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Preparation and characterization of low-cost ceramic microfiltration membranes for water treatment

Boudjemaa Ghouil[1,*], Zineb Khebli[1], Ferhat Bouzerara[1], Dounia Zermani[1], Chahinez Zitouni[1,2], Keltoum Youla[1], Tayeb Idoui[2], Tarek Khennouf[2]

[1]*Laboratory of Condensed Matter Physics and Nanomaterials, Faculty of Exact Sciences and Computer Science, University of Jijel, 18000, Jijel, Algeria*
[2]*Laboratory of Biotechnology, Environment and Health, Faculty of Natural and Life Sciences, University of Jijel, 18000, Jijel, Algeria*

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Abstract

This work presents the preparation of microfiltration (MF) membranes deposited on highly porous supports tailored for microfiltration applications. The supports were prepared from local Algerian clays and calcium carbonate, selected for their abundant availability. Subsequently, these supports underwent coating with microfiltration membrane using the slip casting method employing the same clay powder. The resulting membrane, sintered at 1100 °C, exhibited desirable attributes including a thickness of approximately 27 µm and an average pore size (APS) value of about 0.42 µm, coupled with notable adhesion between the support and the membrane. Furthermore, physicochemical and microbiological tests were conducted on water samples to confirm the effectiveness of the membrane in microfiltration. The results show that the membrane is highly effective in removing turbidity, with a rejection rate of approximately 99%. The pH and conductivity of the water remain stable during filtration. Additionally, the membrane demonstrates significant efficiency in removing heavy metals, with rejection rates of about 68% for iron and 90% for aluminium, as well as it is effective in removing specific bacteria from water.

Keywords: *clay membranes, slip casting, extrusion, structure, permeability and efficiency*

I. Introduction

Porous ceramic membranes are the subject of many research papers [1–8], particularly in recent years [9–19]. The specific properties of ceramic membranes which have attracted the attention of scientists are high thermal stability [9,20], mechanical resistance [2,3], longer life-time and the ease of cleaning [1–3]. In addition, these membranes are characterized by their high chemical stability and resistance to highly corrosive acids and alkali media [21,22].

Clay, kaolin, calcium carbonate ($CaCO_3$), dolomite ($CaCO_3 \cdot MgCO_3$), feldspar and quartz are the raw materials abundantly available in Algeria. Many industries fields are becoming interested in the use of traditional ceramics as raw materials for production of advanced products due to their low price and abundant availability. Moreover, from an economic and energetic point of view, the possibility of replacing the more expensive starting materials like alumina by cheaper raw materials has also advantage of reducing the sintering temperature. For example, the membrane supports prepared from alumina are usually sintered at 1600 °C, while the sintering temperature decreases to about 1250 and 1140 °C when the kaolin and clay are used as starting materials [9–13].

Many studies have already been realized in order to valorise clay, kaolin and quartz, which represent the most abundant raw materials in many countries for the production of ceramic membranes [23–26]. However, the low mechanical resistance of the membrane to filtrate flow induced by the applied pressure difference requires the use of a support that can provide strength to withstand this pressure [2]. On the other hand, microfiltration (MF) ceramic membranes are used for the separation of microorganisms and the removal of par-

*Corresponding author: +213 670 03 85 63
e-mail: *boudjemaa.ghouil@univ-jijel.dz*

ticles from suspensions [14,27,28]. These membranes are widely used on a large scale in many economical fields, especially for purification and concentration in food, pharmacy and chemical industries [29–37].

This study aims to develop a low cost microfiltration ceramic membrane (consisting of macro porous ceramic supports and MF as a top layer membrane) prepared from clay and calcite local raw materials for use in water treatment. The use of these two raw materials has many advantages, such as reducing the cost of the used material and valorisation of our raw materials. The supports were characterized by evaluating their morphology, mechanical and chemical properties. After that, the selected support was used to prepare a MF membrane which was then characterized to show its morphology and structural properties. Furthermore, for estimating the potential of the top layer membrane, especially in the water treatment field, some physicochemical and microbiological tests were also realized.

II. Experimental

2.1. Supports preparation

Local clay and calcium carbonate (CC) powder (99% purity) were used as starting materials. The clay powder was sourced from the Jijel region, Algeria, and the calcium carbonate powder was supplied by ENG/National Aggregates Company, Elkhroub Calcium Carbonate Unit, Constantine, Algeria. The clay powder was ground and sieved through a 150 μm sieve. The average particle size of CC was around 5 μm [4]. The organic additives, Amijel and methocel (methyl cellulose, from Sigma-Aldrich 3050-Spruce Street, Saint Louis, MO 63103 USA) were used in order to improve the rheological properties of the paste that facilitate the forming of supports. The plastic paste preparation was carried out by mixing clay (70 wt.%), CC (30 wt.%) and organic additives (4 wt.% of the total ceramic powder), with the progressive addition of water. This mixture was used to fabricate membrane supports in tubular and flat configurations (Fig. 1), using extrusion and roll pressing methods, respectively. The tubular supports had inner and outer diameters of 6 mm and 10 mm, respectively, while the length of the supports was according to our needs. The flat supports, in the form of discs, had a diameter of 50 mm and a thickness of 2 mm. These flat supports were used in the front filtration tests. After drying at room temperature for 24 h, the supports were sintered for 1 h at different temperatures ranging from 1100 to 1150 °C with a sintering rate of 5 °C/min.

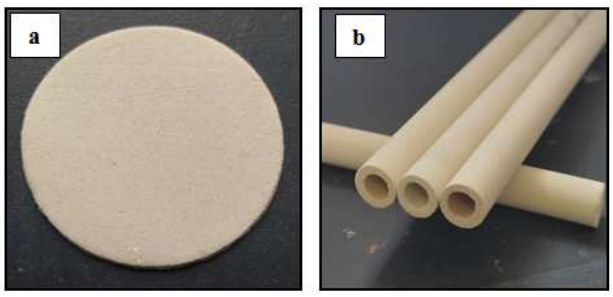

Figure 1. Photographs of the prepared supports: a) flat and b) tubular configurations

2.2. Membrane preparation

Microfiltration membranes were prepared using the same powder employed for the preparation of the supports. This powder was first crushed and sieved through a 40-mesh screen to ensure proper particle size distribution. The membrane was then deposited onto the porous support sintered at 1140 °C using the slip casting method. To prepare the suspension for membrane fabrication, clay powder (15 wt.%) was mixed with distilled water (55 wt.%), and the mixture was stirred for 4 h to ensure uniformity. Subsequently, 30 wt.% of hydroxyethyl cellulose (sourced from Merck Schuchardt OHG, Germany) was added to the clay suspension and the mixture was stirred for an additional 24 h at room temperature. The resulting suspension was then applied to the porous support for 10 min at room temperature and a coating was formed. For tubular membranes, the tube was sealed at one end and filled with the prepared suspension. The obtained coatings were additionally dried vertically for about 24 h at room temperature and subsequently sintered at 1100 °C for 1 h with a heating rate of about 5 °C/min. This temperature was selected because it allows the obtaining of the adequate characteristics of the membrane, precisely, the adhesion between support and membrane.

2.3. Structural characterization

Chemical composition of the clay powder was analysed using X-ray fluorescence spectrometry with a Zetium Malvern Panalytical instrument (Zetium, Malvern Panalytical, Great Malvern, UK). The structural evolution of the powders was also evaluated using differential scanning calorimetry (DSC) and thermogravimetric analysis (TGA) with a PerkinElmer Diamond TGA/DSC system (SDT Q600 TA). These two analyses were carried out under a nitrogen atmosphere with a heating rate of 10 °C/min, from the room temperature to 1100 °C. Phase composition of the samples was characterized using X-ray diffraction (XRD) with a Bruker AXS D8 Advance diffractometer, utilizing Cu Kα radiation (λ = 1.54056 Å). Structural features were determined by Fourier transform infrared (FTIR) spectroscopy on Shimadzu FTIR-8400S instrument.

The total porosity, the average pore size (APS) and the pore size distribution (PSD) were determined by the mercury intrusion porosimetry method (Micromeritics, Model Autopore 9220). The morphology (surface and cross-section) of prepared supports was observed by scanning electron microscopy (SEM) using the Thermo Scientific, Quattro S apparatus, where all samples were gold coated before SEM examination. The mechanical properties of the sintered specimens were evaluated

using a flexural strength test with a PASCO ME-8236 machine (San Lorenzo, CA, USA).

The tangential filtration experiments were realized using a home-made laboratory setup at a room temperature with a pressure obtained from an air gas source (Fig. 2). The permeate flux of the supports was characterized by distilled water flux (*DWF*) which was obtained by the following equation [2]:

$$DWF = \frac{Q}{S \cdot t \cdot P} \quad (1)$$

where, Q is the flux of water, S is the surface of the support membrane, t is the time and P is the pressure. The chemical corrosion resistance of the prepared supports was evaluated by the mass loss after being immersed into acid and alkali solutions at a room temperature for one month. The concentrations of heavy metals in effluents were determined using an atomic absorption spectrometer (Analytik Jena contrAA 800D, Germany).

2.4. Microbiological tests

In order to study the capability of the MF membranes to remove bacteria, Plat Count agar (PCA) and Violet Red Bile Glucose agar (VRBG) culture mediums were used. All materials used for the filtration test were sterilized in autoclave. Then, after the filtration process, the culture mediums were inoculated by the water samples (before and after filtration). Some samples immersed in PCA culture medium were placed at room temperature for 72 h to detect the saprophytic bacteria and the other samples were placed in an incubator at 37 °C for 24 h to detect the pathogenic bacteria. Other samples immersed in VRBG culture medium were placed for incubation at 37 °C for 24 h to detect the *Enterobacteriaceae* family, where the growth of bacteria was detected by visual check of the formation of bacterial colonies on the surface of the culture medium.

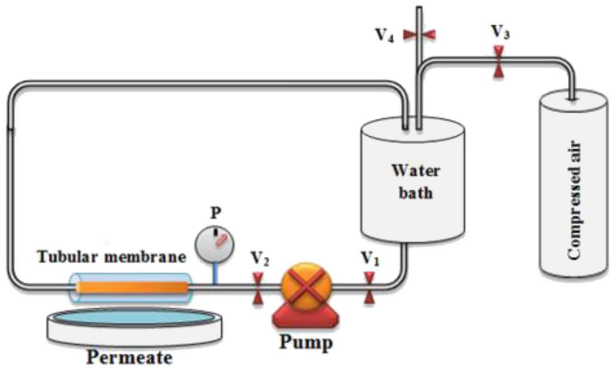

Figure 2. Schematic representation of the experimental setup used in the filtration process with tubular membranes

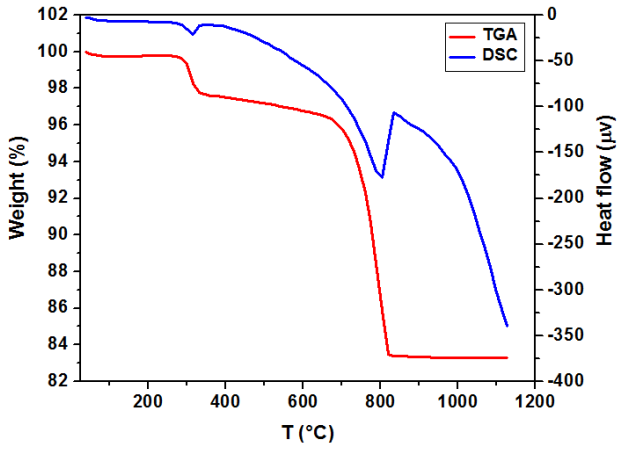

Figure 3. DSC and TGA analysis of the un-calcined support prepared from clay and 30 wt.% calcite

III. Results and discussion

3.1. Structural characterization of supports

The starting clay powder was characterized by different techniques. According to the XRF analysis the used clay powder consists of large amounts of silica and alumina and small portions of Na_2O, CaO, K_2O and Fe_2O_3. The main crystalline minerals existing in this clay are quartz and microline. The clay particles have a random form with different sizes mostly above 10 µm. The FTIR spectrum of the starting clay confirmed that the characteristic bands correspond to Si–O (at around 694 cm^{-1}), Si–O–Si (at around 647 and 1015 cm^{-1}), Si–O–Al (at around 773 and 797 cm^{-1}), Al–OH (at around 913 and 1539 cm^{-1}) and OH (at around 3619 and 3696 cm^{-1}).

Figure 3 presents the thermal analysis of the raw membrane support, prepared from a mixture of clay and 30 wt.% CC. The DSC curve showed two endothermic peaks. The first endothermic peak at 316 °C is probably due to the loss of organic materials (Amijel and Methocel), accompanied by a weight loss of 3 wt.% as shown on the TGA curve. In the temperature range of 650 to 830 °C, the DSC curve shows an endothermic peak at 800 °C. This second endothermic peak is related to the thermal decomposition of $CaCO_3$ into CaO and CO_2, and the TGA curve presented a weight loss of about of 13 wt.% during this process. Finally, it should be mentioned that the total weight loss for this mixture was about 16%.

Phase identification has an important role in the final supports proprieties determination, because the presence of certain phases can improve their physical and mechanical properties [3,6,32]. Figure 4 shows the XRD spectra of the supports sintered at different temperatures (ranging from 1100 to 1150 °C) for 1 h. The main identified phases are anorthite ($CaO \cdot Al_2O_3 \cdot 2\,SiO_2$), gehlenite ($2\,CaO \cdot Al_2O_3 \cdot SiO_2$), wollastonite ($CaSiO_3$) and quartz. The XRD spectrum of the support sintered at 1100 °C indicates that gehlenite is the predominant phase with the presence of anorthite and wollastonite as minor phases. Additionally, the most important observation is that the amounts of anorthite and wollastonite phases increase when the sintering temperature is increased up to 1140 °C, where wollastonite becomes the predominant phase. In contrast, gehlenite phase de-

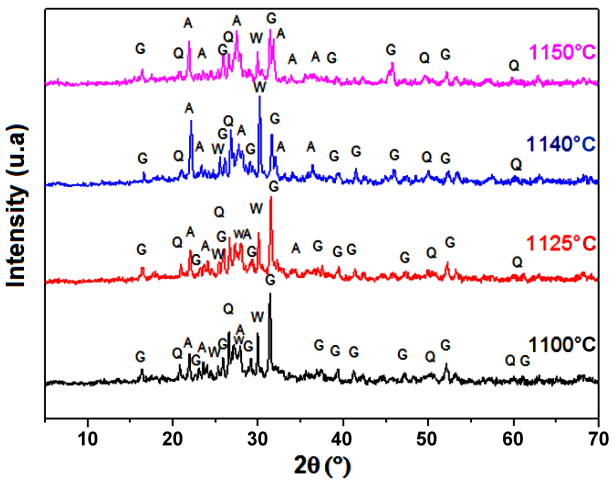

Figure 4. DSC and TGA analysis of the un-calcined support prepared from clay and 30 wt.% calcite

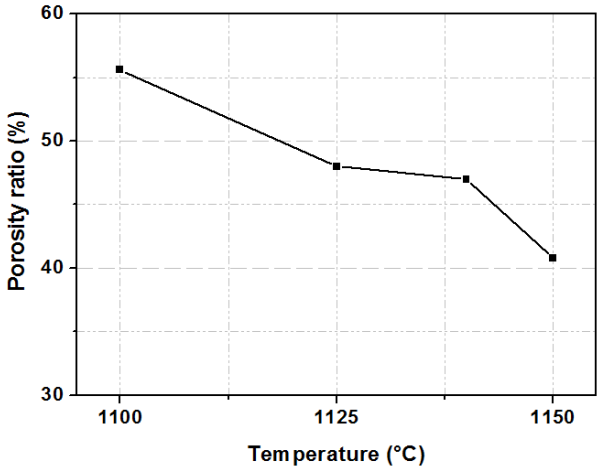

Figure 6. Variation of porosity ratio with a sintering temperature for support samples

creases as the sintering temperature increases. Furthermore, in the XRD spectrum of the support sintered at 1150 °C, an increase in anorthite phase and a decrease in wollastonite phase can be observed. Finally, these identified phases are of great importance because of their promising physical and mechanical properties.

Microstructure of the prepared supports was analysed by SEM and corresponding images of the tubular membrane supports sintered at 1125, 1140 and 1150 °C for 1 h are shown in Fig. 5. The supports sintered at 1125 and 1140 °C have very similar surface morphology, which indicates a homogeneous pore distribution and a highly porous structure. Additionally, no macro-defects such as cracks were observed. Furthermore, from these results an increase in the pore size with the increase of sintering temperatures can be seen. These features are a key condition for the preparation of good quality supports. On the other hand, the images of supports treated at 1150 °C show a high degree of grain adhesion, and some large pores can also be observed, indicating the high densification of support samples at this temperature.

To prepare high performance supports, the major properties to be considered are high porosity ratio, narrow pore size range, high mechanical strength and chemical stability [2,6,20]. Analyses of the sintering experiments showed that the total porosity of membranes decreases (Fig. 6) and the average pore size (ASP) increases with the increase of sintering temperature (Fig. 7). For example, the supports sintered at 1100 °C for 1 h had a porosity ratio of about 55.6% and an APS around 8.5 μm, whereas the supports sintered at 1125 °C had a porosity ratio of about 48% and an APS around 10.7 μm. Moreover, the porosity of the supports sintered at 1140 °C is about 47% and sharp decrease from 47 to 41% was observed when sintering temperature increased from 1140 to 1150 °C. Initially, at lower sintering temperature, porosity may decrease due to the decomposition processes or thermal expansion outweighing the effects of sintering. However, as

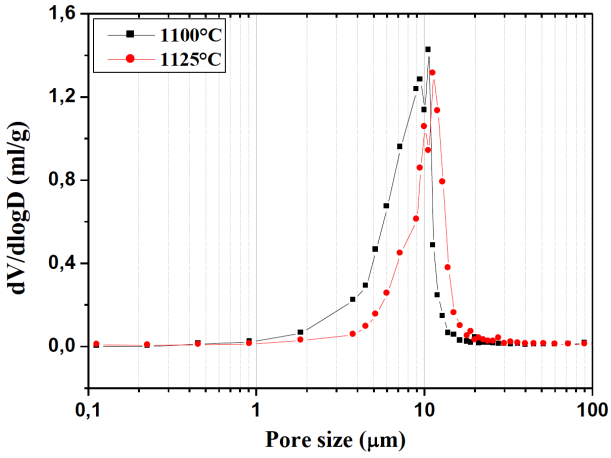

Figure 7. Pore size distribution of support samples sintered at different temperatures for 1 h

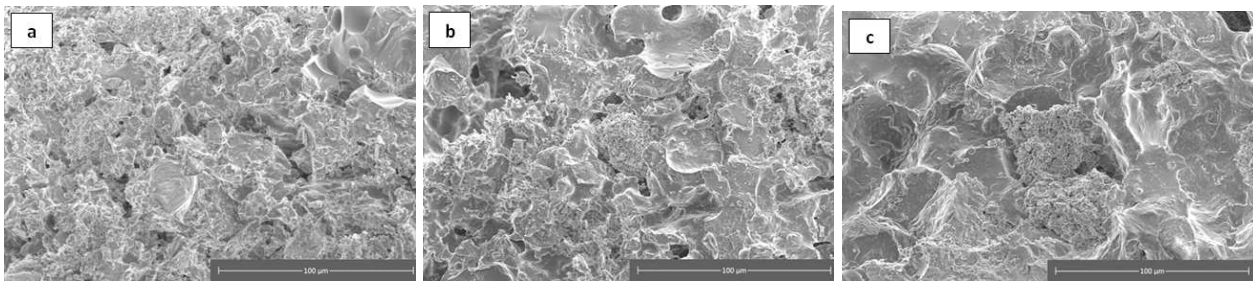

Figure 5. SEM micrographs of supports sintered at different temperatures for 1 h: a) 1125 °C, b) 1140 °C and c) 1150 °C

temperature continues to rise, sintering becomes more dominant, causing particles to consolidate and reduce porosity [2,32]. On the other hand, the PSD curves indicated that the supports had a uniform pore size distribution (Fig. 7), with diameters of the pores ranging from 3.8 to 15 µm. Notably, as the sintering temperature increases, the curve progressively shifts towards the right, suggesting an augmentation in the average pore diameter. In fact, the increase in APS and the uniform PSD were also confirmed by SEM micrographs (Fig. 5).

3.2. Flexural strength of supports

Mechanical strength measurement also has great importance since porous supports should resist the applied pressure during solutions filtration [4,14,31,33]. The effect of sintering temperature on the mechanical propriety of the prepared supports is shown in Fig. 8, where three different stages exist. In the first one, the flexural strength increased from around 6 to 14.5 MPa when the sintering temperature was increased from 1100 to 1125 °C. The second stage was observed in the temperature range from 1125 to 1140 °C in which the flexural strength remains constant (about 15 MPa). The stability of the flexural strength in this temperature range is probably due to stability of the total sample porosity. However, the third stage was observed after 1140 °C and is characterized with sharp increase of the flexural strength up to 25 MPa at 1150 °C. In fact, this increase in the mechanical propriety may be mainly due to the densification of material grains caused by the sintering of the samples. Densification and grain size are generally the main factors that control mechanical strength [2]. In addition, these findings show that the presence of specific phases such as anorthite, gehlenite and wollastonite has strongly affected the mechanical properties of the obtained porous ceramic supports [3].

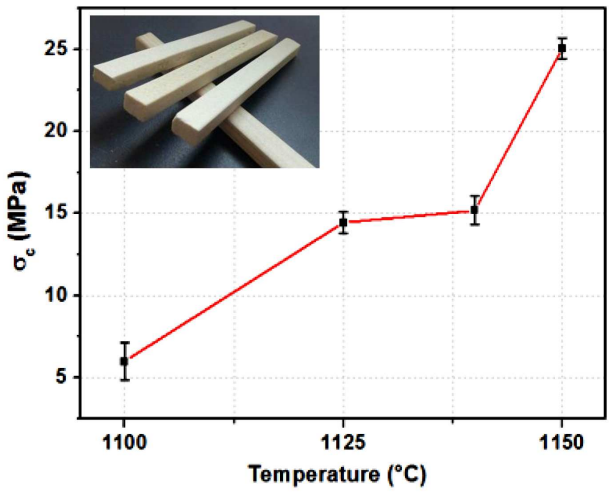

Figure 8. Flexural strength variation as a function of sintering temperature (inset - photograph of the samples used for flexural strength test)

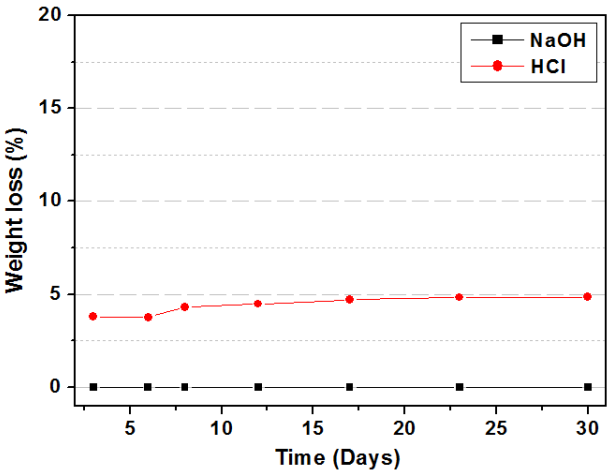

Figure 9. Weight loss of supports sintered at 1140 °C immersed in HCl and NaOH solutions as a function of time

3.3. Chemical resistance of supports

The mass loss of the fabricated support sintered at 1400 °C after being immersed in HCl (pH = 1) and NaOH (pH = 12.5) solutions at room temperature for 30 days was used to evaluate the chemical corrosion resistance. The main remark that can be drawn from the results (Fig. 9) is that these supports present good chemical resistance towards the basic solutions. The mass loss generally remained constant during 30 days. Thus, the mass loss was not large (around 5%) in the first three days for specimens immersed in acid solutions and after that, this loss remained constant and did not exceed 5%. This can be explained by the high alkali content of the considered ceramic compositions. Thus, the prepared supports have good chemical resistance in basic media according to the results of the weight loss during corrosion tests.

3.4. Water permeability and efficiency of supports

One of the most important parameters that describe the separation performance of a membrane supports is its permeability. This parameter is typically used to provide an indication of the capacity of a membrane to process the permeate [2]. According to the Hagen-Poiseuille equation, the water flux through a porous membrane is affected by numerous parameters such as pore size, porosity ratio, pressure difference cross the membrane, membrane thickness and viscosity of water [14,31,33].

Figure 10a shows the variation in water flux with time and pressure for the tubular supports sintered at 1140 °C for 1 h. The most important observations are that the water flux depends on the applied pressure and the stability of the flux was obtained after a few minutes. Moreover, the results indicated that the water permeability of the supports increased with the sintering temperature (Fig. 10b). Specifically, the permeability exhibited small values of about 0.65 and 1.65 $m^3/(h \cdot m^2 \cdot bar)$ for the samples sintered at 1100 to 1125 °C, respectively. However, as the sintering temperature increased to 1140 °C and 1150 °C, the permeability experienced

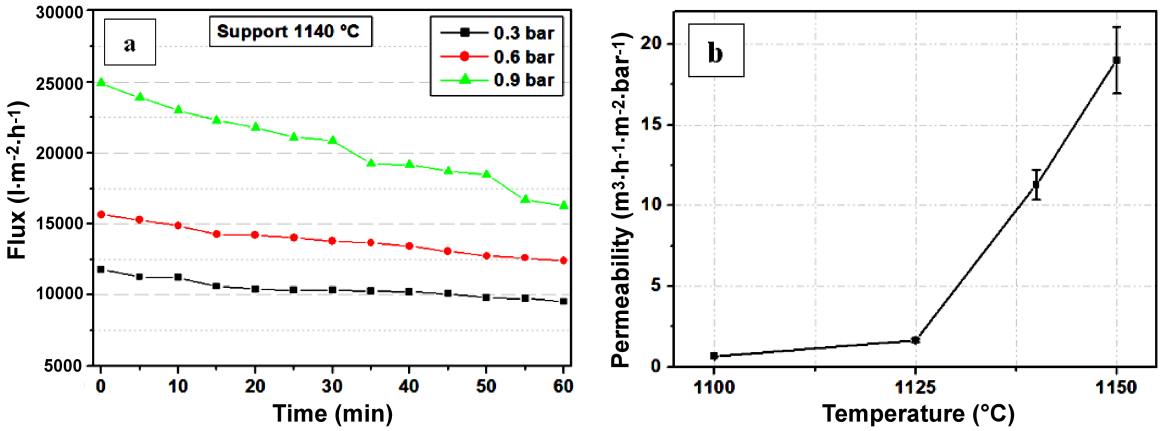

Figure 10. Distilled water flux as a function of time at various working pressures for supports sintered at 1140 °C for 1 h (a) and water permeability of supports sintered at different temperatures (b)

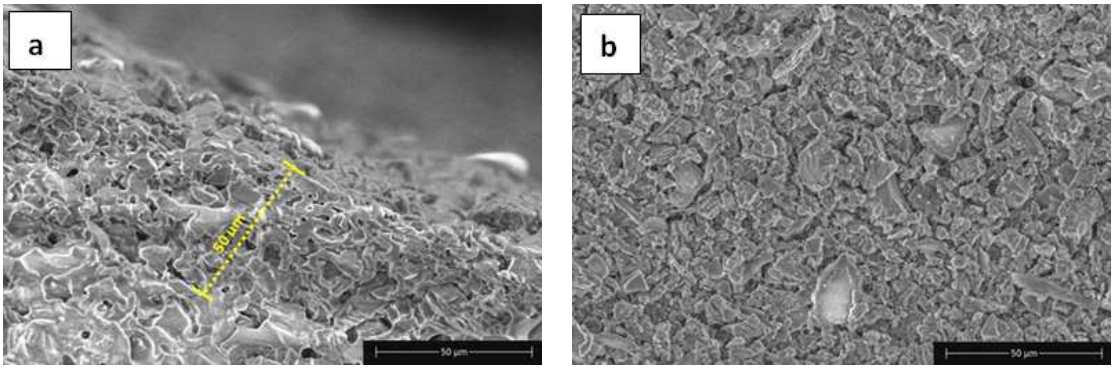

Figure 11. SEM image of the MF membrane: a) cross-section and b) surface

a significant increase, reaching approximately 11.3 and 19 $m^3/(h \cdot m^2 \cdot bar)$, respectively. This increase in permeability can be attributed to the increase in the average pore size, because the porosity ratio was decreased with the sintering temperature.

Finally, the results show that the best conditions to prepare the support were established at sintering temperature of 1140 °C. These supports had interesting characteristics such as a good mechanical strength (25 MPa), a good chemical stability, a high porosity (47%) and permeability of 11.28 $m^3/(h \cdot m^2 \cdot bar)$. The obtained results enable us to conclude that the fabricated membrane supports sintered at 1140 °C are suitable for depositing the membrane layers used in microfiltration field.

3.5. Membrane characterization

SEM micrographs of the MF membrane (surface and cross-section) are shown in Fig. 11. It can be seen that this membrane has a uniform thickness (about 50 µm), a good adhesion with the support (Fig. 11a) and a homogeneous surface without any defects such as cracks (Fig. 11b). Moreover, the results of pore size distribution (Fig. 12) indicate that the membrane has a narrow pore size, ranging from 0.42 to 0.43 µm with an APS of 0.423 µm. The APS value suggests that the membrane is well-suited for application in the field of microfiltration, especially in wastewater treatment.

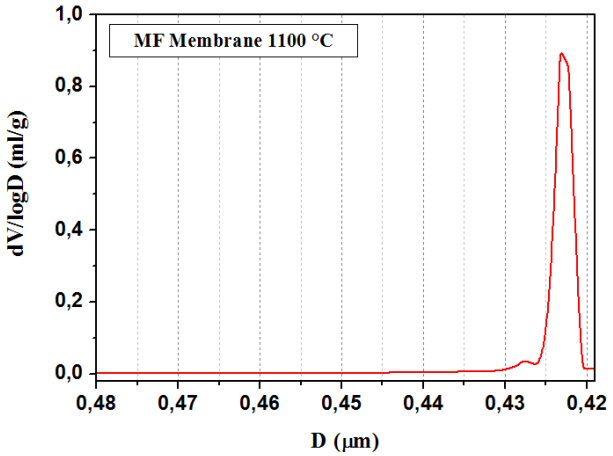

Figure 12. Pores size distribution of the MF membrane

Water flow through the membrane, given as a function of time and mean applied pressure, is shown in Fig. 13. From the results, we can observe an increase in water flux as the applied pressure increases, with a stable flux being obtained after a few minutes. The average water permeability of the membrane is around 1315 ± 50 $l/(h \cdot m^2 \cdot bar)$.

3.6. Membrane efficiency

To estimate the potential of the MF membrane, particularly in water treatment, some physicochemical tests

Table 1. Heavy metal concentrations and some physicochemical characteristics of water sample before and after filtration using the MF membrane

Element	Conductivity [µs/cm]	pH	Turbidity [NTU]	Al [µg/l]	Fe [µg/l]	Zn [µg/l]
Before	785	7.86	173	570.5	336.7	84.59
After	747	7.85	0.3	56.32	107.6	62.87
Rejection rate [%]	-	-	99.9	90.13	68.04	25.68

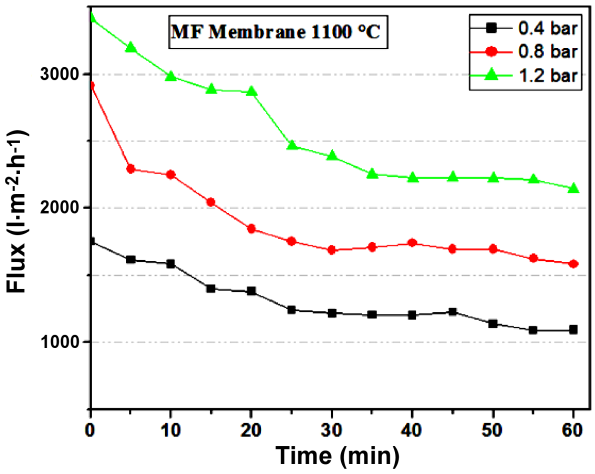

Figure 13. Permeate flux versus time, at three working pressure values, using distilled water for the MF membrane

of water samples were also realized, by measuring pH, turbidity and conductivity during the filtration tests. Table 1 presents some physicochemical characteristics of the water samples which were obtained before and after filtration by using the MF membranes. The results showed that there was no significant change in the pH value of the permeate sample compared to the initial water sample. Furthermore, the MF membrane demonstrated high efficiency in turbidity removal, with a rejection rate of ~99.9% (Table 1). Additionally, a noticeable change in the permeate colour due to the removal of suspended matter was evident (Fig. 14). In contrast, the variation in conductivity value was negligible, as this membrane is not efficient in removing soluble salts from effluents. Moreover, the obtained results indicated that this MF membrane has high efficacy in removing heavy metals, particularly Al, Fe, and Zn, with rejection rates of about 90%, 68%, and 25%, respectively.

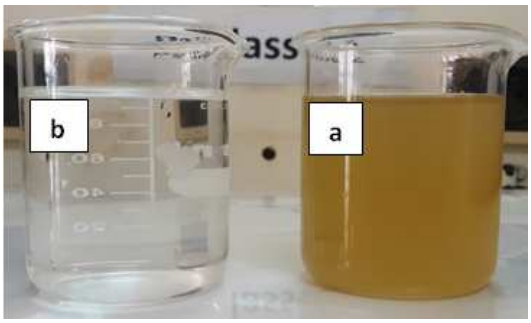

Figure 14. Photograph of water samples: a) before treatment and b) after treatment using the MF membrane

On the other hand, to evaluate the potential of the MF membrane to remove microorganisms from water, some microbiological tests were realized. Figure 15 presents photographs of the VRBG culture medium inoculated by the samples of filtered and unfiltered water after 24 h of incubation. The results showed the absence of *Enterobacteriaceae* bacterial colonies in two tested water samples, indicating the absence of faecal contamination flora.

The separation efficiency of pathogenic bacteria and saprophytic bacteria using the MF membrane was evaluated using PCA culture medium. The removal of bacteria from the sample was indicated by the absence of bacterial colonies on Petri agar plates. From the results of Fig. 16, it is clear that a difference exists between the culture medium containing the filtered and unfiltered water. Bacterial colonies (a dense bacterial lawn) were observed in the culture medium containing the unfiltered water. By contrast, there are no bacterial colonies in the culture medium containing the permeate (filtered) water. In addition, some samples were incubated at room temperature for 3 days to detect the elimination of saprophytic bacteria. Figure 17 presents photographs of filtered and unfiltered water samples inoc-

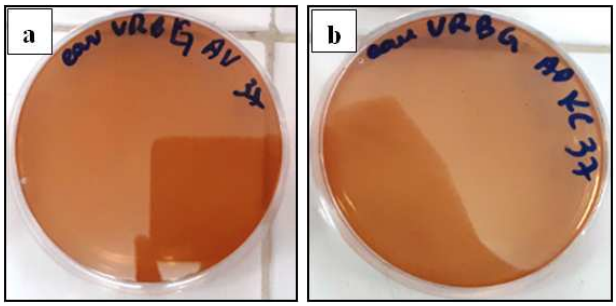

Figure 15. Water samples: a) unfiltered and b) filtered, inoculated on VRBG culture medium, incubated at 37 °C for 24 h

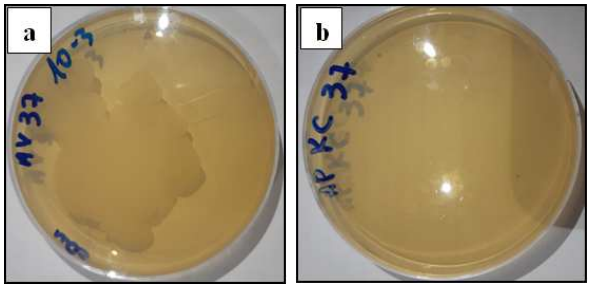

Figure 16. Water samples: a) unfiltered and b) filtered, inoculated on PCA culture medium, incubated at 37 °C for 24 h

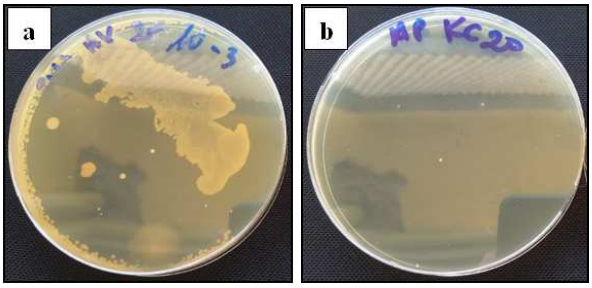

Figure 17. Water samples: a) unfiltered and b) filtered, inoculated on PCA culture medium, incubated at room temperature for 3 days

ulated on PCA culture medium. The results show that there are many bacterial colonies (a bacterial lawn) in the culture medium containing the initial water sample (unfiltered), but on the contrary, the absence of bacterial colonies in the permeate (filtered) water is noticed. The results indicate that the MF membrane has a high efficiency towards removal of these two kinds of bacteria from water.

In recent years, the applications of ceramic membranes for water and wastewater treatment have attracted considerable attention. These membranes have been reported to be thermally stable and resistant to chemicals with a long lifespan, which makes them ideal for the treatment of industrial wastewater and oil/water separation [38]. Among water pollutants, pathogenic microorganisms are one of the most harmful to human health, especially *E. coli* bacteria. *E. coli* bacteria are the most common indicator of faecal contamination in drinking water [39]. Nowadays, ceramic membranes have proven their effectiveness in toxic compound removal from wastewater and retention of *E. coli* bacteria [40]. Our results agree with those reported by Pelagie *et al.* [41]. The results of the use of bio-based ceramic membranes show that the material properties were closely linked to bacterial retention and fouling. The work succeeded in obtaining 100% *E. coli* retention (3.3 log-removal) with the bio-based membrane, calcined at 1000 °C [41].

IV. Conclusions

In this work, ceramic membrane supports with a tubular and flat configurations were prepared by extrusion and roll pressing methods using local low cost raw materials (clay and 30 wt.% calcite). The fabricated supports sintered at 1140 °C had interesting characteristics: flexural strength of 15 MPa, high porosity (about 47%), high permeability (about 11 $m^3/(h{\cdot}m^2{\cdot}bar)$) and good chemical stability. These supports were used for depositing a MF membrane prepared from the same clay using the slip casting method. The MF membrane, sintered at 1100 °C, has following characteristics: thickness of about 27 μm, APS of about 0.42 μm, a narrow pore size distribution and water permeability of 1315 $l/(h{\cdot}m^2{\cdot}bar)$. Furthermore, this membrane showed high performance in eliminating turbidity from water (rejection rate of about 99.9%) and good efficiency in removing heavy metals (rejection rate of about 90% for aluminium). Additionally, to prove the potential of the MF membrane in removing microorganisms from water, some microbiological tests were conducted. The membrane showed good efficacy in rejecting many types of bacteria such as pathogenic flora and saprophytic flora. Finally, these results also enable us to conclude that this low-cost microfiltration membrane is suitable for use in the microfiltration field, particularly for water treatment.

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