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Study of added starch on characteristics of flat ceramic microfiltration membrane made from natural Moroccan pozzolan

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- ✓ ceramic membrane,
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Abstract

This work was conducted to evaluate the starch addition on some physical characteristics of flat ceramic microfiltration membranes made from Moroccan pozzolan. Natural pozzolan was characterized by X-ray fluorescence, X-ray diffraction and Thermo-gravimetric analysis. The starch was added as porosity agent and its amount was varied from 0 to 20 wt.%. Membranes were prepared by dry process using uniaxial pressing method and followed by sintering at temperature of 950 °C. Obtained membranes were characterized by scanning electron microscopy, water porosity, mechanical strength, and water permeability. The experimental results indicate that addition of starch to natural pozzolan increases the porosity, pore size, and permeability of the membrane but accompanied by a decrease of the flexural mechanical strength. The promising aspect of the obtained results allowed using these membranes as filters for clarification of local industrial textile effluent.

1. Introduction

Ceramic porous materials have attracted many researchers in membrane technology due to their important properties such as good mechanical strength, thermal stability, as well as good chemical resistant. Ceramic membranes have been used more and more in different applications like desalination of seawater, wastewater treatment, gas separation and catalysis [1–4].

In elaboration process of ceramic porous membranes, it's crucial in most of the cases to add pore-former agent to raw materials in order to improve essentially the porosity and the permeability. Therefore, decomposition or evaporation or combustion of this pore-former agent during thermal treatment process lead to generation of the porosity in ceramic body [5].

The pore-former agents employed in the manufacturing of ceramic membranes are enormous. Depending on their nature, they could be mineral compounds such as calcium carbonate and dolomite [6] or organic substances as polymethyl methacrylate [7]. They could classify as natural materials as starch [8] and lignite [9] or synthesized product like aluminium hydroxide [10]. Also, they merely could be waste materials for example sawdust [11].

Starch is one of the components frequently used as pore-former agent in the elaboration of porous ceramic membranes. Starch is a natural biopolymer that easily burns out during thermal treatment with low ash residue in the final ceramic matrix. The benefits of used starch as porosity agent include its low cost and its innocuous nature and its environmentally friendly.

Different types of starch are reported in literature as porosity agent in ceramic materials and especially in ceramic membranes. Potato starch was added to kaolin and alumina to prepare low cost microporous ceramic membranes and to study their influence on membrane proprieties [12]. Corn starch was mixed with kaolin and phosphoric acid to develop ceramic membrane supports [13]. The role of starches obtained from potato, pea, maize and wheat on characteristics of low cost ceramic membrane was investigated in term of pore size distribution and permeability coefficient of membrane [14]. Corn starch was added (from 0 to 35 wt.%) to

natural clay and phosphate to elaborate flat ceramic membrane, and investigated the effect of starch addition on water permeability, air permeability, and mean pore size diameter of prepared membrane [15].

In order to reduce the cost of conventional ceramic membranes made from expensive materials like alumina, silica, titania and zirconia. Waste materials and geomaterials could be considered as good alternatives of these oxides [16–19]. The previous works of our laboratory describe utilization of local materials such as clays [20,21], phosphates [22,23], and perlite [24,25] in manufacturing of tubular and flat ceramic membranes.

Natural pozzolans are volcanic rocks mainly composed of silica and alumina. They have been widely used in cement filed because of their pozzolanic effect [26]. A natural pozzolan is chosen in the present work as a raw material for elaboration of ceramic membranes due to its low cost and its abundance in Morocco (Central Middle Atlas).

The aim of this work is the preparation of low cost ceramic flat microfiltration membrane via dry pressing. Moroccan pozzolan was used as a local raw geomaterial and the effect of starch addition (from 0 to 20 wt.%) on distinct characteristics (porosity, pore size diameter, water permeability…) was investigated. The obtained membranes were tested in the filtration of textile effluent generated by Jeans washing process. It should be indicated that membranes without starch were studied in our previous work [27].

2. Material and Methods

2.1. Raw materials

The raw materials used to prepare flat ceramic microfiltration membranes were natural pozzolan extracted from Azrou region which is situated in the Central Middle Atlas in Morocco and a commercial starch ($(C_6H_{10}O_5)n$, Merck KGaA).

2.2. Experimental protocol

The raw natural pozzolan was firstly dried in stove at 110 °C for 24 h then crashed in ball mill (RetschMP 100) and sieved through a sieve of 45 µm. The obtained pozzolan powder was mixed with starch at different weight percentages ranging from 0 to 20 wt.% with a step of 5%. The mixture was introduced in electric mixer at high speed during 15 min for homogenization. The total mass of 5 g of each mixture was spread out in a stainless steel mold of 40 mm in diameter and pressed using uniaxial hydraulic press under pressure of 740 bar during 20 min. The green membranes were later sintered at 950 °C in muffle furnace (Nabertherm L9/13/P320) according to the thermal program shown in Figure 1. The thermal treatment program applied to green membranes is as follow: The temperature increases by a ramp of 3 °C/min from room temperature to 950 °C. There are three holdings of temperature for two hours respectively at 90 °C for elimination of moisture, at 480 °C for calcination of starch, and at 950 °C for sintering.

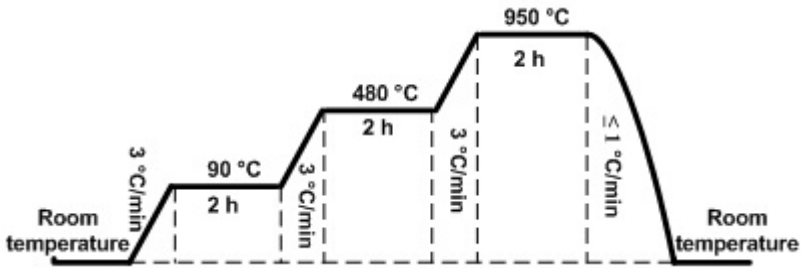

Figure 1: Thermal treatment program

2.3. Filtration test

The dead-end microfiltration mode was applied in filtration test. The experimental equipment made from glass was used for microfiltration at room temperature and at low pressure (from 0 to 0.12 bar). As shown in Figure 2, the membrane, with an effective filtration area of 530 mm^2, was placed in membrane module and the working transmembrane pressure ΔP (bar) was adjusted by height of water between membrane surface and the level of liquid in separating funnel. The pressure gradient across the flat ceramic membrane is given by the following formula (Eq. 1):

$$\Delta P = \rho . g . h \qquad (1)$$

Where ρ is the water density, g is the gravitational acceleration, and h is the height of water.

The water permeability of fabricated membranes was measured using distillated water as testing fluid. The flux J ($L/h.m^2$) and the water permeability L_p ($L/h.m^2.bar$) were calculated according to Eq. 2 and Eq.3:

$$J = \frac{V}{t.S} \qquad (2)$$

$$L_p = \frac{J}{\Delta P} \qquad (3)$$

Where V is the volume of permeate (L), t is the filtration time (h), S the effective filtration area (m^2), and ΔP is the transmembrane pressure (bar).

The prepared membranes were evaluated by microfiltration experiment of industrial textile wastewater which was collected from local textile industry, especially, wastewater of Jeans washing process located in the industrial area of Casablanca, Morocco. Before filtration tests, the raw effluent was sieved through a sieve of 45 µm as pretreatment step. It should be mentioned that the working pressure and the filtration time were fixed respectively at 0.12 bar and 2 h. The parameters such as turbidity, absorbance at wave length of 664 nm and conductivity of permeate were measured every 20 min. Rejection factor of turbidity R_T, color R_{ABS} and conductivity R_C were determined using the following equations: Eq. 4, Eq. 5 and Eq. 6 respectively:

$$R_T = \frac{(T_{effluent} - T_{permeate})}{T_{effluent}} \times 100 \qquad (4)$$

$$R_{ABS} = \frac{(ABS_{effluent} - ABS_{permeate})}{ABS_{effluent}} \times 100 \qquad (5)$$

$$R_C = \frac{(C_{effluent} - C_{permeate})}{C_{effluent}} \times 100 \qquad (6)$$

Where T (NTU), ABS and C (µS/cm) are respectively turbidity, absorbance at 664 nm, and conductivity of effluent and permeate.

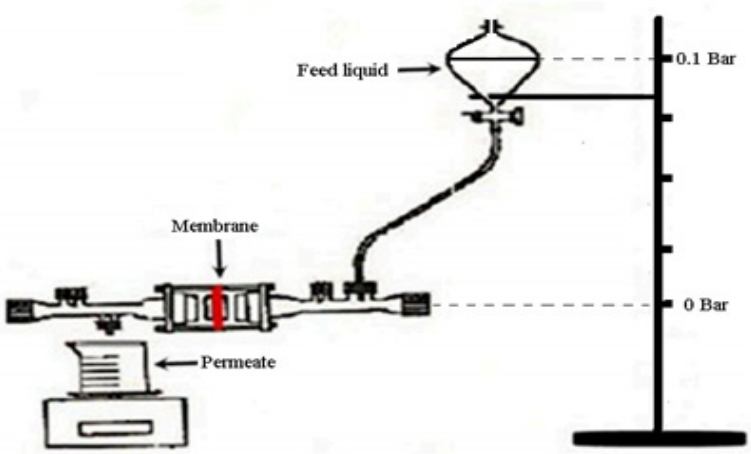

Figure 2: The experimental equipment used for microfiltration at lower pressure

2.4. Analyses

Chemical analysis of natural pozzolan was performed by X-ray fluorescence using sequential spectrometer of X fluorescence based on a scattering of wave length (Bruker S8). Mineralogical composition of natural pozzolan was analyzed by X-ray diffraction with Philips X'Pert PRO using CuKα 1 radiation source (α=1.54060 Å). The gravimetric thermal analysis (GTA) and differential thermal analysis (DTA) were carried out using a thermo-balance of SETARAM type which gives simultaneous curves of DTA and GTA under air atmosphere and αAl_2O_3 as reference from room temperature to 1000 °C at a rate of 10 °C/min. Particle size distribution of pozzolan powder was obtained by dry laser diffraction (master sizer 2000, Malvern Instruments) and the characteristic diameters D10, D50, D90 which are respectively the cut-off particle size below 10%, 50% and 90% of the total particle volume lies were calculated.

The sintered membranes were subjected to various characterization techniques. The measurement of membrane dimensions (diameter and thickness) before and after the thermal treatment was conducted to calculate shrinkage. The microstructure of membranes was observed by scanning electron microscopy (SEM) operating at 10 kV (FEI Company, Quanta 200). The pore size diameter was calculated by image processing of SEM pictures using ImgeJ software (Version 1.44e). The porosity was determined according to ASTM C373-88 method. The flexural mechanical strength was measured in accordance with ASTM C674-88 (Instron 3369).

Textile effluent and permeate were characterized using different instruments. The turbidity was measured by HACH 2100Q Portable Turbidimeter. The absorbance measurement was carried out using ATI UnicamUV2 UV/vis spectrophotometer and the conductivity was analyzed using CyberScan PC 300.

3. Results and discussion

3.1. Characterization of raw natural pozzolan

Chemical composition of natural pozzolan is given in Table 1 by weight percentage (wt.%) of oxides. It shows that natural pozzolan is composed of 65.95 wt.% of SiO_2, Al_2O_3 and Fe_2O_3 superior to 50 wt.% and inferior to 70 wt.%. Therefore, this pozzolan is rightly placed in class C according to ASTM C618.

Table 1: Chemical composition of natural pozzolan

Oxides	SiO_2	Al_2O_3	Fe_2O_3	CaO	MgO	SO_3	K_2O	Na_2O	P_2O_5	L.I*
wt.%	40.76	13.26	11.93	10.38	9.24	0.21	1.36	2.99	0.98	4.09

*Loss on ignition at 1000 °C

The X-ray diffraction patterns of natural pozzolan and sintered pozzolan at 950 °C are shown in Figure 3. For both XRD patterns, it should be mentioned that pozzolan materials are mainly composed by an amorphous phase corresponding to the presence of a large diffused halo. This amorphous phase is probably due to the rapid cooling of the volcanic magma after its eruption. The present of peaks in XRD pattern is associated to the presence of some small crystalline phases which are dispersed in amorphous phase. The crystalline phases presented in natural pozzolan are hematite (Fe_3O_4), augite ($Ca[Mg, Al,Fe]Si_2O_6$) and forstirite ($[Mg_{1.60};Fe_{0.38}]SiO_4$). The same crystalline phases are encountered in sintered pozzolan, except for the forstirite where the iron atom is substituted by magnesium atom to form Mg_2SiO_4 during thermal treatment.

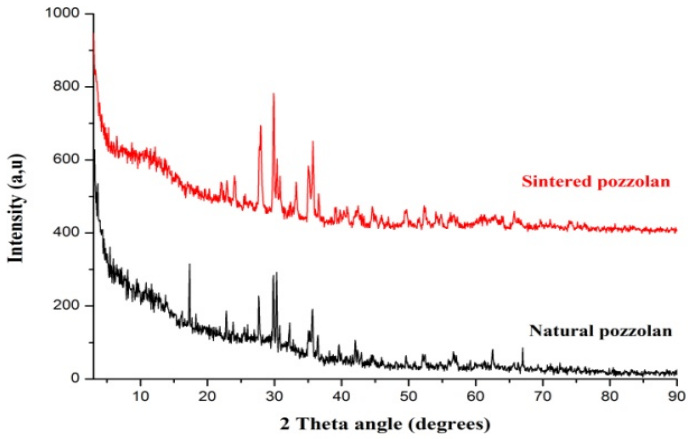

Figure 3: The X-ray diffraction patterns of natural pozzolan and sintered pozzolan at 950 °C

TGA and DTA curves of natural pozzolan are shown in Figure 4. As seen in the figure, the total weight loss of pozzolan is found to be about 4.88 wt.%. The first weight loss of 2.20 wt.% begins from 25 to 142 °C that is attributed to the removal of free and adsorbed water. This phenomenon is accompanied with endothermic behavior. The second weight loss of 1.80% exothermic character starts from 142 to 360 °C. It corresponds to the departure of CO_2 resulting from the combustion of organic impurities. Finally, the third weight loss of 0.88 wt.% with endothermic nature is observed starting from 360 to 800 °C. It is associated with the dehydroxylation of hydroxyl groups structural [28].

Particle size of starting materials strongly affects on the porosity and the pore size of the membrane. The particle size of pozzolan powder used for preparation of ceramic membranes was characterized by laser granulometry. Particle size distribution is shown in Figure 5 and indicates that particle diameters are varied from 1 to 70 µm. Even though the powder was sieved through a sieve of 45 µm, low amount of particles have superior to 45 µm which could be explained by agglomeration of particles. Furthermore, the results indicate that D10, D50 and D90 are respectively 2.12, 14.59 and 38.73 µm.

3.2. Characterization of membranes

The sintering temperature of membranes was optimized in previous work [27] and it was set at 950 °C. After sintering, the resulting membranes changed their color from grey to dark brown (Figure 6). Also we can easily check that the obtained membranes are strength and free of defects. During thermal treatment, starch is burnt out and left a porous structure in ceramic body. The calcination of starch at temperature around 500 °C [16] leads to a small amount of ash less than 0.4 wt.% by weight as result of the decomposition of organic matter to carbon dioxide and water vapor. The starch ash is generally composed of sodium, potassium, calcium

and phosphorus. In addition, the low amount of ash do not seem to exert a significant effect on membrane properties.

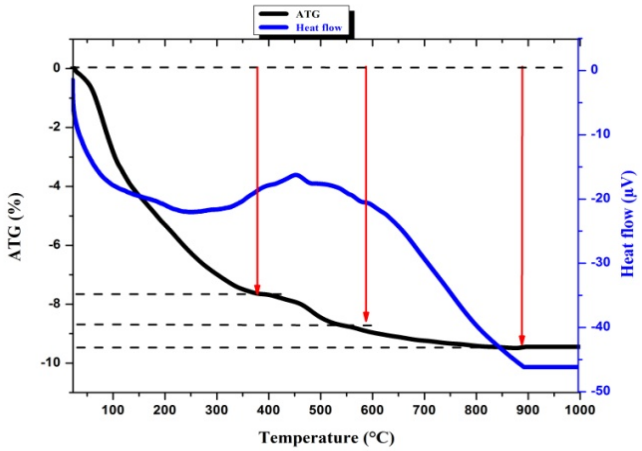

Figure 4: TGA and DTA curves of natural pozzolan

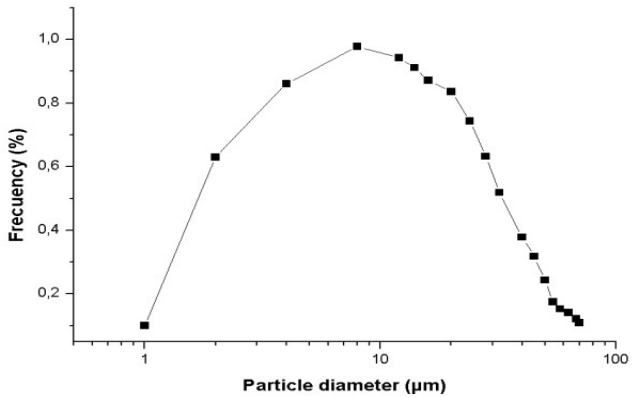

Figure 5: Particle size distribution of pozzolan powder

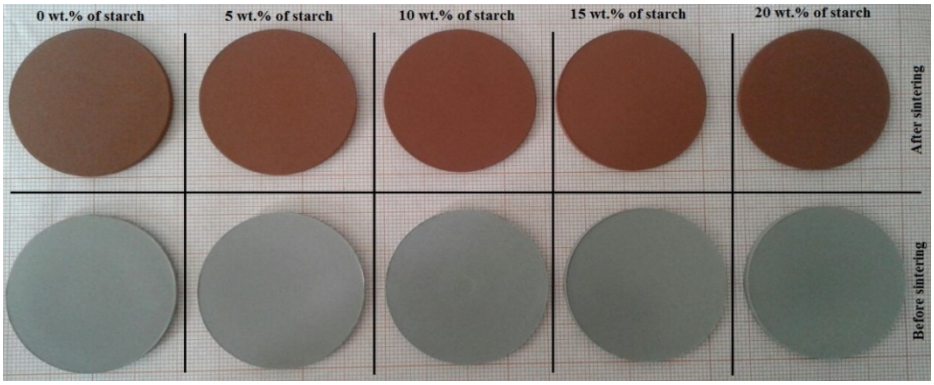

Figure 6: Flat membranes before and after sintering at 950 °C

Figure 7 shows the shrinkage of membrane as a function of starch addition. For the membrane without starch, the value of shrinkage is found to be 4.95% which might be mainly due to loss of water. By adding starch, the shrinkage weakly increases by 0.93% as the added starch increases from 0 to 20 wt.%. This phenomenon is probably due to loss of porosity agent during thermal treatment. As clearly concluded, the shrinkage is managed by moisture rather than starch. The porosity is a preliminary characterization of elaborated membranes. The evolution of porosity in ceramic matrix with starch addition is presented in Figure 8.

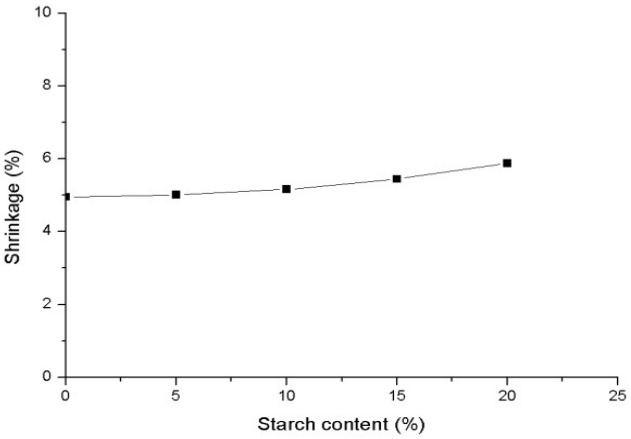

Figure 7: Influence of starch content on shrinkage of membranes

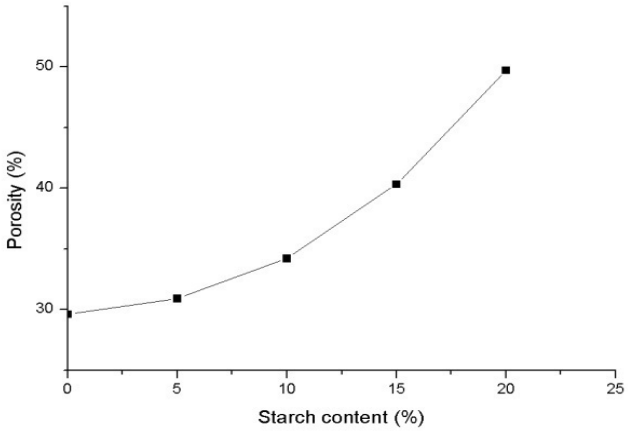

Figure 8: Effect of starch addition on porosity of membranes

As shown in the figure, the porosity is exponentially increased from 29.6 to 49.7% as the added starch increases from 0 to 20 wt.%. It's known that increase of the porosity automatically contributes to enhance the permeability of the membrane.

The pore size diameter of membranes was measured in SEM images using ImageJ software (Version 1.44e). The pores are supposed circular in order to calculate their area average pore diameter using Eq. 7:

$$d = \sqrt{\frac{\sum_{i=1}^{n} n_i \times d_i^2}{\sum_{i=1}^{n} n_i}} \qquad (7)$$

Where n is the number of pores found in SEM image and d_i is the pore diameter (µm) of i-th pore. The average pore diameter of membrane as function of added starch is plotted in Figure 10. The results indicate that the average pore diameter significantly increases from 2.20 to 4.83 µm with increasing of starch addition from 0 to 20 wt.%.

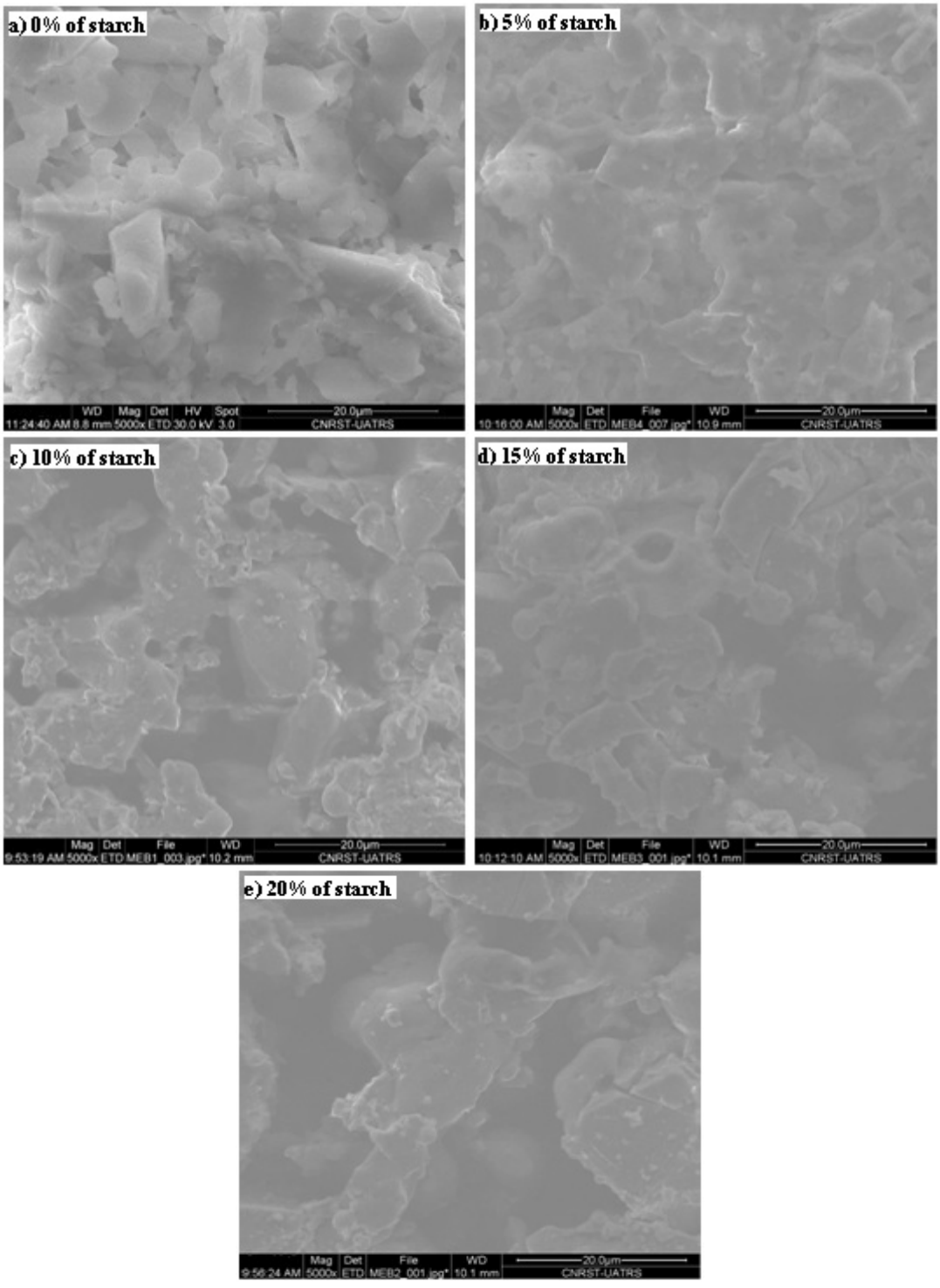

Figure 9: SEM images of membranes at different starch contents

It should be mentioned that pore sizes in ceramic membrane formed by dry process are less than pore sizes formed by wet process using the same amount of starch. This phenomenon may be explained by swilling of starch in the presence of water. In addition, the pressing may contribute to the size reduction of intergranular pores between particles.

The effect of starch on mechanical strength of flat ceramic membranes is illustrated in Figure 11. In contrast to porosity and pore size findings, the starch addition has a dramatic effect on mechanical strength of the membrane. The flexural strength exponentially decreases from 18.58 to 10.24 MPa while starch amount increases of from 0 to 20 wt.%. This reduction in mechanical resistance is explained by the increase of pore volume in ceramic body. The same results were reported in previous study where effects of starch addition on tubular membrane substrate prepared by wet route [8]. Furthermore, the highest mechanical strength is found to be associated with membrane without added starch. Despite the fact that membrane of 20 wt.% of starch content had the lowest mechanical strength it exhibits enough resistance to support the applied hydraulic pressure during microfiltration tests.

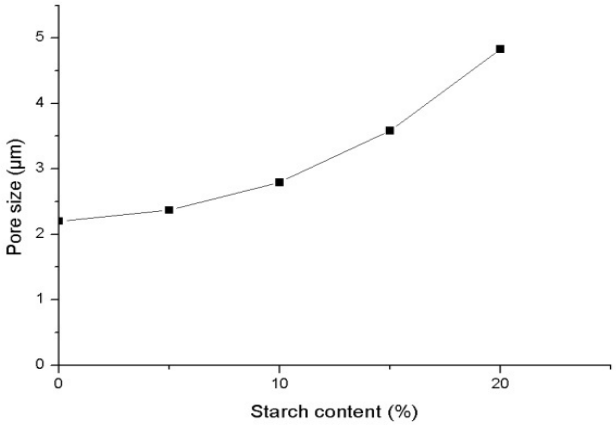

Figure 10: Effect of starch addition on mean pore size diameter membranes

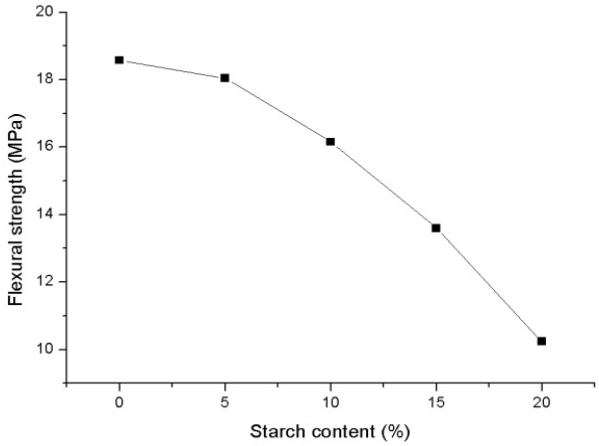

Figure 11: Influence of starch addition on flexural strength of membranes

3.3. Filtration test

Before any microfiltration test, the permeability of the membranes was determined using distillated water. Figure 12 presents the effect of starch addition on water permeability of membranes. From this result, it can be inferred that the permeability increases from 889 to 2274 L/h.m^2.bar as the added starch content increases from 0 to 20 wt.%. This raise of permeability is directly related with the increase of number of pores and pore diameter in ceramic body by adding starch to raw materials. As mentioned earlier, the increase of porosity is an effective way to improve membrane permeation. Therefore, the more permeability is higher, the more membrane can treat important volume of wastewater during reduced time [1].

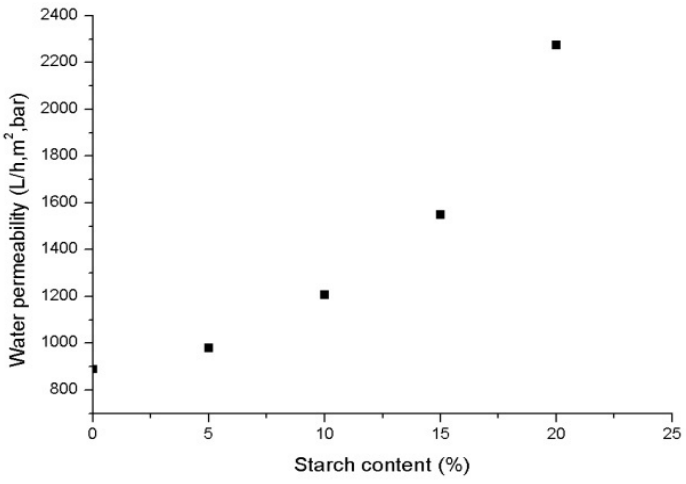

Figure 12: Water permeability of membranes as function of added starch

The wastewaters of textile industries are generally colorful and charged with suspended matter. Thus, they require appropriate treatment in order to remove the pollution. In this study, the textile effluent generated by Jeans washing process was used to evaluate the performance of prepared membranes. Firstly, the textile effluent was characterized in terms of turbidity, conductivity and absorbance (Table 2). Figure 13 presents the variation of permeate flux textile effluent for membranes with different percentages of starch during 2 h of filtration and under pressure of 0.12 bar. From this figure it can be seen that permeate fluxes continuously decrease with the filtration time. This decline can be explained by deposition of suspended particles on membrane surface. Rejection factor of turbidity, color and conductivity of membranes are reported in Table 2. The turbidity of effluent was as high as 93 NTU, indicating relatively the presence of high suspended particles. The rejection ratio of turbidity weakly decreases from 99.1 to 95.3% when the percentage of added starch varies from 0 to 20 wt.%. From color intensity of feed and permeates of industrial effluent measured at λ_{max} = 665 nm, it can be seen that all membranes remove only between 16.0 and 21.4% of soluble dyes. This color reduction is based on the specific affinity more than the pore size diameter of membrane [15]. Also it should be noted that color reduction gradually increases with the filtration time because of the deposition of suspended particles on the membrane surface to thin layer which could play as additional membrane layer. Thus, enhancement of permeate quality. The value of the conductivity of textile effluent is 820 µS/cm. This value is slightly greater than the city water value which corresponds to 780 µS/cm. Non significant conductivity rejection (less than 1.8 %) was obtained for all membranes. This result indicates that the prepared microfiltration membranes are not able to stop ions.

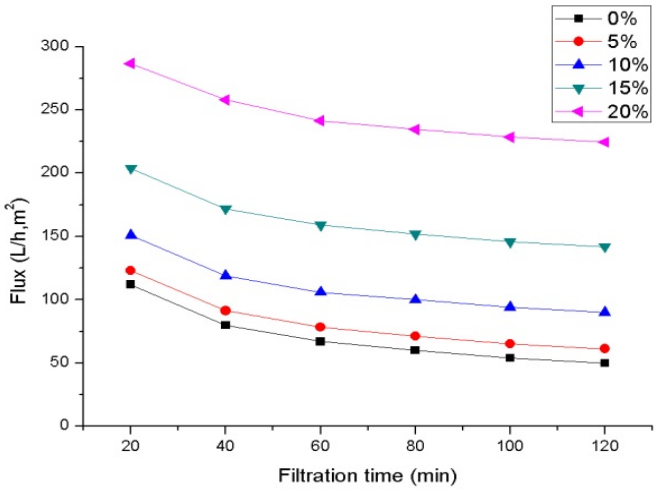

Figure 13: Permeate flux vs. filtration time for membranes with different starch contents

Table 2: Textile effluent analyses and rejection factor of turbidity, color and conductivity of membranes

Sample	Raw textile effluent	Membrane rejection (%)				
		0 wt.%	5 wt.%	10 wt.%	15 wt.%	20 wt.%
Turbidity (NTU)	93	99.1	98.6	98.2	97.4	95.3
Absorbance at 665 nm	0.103	21.4	20.2	18.7	16.8	16.0
Conductivity (µS/cm)	820	1.8	1.7	1.8	1.6	1.7

Conclusion

In this work, starch was added to natural pozzolan as a pore-former agent, to prepare flat ceramic microfiltration membranes. These membranes were elaborated by dry pressing and sintered at temperature of 950 °C. The effect of the starch content on some physical properties of the elaborated membranes was investigated. The addition of starch from 0 to 20 wt.% to ceramic membrane increases the porosity from 29.6 to 49.7%, pore size from 2.20 to 4.83 µm and water permeability from 889 to 2274 L/h.m^2.bar. However, the opposite behavior was observed for mechanical strength that is decreases from 18.58 to 10.24 MPa while starch amount increases of from 0 to 20 wt.%. Finally, the filtration results of textile effluent show the ability of these membranes to remove almost of turbidity (more than 98.2% of turbidity existing in textile effluent especially with the membranes containing from 0 to 10 wt.% of starch), less than 20 % of soluble dyes and less than 1.8% of conductivity.

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