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Flat ceramic microfiltration membrane based on natural clay and Moroccan phosphate for desalination and industrial wastewater treatment

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ABSTRACT

A new microfiltration ceramic membrane has been developed by uniaxially compressing natural Moroccan materials: red clay of Safi and natural phosphate of Youssoufia, which is rich in organic matter. The natural phosphate can be beneficial to porosity formation. The effects of natural phosphate on the morphological and flexural strength of the flat disks sintered at 1100 °C were studied to identify the optimal mass ratio of clay and phosphate via X-ray diffraction, scanning electron microscopy and flexural strength testing. The optimal composition of the flat ceramic membrane contained 40 wt% natural phosphate, which led to a linear thermal expansion coefficient less than $13.42 \times 10^{-6}\,°C^{-1}$, flexural strength of 17.5 MPa and Young's modulus of 40.12 GPa. The average pore diameter and water permeability of the obtained ceramic membrane were respectively 2.5 µm and 928 L/(h·m^2·bar). The microfiltration membrane efficiency was evaluated by filtering three feed solutions types: a tannery beamhouse effluent, a raw seawater and a synthetic solution of aluminium chloride (at high pH). The obtained ceramic membrane exhibited superior turbidity removal efficiency for all studied feeds: effluent (99.80%), seawater (99.62%) and synthetic solution (99.86%). Moreover, this microfiltration membrane showed good microstructure and mechanical stability after filtration experiments.

1. Introduction

Environmental protection has become a critical social issue, and its consideration by all enterprises is necessary for legal and economic reasons [1]. Some of the critical environmental issues are water pollution, industrial emissions of toxic substances, mercury and other toxic heavy metals, and oil in the oceans [2].

The microfiltration membrane technique is an effective solution to this problem and is largely acknowledged as an effective separation method in many economic sectors such as food industries, chemical industries, seawater desalination, and wastewater treatment [3–5].

Due to good thermal and chemical stability, better mechanical resistance, minimal pollution impact, and long-life performance, mineral membranes are preferred in the professional environment over organic membranes [6,7].

Mineral membranes are composed of a porous ceramic support, generally based on alumina or silica. However, a new support made from natural minerals has been considered in recent times due to their low cost [7,8]. Membrane selectivity is achieved by the deposition of a porous thin layer of inorganic oxides such as ZrO_2 and TiO_2. The deposition process should be carefully selected to ensure the separation of solutes through suitable pore radius, while the support provides the mechanical strength [9,10].

From a technical perspective, several authors have investigated the development of low-cost ceramic membranes for microfiltration based on natural materials. Achiou et al. [11,12] manufactured and characterized flat and tubular ceramic microfiltration membranes made from natural Moroccan pozzolan. Majouli et al. [13] elaborated of new tubular ceramic membrane from local Moroccan Perlite for the treatment of industrial wastewaters.

Bouazizi et al. [14] developed a new ceramic membrane from Moroccan bentonite for the microfiltration of industrial wastewater. In

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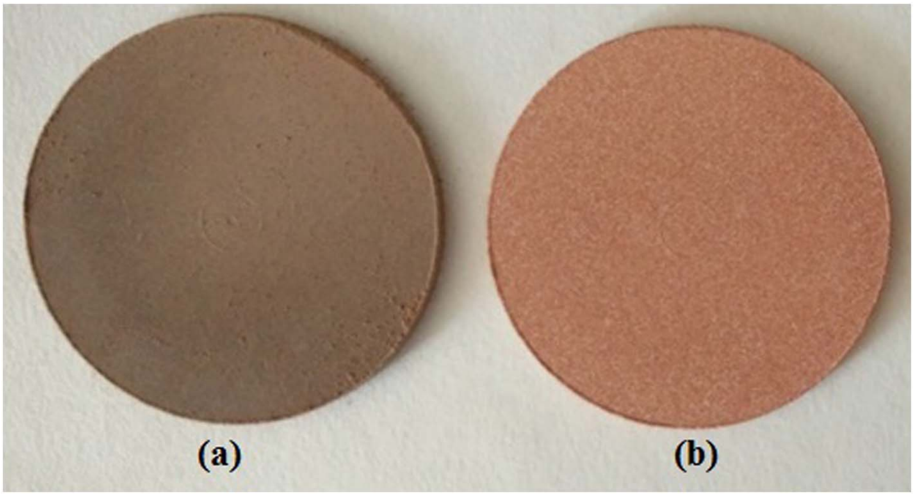

Fig. 1. Images of obtained ceramic membrane: Before (a) and after (b) sintering.

another work [15], they described the development of a new flat ceramic membrane on ceramic support made from bentonite and micronized phosphate for microfiltration and ultrafiltration.

Following the same perspective, we focused on the development of a low cost, porous ceramic support based on two Moroccan natural materials red clay (as a precursor) and natural phosphate (NPh) (as a pore-forming agent). In addition, we demonstrate that the addition of natural phosphate increases the apparent porosity of the prepared material, which leads to a decrease in its tensile strength [16].

Another approach is reported in this paper. The primary purpose of this study is to examine further the effect of natural phosphate on the characteristics (dilatation, pore size, flexural strength, and crystalline phase formation) of flat ceramic microfiltration membrane made from the same materials (clay and natural phosphate) through a similar preparation method, as described in our previous work. The second purpose is to evaluate the performance of microfiltration ceramic membrane, prepared by mixing an optimum amount of clay (60 wt%) and natural phosphate (40 wt%), when applied to the treatment of tannery beamhouse, raw seawater and synthetic aluminium chloride (at high pH) solution.

2. Materials and methods

2.1. Raw materials

The raw materials used to prepare ceramic membrane were Moroccan red clay (RC) quarried from the Safi region and natural phosphate extracted from Youssoufia (Morocco). The major chemical composition of red clay given in weight percentages is as follow: SiO_2 (52.79 wt%), Al_2O_3 (17. 44 wt%), Fe_2O_3 (5.08 wt%). The natural phosphate is mostly rich in P_2O_5 (19.94 wt%) and CaO (39.34 wt%). It also contains SiO_2 (9.26 wt%) and low quantities of Al_2O_3 (0.58 wt%) and Fe_2O_3 (0.28 wt%). The characteristics of raw powders were detailed in our previous paper [16]. The phosphate was chosen as a natural pore-forming agent due to its high content of organic matter, which allowed us to dispense with synthetic additives to create pores [17].

2.2. Powder characterization

Phase identification of the starting powders was carried out in an X-ray powder diffractometer (D2 Phaser) in Bragg-Brentano fixed sample theta-theta geometry using $CuK_\alpha = 1.5406$ Å (40 kV, 40 mA) radiation in the 2θ range from 10° to 100° with a step width of 0.02°.

Dilatometric analysis was carried out by a Setsys evolution vertical dilatometer (Setaram).The samples were prepared from cylindrical pellets, and the surfaces of all the samples were flattened using abrasive SiC discs. The sample dimensions were 6 × 7 × 12 mm. The measurements were taken under dry air atmosphere up to 1100 °C with a heating rate 5 °C/min.

2.3. Preparation of the ceramic membrane

To study the effect of natural phosphate addition (10, 20, and 40 wt %) on the characteristics of ceramic membranes, samples were prepared under the same condition. The dried homogeneous mixture of the clay and phosphate (< 150 μm) was uniaxially compacted under 150 MPa in rectangular and cylindrical moulds forming flat disks, which were then sintered in a programmable oven (Nabertherm L9/13/P320), taking into consideration the DTA/TG results reported in our previous work [16].

Thermal cycling was performed in four steps: (1) annealing at 250 °C for 4 h to ensure the evaporation of residual water; (2) annealing at 450 °C for 1 h to allow thermal degradation of organic matter present in NPh; (3) annealing at 750 °C for 1 h to promote the dehydroxylation of clay minerals and the decomposition of mineral carbonates, and final recrystallization annealing (4) at 1100 °C during 2 h (Fig. 1).

2.4. Ceramic membrane characterization

The XRD analysis of sintered samples was carried out by INEL CPS 120-Curved Position Sensitive diffractometer using CuK_α radiation (λ = 1.5406 A°) (37.5 kV, 28 mA) for an acquisition time of 45 min. The crystalline phases were identified by the JCPDS (Joint Committee on Powder Diffraction Standards) database.

A scanning electron microscope (SEM; Hitachi SC 2500) was used to visualize the microstructure of the developed material in secondary electron mode. The accelerating voltage and magnification range varied from 10 to 30 kV.

The pore size distribution of the ceramic membranes was measured by analysing several SEM images using ImageJ software. A large number of pores (over 400) were examined in order to determine the distribution and the average pore size (nm), as calculated using the following Eq. (1):

$$d = \sqrt{\frac{\sum_{i=1}^{n} n_i \cdot d_i^2}{\sum_{i=1}^{n} n_i}} \tag{1}$$

where n is the number of pores, and d is the pore diameter of each pore.

The flexural strength of the ceramic membranes was measured via the three-point bending test. The sample dimensions were 4.5 × 20 × 50 mm. The tensile strength measured by the Brazilian