



Preparation of Functionally Graded PZT Ceramics Using Tape Casting

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Abstract. Functionally graded ferroelectric ceramics (FGM) have been fabricated for pyroelectric applications by an aqueous tape casting technology. The FGM produced in this study consisted of a porous Lead Zirconate Titanate (PZT) tape-cast ceramic, which was made by starch inclusions; sandwiched between two dense PZT layers by stacking and lamination. This paper investigates the effect of porosity on the microstructure and electrical properties of the PZT FGM samples produced. The microstructure of the porous and laminated sintered structures was studied using Scanning Electron Microscopy (SEM). The grain size of the porous layer tended to decrease with increasing corn-starch content. The dielectric constant and pyroelectric coefficient of the FGM both showed decreasing behaviour with increasing porosity.

Keywords: ceramics, PZT, FGM, tape casting, aqueous

1. Introduction

This paper reports the fabrication of three-layer lead zirconate titanate (PZT) functionally gradient materials (FGMs) by a tape casting technique. The central layer of the FGM was porous with the porosity produced by the addition of starch to the slip [1, 2]. The porous layer was sandwiched between two dense layers by stacking and lamination.

The microstructure and electrical properties of the FGMs are reported here.

2. Experimental Procedure

Three layer FGMs were produced by tape casting using a polyvinyl alcohol binder (89% hydrolysis, Sigma-Aldrich UK). The tapes for the outer layers were produced by aqueous tape casting, details of which can be found elsewhere [3]. To produce the central layer 1–20 wt% of corn-starch (mean particle size 15 μm ,

Sigma-Aldrich UK) was added to the aqueous PZT slips. After tape casting the dried green tapes were cut and stacked for lamination. Lamination of the FGMs was conducted using a pressure of 45 MPa at 50°C for approximately 15 minutes. Samples were burnt out at 500°C in order to remove the organics present in the tape. Samples were sintered at 1250°C for 45 minutes. The total thickness of the sintered FGM samples was approximately 450 μm . The porosity of the samples was measured using the Archimedes method. Microstructures of sintered FGMs were observed by scanning electron microscopy (SEM). The capacitance of poled samples was measured using an impedance analyzer at room temperature in the frequency range 30–20000 Hz. The dielectric constant of the samples was calculated from the capacitance data. The pyroelectric current of poled samples was measured using a digital multi-meter (Keithley 6420) from 24 to 26°C on heating and cooling. The exact thickness of the samples was measured using a flat faced micrometer. The thickness data of each sample was taken into account when determining the pyroelectric coefficient on heating and cooling.

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Table 1. Showing the measured porosity of the PZT FGM samples.

wt% corn-starch added to middle layer	Porosity (%) of FGM
0	2.38
1.5	5.59
2.0	4.58
3	8.56
5	10.11
9	10.42
7	12.05
11	14.68
15	16.00



Fig. 1. Grain size of the porous layer as a function of porosity.

3. Results and Discussion

Table 1 shows the measured porosity of the complete PZT FGM samples after sintering. Porosity increased

from 2 to 16% with increasing corn-starch content. The increase in porosity was due to the cavities formed from the burnout of the corn-starch during the binder burnout stage. These were large enough to be stable during sintering.

Figure 1 shows the grain size of the porous middle layer of the FGM samples as a function of corn-starch content in the green tape. Grain size decreased with increasing corn-starch content.

Figure 2(a) and (b) show fracture surfaces for sintered FGM samples which contained 1.5 and 15 wt% corn-starch respectively in the middle layer. It is apparent that more porosity can be observed in the middle layer of the sample in Fig. 2(b) than in Fig. 2(a). No delamination of the layers was observed after sintering. This indicates good adhesion between the layers as a result of the lamination process.

Figure 3 shows the measured dielectric constant as a function of porosity for the PZT FGM samples. The dielectric constant also showed a decrease with increasing porosity. A similar effect has been shown in bi-layer porous PZT ceramics [4].

Figure 4 show the average pyroelectric coefficient of the PZT FGMs as a function of porosity. The average pyroelectric coefficient decreased as porosity increased. This is consistent with work on the pyroelectric coefficient of low density PZT produced by incomplete sintering [5].

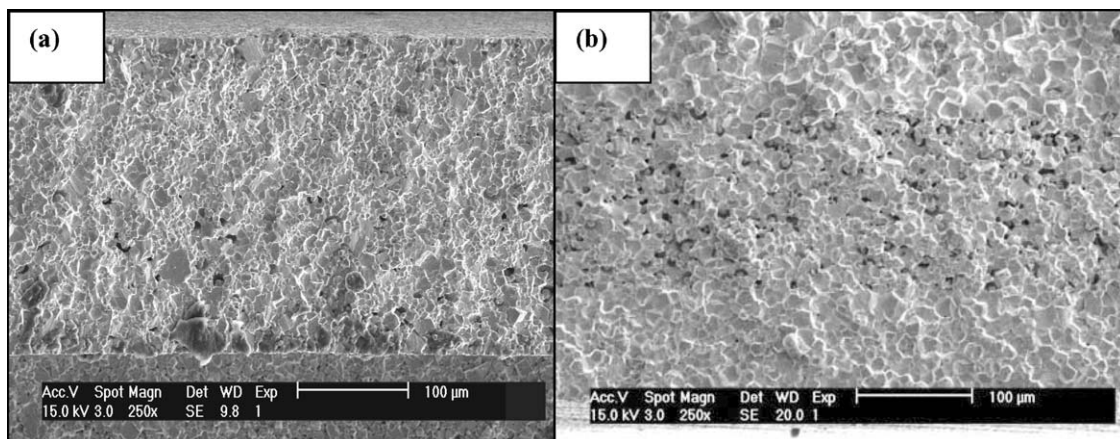


Fig. 2. SEM micrographs of the fractured surfaces of sintered FGMs, a 1.5 wt% corn-starch samples and a 15 wt% corn-starch sample respectively.

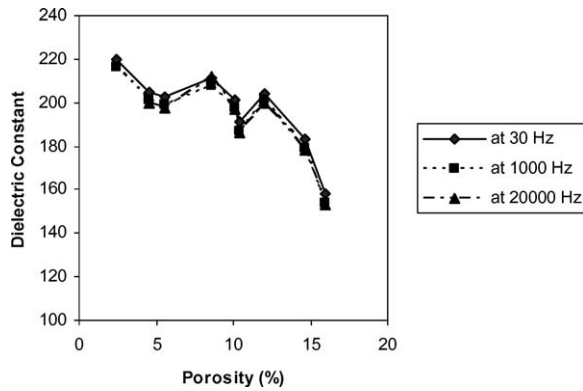


Fig. 3. Dielectric Constant of the samples at 30, 1000 and 20000 Hz as a function of porosity.

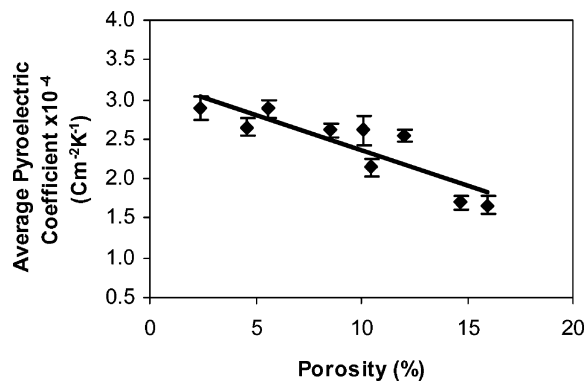


Fig. 4. Showing the average pyroelectric coefficient of the samples as a function of porosity.

4. Conclusions

Porous PZT ceramics have been fabricated successfully by adding corn-starch to tape casting slips. FGMs have been produced by subsequent stacking and lamination of PZT layers such that the porous middle layer of FGM is formed. The addition of corn-starch to PZT slips gave rise to porous PZT ceramics with a maximum porosity of 16%. The grain size of the porous layer decreased on increasing corn-starch content. Electrical characterization of PZT FGM samples showed that the dielectric constant and the pyroelectric coefficient of the samples decreased with increasing porosity.

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References

1. J. Luyten, J. Coymans, A. De Wilde, and I. Thijis, *Key Engineering Materials*, **206–213**, 1937, (2002).
2. S. F. Corbin, *Journal of American Ceramic Society*, **82**, 1693 (1999).
3. A. Navarro, *PhD Thesis*, Cranfield University, United Kingdom (2001).
4. Z. He, J. Ma, R. Zhang, and T. Li, *Journal of European Ceramic Society*, **23**, 1943 (2003).
5. M. Hosseini, S.J. Moosavi, *Ceramics International*, **26**, 541 (2000).