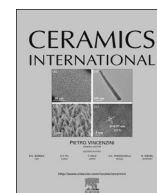




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Effects of slurry composition on the properties of 3-1 type porous PZT ceramics prepared by ionotropic gelation

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ABSTRACT

In this study, 3-1 type lead zirconate titanate (PZT) ceramics with one-dimensional pore channels were produced by ionotropic gelation process of alginate/PZT suspensions. By increasing the sodium alginate concentration from 1.0wt% to 3.0 wt%, the alginate/PZT suspensions turned from Newtonian to non-Newtonian behavior with substantial increase in apparent viscosity. Accordingly, 3-1 type PZT ceramics with porosity decreasing from 56.78% to 41.44% were obtained, while the pore size distribution became non-uniform gradually. Based on the structural features, the 3-1 type PZT ceramics possessed much higher relative permittivity (ϵ_r) than that of 3-0 or 3-3 type PZT ceramics with similar porosities. Increase in the porosity led to a moderate decline in the longitudinal piezoelectric strain coefficient (d_{33}), a reduction in the dielectric loss factor ($\tan \delta$), and a high value of hydrostatic strain coefficient (d_h). As a result, the 3-1 type PZT ceramics possessed a maximal hydrostatic figure of merit (HFOM) value of $5597 \times 10^{-15} \text{ Pa}^{-1}$ when the porosity was 56.78%, which may be of help for low frequency hydrophones applications.

1. Introduction

Porous lead zirconate titanate (PZT)-based ceramics, by introducing air phase into dense PZT ceramics in the form of pores, succeed in bringing about enhanced hydrostatic figure of merit (HFOM) and promoting improved signal-to-noise ratio of PZT ceramics for a host of low frequency hydrophones [1–6]. Based on the association between pores and PZT phase, porous PZT ceramics can be broadly classified into three types, i.e., 3–0, 3–1, and 3–3 type (the first number represents the connectivity of active PZT phase and the second is that of passive air phase). Traditionally, 3–0 and 3–3 type porous ceramics are able to be labeled as foam ceramics, whilst 3–1 type porous ceramics can be defined as honeycomb ceramics [7,8].

Compared with 3–0 and 3–3 type PZT ceramics, 3–1 type PZT ceramics always preserve the structure of unidirectional oriented pore channels, which is considered to favor the increase of the volume fraction of active materials along the poling direction and result in superior electric properties [9,10]. Due to high demands for well-ordered pore alignment and uniform pore size distribution, there are very few methods adopted to prepare 3–1 type PZT ceramics. In recent years, Guo et al. have developed unidirectional freeze-casting process,

employing Tert-butyl alcohol as crystallization regulator, to produce 3–1 type PZT ceramics with superior dielectric and piezoelectric properties [11]. But, the disadvantages of thin pore walls and uncontrollable pore size distribution have hindered the popularization of this technique. Lately, our group has successfully fabricated 3–1 type PZT ceramics with unidirectionally aligned pore channels by ionotropic gelation process of alginate/PZT slurries, which took the advantage of a phenomenon that hexagonally ordered alginate capillary hydrogels can be formed when the alginate sol is brought into contact with a salt solution containing calcium cations [12]. However, we have only discussed the effect of solid content of alginate/PZT slurries on the structure and piezoelectric properties of 3–1 type PZT ceramics, there are further investigations needed to comprehend the technique thoroughly. Hence, in the present work, the influence of slurry composition, e.g. sodium alginate concentration, on the structural, dielectric, and piezoelectric properties of 3–1 type PZT ceramics was investigated.

2. Experimental procedure

PZT-5H powders (Bao Ding Hong Sheng Acoustics Electron Apparatus Co. Ltd., Hebei Province, China) with a mean particle size

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Table 1
Chemical composition (wt%) of the original PZT powders.

Analyte	Result (%)
PbO	64.60%
ZrO ₂	11.27%
TiO ₂	10.72%
NbO	9.54%
SrO	2.26%
MgO	1.22%
Others	0.39%

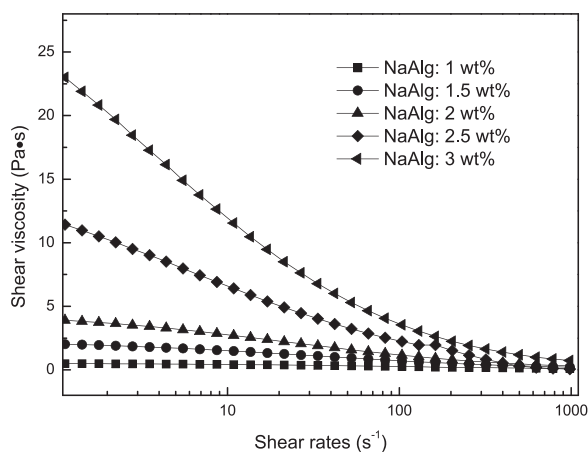


Fig. 1. The effect of sodium alginate concentration on rheological behavior of alginate/PZT suspensions.

of 1.87 μm and a density of 7.6 g/cm^3 were employed in the experiment. The stoichiometric starting composition of the PZT powders is listed in Table 1, which is presumed to be near the morphotropic phase boundary (MPB) and makes it suitable to be utilized as a host of hydrophones for the excellent electromechanical coupling coefficients. Deionized water with conductivity of 1.02 $\mu\text{S}/\text{cm}$ was used as the solvent. The sodium alginate (NaAlg, Sinopharm Chemical Reagent Co., Shanghai, China) was dissolved in deionized water with different concentrations (1.0, 1.5, 2.0, 2.5 and 3.0 wt%). 1.0 mol/L solutions of calcium chloride (CaCl_2 , Sinopharm Chemical Reagent Co., Shanghai, China) was employed as gelling agents.

10 wt% suspensions with PZT powder and the premixed alginate solutions were prepared by ball-milling for 6 h, followed by degassing in a rotary evaporator under vacuum. Then 60 mL of the prepared slurry was placed in cylindrical beakers and sprayed with calcium

chloride solution until a thin alginate gel layer was formed on the top, which allowed the directional diffusion of Ca^{2+} ions gradually. Afterwards, 20 mL of CaCl_2 solution was added on the thin gel layer and it took 48 h to achieve hexagonally structured anisotropic capillary gels. Then the cylindrical gel body was washed in deionized water until the resultant water was free of Na^+ , Cl^- , and Ca^{2+} .

After that, the densification of gel bodies through ion exchange was carried out by soaking in 1 mol/L gluconolactone (Anhui Xing Zhou Medicine Food Co., Anhui, China) solutions for 48 h, which hydrolyzes slowly to form acid. Solvent exchange from water to tertiary butanol (Tianjin Wing Tai Chemical Reagent Co., Tianjin, China) was achieved by immersing the wet gels into TBA for 12 h, and freeze-drying was subsequently adopted to achieve a low shrinkage of the samples. After drying, the green bodies were sintered at 1150 $^\circ\text{C}$ for 2 h to obtain 3-1 type PZT ceramics.

The rheological behavior of slurries was evaluated using Kinexus pro rotational viscometer (Malvern Instruments Ltd., UK). The porosity and density of sintered specimens were determined by the Archimedes method. The microstructures were directly observed on the surfaces under the scanning electron microscopy (SSX-550; Shimadzu Corp., Kyoto, Japan). Pore size distribution was analyzed by a mercury intrusion method (AutoPore IV 9500; Micromeritics Instrument Corporation, GA). For the measurement of electric properties, porous PZT ceramics were machined to be disk-shaped with a diameter of 10 mm and a thickness of 1.5 mm. Both surfaces of the samples were coated with a thin silver paste and heated at 550 $^\circ\text{C}$ for 20 min to form the Ag electrodes. To minimize the penetration of silver paste into the inner parts of porous PZT ceramics, the silver paste was made highly viscous by adding into ethyl cellulose (EC), and the disk-shaped specimens were placed vertically during the whole operation process. Then the Ag-pasted specimens were poled at 120 $^\circ\text{C}$ for 20 min by applying a DC field of 10 kV/mm in a bath of silicon oil, and aged for 24 h before testing. Here, one important thing to note is that the axially polarized porous PZT ceramics should be ultrasonic cleaned for 3 min with acetone to remove the silicone oil thoroughly. The longitudinal piezoelectric coefficient (d_{33}) was measured by a quasi-static d_{33} -meter (ZJ-3A; Institute of Acoustics, Chinese Academy of Science, Beijing, China). The relative permittivity (ϵ_r) and dielectric loss factor ($\tan \delta$) were measured using an impedance bridge (HP-4194A; Hewlett-Packard Development Company, CA). The parameters of hydrostatic strain coefficient (d_h) and hydrostatic figure of merit (HFOM) were determined from the measured values and other physical parameters.

3. Results and discussion

3.1. Rheological behavior of slurries

Fig. 1 gives the plots of apparent viscosity versus shear rate for aqueous alginate/PZT suspensions with sodium alginate concentration ranging from 1.0 wt% to 3.0 wt%. All measurements were carried out in suspensions containing 10 wt% PZT. As shown in Fig. 1, when the concentration of sodium alginate was less than 2.0 wt%, the suspensions resembled Newtonian fluid nature with reduced apparent viscosity. As the concentration of sodium alginate increased to 2.5 wt%, the suspensions exhibited non-Newtonian behavior with apparent viscosity increasing obviously at low shear rates. As is known to us, with the increase in sodium alginate concentration, there were more amounts of organic macromolecules in the corresponding alginate/PZT slurries, which have played a role of binder and tangled together in the static state. By applying a sequential set of shearing forces, the molecular entanglements were unlocked, and the sodium alginate molecules rearranged in line along the direction of shear force, hence, there was a declining tendency of shearing stress between flow layers, which has given rise to the shear thinning behavior of alginate/PZT suspension over the applied shear rates. Nonetheless, it should be noticed that, the

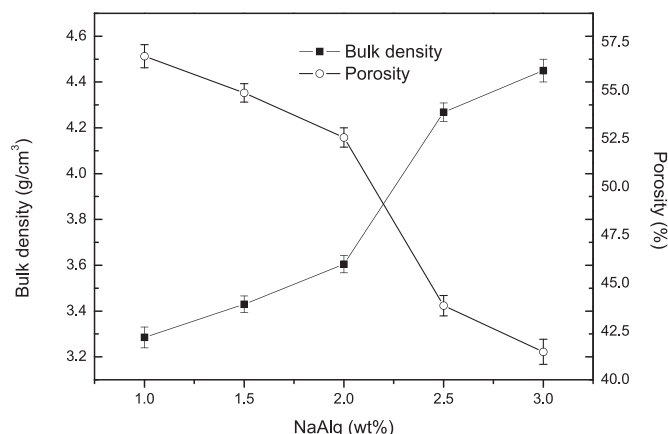


Fig. 2. Variations of porosity and bulk density of 3-1 type PZT ceramics with sodium alginate concentration.

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