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Effect of sodium phosphate addition on mechanical properties of porous Sigue quartz sand

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ABSTRACT

Porous ceramic separation membranes exhibit many excellent properties, such as a high structural durability, a good thermal stability, a long operational life, and an especially high chemical stability in the corrosive media. In order to prepare low-cost membranes, many researchers have used low-cost starting materials such as clay, dolomite, and kaolin. This work addresses the development of low-cost ceramic microfiltration membrane supports from inexpensive raw materials, such Sigue quartz sand (SQS) and sodium phosphate (SP), using a uniaxially dry compaction method. The prepared samples were sintered at different temperatures ranging between 900 and 1,400˚C. The raw materials and the prepared samples were characterized using X-ray diffraction (XRD), optical microscopy, Hg-porosimetry, and tensile strength using a diametral compression test. Subsequently, the effect of sintering temperature and SP amounts on support proprieties, such as the shrinkage, the phase transformation, the porosity, and tensile strength, were also investigated. It was observed that with increasing sintering temperature, dimensions of sintered samples increased; this was due to the phase transformation. XRD confirmed that SQS was transformed into cristobalite phase when both the sintering temperature and the holding time were increased. The porosity and average pore size (APS) increased in all cases (2 and 3 wt% SP) in the range of temperatures between 1,100 and 1,400˚C. For example, the porosity increased from 25.4 to 32.9%, and APS increases from 11 to 27 μm in the case of 3 wt% SP. The flexural strength was found to be acceptable. The value varied from 16 to 20 MPa, which corresponds well with the relatively high porosity and APS.

Keywords: Sodium phosphate; Quartz sand; Mechanical properties; Sintering; Porous ceramics

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1. Introduction

Porous ceramic separation membranes exhibit many excellent properties, such as a high structural durability, a good thermal stability, a long operational life, and an especially high chemical stability in corrosive media, such as strong alkaline and acidic solutions [1–3]. In order to prepare low-cost membranes many researchers have used low-cost starting materials such as clay, dolomite, and kaolin [4–7]. In this work, the long-term goal is the development of a low-cost ceramic microfiltration membrane supports from inexpensive raw materials, such as Sigue quartz sand (SQS) and sodium phosphate (SP) using a uniaxially dry compaction method. Silica is a mineral used in many domains. The traditional view is that there are three distinct families of silica structures, which are stable at an ambient pressure: quartz, tridymite, and cristobalite. Although, quartz is believed to be in stable phase at temperatures below 870˚C, both tridymite and cristobalite can be cooled to low temperatures, where they are metastables. The conversion of quartz into tridymite can be occurred at temperatures more than 870˚C with mineralizer. It seems, besides that, the temperature of transition depends on nature and concentrations of the mineralizer. The transformation of pure quartz, under the only action of temperature, always leads to cristobalite. Theoretically, the temperature must exceed 870˚C; practically, it is generally necessary to reach 1,000˚C to put into evidence the presence of cristobalite, and the speed of transformation increases with the increase in the sintering temperature. However, the cristobalite maintained at a temperature ranging between 870 and 1,470˚C is transformed into stable tridymite in this temperature range. Moreover, the transformation rate is always much smaller than that of quartz into cristobalite. Calcite ($CaCO_3$), dolomite ($CaCO_3{\cdot}MgCO_3$), bones (natural derived hydroxyapatite: HA: $Ca_{10}(PO_4)_6(OH)_2$), kaolin, feldspar, and quartz are abundantly available raw materials. Many works have already been published for appreciating these native raw materials including ceramic membranes [1–7], advanced ceramics [8–10], and bioceramics [11–17]. For example, ceramic filters are generally constituted of a thick support (2,000 μm) and mono- or multi-thin membranes (from 10 to 40 μm for each one).

Therefore, replacing the more expensive starting materials, mentioned above, by other cheaper raw materials used in supports (which constitute about 99% of the filter mass) is important. So, what do low-cost raw materials mean? The price of alumina is at least 100 times greater than that of kaolin. Another important advantage is the substantial gain in energy obtained by decreasing the sintering temperature from 1,600 to 1,250˚C [1,7], when alumina supports were replaced by the supports made from kaolin. Besides this, about 50% of the prepared supports are pores, which may also be considered as a gain in mass. The relatively lower theoretical density of the prepared supports (2.8 g/cm^3) when compared to that of alumina (3.98 g/cm^3) is also another interesting advantage. More recently, it has been demonstrated that besides these advantages the fabricated membrane supports have also comparable mechanical strength than that of alumina [7]. Indeed, a flexural strength of 87 ± 2 MPa was obtained for 100 wt% Al_2O_3 samples sintered at 1,620˚C for 2 h [18], while nearly the same flexural strength value (87 ± 6 MPa) was also measured for compacts sintered only at 1,250˚C for 1 h, using the proposed process.

Generally, membrane supports should have a total porosity ratio of about 45% as reported in the literature [1,7]. According to the previous results, there is a relationship between the porosity ratio, pore sizes, sintering temperatures, and mechanical properties [7]. Since porous supports should resist the applied pressure during solution filtrations, a higher mechanical strength is also of great importance.

The aim of this work is an attempt to use SQS as a cheaper raw material for ceramic membrane supports production.

2. Experimental procedures

2.1. Preparation of specimens

The starting material is sand (derived from SIGUE region) whose composition (wt%) is shown in Table 1.

Fig. 1 shows a schematic drawing of the main steps employed for the sample preparation, used in this work. Firstly, SQS was crushed with a mortar for 20 min, to make it finer for the compaction. Afterwards, two mixtures were prepared. The first mixture was constituted of about 98 wt% sand and

Table 1
Chemical composition of SQS, using X-ray fluorescence analyzer

Composition	wt%
SiO_2	98.25
Al_2O_3	0.50
TiO_2	0.12
Fe_2O_3	0.25
CaO	0.80
MgO	0.08

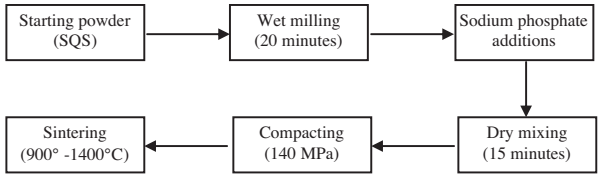

Fig. 1. A schematic diagram showing the process used for samples preparation.

2 wt% SP, whereas the second one contained 97 wt% sand and 3 wt% SP. The materials were then dry mixed for 15 min. The mixtures were afterwards compacted in the form of discs (13 mm in diameter and 5–6 mm in thickness) at 140 MPa. Subsequently, the compacted samples were fired for 1 h at temperatures ranging between 900 and 1,400˚C. Some samples containing 2 wt% SP were sintered at 1,200˚C for different times (1, 3, 5, and 7 h).

2.2. Characterizations

Various analyses were used for the characterization of sintered samples. Phase compositions of samples were identified by X-ray diffraction (XRD) (BRUKER, D8 ADVANCE) (Karlsruhe, Germany) with a CuK α radiation ($\lambda = 0.154$ nm) and a Ni filter, working voltage 40 kV, and working current 30 mA. The shrinkage was determined by the diameter measurement of the dry circular pellet before and after the sintering process. The morphology of the sintered pellet was investigated using an optical microscopy. For this, a sintered pellet was coated with resin in a vacuum, polished on one side using a silicon carbide abrasive paper and diamond paste, and in the final stage, the polished side was coated with a thin gold layer. The porosity and the pore size of fired samples were measured by Hg-porosimetry (Micromertics, AutoPore IV 9500).

The tensile strength testing of sintered specimens was performed using a diametral compression test (FORM TEST SEIDNER D 79-40) (Germany). One of the fundamental aspects of this test is the relatively small proportion of the specimen volume which reaches the peak stress at fracture.

In its simplest form, a right-circular cylindrical specimen is compressed diametrally between two flat platens. A biaxial stress state is produced within the test specimen, and on the assumption of an ideal line loading; the vertical plane is subjected to a uniform horizontal tensile strength of magnitude.

$$\sigma_t = 2P/\pi dt \quad (1)$$

where σ_t (MPa) is the maximum tensile stress, P (N) is the applied load at fracture, d (mm) is the specimen diameter, and t (mm) is the specimen thickness. The correspondence between measured tensile strength (σ_t) value and its equivalent three-point flexural (bending) strength (σ_f) is given by the following equation:

$$\sigma_f\ (\text{MPa}) = 2.7\sigma_t\ (\text{MPa}) \quad (2)$$

It should be noticed that this deduced flexural strength is needed for a comparison since; it is generally used by the major investigators and vice versa [19,20]. It should also be noticed that hundreds of samples within the two different shapes have been tested separately. So, it has also been confirmed that the conversion ratio between flexural strength and the tensile strength of samples is always interchangeable.

3. Results and discussion

Fig. 2 shows the variations of porosity and average pore size (APS) in terms of temperature of samples containing 3 wt% SP. The porosity and APS values increase considerably in the temperature range between 1,100 and 1,400˚C. For example, the porosity ratio increases from 25.4 to 32.9% whereas, APS increases from 11 to 27 μm in the case of 3 wt% SP. This increase may be attributed to the consolidation of the grains among themselves under the effect of sintering with the glassy phase and the phase transformation. Moreover, the effect of SP (wt%) additions on the porosity and APS is shown in Fig. 3.

This SP addition allows the formation of a more porosity and increases the APS. A typical overview of

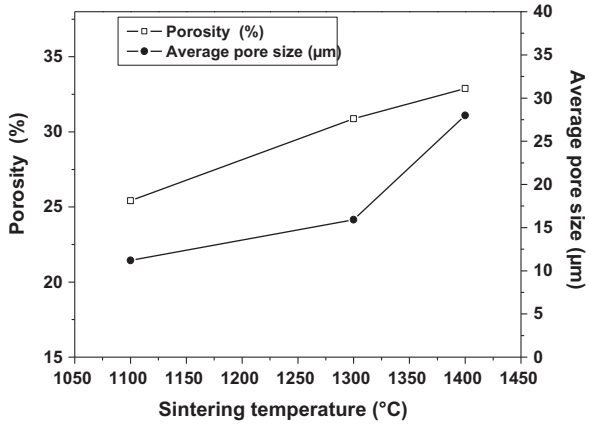

Fig. 2. Effect of sintering temperature on the porosity (%) and APS of sintered samples containing 3 wt% SP at various temperatures for 1 h.

the microstructure of samples containing 2 wt% SP sintered at 1,200˚C for 1 h is given in Fig. 4. This micrograph shows the distribution modal of pore size within the main phase, and open and communicating pores through the sintered samples.

The effect of both the SP content and the sintering temperature on the tensile strength is given in Fig. 5. Indeed, in the case of 2 wt% SP, the tensile strength increased in the range between 900 and 1,100˚C. After 1,100˚C, it decreased considerably. These variations may be closely related to sintering phenomena and the phase transformation. In the first stage, the sintering is a controlling factor, which may improve mechanical properties, at lower temperatures. Nevertheless, by increasing the temperature, the phase transformation of quartz into cristobalite occurs more rapidly, as porosity and pore size increase too as shown in Figs. 2 and 3. All these factors may affect the mechanical properties and especially, cristobalite has lower mechanical properties than that of quartz.

Fig. 6 shows the effect of sintering time on the tensile strength. The tensile strength increased slightly with increasing sintering time. The increase in strength may be mainly due to the densification of the samples when transformation is completed. In order to confirm the direct correlation between the porosity and the tensile strength; the evolution of phase transformation was examined with increasing sintering temperature in the range between 900 and 1,400˚C. It was found that the only phase which exists up to 1,100˚C is quartz. From 1,200˚C to higher temperature, cristobalite appears and the transformation rate increases with the increase in temperature. The transformation is almost completed at 1,400˚C for 1 h (Fig. 7).

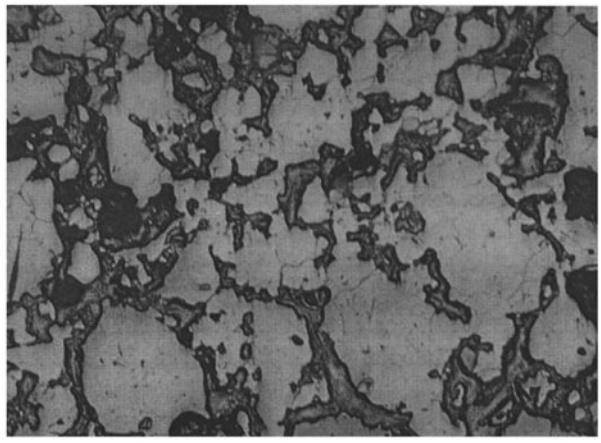

Fig. 4. Optical micrograph showing a microstructure of sample containing 2 wt% SP sintered at 1,200˚C for 1 h.

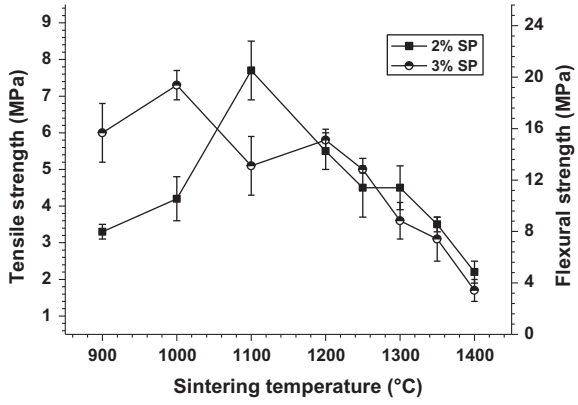

Fig. 5. Tensile strength and flexural strength of SQS containing 2 and 3 wt% SP sintered at different temperatures for 1 h.

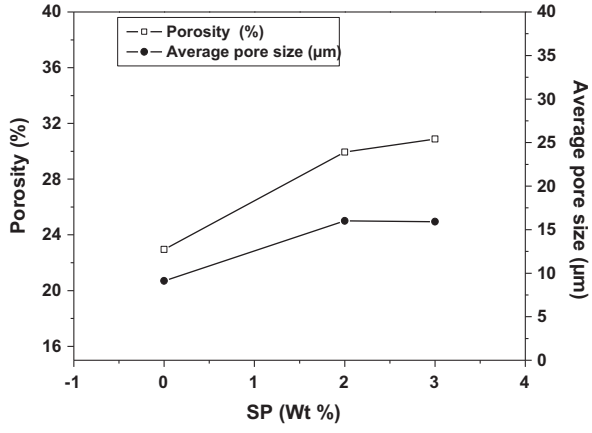

Fig. 3. Effect of SP wt% on the porosity (%) and APS of samples sintered at 1,300˚C for 1 h.

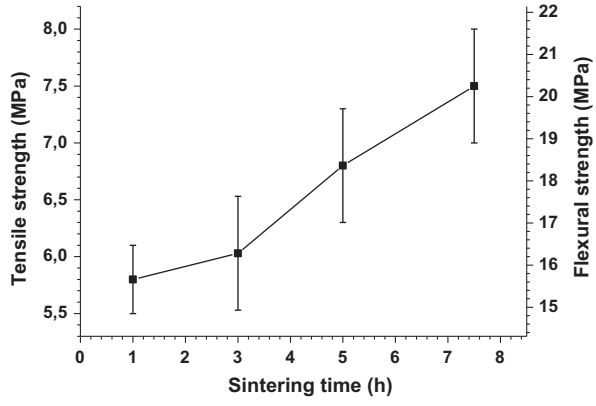

Fig. 6. Tensile strength and flexural strength of SQS + 2 wt% SP sintered at 1,200˚C for different times.

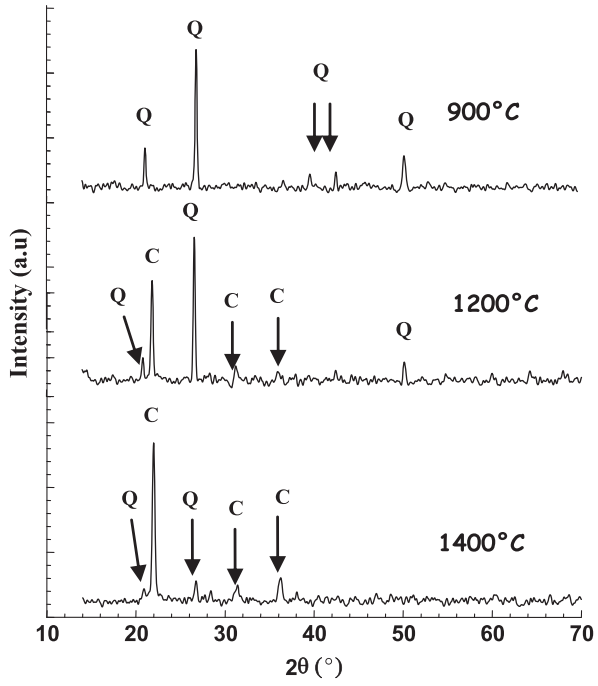

Fig. 7. XRD patterns of samples of SQS + 2 wt% SP sintered at various temperatures for 1 h.

Fig. 7 illustrates the XRD patterns of samples containing 2 wt% SP as a function of sintering temperature. It was clearly observed in XRD patterns that both SP contents and sintering temperatures promoted transformation phenomena. Additionally, the dilatation (i.e. the increase in diameter) of the samples was evaluated using diameter measurement of samples before and after sintering. As shown in Fig. 8, an increase in the sintering temperature caused an

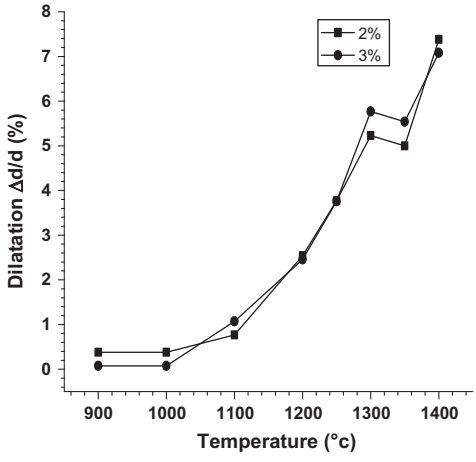

Fig. 8. Linear dilatation percent $\Delta d/d$ (%) of samples sintered at various temperatures for 1 h.

increase in the dilatation of samples. It was observed that the change in the dimension of the samples was lower in the range from 900 to 1,100˚C. The increase in the diameter for the samples sintered at 1,100˚C was found to be between 0.77 and 1.07% in the cases 2 and 3 wt% SP, respectively. However, these changes reached between 7.38 and 7.08%, when samples were sintered at 1,400˚C. At lower temperatures, the sintering process plays a more important role than the transformation. Hence, there is a small change. Nevertheless, at higher temperatures, the transformation is becoming important. The increase in the dilatation is more obvious; since, the transformation of quartz into cristobalite accompanied by the volume increase. When the phase transformation is achieved, the densification will continue and may justify the mechanical properties enhancement in samples sintered for a few hours (Fig. 6).

Moreover, it should also be mentioned that using ceramics (oxides) in this work and others [21] instead of metallic products [22,23] is well justified, particularly for water filtration or purification. In fact, the utilization of these metallic products as a support or membranes is strictly forbidden since: they will react instantaneously with the filtered water.

The results obtained in the current study are in a good agreement with other published works [24–26]. The SP addition plays a role in the material densification. Indeed, a 2 wt% SP addition remarkably helps SQS powders to be sintered earlier (from 900˚C). On the other hand, SQS alone cannot be sintered at a higher temperature (1,400˚C). The effect of additions was observed in a work on refractories containing zirconia (ZrO_2) carried out by Volceanove et al. [24]. Another role of SP is to promote the transition from one phase to another (e.g. from quartz to cristobalite). A similar effect of phosphate addition on transformation from μ to α cordierite was studied by Met et al. [25]. They found that the P_2O_5 additive promotes the μ-cordierite-α-cordierite transition at lower concentrations, whereas it retarded the transition at higher concentrations. Finally, it can be said that the cristobalite crystallization temperature (1,200˚C for 1 h) is in a good agreement with that reported by San et al. [26] (1,200˚C for 5 h).

4. Conclusions

In this work, ceramic membrane supports were prepared by using SQS. Moreover, these supports are characterized by a reduced manufacture cost since the used raw materials are very abundant (in Algeria). It was also demonstrated by using XRD patterns that both SP contents and sintering temperatures promoted

the phase transformation. It has also been found that both the support porosity (about 33%) and their flexural strength (about 22 MPa) were acceptable. Based on the above-mentioned advantages, SQS can be used as supports for membranes of the microfiltration and/or ultrafiltration.

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