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ORIGINAL RESEARCH PAPER

PROCESSING AND CHARACTERIZATION OF TUBULAR CERAMIC SUPPORT FOR MICROFILTRATION MEMBRANE PREPARED FROM PYROPHYLLITE CLAY

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Abstract: Tubular macroporous support for ceramic microfiltration membranes were prepared by extrusion followed by sintering of the low cost pyrophyllite clay. Clay powders mixed with some organic additives can be extruded to form a porous tubular support. The average pore size of the membrane is observed to increase from 5 µm to 10.8 µm when sintering temperature increase from 900 °C to 1200 °C. However, with the increase in temperature from 900 °C to 1200 °C, the support porosity is reduced from 47% to 30% and flexural strength is increased from 4 MPa to 17 MPa. The fabricated macro-porous supports are expected to have potential applications in the pre-treatment and also can be used like support for membranes of ultra-filtration.

Keywords: *ceramic, clay, membrane, microfiltration*

INTRODUCTION

Due to their potential application in a wide range of industrial processes, membrane technologies have received an increasing interest. Numerous application have been proposed of which micro-filtration and ultra-filtration are critical technologies in chemical and biochemical processing that are regarded economically competitive due to the availability of membranes with higher flux and lower process cost [1, 2]. The use of ceramic membranes has many advantages such as higher thermal and chemical stability, pressure resistance, long lifetime and catalytic properties from their intrinsic nature [3]. A membrane support provides a mechanical strength to a membrane top layer to withstand the stress induced by the pressure difference applied over the entire membrane and must simultaneously have a low resistance to the filtrate flow.

The market support is generally manufactured from the compounds such as alumina (Al_2O_3), silica, zirconia which have a relatively elevated cost. To circumvent the issue of support cost, recent research in the fabrication of inorganic membranes is focused towards the utilization of less expensive materials such as apatite powder, dolomite, and kaolin [4 – 7].

In this work, we report the fabrication of tubular macro-porous ceramic support from pyrophyllite by extrusion followed by sintering. The properties of porous pyrophyllite were discussed as a function of sintering temperature in order to optimize the preparation conditions.

MATERIAL AND METHODS

After collecting of the raw clay, stones and other heavy particles were removed from the samples. The clay material was crushed, about 250 g of the powder was then dispersed in 2 L of deionised water; after sedimentation of the clay particles, the top part was sieved through a 50 µm sieve to remove the larger non-clay fractions and to obtain the fine clay fraction. The sample were dried at 100 °C and stored in a dessicator.

The particle sizing Accusizer model 770 (particle sizing system Santa Barbara) was used to determine the particle size distribution. The XRD analysis of the treated clay were carried with a Siemens D5000 Diffractometer (Cu K ray, Ni Filter)

Thermogravimetric analysis (TGA) and differential scanning calorimetry (DSC) were performed with simultaneous DSC-TGA 2960 TA instrument. The sample was heated from room temperature to 1050 °C at a rate of 5 °C/min under static atmospheric conditions; α-Al_2O_3 was used as reference.

The pore size of the support was determined by mercury porosimetry (Micrometrics Autopore II 9215).

RESULTS AND DISCUSSION

Powder characterization

The chemical composition of the clay in weight percentages of oxides is given in Table 1; it reveals that the clay powder is essentially formed of large amount of silica with alumina and iron oxide in lower proportion.

Table 1. *Chemical composition of clay [% w]*

SiO_2	Al_2O_3	Fe_2O_3	Na_2O	CaO	K_2O	MnO
65.4	22.1	3.12	1.75	1.58	6.04	0.01

The particle size distribution of this matter (Table 2), has a range from 1 to 40 µm.

Table 2. *Percentage of particle size distribution for clay*

Diameter [µm]	**Particle size distribution [%]**
[10, 40]	50
[5, 10)	30
< 5	20

Figure 1 presents the XRD patterns of the treated clay; it shows that pyrophyllite, kaolinite and quartz are the main minerals present in the clay.

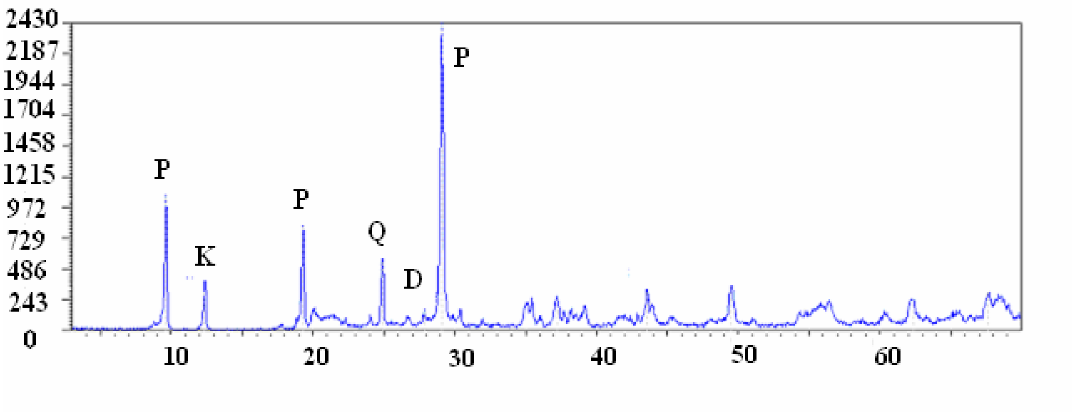

Figure 1. *X-Ray diffraction diagram of the clay (P: Pyrophyllite; K: Kaolinite; Q: Quartz; D: Dickite)*

The objective of thermal analysis is to identify temperature regimes where predominant losses (and hence transformations) occur in the membrane. Thereby, an understanding could be developed for analyzing the effect of various temperature regimes on the porous structure, pore diameter and mechanical strength of the membrane. Figure 2 presents TGA and DTA of the pyrophyllite clay when subjected to thermogravimetric analysis by heating the dry clay in an α-alumina crucible from room temperature to 1000 °C at a heating rate of 10 °C/min.

The weight loss is of the sample is observed to 8%, which can be correlated with the deshydroxylation of the pyrophyllite and also with the transformation of kaolinite into metakaolinite.

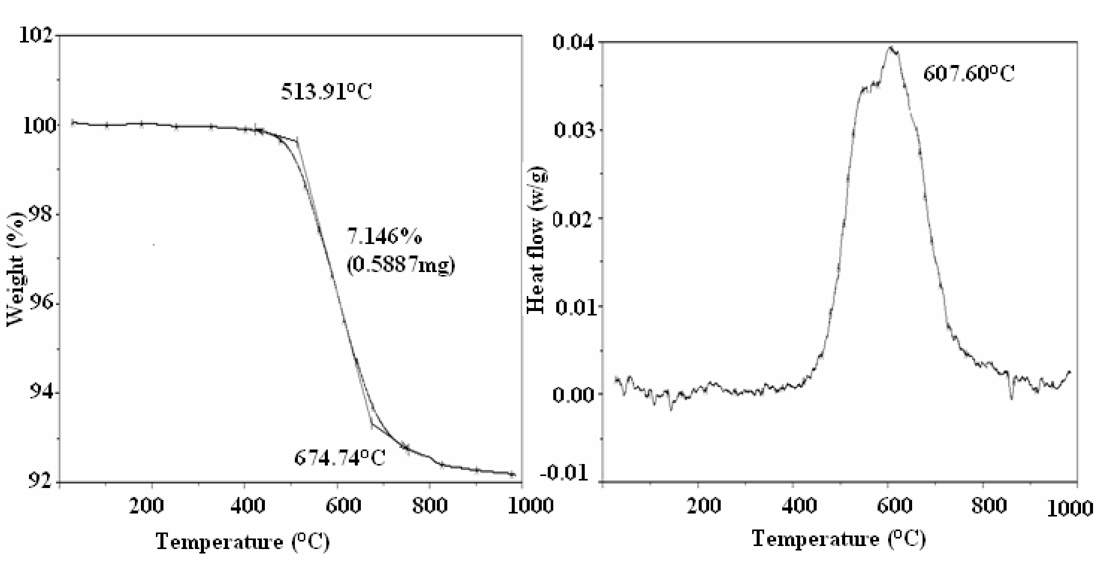

Figure 2. *TGA-DSC of the clay*

During the DTA treatment, we observed two endothermic peaks at 560 °C and 607 °C, which can be explained by the transformation of kaolinite into metakaolinite and the deshydroxylation of pyrophyllite respectively.

Preparation of clay support

The optimal formulation of the ceramic precursor paste was carried out in an empirical way. The plastic paste was prepared from the clay powder mixed with the organic additives and water. Plasticizer and binder are required to prepare paste with rheological properties allowing the shaping by extrusion. We report the main steps of the preparation:

a) mixing of 81.7% (w/w) clay, 10% (w/w) starch, 4% (w/w) Methocel, 4% (w/w) Amijel and 0.3% (w/w) PEG 1500;
b) adding 37.2% (w/w) water and 0.24% (w/w) Zusoplast 126/3;
c) Mixing for 30 min;
d) Ageing of the paste: the paste is kept at room temperature in a closed box for 48 h under high humidity.

The tubular support is then obtained by the extrusion of the prepared ceramic paste. After extrusion, the wet support is placed on rollers to obtain a good homogeneity with room temperature. Finally, a thermal treatment was carried out in a programmable furnace at different final temperatures. The adapted firing treatment was established from the thermal analysis data (Figure 2). Two steps have been used, the first for the decomposition of organic additives, the second for the sintering of ceramic.

The sintering temperature is an important parameter which controls the pore diameter of the support but also its mechanical resistance. The thermal expansion depends also on the firing treatment; therefore, the best properties of the final support are achieved by adjusting the conditions for sintering. Figure 3 shows the evolution of the pore diameter with the firing temperature. The pore diameter increase as the temperature increases; it reaches a maximum 10.6 µm at 1200 °C.

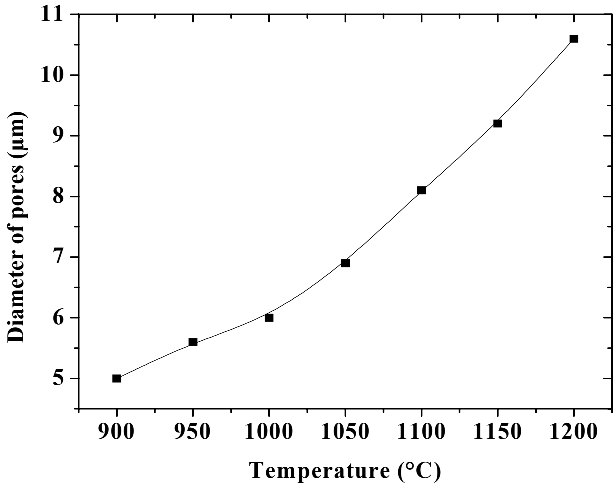

Figure 3. *Pore diameter versus firing temperature of the support*

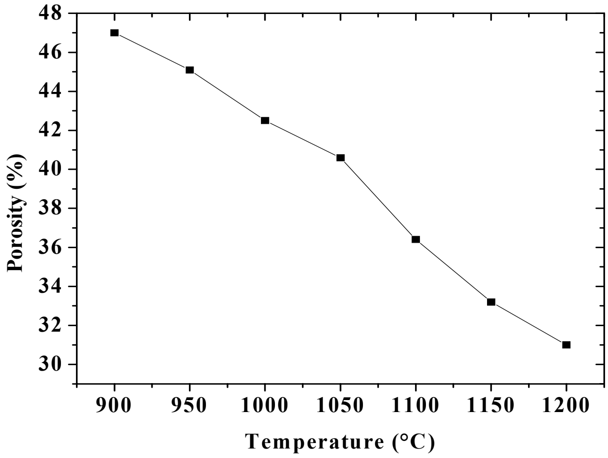

Figure 4. *Porosity versus firing temperature of the support*

The porosity (Figure 4) decreases from 47% at 900 °C to 31% at 1200 °C.

The first part of the curves corresponds to an opening of the pores with temperature, whereas the last part is caused by the beginning of the material densification.

The mechanical resistance test was performed using the three points bending strength to control the resistance of the support tube fired at different temperatures.

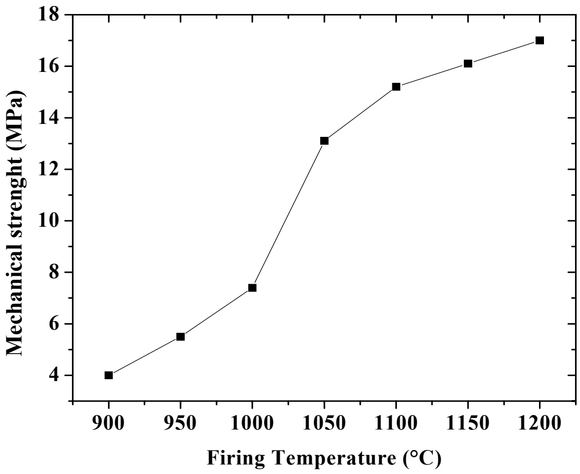

***Figure 5.** Mechanical strength versus firing temperature of the support*

The mechanical strength reported in Figure 5 increases with increasing sintering temperature and reaches 15 MPa at 1200 °C.

CONCLUSION

In this work a new support of micro-filtration membrane was prepared from natural powders derived from Moroccan clay. This kind of ceramic support presents a very interesting porous volume and average pore size.

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