic 2017; Zacharof et al. 2016).



**Figure 2. VFAs concentration and recovery percentages for Sequencing (a,c) and Recirculation (b,d)**

#### 4. Conclusion

In the current study, the effects of recycling anaerobic digested liquid effluent on the recovery efficiency of VFAs were investigated by applying a two-stage recyclability consortium by maintaining a circular economy framework network. The results revealed that the first stage, namely sequencing batch nanofiltration, is favoured for VFAs retention (more than 96%) compared to the recirculation process. However, this study paved the way for the enhancement of VFAs recovery in the nanofiltration process.

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