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Low-cost ceramic membranes manufacture using INKJET technology for active layer deposition and validation on membrane bioreactors

M.J. Luján-Facundo [a,*], J.A. Mendoza-Roca [a], A. Bes-Piá [a], E. Zuriaga-Agustí [b], Sergio Mestre [c,d], María-Dolores Palacios [d]

[a] Instituto de Seguridad Industrial, Radiofísica y Medioambiental, Universitat Politècnica de València, Camino de Vera, s/n, Valencia 46022, Spain
[b] FACSA, C/ Mayor, 8-84, 12001 Castellón, Spain
[c] Departamento de Ingeniería Química, Universitat Jaume I, 12006 Castellón, Spain
[d] Instituto Universitario de Tecnología Cerámica, Universitat Jaume I, 12006 Castellón, Spain

A R T I C L E I N F O

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A B S T R A C T

The fabrication of eco-friendly ceramic membranes based on low-cost raw materials, using digital INKJET printing techniques for active layer deposition and its validation in a membrane bioreactor (MBR) has been studied. The raw materials used in the manufacture of the support layer were UA50/2 clay, chamotte, calcium carbonate and potato starch. A MBR laboratory plant was operated to treat municipal wastewater with three ceramic membranes with different grammages in the selective layer deposition. The membrane performance at laboratory scale was evaluated in terms of transmembrane pressure (TMP) evolution for a constant permeate flux, permeate quality and mixed liquor characteristics. From these experiments, it was selected the membrane which obtained the lowest TMP profile maintaining appropriate water quality parameters (chemical oxygen demand (COD) removal percentage around 90% and turbidity around 0.06 NTU). This membrane was also operated at pilot plant scale in order to validate it at higher scale. Results indicated that TMP values were in the range of 0.06 and 0.1 bar, COD removal percentage was around 98% and microbiology analysis demonstrated that the quality of the effluent, according to European regulation 2020/741, can be classified as Class A and it can be reused for non-potable purposes.

1. Introduction

Water is a precious substance for human life and plays an essential role in many aspects of the economy. Water requirements will significantly increase by 2050 due to population growth, increasing water pollution and cyclical droughts (García-Pacheco et al., 2018; Morón-López et al., 2019). In this context, since water resources are limited, the study of alternatives for the current wastewater treatments and the promotion of the water reuse are essential to achieve the environmental protection and to enhance circular economy.

Membrane techniques are among the most interesting separation technologies used for wastewater treatment. Specifically, membrane bioreactor (MBR) is used for industrial and municipal wastewater treatment mainly due to the high treated effluent quality, small footprint, complete rejection of suspended solids, high organic matter removal and relatively low cost for the advantages it can offer (Jeong et al., 2018; Judd, 2010). This technology combines a biological reaction and a membrane filtration process in a single compact reactor (immersed membranes) or as two independent steps when membrane module is external. Membranes are configured in MBR as flat-sheets or hollow fibers (for submerged configuration) and as tubular membranes (for external configuration)(He et al., 2019).

Regarding membrane material, the two main material types used for fabrication are polymers and ceramic materials. The second ones, are produced from inorganic materials such as alumina, titania and zirconia (Mestre et al., 2019). The application of ceramic membranes instead of polymeric membranes for MBR offers several advantages such as the long lifespan, high mechanical stability, cleaning agent resistivity, narrow and well-defined pore size distribution, higher porosity, high flux, hydrophilic properties, anti-fouling performance and recyclability as raw ceramic materials (He et al., 2019; Ren et al., 2019; Shang et al., 2017; Wang et al., 2019; Zhang et al., 2017). Ceramic MBR is used for many different applications such as the treatment of urban (Xing et al., 2001), textile (Chougui et al., 2019), food industry (Samaei et al., 2018)

* Corresponding author.
E-mail address: malufa@etsii.upv.es (M.J. Luján-Facundo).

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and pharmaceutical industry wastewaters (Zaviska et al., 2013). However, ceramic membranes are more expensive than polymeric ones due to the raw materials and the manufacturing difficulty (Jeong et al., 2017). Mainly for these reasons, the market for ceramic membranes is not as extended as the one of polymeric membranes (He et al., 2019). For these reasons, a worldwide line of research is focused on the development of ceramic membranes from low-cost raw materials, such as kaolin, clays, diatomaceous earth, bauxite, calcite, dolomite, etc. (Mestre et al., 2019). In this work, clay, calcite and chamotte were used as precursors of the membrane (support, intermediate and selective layer) and potato starch as pore generator. All raw materials are cheap, easily available, and their processing requires milder conditions than Al_2O_3, TiO_2 or ZrO_2 based membranes.

Ceramic membranes have an asymmetrical porous structure consisting of three layers. This structure usually consists of a macroporous support, which offers a high mechanical strength, an intermediate mesoporous layer to bind the inner and the outer layer and an active layer ensuring the separation process (Issaoui and Limousy, 2019; Mestre et al., 2019). The main methods for active layer deposition are casting and dip-coating (He et al., 2019).

Inkjet printing has emerged as an attractive method for the manufacture of functional substrates and for surface modification. Inkjet technique is a special deposition of fine ink bed on a substrate based in ^{2}D–^{3}D printing technology and following the previous design made by the 2D or 3D modelling software (Balogun et al., 2019; Yus et al., 2019). When applied to ceramic processing, the ink is transformed in a powdered or a continuous material adhered to the surface in the firing step. The main advantages of this printing technique is the low production costs and precise deposition control (Li et al., 2019). In this way, it is not necessary to elaborate a specific precursor for every kind of selective layer to control its properties. Digital printing allows a good control of the deposited quantity of ink and its spatial distribution. Consequently, if the nature of a variety of selective layers is the same, they could be obtained from the same ink. Second, the changes in the selective layer could be implemented changing a digital file, not adjusting, or changing machinery in the production line. These reasons had favored a broad adoption of inkjet printing in other areas of ceramics production, like tiles (Molinari et al., 2020), and could be a useful tool for membrane manufacture.

As a consequence, it is very interesting to study its application to the fabrication of new ceramic-based membranes, and to validate their application for wastewater treatment. The fact that the membranes are made of cheap, abundant and sustainable materials (such as fly ash, rice husk or ball clay) reduces their cost and the possibility to reuse the spent membrane as raw material for other ceramics promotes circular economy (Hubadillah et al., 2022). Thus, the research on new membrane materials would allow savings and solve many environmental problems. In this way, several authors have studied the application of low cost ceramic membranes made of kaolin for wastewater treatment (Kaur et al., 2015). In addition, Malik et al. (2020) fabricated low cost ceramic membranes by means of mixing several materials as kaolin, fly ash and dolomite. Hubadillah et al. (2016) investigated the viability of using kaolin as starting material for ceramic membrane support fabrication by means of phase inversion/sintering technique. However, the application of Inkjet technology for the fabrication of ceramic membranes has not been investigated yet.

The aim of this work is to develop ceramic membranes based on low-cost ceramic raw materials using digital inkjet printing techniques to deposit the selective layer and further validation on MBR. Different studies have been carried out on the use of ceramic membranes in MBRs (Ninomiya et al., 2020; Wan et al., 2023; Wang et al., 2021), including low-cost membranes (Ugarte et al., 2022), but polymeric membranes dominate industrial installations. The most innovative aspect of this article is the use of ceramic inkjet technology in membrane manufacture and validation of this new membrane at laboratory and pilot plant scales.

2. Materials and methods

2.1. Laboratory and pilot plants

On one hand, a laboratory plant was employed to study the behaviour of three low-cost ceramic membranes for MBR application. On the other hand, the membrane with the best performance was validated at pilot scale using the pilot plant described in this section.

2.1.1. Laboratory plant description

To study the viability of the new eco-friendly ceramic membranes, a submerged MBR system was built at laboratory scale (Fig. 1). The total volume of the biological reactor was 54 L divided into two compartments connected at the bottom part. A flat membrane with an effective area of 0.036 m^2 was located inside the reactor. In the first compartment, a mechanical stirrer (Velp Scientifica, Spain) was used to agitate the mixed liquor and an air pump EHEIM 100 (Spain) provided air in order to keep an oxygen concentration in the bioreactor around 2 $mg \cdot L^{-1}$. In the second compartment, a diffuser located below the membrane module was connected to a compressor in order to provide air bubbles to minimize the membrane fouling by air scouring and to maintain aerobic conditions.

The laboratory plant was also equipped with a peristaltic pump that could work in two modes (suction and impulsion modes), a pressure transducer, a temperature probe and two storage tanks. One tank (with a capacity of 50 L) contained the feed wastewater and the second one (volume = 5 L) collected the permeate stream. The peristaltic pump in suction mode was used to draw the permeate from the membrane and in impulsion mode to take this permeate from the 5 L tank for membrane cleaning by backwashing. Temperature, permeate flow rate and pressure in the permeate side of the membrane were registered every minute during each experiment using the acquisition data system provided by the automation system.

2.1.2. Pilot plant description

Pilot plant employed in this study presented the same configuration (it is also a submerged membrane bioreactor) and elements as the laboratory plant but at a higher scale. Fig. 2 shows an image of the pilot plant, which was located in a WWTP of Murcia region, Spain. The main components of this pilot plant are a cylindrical container with a capacity of 1500 L and a membrane module with a capacity of 32 flat membranes with an effective area of 0.19 m^2 per membrane. The pilot plant also includes: four air diffusers located at the bottom of the tank connected to two air blowers, a peristaltic pump to extract the permeate, a permeate tank (from which permeate was fed to the membranes for the backwashing), a mass flowmeter to measure the permeate flow and a pressure gauge in order to check the variation of the pressure in the permeate side. The distribution of all these elements can be observed in Fig. 2. The pilot plant was also equipped with an electrical panel and an automation software to register the pressure in the permeate side of the membranes and the permeate flow rate.

2.2. Low-cost ceramic membrane manufacturing

Ceramic membranes made with low-cost materials were designed with a three layer structure (high-porosity support, intermediate layer and selective layer). It is important to highlight that the raw materials were cheaper than the refractory oxides currently used in the industrial manufacture of ceramic membranes. The raw materials used in the manufacture of the support layer were UA50/2 clay (40 wt%), chamotte (20 wt%), calcium carbonate (15 wt%) and potato starch (25 wt%). These materials were kneaded with water and a small proportion (0.2 wt % with respect to the solid) of an additive (Biokeram) to obtain a homogeneous mass. This mass was extruded to form rectangular supports of 20 × 60×0.7 cm. After drying in an oven, the supports were fired in a laboratory muffle with a slow cycle designed to allow the starch

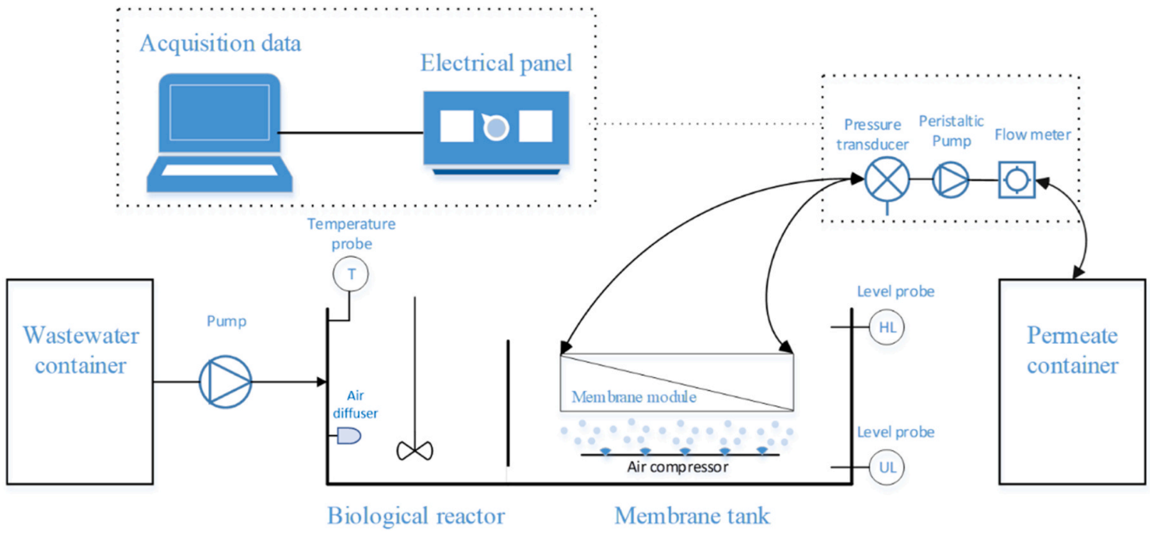

Fig. 1. Scheme of the Laboratory plant.

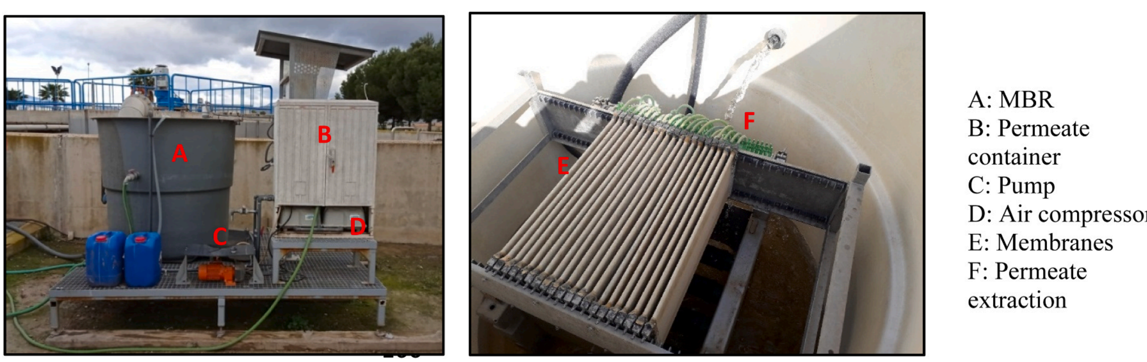

Fig. 2. Pilot plant and membrane module included in the pilot plant scale.

oxidation and then the sintering of the ceramic fraction by treatment at 1165 ºC for one hour. The fired supports were cut up to 20 × 8×0.7 cm pieces for subsequent tests.

The raw materials for the intermediate layer were UA50/2 clay previously calcined at 700 ºC (90 wt%) and bentonite (10 wt%). An aqueous suspension was prepared in a ball mill with a solids content of 43 wt%. This suspension was applied by spraying until both sides of the support were covered homogeneously. After drying in an oven, the samples were fired at 1060 ºC for 1 h.

The ink for the selective layer was formulated with the UA50/2 clay calcined at 900 ºC, an industry-standard medium for ceramic inkjet inks and various additives. The raw materials were processed in a microballs mill to obtain an ink with optimal physical properties and suitable particle size distribution (99 wt% below 1 µm) for printing. The printing tests were made in a laboratory ceramic inkjet printer of the company SYSTEM Spain S.A. The ink was applied to the supports already coated with the intermediate layer, with grammages of 30, 60 and 80 $g{\cdot}m^{-2}$ (named as Membrane 30, 60 and 80, respectively). Two replicas of each of them were made. The samples were fired at 900 ºC to sinter the selective layer. These 3 membranes were evaluated at the MBR laboratory scale and the one obtaining the best results was validated in the MBR pilot plant.

2.3. Membrane characterization

The morphology of the laboratory membranes was characterized by means of a Field Emission Gun Scanning Electron Microscope (FEG-SEM) model Quanta 200 F, FEI. The working conditions for the analysis of the three laboratory membranes used (Membrane 30, Membrane 60 and Membrane 80) were backscattered mode, acceleration voltage of 20 kV, and chamber pressure of 0.3 mbar.

2.4. Composition of the wastewater

The treatment of municipal wastewater was used to check the efficiency of the low-cost ceramic membranes. For the laboratory experiments, simulated municipal wastewater was selected as influent to maintain the stability of the influent water characteristics. The synthetic wastewater prepared for the MBR experiments consisted of meat extract, bacteriological peptone (both supplied by Panreac, Spain) and trisodium phosphate 12-hydrate (from Panreac, Spain). The concentrations of these compounds were fixed in order to achieve a COD:N:P ratio of 100:5:1 to ensure the appropriate nutrients amount. These chemicals were mixed and dissolved in tap water. COD of the simulated wastewater was varied between 500 and 1000 $mg{\cdot}L^{-1}$ depending on the mixed liquor suspended solids (MLSS) fluctuation in order to maintain the organic load around 0.1 g COD·g $SS^{-1}{\cdot}d^{-1}$.

As commented above, from laboratory experiments, the best membrane was selected in order to validate it at pilot scale. The pilot plant was fed with the pretreated wastewater influent to the WWTP. Table 1 shows the characterization of both synthetic and actual wastewater used in the experiments.

Table 1
Characteristics of the synthetic and municipal wastewater.

	Synthetic wastewater	Municipal wastewater
pH	7.8 ± 0.24	7.9 ± 0.15
Conductivity (µS·cm^{-1})	2958 ± 89	3632 ± 223
COD (mg·L^{-1})	750 ± 198	754 ± 29

2.5. Experimental set-up

2.5.1. Laboratory plant operation

The activated sludge from a WWTP located in Comunidad Valenciana (Spain) was used as seed sludge for the MBR. Synthetic wastewater was prepared according to Section 2.4. The total duration of each test was 4 days.

The MBR was operated at a constant flux mode (31 LMH) and the permeate side pressure was registered to control membrane fouling. Initially, the experimental cycles were performed as follows: 8 min of filtration (suction step), 1 min of relaxation and 2 min of backwashing. However, from the first 24 h of operation on, the backwashing step was increased until 4 min and a commercial NaClO solution with a concentration of 2.54 g·L^{-1} was added every day to the backwashing tank (permeate tank) in order to prevent a more severe fouling in the membrane. It has to be taken into account that it is expected a higher fouling in comparison with polymeric membranes. For example, Ninomiya et al. (2020) considered chemical cleaning frequencies between 6 and 120 h.

2.5.2. Pilot plant operation

F/M ratio was also fixed at 0.1 g COD·g SS^{-1}·d^{-1} and real municipal wastewater was treated. The total duration of the experiment was 10 days. The MBR was operated also at a constant flux mode (32 LMH) and the pressure in the permeate side was monitored to control the process. The cycles and the cleaning phase in the MBR were carried out in the same way as for the laboratory plant scale except for the filtration step, that in this case had a duration of 12 min.

2.6. Analytical methods

2.6.1. Effluent quality

In order to control the water quality in the MBR effluent, several water quality parameters were measured. Samples were filtered through a 0.45 µm filter before the analysis of chemical oxygen demand (COD), total phosphorous (TP), total nitrogen (TN), and ammonium nitrogen (NH_4^+-N). These parameters were measured by means of kits from Merck (Spain). In addition, pH, conductivity and turbidity were analysed. The pH was measured with the pH-Meter GLP 21$^+$ and the conductivity with the conductivity meter EC-MeterGLP31$^+$ (both from Crison instruments, Spain). Turbidity was determined with a Turbidimeter D-112 from Dinko Instruments. COD and turbidity were analysed every day and the rest of parameters were measured three times per test or week for the laboratory and pilot scale, respectively.

2.6.2. Mixed liquor characterization

In this study, to control the concentration of the biomass for ensuring working at the same mass load, mixed liquor suspended solids and mixed liquor volatile suspended solids (MLVSS) concentrations in the biological reactor were measured every day following the protocol described in APHA (2005). In addition, pH and conductivity measurements were also carried out in the mixed liquor.

In the experiments carried out at laboratory scale, the mixed liquor was also characterized in terms of soluble microbial products (SMP) concentration. In this way, SMP concentration was studied in terms of proteins and carbohydrates. Protein concentrations were measured by means of Bicinchonic acid (BCA) assay test from Novagen. Carbohydrates were determined using the Antrone method (Frolund et al., 1996). Finally, DNA concentrations in the biological reactor was quantified using Quant-it™ dsDNA HS (0.2–100 ng) kit from Invitrogen (Spain).

2.6.3. Microbiological analysis

In order to check the water quality in the MBR effluent operated at pilot scale, several bacteriological indicators were measured: *Echerichia Coli* (UFC/mL), *Total Bacteriophages* (Plaque-Forming Particles, PFP·mL^{-1}), somatic *Coliphages* (PFP·mL^{-1}) and specific *Bacteriophages* (PFP·mL^{-1}). For all of them, a filtration and subsequent incubation in the appropriate culture medium was carried (AENOR, 2008). *Echerichia Coli* was determined following the APAT CNR IRSA 7030 procedure (APAT CNR IRSA, 2003). *Clostridium Perfingens* was measured according to UNE-EN ISO 14189. Total and specific *Bacteriophages* were analyzed following UNE-EN ISO 10705 and, *Coliphages* were examined using the UNE-EN ISO 10705–2:2000 method.

3. Results

3.1. Microstructure of the ceramic membranes

The characterization of the ceramic membranes by SEM confirmed the three-layered structure (Fig. 3). The micrographs showed the membrane support, with large pores and large particles between 100 and 500 µm. An intermediate layer of about 15 µm of thickness was located on the top of the support, and over it the active layer. The surface of the intermediate layer was not completely flat. It was a consequence of the high size of the chamotte particles present in the support, which caused undulations on the surface of the substrate after sintering. As a consequence, the intermediate layer could not completely dampen. This phenomenon provoked irregularities in the thickness of the active layer, but, on average, the Membrane 30 (Fig. 3b) presented an active layer approximately 2 µm thick, the Membrane 60 (Fig. 3c) had a thickness around 6–7 µm, and Membrane 80 (Fig. 3d) reached a thickness between 8 and 10 µm.

3.2. TMP evolution of the laboratory MBR

The variation of the pressure during the experimental time for each membrane was continuously monitored and these results are shown in Fig. 4. It is important to highlight that only the last value of TMP at the end of each filtration step of the cycle has been represented. MBR was operated at a fixed flux of 31 LMH and a chemical cleaning with NaOCl solution (2.54 g·L^{-1}) was carried out every day (this corresponds to the red vertical line in Fig. 4, dividing the experiment in 4 stages).

The TMP gradually increased over experimental days, then dropped drastically after the first chemical cleaning (end of stage 1). It can be observed in Fig. 4 that in the stage 1 (first 24 h of operation), membrane TMP was maintained between 0.22 and 0.63 bar, 0.37–0.75 bar and 0.4–0.81 bar for Membrane 30, Membrane 60 and Membrane 80, respectively. The fast increase in this stage of the TMP was due to both the membrane fouling and the high pressure loss caused by the membrane itself (due to the thickness of the three layers), especially for Membrane 80. From stage 2 (from the 2nd day of operation), the TMP decreased significantly, mainly in the cases of Membrane 30 and 60. This was due to the use of NaClO every 24 h to clean the membrane in the backwashing step, which resulted effective for foulants removal. During the stage 3 and 4 (the last 2 days of the experiment) the TMP reached a constant value around 0.5, 0.55 and 0.6 bar, for Membrane 30, 60 and 80, respectively. It can be also observed that sudden TMP increases occurred during the tests, especially for Membrane 80 (for example around cycle number 100). It could be due to pore blocking, which was produced in more extent in the membrane with the thickest active layer. The backwashing of the following cycles eliminated this fouling gradually and this is the reason why TMP decreased.

Ugarte et al. (2022) published a work also using low-cost ceramic membranes, obtaining TMP values for the 5th day of operation between

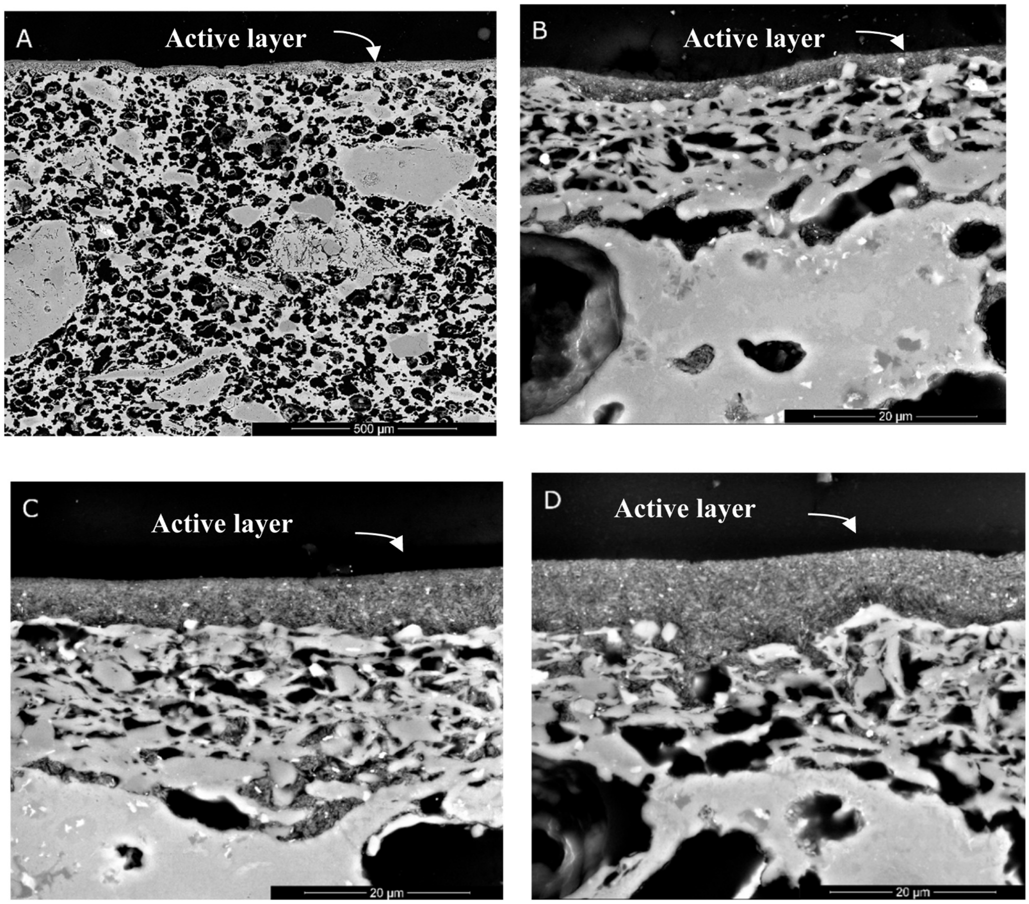

Fig. 3. FEG-SEM images of a) Membrane 30 complete structure, b) Selective layer of Membrane 30, c) Selective layer of Membrane 60 and d) Selective layer of Membrane 80.

0.25 and 0.45 bar. These authors worked at permeate flux of 15 LMH, lower than the permeate flux fixed in this study (31 LMH), which could be directly related with the lower TMP obtained. For the three membranes tested in this study, results from TMP variation (specially for the stage 1) suggest a progressive membrane fouling that resulted in an increase in irreversible fouling which could not be removed in the backwashes with permeate (Villarroel et al., 2013). However, M-30 membrane showed the highest resistance to fouling since TMP profile was lower during the all the process.

3.3. Overall treatment performance of the laboratory MBR tests

The turbidity of the permeate was measured regularly and results for the three membranes were all lower than 0.36 NTU. The turbidity average value was 0.06, 0.37 and 0.055 NTU for Membrane 30, Membrane 60 and Membrane 80, respectively. It can be noted that all the values are very low except for Membrane 60. However, even for this membrane, these values demonstrated the high water quality (in the same order of magnitude of a commercial membrane) in terms of turbidity. Thus, according to turbidity values, Membrane 30 and Membrane 80 showed the best results. In addition, these results are in concordance with previous studies focused on municipal wastewater using MBR such as Isik et al. (2022) and Arévalo et al. (2009) who reported turbidity values in the effluent around 0.3 in both cases.

Fig. 5 shows the COD removal efficiency during the experimental time for the three membranes tested. As it can be observed, COD removal efficiencies quickly reached a value around 90% for the three membranes. These are typical COD removal efficiency values, which have been reported by different authors (Oztemur et al., 2020; Ugarte et al., 2022).

3.4. Mixed liquor characterization in laboratory MBR

Regarding the MLVSS variation, the percentage of MLVSS showed a relatively constant value (around 75%) during the MBR operation for the three membranes tested. On one hand, 75% of MLVSS is characteristic of a low organic load process. On the other hand, no variations in the percentage of MLVSS indicated that biomass activity is stable (Fan et al., 2015). Thus, biological process was stable irrespective of membrane type. These results were in concordance with those previously published by Villain et al. (2014) treated synthetic urban wastewater with a MBR, who reported a MLVSS percentage of the acclimated sludge of 82%.

The membrane fouling in MBR is complex and depends on the nature of the foulants and the interactions between membrane and foulants. However, the main substances that affect membrane fouling in the membrane bioreactor process are SMP (Teng et al., 2019, 2020). The decrease in the membrane water flux was mainly attributed to a combination of extracellular polymeric substances, SMP, and fine sludge particles attached to the membrane surface and pores (Zhang et al., 2017). In this way, to study the influence of sludge properties on each membrane, the SMP concentrations were measured in terms of proteins and carbohydrates (Fig. 6 and Fig. 7 respectively). It is important to highlight that for the tests M-30, even with a higher SMP concentration in the reactor, the values of the TMP were lower, as it was commented in Section 3.2. In addition, the results indicated that in the tests for M-30, proteins were almost constant, and carbohydrates slightly decreased at the end of the experiment what influenced positively on membrane

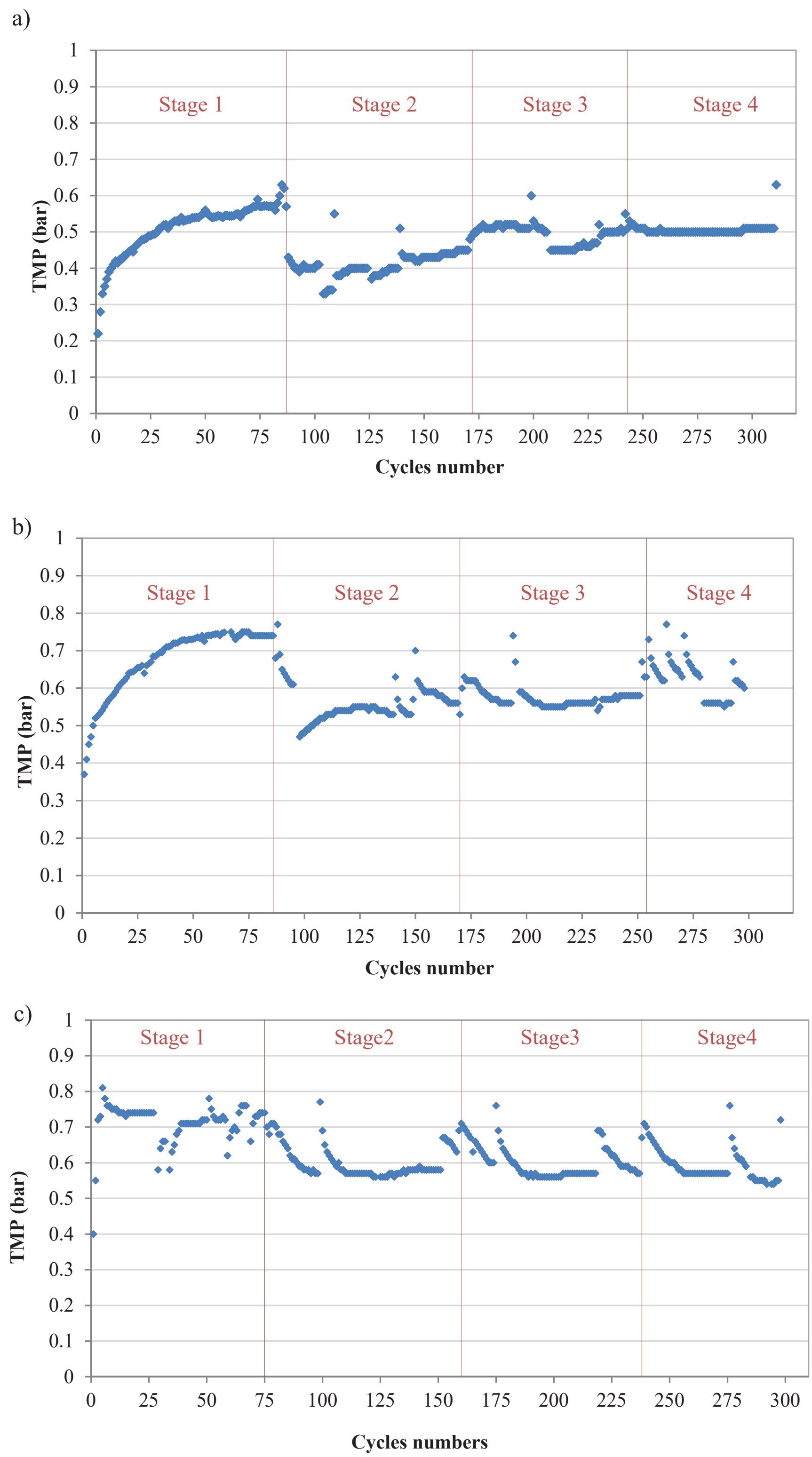

Fig. 4. TMP evolution for each cycle during the laboratory MBR test for a) Membrane 30, b) Membrane 60 and c) Membrane 80.

fouling. For M-60, the SMP concentrations were practically constant in all the tests and, for the test with M-80, there was not a clear tendency. However, in general terms, SMP concentrations were low (19.47 mg·L^{-1} and 23.10 mg·L^{-1} as a maximum value for proteins and carbohydrates, respectively) for all the tests. These results follow the same tendency that a previous study with a MBR published by Teng et al. (2020), who reported a total concentration in proteins and polysaccharide between 13 and 27 mg·L^{-1}. The lowest values of SMP obtained for Membrane 80 (especially those of 3rd operation day) could be due to a higher accumulation of these substances inside membrane pores (adsorption) due its higher grammage in comparison with the other membranes.

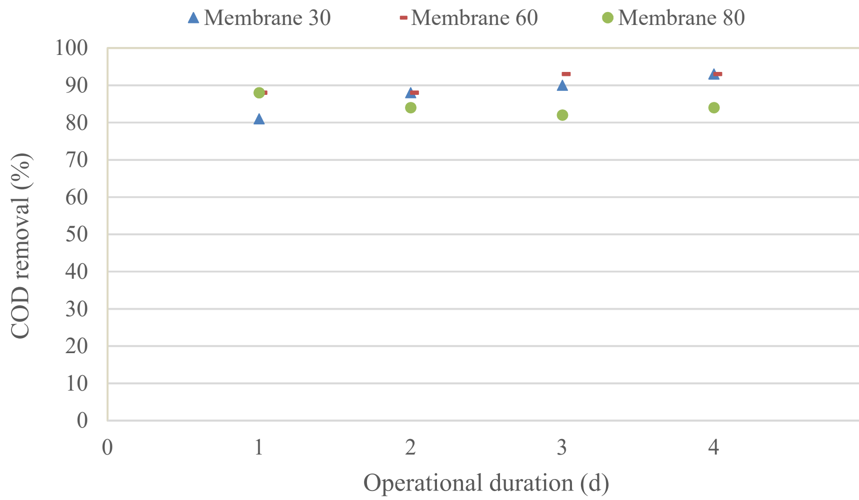

Fig. 5. COD removal percentage during the laboratory MBR tests.

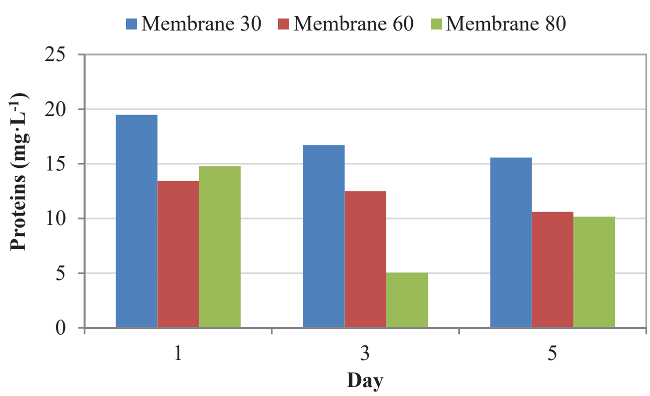

Fig. 6. Proteins concentrations for the three membranes.

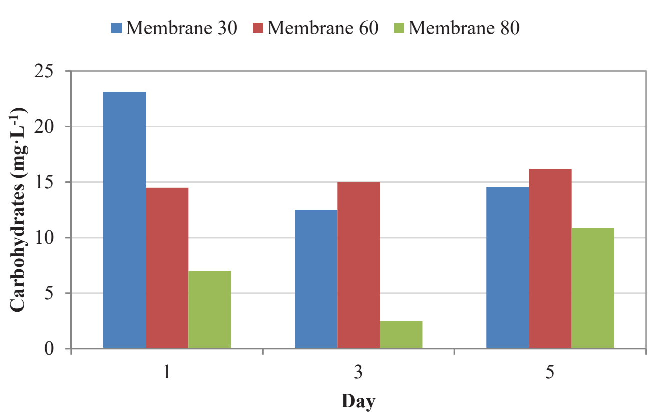

Fig. 7. Carbohydrates concentrations for the three membranes.

3.5. *Membrane validation in the MBR pilot plant*

As it was commented above, M-30 membrane showed the highest resistance to fouling since TMP profile was lower during all the process. Thus, according to the results showed in previous sections in terms of TMP and water quality parameters, the membrane M-30 was selected as the best option for MBR application for its validation in pilot plant. In addition, this membrane is more environmentally friendly since it has the thinnest active layer, saving in the inkjet material used. Fig. 8 reflects the TMP evolution during the experiment. It can be observed that

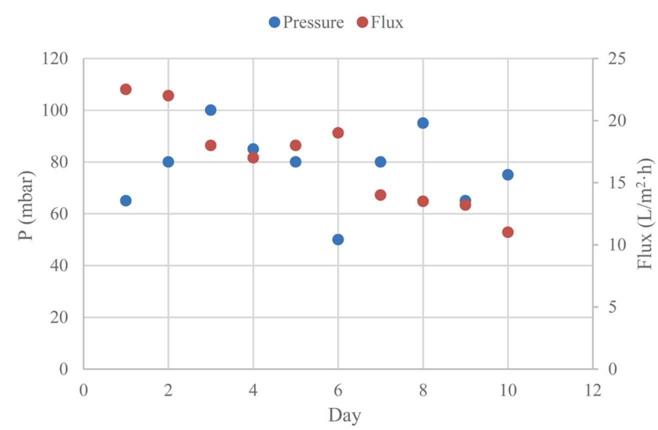

Fig. 8. TMP variation for M-30 at MBR pilot scale.

TMP values were lower than those obtained at laboratory scale (Fig. 4). This was due to two main reasons: the higher air scouring flow rate per membrane area employed at pilot plant scale than at laboratory scale, and the lower operating permeate flux in pilot plant, which could not be fixed independently of the TMP (the reason why there is not a steady trend), ranging between 12 and 22 LMH. In spite of these considerations, it has to be highlighted that TMP values in the range of 0.06–0.1 bar were obtained during the tests, being a key result for implementing these membranes in MBR processes in a next future.

Table 2 reflects the water quality parameters in terms of COD, turbidity and microbiological analysis in the influent and effluent of the

Table 2
Water quality parameters for the MBR pilot plant.

	Influent	Effluent	Removal (%)	Logarithmic removal units (log)
Turbidity (NTU)	259	0.41	99.84	-
COD (mg·L)	754	15	98.01	-
E. Coli (UFC/100 mL)	6,700,000	24	100.00	5.45
Total Bacteriophages (P.F.P/mL)	53,000	2	100.00	4.42
Somatic Coliphages (P.F.P/mL)	37,000	2	99.99	4.27
Specific Bacteriophages (P.F.P/mL)	16,000	20	99.88	4.2

MBR pilot plant. As it can be observed, removal percentages for all the parameters are higher than 98%, which indicated an excellent membrane (M-30) performance. The wastewater treated in this system has the aim to be reused for non-potable purposes such as irrigation or street cleaning. In this case, at European level, from year 2023 it is necessary to follow the European regulation 2020/741. According to this legislation, this effluent meets the values required to be classified as Class A, which is the highest quality in this regulation.

4. Conclusions

In this article, the fabrication and further validation of MBR with ceramic membranes based on low-cost raw materials using digital inkjet printing techniques was studied. Regarding experiments carried out at MBR laboratory scale, Membrane 30 (with an intermediate layer grammage of 30 g/m^2) was the best option since it obtained the lowest TMP profile and good water quality parameters (COD percentage removal around 90% and turbidity around 0.06 NTU). In addition, SMP concentrations did not affect significantly the MBR performance during the use of this membrane. From membrane validation at pilot plant scale, results indicated that TMP values were in the range of 0.06 and 0.1 bar, COD percentage removal efficiency was around 98% and microbiology analysis demonstrated that the quality of the effluent, according to European regulation 2020/741, can be classified as Class A and can be reused for non-potable purposes. As a main conclusion, the three ceramic membranes manufactured with low-cost raw materials using digital inkjet technology were suitable to treat urban wastewater taking into account results of water quality parameters and mixed liquor characteristics. Longer experiments will have to be performed to prove that these inorganic low-cost membranes may have a niche in the MBR market.

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.

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