



# Composite ceramic membranes from natural aluminosilicates for microfiltration applications

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## Abstract

This work concerns to the development and characterisation of support, active layer and tubular composite membranes (CM) from natural aluminosilicates as principal components (clay, bentonite, feldspar, quartz, alumina). The selection of these raw materials was primarily based on their low cost and they are locally produced. In the substrates preparation, the effect of materials compositions, additives, particle sizes, paste rheological properties, and drying-sintering temperatures was investigated. The consolidated ceramic substrates were characterised by SEM, DTA–TG, X-Ray diffraction, Hg intrusion, mechanical resistance, and water flux measurements. Extrusion has been used as the forming process of tubular support. The CM was fabricated depositing a thin active layer by slip-casting method on the support. The CM sintered at 1200 °C showed the best structural characteristics, porosities of 50%, active layer pore size between 0.08 and 0.55 μm. The CM hydraulic permeabilities (10–274 L/h m<sup>2</sup> kPa) were comparable and greater than several inorganic commercial membranes and CM obtained from other researches. The CM microfiltration effectiveness was tested with different substances from food industry, i.e. slaughterhouse wastewater treatment and goat milk pasteurisation. The obtained results, insoluble residue rejections (100%) and high bacterial removal (87–99%), make the ceramic CM suitable for microfiltration processes.

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## 1. Introduction

Membrane separation processes extend more every day in industrial uses, with new requirements concerning to materials and preparation procedures. The interest in separation by the use of inorganic membranes has rapidly increased during the last two decades. Due to their potential application in a wide range of industrial processes such as water and effluent treatments [1–5], drink clarification [6–8], milk pasteurisation [9–13], biochemical processing [14,15], inorganic membrane technologies have achieved an increasing importance. Ceramic membranes have many known advantages such as high thermal and chemical stability, pressure resistance, long lifetime, and catalytic properties from their intrinsic nature [16,17].

The main process to prepare ceramic membranes includes first the obtaining of a good dispersion of small particles and then, the deposition of such dispersion on a support by a slip casting method. The support provides mechanical strength to a membrane top-layer (or active layer) to withstand the stress induced by the pressure difference applied over the entire membrane and simultaneously has a low resistance to the filtrate flow. Furthermore, the supports require chemical stability, heat shock-stability and high porosity, among others. For example, ceramic filters can be steam cleaned in applications that require sterility. The high hardness of ceramics also gives them better wear resistance. These properties are similar to those put forward for the use of dense ceramics as structural engineering materials. Macroporous ceramics can be fabricated by ceramic compacts partially sintered which are produced by conventional ceramic techniques. In this case the pore structure is controlled by the packing of particles. The pore size is commonly controlled by the particle size and the

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densities are limited to those given by the green density. Usually, the suspensions are prepared from a small distribution of particles and they may be used only after a complete stabilisation. Besides, it is necessary some adequate processes such as mixing, aging and removing the bigger diameter particles. Sometimes organic surfactants are added to avoid the flocculation of the suspensions.

Most of the supports are generally manufactured from compounds such as alumina ( $\text{Al}_2\text{O}_3$ ) [18–28], cordierite ( $2\text{MgO} \cdot 2\text{Al}_2\text{O}_3 \cdot 5\text{SiO}_2$ ) [29–31] and mullite ( $3\text{Al}_2\text{O}_3 \cdot 2\text{SiO}_2$ ) [32–33]. These materials have relatively elevated prices which significantly contribute to the cost of membrane modules for industrial applications. To reduce the cost of ceramic membrane fabrication, recent research works are focused on the use of cheaper raw materials such as apatite powder [34], natural raw clay [35–37], dolomite, kaolin [38,39] and waste materials such as fly ash [40–42]. Regarding to mesoporous and microporous ceramics, these materials have pore sizes that range from 50 nm to below 1 nm. This interval of pore size is commonly beyond the pore size ranges for which the powder processing routes are suitable. Alternative approaches include the sol–gel process or the preparation of synthetic zeolites and pillared clays.

To obtain the composite ceramic membrane the active layer has to be deposited on the support. This active layer used in liquid permeation consists of a porous layer, usually symmetric, with a pore density of  $10^9$  pores/cm<sup>2</sup> approximately and a porosity (or void volume fraction) between 30% and 70%. The pore size is included in a range of 50–1000 nm in the case of MF and 2–50 nm for ultrafiltration (UF) membranes [43].

In order to decrease the membrane cost and to make use of regional raw materials, this work aims to prepare composite ceramic membranes using natural ball clay, bentonite, feldspar, quartz, alumina, and chemical additives. The different substrates (support, active layer, and composite membrane) are prepared controlling the particle size, the paste formulation and the thermal treatment. Their structural and functional properties are determined by different techniques. The most important parameters used in the characterisation of these substrates are: surface and internal morphology, chemical transformation during the thermal process, mean pore size, pore size distribution, porosity and water permeability. The performance of the ceramic composite membranes is analysed through microfiltration tests of different substances from the food industry.

Fig. 1 shows a schematic representation of the sequential steps of the support, active layer, and the procedure of composite membrane preparation. The main manufacture steps include: a) paste formulation, b) paste conditioning (milling, sieving and suspension stabilisation), c) casting, d) drying and sintering.

## 2. Materials and characterisation techniques

### 2.1. Raw materials

Clay, quartz, and feldspar used in the present work were provided by Piedra Grande (Argentina). The chemical composition determined by Induction Coupled Plasma (ICP) and

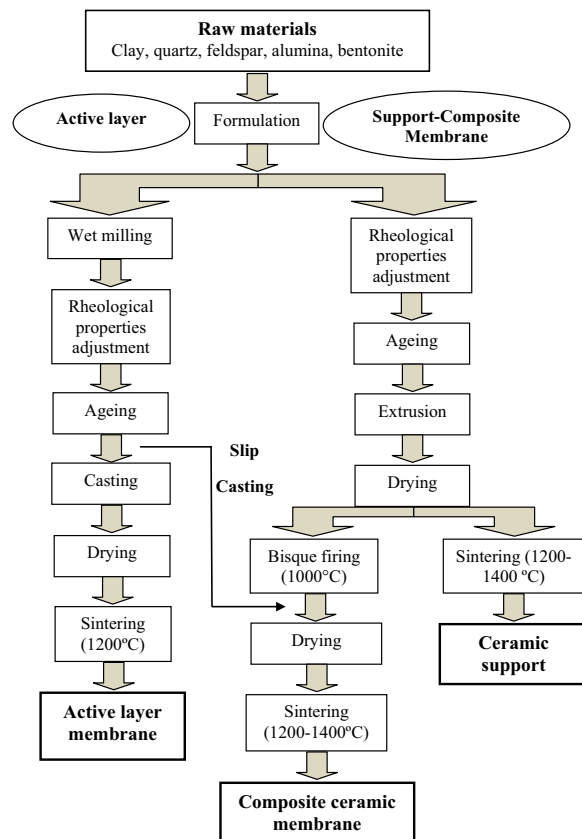


Fig. 1. Schematic representation of support, active layer and composite ceramic membrane preparation.

Atomic Absorption with Graphite Furnace (AA-GF), expressed as weight percentages are [43]; clay: 61%  $\text{SiO}_2$ , 25%  $\text{Al}_2\text{O}_3$ , 0.7%  $\text{Fe}_2\text{O}_3$ , 1.6%  $\text{CaO}$ , 0.45%  $\text{MgO}$ , 0.5%  $\text{TiO}_2$ , 1.1%  $\text{K}_2\text{O}$ , 0.15%  $\text{Na}_2\text{O}$  and 9.5% of solids lost by calcinations; quartz:  $\text{SiO}_2$  was present in a 96–98% (w/w), and low quantities of  $\text{Al}_2\text{O}_3$  (0.5–1% (w/w)) and alkalis (0.1–0.3% (w/w)); feldspar: 65–70%  $\text{SiO}_2$ , 16–20%  $\text{Al}_2\text{O}_3$ , 10–14% of alkalis with a Na/K ratio of 3–4, 1–2% oxides of Fe, Mg, Ca and Ti, and 0.2–0.6% component losses by calcinations. Bentonite was supplied by Santa Gema Company (Argentina), and it is composed primarily by silica and aluminium oxide, 0.2–0.5%  $\text{Fe}_2\text{O}_3$ , 1–2%  $\text{CaO}$ , 1.5–2.5%  $\text{Na}_2\text{O}$ , 3–5%  $\text{MgO}$ , 1–2% carbonates of Ca and Mg, and humidity of 10–12%. Alumina was provided by Alcoa S.A. (Argentina), and it was obtained from bauxite by the so-called complex process Bayer. It contains more than 98% of  $\text{Al}_2\text{O}_3$ , 0.5–1%  $\text{Na}_2\text{O}$  and 0.05–0.07% ( $\text{Fe}_2\text{O}_3 + \text{SiO}_2$ ). The particle size ranges of these materials obtained by X-Rays analysis are informed in Table 1. The additives magnesium silicate ( $\text{MgSiO}_4$ ), sodium silicate ( $\text{Na}_2\text{SiO}_4$ ), sodium carbonate ( $\text{Na}_2\text{CO}_3$ ), and glycerine were acquired from Aldrich (Argentina).

### 2.2. Characterisation techniques

To determine the chemical and physical properties of the green pastes and consolidate ceramic substrates, different techniques were used. The substrates were characterised by studying both their structure and functionality.

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