PREPARATION AND CHARACTERISATION OF FUNCTIONALLY GRADED PIEZOCERAMICS

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Introduction

- Functionally graded materials (FGM) may be used when a trade-off must be found between different properties that are important for the performance
- In the case of piezoceramics, a gradient in the piezoelectric activity through the material has the specific consequence that the even harmonics otherwise forbidden can be excited; this may be exploited for making ultrasonic transducers with a large bandwidth [1,2]
- In the present work, two types of gradients have been introduced into a soft PZT piezoceramic: a gradient in porosity and a gradient in composition

FEM modelling

- A soft PZT disc consisting of 5 layers has been modelled in COMSOL Multiphysics v. 3.5
- The impedance and displacement have been simulated by harmonic analysis in three cases: 1) all 5 layers identical; 2) linear gradient in *d* tensor; 3) non-linear gradient in *d* tensor
- In the two gradient cases, even harmonics appear (Fig. 1) and asymmetric displacement is seen

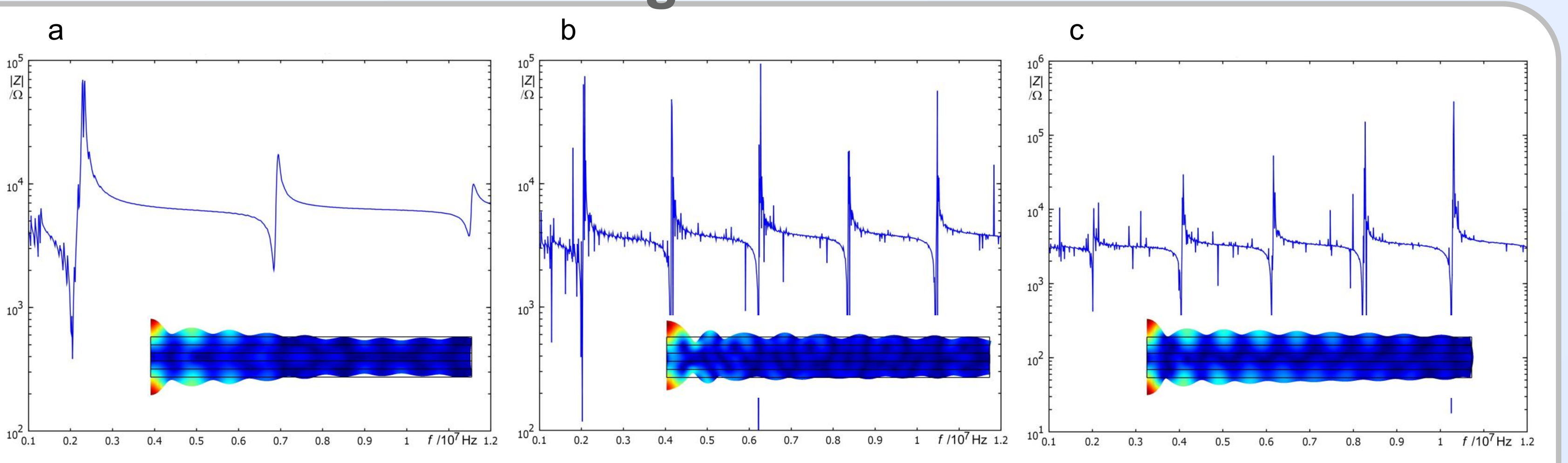

Fig. 1. Simulations of piezoelectric discs consisting of 5 layers. (a) all 5 layers identical. (b) linear gradient in *d* tensor: [1, 0.8, 0.6, 0.4, 0.2]**d*. (c) non-linear gradient in *d* tensor: [1, 0.6, 0.3, 0.01, 0.01]**d*. The inserts show displacement at the thickness resonance.

Experimental work and microscopy

- In order to obtain a *porosity* gradient, Pz27 [3] discs were pressed with 5 layers of increasing porosity: 4 %, 11 %, 18 %, 25 %, 32 % (nominal vol. percentage after sintering, relative to theor. density)
- After plane grinding, only the 4 most dense layers remained, and these are barely visible by optical and electron microscopy (Fig. 2)
- The gradient in *composition* was prepared by unidirectional diffusion of Mg through the thickness of a Pz27 disc
- In practice this was done by pressing a disc of MgO together with a Pz27 disc (uniaxial pressure 155 MPa) and co-sintering them at T > 1240 ºC to obtain a dense microstructure (cf. Fig. 3)
- Unfortunately, the level of Mg in the PZT matrix was too low to be detected with certainty by EDS, as seen from Fig. 4

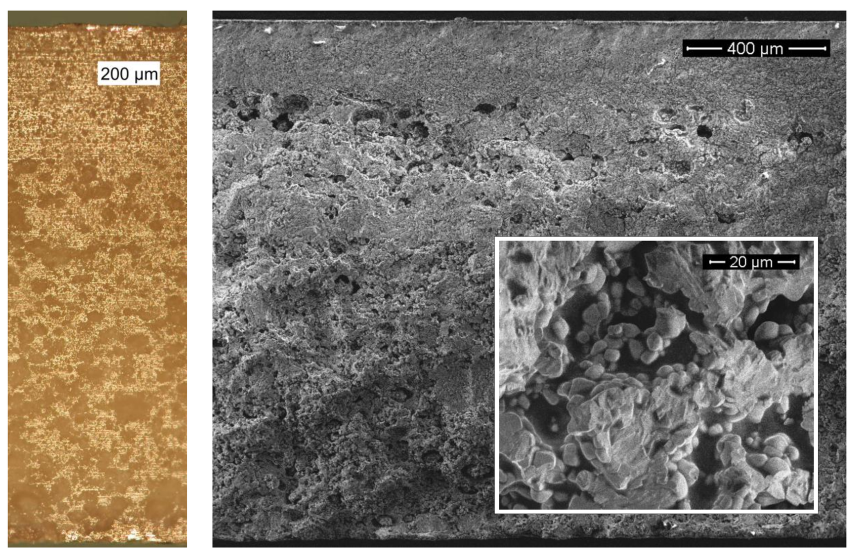

Fig. 2. Images of a sample with a gradient in porosity: edge (OM) and fracture surface (SEM).

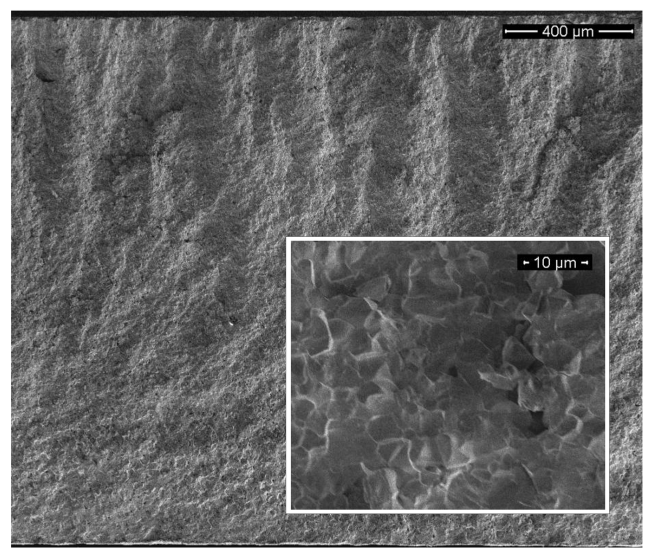

Fig. 3. SEM images of a sample with a diffusion gradient of Mg.

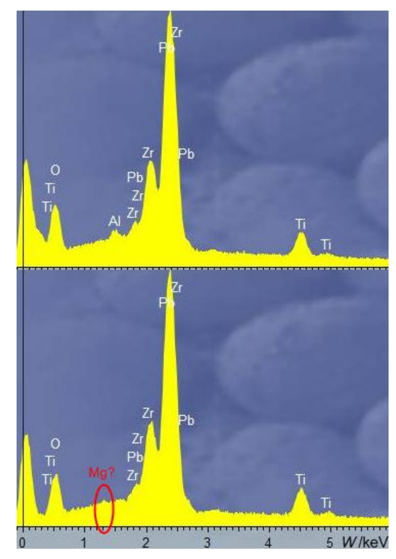

Fig. 4. EDS spectra of reference (top) and Mg-doped sample (bottom).

Electrical characterisation

- As shown earlier [4], the appearance of even harmonics in the thickness resonance spectrum is a good indicator of a gradient in piezoelectric properties
- For samples with a *porosity* gradient, even harmonics were insignificant (see Fig. 5), but in the case of a *diffusion* gradient, even harmonics were clearly seen (Fig. 6)
- In order to somehow quantify the even harmonics, an empirical parameter r_i has been defined as the height of the phase peak of the i'th harmonic resonance relative to the height of the fundamental thickness resonance: $r_i = [\theta_i - (-90^o)]/[\theta_1 - (-90^o)]$
- In Table 1, r_2 and other characteristic parameters are summarised and compared to values obtained by thermal, directional depoling

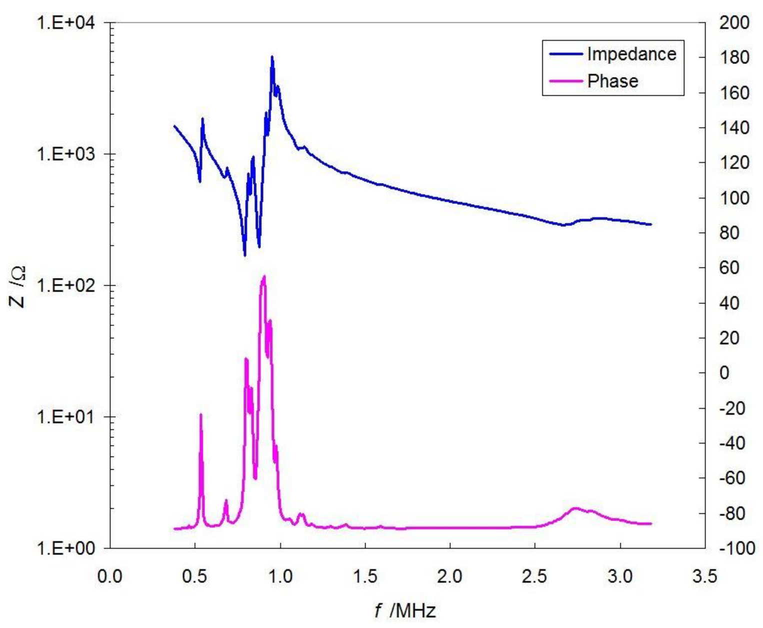

Fig. 5. Impedance spectrum of sample with a porosity gradient. No even harmonics are seen.

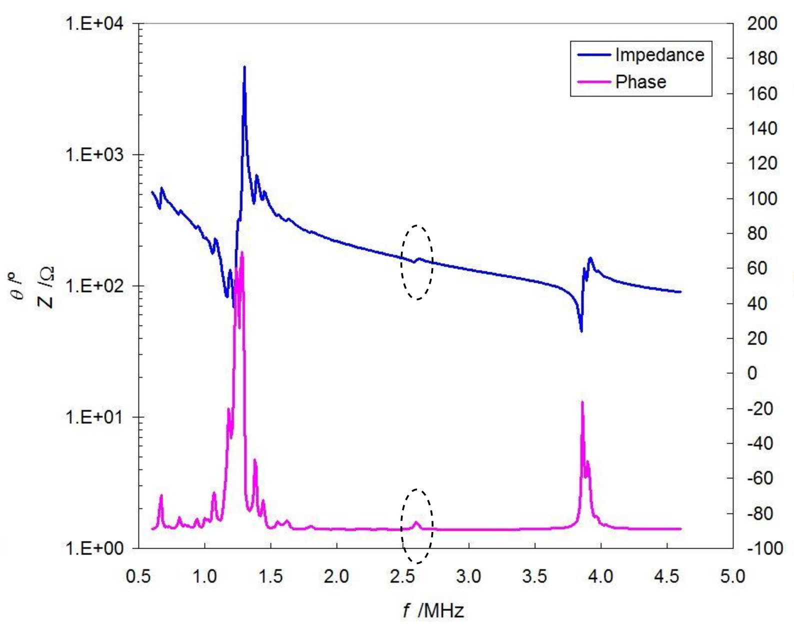

Fig. 6. Impedance spectrum of sample with a diffusion gradient, showing a second harmonic.

Table 1. Summary of key data for samples with a gradient in porosity (Pz27-por) and Mg conc. (Pz27-Mg) compared to an earlier sample prepared by thermal, directional depoling (diameter, height, thickness coupling factor and rel. height of 2nd harm.)

	Pz27-por	Pz27-Mg	Pz59-tdd [4]
D /mm	8.00	11.1	15.7
h /mm	1.50	1.66	1.00
k_t	0.54*	0.37	0.32
r_2 ¤	<1.5 %	3.1 %	18 %

*) value uncertain due to parasitic modes
¤) defined in the left side of this box

Conclusions and outlook

- FEM modelling showed that even harmonics may appear in the thickness resonance in the case of linear and non-linear gradients in piezoelectric properties
- Discs with a porosity gradient have been successfully prepared by uniaxial pressing, but they did not show even harmonics in the thickness resonance
- Experience with porous PZT [3] shows that elastic and dielectric properties are much more affected by porosity than piezoelectric properties, so this result seems to indicate that even harmonics are more sensitive to a piezoelectric gradient than to stiffness and permittivity gradients; further FEM modelling would be useful to investigate this
- The samples with a porosity gradient showed a very high thickness coupling factor (> 0.5), which may be exploited in combination with improved acoustic matching to soft matter (e.g. body tissue) exhibited by the porous side
- A diffusion gradient of Mg caused even harmonics in the thickness resonance to appear, although less pronounced than previously seen for depoling by a thermal gradient
- The mechanism is believed to be a doping effect (Mg^{2+} acting as a *B*-site acceptor), gradually changing the piezoelectric properties

Literature

[1] Yamada, K., Yamazaki, D. & Nakamura, K. (2000): Broadband ultrasound transducers using a plate with a graded piezoelectric constant formed by an internal temperature gradient. In Proc. of the 2000 IEEE Ultrasonics Symposium, 1017-1020.

[2] Yamada, K., Sakamura, J. & Nakamura, K. (2001): Equivalent network representation for thickness vibration modes in piezoelectric plates with a linearly graded parameter. IEEE Trans. UFFC, 48 [2] 613-616.

[3] http://www.ferroperm-piezo.com (2010).

[4] Ringgaard, E., Zawada, T., Hansen, K., Lang, S.B., Feuillard, G. & Le Clézio, E., (2009): Preparing functionally graded piezoceramics by partial, directional depoling – Part I: Electrical and microstructural characterization. Presented at PIEZO 2009, Zakopane (PL), 1 - 4 March 2009.

Acknowledgement

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