Research Article

Preparation and Sintering Effect in Quartz-Barium Titanate Porous Ceramics and Permeability Modulation Using an Implanted Electrode

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Barium titanate and quartz mixed in different proportions were used to create porous piezoelectric ceramics. Three different sintering temperatures were used for the ceramics preparation; a nichrome wire was used as internal electrode in porous ceramics. Characteristics as porous area, porosity, and its relationship with quartz percentage and sintering temperatures were studied. Porous ceramics with an implanted electrode were created, by applying an alternating voltage in the internal electrode that controlled the liquid permeability coefficient, calculated by the Darcy Law.

1. Introduction

Piezoelectric ceramics should have high density in order to obtain good mechanical and piezoelectric responses, which means that porosity represents a fail; moreover, there are some fields where porosity is an advantage; examples of this are filtering, high temperature applications, and piezoelectric transducers where the target acoustic impedance is low [1]. Usually ceramic preparation is expensive and it is difficult to control its structural properties. This is the reason to research new methods to obtain ceramics is necessary. In the ceramics fabrications process is necessary to pay attention to the initial components, preparation, and sintering conditions, since those steps are essential to obtain specific properties in accordance with the desired applications [2].

Several methods have been proposed in order to obtain porous ceramics; sacrificial template method, replica technique, and direct foaming can be mentioned as examples of this [3–5]. In this paper, a method is proposed based on the combination of two different piezoelectric materials and their differences on densification processes and fusion temperatures, in order to obtain porous piezoelectric ceramics. By the use of adequate material proportions and suitable processing

conditions it is possible to get porous ceramics in the micro- and nanofiltration [6].

Using metallic wire immersed in the ceramic as third electrode, it is possible to modify the dielectric constant in the ceramic; it is due to the domains concentration around the wire that magnifies the sensibility in the area near the electrode [7]. Some metallic materials used as third electrode in this ceramic kind are platinum (Pt) and nichrome (NiCr); these materials are used due to low chemical reactivity, stability in electric properties, high conductivity (electrical and thermal), and high fusion temperature [8–10]; a ceramic with third electrode called ceramic controlled piezoelectric offers the advantage that increases the analysis surface because the metallic implant works as third electrode [11–14].

2. Materials and Methods

2.1. Samples Preparation. The oxide method was used to obtain piezoelectric ceramics [15]. Barium titanate (BaTiO_3) and Quartz (SiO_2) with purity of 99% or more were used; ceramics were created with quartz in proportions of 20%, 18.75%, and 10.63%, and these percentages were selected

based on the mechanical resistance of final ceramics. All the raw materials and wire implants were obtained from Sigma Aldrich.

The particle size of the used compounds was controlled by sieving. Also mentioned compounds were mixed in controlled proportions and crushed in an agate mill for 40 minutes; polyvinilic alcohol was added in a rate of 0.075 mL per gram of mixture; afterwards this mixture was pressed forming two layers: the first contains BaTiO_3 and the second is a mixture of BaTiO_3 and SiO_2 in the rates previously mentioned, at 3500 kg/cm^2 in a circular mold to obtain samples with 10 mm diameter and 2 mm thickness. During the fabrication process and previous to the sintering stage NiCr wire was added in transversal form between layers. Samples were sintering in air with heat rate of 5°C/min from room temperature to 600°C , a stay of 30 minutes and new ramp with heat rate of 10°C/min until final sintering temperature, and a final stay in the sintering temperature of 60°C ; finally the samples were cooled in natural way; in this form ceramics of two layers and immerse electrode were created as Figure 1 shows; three sintering temperatures were used: 800, 1000, and 1200°C . Ceramics without third electrode were created in order to serve as references.

The obtained ceramics were poled by applying an electric field of 1 kV per millimeter thickness at 100°C in an hour silicone oil bath.

3. Characterization

In order to know the size and number of pores in sintered ceramics an optic microscope (DTM300) with coupled photographic camera was used. The obtained images were processed by the use of software (Ts View tucsen and NI Vision Builder Al) as way to avoid measurement errors due to human perception.

By the use of theoretical density, porosity p was calculated with

$$\%p = \left(1 - \frac{p_m}{p_t}\right) \times 100, \quad (1)$$

where p_m and p_t are measured and theoretical densities, respectively.

The fine lands method was used to calculate the liquids permeability coefficient for ceramics with and without third electrode; the method consists of placing water in graded container sealed in its bottom with the element which will be characterized. Measuring the time used to reach certain altitude, by the use of Darcy law equation (2) [16], it is possible to get the liquids permeability coefficient. Figure 2 shows the experimental setup used to obtain this coefficient:

$$\%p = \left(1 - \frac{p_m}{p_t}\right) \times 100 \left[\frac{\text{cm}}{\text{s}}\right], \quad (2)$$

where a is the burette area (cm^2), L is the ceramic thickness (cm), A is the ceramic area (cm^2), h_1 and h_2 are the initial and final water altitude, respectively.

FIGURE 1: (a) NiCr electrode and porous ceramic with (b) BaTiO_3 and (c) $\text{BaTiO}_3/\text{SiO}_2$ layers.

In the case of ceramics with third electrode, the experimental setup previously explained was used and the third electrode was excited with AC voltage with 5 V p-p amplitude and frequencies of 100 Hz, 500 Hz, and 1 MHz as Figure 2 shows.

4. Results and Discussion

Mixed oxide method was used to obtain porous ceramics, by using different sintering temperatures as well as different quartz proportion porous ceramics with different pore size, and as a result permeability coefficients were obtained and characterized.

Figure 3 shows ceramics photographs with 20% quartz, where it is observed that, by increasing the temperature, the number of pores decreases and its area increases; this is because at high temperature the number of grains decrease as a consequence of the compounds start to melting and make the ceramic structure more dense in some areas; low sintering temperatures can be considered incomplete sintering having as a consequence incomplete densification in both ceramic components.

Figure 4 shows the relation between pore area and pore number for ceramics sintered at 1200°C with quartz percentage of 18.75 and 20%; it is possible to see that pore number as well as area increases for those ceramics that have superior percentage of quartz.

Samples porosity as function of sintering temperature is shown in Figure 5; it is possible to see the increment in porosity when sintering temperature increases for those ceramics containing 18.75 and 20% quartz, while for ceramics with 10.63% quartz porosity decreases when sintering temperature increases.

For ceramics with third electrode, liquids permeability coefficient was measured; Figure 6 shows the obtained coefficients when alternating voltage that was applied in the third electrode for ceramics sintered at 1000°C and quartz percentage of 10.63 and 18.75%. It is clear that permeability coefficient for ceramics with 18.75% quartz is lower than for ceramics with 10.63% quartz; this is due to the density increment for this ceramics.

FIGURE 2: Experimental setup used to determine the liquids permeability coefficient, (a) porous ceramic without implant, (b) porous ceramics with implant; (A) container, (B) burette, (C) porous ceramic, (D) support, and (E) signal generator.

FIGURE 3: Ceramic digital photographs ($\times 10$) with scale bar 800 microns for sample with 20% quartz and sintering temperatures of (a) 1200°C, (b) 1000°C, and (c) 800°C.

FIGURE 4: Area and pore number for ceramics sintered at 1200°C.

FIGURE 5: Porosity changes as sintering temperature function.

FIGURE 6: Liquids permeability coefficient for porous ceramic sintered at 1000°C.

Figure 7 shows the liquids permeability coefficient for ceramics sintered at 1200°C and 10.63% quartz; the measurements point out that the highest liquids permeability coefficients were founded in ceramics sintered at 1200°C with a 10.63% percentage of quartz.

For ceramics stimulated with AC signal, it is important to point out that, in all cases, when the frequency of signal increases, the permeability coefficient increases; this is due to the fact that ceramic vibration allows increasing the liquid flux through the ceramic.

5. Conclusions

It was observed that it is possible to get porous ceramics by mixing BaTiO₃ and SiO₂ and changing the rates of the mixture as well as changing the sintering temperature. There is also a strong relationship between manufacture process and

FIGURE 7: Liquids permeability coefficient for porous ceramic sintered at 1000°C.

physical ceramic properties like number and pore area and liquids permeability coefficient; obtained data is the base to develop porous ceramics adequate for different applications.

It is practical to modify the sintering temperature and rate of quartz used in the porous ceramic fabrication as a way to control the desired porosity in the ceramics and consequently its permeability coefficient.

In the case of ceramics without third electrode, it is not necessary to stimulate the ceramic; the temperature at which this kind of ceramic works is suitable not only for biomedical applications but also for industrial applications, as high temperature filter.

In the case of ceramics with metallic implant as third electrode by applying an AC signal, it is possible to modify the permeability coefficient by means of varying the signal frequency; this characteristic can be used for the development of controlled filters in many fields.

Conflict of Interests

The authors declare that there is no conflict of interests regarding the publication of this paper.

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