Journal of Membrane Science and Research 9 (2023) 556692

Journal of Membrane Science & Research

journal homepage: www.msrjournal.com

Research Paper

Properties of Flat Ceramic Membranes and Their Application for Municipal Digestate Liquid Fraction Purification

Urbanowska Agnieszka [1,*], Kabsch-Korbutowicz Małgorzata [1]

[1] *Wroclaw University of Science and Technology, Faculty of Environmental Engineering, Department of Environment Protection Engineering, Wroclaw, Poland*

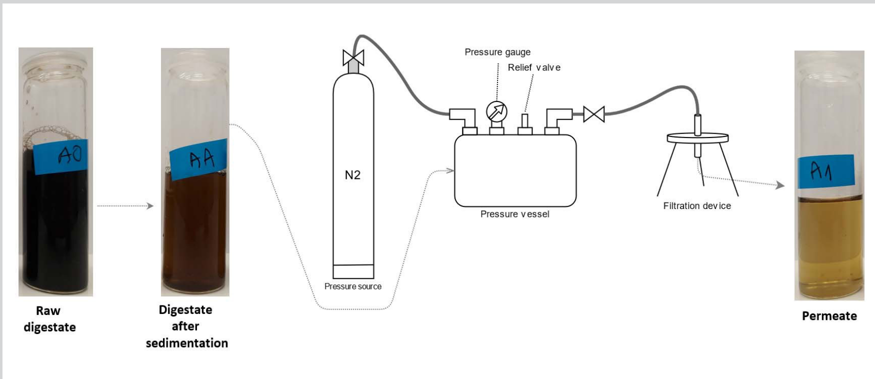
Article info

Received 2022-07-07
Revised 2022-09-14
Accepted 2022-09-29
Available online 2022-09-29

Keywords

Flat ceramic membranes
Digestate
Municipal biogas plant
Microfiltration
Ultrafiltration

Graphical abstract



Highlights

- Flat ceramic membranes can be applied in municipal digestate liquid fraction purification
- An increase in the size of the transmembrane pressure (TMP) results in an increase in the permeate flux
- The separation efficiency of the tested MF and UF membranes was found to depend on the membranes' cut-off
- Neither the increase of TMP, nor the duration of the filtration process had any influence on separation efficiency
- An increase in TMP resulted in a decrease in the permeability of the relative membrane

Abstract

Due to the increasing water scarcity in agriculture, digestate is not only considered as an alternative fertilizer, but also as a potential water source. Unfortunately, it requires treatment to such an extent that the contaminants from the fermented biomass do not return to the environment. The aim of this study was to extend the characteristics of the flat ceramic membranes provided by manufacturers and to evaluate their applicability to the treatment of the municipal digestate liquid fraction. The digestate liquid fraction from a biogas plant processing municipal waste organic fraction located in the Lower Silesia province (Poland) was tested. A Sterlitech laboratory plant operating in the dead-end mode at a TMP of 0.1 - 0.4 MPa was used to carry out the pressure membrane filtration process. The tested digestate was subjected to 72 h of sedimentation before testing. Six micro- and ultrafiltration flat ceramic membranes from Tami Industries were used in the experiments. The membranes used in this study were hydrophilic (a wetting angle of less than 59.6°) and the average pore radius ranged from 0.035 to 0.29 µm, depending on the membrane type. The performed experiments confirmed the applicability of the tested membranes for municipal digestate purification, although a deterioration in permeate quality was observed as the pore size of the membranes increased. The best separation was observed for the 1 kDa membrane, the average pore diameter of which was 35.53 nm.

1. Introduction

Demographic growth, a consumption-based lifestyle, economic and industrial development, and climate change are some of the reasons why many regions of the world are struggling to provide adequate water - not only for municipal and industrial purposes but also for agriculture. This imposes, on present and future generations, a great responsibility for the natural environment and the restoration of the ecological balance. To stop the progressive degradation of the natural environment, the solving of environmental problems that have arisen as a result of industrial activity is becoming a prioritized activity. This need has been recognized through changes in environmental protection strategies towards so-called "clean production". Their implementation creates the necessity to search for innovative solutions that can fulfil certain assumptions, in particular those concerning the reduction

* Corresponding author: agnieszka.urbanowska@pwr.edu.pl (U. Agnieszka)

DOI: 10.22079/JMSR.2022.556692.1549

of pollutant emissions, as well as those with regards to increasing the management efficiency of energy and natural resources, including water [1,2].

Currently, pressure-driven membrane processes are widely used in many branches of industry and environmental protection to remove various pollutants. They make it possible to obtain water with a high degree of purity [3], as well as to remove organic and inorganic contaminants and microorganisms at relatively low costs and with small installation dimensions [4,5]. It is also important that membranes can be used to purify solutions in which the contaminant concentrations fluctuate over a wide range [6]. In addition to their many advantages, membrane processes have the disadvantage of membrane blocking (fouling). Fouling is a result of the deposition of organic and inorganic compounds and microbiological contaminants on the membrane's surface and inside its pores. This results in both a decrease in the permeate flow rate and a decrease in purification efficiency [7]. One of the main factors determining the separation efficiency of a membrane process is the used membrane type. In membrane separation techniques, only synthetic membranes are used. They are characterized by different functions and structures in comparison with natural membranes (cell membrane, plasma). Both polymeric and ceramic materials are used to make membranes [8]. Polymeric membranes can have different structures, depending on both the polymer used (e.g., polyethersulfone, polysulfone, polyamide, cellulosic materials) and the manufacturing method itself. The main advantages of polymeric membranes include the ease of processing the material, their relatively low price, and the wide variety of properties [4,5]. Most polymeric materials have certain limitations related to their tolerance to pH, hydrolysis, and oxidation. The hydrophobicity of some polymers is also a disadvantage. These properties mean that polymeric membranes are not resistant to fouling, which results in reduced membrane performance and selectivity [9]. The choice of material and membrane manufacturing method depends on the intended use of the membrane, as well as on the conditions under which they are to be operated (mainly: temperature, pH, and presence of substances that have a negative influence on the membrane's surface, etc.). Ceramic membranes can be an alternative to polymeric membranes, and are characterized by high chemical, mechanical, biological, and thermal resistance; steam stabilizability; longevity; and the possibility of using spent membranes as ceramic material in other applications [10]. Their usage enables raw material and energy savings [11]. A ceramic membrane is structurally asymmetric [12], and consists of a macroporous support and a thin surface layer that determines the membrane's separation properties. Commercially available ceramic membranes have a pore size ranging from 0.005 to 1 µm. Typically, the support layer is 1 - 3 mm thick, while the skin layer is a few µm thick (ultrafiltration membranes) and usually formed from ZrO_2, Al_2O_3, TiO_2, or CeO_2 [10,13]. One pressure-driven membrane process application is liquid digestate fraction purification, which is a by-product (waste) generated in biogas plants during the process of anaerobic methane fermentation. It mainly contains undigested organic matter residues and mineral components in amounts comparable to their content in the substrates used in the biogas plant. It is estimated that the amount of digestate produced is 85-95% of the weight of the substrates used (the more fermentable organic substances in the feedstock, the smaller the amount of digestate) [14]. The process of biogas production includes a reduction in the organic matter content, an increase in the mineral content with regard to dry matter, a fragmentation of solids, complete or partial hygienization, and the decomposition of odor-forming compounds [15]. The digestate includes all the compounds that did not undergo the fermentation process, and thus contains all the impurities contained in the substrate. Therefore, good (or poor) quality substrate subjected to the fermentation process will lead to good (or poor) quality digestate [16].

In agricultural biogas plants, animal excreta (pig or cattle manure), agri-food waste (fruit and vegetable waste, food, slaughterhouse waste), and lignocellulosic biomass can be distinguished among the main input components. Municipal biogas plants, on the other hand, use a selected biodegradable fraction of municipal waste and combine a stream from households (called kitchen waste) and urban greenery (called green waste). A municipal biogas plant based on the organic fraction is an installation similar to an agricultural biogas plant but is expanded with additional technological modules related to the pre-treatment of waste. The two types of biogas plants also differ with regards to location - municipal biogas plants are built in or adjacent to urbanized areas, where there is better infrastructure for obtaining high-energy waste, while agricultural biogas plants are located in rural areas that are abundant in suitable raw materials. One of the rational ways to manage digestate is to recover water and nutrients, which can then be used in agriculture. However, in order for the recovered water to be used to irrigate crops, it must be treated so that any contaminants from the fermented biomass do not return to the environment. Both conventional physico-chemical processes and membrane processes can be useful in this area. While the issue of treating the liquid fraction of digestate from agricultural biogas plants is present in the literature [3,17–21], there are only a few reports on the treatment of digestate liquid from municipal waste biogas plants. Since the two types of digestate are fundamentally different, research related to municipal digestate, regardless of the results obtained for agricultural digestate, should be conducted. Moreover, digestate liquid fraction purification using polymeric membranes has already been reported [17,22], but to our knowledge, there are no studies on the application of flat ceramic membranes in this area.

Another of our papers [23] focused on the effectiveness of the treatment of the digestate liquid fraction from municipal waste biogas plants using polymeric membranes, but the purification efficiency turned out to be unsatisfactory. Considering the advantages of inorganic membranes, it can be seen to be important to conduct research on the possibility of purifying municipal digestate liquid using flat ceramic membranes. Moreover, it is also justified to extend the characteristics (provided by manufacturers) concerning the membranes.

2. Materials

The ceramic flat micro- (Ceram 0.45 µm, Ceram 0.14 µm) and ultrafiltration (Ceram 1 kDa, Ceram 5 kDa, Ceram 15 kDa, and Ceram 50 kDa) membranes from Tami Industries were used in this study. Their detailed characteristics are included in our earlier publication [24]. A sample SEM image of the Ceram 5 kDa membrane can be seen in Fig. 1.

The digestate liquid fraction from a municipal waste biogas plant located in the Lower Silesia province of Poland (50°53'15.5 "N 17°23'28.0 "E) was tested. This fraction was separated from the digestion pulp with the use of sedimentation centrifuges. The characteristics of the test solution are presented in Table 1. As can be seen, the digest is characterized by high salinity and a high content of organic compounds (COD, DOC), including biodegradable ones (BOD5). It also contains substantial amounts of microorganisms. A microscopic image of the liquid fraction of the digestate with, among others, visible Cercomonas sp. flagellates is shown in Fig. 2.

3. Experimental methods

To perform the pressure membrane filtration process, a Sterlitech laboratory installation with a 316 SS pressure vessel (3.8 dm^3) was used [24]. This system is designed to work with flat ceramic membranes. The process was conducted in the dead-end mode at TMPs ranging from 0.1 - 0.4 MPa. The tested digestate was subjected to 72 h of sedimentation prior to testing. All separation experiments were duplicated. All the ceramic flat membranes were subjected to a conditioning procedure before being used in this study in order to prepare them for proper operation. This included alkaline cleaning by placing the membranes in NaOH solution (15-20 g/dm^3) at 80°C for 30 min, rinsing with redistilled water until the pH was neutral, and then acid cleaning and rinsing again until the pH was neutral. The MF and UF membranes were cleaned with 0.39 mol/dm^3 HNO_3 or 0.038 mol/dm^3 H_3PO_4 at 50°C for 15 min, while the fine UF membranes were cleaned with a 0.0076 mol/dm^3 H_3PO_4 at 50°C for 15 min (as recommended by the manufacturer).

After each process, the membranes were chemically cleaned with 0.1 mol/dm^3 NaOH solution (Avantor Performance Materials Poland S.A., Gliwice, Poland) and then rinsed with redistilled water until the initial permeate flux values were obtained. To extend the characteristics of the tested membranes, their wettability was determined. Wettability was measured as the wetting angle using the ASTM D5946 method and consisted of placing a 5 µl drop (approximately) of Milli-Q redistilled water (Millipore, USA) on the membrane's surface using a Hamilton microsyringe while maintaining the same minimum needle height above the membrane's surface and the bevel direction of the needle tip. These measurements were performed using a PGX 50372 device (Fibro System AB, Sweden). A minimum of 7 drops were deposited on each membrane, each time on a "fresh" surface.

To determine the pore size of the membranes, standard porometric tests were performed for all the flat ceramic membranes using a POROLUX™1000 porometer (Belgium) based on the step/pressure stabilization method. The POROLUX™1000 porometer is shown in Fig. 3. The first bubble point (FBP) pore diameter measurement was performed using the step/stability method. It allows measuring the actual FBP. The procedure for measuring the FBP is as follows: the pressure in the chamber increases linearly by delivering a constant flow of gas before the largest pore opens, and then, as the gas stream passes through the sample (through the largest pores), the pressure growth decreases. The FBP is measured at the pressure at which it occurs. The associated tests were conducted at a flow rate of 30 cm^3/min and a deviation of 30%. Calculation of this point for the pressure at which the first continuous gas bubbles are detected is defined by the ASTM F-316-03 standard. Porefil fluid (γ = 16.1 mN/m) was used as the wetting liquid. The pressure rise gradient was 30 s/bar.

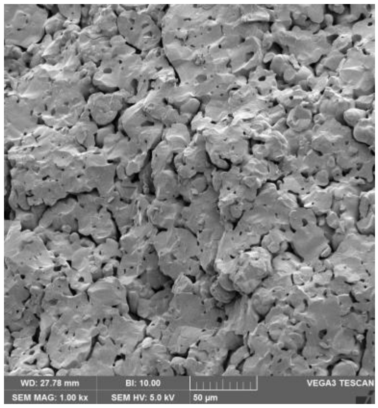


Fig. 1. SEM image (1000x) of the Ceram 5 kDa ceramic membrane

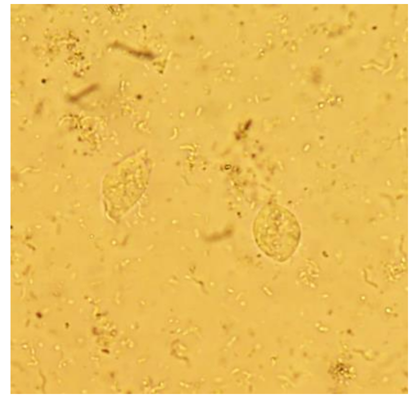


Fig. 2. Microscopic image of the digestate liquid fraction (magnitude 600x)

Table 1
The properties of the liquid digestate fraction from the municipal waste biogas plant

Index	Value
pH	7.2
Conductivity, mS/cm	22
Chemical oxygen demand (COD), mg O_2/dm^3	6,190
Biochemical oxygen demand (BOD_5), mg O_2/dm^3	2,170
Dissolved organic carbon (DOC), mg C/dm^3	3,050
N-NH_4^+, mg N/dm^3	1,742
N-NO_2^-, mg N/dm^3	6.25
N-NO_3^-, mg N/dm^3	below the limit of detection
PO_4^{3-}, mg/dm^3	18.9
SO_4^{2-}, mg/dm^3	38
P, mg/dm^3	15.9
Na, mg/dm^3	1,650
K, mg/dm^3	1,560
Ca, mg/dm^3	421
Mg, mg/dm^3	230
Cl^-, mg/dm^3	2,246
Fe, mg/dm^3	2.5
Mn, mg/dm^3	4.0
Cu, mg/dm^3	0.096
Zn, mg/dm^3	0.630
Li, mg/dm^3	6
Hg, mg/dm^3	0.0036
Co, mg/dm^3	0.137
Ni, mg/dm^3	0.250
Ba, mg/dm^3	0.240
As, mg/dm^3	0.0005
Cr, mg/dm^3	0.230
Pb, mg/dm^3	0.025
Cd, mg/dm^3	0.001
Mesophilic bacteria, CFU/cm^3	$111 \cdot 10^6$
Thermophilic bacteria, CFU/cm^3	$163 \cdot 10^2$

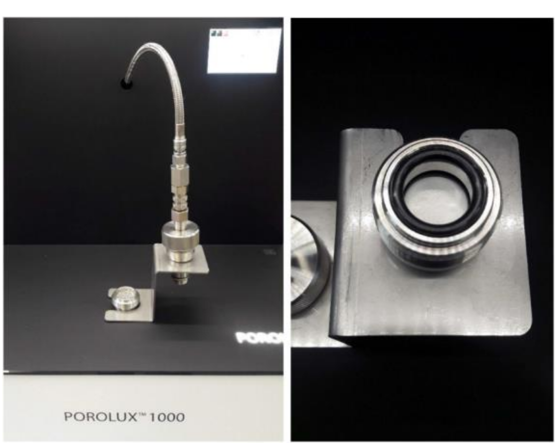


Fig. 3. The POROLUX^{TM}1000 porometer with a sample holder used in this study

The preparation procedure for the porometric tests involved cutting the samples to an external diameter of 25 mm. The samples were then soaked in Porefil for 10 min and placed under a vacuum to ensure complete impregnation. Due to the thickness of the membranes, they were placed in a special sample holder that is used for materials of significant thickness. The samples were supported by a standard support mesh.

4. Results and discussion

4.1. Properties of the flat ceramic membranes

To gain a deeper understanding of the differences in the transport and separation properties of the tested ceramic membranes, their characteristics, which were provided by the manufacturers [24], were extended by the authors' wetting angle measurements. Based on the wetting angle value, it is possible to determine the surface character of the membranes: the higher the value, the less susceptible the membrane is to wetting. For all the tested ceramic membranes, the wetting angles were 59.6° or below, which indicates the hydrophilic character of these membranes (Table 2). In general, as the pore diameter of the tested membranes increased, a decrease in the wetting angle value was observed, which indicates increasing hydrophilicity. A deviation from this trend was found for the microfiltration membranes, where, despite significant differences in the diameters of the pores (Table 3), the wetting angle values were similar.

Membrane properties (i.e., pore size, hydrophobicity, permeability, and charge) significantly affect their separation ability. To compare the effectiveness of the flat ceramic membranes to purify the liquid fraction of the digestate, the pore sizes of the membranes were determined. The results of the measurements are presented in Table 3 and Fig. 4, while pictures of the membranes after the measurements with the POROLUX^{TM}1000 porometer are shown in Fig. 5.

Table 2
Wetting angle measurement results of the flat ceramic membranes

Membrane type	Wetting angle, °
Ceram 1 kDa	59.6
Ceram 5 kDa	57.6
Ceram 15 kDa	43.8
Ceram 50 kDa	42.4
Ceram 0.14 µm	36.6
Ceram 0.45 µm	36.7

Table 3
Pore size measurement results of the flat ceramic membranes

Membrane type	Maximum pore size (FBP), nm		Average pore size, nm		Smallest pore size, nm	
	value	standard deviation	value	standard deviation	value	standard deviation
Ceram 1 kDa	393.8	198.5	35.53	1.38	24.03	0.93
Ceram 5 kDa	162.8	50.9	37.69	0.88	21.96	0.22
Ceram 15 kDa	307.7	186.7	52.45	8.06	38.52	2.83
Ceram 50 kDa	381.9	120.5	67.09	2.00	36.22	1.10
Ceram 0.14 µm	160	20	122	1	95	9
Ceram 0.45 µm	820	50	290	30	150	20

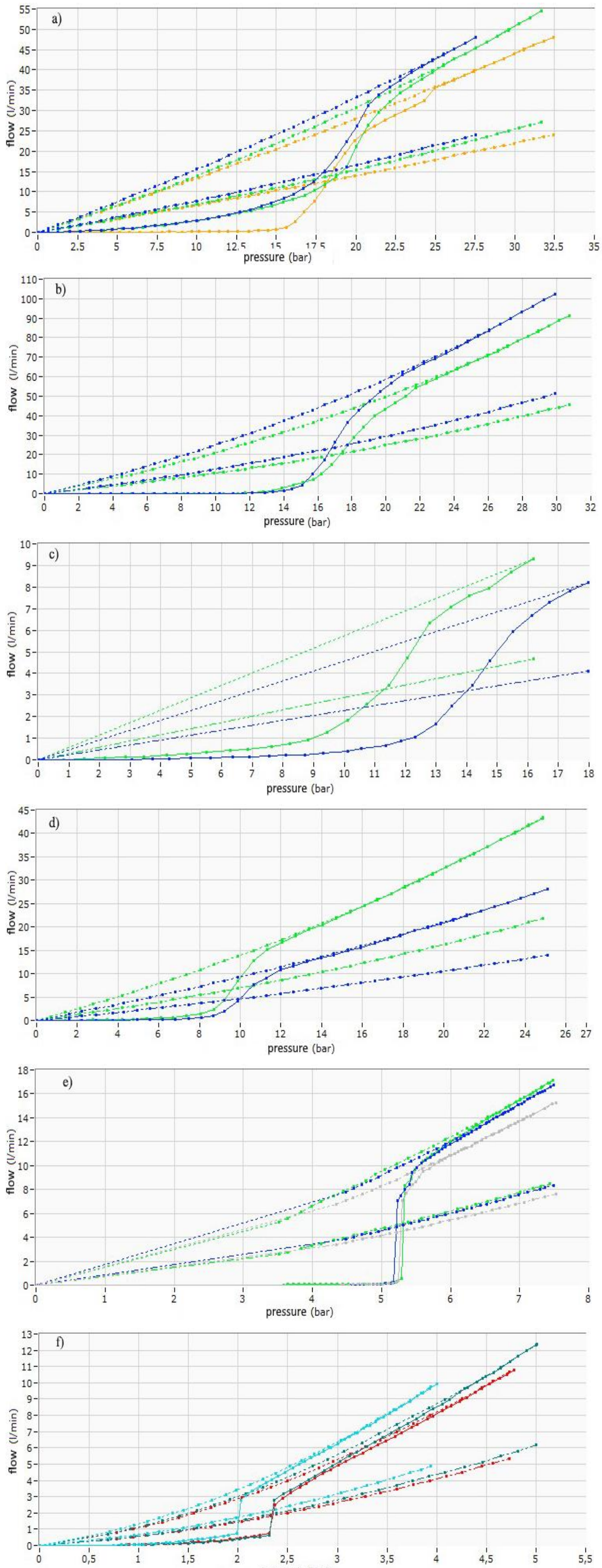


Fig. 4. Gas flow during the porometric measurements for the flat ceramic membrane samples: a) Ceram 1 kDa, b) Ceram 5 kDa, c) Ceram 15 kDa, d) Ceram 50 kDa, e) Ceram 0.14 µm, and f) Ceram 0.45 µm (continuous graphs - wet curve, dashed graphs - dry curve and half of the dry curve to determine the average pore size)

The procedure used to determine the pore size uses Capillary Flow Porometry (CFP), which involves pushing an inert gas through a wetted membrane at increasing pressure [25]. This causes the liquid to be displaced from successive pores in the membrane (starting with the pores with the largest diameters, and ending with the smallest pores). The observed effect is an increase in the flux of gas through the membrane. As can be seen in Fig. 4, as the membrane's pore diameter increases, the pressure at which gas flows through the sample decreases. The maximum pore size (FBP) is determined when gas flow through the sample is observed. The simplified Young-Laplace formula ($P = 4 \cdot \gamma / D$, where D is the pore size diameter, P is the measured pressure, and γ is the surface tension of the wetting liquid) enables the pore diameter to be calculated.

The measurements showed that the average pore diameter of the tested membranes ranged from 35.53 to 67.09 nm for the ultrafiltration membranes, and from 122 to 290 nm for the microfiltration membranes, while the diameter of the maximum pores was in the 162.8 – 393.8 and 160 – 820 nm ranges, respectively. Analysis of the variation in the average diameter of the studied membranes shows an evident trend of change: an increase in the nominal cut-off value results in a larger average pore diameter. It is difficult to explain why such a trend is not observed when analyzing changes in the maximum pore diameter.

The POROLUX™1000 porometer results obtained for most of the membranes (Ceram 1 kDa, Ceram 5 kDa, Ceram 50 kDa, Ceram 0.14 µm and Ceram 0.45 µm - Fig. 4 a, b, d, e, f) show good agreement in the terms of average pore size and flow. Only the results obtained for the Ceram 15 kDa membrane (Fig. 4 c) show a slight spread. Furthermore, for the Ceram 1 kDa membrane, a spread in pore size was observed in the FBP. For the test shown as the orange curve in Fig. 4 a), a cracking sound of the sample was heard, and a small flow spike was observed. Higher FBP values may indicate the presence of a small number of larger voids (pre-FBP) or may be related to small defects that led to the sample fracturing during the measurements. It was similar for the Ceram 50 kDa membrane (Fig. 4 d), where a pore size spread in FBP was also observed. For ultrafiltration membranes Ceram 5 kDa and Ceram 50 kDa and microfiltration membranes Ceram 0.14 µm and Ceram 0.45 µm, the samples did not break during the measurement (Fig. 5 b, d, e, f). In contrast, all the UF membrane samples with 1 and 15 kDa cut-offs broke into two or more pieces at 24 bar and 16-18 bar, respectively (Fig. 5 a, c).

To analyze the transport properties, and subsequently, their susceptibility to fouling, the permeate flux for redistilled water was determined. Fig. 6 shows the effect of TMP on the ceramic membrane permeate flux volume. An increase in the TMP resulted in a linear increase in the permeate flux (for all the tested membranes and in the analyzed TMP range). An increase in the cut-off value of the membranes, and thus an increase in the membrane's pore radius, increased their hydraulic efficiency. The highest permeate flux (5.9 $m^3/m^2 \cdot d$) was achieved for the Ceram 0.45 µm MF membrane, while the lowest (0.35 $m^3/m^2 \cdot d$) was measured for the Ceram 1 kDa UF membrane (at the highest tested pressure of 0.4 MPa).

4.2. Purification of the municipal digestate liquid fraction using flat ceramic membranes

The effectiveness of the digestate liquid fraction treatment in the MF and UF processes using flat ceramic membranes was determined by analyzing the influence of the membrane's cut-off and process parameters (TMP, process duration) on the COD, BOD_5, and DOC value changes. All samples of the digestate liquid fraction were subjected to 72 hours of sedimentation prior to membrane filtration.

The digestate purification (using sedimentation) results for the flat ceramic membrane filtration (Fig. 7) showed that the tested membranes could be used for digestate purification. However, a deterioration in the quality of the digestate could be observed as the membrane's pore size increased. The sieve mechanism mainly separated organic substances, [26] and therefore the separation efficiency was significantly affected by the ratio between the contaminant's particle size and the membrane's pore diameter, which was e.g., about 35.5 nm for the Ceram 1 kDa membrane, about 37.7 nm for the Ceram 5 kDa membrane, and about 67.1 nm for the Ceram 50 kDa membrane (Table 3). The highest organic compound concentration was obtained in the permeate for the MF membranes (pore diameters of 0.12 and 0.29 µm, respectively for the Ceram 0.14 µm and Ceram 0.45 µm membranes). It was observed that the use of denser membranes resulted in improved digestate purification efficiency. The smaller the cut-off value of the membrane, the fewer organic particles could pass into the permeate, as was also reported by other researchers [27]. The best separation efficiency was observed when the Ceram 1 kDa membrane was used. For example, at a TMP of 0.2 MPa, the concentration of the biodegradable fraction of organic substances, expressed as BOD_5, was reduced from 2 170 to 930 g O_2/m^3, while COD and DOC were reduced from 6 190 to 3 440 g O_2/m^3 and 3 050 to 1 330 g C/m^3, respectively (Figs. 7 and 8).

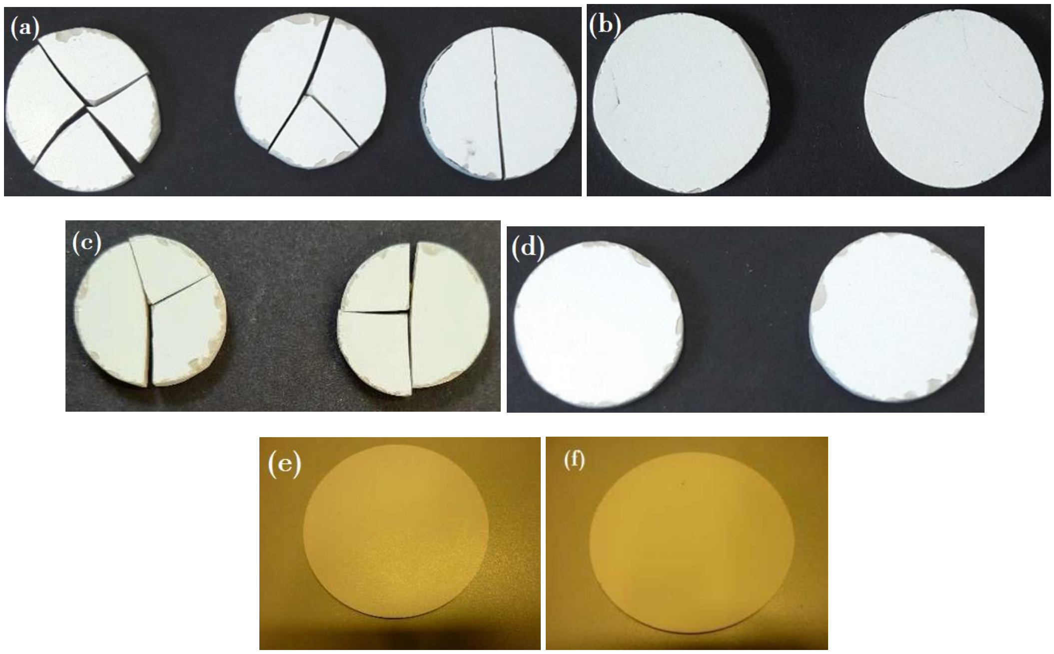


Fig. 5. Photographs of samples of the ceramic membranes after measurements with the POROLUX^{TM}1000 porometer: a) Ceram 1 kDa, b) Ceram 5 kDa, c) Ceram 15 kDa, d) Ceram 50 kDa, e) Ceram 0.14 µm, and f) Ceram 0.45 µm

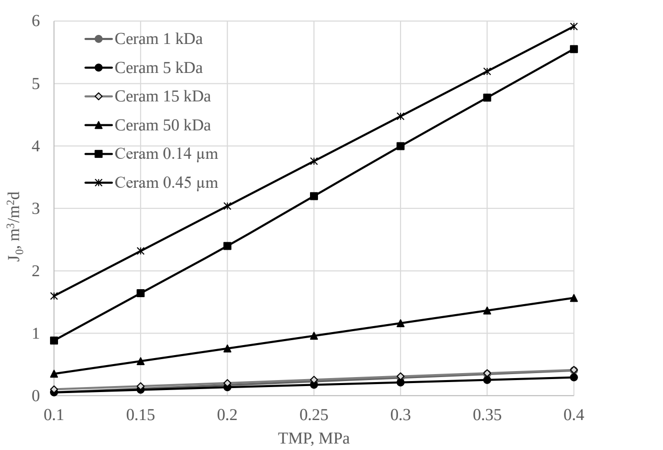


Fig. 6. Effect of TMP on the permeate flux of the ceramic membranes for redistilled water

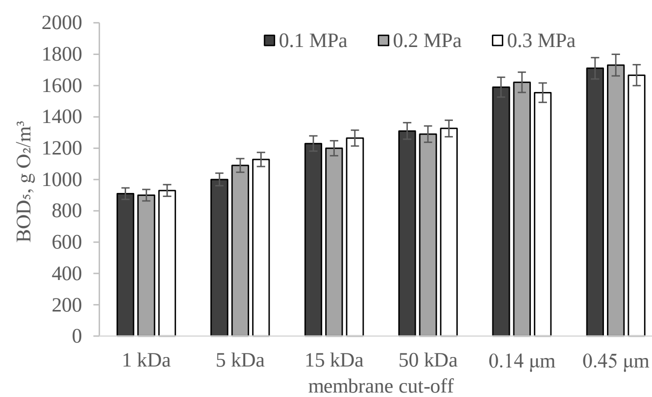


Fig. 7. The effect of TMP on the BOD_5 of permeate from the digestate liquid fraction sedimentation – flat ceramic membrane filtration process

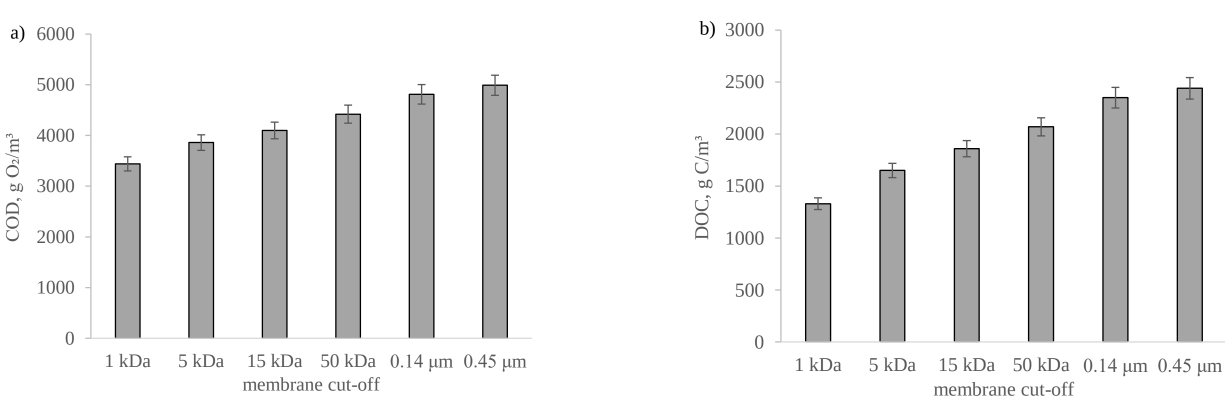


Fig. 8. The effect of the membrane type on the COD (a) and DOC (b) changes for the liquid fraction of digestate sedimentation – flat ceramic membrane filtration (TMP 0.2 MPa)

When comparing the removal efficiency of organic compounds from the liquid fraction of the digestate with a previous study [28], it was found to be higher for ceramic membranes than for polymer membranes with a comparable cut-off value. Although the COD, BOD_5 and DOC values obtained exceed local regulations set for water used in agriculture [29,30] and more so for drinking water, such a high degree of organic compound removal means that the analyzed solution is well prepared for the next treatment stage, or also that it can be diluted with water from another source before being used to irrigate plants. It was also observed that the value of the transmembrane pressure did not affect the quality of the permeate, with the content of organic compounds remaining at a comparable level in the tested TMP range (0.1 - 0.3 MPa) (Fig. 7).

It was also verified whether the duration of the membrane filtration process influences the effectiveness of eliminating organic pollutants from the treated solution (Fig. 9). These studies have shown that separation efficiency is practically unchanged over time. Longer filtration did not affect the membrane's separation properties - the values of BOD_5 in the permeate remained at a constant level. Comparable results for COD and DOC were shown in [24]. The constant efficiency of separating organic substances during membrane filtration also indicates that it was the sieve mechanism that determined the separation, and not, for example, sorption on the membrane, the effect of which was observed for some membranes at the initial time of membrane filtration.

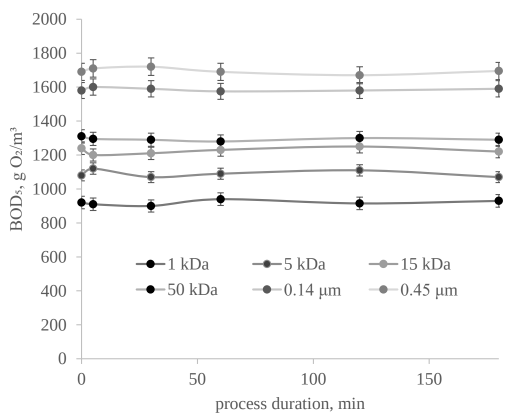


Fig. 9. Influence of the duration of the membrane filtration on the BOD_5 of permeate during the digestate liquid fraction treatment (Δp = 0.2 MPa)

From the point of view of the liquid fraction of digestate purification using membranes, attention should not only be paid to separation but also to transport properties. The tested membranes not only differed in terms of absolute hydraulic efficiency (Fig. 10), which resulted mainly from the differences in pore diameters, but also from their susceptibility to fouling. Fig. 11 shows the effect of the TMP on the membrane's relative permeability and enables the fouling intensity to be investigated. The analysis of the obtained test results shows that for all the membranes, an increase in the TMP value resulted in a decrease in the J/J_0. This effect was most evident when using the Ceram 5 kDa and Ceram 15 kDa membranes. Moreover, it was found that an increase in the membrane's cut-off, and thus an increase in the membrane's pore diameter, caused a decrease in the membrane's relative permeability (except for the Ceram 1 kDa membrane). It was most visible for MF membranes (Ceram 0.14 and Ceram 0.45 µm) and practically regardless of the pressure applied, J/J_0 was approx. 0.01. These membranes were the most susceptible to fouling. This is confirmed by some reports in the literature, according to which membranes with larger pore diameters (in this case the MF membranes) can be more susceptible to fouling than in the case of more compact membranes, e.g., UF membranes [31,32]. Fouling resulting from the blocking of the pores of the membrane by particles from the feed penetrating them dominates in the case of MF membranes. This is also confirmed by the results of our research regarding the average pore size of membranes (Table 3). A photograph of the formed filtration cake on the Ceram 0.45 µm flat ceramic membrane is shown in Fig. 12.

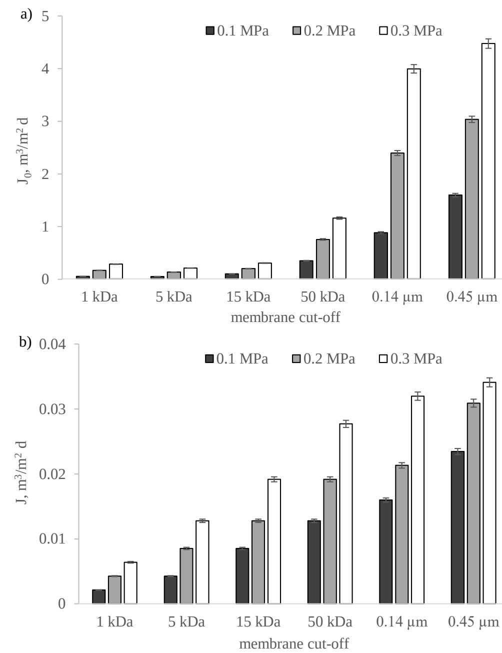


Fig. 10. The effect of TMP on the permeate flux of the flat ceramic membranes for (a) redistilled water and (b) the digestate liquid fraction

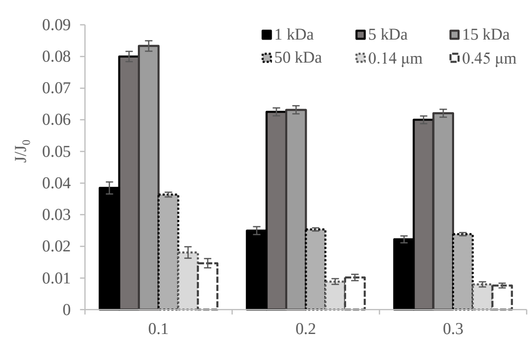


Fig. 11. The influence of TMP on the flat ceramic membrane's relative permeability during the treatment of the digestate liquid fraction

Fig. 12. The new Ceram 0.45 µm ceramic membrane (a) and the filtration cake formed on its surface during the treatment of the digestate liquid fraction (b).

5. Conclusions

The results of this study showed that flat ceramic membranes can be applied in the purification of the municipal digestate liquid fraction. The tests and analyses showed that the flat ceramic micro- and ultrafiltration membranes used in this study were hydrophilic (a wetting angle of less than 59.6°) and that the average pore diameter ranged from 0.035 to 0.29 µm, depending on the membrane type. It was shown that an increase in the size of the transmembrane pressure (TMP) results in an increase in the permeate flux during both redistilled water and liquid fraction digestate membrane filtration. The separation efficiency of the tested MF and UF flat ceramic membranes was found to depend on the membrane type (cut-off). The best separation was observed for the Ceram 1 kDa membrane. It was also found that neither the increase of TMP nor the duration of the filtration process had any influence on the separation efficiency. The tested membranes differed not only in hydraulic efficiency, due to differences in pore diameters, but also in their susceptibility to fouling. It was observed that an increase in TMP resulted in a decrease in the J/J_0 value (which means a higher intensity of fouling). For the tested membranes (except for the Ceram1 kDa membrane), an increase in the membrane cut-off value, and thus an increase in their pore size, increased the fouling intensity.

Acknowledgments

The research was conducted as part of the interdisciplinary project Sustainable technology for the staged recovery of agricultural water from high moisture fermentation products RECOWATDIG carried out under the Water Joint Programming Initiative co-financed by 20 Funding Partner Organisations from 18 countries and with the support of the European Commission (full list: http://www.waterjpi.eu/images/documents/JC2018/water jpi 2018 joint_call_participating_countries.pdf).

References

[1] European Commission Roadmap to a Resource Efficient Europe; 2011; Vol. 147

[2] EC Europe 2020 - A Strategy for Smart, Sustainable and Inclusive Growth; 2010; Vol. COM (2010);

[3] T. Gienau, U. Brüß, M. Kraume, S. Rosenberger, Nutrient Recovery from Biogas Digestate by Optimised Membrane Treatment. Waste Biomass Valori. 9 (2018) 2337–2347. https://doi.org/10.1007/s12649-018-0231-z.

[4] T. Iwashita, H. Katayanagi, N. Miki, Needle-Type in Situ Water Content Sensor with Polyethersulfone Polymer Membrane, Sens. Actuators B Chem. 154 (2011) 41–45. https://doi.org/10.1016/j.snb.2009.11.019.

[5] R. Rodil, S. Schrader, M. Moeder, Non-Porous Membrane-Assisted Liquid-Liquid Extraction of UV Filter Compounds from Water Samples, J. Chromatogr. A 1216 (2009) 4887–4894. https://doi.org/10.1016/j.chroma.2009.04.042.

[6] M. Peter-Varbanets, C. Zurbrügg, C. Swartz, W. Pronk, Decentralized Systems for Potable Water and the Potential of Membrane Technology, Water Res. 43 (2009) 245–265. https://doi.org/10.1016/j.watres.2008.10.030.

[7] J. Wiszniowski, D. Robert, J. Surmacz-Gorska, K. Miksch, J. Weber, Landfill Leachate Treatment Methods: A Review. Environ. Chem. Lett. 4 (2006) 51–61. https://doi.org/10.1007/s10311-005-0016-z.

[8] R.W. Baker, Membrane Technology and Applications; John Wiley & Sons, Ltd: Chichester, UK, 2012.

[9] D. M. Warsinger, S. Chakraborty, E. W. Tow, M. H. Plumlee, C. Bellona, S. Loutatidou, L. Karimi, A. M. Mikelonis, A. Achilli, A. Ghassemi, et al., Review of Polymeric Membranes and Processes for Potable Water Reuse. Prog. Polym. Sci. 81 (2018) 209–237. https://doi.org/10.1016/j.progpolymsci.2018.01.004.

[10] Z. He, Z. Lyu, Q. Gu, L. Zhang, J. Wang, Ceramic-Based Membranes for Water and Wastewater Treatment. Colloids Surf. A: Physicochem. Eng. Asp. 578 (2019) 123513. https://doi.org/10.1016/j.colsurfa.2019.05.074.

[11] R. Sondhi, R. Bhave, G. Jung, Applications and Benefits of Ceramic Membranes. Membr. Technol. 2003 (2003) 5–8. https://doi.org/10.1016/S0958-2118(03)11016-6.

[12] S. K. Amin, M. Hassan, H. Abdallah, An Overview of Production and Development of Ceramic Membranes Solid Waste Management and Pollution Prevention View Project 1st International Conference for Membrane Technology & Its Applications View Project. Int. J. Appl. Eng. Res. 11 (2016) 7708–7721.

[13] D. Zou, Y. Fan, State-of-the-Art Developments in Fabricating Ceramic Membranes with Low Energy Consumption. Ceram. Int. 47 (2021) 14966–14987. https://doi.org/10.1016/j.ceramint.2021.02.195.

[14] I. A. Łucka, A. U. Kołodziej, M. Szymańska, K. Pilarski, Agricultural use of digestate from agricultural biogas plants, in: Use of biomass in the energy industry - economic and ecological aspects (in Polish). The Polish Economic Society, Koszalin, Poland (2011) 277–304.

[15] P. Merlin Christy, L. R. Gopinath, D. Divya, A Review on Anaerobic Decomposition and Enhancement of Biogas Production through Enzymes and Microorganisms. Renew. Sust. Energ. Rev. 34 (2014) 167–173. https://doi.org/10.1016/j.rser.2014.03.010.

[16] M. Koszel, E. Lorencowicz, Agricultural Use of Biogas Digestate as a Replacement Fertilizers. Agric. Agric. Sci. Procedia 7 (2015) 119–124. https://doi.org/10.1016/j.aaspro.2015.12.004.

[17] E. Monfet, G. Aubry, A. A. Ramirez, Nutrient Removal and Recovery from Digestate: A Review of the Technology. Biofuels 9 (2018) 247–262. https://doi.org/10.1080/17597269.2017.1336348.

[18] P. Świątczak, A. Cydzik-Kwiatkowska, M. Zielińska, Treatment of Liquid Phase of Digestate from Agricultural Biogas Plant in a System with Aerobic Granules and Ultrafiltration. Water 11 (2019) 104. https://doi.org/10.3390/w11010104.

[19] D. Đurđević, I. Hulenić, Anaerobic Digestate Treatment Selection Model for Biogas Plant Costs and Emissions Reduction. Processes 8 (2020) 142. https://doi.org/10.3390/pr8020142.

[20] M. Szymańska, E. Szara, T. Sosulski, A. Wąs, G. W. P. van Pruissen, R. L. Cornelissen, M. Borowik, M. Konkol, A Bio-Refinery Concept for N and P Recovery—A Chance for Biogas Plant Development. Energies 12 (2019) 155. https://doi.org/10.3390/en12010155.

[21] P. Świątczak, A. Cydzik-Kwiatkowska, M. Zielińska, Treatment of the Liquid Phase of Digestate from a Biogas Plant for Water Reuse. Biores. Technol. 276 (2019) 226–235. https://doi.org/10.1016/j.biortech.2018.12.077.

[22] C. Yue, H. Dong, Y. Chen, B. Shang, Y. Wang, S. Wang, Z. Zhu, Direct Purification of Digestate Using Ultrafiltration Membranes: Influence of Pore Size on Filtration Behavior and Fouling Characteristics. Membranes 11 (2021) 179. https://doi.org/10.3390/membranes11030179.

[23] A. Urbanowska, M. Kabsch-Korbutowicz, M. Wnukowski, P. Seruga, M. Baranowski, H. Pawlak-Kruczek, M. Serafin-Tkaczuk, K. Krochmalny, L. Niedzwiecki, Treatment of Liquid By-Products of Hydrothermal Carbonization (HTC) of Agricultural Digestate Using Membrane Separation. Energies 13 (2020) 262. https://doi.org/10.3390/en13010262.

[24] A. Urbanowska, M. Kabsch-Korbutowicz, The Use of Flat Ceramic Membranes for Purification of the Liquid Fraction of the Digestate from Municipal Waste Biogas Plants. Energies 14 (2021) 3947. https://doi.org/10.3390/en14133947.

[25] M. Halich, E. Vogt, C. Müller, A. Cano-Odena, D. Pattyn, P. Hellebaut, K. van der Kamp: Capillary flow porometry - assessment of an alternative method for the determination of flow relevant parameters of porous rocks, Annual Symposium of the Society of Core Analysts, Napa Valley, USA. September 2013.

[26] D. M. Kanani, W. H. Fissell, S. Roy, A. Dubnisheva, A. Fleischman, A. L. Zydney, Permeability–Selectivity Analysis for Ultrafiltration: Effect of Pore Geometry. J. Membr. Sci. 349 (2010) 405–410. https://doi.org/10.1016/j.memsci.2009.12.003.

[27] P. Ghosh, R. C. Pradhan, S. Mishra, Clarification of Jamun Juice by Centrifugation and Microfiltration: Analysis of Quality Parameters, Operating Conditions, and Resistance. J. Food Process Eng 41 (2018) e12603. https://doi.org/10.1111/jfpe.12603.

[28] A. Urbanowska, I. Polowczyk, M. Kabsch-Korbutowicz, Treatment of Municipal Waste Biogas Plant Digestate Using Physico-Chemical and Membrane Processes. Desalin. Water Treat. 214 (2021) 214–223. https://doi.org/10.5004/dwt.2021.26661.

[29] The Ordinance of the Polish Minister for Maritime Affairs and Inland Navigation of 12 July 2019 on Substances Particularly Harmful to the Aquatic Environment and the Conditions to be Fulfilled when Discharging Wastewater into Waters or the Earth and when Discharging Rainwater or Meltwater into Waters or Watercourses (in Polish)

[30] Council Directive 91/271/EEC of 21 May 1991 concerning urban waste-water treatment

[31] T. Miyoshi, K. Yuasa, T. Ishigami, S. Rajabzadeh, E. Kamio, Y. Ohmukai, D. Saeki, J. Ni, H. Matsuyama, Effect of Membrane Polymeric Materials on Relationship between Surface Pore Size and Membrane Fouling in Membrane Bioreactors. Appl. Surf. Sci. 330 (2015) 351–357. https://doi.org/10.1016/j.apsusc.2015.01.018.

[32] L. Jin, S. L. Ong, H. Y. Ng, Comparison of Fouling Characteristics in Different Pore-Sized Submerged Ceramic Membrane Bioreactors. Water Res. 44 (2010) 5907–5918. https://doi.org/10.1016/j.watres.2010.07.014.