

**E#≋₹**S

Journal of the European Ceramic Society 30 (2010) 101–105

www.elsevier.com/locate/jeurceramsoc

# Study of vibratory behavior of interconnected porous PZT by impulse method

K. Boumchedda <sup>a,\*</sup>, A. Ayadi <sup>a</sup>, T. Aouaroun <sup>a</sup>, G. Fantozzi <sup>b</sup>

<sup>a</sup> Laboratory of Minerals Materials and Composites (LMMC), FSI, University "M'hamed Bouguara" of Boumerdes, 35000, Algeria b Materials Engineering and Science (MATEIS), INSA de Lyon, 69621 Villeurbanne Cedex, France

Received 28 March 2009; received in revised form 6 June 2009; accepted 13 July 2009 Available online 8 August 2009

#### Abstract

Performances in ultrasonic active transducers of interconnected porous lead zirconate titanate (PZT) piezoelectric disks with a porosity ranging from 30 to 70%, and polarized along their axial axis, are investigated. The characterization method used is based on the measurement of the voltage, which appears between the two faces of the piezoelectric element when it is excited by a current impulse. The device used, allows the acquisition of axial and radial vibrations of the transducer, and from these data, electromechanical and acoustic parameters are deduced. One observes that interconnected porosity causes the disappearance of the radial vibrations, and for large porosities the disk vibrates exclusively according to the axial mode.  $k_t$  is increased, the acoustic impedance is reduced, and the axial propagation velocity reaches  $\sim$ 2500 m s<sup>-1</sup> for 30% of porosity. These results show that interconnected porous PZT are suitable for making ultrasonic active transducer, such as biomedical imaging devices. © 2009 Elsevier Ltd. All rights reserved.

Keywords: PZT; Porosity; Piezoelectric properties; Ultrasonic sensors

# 1. Introduction

In this paper, interconnected porous lead zirconate titanate (PZT) piezoelectric ceramics are characterized by an original method developed by Jayet et al. 1 to determine the dielectric, piezoelectric and mechanic properties on a wide range of porosity (30–70%). This method is based on the measurement of the voltage that appears between the two faces of the thin piezoelectric element when excited by a current impulse. It allows the study of the vibratory behavior of these ultrasonic transducers. Usually, one uses the resonance method which gives access to the various electromechanical parameters by measurement of the impedance of a transducer for various frequencies.<sup>2</sup> The resonance method, however, is valid only in the case of dense piezoelectric ceramics, since in the case of porous ceramics it is very difficult to distinguish parallel resonance frequencies from series ones. In our investigation, we used the impulse method where immediate response of the sensor is recorded. In this method, the parasitic phenomena of damping of the waves and

radial vibrations are strongly reduced. This study makes a new contribution to the knowledge of the vibratory behavior of interconnected porous ceramics discs that are polarized according to their axial axis and excited by a current impulse according to the same axis. These materials, which have tri-dimensionally porous structure, present many advantages over dense ceramics in ultrasonic applications such hydrophone, biomedical imaging; extensive studies are done for better controlling their manufacture and electromechanical behavior according to porous volume and to the pore shapes. 4–9

Ultrasonic systems for medical imaging employ piezoelectric transducers (1–30 MHz), which operate in the pulse-echo mode to transmit ultrasonic pulses into the body and also to receive the echoes produced by reflections from internal structures. Optimal design of such transducers requires a piezoelectric ceramic with high thickness electromechanical coupling coefficient  $k_t$  and minimal planar electromechanical coupling coefficient  $k_p$  so to make the ratio  $k_t/k_p$  as large as possible. Large values of piezoelectric charge coefficient  $d_{33}$  and piezoelectric voltage coefficient  $g_{33}$  are highly desirable in order to get a large value of a figure of merit ( $d_{33} \times g_{33}$ ) for pulse echo transducers. The transducer's acoustic impedance must be near that of body tissue ( $\sim 1.5 \times 10^{-6}$  kg m $^{-2}$  s $^{-1}$ ) for strong acoustic coupling, mini-

<sup>\*</sup> Corresponding author. Tel.: +213 554 17 17 51; fax: +213 24 81 82 70. E-mail address: boumchedda\_k@yahoo.fr (K. Boumchedda).

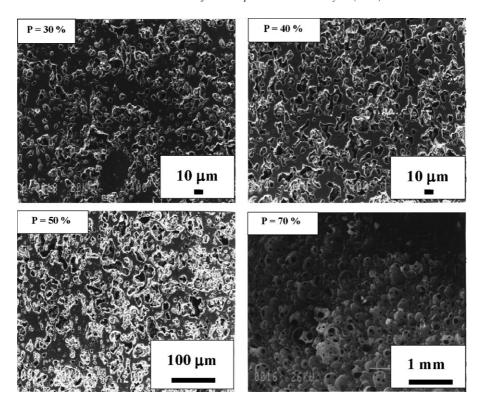


Fig. 1. Microstructure of interconnected porous PZT ceramics.

mizing the reflection of acoustic signal at the transducer/skin interface.

In earlier studies progress was made in investigating the electromechanical applications of porous PZT. Using the Berlincourt- $d_{33}$ -Meter (CPDT 3300), we showed that  $d_{33}$  keeps the same value as that of a dense PZT on a wide range of porosity, and using resonance method we found that  $k_t$  increases strongly with increasing porosity while  $k_p$  decreases. <sup>12,13</sup> The data presented in this paper are collected using the impulse method, which allows us to get better informed about the optimal design and the ideal functioning of axial ultrasonic transducers such as biomedical imaging devices.

## 2. Experiments and measurements

Interconnected porous PZT disks characterized in this paper were obtained by mixing glucose with a commercial PZT powder followed by die pressing with 25 mm in diameter and 1–3 mm in thickness, and sintering at  $1310\,^{\circ}\text{C}$ . The porosity of samples varies from 30 to 70% and the pore size varies from 50 to 500  $\mu m$  (Fig. 1). The technique developed to fabricate porous ceramic has been reported earlier.  $^{12}$ 

We have used an original method developed by Jayet et al. 1 to characterize interconnected porous PZT, which allows to study the vibratory behavior and to determine dielectric, piezoelectric, and acoustic parameters. It is based on the measurement of the voltage which appears between the two faces of a thin piezoelectric element (thickness  $a \ll$  diameter d) when it is excited by a current impulse. The theoretical curve representing the voltage U(t) versus time is given in Fig. 2, and from this curve different electromechanical and acoustic parameters in axial and

radial modes of a transducer vibrations are deduced, such as; the propagation velocity  $V_a$ , the axial resonance frequency  $F_a$ , the elastic constant at a constant electric displacement  $C_{33}$ , the acoustic impedance  $Z_a$ , the relative permittivity  $\varepsilon_r$ , the thickness electromechanical coupling coefficient  $k_t$ , and the piezoelectric deformation coefficient  $h_{33}$ . The porosity is calculated from this relation: p=1-q, p represents the fraction of vacuum and q represents the fraction of PZT. q is calculated from the ratio of porous PZT density d with respect to theoretical density  $d_{th}$ ;  $q=d/d_{th}$ . To determine the figure of merit  $d_{33} \times g_{33}$  for pulse echo transducers; the piezoelectric coefficients  $d_{33}$  and  $g_{33}$  are calculated from these formulas:

$$g_{33} = \frac{h_{33}}{C_{33}} \tag{1}$$

$$d_{33} = g_{33}\varepsilon_0\varepsilon_r \tag{2}$$

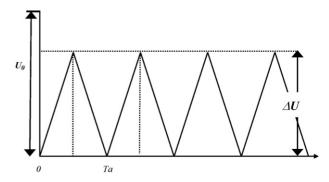


Fig. 2. Theoretical voltage  $\Delta U$  in a piezoelectric element excited by a current (Ta: axial resonance period).

#### 3. Results and discussion

The voltage produced by interconnected porous PZT and dense PZT along the axial and radial axis is reported in Fig. 3. We made the following observations.

In axial vibration, porous ceramics develop more voltage than dense ones. Furthermore, the larger the porosity the higher is the generated voltage. For p = q = 50%, the generated voltage is 2.5 larger than that of dense PZT ceramics, whereas in radial vibration, the voltage is decreased by 10 times. Beyond 50% of porosity; the voltage generated in radial vibration becomes negligible and the transducer vibrates exclusively according to its central axis. We observe also an increase in the resonance period  $T_a$  with increasing porosity volume, which causes a reduction in the axial frequency of resonance  $(F_a = 1/T_a)$ . According to Table 1, the ratio of the amplitude of the first radial vibration to the amplitude of the first axial vibration R, decreases strongly with increasing porosity;  $R \sim 0.66$  for dense PZT is reduced to 0.11 for p = 30%, and R < 0.05 for p > 50%. The ratio of the axial resonance frequency  $F_a$  to the radial resonance frequency  $F_r$  also increases strongly with increasing porosity volume;  $F_a/F_r = 10$  for dense PZT, is increased to values greater than 20 for p > 30%. These results show that radial vibrations of interconnected PZT decrease with increasing porosity volume, and for strongly porous PZT the radial vibrations are non-existent  $(R \ll 0.05)$ , and the vibrations are exclusively in the axial direction. In contrast, dense PZT discs vibrate in the axial and radial directions in the same way in spite of a low discs thicknesses  $(a \ll d)$ .

Electromechanical and acoustic parameters are deduced from these data (Fig. 3) at different transducers thicknesses (a = 1, 1.5, and 3 mm), and plotted versus the fraction of PZT q to investigate the interconnected porous PZT. According to Fig. 4, we observe for a = constant, the axial resonance frequency  $F_a$ increases proportionally with the increase of the PZT fraction q, and for PZT of low porosity (q>0.7), the thickness increase reduces strongly the axial frequency:  $F_a$  decreases by four times when a is increased by three times. For PZT of high porosity, the thickness of the transducer seems not to have an influence on  $F_a$ . In the other hand, the values of the axial propagation velocity  $V_a$  (Fig. 5), the acoustic impedance  $Z_a$  (Fig. 6), the elastic constant  $C_{33}$  (Fig. 7), and the piezoelectric stiffness  $h_{33}$  (Fig. 8), decrease with the introduction of porosity in PZT transducers. In this case, the thickness has no influence on these parameters for a wide interval of porosity (from 30 to 70%). One observes also, that interconnected porosity improves the electromechanical coupling factor  $k_t$ , and the maximum value is obtained in an interval of porosity from

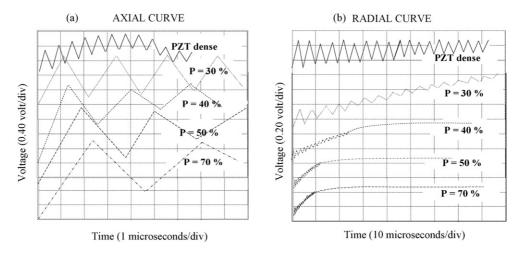


Fig. 3. Voltage produced in porous piezoelectric PZT excited by a current impulse: (a) in axial axis and (b) in radial axis.

Table 1 Electromechanical parameters of porous PZT and dense PZT (thickness a = 1 mm).

| Parameters   |  | Dense PZT | Porous PZT | 1    |        |       |
|--|--|-----------|------------|------|--------|-------|
| Density  | $d (g cm^{-3})$  | 7.25      | 5.30       | 4.40 | 3.70   | 2.40  |
| Fraction of PZT  | $q = (d/7.4^{a})$  | 0.98      | 0.71       | 0.60 | 0.50   | 0.30  |
| Porosity   | $p = (1 - q) \ 100 \ (\%)$                                       | ~2        | ~30        | 40   | 50     | 70    |
| Ratio of the amplitude of the first radial vibration to the amplitude of the first axial vibration | R  | 0.66      | 0.11       | 0.05 | < 0.05 | ≪0.05 |
| Relative permittivity  | $arepsilon_r$  | 1300      | 627        | 490  | 390    | 205   |
| Ratio of the axial resonance frequency to the radial resonance frequency                           | $F_a/F_r$  | ~10       | 16         | 20   | 22     | 35    |
| Piezoelectric voltage coefficient  | $g_{33} (10^{-3} \mathrm{V} \mathrm{m} \mathrm{N}^{-1})$         | 23        | 49         | 61   | 91     | 200   |
| Piezoelectric charge coefficient   | $d_{33} (10^{-12} \mathrm{pC}\mathrm{N}^{-1})$                   | 270       | 267        | 264  | 262    | 274   |
| Figure of merite   | $d_{33} \times g_{33} \ (10^{-12} \mathrm{m}^2 \mathrm{N}^{-1})$ | 62        | 131        | 161  | 238    | 548   |

 $<sup>^{\</sup>rm a}$  7.4 g cm $^{\rm -3}$  is the theoretical density of PZT.

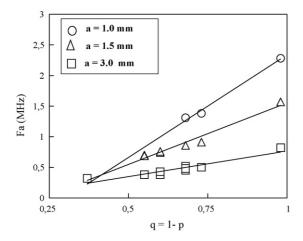


Fig. 4. Axial frequency (Fa) as a function of fraction of PZT (q) at various thicknesses.

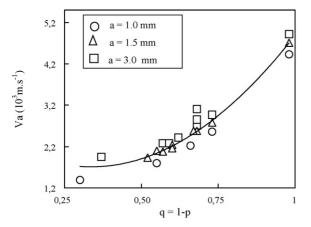


Fig. 5. Axial propagation velocity (Va) as a function of fraction of PZT (q) at various thicknesses.

30 to 40% (Fig. 9). However, porosity strongly reduces the frequency constant Faxa, and a compromise must be found between porous volume and frequency of ultrasonic transducers according to conditions of applications (Fig. 10). In ultrasonic

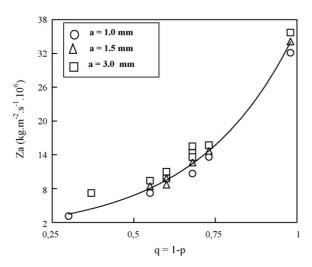


Fig. 6. Axial acoustic impedance (Za) as a function of fraction of PZT (q) at various thicknesses.

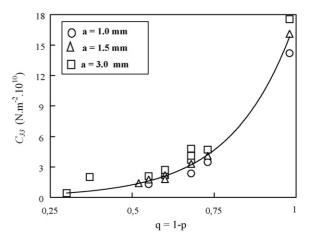


Fig. 7. Elastic constant  $(C_{33})$  as a function of fraction of PZT (q) at various thicknesses.

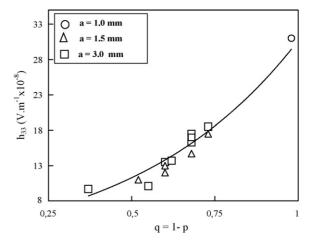


Fig. 8. Piezoelectric stifness constant  $(h_{33})$  as a function of fraction of PZT (q) at various thicknesses.

biomedical imaging applications, the transducers vibrate at frequencies ranged from 1 to 30 MHz. So to make sure a frequency is higher than 1 MHz, the porosity should not be higher than 50%.

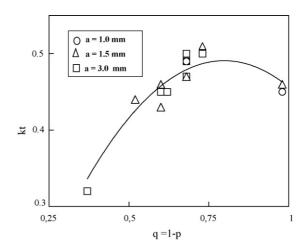


Fig. 9. Axial electromechanical coupling factor  $(k_t)$  as a function of fraction of PZT q at various thicknesses.

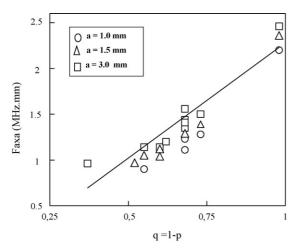


Fig. 10. Axial frequency constant (Faxa) as a function of fraction of PZT (q) at various thicknesses.

One observes that the impulse method is useful to optimize the axial performances of transducers according to porous volume and the thickness of the PZT disc. The radial vibrations generated in a dense PZT thin disc when axially excited, harm the performances of the transducer functioning in a thickness mode (axial vibration), however, the creation of a relative high small volume fraction of porosity ( $\sim$ 30%), considerably reduces radial vibrations, enhanced the figure of merit for pulse echo transducers  $d_{33} \times g_{33}$  and reduces the acoustic impedance  $Z_a$ , which improves the acoustic coupling with media (water, or biological tissue). The optimal design of transducers such medical imaging can be realized with porous PZT discs having 30–50% of porosity which vibrate at 1 MHz and more with a propagation velocity  $V_a > 2000 \, \mathrm{m \, s^{-1}}$ .

# 4. Conclusion

Interconnected porous piezoelectric PZT discs excited along their axis have been characterized and investigated by the impulse method over the porosity range of 30–70%. The results show that introduction of porosity in PZT reduces radial vibrations in favour of axial vibrations, and for large porosities (>70%) the discs vibrates exclusively along the axial

axis. Porosity improves  $k_t$ ,  $g_{33}$  and  $d_{33} \times g_{33}$ , reduces  $Z_a$  to values of better acoustic coupling with biological tissue or with water. The porosity, however, reduces the velocity and the frequency. A compromise must be found between porous volume, thickness, and frequency of PZT discs, depending upon the applications sought. For biomedical ultrasonic active transducers, we found that optimal axial performances are found in an interval of porosity from 30 to 50%.

### References

- Jayet, Y., Baboux, J. C., Perdrix, M. and Lethiecq, M., Use an impulse method to measure different parameters of a thin piezoelectric element. *Ultraso. Int. 87 Conf. Proc.*, 1987, pp. 849–855.
- Ire Standards on Piezoelectric Crystal, Measurements of piezoelectric ceramics. In Proc. IRE, vol. 49, 1961, pp. 1162–1169.
- Boumchedda, K., Hamadi, M. and Fantozzi, G., Properties of a hydrophone produced with porous PZT ceramic. *J. Eur. Ceram. Soc.*, 2007, 27, 4169–4171.
- Bowen, C. R., Perry, A., Lewis, A. C. F. and Kara, H., Processing and properties of porous piezoelectric materials with high hydrostatic figures of merit. *J. Eur. Ceram. Soc.*, 2004, 24, 541–545.
- Galassi, C., Processing of porous ceramics: piezoelectric materials. J. Eur. Ceram. Soc., 2006, 26, 2951–2958.
- Zeng, T., XianLin, D., ShuTao, C. and Hong, Y., Processing and piezoelectric properties of porous PZT ceramics. *Ceram. Int.*, 2007, 33, 395–399.
- Zhang, H. L., Jing-Feng, L. and Zhang, B.-P., Microstructure and electrical properties of porous PZT ceramics derived from different pore-forming agents. *Acta Mater.*, 2007, 55(1), 171–181.
- Bowen, C. R. and Kara, H., Pore anisotropy in 3–3 piezoelectric composites. *Mater. Chem. Phys.*, 2002, 75, 45–49.
- Bowen, C. R. and Topolov Yu, V., Piezoelectric sensitivity of PbTiO<sub>3</sub>-based ceramic/polymer composites with 0–3 and 3–3 connectivity. *Acta Mater.*, 2003, 51, 4965–49767.
- Safari, A., Ting, S. M. and Livneh, S. S., Development of piezoelectric composites for transducers. In *Journées d'études-8 et 9 Juin*, 1993.
- Smith, W. A., Shaulov, A. A. and Auld, B. A., Tailoring the properties of composites piezoelectric materials for medical ultrasonic transducers. In *Proc.* 1985 IEEE Ultrasonics Symp., 1985, pp. 642–647.
- Boumchedda, K., Abadlia, M. T., Kondratas, A. and Fantozzi, G., Elaboration and parameter analysis of porous PZT piezoelectric ceramics. *Mechanika*, 2004, 45(1), 58–62.
- Boumchedda, K., Paletto, J. and Fantozzi, G., Study of proprieties of porous piezoelectric ceramics and impregnated with polymer. *J. Phys. IV France*, 2005, 123, 171–176.