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Manufacturing of tubular ceramic microfiltration membrane based on natural pozzolan for pretreatment of seawater desalination

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ABSTRACT

This work describes manufacturing of low-cost tubular ceramic microfiltration membrane made from natural pozzolan. Firstly, the tubular ceramic support was prepared by extrusion technique followed by sintering at low temperature of 950 °C. The extruded paste is essentially composed of pozzolan powder (< 200 μm), clay (< 68 μm), some organic additives and water. This formulation is conducted to obtain a ceramic support with uniform porous structure and high water permeability. Secondly, the membrane layer was prepared by cross-flow filtration of suspension containing the pozzolan powder (< 45 μm), polyvinyl alcohol and water, thereafter the membrane was dried and sintered at 950 °C. Scanning electron microscope analysis showed that the membrane has homogenous structure and is free from any defect. In addition, the average pore size and water permeability parameters were respectively 0.36 μm and 1444.7 L/h·m^2.bar.

Manufactured membrane is expected to have challenge application in pretreatment of seawater for desalination. Membrane performance was evaluated by filtration of raw seawater in the aim of reducing its turbidity and its chemical oxygen demand. Experimental results showed a high rejection of turbidity (98.25%) as well as a good retention of chemical oxygen demand (70.77%).

1. Introduction

The ceramic membranes have attracted the attention of both academic and industrial researchers due to their interesting proprieties (high thermal stability, chemical inertness, good mechanical resistance and long lifetime) compared with organic membranes [1]. Generally, ceramic membranes are made from industrial oxides such as alumina and silica whish are severely restricted due to their high cost. Moreover, they very often need the high sintering temperature that increase energy consumption [2]. On the other word, low-cost raw materials are very wanted in manufacturing of ceramic membranes for the reason that reducing their price. Geomaterials and waste materials may be used as starting materials [3–5].

In context of preparing of ceramic membrane from local geomaterials, plenty works have reported in bibliography. Local clays were used to develop flat and tubular ceramic membranes for microfiltration process [6,7]. Natural phosphates were used to prepare low cost ceramic membranes, and applied for removal of bacteriological pollution presented in water and textile effluent [8,9]. Perlite was selected as raw material for elaboration of plane and tubular configuration of membranes for industrial wastewaters treatment [10,11]. Additionally, other geomaterials were successfully studied as starting materials for fabrication of ceramic membranes such as zeolite [12], cordierite [13] and kyanite [14].

Preparing of homogeneous membrane (support and membrane layer) made from materials of the same nature presents a serious challenge in term of optimization of sintering process, avoidance microstructure defects as well as obtaining good adherence of membrane layer on the support [15,16,12]. The tubular ceramic membranes are essentially composed of support and membrane layer. The tubular supports can be shaped by extrusion or isostatic pressing techniques [11,17]. The membrane layers were generally prepared by slip-casting [18], dip-coating [19], and spin-spraying [20]. The cross-flow filtration [21] may be considered an alternative coating approach to prepare membrane layer on inner surface of tubular support. This approach is based on filtration of selected suspension. As a consequence, solid particles accumulate on the support and growth to make easily adherent layer.

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Microfiltration process has widely applied in many industrial applications including producing of drinking water and treatment of wastewater for the reason that it can operate at high flux with law turbidity in filtrated water [2,22]. Production of drinking water by seawater desalination becomes more and more a worldwide preoccupation, especially in countries that suffer from few rainfalls like Morocco. The efficiency and lifetime of seawater desalination installation require a pretreatment that prevents or minimizes the bioencrassage, scaling and clogging of the reverse osmosis membranes [23,24]. The classical methods (i.e. the coagulation and flocculation processes) for pretreatment of seawater for desalination generate a huge amount of sludge that must be eliminated later [25]. Microfiltration membrane could substitute conventional pretreatment processes like coagulation and flocculation that are more expensive [26].

The natural pozzolan is selected as a raw material to prepare ceramic microfiltration membrane because it is a cheaper and an environment-friendly raw material, and Morocco has large reserves of pozzolan. Morever, The chemical composition of the pozzolan (especially rich in MgO and SiO_2, $Al2O_3$) and its low quantity in lime fraction lead to membrane with good chemical and mechanical resistance compared with clay membranes.

This work describes manufacturing of tubular ceramic microfiltration membrane made from natural Moroccan pozzolan. The support was prepared by extrusion method and the membrane layer was deposed on inner surface of tubular support by cross-flow filtration process. The obtained membrane was characterized and applied for filtration of raw seawater as pretreatment for desalination.

2. Experimental

2.1. Raw materials

In this work, the natural pozzolan (collected from Azrou region (Morocco)) is used as starting material for elaboration of tubular microfiltration membrane without any chemical treatment. The raw pozzolan was dried at 105 °C for 24 h, crushed in ball mill and sieved. Fraction inferior to 200 μm was used for elaboration of tubular support and fraction inferior to 45 μm was used for preparation of microfiltration layer. This geomaterial was characterized and detailed in previous work [27]. The chemical composition of natural pozzolan given in weight percentages of major oxides is as follow: 40.7 wt% of SiO_2, 13.4 wt% of Al_2O_3, 12.1 wt% of Fe_2O_3, 9.0 wt% of MgO and 8.4 wt% of CaO. The additive materials used in manufacturing of tubular support were fine clay of Fès (Morocco) [28], starch (Merck KGaA), Amijel derived from starch (Cplus 12072, Cerestar) and Methocel derived from methycellulose (The Dow Chemical Company). The chemical products used in preparation of microfiltration membrane were polyvinyl alcohol (PVA) (Rhodoviol 25/140, Prolabo) and acetone (Solvachim).

2.2. Elaboration of tubular support

A monocanal tubular support of 10 mm in exterior diameter, 2 mm in thickness and 150 mm in length was obtained by the extrusion process. The composition of solid phase is composed of 78 wt% of pozzolan (≤ 200 μm), 10 wt% of clay as plasticizer, 6 wt% of starch as porosity agent, 3 wt% of Amijel as binder and 3 wt% of Methocel as plasticizer. The powders mixture was homogenized using electrical mixer at high speed during 15 min. Then, 22 wt% of distilled water relative to total mass (water and solid mixture) was gradually added to the solid mixture for the formation of desired plastic paste. The obtained paste was conserved in closed container for aging during 24 h in order to reach homogeneity and improve the quality of paste by migration of organic additives and water. Then, the paste was shaped into a tubular configuration support using ram extruder, and it was dried and sintered in a programmable furnace.

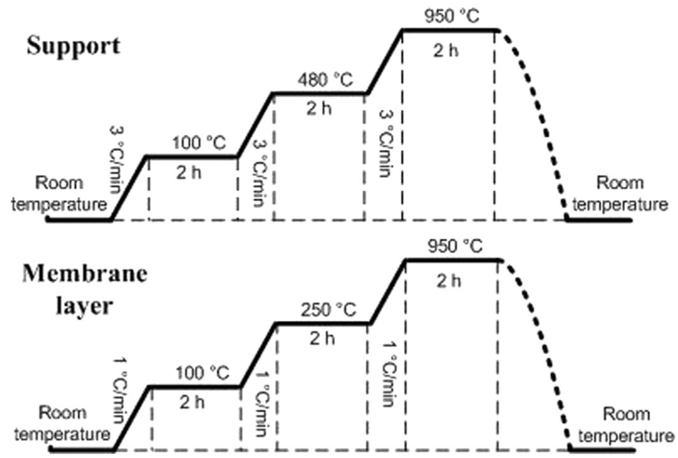

Fig. 1. Thermal treatment applied for support and membrane layer.

2.3. Preparation of the microfiltration membrane

The microfiltration membrane was deposed on inner surface of tubular support by cross-flow filtration technique. The suspension of pozzolan was prepared by mixing 10 wt% of pozzolan powder (≤ 45 μm) and 90 wt% of distillated water containing 3 wt% of PVA as binder and 1 wt% of acetone as antifoaming using magnetic stirring at 250 tr/min during 30 min. After 4 h of rest, the supernatant was carefully recovered by siphoning and used as a feed for cross-flow filtration coating at pressure of 0.5 bar during 30 min. The green membrane was dried and sintered.

2.4. Thermal program

The thermal treatment is an extremely important step in the elaboration ceramic membrane process which is generally composed by two parts: drying and sintering. The thermal program applied in this work is described in Fig. 1. The support was dried overnight in a rotary dryer with 5 tr/min in order to get uniform dying at temperature inferior to 40 °C. The sintering process included temperature plate at 100 °C for elimination de residual moisture, plate at 480 °C for burn out of organic additives and plate at 950 °C for sintering. The sintering temperature was previously optimized and fixed at 950 °C [27]. The heating rate and time length of temperature plates are respectively 3 °C/min and 2 h for each plate. The same thermal treatment was applied to membrane layer, except the plate at 480 °C was changed by plate at 250 °C to eliminate completely the PVA, and heating rate was decreased to 1 °C/min that is needed to avoid the formation of cracks on microfiltration layer.

2.5. Filtration test

The tangential filtration experiment was carried out using a laboratory pilot made from stainless steel. The schematic diagram of microfiltration plant is shown in Fig. 2. It is essentially made up of circulation pump, feed tank of three liters, air compressor, membrane model and manometers. Transmembrane pressure was varied from 0 to 2 bar using pressure regulator. The filtering surface area is about 24.17 cm^2 for all filtration experiments. It should be noted that three membrane samples were used for flirtation test in order to confirm the reproducibility of experimental results and all values given in filtration part are arithmetic average. Also, all filtration experiments were conducted at room temperature (21 ± 1 °C).

Before each filtration test, the support and microfiltration membrane were conditioned in distilled water for at 24 h in order to reach faster stable flux at the beginning of the filtration experiment. Water permeability of support and membrane were measured using distilled

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